

# **Transfer of Radionuclides from Soil to Vegetation in Selected Irish Ecosystems**

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This project is submitted in fulfillment of the HETAC requirements for the award of Doctor of philosophy by research and thesis.

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## ABSTRACT

This thesis details the findings of a study relating the transfer of  $^{238}\text{U}$ ,  $^{228}\text{Ra}$  ( $^{232}\text{Th}$ ),  $^{226}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  from soil to vegetation in an Atlantic blanket bog, upland blanket bog and semi-natural grassland situated along the north-west coast of Ireland. The results of this study provide information on the uptake of these radionuclides by the indigenous vegetation found present in these ecosystems. The ecosystems chosen are internationally recognizable ecosystems and provide a wide variety of vegetation species and contrasting soil physiochemical properties which allow the influence of these parameters on radionuclide uptake to be assessed. The levels of radionuclides in the soil and vegetation were measured using gamma spectrometry, alpha spectrometry and ICP-MS. The nutrient status of the vegetation and soil physiochemical properties were measured using atomic absorption, flame photometry and other analytical techniques.

The results of the study indicate that the uptake of  $^{238}\text{U}$  and  $^{228}\text{Ra}$  ( $^{232}\text{Th}$ ) by vegetation from all three ecosystems was negligible as the levels in all vegetation was below the limits of detection for the methods used in this study. These results appear to indicate that the vegetation studied do not possess the ability to accumulate significant levels of these radionuclides however this assumption cannot be upheld in the case of the Atlantic blanket bog as the levels in the soil of this ecosystem were too low for detection. Similar results were obtained for  $^{226}\text{Ra}$  uptake in both the Atlantic blanket bog and grassland for all vegetation with the exception of *H. lanatus* from the grassland ecosystem. Radium-226 uptake in upland blanket bog was higher and was detectable in the majority of vegetation indigenous to this ecosystem. Transfer factor values ranged from 0.07 to 2.35 and the TF values for *E. tetralix* were significantly higher than all other vegetation studied. This species of heather demonstrated the ability to accumulate  $^{226}\text{Ra}$  to a greater extent than all other vegetation. The uptake of  $^{226}\text{Ra}$  by upland blanket bog vegetation appears to be significantly influenced by a range of soil physiochemical properties. The nutrient status of the vegetation, in particular the calcium content in the vegetation appears to have a negative impact on the uptake of this radionuclide.

Potassium-40 was detectable in all vegetation present in the three ecosystems and the levels in the grassland soil were significantly higher than the levels in both bogland soils. Transfer factor values for Atlantic blanket bog vegetation ranged from 0.9 to 13.8 and were significantly higher in *E. vaginatum* in comparison to *C. vulgaris*. Potassium-40 TF values for upland blanket bog vegetation on average ranged from 1.4 for *C. vulgaris* (stems) to 5.2 for *E. vaginatum* and were statistically similar for all species of vegetation. Transfer factor values for grassland vegetation ranged from 0.7 to 3.8 and were also statistically similar for all species of vegetation indicating that the transfer of  $^{40}\text{K}$  to vegetation within the upland bog and grassland ecosystem is not dependent on plant species. Comparisons of  $^{40}\text{K}$  TF values for all three ecosystems indicate that the uptake in *E. vaginatum* from the Atlantic blanket bog was statistically higher than all other vegetation studied. This appears to indicate that *E. vaginatum* has the ability to accumulate  $^{40}\text{K}$ , however, this species of vegetation was also present in the upland blanket and did not demonstrate the same behaviour. The uptake of  $^{40}\text{K}$  by vegetation from all three ecosystems was significantly affected by a range of soil physiochemical properties and in some cases the results were contradictory in nature possibly indicating that the affect of these parameters on  $^{40}\text{K}$  uptake is species dependent. The most obvious trend in the data was the influence of soil CEC and magnesium levels in vegetation on  $^{40}\text{K}$  TF values. A positive correlation was apparent between the CEC of the soil and  $^{40}\text{K}$  uptake in vegetation from both the Atlantic blanket bog and grassland ecosystem. A similar

trend was apparent between magnesium levels in vegetation and  $^{40}\text{K}$  TF values for the upland blanket bog and grassland vegetation.

Caesium-137 levels were found to be significantly higher in the two bogland soils in comparison to the grassland soil and levels of  $^{137}\text{Cs}$  decreased with increasing soil depth. Transfer factor values for Atlantic blanket bog vegetation ranged from 1.9 to 9.6 and TF values were significantly higher in the leaves of *C. vulgaris* in comparison to all other vegetation from this ecosystem. Caesium-137 TF values for the upland blanket bog vegetation on average ranged from 0.29 for *E. tetralix* to 1.6 for *C. vulgaris*. Uptake by the leaves of *C. vulgaris* was significantly higher than all other vegetation present thereby supporting the trend found within the Atlantic blanket bog vegetation. These results appear to indicate that the leaves of *C. vulgaris* have the ability to accumulate significant quantities of  $^{137}\text{Cs}$  and also that the uptake of  $^{137}\text{Cs}$  by this vegetation is dependent on plant compartment as the stems of this vegetation contained significantly lower levels than the leaves in both ecosystems. The uptake of  $^{137}\text{Cs}$  by grassland vegetation was very low and was only detectable in a fraction of the vegetation sampled. Caesium-137 TF values for grassland vegetation were in general lower than 0.02. The impact of soil physiochemical properties and nutrient status of vegetation on  $^{137}\text{Cs}$  uptake by vegetation appears to be complex and in some cases contradictory. The most apparent trend in the data was the positive influence of vegetation nutrients on  $^{137}\text{Cs}$  uptake in particular the magnesium levels present in the vegetation and to a lesser extent the calcium levels present.

The results in general indicate that the uptake of  $^{226}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  by the chosen vegetation is varied and complex and is significantly dependent on the species of vegetation, soil radionuclide concentration, soil physiochemical properties and the nutrient status of the vegetation.

## DECLARATION

The work reported in the thesis was carried out by Carmel Moran-Hunter during the period the period September 2001 to May 2008. Aside from the analysis conducted by Bord na Mona Laboratories, the work was carried out solely by Carmel Moran-Hunter. The thesis has not previously submitted to any other institution or awarding body for consideration for the awarding of any degree.

Signed:

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## Acknowledgements

I wish to acknowledge the following people for their support, assistance and encouragement throughout this project:

- My project supervisor, John O' Dea for his ongoing support and encouragement.
- Dr. Mark Dowdall and Dr. Paddy Fleming for their assistance and advice throughout the project.
- Dr. John Bartlett for his assistance and support.
- Dr. Luis León Vintó and the staff of the Dept. of Physics, UCD for their technical advice and support.
- The lecturing staff of the School of Science, IT Sligo.
- The technical staff of the School of Science, IT Sligo for their invaluable assistance throughout the project.
- The library staff at IT Sligo.
- The technical and sales staff of Canberra Industries.
- The research department with special thanks to Jean, Aoife, Bosco, Pat Maughan, Shiela, Tasneem, Angel and Michael for making the life of a postgrad. that bit easier. Also to Caroline and Mia for their support and encouragement.
- Mum, Dad, Marie, Ken, Peter, Caroline, Brendan, Gerard and the Moran-Hunter family for their support throughout the last few years.
- And especially Trevor and Richard for every single thing.

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## 1.0. INTRODUCTION.

### 1.1 General Introduction and Study Objectives

The phenomenon “radioactivity” was discovered a little more than a century ago and since then a significant amount of research has been carried out on the effects of radiation in the environment and its applications within such disciplines as medicine, geology and archaeology. Radionuclides occur naturally within the environment and are found predominately in rock from which they are leached into the soil, ground water and surface water. The discovery of nuclear fission in the 1930s which marked the dawn of the atomic age brought about the introduction of a range of radionuclides which up until then were not present in the environment. The impact of these radionuclides, both natural and anthropogenic on humans and their environment is of great concern due to the ionising nature of the radionuclides and possible toxic effects of the individual radionuclides. This study will focus on the transfer of both natural and anthropogenic radionuclides within natural terrestrial ecosystems within Ireland.

Traditionally, radiological protection has been firmly focused on the protection of man. This has arisen primarily because the relevant advisory body the International Commission on Radiological Protection (ICRP) has maintained a very strong bias towards human health issues stating the belief that “*if man is adequately protected than other living things are also likely to be sufficiently protected*”. In recent years there has been increasing pressure on the ICRP to explicitly demonstrate environmental protection. Thus they are currently focusing their attention on protection of the environment as a whole as opposed to just protecting humans. They are looking more closely at the effects on the non-human environment on the basis that the levels set out in the current radiation protection standards which are focused on mainly human protection may be too high for other natural organisms or vegetation.

The EC have part-funded two related projects within the 5<sup>th</sup> Framework Programme. i.e., 1) Framework for Assessment of Environmental Impact (FASSET) and 2) Environmental Protection from Ionizing Contaminants in the Arctic (EPIC). FASSET's main aim is to create a framework for assessment of the effects on biota and ecosystems by ionizing radiation. EPIC's main aim, while being focused in the Arctic, is to develop a methodology for the protection of natural populations of organisms in Arctic ecosystems by gathering information on the environmental transfer and fate of selected radionuclides through aquatic and terrestrial ecosystems. Both projects are adopting the same general approach – development of a framework based on the protection on an ecosystem approach as opposed to just the protection of man. A major part of these projects and also of environmental radiation protection herein will be the collation of information in regard to the transfer of radionuclides from soil to vegetation within entire ecosystems. This area of research has been greatly neglected on a worldwide scale (FASSET, 1999).

Environmental surveys carried out in relation to soil to plant transfer have mainly been concerned with the fate of anthropogenic radionuclides namely  $^{137}\text{Cs}$  and very little attention has been given to the fate of natural radionuclide within terrestrial ecosystems. For this reason the radionuclides under investigation within this study are the uranium ( $^{238}\text{U}$  and  $^{226}\text{Ra}$ ) and thorium ( $^{232}\text{Th}$  and  $^{228}\text{Ra}$ ) series radionuclides along with the radionuclide,  $^{40}\text{K}$ , and the anthropogenic radionuclide  $^{137}\text{Cs}$ . Knowledge of the distribution and behaviour of these radionuclides is important for a number of reasons. In the radioecological context radioisotopes of caesium are among the most hazardous radionuclides being released into the biosphere following weapons testing and nuclear accident (Chernobyl) fallout. Isotopes of caesium are chemical analogues of potassium and consequently may be incorporated in the nutrient cycle of plants thereby leading to increased exposure to animals and humans. Uranium series radionuclides are the precursors of radon gas and improved knowledge of the behaviour of  $^{226}\text{Ra}$  ( $^{238}\text{U}$  daughter) in soils may provide for more accurate estimation of the radon production potential of soils in the country. Uranium being a heavy metal, presents similar hazards in the environment as other heavy metals contaminants.

Very few studies have been carried out on the fate of natural radionuclides within Ireland and most of the work pertains to concentration in soil and this in itself is very limited.

McAuley and Marsh (1992) carried out a national survey of natural radionuclide activities within the soil based on the 10 km<sup>2</sup> national square grids but only looked at the distribution within the soil as is the case with Dowdall (2000) where an extensive survey was carried out on the spatial analysis and speciation of uranium and thorium series radionuclides in soil in Donegal. Mitchell *et al* (1990) assessed the total deposition of <sup>137</sup>Cs over Ireland arising from weapons fallout and Chernobyl and the Radiological Protection Institute of Ireland (RPII) have studied <sup>137</sup>Cs distribution within woodland ecosystems and semi-natural grassland (Rafferty *et al*, 1994a, Rafferty *et al*, 1994b, Rafferty *et al*, 1994c, Rafferty, 1996 and Rafferty *et al*, 1997).

This study provides an integrated assessment of the distribution of selected natural and anthropogenic radionuclides within the soil and their transfer to specific vegetation within three internationally recognized ecosystems.

The primary objective of this study is to determine the transfer of selected natural and anthropogenic radionuclides from soil to vegetation in Irish ecosystems. The study will focus on the uptake of <sup>238</sup>U, <sup>232</sup>Th, <sup>226</sup>Ra, <sup>40</sup>K and <sup>137</sup>Cs by vegetation indigenous to three natural ecosystems in Ireland. It will also focus on the impact of the parameters listed below on radionuclides uptake by vegetation to ascertain if any of these parameters inhibit or enhance the uptake of the selected radionuclides:

1. Soil radionuclide concentration
2. Soil physiochemical properties
3. Species of vegetation
4. Nutrient status of vegetation

The potential transfer of these radionuclides into the human food-chain will also be discussed with respect to the levels found in the vegetation in comparison to levels outlined in current legislation. The conclusions derived from this study should provide

valuable information on the fate of these radionuclides within the chosen ecosystems and also on their behaviour within similar ecosystems around the world.

## 1.2 Ecosystem Selection

The primary objective of this study was to investigate the fate of radionuclides in natural or semi-natural ecosystems within Ireland. The three ecosystems chosen were (1) Atlantic blanket bog, (2) upland blanket bog and (3) semi-natural pasture (wet grassland). These ecosystems were chosen due to their extensive coverage within Ireland. Peatlands are natural habitats in Ireland with bogs being one of Ireland's most characteristic features covering approximately 17% (1/6<sup>th</sup>) of the island. Approximately 60% of Ireland is covered by grassland of one type or another with this percentage rising if annual crops such as cereals and amenity grass like golf courses and lawns are included.

Peat bogs are particularly important in relation to radionuclide distribution as the association between natural radionuclides (uranium and thorium series) and organic matter has been well established by a number of authors and peat bogs have long been accepted as acting as uranium "sinks" with significant amounts of uranium accumulating over long periods of time (Douglas, 1991, Schulz, 1965, Hansen and Stout, 1968, Rusonova, 1962, Talibudeen, 1964, Dowdall, 2000). Organic matter has the ability to adsorb caesium in a non-specific manner resulting in increased uptake of caesium by vegetation dominant on organic soil and in turn increased uptake by animals grazing in these regions (Livens and Loveland, 1988)

Distribution of radionuclides in grasslands is also very pertinent as agriculture in Ireland is based on grass, with 93% of farmland being devoted to the production of grass (Rath and Peel, 2005). Radionuclide uptake by pasture vegetation has the ability to enter the human food chain either by being retained in the meat of grazing cattle or through entering the milk of lactating cows and being consumed by humans in this manner. The grassland chosen has been grazed in the past by dairy cattle, which produced milk for sale and by sheep which was sold for meat production purposes. The presence of high levels

of radionuclides in the grasses within this region would give a general indication of the uptake of radionuclides by the animals grazing here.

Radionuclide uptake has been shown to be dependent on a number of parameters the most important being: plant species, chemical and physical nature of soil, time of sampling and speciation and concentration of radionuclide in soil (Arkhipov *et al*, 1984, Mortvedt, 1994, Schulz, 1965). The physical and chemical characteristics of the bog and grassland ecosystems are very different from each other thereby providing the opportunity to determine the characteristics affecting the uptake of radionuclides by indigenous vegetation.

### 1.2.1 Atlantic Blanket Bog: Cloonty Bog, Ballintrillick, Co. Sligo.

Cloonty Bog defined as an Atlantic blanket bog (Natura 2000 Code 7130, European Commission 1999, Habitat Category Code PB3, Fossitt 2000) is located in Ballintrillick, Sligo (Grid Ref. G 733 528).



Figure 1 Atlantic Blanket Bog: Cloonty Bog (OSI map, Discovery Series 16, Scale 1:50,000)

Blanket bogs are defined as extensive bog communities or landscapes on flat or sloping grounds with poor surface drainage, in oceanic climates with heavy rainfall, characteristic of western and northern Britain and Ireland. They often cover extensive areas with local topographic features supporting distinct communities. They are broken down into two

distinct types: 1) Hyper Atlantic bogs of the western coastlands of Ireland, western Scotland and its islands, Cumbria, and northern Wales and 2) Blanket bogs of high ground, hills and mountain in Scotland, Ireland, western England and Wales (European Commission, 1999). Atlantic blanket bog is more restricted in its distribution within Ireland than upland blanket bog and is largely confined to wetter regions along the western seaboard where the annual rainfall exceeds 1250 mm and occurs on flat or gently sloping ground below altitude 150 m. Peat depths vary considerably (1.5 – 7 m) depending on the underlying topography, and are usually intermediate between those of raised and upland blanket bog. The vegetation of lowland blanket bog is typically ‘grassy’ in appearance and is characterized by abundant Black Bog-rush (*Schoenus nigricans*), Purple Moor-grass (*Molinia caerulea*) cotton grasses (*Eriophorum* spp.), Deergass (*Trichophorum caespitosum*) and White Beak-sedge (*Rhynchospora alba*). Heathers are common but are not as abundant as in upland blanket. Broadleaved herbs and Bog-myrtle mat also be locally abundant. *Sphagnum* mosses (*Sphagnum auriculatum*, *S. magellanicum*, *S. compactum*, *S. papillosum*, *S. nemoreum*, *S. rubellum*, *S. tenellum*, *S. subitens*) and mucilaginous algae, collectively known as *Zygodogonium ericetorum* may also be present; the latter increases in abundance in overgrazed areas where *Sphagnum* mosses are rare or absent (European Commission 1999, Fossitt 2000).

Sections of Atlantic blanket bogs (and all bogs) have been transformed into cutover bogs (Habitat Category Code PB4, Fossitt 2000), which are areas where part of the original mass of peat has been removed through turf cutting or other forms of peat extraction. Turf cutting activity is characterized by vertical face banks or rectangular peat ramparts where the cutover section is at a distinctly lower level than the uncut high bog. These banks vary in height, depending in part on the depth of peat, and remain in evidence for a long time after turf cutting ceases. Cutover bog is a variable habitat, or complex of habitats, that can include mosaics of bare peat and revegetated areas with woodland, scrub, heath, fen and flush or grassland communities. The nature of recolonising vegetation depends on a number of factors including the frequency and extent of disturbance, hydrology, the depth of peat remaining, and the nature of the peat and the underlying substratum (Fossitt, 2000). The habitat ‘Depressions on peat substrates of the

*Rhynchosporion*' (Natura 2000 Code 7150, European Commission 1999) can occur in pockets on cutover bogs, mostly in association with areas of cutover raised bog. The plant species dominant in the latter habitat are *Rhynchospora alba*, *R. fusca*, *Drosera intermedia*, *D. rotundifolia*, and *Lycopodiella inundata* (European Commission, 1999).

Cloonty bog is divided into two regions; undisturbed peat and cutover bog. The underlying bedrock geology is lower and middle carboniferous, mostly limestone as illustrated in Figure 1 (Appendix 3). The dominant vegetation species found in both regions of the site is a mixture of *Calluna vulgaris* (ling heather), *Eriophorum vaginatum* (bog cotton) and *Sphagnum* mosses.

*Calluna vulgaris* also known as ling heather is a diffuse, purple flowered, evergreen shrub generally up to 60 cm in height although it may approach 1 metre or more. It has numerous ascending branched stems, rooting at the base and bearing numerous short shoots. It is dominant over large areas on well-drained acid soils. *Calluna vulgaris* prefers atmospheric humidity and flourishes on soils poor in mineral salts and areas where it is free of competition from taller, more quickly growing plants. While it has a widespread and universal distribution, it only attains substantial growth and abundant flowering in drier areas (Webb *et al*, 1996).

*Eriophorum vaginatum* known as bog cotton or haretail cotton grass is a perennial plant, which grows in tussocks and spreads by rhizomes. It is found on wet peaty soils, particularly blanket bogs and very frequently found in the north, west, and centre and occasionally in the south and east of Ireland. It is easily distinguished from the other cotton grasses by single flower spikes. It grows to a height of 30 – 50 cm, flowers from April to May and fruits from May to June (Webb *et al*, 1996).

*Sphagnum* mosses were the third species of vegetation sampled and due to the large quantities of sample required by gamma spectrometry the moss was not separated into individual species. Mosses have leafy stems but the fine hair-like structures anchoring them to the soil are not true roots. Peat mosses (*Sphagnum*) have their assimilatory cells

interspersed with large, empty, hyaline cells capable of holding large amounts of water (Jahns, 1983).

### 1.2.2 Upland Blanket Bog: Cronamuck Valley, Donegal

The second ecosystem classified as an upland blanket bog (Natura 2000 Code 7130, European Commission 1999, Habitat Category Code PB2, Fossitt 2000) is located in the Cronamuck valley in Donegal along the Atlantic coast of Ireland. The region lies on the north-eastern part of the Barnesmore pluton, an isolated granite some 52 km<sup>2</sup> in area which forms the Bluestack mountain range in central Donegal (Grid Ref H 022 928). The region has been the subject of uranium prospecting efforts in the past and studies have shown that areas within this region show elevated levels of natural radioactivity (Dowdall, 2000).

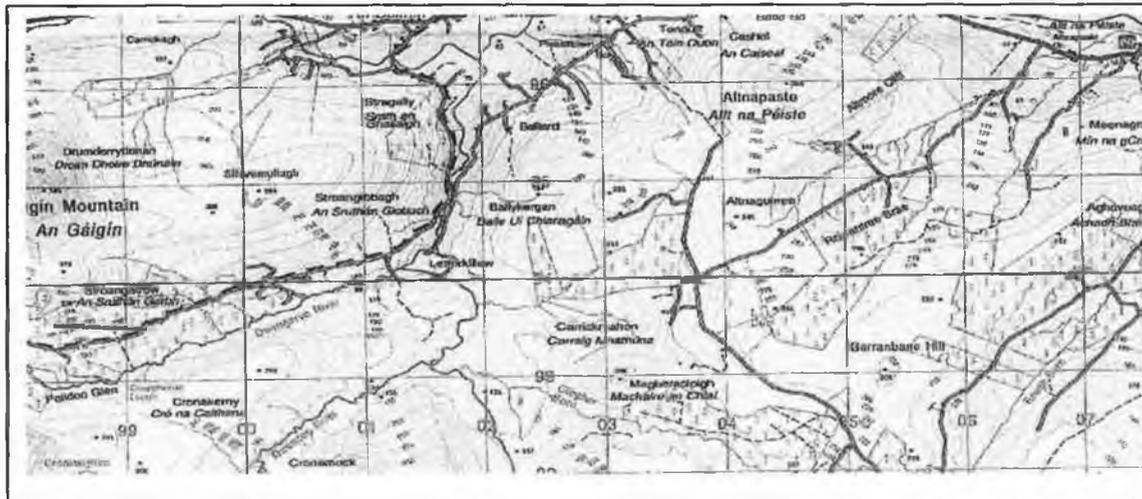


Figure 2 Upland Blanket Bog: Cronamuck Valley (OSI map, Discovery Series 11, Scale 1:50,000)

Upland blanket bogs occur on flat or gently sloping grounds above altitude 150 m and is widespread on hills and mountains throughout Ireland. The 150 m altitude limit serves to distinguish upland from lowland blanket bog but is loosely applied. Peat depths vary and normally fall in the range of 1 – 2 m, but can be deeper in pockets. Vegetation is typically

dominated by Deergrass (*Trichophorum caespitosum*), cotton grasses (*Eriophorum vaginatum*) and dwarf shrubs such as Ling (*Calluna vulgaris*), Cross-leaved heath (*Erica tetralix*) and bilberry (*Vaccinium myrtillus*). Purple moor grass (*Molinia caerulea*) and Crowberry (*Empetrum nigrum*) may be locally abundant. Cover of *Sphagnum* mosses is usually high in undamaged in areas of undamaged bog (Fossit, 2000).

The blanket bog in Donegal was chosen based on results from previous studies which showed regions within the site to contain elevated levels of natural radioactivity in particularly  $^{238}\text{U}$  and  $^{226}\text{Ra}$  (Dowdall, 2000). However, soil radioactivity levels vary greatly within this region and the levels found within this study for  $^{226}\text{Ra}$  were below the national average of 46.1 Bq/kg for Ireland (McAuley and Marsh, 1992). The dominant vegetation within this upland blanket bog is a mixture of *Calluna vulgaris* (ling heather), *Eriophorum vaginatum* (bog cotton), *Myrica gale* (bog myrtle), *Erica tetralix* (cross-leaved heath), *Molinia caerulea* (purple moor-grass) and *Sphagnum* mosses. Information on *Calluna vulgaris*, *Eriophorum vaginatum* and *Sphagnum* mosses can be reviewed in Section 1.2.1. Detailed descriptions of *Myrica gale*, *Erica tetralix* and *Molinia caerulea* are given below.

*Myrica gale* also known as bog myrtle is the most characteristic bushy species of Irish bogs and can also be found along some lake shores. It is most common on the Atlantic coasts of Ireland and Scotland. It is a small bushy species which obtained its name from the myrtle-like aroma given off by its shoots when crushed. It grows to a height of approximately 1 m, is deciduous and has hairless, long and narrow dark green, slightly glossy leaves. Plants flower from April to May (Philips, 1977, Wyse- Jackson, 1993).

*Erica tetralix* more commonly known as cross-leaved heath is found on raised and blanket bogs as a low, sprawling shrub. It is common throughout Britain and Ireland and in Atlantic areas of Europe from southern Portugal to northern Norway. It is a perennial shrub with pink bell-shaped drooping flowers borne in compact clusters at the ends of its shoots, and leaves in whorls of four. The flowers appear between June and October. The plants generally grow to a height of 30 – 40 cm (Philips, 1977).

*Molinia caerulea* more commonly known as purple moor-grass is a perennial grass which grows to a height of 90 cm. Its leaves are flat and grayish, approximately 3-6 mm wide, not persisting through the winter. Flowering occurs between July and September and the grass is commonly found on wet heaths and moors on acidic soils with a fluctuating water table (Fitter *et al*, 1984).

### 1.2.3 Wet Grassland: Doonally, Sligo.

The wet grassland habitat, which is a semi-natural pasture, is located in Doonally, Sligo (Grid Ref. G 725 385). The underlying bedrock geology is lower and middle carboniferous, mostly limestone as illustrated in Figure 1.2.2.1 (Appendix 3).

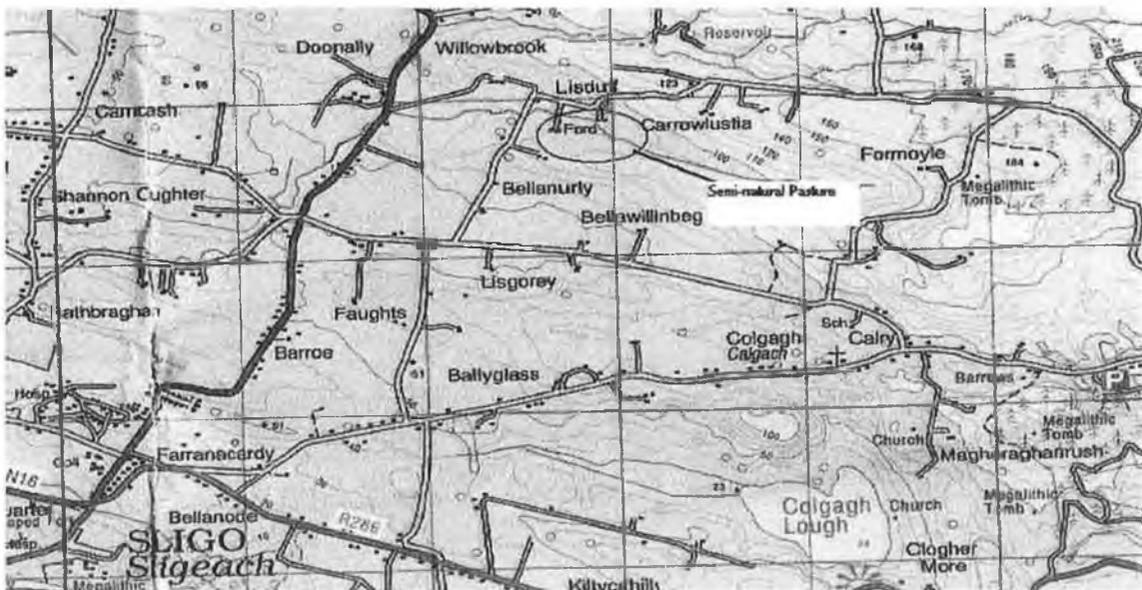


Figure 3: Wet Grassland Site: Doonally (OSI map, Discovery Series 16, Scale 1:50,000)

Wet grasslands (Habitat Category Code GS4, Fossitt 2000) can be found on flat or sloping ground in upland and lowland areas. It occurs on wet or waterlogged mineral or organic soils that are poorly drained or, in some cases, subjected to seasonal or periodic flooding. On sloping ground, wet grassland is mainly confined to clay-rich gleys and

loams, or organic soils that are wet but not waterlogged. Wet grassland often contains abundant rushes (*Juncus effuses*, *J. acutiflorus*, *J. articulatus*, *J. inflexus*) and/or small sedges (*Carex flacca*, *C. hirta*, *C. ovalis*), in addition to grasses such as Yorkshire-fog (*Holcus lanatus*), Creeping Bent (*Agrostis stolonifera*), Marsh Foxtail (*Alopecurus geniculatus*), Rough Meadow-grass (*Poa trivialis*) and Tufted Hair-grass (*Deschampsia caespitosa*). Purple Moor-grass and broad land herbs may also be present. Wet grassland may also contain examples of the annexed habitat 'Molinia meadows on calcareous, peaty or clayey-silt-laden soils: *Molinion caeruleae* (Natura 2000 Code 6410, European Commission 1999).

The grassland site was located at a fairy fort, which is joined to other grazing fields by a deep depression, thus leaving the area inaccessible to heavy machinery. The field was chosen on this basis, as the soil has not had any chemical fertilizer applied to it. Artificial fertilizers increase the concentration of both uranium and potassium in the soil, and thus would affect the natural levels of these radionuclides in the soil and also have significant implications on the chemical reactions in the soil. The dominant vegetation in this area was *Holcus lanatus* (Yorkshire fog), *Juncus effuses* (soft rushes), *Urtica dioica* (Stinging Nettle), *Crataegus monogyna* (Hawthorn tree), *Acer pseudoplatanus* (Sycamore tree) and moss.

*Holcus lanatus*, also known as Yorkshire fog is a very common, greyish softly downy tufted perennial grass. It is common in Ireland on pasture, roadsides, waste ground, and open-woodland on all types of soil. It is covered with soft hairs, which gives the plant a soft velvety feel. It often appears flushed pink or purple or may be almost white. Its height can vary from 20 cm to 100 cm and flowers between May and August (Phillips 1980, Fitter *et al*, 1984).

*Juncus effuses* also known, as soft rush is a native perennial forming thick tufts common throughout Britain and Ireland in damp often-grassy places and differs from hard rush by being softer, smooth and light yellowish green. Its height can vary from 30 cm to 150 cm and flowers from June to August (Phillips, 1980, Fitter *et al*, 1984).

*Urtica dioica* also known as stinging nettle is a dark green perennial 60-100 cm high, with oval pointed strongly toothed, often cordate leaves 5-10 cm long. Commonly found on roadsides, thickets and in waste areas. Plants have a covered stem with stinging hairs that release irritant histamine-containing juices when broken (Webb *et al*, 1996, Rose 2006).

*Crataegus monogyna* commonly known as the hawthorn tree is a bush or small tree with spiny branches. Leaves are dull green, somewhat triangular, with rather acute lobes. Flowers are generally 12 mm across and usually white while rarely pink. Commonly found in hedges, fields and rocky places (Webb *et al*, 1996).

*Acer pseudoplatanus* also known as the Sycamore tree produces a valuable pale creamy coloured wood. The leaves are typically maple shaped, 9 – 12 cm across with five pointed lobes and a coarsely toothed edge. The flowers are smallish and green and occur in drooping heads; they are useful source of early spring nectar for honey bees. Although the sycamore is one of the most familiar Irish trees, it is not native, having been introduced to the British Isles in the fifteenth or sixteenth century. It grows to be a large and occasionally a magnificent tree with greyish bark. More often it is found as a small weedy tree of waste places, woods, hedges and gardens. Although native to the European mountains it is ideally suited to the Irish climate (Wyse Jackson, 1993).

The grassland also contained moss species which were unidentified.

### 1.3 Chemistry of Soil

Soils consist of minerals, organic matter, water and air arranged in a complicated physiochemical system that provides the mechanical foothold for plants in addition to supplying nutritive needs. A soil system can be classified into horizontal layers known as

soil horizons, which differ in their physical characteristics. A diagram of the soil horizons can be seen below in Fig. 4.

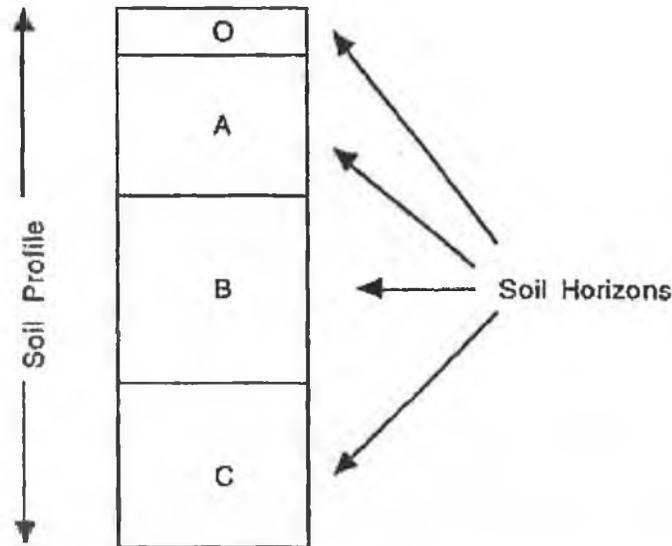


Figure 4: Major Soil horizons found in a soil profile.

The “O”- horizon refers to a surface layer of raw or partly decomposed organic material. The content of organic matter in this horizon is several times greater than the underlying horizons. The “A”- horizon also known as the surface soil is generally 30 – 60 cm in depth and is the part of the soil in which living matter (plant roots and microbes) is most abundant. The “B”- horizon, known as the subsoil extends to approximately 1 meter below the surface. It is a zone of an accumulation of nutrients and clay particles and usually has a relatively high content of iron and aluminium oxides and clay particles. The “C”- horizon refers to the parent material beneath the “A” and “B”- horizons, the upper part of which is usually loose and slightly weathered rock (Eisenbud, 1987).

### 1.3.1 Inorganic component of Soil

The inorganic content of soil may fall into different textural classes depending on the percentages of sand, silt and clay present. The textural classes into which the soil may be differentiated can be seen in Fig. 5.

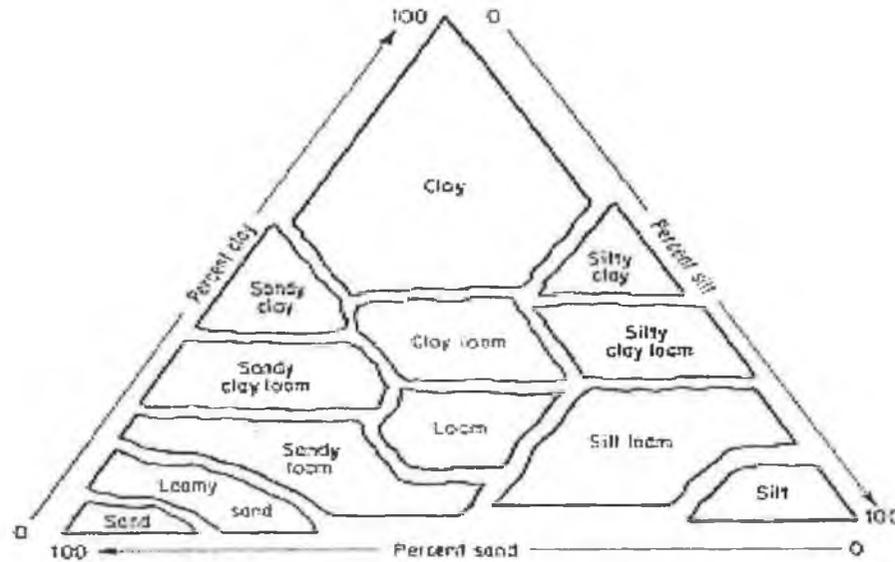


Figure 5: Soil Texture triangle

Sand consists largely of primary minerals such as quartz and has a particle size range from  $60\ \mu\text{m}$  –  $2\ \text{mm}$ . Silt consists of particles in the size range of  $2$  –  $60\ \mu\text{m}$  and clay particles are less than  $2\ \mu\text{m}$  in diameter. The sand and silt portion of the soil are essentially chemically inert compared to the clay and organic fractions, which are the portions responsible for the chemical reaction in the soil. The sand and silt portions play an important role in providing a skeleton for the soil.

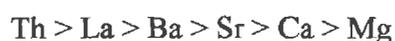
As clay particles are the inorganic fraction most responsible for chemical reactions in the soils they shall be discussed in more detail. They are minute, plate – shaped aluminosilicate crystals consisting of silicon, aluminium, magnesium, oxygen and hydrogen. They may also contain potassium, calcium and other elements. An essential characteristic of the plate like particles of secondary silicates is the abundance of negative surface charges. The resultant ability of these clay particles to attract positive ions to their

surface is one of the most important properties of soils. The majority of nutrient ions in the soil are adsorbed onto the soil particle surfaces as opposed to dissolved in the soil water. This allows the soil to hold a much larger reservoir of nutrients in the soil and also prevents the loss of nutrients by leaching. The cations present in the soil water can exchange with those adsorbed onto the soil. The ability of a soil to hold and exchange cations with the soil water is quantified using the Cation Exchange Capacity (CEC)<sup>1</sup>. Soils with high clay content in comparison to sand or silt content will have a high cation exchange capacity due to the large surface area of the clay and also the presence of the negative charges within the crystals (Eisenbud, 1987, Kohnke & Franzmeier, 1995).

The rate at which a cation is adsorbed to a clay colloid is dependent on a range of factors, some of which are: surface potential, valence, and hydrodynamic radius. When a mixture of monovalent and divalent ions is present in a soil solution adsorption is usually shifted in favour of the divalent ions. As the surface potential increases the concentration of divalent ions adsorbed will increase. The hydrodynamic radius also affects specific adsorption of the cations. Generally ions with smaller hydrated sizes are preferably adsorbed. The following decreasing order of preference, for adsorption of monovalent cations by clay has been reported (Tan, 1992)



The rate of adsorption for caesium is the highest, as this element is smallest in hydrated size. The ion has a thin hydration shell, which makes a close approach to the clay surface possible. Lithium on the other hand, has the largest hydrodynamic radius and its thick hydration shell increases the distance from the ion to the clay surface. The decreasing order of preference for adsorption of divalent cations on clay is shown below.




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<sup>1</sup> Cation Exchange Capacity (CEC) expressed in milliequivalents per 100g of soil (meq/100g) is a measure of the quantity of readily exchangeable cations neutralizing charge in the soil (Rhoades, 1982).

These series may change depending on the type of clay.

The cations adsorbed to the clay surface due to electrostatic attraction between the negative clay surface and the positive cations are generally free to distribute themselves through the liquid phase by diffusion. However under certain circumstances the adsorbed cations are held so strongly that they cannot be recovered by exchange reactions. These cations are referred to as fixed cations. Fixation can occur with all cations but is most important for  $K^+$  and  $NH_4^+$ , where fixation occurs by a similar mechanism. The most important mode of fixation for these ions is the entrapment of these ions in the intermicellar regions of the clays. When  $K^+$  and  $NH_4^+$  penetrate the intermicellar space, they will fit snugly into the holes. Upon closure of this space these cations will then become trapped between the clay layers. They are then relatively non-exchangeable and are referred to as fixed. Some of the soil minerals responsible for this mode of interaction are micas, montmorillonites, illites and vermiculites (Tan, 1992). Although the quantity of potassium retained by fixation can assume high proportions, sufficient potassium is generally available in the plant available form. Depending on the conditions, significant amounts of the fixed K can be released and made available for plant growth. The presence of organic matter such as humic acid and fulvic acid in soils can accelerate this release. Recent studies have also shown that caesium is very strongly adsorbed to clay particles particularly illite and as is further discussed in Section 1.10.1, "Caesium in the Soil".

### 1.3.2 Organic component of Soil

Soil organic matter is generally broken down into humified and non-humified materials. The non-humified substances are the compounds found in plants and other organisms such as amino acids, proteins lipids, nucleic acids and lignin. These compounds are subject to degradation and decomposition reactions. They can, however, be adsorbed by soil inorganic components such as clay, or they may occur in anaerobic conditions. Under these conditions these compounds will be relatively protected against decomposition (Tan, 1992).

The humic fraction of the soil is of greater importance in relation to this study, as the components of this fraction have the ability to retain radionuclide cations under specified pH ranges. The humified fraction also known as humus is the end product of decomposition of plant materials in soil. It can be subdivided into constituent components according to their solubility in various reagents, most commonly alkali and acidic conditions (Table 1).

<b>Fraction</b>	<b>Alkali Solution (0.1 M NaOH)</b>	<b>Acid Solution (0.1 M HCl)</b>
Fulvic Acid	Soluble	Soluble
Humic Acid	Soluble	Insoluble
Humin	Insoluble	Insoluble

Table 1: Classification of Humic Substances (Tan, 1992).

#### 1.3.2.1 Fulvic Acid

Fulvic acid contains less carbon and more nitrogen and oxygen than humic acids and it seems likely that while its structure is broadly similar to that of humic acid it has a smaller proportion of aromatic units and greater peripheral aliphatic chains (Cressar, Killham & Edwards, 1993).

#### 1.3.2.2 Humic Acid

Elemental analysis of humic acid has shown it to consist largely of carbon, oxygen (50 and 40 % respectively), phosphorous and sulfur (< 1% each) The structure of humic acid is polyphenolic with an array of functional groups including carboxyls, hydroxyls amides and carbonyls (Cressar, Killham & Edwards, 1993).

#### 1.3.2.3 Humin

The alkali insoluble fraction of the soil is the least studied of the three fractions. This is probably due to the extreme difficulty in obtaining “pure” humin, which is free of

inorganic impurities. The structure of humin is broadly similar to that of humic acid but contains less nitrogen. The insolubility of humin is more likely to be due to binding to mineral impurities than to fundamental differences between humin and other soil humic fractions.

The chemical behaviour of humic matter is controlled by the two functional groups, the carboxyl and phenolic – OH groups. The carboxyl group starts to dissociate its proton ( $H^+$  ion) at pH 3.0 and the humic molecule becomes electro negatively charged. The phenolic group dissociates at pH 9.0. Several reactions and interactions can occur with cations due to the presence of these charges. At low pH values, cation exchange mechanisms predominate. However, as the pH increases complex reactions and chelation mechanisms tend to predominate (Tan, 1992). The dissociation of the humic molecules at specific pHs and the interactions of metal cations with these molecules are shown in Figures 6 and 7 respectively.

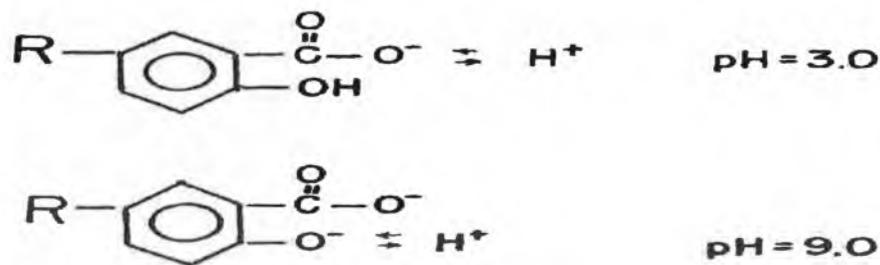


Figure 6: Dissociation of humic molecule at pH 3.0 and 9.0.

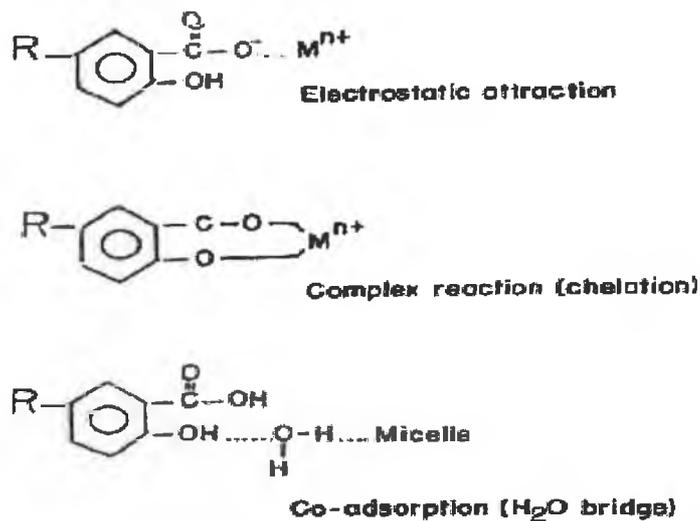


Figure 7: Interaction between humic acid and a metal cation and humic acid and a clay micelle.

#### 1.4 Essential Plant nutrients

In order to understand the uptake of radionuclides by plants it is imperative to have an understanding of plant growth and the nutrients essential for optimum plant growth. As some of the radionuclides of interest are analogues of the essential plant nutrients it is probable that these elements may be taken up in a similar manner or that the nutritive status of the plant may affect their uptake.

There are 13 essential elements required for plant growth. These are broken down into constituent groups depending on how they are obtained, or the quantities required by the plants.

<b>Primary Nutrients</b>	<b>Secondary Nutrients</b>	<b>Tertiary or Micronutrients</b>
Nitrogen (N)	Calcium (Ca)	Boron (B)
Phosphorous (P)	Magnesium (Mg)	Chlorine (Cl)
Potassium (K)	Sulfur (S)	Copper (Cu)
		Iron (Fe)
		Manganese (Mn)
		Molybdenum (Mo)
		Zinc (Zn)

Table 2: Essential elements for plant growth

The primary and secondary nutrients are the constituents of many plant components such as proteins, nucleic acids and chlorophyll and are also essential for processes such as energy transfer, enzyme function and the maintenance of internal turgor pressure. These nutrients, also known as macronutrients, are taken up in large amounts by the plant. The micronutrients, are taken up in relatively small amounts by the plants, have a variety of essential functions in plant metabolism (Barnes & Poore, 1994). Additional mineral nutrient elements, which are beneficial to plants but not necessarily essential, include sodium, cobalt, vanadium, nickel, selenium, aluminium and silicon. The nutrients differ by: 1) how they are absorbed by the plant, 2) their functions in the plant, 3) their mobility in the plant and 4) plant deficiency or toxicity symptoms.

Optimum plant growth requires that nutrients be available in soluble form and in appropriate and balanced amounts. Most agricultural soil contains considerable reserves of nutrients but these are largely tied up in the organic humus or as inorganic ions bound to colloidal clay and humus. Plants can only absorb nutrients as water soluble ions and these reserves are mostly unavailable, although a small proportion are released into the soil solution each year through biological activity and chemical processes (Barnes & Poore, 1994).

## 1.5 Uranium in the Terrestrial Environment

Natural uranium (U) mined from the earth contains the isotopes  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{234}\text{U}$ . The majority (99.2745%) of all the atoms in natural uranium are  $^{238}\text{U}$ . Most of the remaining atoms (0.72%) are  $^{235}\text{U}$ , and a slight trace (0.0055%) is  $^{234}\text{U}$  (Mortvedt, J.J., 1994).

Uranium exists in four valence states,  $\text{U}^{3+}$ ,  $\text{U}^{4+}$ ,  $\text{U}^{5+}$ , and  $\text{U}^{6+}$ , the tetra and hexavalent states being predominant in nature. The hexavalent state is dominant in oxidizing environments such as surficial materials and rivers.

### 1.5.1 Chemistry of Uranium in Soils

Uranium is considered to have some degree of mobility in soils. The uranium content in soil is related to the bedrock from which the soil is formed (ATSDR, 1990). It may be present in soil as a cation and as such will tend to be adsorbed on soil minerals. The degree of adsorption and desorption of an individual cation will depend, in part, on the adsorption energy of the cation and the amount and species of all other ions present. Other factors such as type of clay mineral and the pH of the soil will affect the tenacity with which a particular ion is held. The most likely valence state of uranium in soils is hexavalent and, in the absence of large amounts of organic matter, uranium is generally considered to be mobile and transported as a divalent uranyl ( $\text{UO}_2^{2+}$ ) ion (Schulz, 1965 Bondetti and Sweeton, 1977, Sheppard and Evenden, 1988). Sheppard (1980b) also supports this view and state that hexavalent uranium is invariably found associated with oxygen as the very stable uranyl ion ( $\text{UO}_2^{2+}$ ) and occurs whenever the electrochemical potential is oxidizing such as in weathered rock and surficial material. Tetravalent uranium is found in reducing environments such as in unweathered rock where a reducing potential is maintained. Alloway (1990) states that uranium may be classified as being moderately mobile in oxidizing conditions across the whole pH range and immobile in reducing conditions. The formation of the oxycation  $\text{UO}_2^{2+}$  is probably responsible for the solubility of uranium over a broad range of pH. Also, several organic acids may increase the solubility of uranium in soils. Its solubility may be limited by the formation of slightly soluble precipitates (e.g. phosphates, oxides) and adsorption to clays

and organic matter (Langmuir, 1978). Uranyl carbonate complexing results in a wide range of uranium solubility. The oxidized  $U^{6+}$  (uranyl) ion complexes readily with carbonate and sulphate ions and are readily transported in the hydrological cycle. In reducing waters, uranium is in the  $U^{4+}$  state and as such it has an extremely strong tendency to precipitate and to remain immobile (Gascoyne, 1992). The dominant aqueous species are thought to be  $UO_2^{2+}$ ,  $UO_2(CO_3)_3^{4-}$ ,  $UO_2(CO_3)_2^{2-}$ ,  $UO_2(HPO_4)_2^{2-}$ , with  $UO_2(CO_3)_3^{4-}$  or phosphate complexes forming the dominant species in alkaline waters (Langmuir, 1978). Hostetler and Garrels (1962) reported that U is transported in acid-oxidizing solutions as  $UO_2^{2+}$  or  $UO_2(OH)^+$  ions, as a  $UO_2(CO_3)_2^{2-}$  in neutral solutions, and as a  $UO_2(CO_3)_3^{4-}$  in alkaline solutions. In contrast the reduced tetravalent form of uranium behaves similarly to immobile tetravalent thorium (Hansen and Huntington, 1969). Yamamoto *et al* (1973) also reported adsorption of uranium greatly reduced by the presence of carbonate ions in soil solution where a concentration of  $500 \mu\text{g g}^{-1}$  of carbonate ion in solution reduced the adsorption of uranium by 75%. Precipitation – sorption reactions also increase with increased soil pH and Guimond and Windham (1975) reported greater precipitation of uranium from effluents of wet process phosphorous (P) manufacture when the pH was increased to levels greater than 6. In the soil-water-plant environment the most concern is over oxides of uranium and uranium salts ( $CO_3^{2-}$ ,  $F^-$ ,  $SO_4^{2-}$ ,  $Cl^-$ ,  $NO_3^-$ ,  $PO_4^{3-}$ ) (Sheppard, 1980b)

The clay content of soil is thought to play a major role in the retention of uranium. Mortvedt (1994) stated that divalent cations (uranyl cation) are sorbed by soil clays, through exchange mechanisms, so relative rates of adsorption generally increase as clay content increases in soils. As mobility in soils is inversely related to sorption, downward movement of the uranyl cation would be lower in fine-textured (high clay) soils. Sheppard and Evenden (1988) also stated that retention of uranium is greater in fine-textured soils as they have a higher density of negatively charged sorption. Megumi and Mamuro (1977) reported that uranium content increases with decreasing particle size below  $100 \mu\text{m}$  implying that surface reaction may be an important retention mechanism and that adsorption of uranium is dependent on cation exchange capacity. However, this view is not supported by Masuda and Yamamoto (1971) who found that adsorption of the

uranyl ion was not dependent on the cation exchange capacity of the soil. Kiss *et al* (1988) used the daughter nuclides of  $^{238}\text{U}$  ( $^{214}\text{Pb}$  and  $^{214}\text{Bi}$ ) to determine the uranium concentrations of the native soils (unfertilized) of southern Saskatchewan, Canada. The nuclide  $^{214}\text{Bi}$  was used as an indicator for the presence of uranium and given the possible loss of radon from the soil and the probable disequilibrium between  $^{226}\text{Ra}$  and  $^{238}\text{U}$ , this nuclide was only used as a general indicator. They concluded that heavy clay deposits in the Regina area were also characterized by higher activity of  $^{214}\text{Bi}$  and that below average levels of  $^{214}\text{Bi}$  were generally associated with areas of sandy and loamy textured soils. They found that the radioactivity decreased as soil texture became coarser and regions, which had elevated radioactivity not in-line with this, were probably due to local parent material composition. Megumi (1979) and Jasinska *et al* (1981) concluded that the majority of radionuclides are attached to a soil fraction of  $< 20\mu\text{m}$  (silt and clay), and that this phenomenon is independent of soil type.

Uranium concentration in soils is found to be related to soil organic matter (Talibudeen, 1964). Evidence of sorption of uranium contaminants in phosphate fertilizers by soil organic matter was reported by Rothbaum *et al* (1979). Soil samples from a long-term experiment were separated into two density fractions and analyzed for uranium. In soils without added P, the light, highly organic fraction contained slightly less uranium than the heavy, low-organic fraction. However, the uranium added in super phosphates over a period of 85 years increased the uranium concentration in the light fraction of soil four times more than that in the heavy fraction. Analyses of soils from a long-term pasture experiment in New Zealand showed that the uranium from super phosphate remained in the top 5 cm of soil, which was high in organic matter. Rothbaum *et al* (1979) emphasized that their results found virtually all the uranium applied in super phosphate remained in the surface horizon of soils, but felt that this finding did not necessarily conflict with the work of Spalding and Sackett (1972) who found significantly increased contents of uranium in rivers draining intensively fertilized and farmed agricultural land in south-western USA. They argued that the enrichment observed by Spalding and Sackett (1972) could be due to losses by leaching or soil erosion, and that in the absence

of organic matter, uranium is generally considered to be mobile and is transported as a hexavalent carbonate complex or as a divalent uranyl ion.

Szalay (1964) concluded that the insoluble humic acid content of peat is the geochemical agent responsible for the bioaccumulation of uranium in bioliths. The value given for the geochemical enrichment of  $\text{UO}_2^{2+}$  in peat was  $10^4/1$ . The geochemical enrichment factor (GEF) is defined as the equilibrium distribution ratio of the uranyl ion between the aqueous and humic phases at pH of about 5 and at low concentrations of the ion, i.e.

$$\text{G.E.F.} = [\text{UO}_2^{2+}]_{\text{peat}} / [\text{UO}_2^{2+}]_{\text{aq}}$$

Szalay (1964) considers this factor to be the governing factor in the association of uranium with organic substances in nature. Laboratory studies indicated that the sorption process is a cation exchange process that is identical to the cation exchange properties of humic acids which has long been observed for calcium, potassium and other cations in soil chemistry. As outlined previously, cation exchange properties of humic acids originate in their carboxylic groups. Armands (1961) also reported GEF factors of 9000/1 for peat soils in a bog in north Sweden. Titaeva (1967) postulated that, under oxidizing conditions, uranium is bound to peat by ion exchange. Peat readily extracts uranium from natural waters, reducing  $\text{U}^{6+}$  to  $\text{U}^{4+}$  after sorption onto the organic fraction. Sheppard (1980b) states that decaying plant material or organic mat at the soil surface is an important factor in determining the distribution of uranium in soil. On upland areas this mat varies from non-existent to a thin veneer. However, in low-lying areas it can contribute to a significant accumulation of uranium near the surface. Retention of uranium by organic matter can also cause enrichment lower in the profile such as in a  $\text{B}_{\text{th}}$  horizon, which contains illuviated organic material. The nature and strength of the uranium-organic matter association that is established depends upon the degree of carbonization attained by the humic material (Sheppard, 1980b). Dowdall and O'Dea (1999) conclude that for peat soil with elevated natural radioactivity in Donegal, Ireland the redox potential (Eh) of the soil is a major factor controlling either the enrichment or depletion of uranium within the peat. Results showed that accumulation of uranium

within the soil profile increased as the potential of the soil became more reducing. Durrance (1986) also concluded that uranium accumulation correlated with reducing environment of organic matter. Sheppard and Evenden (1988) state that organic matter plays a two-fold role on the behaviour of uranium in soils. Organic soils are effective at retaining uranium and high concentrations of uranium have been noted in peat bogs with very little distributions of the uranium from point of entry. In contrast, organic complexes and colloids can increase the mobility of uranium in mineral soils.

Jenkins et al (1990a) studied the uranium and thorium levels in British arable and grassland soils. The uranium and thorium concentrations showed no significant difference in the arable or grassland soils surveyed indicating that their study does not support the view that the regular use of phosphate fertilizers on arable land is leading to a build-up of uranium in arable soils. Higher Th/U ratio in arable compared than grassland soils may indicate a preferential loss of uranium from the soil due to leaching and/or plant uptake. Above average concentrations uranium and thorium were associated with areas of high silt and clay content. The combined clay and silt content of grassland soils showed a positive correlation with uranium concentrations. They indicated that the variability of uranium in British grassland soils is generally associated with the  $< 20\mu\text{m}$  (i.e. silt and clay) soil fraction and is independent of soil type: only soils high in carbonate ion or organic matter and those associated with igneous formation appear to deviate from this general rule. The highest concentration of uranium was found in an arable peat, however the peat soil used for the grassland survey was from an upland site and did not show any accumulation of uranium. They authors suggested that this may be due to a combination of high rainfall (1520 mm average rainfall) and low pH (3.2), both factors favouring the leaching of uranium. In both surveys the lowest uranium concentrations were associated with rendzina and brown sands where the parent materials (chalk and sandstone respectively) are both known to contain relatively low levels of uranium. In addition, the high carbonate content of rendzina may lead to a reduction of uranium in these soils due to complexation with carbonate ions and enhanced leaching. Uranium in arable soils ranged from 0.6 to 4.9  $\mu\text{g g}^{-1}$  with an average and standard

deviation of  $2.0 \pm 0.7 \mu\text{g g}^{-1}$ . The uranium in grassland soils ranged from 0.9 to  $3.9 \mu\text{g g}^{-1}$  with an average and standard deviation of  $2.3 \pm 0.8 \mu\text{g g}^{-1}$ .

### 1.5.2 Uptake of Uranium by Plants

Uptake of uranium and thorium series radionuclides by terrestrial plants is relatively low but varies considerably among their daughter elements. Plant uptake of uranium is generally greater than that of thorium and polonium (Mortvedt, 1994). Cannon (1965) concluded that the amount of uranium absorbed by plants varies with the species, time of year, part of plant, availability of uranium in the soil, and chemical composition of the underlying rock. The data also suggested that the type of rooting system and soil moisture conditions influence uranium absorption.

As mentioned previously the nature of the soil and in particular the organic matter and clay content plays a significant role in the mobility of uranium within the soil profile and therefore has a significant effect on the uptake by vegetation growing on the soil. The majority of literature shows that uptake by vegetation is lowest on clay soil, highest on sandy soil and peat soil situated within this range. Sheppard and Evenden (1988) reviewed soil to plant concentration factors for studies subsequent to the review and concluded that concentration ratios (CR:  $[\text{U}]_{\text{plant d.w.}} / [\text{U}]_{\text{soil d.w.}}$ ) values are lowest for fine textured soils as opposed to coarse, peat and tailings soil. Sheppard *et al* (1989) analyzed for U content in a number of different vegetation grown on a number of different soils and found that in general the CR values were high for sands, mostly likely due to low sorption capacity of sands and low for finer textured soils that sorb more of the radionuclides. Sheppard *et al* (1983) grew Swiss Chard in two soils of different and found that the uptake in sandy soil was much higher than that in peat. Cannon (1965) found that the variation in uranium content of the same species (based on a small number of samples) growing on different soil types was quite striking. Plant ash of *Atriplex confertifolia* growing on a colluvial substrate containing 3070 ppm uranium contained 3.0 ppm uranium while plant ash of *Astragalus pressii arctus* growing on shale (80 ppm

uranium) contained 70 ppm uranium. Other plant ash of *Atriplex confertifolia* growing on different substrate types (from 0.9 ppm to 3070 ppm) were found to have 0.26 to 5.9 ppm of uranium, but with no consistent relationship between the amount of uranium in the soil rocks and the amount measured in the plant ash (Cannon 1965).

The translocation of uranium within the plant is of great importance and work carried out in the first half of the 20<sup>th</sup> century indicated that uranium was deposited on the roots and translocation to other plant portions was negligible. A review of the current literature indicates that uranium concentration is highest in roots and leaves followed by stems and lowest in fruits. Acqua (1912) indicated that uranium is adsorbed by the plant roots and stored as a yellow deposit in the cell nuclei of the meristem. This results in destruction of the chromatin and cessation of cell nuclear activity preventing uranium translocation. Vinogradov (1959) pointed out that the apparent selectivity of uranium to roots is attributable to insoluble calcium-uranyl phosphates, which are deposited on the roots surfaces, allowing only a small portion to enter the root sap. However this conflicted with the findings of Cannon (1952) who found that considerably more uranium was absorbed by plants from deposits of calcium-uranium carbonate and limestone than from deposits of calcium-poor sandstone and shale. Alloway (1990) stated that most of the uranium in plant roots apparently precipitates in the tips as autunite,  $\text{Ca}(\text{UO}_2)_2\text{PO}_4$  and that which reaches the plant shoot may be held as a protein complex

Dunn (1986) reported that uranium concentrations in several trees species decreased in the following order: twigs > leaves > roots > trunk. In other words, uranium migrates towards the outer extremities of trees. He cautioned that such vegetation should be sampled by taking similar amounts of growth, e.g. the latest 10 years from each tree, to even out seasonal differences. Drobhov (1964) illustrated by autoradiographs that uranium in the green portion of the plant migrates to those parts where development is the most intense: growing tips, young leaves and seeds. Hoffman (1964) reported results for uranium concentrations in various parts of plants that were in agreement with Drobhov (1964) in that the uranium content of seeds and roots tended to be higher than that of leaves, with seeds of grapes found to be particularly high in uranium but this is not

in agreement with Dunn (1986). Tracy *et al* (1983) studied produce from gardens contaminated by uranium processing in Port Hope, Ontario and found that total uranium concentration in leaves and roots were generally greater than that for fruit. Sheppard and Evenden (1988) carried out a review of all relevant soil to plant concentration factors for uranium and concluded that CR factors were higher for root crops than fruit, cereal, shrub or leafy crops. Arkhipov *et al* (1984) found that the coefficient of accumulation (i.e.  $[U]_{\text{soil d.w.}} / [U]_{\text{plant d.w.}}$ ) for  $^{238}\text{U}$  for various barley plant organs grown on different types of artificially contaminated soil decreased in the following order: roots > straw > grain. The authors concluded that the high accumulation coefficient values associated with the roots were apparently due to the fact that when plants were growing in containers and the root system spreads out in a small volume of soil uniformly contaminated with naturally occurring radionuclides in a soluble form, conditions arise that promote increased absorption of radionuclides by the roots.

Sheppard *et al* (1989) found that fruit crops tended to have very low CR ( $[U]_{\text{plant d.w.}} / [U]_{\text{soil d.w.}}$ ) values followed by root crops with leafy crops tending to have slightly higher CR values. CR factors were shown to be clearly affected by the degree of plant washing and peeling of the root crops. Schreckhise (1980) grew peas and barley in pot culture under green house conditions in a silty loam soil with moderate CEC. CR ( $[U]_{\text{plant d.w.}} / [U]_{\text{soil d.w.}}$ ) factors for peas in terms of leaves, stem/pod and seed were  $170 \times 10^{-4}$ ;  $33 \times 10^{-4}$  and  $0.5 \times 10^{-4}$  respectively and for barley in terms of seed and stem/leaf were  $1.6 \times 10^{-4}$  and  $36 \times 10^{-4}$  respectively. The CR values for both plants decreasing in the following order: leaf > stem > seeds.

Nicholson *et al* (1990) studied soils and vegetation from Cornwall and Sutherland for uranium content. These are two areas in the United Kingdom, which have naturally high levels of uranium and thorium. These high levels are associated with the Late Caledonian Helmsdale Granite, which is known to contain uranium minerals, which are relatively soluble under low to neutral pH conditions. The vegetation sampled was a mixture of root, leafy and cereals intended for human consumption. The uranium uptake factors ( $[U]_{\text{plant d.w.}} / [U]_{\text{soil d.w.}}$ ) were in the order of  $10^{-3}$  to  $10^{-4}$ . Trends were difficult to observe in the

limited set of data but the following trend appears to be present for the Scottish vegetation for uranium: root vegetables > leafy vegetables > cereals. The higher values observed for the root vegetables were unlikely to be due to soil contamination as the vegetation was thoroughly washed and only the West Helmsdale potatoes have significantly greater concentration of U in the peel than the flesh. A sequential extraction procedure was carried out on the soil to assess how uranium was distributed through the following phases 1) exchangeable, 2) bound to carbonates, 3) bound to Fe-Mn oxides, 4) bound to organic matter and 5) residual. The results showed that very little of the uranium present were associated with the exchangeable phase which appears consistent with the low uptake factors obtained and that the uranium content was uniformly distributed throughout the non-exchangeable phases. This leads to the conclusion that changes in soil conditions (pH and Eh) could therefore increase the availability of uranium to plants.

Kohler *et al* (2000) studied the uptake of primordial radionuclides in tomatoes from soils contaminated by former uranium mining activities. The concentration of both soils is much higher than the average in the upper earth crust (30 Bq/kg  $^{238}\text{U}$ ). One of the soils had a lower uranium content (2700 Bq/kg) than the other (6200 Bq/kg) as the uranium content was significantly reduced due to its origin from an ore processing facility and their second soil originating from a former tailing dam. The vegetation was broken down into constituent parts such as roots, stalks and fruits. The transfer factors (TF:  $([\text{U}]_{\text{plant d.w.}} / [\text{U}]_{\text{soil d.w.}})$  for  $^{238}\text{U}$  showed no difference between the two soils whereas the transfer of  $^{238}\text{U}$  decreased from the roots over the stalk to the fruits. For tomato roots the transfer factors were  $0.063 \pm 0.002$ , tomato stalk were  $0.024 \pm 0.023$  and tomato fruits the transfer factors were  $0.0007 \pm 0.0006$ . The average transfer factor for  $^{238}\text{U}$  of 0.032 was similar to the value for uranium of 0.013 published by Sheppard *et al* (1989).

Linsalata *et al* (1989) analyzed  $^{238}\text{U}$  concentration in animal feed stuff grown on a farm in New York State. The CR  $([\text{U}]_{\text{plant d.w.}} / [\text{U}]_{\text{soil d.w.}})$  factors given for second cutting of alfalfa was  $16 \times 10^{-4}$ , and for unwashed and washed pasture grass were  $137 \times 10^{-4}$  and  $47 \times 10^{-4}$  respectively indicating the effect of soil contamination on grass. Ewers *et al* (2003) carried out an extensive review of the data of transfer factors for natural radionuclides to

vegetation for data pertaining to the UK and calculated geometric means for U uptake into a number of different vegetation groups. The geometric mean calculated from the relevant literature for uranium uptake into pasture, brassicas, root crops, potato tubers and cereal crops were  $60 \times 10^{-4}$ ,  $30 \times 10^{-4}$ ,  $30 \times 10^{-4}$ ,  $30 \times 10^{-4}$  and  $10 \times 10^{-4}$  respectively. Pietrzak-Flis and Suplinska (1995) compared U uptake in pasture grown in a tent to that grown outdoors and the mean CR values for outdoor and tent pasture  $45 \times 10^{-4}$  and  $27 \times 10^{-4}$  respectively. Moffet and Tellier (1977) published CR of  $15 \times 10^{-4}$  for uranium uptake into grass species growing on U mill tailings.

Jenkins et al (1990b) determined concentrations for uranium in British arable crops and grass and found them to be with the range of  $< 0.3 - 45 \text{ ng g}^{-1}$ . The concentrations for uranium in the different crop types increased in the order; maize = beet < potato flesh < carrots < beans < wheat < barley < onions < peas < potato peel < grass. Transfer factors ( $\text{TF} = [\text{U}]_{\text{plant d.w.}} (\text{ng g}^{-1}) / [\text{U}]_{\text{soil 0-30 cm d.w.}} (\text{ng g}^{-1})$ ) for uranium ranged from  $< 0.0001$  to  $0.0174$  with a mean TF value of  $0.0031 \pm 0.0044$ . On closer examination, it was found that the TF values on all crops except grass, potato peel and peas were generally  $< 0.001$ . The TF values for grass; potato peel and peas were between  $0.001$  and  $0.015$ .

Vera Tomé *et al* (2003) presented TF ( $U_{\text{plant activity d.w.}} (\text{Bq/kg}) / U_{\text{soil activity d.w.}} (\text{Bq/kg})$ ) for different natural radionuclides belonging to the uranium and thorium series for grass-pasture in an area where a disused uranium mine is located in a Mediterranean region. Statistical analysis of the TF values obtained showed that TF values corresponding to the two soils types (granite and alluvial) were statistically indistinguishable and the same was the case for the two study areas (region affected by mine and region unaffected by mine). The transfer factors for the two uranium isotopes  $^{238}\text{U}$  and  $^{234}\text{U}$  analyzed were also found to be statistically indistinguishable with mean values of  $0.067$  and  $0.072$  respectively. Ibrahim and Whicker (1992) obtained similar TF ( $U_{\text{plant mean activity}} (\text{g}^{-1}) / U_{\text{soil mean activity}} (\text{g}^{-1})$ ) for native plants growing on a uranium mine with transfer factors for  $^{238}\text{U}$  in the range of  $0.04$  to  $0.81$ . These results are compatible with those reported for uranium in grass by IUR (1994) of  $0.0023$  to  $0.23$ .

Lopatkina *et al* (1970) found that primitive plants (mosses, algae and microorganisms) tend to accumulate more uranium than higher plants, and plant debris (dead material) accumulates more uranium than living material in contact with uranium-enriched water. Coker and DiLabio (1979) found that *Sphagnum* moss growing on the surface of a uraniumiferous peat bog concentrated uranium to a level of  $6.5\mu\text{g g}^{-1}$  where the uranium concentration in the light brown fibrous peat below the moss contained 0.9 to  $1.6\mu\text{g g}^{-1}$  and the more humified peat lower in the profile contained up to  $1290\mu\text{g g}^{-1}$ .

Mahon and Mathewes (1983) analyzed vegetation growing in a region of high natural radioactivity, for accumulation of uranium. The majority of soil was upland till or upland till/lowland till (some bog) soil type. Four separate groups of vegetation type were analyzed and these are as follows: forbs and grasses, shrubs, deciduous trees, and coniferous trees. The species of forbs and grasses analyzed were species prevalent in the area: Pinegrass, Fireweed, Lupines (*Lupines arcticus*), and Asters (*Aster subspicatus* group). The shrubs species analyzed were: Grouseberry, raspberry (*Rubus idaeus* L.), gooseberry, and wild strawberry. The coniferous tree species were: scoulters willow and Mountain alder. The deciduous tree species sampled were: Lodgepole pine, alpine fir, Douglas fir and western Larch. Based on concentration factors, which determine the concentration of radionuclide in the vegetation relative to that in the soil, the only species, which showed any ability to accumulate uranium, were pine and aster. The aster species was picked due to its known ability to accumulate metal therefore it was expected to concentrate high levels of uranium. The range of concentration factors ( $[\text{U}]_{\text{plant d.w.}} / [\text{U}]_{\text{soil d.w.}}$ ) for all species was 0.002 – 0.018. These values were a factor of 10 lower than those obtained by van Netten and Moorley (1981) for uranium transfer from soil to barley for soils in the same region. Mahon and Mathewes (1983) stated that the data for the aster samples may have been misleading in that only two samples were collected, both from areas from widely differing uranium content (390 ppm and 9 ppm) as concentration factors values decrease as soil content of radionuclide increases.

Sheppard *et al* (1989) analyzed vegetation for uranium content in a number of different experiments: 1) outdoor lysimeter setting where different soil types were contaminated with natural uranium and a number of different plants species were grown, 2) uptake of uranium from soils contaminated by a former Ra processing facility, 3) a controlled lysimeter setting and 4) an uncontrolled home garden setting and found the overall geometric mean CR ( $[U]_{\text{plant d.w.}} / [U]_{\text{soil d.w.}}$ ) value for barley, blueberry, corn and potato to be 0.013. Morishima *et al* (1977) collected several field-grown vegetables and their soil substrates from three districts in Japan. The transfer factors ranges were as follows; spinach:  $26 - 190 \times 10^{-4}$ , Chinese cabbage:  $19 - 68 \times 10^{-4}$ , cabbage:  $13 - 21 \times 10^{-4}$ , potato:  $14 - 45 \times 10^{-4}$  and wheat (one site):  $12 \times 10^{-4}$ .

Sheppard and Evenden (1988) reviewed all soil to plant concentration factors subsequent to the review and showed that small-scale studies yielded CR values six-fold higher than the field studies and therefore it is imperative that studies be conducted under very realistic conditions with large volumes of soil and appropriately aged contamination.

Chemical composition of both the soil and plant is seen to play a significant role in the uptake of radionuclides by plant and a review of the current literature indicates that calcium and phosphorous are the two elements which have the greatest effect on the uptake of U by plants. A significant positive correlation ( $p < 0.001$ ) was found between the uranium and phosphate content and the uranium and calcium content in crops studied by Jenkins *et al* (1990b). Morishima *et al* (1977) also showed a positive correlation for uranium with both phosphorous and calcium in leaf vegetables. One of the main reasons for a correlation in the British soils may be due to the trace amounts of uranium present in phosphate fertilizers and the liberal application of fertilizer to British soils. Jenkins *et al* (1990b) found that the positive correlation for calcium and uranium reported by Morishima *et al* (1977) in leafy vegetables also holds true for cereals and roots crops. In general, they found that the plant materials with relatively high carbohydrate concentrations (e.g. grains and potato) contain relatively low levels of calcium and corresponding low levels of uranium. Hence storage organs and the edible portion of many arable crops such as tubers and grain will contain low concentrations of uranium

compared with the foliage and the skins of root crops. As calcium in a plant is usually associated with cell walls, it is reasonable to assume that uranium is also incorporated into the structures (Jenkins *et al* 1990b). Mengel and Kirkby (1979) found that phosphorous behaved in a similar manner to uranium and Morishma *et al* (1977) found that there was a linear relationship between uranium and phosphorous concentrations in leafy vegetables grown on farm fields ( $r = 0.75$ ). However, it is not clear if some of the plant uranium may have originated in phosphorous fertilizers. Prister and Prister (1970) found that plants high in potassium and calcium have the lowest concentration of uranium in their ash, which is in conflict with the above findings. The CR obtained by Yamamoto and Masuda (1974) for uranium uptake by Chinese cabbage from three soil types (volcanic, sandy and alluvial) that were artificially enhanced with uranium indicated that low CR values correspond with high CEC.

Sheppard and Evenden (1988) found that the influence of the soil concentrations of uranium on concentration ratios suggest a saturation-type mechanism and indicate that, for uranium, most plants conform to the accumulator strategy. As soil concentrations get progressively higher, the plants accumulate relatively less of the radionuclides (CR values decreased as the soil concentration increased). Sheppard *et al* (1983) and Yamamoto and Masuda (1974) also found a decreased CF with increased uranium concentration in soil. Tracy *et al* (1983) observed the same trend. Davies and Dollard (1988) also noted a decrease in CF with increase in soil uranium and suggested that uranium may behave as an essential element. They concluded that while evidence for a physiological role for uranium is not apparent (Petersen and Girling 1981) it is possible that selective uptake could occur through sites specific for other essential trace elements.

Soil contamination of certain crops (grass, peas and some grains) may contribute a significant proportion of the measured uranium. The difference in height, shape and density of a crop will affect the amount of soil contamination on the plants. Thus ground hugging crops such as peas will be more prone to soil contamination through rain-splash and resuspension than taller crops such as beans and grains (Jenkins *et al*, 1990b).

Plant species differ considerably in their uptake of radionuclides. Some species absorb such significant amounts that they are used as biological indicators in geochemical experiments. Dunn (1986) reported that the twigs (minus the leaves) of black spruce (*Picea mariana*) contained uranium concentrations  $> 100\text{mg/kg}$  ash weight in comparison with background levels of  $<1$  to  $60\text{ mg/kg}$ . This knowledge was used to locate a massive U biogeochemical anomaly in Canada (Dunn, 1986).

The concentrations of uranium and radionuclides in general may change with time and Dunn (1981) found that uranium accumulation progressed slowly as the twigs were actively growing and then decreased owing to translocation to other parts of the tree as the twigs became less active. Cannon (1952) also stated that through the growing season, the uranium content tends to increase in evergreens but decreases in most deciduous species.

Hoffman (1964) analyzed the leaves of several deciduous trees throughout the summer growing season and noted a consistent decrease in uranium content in the fall. However no explanation was made as to whether this decrease could be attributed to dilution effect or translocation.

Effects due to excess amounts of uranium in plants include abnormal numbers of chromosomes, unusually shaped fruits, sterile apetalous forms and stalked leaf rosette (Alloway, 1990).

## 1.6 Radium in the Terrestrial Ecosystem

Radium occurs in the environment as radioactive nuclides of which  $^{224}\text{Ra}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  are the common products of the uranium and thorium decay chains with  $^{226}\text{Ra}$  the most stable of these nuclides. Radium, which is chemically similar to barium, has only one oxidation state, +2. Radium hydroxide,  $\text{Ra}(\text{OH})_2$  is the most soluble of all the alkaline earth hydroxide including the hydroxides of thorium and actinium. Radium forms a water-soluble chloride and bicarbonate but it co-precipitates with barium as a sulphate or a chromate.

### 1.6.1 Chemistry of Radium in Soil

Radium is a member of the alkaline earth group and oxidized members of this group are always present in the 2+ oxidation state. Radium is found to be present as  $\text{Ra}^{2+}$  over the normal soil pH range of 4 to 8. It behaves similarly in soils to calcium, strontium, uranium, barium and lead (Bondetti and Sweeton, 1977 and Schulz, 1965). Tashayev *et al* (1977) found that mainly barium rather than calcium controlled the behaviour of  $^{226}\text{Ra}$  in the soil under investigation. They found a direct positive correlation ( $r = 0.5-0.9$ ) between  $^{226}\text{Ra}$  and barium and none between  $^{226}\text{Ra}$  and calcium or barium and calcium. They also found that most of the  $^{226}\text{Ra}$  was strongly bound in the soil and its mobile forms could be arranged in the following sequence: acid-soluble > exchangeable > water > soluble. Other findings were that a high content of calcium in the soil reduced the rate of uptake of  $^{226}\text{Ra}$  by the plant while the behaviour of  $^{226}\text{Ra}$  and barium were virtually identical in the soil-plant system. Menzel (1965) suggested that radium is more strongly adsorbed in soil than calcium. Tashayev *et al* (1978) studied the speciation of  $^{226}\text{Ra}$  in soils with high concentrations of the isotope and found that  $^{226}\text{Ra}$  is firmly fixed within the soil, and that the ratio of the water soluble and exchangeable proportions varies significantly with depth. Another conclusion was that organic acids present in the soil increase the mobility of the isotope but no explanation for this was given. Mays and Mortvedt (1986) reported that the concentration of  $^{226}\text{Ra}$  in the surface 15-cm layer of soil increased from 35 to 73  $\text{Bq Kg}^{-1}$  about 1 year after application of phosphogypsum at a rate of  $112 \text{ t ha}^{-1}$  and the levels in the subsurface layers were not affected indicating that the radium was retained in some manner in the surface layer. Guimond and Windham (1975) reported greater precipitation of  $^{226}\text{Ra}$  from effluents of wet process phosphorous manufacture when the pH was increased to levels > 6.0. Havlik *et al* (1968) listed water temperature, rate of flow, size of radioactive particles and type of medium as the most important factors in the liberation of radium from rocks and water. Salts of radium such as sulphate are transported in water and move attached to clay particles or as radiocolloids.

Mortvedt (1986) reported that soil texture had a significant effect on the movement of radium in soil where divalent cations ( $\text{Ra}^{2+}$ ) were sorbed by soil clay through exchange mechanisms and relative rates of sorption generally increased as clay content in the soil increased. Since mobility in soils is inversely related to sorption, downward movement of radium would be lower in finely textured soils i.e. clay soils.

Hansen and Huntington (1969) measured the distribution of thorium and radium in the soils of the High Sierra, California and found that radium was more irregularly distributed than thorium, and these patterns resulted not only from radium interaction with soils but also the effects of soil chemistry of  $^{238}\text{U}$ ,  $^{234}\text{U}$ ,  $^{230}\text{Th}$ , and  $^{226}\text{Ra}$ . Its distribution was explained in terms of uranium retention by organic matter, mobilization of  $^{230}\text{Th}$ , plant absorption of radium, and time. Increased concentration of radium was found in the uppermost horizons and this may be the result of uranium retention and subsequent accumulation of  $^{226}\text{Ra}$  by decay, but there exists the possibility that the trees on these soils can absorb radium, enriching the surface soil with radium from their litter.

Greeman *et al* (1999) studied the geochemistry of radium in soils of the Eastern United States and found that uranium, thorium and radium are enriched in soils relative to underlying parent rocks, especially in carbonate soils where a 10-fold enrichment is common. This enrichment in radium was found to be a major factor in causing Rn in soil gas of carbonate-derived soils to be 3 to 10 times higher than soils over most shale, sandstone and igneous rocks in Pennsylvania. Radium was found to be strongly enriched in the exchangeable fraction of soils in which it is largely unsupported by uranium especially in the organic rich "A" horizon. This enrichment in the "A" horizon was found to be largely caused by cycling through vegetation, which preferentially incorporates radium relative to uranium, followed by strong retention of radium in the resulting humic material. A high proportion of radium occurred as exchangeable ions and in organic matter in soils of this region, in contrast to uranium and thorium, which are most enriched in iron-oxides. Radium is evidently bound in organic matter. Deeper in the soil profile, decay loss of unsupported radium in the exchangeable-organic fraction appeared to be

replaced by incorporation of radium leached from minerals, and perhaps in part by radium uptake and fixation in roots.

Titaeva (1967) reported that uranium is bound to the humic and fulvic acids that are soluble in alkali solutions, whereas radium is associated with the insoluble residue. Under oxidizing conditions these elements appear to be retained by ion exchange, but in the presence of high concentrations of calcium in the water, the radium in peat and in the insoluble residue becomes nearly unexchangeable. Nathwani and Phillips (1979) concluded that in the presence of elevated  $\text{Ca}^{2+}$  radium absorption by soils was severely retarded. Rusanova (1962) indicated that the soil horizons enriched in organic matter accumulate radium from the groundwater and only a small proportion of the radium is found in the salt or hydrochloric extracts. The study showed that rapid desorption of radium took place at pH 3, and as the pH increased desorption decreased with a slight increase at pH 8 with the conclusion that soils with a higher content of adsorbed bases have a greater ability to fix minute amount of radium. This conclusion was also supported by Verkovskaja *et al* (1967) who stated the higher the content of organic substances and adsorbed bases in a soil horizon the higher the radium content.

Mc Auley and Marsh (1992) measured radium activities in the Republic of Ireland and found activities in the range 6 – 292 Bq kg<sup>-1</sup> (dry weight), with a national average of 46.1 Bq kg<sup>-1</sup>. Regions of high levels tended to cluster and these clusters are mainly situated in the west and northwest of the country with a smaller cluster in the northeast. The majority of the country had levels of less than 65 Bq kg<sup>-1</sup>. The authors concluded that elevated <sup>226</sup>Ra correspond to regions of either granite or limestone rock types. The only exception is in the northwest where the predominant rock type is upper avonian shale bordering on limestone. The uranium series disequilibrium ratio (ratio of <sup>226</sup>Ra to <sup>238</sup>U in soil) was measured. The results given show a degree of uranium series disequilibrium in soil and are in the range 0.11 to 21, with most of the country lying in the interval 0.2 – 2.0. The authors noted that the strongest disequilibrium existed in areas where <sup>226</sup>Ra activities are highest.

## 1.6.2 Uptake of Radium by plants

For naturally occurring  $^{226}\text{Ra}$  it seems likely that uptake by roots is influenced by soil type, as well as factors such as soil pH, content of other alkaline earth in the soil, plant species, and chemical form of  $^{226}\text{Ra}$  in the soil. Mortvedt (1986) also suggested that uptake of radium was competitively depressed by  $\text{Ca}^{2+}$ . Menzel (1965) concluded that when radium was added to soil in water-soluble form it was found to be concentrated to the same degree in soil and vegetation. Numerous studies have been performed to investigate the behaviour of  $^{226}\text{Ra}$  in the soil-plant system and its incorporation into plant tissues from nutrient solutions. However, experiments quantifying uptake from nutrient solutions are less useful for the purpose of this study, as retention of  $^{226}\text{Ra}$  by soil particles and its interaction with soil components are important elements in determining plant uptake.

In general the conclusion obtained from the literature is that the concentration of radium is found to be higher in leaves and stems than in roots and lowest in grains. Kirchmann et al (1980) reported that  $^{226}\text{Ra}$  concentrations were 5 to 16 times higher in winter wheat straw than grain. They also found that sugar beet foliage contained more  $^{226}\text{Ra}$  than the root tissues. Uptake of  $^{226}\text{Ra}$  was somewhat higher in wheat (*Triticum spp.*) straw and swiss chard (*Beta vulgaris* L.) grown on soil pH 7.2 but not in wheat grain or fescue (*Festuca arundinacea* L.) at the same soil pH levels (Mortvedt, 1986). Lima and Penna-Franca (1988) studied uptake of  $^{226}\text{Ra}$  by vegetables from natural and artificially contaminated soil in a highly radioactive region known as Poços de Caldras in Brazil. The plant species analyzed was kale (leaf and stem), carrot (leaf and root) and brown bean (grain, husk and root). The distribution of  $^{226}\text{Ra}$  among the vegetal structures followed the same pattern in all plants both from natural and artificially contaminated radium. Foliar parts of kale and carrot presented higher  $^{226}\text{Ra}$  concentration than their stems and roots respectively. These results although confirmed by Sheppard (1980a), contradict the idea presented by Gunn *et al* (1970) and D'Souza *et al* (1970) which postulates that  $^{226}\text{Ra}$  is discriminated against in the passage towards the plant apex, and has no metabolic role in vegetal systems. Only brown bean shows a radium acropetal

gradient (travels towards the apex). Tracy *et al* (1983) assessed the concentration levels of  $^{226}\text{Ra}$  in regions of contamination and also regions with natural background radiation and assessed the transfer of  $^{226}\text{Ra}$  to garden produce in the area. The  $^{226}\text{Ra}$  concentration was highest in leafy vegetables followed by root and stem vegetables. The concentration in fruit from contaminated gardens showed little difference from those in control gardens. The exceptions were raspberries, strawberries and perhaps apples, which are all fruit from perennial plants that can accumulate radium from year to year.  $^{238}\text{U}$  and  $^{210}\text{Pb}$  were also analyzed during the course of this study and the results show that uptake was strongest for  $^{226}\text{Ra}$  followed by  $^{210}\text{Pb}$  and then  $^{238}\text{U}$ . The mean concentration factors ( $[\text{Ra}]_{\text{plant d.w.}} / [\text{Ra}]_{\text{soil d.w.}}$ ) were  $1.1 \times 10^{-3}$ ,  $0.37 \times 10^{-3}$  and  $0.075 \times 10^{-3}$  respectively. Sam and Eriksson (1995) studied the uptake of  $^{226}\text{Ra}$  by cultivated crop plants from regions of natural radioactivity in Western Sudan. Soil/plant CR's ( $[\text{Ra}]_{\text{plant activity d.w. (Bq/kg)}} / [\text{Ra}]_{\text{soil exch. activity d.w. (Bq/kg)}}$ ) were related to the exchangeable radium in the soil and showed ranges of 0.09 – 3.31 and 0.01 – 0.34 for vegetative and edible crops parts respectively. Among the species analyzed the vegetative plant purslane (*Portulaca oleracea*) displays a higher affinity for radium uptake relative to other species and concluded that there is a tendency for a lower transfer of radium to seeds and fruits than to the vegetative parts of plants. Nevertheless the orka plant (*Abelmoschus culentus*) shows a comparatively high capacity for accumulating radium in the fruit with a CR of 0.34. The authors suggested the use of purslane and orka plants as indicator plants in regions with climatic conditions similar to Sudan. Makrose *et al* (1993) concluded that uptake of  $^{226}\text{Ra}$  by tomato plants portions decreased in the following order: leaves > root > stem > fruit. The order remained the same whether TR was evaluated on total radium or on available radium in soil. Popova *et al* (1964) and Mistry *et al* (1965) have found similar behaviour. Kohler *et al* (2000) studied the uptake of primordial radionuclides in tomatoes from soils contaminated by former uranium mining activities and found generally the same trend as Makrose *et al* (1993). The transfer of  $^{226}\text{Ra}$  as with the other natural nuclides studied decreased from the roots over the stalk to the fruits. For tomato roots, stalk and fruits the transfer factor ( $[\text{Ra}]_{\text{plant activity d.w. (Bq/kg)}} / [\text{Ra}]_{\text{soil activity d.w. (Bq/kg)}}$ ) were  $0.061 \pm 0.009$ ,  $0.03 \pm 0.01$  and  $0.0021 \pm 0.0017$  respectively. The average value for all plant parts was 0.035, which is in accordance with the value of 0.022 published by Sheppard *et al* (1989).

Beaza *et al* (1996) studied the bioavailability of radionuclides ( $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{228}\text{Th}$ ) in soil and *Cistus ladanifer* (trees) in a Mediterranean ecosystem. The soil was analysed for radioactive concentrations in the total and available fraction at different depths between 0 and 19 cm. The vegetation was broken down into fruit, young leaf, mature leaf, trunk, root and litter fractions. There was no significant variation of the radionuclides with depth to 19 cm in either total or available fractions. The authors found a secular equilibrium between  $^{228}\text{Ra}$  and  $^{228}\text{Th}$  in the total fraction of soil which is broken in favour of  $^{228}\text{Ra}$  in the available fraction of the soil, and, as a consequence in the different plant fractions. Of the plant fractions analyzed, the mature leaves and trunk-roots had the highest levels for the three radionuclides. The fruit and young leaves had the lowest concentrations. The author obtained a  $^{226}\text{Ra}/^{228}\text{Ra}$  discrimination factor between 2.4 and 3.8 for all the fractions analyzed. This indicates that the plant has a preferential assimilation of  $^{226}\text{Ra}$  over  $^{228}\text{Ra}$ .

Arkhipov *et al* (1984) concluded that barley roots was found to accumulate tens to hundreds of times more than the above ground parts with the straw accumulating ten times more than that of the grain from a range of soils artificially contaminated with uranyl nitrate. The authors suggested that the high accumulation coefficient (i.e.  $[\text{Ra}]_{\text{plant d.w.}} / [\text{Ra}]_{\text{soil d.w.}}$ ) associated with the roots is apparently due to the fact that when plants are growing in containers and the root system spreads out in a small volume of soil which is uniformly contaminated with naturally occurring radionuclides in soluble form conditions arise that will promote increased absorption by the roots. Results indicated that  $^{238}\text{U}$  and  $^{226}\text{Ra}$  accumulate in plants in large quantities while  $^{232}\text{Th}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  accumulate in smaller quantities.

The mobility of  $^{226}\text{Ra}$  within plant tissues appears to be high during transport from root to shoot, but low after deposition within leaf tissues. This results in acropetal concentration and a potential for enhanced uptake by animals from forage and leafy vegetables (McDowell-Boyer *et al*, 1980).

Radhakrishna *et al* (1996) calculated soil to plant transfer factors for  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  based on the geometric mean values of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in soil and vegetables. The transfer factors for  $^{226}\text{Ra}$  in vegetables (not specified) and rice were  $4 \cdot 10^{-3}$  and  $7 \cdot 10^{-3}$  respectively. The authors suggested that the relatively higher values quoted for rice may have been due to the longer growing period of the paddy. The transfer factors for  $^{228}\text{Ra}$  in vegetables and rice were 0.017 and 0.028 respectively. The authors concluded that the transfer coefficients for  $^{226}\text{Ra}$  were an order of magnitude lower than that of  $^{228}\text{Ra}$ , which indicated preferential transfer and uptake of  $^{228}\text{Ra}$  in vegetation sampled but no possible explanation was given for this. McDowell-Boyer *et al* (1980) in their food-chain transport parameters review gave a  $^{226}\text{Ra}$  concentration factor (CR) of  $8 \times 10^{-3}$  for vegetables and Watson *et al* (1984) determined a TR for  $^{226}\text{Ra}$  for fruits and vegetables of  $3 \times 10^{-3}$  and  $1.2 \times 10^{-2}$  respectively.

Mahon and Mathewes (1983) analyzed the concentration of  $^{226}\text{Ra}$  in a range of sample species. The four sample groups analyzed were: forbs and grasses, shrubs, deciduous trees and coniferous trees. The forbs and grasses analyzed were: pine grass, fireweed, lupines and asters. The shrubs analyzed were: grouseberry, raspberry, gooseberry, wild strawberry and Oregon boxwood. The deciduous trees sampled were: scoulers willow and mountain alder. The coniferous trees sampled were lodgepole pine, alpine fir, Douglas fir and western larch. Grouseberry, larch, fireweed, and grass accumulated  $^{226}\text{Ra}$  from the soil to a greater extent than any of other species. The range of concentration factors ( $\text{Ra}_{\text{plant activity d.w.}} / \text{Ra}_{\text{soil activity d.w.}}$ ) for all species was 0.01 – 0.24

Concentration factor ( $\text{Ra}_{\text{plant activity fresh.w.}} / \text{Ra}_{\text{soil activity d.w.}}$ ) results from the study of Bettencourt *et al* (1988) for  $^{226}\text{Ra}$  for cabbage leaves ranged from 0.007 to 0.11. Concentration factors for herbage (grass and watercress - relative to fresh weight of vegetable) under the same conditions were higher and ranged from 0.09 - 0.5. The lowest value given for the cabbage corresponds to cabbage grown in the zones with the highest  $^{226}\text{Ra}$  concentrations in soils. An explanation given by the authors is that high barium content is found in the area. However, the authors carried out calcium and barium analysis and no significant correlation was found between the CF's and either element.

Ewers *et al* (2003) carried out an extensive review of the data of transfer factors for natural radionuclides to vegetation for data pertaining to the UK and calculated geometric means for Ra uptake into a number of different vegetation groups. The geometric mean calculated from the relevant literature for radium uptake into pasture, salad vegetable, brassicas, root crops, potato tubers and cereal crops were  $300 \times 10^{-4}$ ,  $200 \times 10^{-4}$ ,  $600 \times 10^{-4}$ ,  $300 \times 10^{-4}$ ,  $30 \times 10^{-4}$  and  $80 \times 10^{-4}$  respectively. IAEA Technical Report 364 recommends a soil-to plant transfer factor for Ra in pasture of  $8 \times 10^{-2}$  (IUR, 1994).

Amaral *et al* (1988) studied the transfer of  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$  to forage and milk in a region of high natural radioactivity in Brazil known as the Poços de Caldas plateau. Soil to plant transfer factors for  $^{226}\text{Ra}$  was based on a geometric mean of the values for soil and unwashed forage. The value given for  $^{226}\text{Ra}$  was  $7 \times 10^{-2}$ , which is in complete agreement with the values given by McDowell-Boyer *et al* (1980) and Ng (1982). Transfer factors for  $^{210}\text{Pb}$  based on a geometric mean value were  $3 \times 10^{-1}$  which is one order of magnitude higher than the  $^{226}\text{Ra}$  value even though the soil nuclide values are within the same range. The authors indicate that the  $^{210}\text{Pb}$  deposition, due to the  $^{222}\text{Rn}$  and further foliar absorption is an important contributor to the forage lead uptake.

Vera Tomé *et al* (2003) studied the transfer factors for natural radionuclides and stable elements in a Mediterranean area. The mean value obtained by the authors for  $^{226}\text{Ra}$  was 0.17, which is similar to the mean value by Linsalata *et al* (1989) for radium in grass of 0.08 with a range between 0.016 and 0.4. Lower mean values for  $^{226}\text{Ra}$  in vegetables and rice of approximately 0.001 in both cases have been found (Radhakrishna *et al*, 1996). However, transfer factors for radium in native plants growing in a uranium mine are in the range 0.07 to 0.15 (Ibrahim and Whicker, 1992). The uptake of  $^{226}\text{Ra}$  was higher than that for the isotopes of uranium and thorium and this is in agreement with the work of Linsalata *et al* (1989) who state that the TF values for elements in oxidation 2+ (Ra) are always greater than for those elements in 4+ (Th) oxidation state.

Chemical composition of the soil and plant are seen to play a significant role in the uptake of radionuclides by plants and a review of the current literature indicates that calcium has the greatest effect on the uptake of radium. Some authors have indicated that barium has a greater effect on the availability of radium to plants but a definitive conclusion on this cannot be made due to the lack of barium analysis carried out in the studies. Arkhipov *et al* (1984) found that as the content of humus and exchangeable calcium in the soil increases and as the soil solution reaction approached neutral, the accumulation coefficient for  $^{226}\text{Ra}$  decreased. The explanation given was that soils that are rich in humus and close to neutral pH, with a predominance of calcium ions in their soil/absorption complex, have the highest exchange capacity. In these conditions, radionuclides are absorbed to a greater degree by the solid phase of the soil and the plant takes up a smaller quantity of radionuclide. Makrose *et al* (1993) data showed that addition of calcium to soil appears to have inhibited the availability of radium, which is akin to isotopic dilution in view of the similar chemical characteristics of radium and calcium. Blanco Rodríguez *et al* (2002) carried out multivariate regression analysis on transfer factors versus stable elements concentrations in plants and in substrate and concluded that the uptake of radium depends mainly on the concentration of iron in the plant and of phosphorous and alkaline-earths in the substrate. Rusanova (1962) concluded that radium behaves similarly to strontium and suggested that since large quantities of available calcium in the soil prevents strontium from entering the plant it will also reduce radium uptake. Kirchmann *et al* (1966) found no relationship between calcium and radium ions but did note that a reduction in radium uptake was strongly dependent on pH of the solution, which is in turn influenced by the calcium concentration. Taskayev *et al* (1977) concluded that the behaviour of radium in the soil-plant system is more related to barium than calcium. This is supported by Gunn and Mistry (1970) who showed that complexing agent increased the translocation of radium in plants but did not affect the mobility of calcium and strontium. Sheppard (1980a) suggested that the resemblance between the behaviours of radium and barium may be due to the sizes of their ionic radii being closer to that of either calcium or strontium. Vasconcellos *et al* (1987) also found that the  $^{226}\text{Ra}$  plant uptake was also not well correlated to exchangeable calcium in soil. However, the highest average calcium and  $^{226}\text{Ra}$  were found in the same vegetables:

brown beans and carrots. Bettencourt *et al* (1988) found that the lowest value obtained for cabbage leaves corresponds to cabbage grown in the zones with the highest  $^{226}\text{Ra}$  concentrations in soils. An explanation given by the authors is that high barium content is found in the area, however, the authors carried out calcium and barium analysis and no significant correlation was found between the CF's and either element.

The effect of soil radium concentration on uptake by plants is unclear and conflicting observations have been reported in the literature possibly due to the differences in chemical composition of the soil. Very different results have been shown as to whether total radium or available radium is a better indicator of radium uptake by plants. Blanco Rodríguez *et al* (2002) studied the linearity in soil to plant transfer factors (i.e. a linear relationship would imply that if soil concentration increases then uptake into plant should increase) for  $^{226}\text{Ra}$  and found that the linearity assumption for different isotopes of radium can be considered valid over a concentration range of two orders of magnitude. Otherwise, there is a nonlinear variation in the TF values with changing substrate concentration. This variation was found to be due to the influence of other variables, such as the chemical form and the chemical environment of the soils reflected in the concentration of stable elements in the substrate. Makrose *et al* (1993) found a positive correlation between radium in soils and plants when TRs were based on available radium in soil. However, Lima *et al* (1988) concluded that either the method used by the authors for determining plant availability of radium was not suitable or that other physico-chemical parameters have more influence on  $^{226}\text{Ra}$  uptake than the readily available fraction of radium.

Kohler *et al* (2000) concluded that soil concentration had no effect on transfer factors for  $^{226}\text{Ra}$  as the two soils studied have very different concentrations of radium present (3800 Bq/kg and 13800 Bq/kg) and there was no significant difference between the transfer factors from both soils. Vasconcellos *et al* (1987) studied uptake of  $^{226}\text{Ra}$  by vegetables from soils located in regions of high natural radioactivity in Brazil. Plant uptake of  $^{226}\text{Ra}$  was not well correlated with levels of  $^{226}\text{Ra}$  in soil either the total or exchangeable forms.

The authors concluded that the results reported in their paper does not support the use of radium CRs based on the 'exchangeable' fraction in soils as opposed to the traditional CRs which are related to the total radium in soil. They found that the variability in CRs for each vegetable studied is about the same using either method of calculation. Madruga *et al* (2001) suggested that the nonlinear functions describing the plant/tailings concentration ratios could possibly indicate that radium is behaving as an 'accumulator' element in plant uptake. The term 'accumulator' does not infer that the elements are accumulated to any great extent, but it infers that the uptake response of plants to soil activity is a non-linear function increasing to single or multiple saturation values.

Bettencourt *et al* (1988) concluded that radium uptake appears to be more important for plants grown in contaminated soils than for those grown in zones of natural radioactivity, possibly due to varying physico-chemical form of radium and its availability in soil. They found that radium uptake from contaminated soils depends on its concentration in the substrate,  $C_s$ , in a manner expressed by the following equation. The concentration in the plant  $C_p$  can be expressed as a function of  $C_s$  as follows:

$$C_p = a C_s^b$$

the parameters  $a$  and  $b$  are dependent on the plant. Makrose *et al* (1993) however concluded that the order of available radium to plants was as follows: soil > tailings > processed uranium ore and concluded that transfer of radium to plant from tailings is far lower compared to that from normal soil.

## 1.7 Thorium in the Terrestrial Environment

Of the six thorium radioisotopes found in nature  $^{232}\text{Th}$  is of greatest importance, the other radioisotopes being relative minor components of the three decay chains –  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{235}\text{U}$ . Thorium is only found in the tetravalent state in nature. Thorium forms insoluble hydroxides, fluorides and phosphates, while soluble thorium compounds include nitrates, chlorides and sulphates. Thorium also forms metal complexes with citric acid, oxalic acid

and substances such as acetyl acetone, which render it very mobile. In a similar manner, thorium can be leached from soils and soil-clay fractions by various solutions.

### 1.7.1 Chemistry of Thorium in Soil

Thorium has generally been considered to be a lithophilic element of low geochemical mobility. Quadivalent thorium is adsorbed tenaciously on cation exchange resins and is one of the last elements to elute when resin columns are leached with 3N HCl. However, it will form complexes with many substances such as oxalic acid, citric acid and acetyl acetone forming metal complexes that can easily be eluted from exchange resins (Hansen and Huntington, 1969). Schulz (1965) studied the soil chemistry of radionuclides and found thorium behaves similarly to rare earth elements in that it is sorbed by soil in a very immobile manner. The dominant species of thorium in soils is cationic, specifically  $\text{Th}^{4+}$  and  $\text{Th}(\text{OH})_2^{2+}$ . Since soils have a net negative charge, these species are strongly sorbed. The retention of these species is greater in fine-textured soils with their high density of negatively charged sorption sites. Kiss *et al* (1988) reported that  $^{208}\text{Tl}$  (daughter of  $^{232}\text{Th}$ ) activities in soils under native grassland in Saskatchewan decreased as the clay content of the surface soils decreased. As thorium is quite immobile in soils any mechanism that increases their mobility becomes important in root uptake. Such mechanisms include the formation of complexes and association with colloids (Sheppard and Evenden, 1988). Accumulations of thorium are often associated with a gleyed horizon, suggesting that thorium accumulates under reducing conditions. Koczy (1963) stated that thorium forms radiocolloids which attach to other particles and are transported in this manner.

Hansen and Huntington (1969) investigated the impact of organic matter on the movement of thorium in soils and examination of  $^{232}\text{Th}$  in the soil profiles supported the idea of thorium movement under the influence of soil organic matter. They found that thorium appeared to have complexed with organic matter and leached downwards in the soil. This process is similar to what occurs to iron in organic soils during podzolation in

which iron is complexed with organic substances and transported downward in solutions, precipitating in the illuvial horizon. However, the authors reported that thorium distribution in the soil profile is apparently less affected by change in pH than is the iron distribution. They reported distinct thorium accumulations in soil horizons immediately below layers with high organic matter content. Bondietti (1974) used a calcium citrate solution to desorb thorium from soil and showed that this solution desorbed 30% of thorium from both montmorillonite and kaolinite but only desorbed 1% from a calcium humate material therefore highlighting the strength of the interaction between thorium and humic material. The relationship between humic material and thorium is countered by Tyuryukanova and Kalugina (1971) who found that soils high in humic acids (peats and mountain podosols) had low thorium levels relative to alluvial soils and stated that the low thorium content in peaty soils is apparently due to low biogenesis and weak assimilations in bog plants. Hansen and Stout (1968) showed that thorium leached from organic layers reprecipitate in zones containing less organic matter and Barnov *et al* (1964) showed that soils high in organic matter have the lowest Th/Ra ratio. Hansen and Stout (1968) reported that upper horizons of upland soils have higher concentrations of uranium than thorium, while thorium concentrations were higher in the “B” horizons. Alluvial soils tended to have higher and more evenly distributed concentrations of thorium, indicating its greater mobility as organic complexes. Sheppard and Evenden (1988) stated that the role of organic matter on the mobility of thorium has two distinct aspects. Organic soils are effective at retaining thorium in the soils whereas in contrast, organic complexes and colloids increase the mobility of thorium in mineral soils.

Rancon (1973) identified four types of soil-thorium adsorption reactions 1)  $\text{Th}(\text{OH})_4$  precipitation caused by calcareous soil buffering, 2) adsorption from dilute solutions onto clay soils, 3) adsorption to organic components under acidic conditions, and 4) low adsorption under alkaline conditions due to the dissolution of humic acids.

Barnov *et al* (1964) concluded that thorium transport occurs largely in the colloidal state and Verkovskaja *et al* (1967) concluded that plant cycling of thorium was not as important as it was for radium in the movement of the element in the soil.

Jenkins *et al* (1990a) studied the uranium and thorium levels in British soil and found that the concentration of thorium in arable and grassland soil ranged from 2.5 to 14.0  $\mu\text{g g}^{-1}$  and 3.1 to 13.0  $\mu\text{g g}^{-1}$  respectively. The highest thorium levels were reported in calcareous pelosol soil type for both arable and grassland studies. Lowest levels were associated with rendzina and brown sands where the parent material chalk and sandstone respectively are both known to contain relatively low levels of thorium. Th/U ratio from this study indicated preferential loss of uranium from arable soils either through leaching and /or plant uptake but only total uranium and thorium levels were studied and this is likely to differ greatly from that proportion of these elements available to plants. The authors found that the combined clay and silt content of the grassland soils showed a significant positive correlation with thorium concentrations. Thorium concentrations were associated with igneous outcrops and shale deposits.

### 1.7.2 Uptake of Thorium by plants

Thorium is generally considered to be immobile in soils and to be adsorbed in a very tenacious manner therefore is generally not taken up to any great degree by plants.

Squire (1963) compared  $^{226}\text{Ra}$  and  $^{230}\text{Th}$  uptake in water culture experiments for maize shoots and after 5 weeks in solution only 0.001% of  $^{230}\text{Th}$  was taken up in comparison with 3% of  $^{226}\text{Ra}$  (Osburn, 1965). However, as noted previously uptake factors from nutrient solutions do not give a clear indication of vegetation uptake under real conditions due to the interaction of the radionuclides with soil particles.

Sheppard and Evenden (1988) conclude that the mobility of Th is restricted in plants largely because of adsorption on cell wall materials. Therefore, concentrations are often higher in tissues found lower on the plant and highest on the root surfaces. For this reason root crops usually have higher CR values than cereal grain crops. Nicholson *et al* (1990) noted a trend amongst the limited data set available where thorium uptake factors decreased in the following order: root vegetables > leafy vegetables > cereals. They concluded that the higher uptake in the root vegetables was unlikely to be soil

contamination as vegetation was thoroughly washed. Studies of uranium and thorium speciation in soil were carried out by Nicholson *et al* (1990) and results indicated that the majority of thorium recovered by the sequential extraction resided in the residual phase. However, the authors indicated that thorium recoveries from the extraction procedure were quite low, and one possible reason given for this was incomplete dissolution of the residual phase. The transfer factors for the Scottish vegetation (potato peel and flesh, kale, sward, carrot, turnip and oats) were of the order  $10^{-3}$  to  $10^{-4}$ . They also stated that most of the thorium accumulates in aged tissues such as bark, branches and wood, and the least is observed in annual organs such as the leaves.

Arkhipov *et al* (1984) found that barley roots were found to accumulate tens to hundreds of times more than the above ground parts with barley straw accumulating ten times more than that of barley grain and the authors suggested that the accumulation by roots was due to the spreading out of plant roots in containers of soil uniformly contaminated with thorium. Arkhipov *et al* (1984) reported coefficients of accumulation (i.e.  $[\text{Th}]_{\text{plant d.w.}} / [\text{Th}]_{\text{soil d.w.}}$ ) of  $^{232}\text{Th}$  by the above ground mass of barley under experimental greenhouse conditions of the order of  $n \times 10^{-3}$ . Sheppard and Evenden (1988) carried out a review on all the available data on plant/soil concentration factors available to them and found that the only significant difference among soils or plant types was that annual plants showed a higher CR values than trees. However, this may largely be due to differences in soil concentrations among the studies.

Jenkins *et al* (1990b) studied the uranium and thorium concentrations in British arable crops and grass. The transfer factors ( $\text{TF} = [\text{Th}]_{\text{plant d.w.}} (\text{ng g}^{-1}) / [\text{Th}]_{\text{soil 0-30 cm d.w.}} (\text{ng g}^{-1})$ ) observed for thorium for all vegetation (potato flesh and peel, carrot, onion, beet bean, pea, wheat, barley, maize and grass) ranged from  $<0.00005$  to  $0.0152$  with a mean value of  $0.0019 \pm 0.0034$ . Transfer factors for all crops except grass, potato peel and peas were generally lower than  $<0.001$  with the transfer factors for the other three between  $0.001$  and  $0.015$ . Statistical analysis showed a preferential uptake of uranium on comparison to thorium assuming similar fractional availabilities of both in the soil. The authors highlight the fact the peas used were shelled in the field by machinery which possibly

lead to soil contamination and the possible explanation for the transfer factor for peas ( $38.2 \times 10^{-4}$ ) being higher than that found by Nicholson *et al* (1990) where the peas were hand-shelled (non detectable levels).

Ibrahim and Whicker (1988) assessed the levels of thorium uptake by plants growing at a uranium production site. For  $^{230}\text{Th}$ , CR values ranged from 0.01 at the reclamation area to 2.9 for plants growing at the edge of the tailings impoundment. Vegetation uptake of  $^{238}\text{U}$  was comparable to  $^{230}\text{Th}$  ( $^{238}\text{U}$  daughter) at all sites except at the tailings impoundment. The authors suggested that several factors may have led to the exceptionally high CR values observed for  $^{230}\text{Th}$  in plant growing at the edge of the tailing impoundment. Acidic and wet conditions at this site may have enhanced the solubility and availability of  $^{230}\text{Th}$  for plant uptake. Higher uptake was also noted for  $^{230}\text{Th}$  in beans grown in spiked nutrient solution than for plants grown in soil due to the lack of exchange sites. Additionally foliar deposition of pond water spray contained elevated  $^{230}\text{Th}$  concentration and subsequent foliar absorption may be another important uptake mechanism at this location. The concentration ratio for  $^{238}\text{U}$  and  $^{230}\text{Th}$  in plants growing on exposed tailings were greater than those reported for  $^{226}\text{Ra}$  and  $^{210}\text{Po}$  at this location. The authors postulated that the higher concentration of sulphate in the tailings impoundment may have promoted the precipitation of  $^{210}\text{Po}$  and  $^{226}\text{Ra}$  as sulphate and that thorium and uranium may have formed complexes with  $\text{Cl}^-$  and  $\text{SO}_4^-$  in tailings and prevented the formation of insoluble hydroxide species. The authors concluded that exceptionally high CR values for  $^{230}\text{Th}$  in vegetation growing at the tailings edge reported in their study demonstrates that, under certain conditions, above ground vegetation can accumulate thorium to a much greater extent than previously reported. Comparisons made between the CR ratio of  $^{232}\text{Th}$  and  $^{228}\text{Th}$  in plant from all locations around the uranium production site indicated higher assimilation for  $^{228}\text{Th}$  by a factor of 3-7 relative to that of  $^{232}\text{Th}$ . A possible reason given for this may be due to the in growth of  $^{228}\text{Th}$  from  $^{228}\text{Ra}$  and/or  $^{228}\text{Ac}$ , which are preferentially taken up by plants in addition to the direct uptake of  $^{228}\text{Th}$ . The  $^{230}\text{Th}$  concentration ratio in plants from the background locations was also elevated relative to  $^{232}\text{Th}$  values. The authors suggested that  $^{230}\text{Th}$  is associated to a considerable

degree with its parent material (natural uranium) and that isotopic exchange between  $^{230}\text{Th}$  and  $^{232}\text{Th}$  (in the natural thorium minerals) cannot be expected in soil.

Vera Tomé *et al* (2003) presented TF factors for different natural radionuclides belonging to the uranium and thorium series for grass-pasture in an area where a disused uranium mine is located in a Mediterranean region. Statistical analysis of the TF values obtained showed that TF values corresponding to two soils types (granite and alluvial) were statistically indistinguishable and the same was the case for the two study areas (region affected by mine and region unaffected by mine). The transfer factors for the two thorium isotopes  $^{232}\text{Th}$  and  $^{230}\text{Th}$  were statistically indistinguishable with mean values of 0.058 and 0.056 respectively. The TF for  $^{228}\text{Th}$  (mean value 1.65) were two orders of magnitude higher than those for  $^{232}\text{Th}$  and  $^{230}\text{Th}$ . The excess of  $^{228}\text{Th}$  over  $^{232}\text{Th}$  in vegetation must be due to the higher absorption of radium in particular  $^{228}\text{Ra}$ . Therefore the excess of  $^{228}\text{Th}$  arises from  $^{228}\text{Ra}$  decay in the plant and the uptake of  $^{228}\text{Th}$  itself (Vera Tomé *et al*, 2002). The radium uptake is greater than for thorium by about two orders of magnitude. These differences have been attributed to the different solubilities of the elements with oxidation states +II. Linsalata *et al* (1989) also found that TF values for elements in oxidation state +II (Ra) are always greater than those for elements in oxidation state +IV (Th).

Linsalata *et al* (1989) reported a mean concentration ratio (CR) value of  $6 \times 10^{-5}$  for edible vegetables growing in an area of enhanced radioactivity and these results are three orders of magnitude lower than Vera Tomé *et al* (2003) results. Zararsiz *et al* (1997) studied the uptake of thorium by thyme, forage and barley stems around a thorium ore deposit, and obtained TF values for unwashed plants of the same order of magnitude as those obtained in the study by Vera Tomé *et al* (2003) and an order of magnitude lower for washed plants. The mean TF value for thorium in grass is 0.011, with a range between 0.0011 and 0.11 was published by IUR (1994). Pietrzak-Flis and Suplinska (1995) compared thorium uptake in pasture grown in a tent to that grown outdoors and gave differences in mean CR values of less than a factor of 2. The mean CR values for thorium in pasture grown outdoors and in a tent were  $37 \times 10^{-4}$  and  $23 \times 10^{-4}$  respectively. Zararsiz

*et al* (1997) showed that there was relatively little difference in thorium uptake by pasture from contaminated alkaline and acidic soil types with CR of  $25 \times 10^{-4}$  and  $17 \times 10^{-4}$  respectively. Moffet and Tellier (1977) published CR of  $24 \times 10^{-4}$  for thorium uptake into grass species growing on uranium mill tailings.

Ewers *et al* (2003) carried out an extensive review of the data of transfer factors for natural radionuclides to vegetation for data pertaining to the UK and calculated geometric means for thorium uptake into a number of different vegetation groups. The geometric mean calculated from the relevant literature for thorium uptake into pasture, brassicas, root crops, potato tubers and cereal crops were  $30 \times 10^{-4}$ ,  $30 \times 10^{-4}$ ,  $20 \times 10^{-4}$ ,  $20 \times 10^{-4}$  and  $3 \times 10^{-4}$  respectively.

Sheppard *et al* (1989) applied  $^{230}\text{Th}$  at  $8 \text{ Bq kg}^{-1}$  to several soils varying in texture and organic matter content and found an overall geometric mean concentration ratio (CR) of 0.0022 for Th. They concluded that all their data indicated that thorium is about as plant available as uranium and not ten fold less as suggested by previous authors.

Relationship between thorium uptake and chemical parameters in soil and vegetation were limited and no clear conclusion could be obtained from the literature reviewed. Linsalata *et al* (1989) reported a geometric mean transfer factor for vegetation (corn, potato, manioc, squash, brown beans, carrot and couve) growing in Pocos de Caldas, Brazil of  $0.6 \times 10^{-4}$ . They found correlations between the CRs for thorium and the rare earth elements and the reasons for the correlation between the elements with such diverse chemistries is unclear but they postulated that it is apparently related to the essential mineral requirements or mineral status of the different vegetables sampled. This conclusion is based on the significant correlations obtained between the Ca content of the dried vegetables and the CRs for thorium and the rare earth elements. Arkhipov *et al* (1984) found from studying the uptake of thorium by barley from a range of artificially contaminated soil that as the content of humus and exchangeable calcium in the soil increases and as the soil solution reaction approaches neutral, the accumulation coefficient for  $^{232}\text{Th}$  decreased.

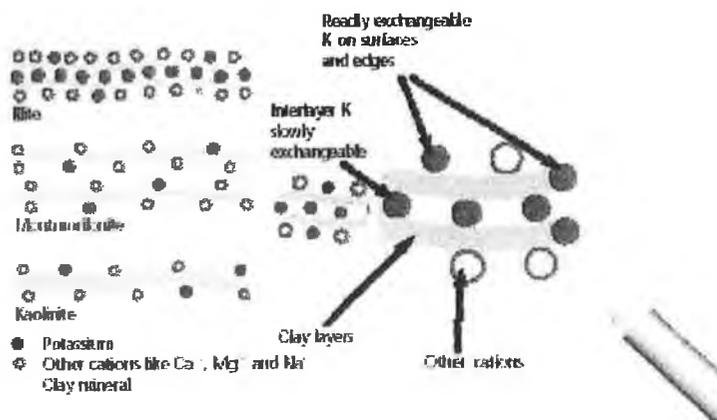
## 1.8 Potassium in the Terrestrial Environment

Potassium (K) is an alkali metal and is chemically similar to caesium and rubidium. It is an important constituent of soil and is widely distributed in nature being present in all plant and animal tissues. The three main isotopes of potassium are  $^{39}\text{K}$ ,  $^{40}\text{K}$  and  $^{41}\text{K}$ , with  $^{40}\text{K}$  being a radioisotope and the others stable isotopes.  $^{39}\text{K}$  makes up 93% of natural occurring potassium while  $^{41}\text{K}$  accounts for essentially all the rest.  $^{40}\text{K}$  only accounts for approximately 0.012% of naturally occurring potassium. There are several other radioactive isotopes of potassium but all have half-lives of less than one day so are therefore not of environmental concern. The half-life of  $^{40}\text{K}$  is 1.3 billion years.

### 1.8.2 Potassium in Soil

The behaviour of the potassium (K) in the soil is related to the type and amount of clay and soil organic matter. The type of clay depends on the parent rock, igneous or sedimentary, and the extent to which the mineral particles have undergone change (weathering) over many millennia. The clay-sized particles can be pictured as made up of many layers, each composed of interlocking silicon and oxygen atoms or aluminium and oxygen atoms. In some types of clay (illite) there are spaces between the layers into which positive ions, like potassium, can migrate and be held there by negative charges on the layers. In other clays (kaolinite) the layers are so tightly compacted that ions like potassium cannot enter between them. In both types of clay, potassium can be held at the edges of the clay layers. Also at the edges, the layers tend to open up, allowing ready ingress of potassium. The positive potassium ions held at the edges of the clay layers and towards the outer edge of any interlayer space can be replaced relatively easily by other positively charged ions. When this occurs, the potassium is released into the soil solution from where it is taken up by plant roots.

## Potassium and clay minerals



Clay minerals consist of lattices and layers and cations are held in various positions in and around the layers.

Figure 8: Potassium and Clay minerals

Potassium found in soil is in 1) soil solution, 2) exchangeable potassium (plant available), 3) non-exchangeable but available potassium (slowly available) and 4) mineral potassium (not available). While most soils contain large amounts of potassium in total, most of this is present in the soil minerals and therefore it must be weathered before it is made plant available. Unlike most other major nutrients potassium is not a component of and therefore cannot be stored in the soil organic matter. Furthermore, most soils do not have a large capacity (CEC) to store potassium in its inorganic form and therefore additional fertilizer potassium is required on most agricultural soils (Johnston 2003). Potassium and in particular <sup>40</sup>K in soil are discussed in more detail in Section 1.9.1.

### 1.8.3 Potassium in Plants

Of all the nutrients, potassium is present in plants in the highest concentrations. Potassium has two roles in the functioning of plant cells. Firstly, it plays a part in the activation of enzymes which are fundamental to metabolic processes, especially the production of proteins and sugars. Only small amounts of potassium are required for this

biochemical function. Secondly, potassium is the "plant-preferred" ion for maintaining the water content and hence the turgor (rigidity) of each cell, a biophysical role. A large concentration of potassium in the cell sap (i.e. the liquid inside the cell) creates conditions that cause water to move into the cell (osmosis) through the porous cell wall

Turgid cells maintain the leaf's vigor so that photosynthesis proceeds efficiently.

Photosynthesis is the process by which plants harvest the energy of sunlight to produce sugars. These sugars contain carbon derived from the carbon dioxide in the air that has entered the leaf through the stomata, tiny openings mainly on the underside of the leaf. These tiny openings are surrounded by "guard cells" and it is only while they are turgid that the stomata remain open and carbon dioxide can pass through into the leaf. But most of the water transpired by the plant is lost through the stomata when they are open. Thus, if there is a water deficit, the plant needs to close the stomata to conserve water. The plant controls the opening /closing of the stomata by regulating the concentration of potassium in the guard cells. A large concentration of potassium ensures turgid cells and open stomata. When the potassium in the guard cells is lowered, they become limp and the stomata close.

A high osmotic potential in plant cells is also needed to ensure the movement through the plant of nutrients required for growth, and the sugars produced by photosynthesis, for example, the transport of sugar to grains, beet roots, tubers, and fruit. By maintaining the salt concentration in the cell sap, potassium helps plants combat the adverse effects of drought and frost damage and insect and disease attack. It also improves fruit quality and the oil content of many oil-producing crops (Johnston, 2003).

Potassium influences the uptake of other elements and affects both respiration and transpiration. It also encourages the synthesis and translocation of carbohydrates thereby encouraging cell wall thickening and stem strength. A deficiency can cause lodging in cereals and yellowing of the leaf tips and margins and is usually the result of leaching and continuous cultivation. This fact can be rectified by fertilizer application (Fitzpatrick, 1986).

The behaviour of potassium in the plant will be further discussed with caesium in section 1.9.1.

## 1.9 Caesium in the Terrestrial Environment

Caesium, is an alkali metal is the most alkaline of all elements and is chemically similar to potassium and rubidium. There are 11 major radioactive isotopes of caesium but only three have half-lives long enough to warrant concern:  $^{134}\text{Cs}$ ,  $^{135}\text{Cs}$  and  $^{137}\text{Cs}$ . Each of these isotopes decays by emitting a beta particle, and their half-life range from 2 years to 2 million years. The half-lives of its other radioisotopes are less than 2 weeks. In relation to environmental management the isotope of most concern is the  $^{137}\text{Cs}$ , which has a half-life of 30.2 years. Its decay product,  $^{137\text{m}}\text{Ba}$  stabilizes itself by emitting an energetic gamma ray with a half-life of about 2.6 minutes. It is this decay product that makes caesium an external hazard.  $^{134}\text{Cs}$  and  $^{135}\text{Cs}$  are typically of less concern because of their radiological decay characteristics. Caesium is naturally present as  $^{133}\text{Cs}$  in various ores and to a lesser extent in soil. The three radioactive isotopes identified above are produced by nuclear fission.  $^{137}\text{Cs}$  is found in the terrestrial environment as a result of fallout from weapons testing and the Chernobyl accident.

A large body of literature exists on the fate of caesium in both soil and vegetation systems compared to the other radionuclides of interest ( $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$ ). The majority of this literature is relatively recent (post Chernobyl 1986) and this is reflected in the following literature review on caesium.

### 1.9.1 Chemistry of Caesium in Soil

Caesium, like potassium and rubidium are alkali metals and along with  $\text{NH}_4^+$  will tend to behave in a similar fashion in soil. Schulz (1965) stated that the energy of adsorption for these ions decreases in the following order:  $\text{Cs}^+ > \text{Rb}^+ > \text{K}^+ = \text{NH}_4^+$ . Schulz (1965) found that ions such as  $\text{Ca}^{2+}$  are about as effective as  $\text{Cs}^+$  itself in exchanging for macro-

amounts of caesium adsorbed on a soil. However where carrier-free  $^{137}\text{Cs}$  is adsorbed on a soil, calcium or divalent ions are quite ineffective in replacing the adsorbed  $^{137}\text{Cs}$ .  $\text{NH}_4^+$ ,  $\text{K}^+$  and  $\text{Cs}^+$  were far more effective as replacing ions (Schulz, 1965). As caesium is a Group 1 element, simple cation exchange processes in soil mainly govern it.

The behaviour of Cs in soil appears to be highly dependent on the clay and organic matter content in the soil. The clay content plays a very significant role in rendering the Cs unavailable to vegetation and the general consensus in the literature is that Cs is bound to peat in a weak non-specific form rendering the majority of Cs present in peat available to plant uptake. The most important physiochemical interaction within soils is the specific and almost irreversible attachment to clay minerals, notably the 2:1 clay mineral illite. Illite has a stacked layer structure. Each unit layer in turn is a gibbsite (Al-O or Al-OH) sheet sandwiched between two silica sheets. The sandwiches are bound together by potassium ions in so called inter-layers. The top and bottom of the two outer plates in a stack create surfaces on which simple ion exchange sorption can take place, so-called planar sites (Regular Exchange Sites: RES). Brouwer *et al* (1983) indicated that there are several different types of sites on illite and of these, two are of significant importance in relation to caesium retention, i.e. Regular Exchange Sites (RES) and Frayed Edge Sites (FES). The RES as mentioned previously are high capacity sites but are not as Cs selective as the FES. The FES, which is extremely selective for caesium, is located at the edges of the interlayer regions, where slightly wedge-shaped spaces exist between the outside edges of an adjacent pair of layers. The special shape of these sites limits entry and sorption to relatively small ions. As charged ions in water attract a sheath of water molecules around them by electrostatic attraction, most (hydrated) ions are too large to enter the space easily. However, cations such as  $\text{Cs}^+$  and to a lesser extent  $\text{K}^+$ ,  $\text{Rb}^+$ , and  $\text{NH}_4^+$ , with low hydration energy, can easily lose the hydration sheath and enter the so-called Frayed Edge Sites (FES). In contrast, ions such as  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{Sr}^{2+}$ , which are surrounded by a large and stable hydration shell, are not selectively sorbed. This selection by size introduces a positive bias towards caesium on the FES sites so that, whereas on the planar sites caesium competes on about equal terms with ions like  $\text{K}^+$ ,  $\text{Na}^+$  and  $\text{NH}_4^+$ , i.e. the selectivity coefficient is about 1, at the FES the selectivity coefficient for  $\text{Cs}^+$

relative to  $K^+$  is about 1000 and relative to ammonium is about 200 (DePreter, 1990). Because of the very high selectivity of FES for radiocaesium and the very low concentration of stable caesium in natural systems, almost all the radiocaesium in most soils and sediments will concentrate at these sites, even though they may constitute a very small fraction of the total number of sites (Smith, 2001). Additionally slow but almost irreversible penetration into the 'interlayer spaces' between the plate-like components of the mineral can take place, during which the potassium inter-layer ions can be exchanged for caesium ions. It is this process which leads to long-term fixation of radiocaesium in soils, rendering caesium physically immobile and biologically unavailable. The process of fixation of clay minerals was considered, pre-Chernobyl, to be able to render the majority of radiocaesium unavailable for uptake by crops over the long term in mineral soils under 'average' western European agriculture (Shaw and Bell, 2001).

Frayed edge attachment and inter-layer fixation is known to occur within soils containing even moderate to low quantities of illite. The degree to which soils of different characteristics can retain radiocaesium attached to FES has been defined on the basis of the relative availability of FES to 'capture' caesium ions, and has been coined as the Specific Radiocaesium Interception Potential (SRIP) (Cremers *et al*, 1988). Organic soils and peat of upland areas of Europe have been shown to possess a demonstrably low SRIP. Livens and Loveland (1988) produced overlay maps which demonstrated convincingly that the area in the south west corner of the Cumbrian Mountains, in which contamination of sheep was so high as to lead to a ban on sale and slaughter, coincided with both relatively high level of  $^{134}Cs$  and  $^{137}Cs$  deposition and with soils of low radiocaesium 'immobilization capacity'. These soils tended to be high in organic matter, had relatively low potassium contents, had relatively high ammonium contents and had a relatively low content of illitic minerals.  $NH_4^+$  has the ability to displace  $Cs^+$  ions from the FES of illite and therefore has a tendency to increase the bioavailability and mobility of radiocaesium within the soil. Soils of somewhat opposite characteristics were found in the area, which received similar levels of Chernobyl deposition, yet which did not lead to high levels of radiocaesium transfer to vegetation and animals. (Shaw and Bell, 2001, Valcke and Cremers, 1994, Comans and Hockley, 1992).

Sheppard and Thibault (1990) published estimates of  $K_d$  ( $K_d = [Cs]_{ads\ soil} / [Cs]_{aq}$ ) values for four generic soil types and they showed that for Cs the  $K_d$  value tended to decrease with soil type in the following order: loam > clay > sand = organic, reflecting the tendency for clay minerals in loam and clay soils to 'intercept' and 'fix' radiocaesium ions on frayed edge sites and inter-layer sites respectively. Roca and Vallejo (1995) studied the migration of radionuclides in soil contaminated with  $^{134}Cs$ . The fixation of  $^{134}Cs$  was found to be more effective in sandy-loam soil than in sandy soil in agreement with the higher clay and organic matter content of the former. The sandy loam soil showed less migration and a more rapid immobilization of  $^{134}Cs$  than sandy soil. Ratios of exchangeable  $^{134}Cs/K$  were higher in the sandy soil. The authors concluded that this was due to 1) greater immobilization of radionuclides in non-exchangeable form in the sandy-loam soil and 2) lower K concentration in the sandy soil. The lower immobilization of radiocaesium in the sandy soil resulted in the element being more plant available.

The ability of organic matter to adsorb Cs is well known but the mode of interaction is less well understood than that of clay. In some organic soils, caesium is apparently less strongly bound and absorption by plants is higher from peat soils than from mineral soils. This was highlighted by the unexpectedly high plant uptake in upland peaty areas after the Chernobyl accident. Staunton *et al* (2002) illustrated that, despite the weak, non-specific interaction between soil organic matter and Cs, the former decreases the affinity of clay minerals, thereby reducing their ability to immobilize Cs in soil. The authors conclude that this is one of the factors that contribute to the small immobilization of radiocaesium in organic soils and hence its large availability to plants and other soil biota. Chibowski and Zygmunt (2002) state that organic matter does not play a significant role in the process of caesium retention and that the adsorption of  $^{137}Cs$  on the organic component is low and fully reversible. They also state that the presence of small quantities of minerals in organic soils appears to be of primary importance and as the mineral content increases the adsorption of Cs clearly increases as well. Hird *et al* (1995) argued that even in highly organic soil horizons sufficient frayed edge sites exist to

account for observed patterns of Cs retention and this view is supported by Shand *et al* (1994), Cheshire *et al* (1998) and Hird *et al* (1996). Hird *et al* (1995) further postulate that the lack of Cs fixation is attributable to interaction between organic matter and the frayed edge sites. Rigol *et al* (1998) concluded that the mineral fraction was responsible for Cs sorption in the presence of humic substances and also that removal of humic substances increased Cs sorption. Shand *et al* (1994) studied the distribution of radiocaesium in organic soils and found that even in highly organic peat, the mineral components were important for the eventual fixation of radiocaesium and oxidizable organic matter corresponding to humin also strongly retained caesium. The authors suggest two forms of organic matter, which could fall within this criterion, are charcoal and fungal hyphae residues. The ability of fungi to concentrate caesium to a very high level in the living tissue and to retain a proportion of the element in a form non-exchangeable with caesium chloride has been demonstrated by Dighton *et al* (1991). Thiry and Myttenaera (1993) suggest that caesium fixation in OAh horizon is caused partly by absorption by microorganisms and partly by the humic-clay component. Recently Sanchez *et al* (2000) proposed that microbial content accounted for Cs retention by highly organic matter.

Rigol *et al* (2002) studied the effect of organic matter on root uptake of radiocaesium and have found that the specific sites in clay control radiocaesium adsorption in organic soils and those organic compounds have only an indirect but significant effect. They found that only in organic soils with more than 95% organic matter content and negligible clay contents does adsorption occur mostly in non-specific sites.

Sanchez *et al* (1988) studied the interaction of selected radionuclides with peat under acidic precipitation conditions and found that radiocaesium has a low sorptive coefficient in organic rich peat at pH 4, however, peat samples with some mineral components, which may contain significant exchangeable sites for Cs<sup>+</sup>, are able to retain this radionuclide. The authors obtained a sorption ratio ( $R_d$ : undefined) of  $1 \times 10^5$  ml/g at pH 4 in Spruce Flats peat, which is comparable to  $R_d$  reported for illitic shale at neutral pH values by Tamura (1972).

Lofts *et al* (2002) studied the role of humic acid in radiocaesium distribution in a British upland peat soil and found that the  $\text{Cs}^+$  cation associates with humic acid in a non-specific manner by accumulating in the counter ion swarm adjacent to the negatively charged humic molecules. The main control of  $\text{Cs}^+$  binding is the concentrations of competing counter ions and the magnitude of the humic molecular charge. The type of competing ion also influences the  $\text{Cs}^+$  binding, but this is a less important effect. The simulations carried out by the authors indicate that even when illite FES are present in very small amounts ( $< \approx 1\%$ ) they may still take up significantly more Cs than humic substances. They concluded that the role of humic substances in direct sorption of Cs is likely to be minor even in highly organic soils with negligible illite, given the possible role of microbial uptake identified by Sanchez *et al* (2000).

Barber (1964) suggested that when organic matter is responsible for a large fraction of the total exchange capacity of the soil, the adsorption and subsequent fixation of  $^{137}\text{Cs}$  on the clay minerals are much reduced. Livens *et al* (1991) examined the distribution of Cs in soils in upland areas of Europe and found that most of the soils contained illite or micaceous minerals that in principle can take up radiocaesium. However in the conditions encountered in these soils, extensive sorption of Cs to clays does not occur. The authors suggested that this may be due to recycling of radiocaesium in the uppermost organic horizons, which prevents it reaching the clay-rich, deeper horizons, or it may be due to the prevention of Cs-clay interaction by organic matter. The organics can either take up radiocaesium in competition with the minerals or they may react with the minerals and block the Cs uptake sites. Hird *et al* (1996) showed that the total number of sites for the potential fixation of radiocaesium was actually greater in acid organic soils of the UK uplands than in many lowland mineral soils. However they determined that the physical concentrations of stable caesium or potassium were too low in the former soils to induce collapse of the interlayer of clay minerals, a process that is required to trap and therefore fix radiocaesium. One of the main differences between these two groups of soils is that the potassium status is likely to be much higher in lowland soils, especially agricultural soils receiving fertilizers, so the likelihood of potassium-induced fixation of radiocaesium is higher in lowland mineral soils.

Gastberger *et al* (2000) found that the vertical migration velocity of  $^{137}\text{Cs}$  is much lower at alpine pasture sites than at lowland pasture sites. The low vertical migration velocity of radiocaesium in Alpine soils is a well-known phenomenon and according to the authors is due to a combination of factors. Low biological activity in Alpine soils in regard to the absence of earthworms, which are very abundant in the undisturbed soils of the permanent lowland pastures, is an important factor. According to Muller-Lemans and van Dorp (1996) it takes approximately 5 – 20 years for earthworms to turn over the topsoil of grassland once, resulting in intensive and more or less homogenous mixing. Furthermore the low migration velocities can also be partly explained by climatic factors. Alpine pastures are characterized by an extended snow cover (6-7 months), which is accompanied by frozen soil profiles therefore preventing intensive migration process due to lack of percolating water during most of the year.

### 1.8.2 Uptake of Caesium by plants

van Bergeijk *et al* (1992) suggested that the relationship between soil properties and soil to plant transfer of caesium is a complicated matter in which many parameters are involved. Clay fraction, type of clay mineral, organic matter content and amount of soluble and exchange calcium, potassium and ammonium are regarded as the main soil factors influencing transfer. Other factors, which appear to influence the uptake, are pH and moisture content of soil and species of vegetation.

Clay and organic matter play a significant role in the retention or mobilization of caesium within the soil as seen in the previous section and will obviously affect the availability of caesium in the soil and therefore it's transfer into vegetation.

van Bergeijk *et al* (1992) found that organic matter appears to affect the transfer factor of  $^{137}\text{Cs}$  positively. The authors postulated that this may be due to the large cation exchange capacity of organic matter and the spatial distribution of organic substances around clay particles, which prevents adsorption and subsequent fixation of  $\text{Cs}^+$  on the clay minerals and as the interaction of  $\text{Cs}^+$  with organic substances is weak, the ions remain available to

plants. Albers *et al* (1998) studied uptake of  $^{137}\text{Cs}$  by plants inhabiting alpine pastures in Germany and found a positive correlation between  $^{137}\text{Cs}$  content in plants and organic matter in soil. Evans and Dekker (1967) observed positive as well as negative effects of the soil organic matter content on the transfer of caesium. Tsukada and Nakamura (1999) could not find a correlation between  $^{137}\text{Cs}$  in potato and organic matter in soil. Carini (2001) concluded that highest transfer factors for fruit is attributable to peat and light textured soil and to calcareous soil with low potassium content in tropical or subtropical regions. Caesium tends to remain more available developed on organic soils with a distinct root matt than on sandy soils. There have also been reports of a continued uptake of caesium derived from atmospheric nuclear weapons testing by plant species which inhabit acid heaths and bogs. Seymour *et al* (1999) found that, in a deciduous forest ecosystem, the bulk (> 90%) of  $^{137}\text{Cs}$  is in the surface organic horizons and is present in a non-labile form most likely as a result of complexing with decomposing organic matter and very little is leached to the underlying mineral horizons.

Gerzabek *et al* (1998) found significant negative correlations between  $^{137}\text{Cs}$  TF into agricultural crops and clay content. Albers *et al* (1988) found a negative correlation of  $^{137}\text{Cs}$  content in plant with gravel content in soil. Livens *et al* (1991) concluded that the presence of micaceous minerals or illite clay do not significantly inhibit caesium uptake, either because of recycling in the organic surface horizon or because of clay-organic complex formation. Delvaux *et al* (2000) found that the transfer factors was very strongly negatively correlated to the Radiocaesium Interception Potential (RIP) which is defined as the product of the trace  $\text{Cs}^+$  to  $\text{K}^+$  selectivity coefficient in the FES and the FES abundance ( $\text{RIP} = K_c^{\text{FES}}(^{137}\text{Cs}^+ / \text{K}^+ [\text{FES}]) (\text{mol/kg})$ )

Kruyts and Delvaux (2002) concluded that in  $^{137}\text{Cs}$  contaminated forest soils the surface horizons constitute a major potential source for  $^{137}\text{Cs}$  soil-to-plant transfer and gave four possible reasons for this: (1) these horizons have large contents of organic matter, which does not specifically retain trace  $^{137}\text{Cs}^+$  ions, (2) they have a very small number of caesium-specific FES which can act in deeper mineral horizons as a very efficient mineral sink for radiocaesium, (3) these horizons are usually acidic and potassium-

depleted thus favouring root uptake of trace  $Cs^+$  and (4) plant roots usually extensively explore these horizons extensively. Coughtrey *et al* (1989) stated that in the case of non-organic soils the uptake of caesium from soil by plants is limited, because the element is bound initially to natural clay minerals by cation exchange and subsequently can be incorporated into the lattice of certain minerals.

Frissel and Heisterkamp (1989) provided statistical summaries of recommended values for TFs for generic crop types growing on generic soil type. The soil types groups were sand, clay and peat, and the vegetative groups were grass, cereals, and crops (green and root). The TF values for grass samples decreased in the order: peat > sand > loam. The order was the same for cereals but TF values in each case were one order of magnitude lower in the cereals than the grass. The TF values into green vegetables decreased in the following order: sand > peat > clay and root vegetables decreased in the order: peat > sand > clay.

van Bergeijk *et al* (1992) concluded that soil pH hardly any effect on the TF values in the pH range 3.9 – 8.4. Frissel *et al* (2002) suggest that transfer factors are not affected by change in pH unless the pH value drops below 4 or 5 where an increase in TF appears to result from lower pH. Frissel *et al* (2002) state that soils which almost entirely consist of organic matter show higher Cs TF values, this being due to the presence of organic matter, absence of nutrients and lower pH. Strebl *et al* (2002) found a negative correlation with radiocaesium soil-plant transfer factor and pH and a negative correlation with TF values and exchangeable fractions of Mg, Ca and Na. The authors pointed out that this could be interpreted as a pH effect on cation exchange capacity where higher pH values of carbonate containing soils correlate positively with the exchangeable fraction of calcium and magnesium. Gerzabek *et al* (1998) found significant negative correlations between  $^{137}Cs$  TF values in agricultural crops and pH.

Ehlken and Kirchner (1996) found a negative correlation between  $^{137}Cs$  and soil moisture content and suggested that this is due to the influence of evapotranspiration resulting in

higher trace element concentrations in the remaining solution and thus in higher root uptake and these findings were supported by Ehlken and Kirchner (2002).

Rafferty *et al* (1997) conclude that the behaviour and stability of the microbial community of the forest floor is essential to the persistence, bioavailability, mobility and distribution of the  $^{137}\text{Cs}$  pool in forest soils. Albers *et al* (1998) concluded that microbial activity increases the uptake of radiocaesium.

Bunzl *et al* (2000) determined the  $^{137}\text{Cs}$  concentrations in soils and plants in alpine pastures and found no correlation between soil concentration and plants, however during this sampling individual plant species were not selected from the plant community, which may be masking possible trends. Ciuffo *et al* (2002) could not find a statistically significant linear relationship between soil activity of  $^{137}\text{Cs}$  or  $^{40}\text{K}$  in soil and grass.

Schulz (1965) stated that the mobility and subsequently availability of a given ion in soil is dependent on the complementary ions present in soil. Schulz (1965) categorizes the complementary ions into two groups (1) elements ( $\text{H}^+$ ,  $\text{Na}^+$ , and the alkaline earths,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{Ba}^{2+}$ ) which will liberate about 10% of tightly bound  $\text{Cs}^+$  and (2) elements ( $\text{Cs}^+$ ,  $\text{NH}_4^+$ ,  $\text{K}^+$  and  $\text{Rb}^+$ ) which can liberate between 20 – 80% of  $\text{Cs}^+$  to the liable form. As typical agricultural soils in the neutral pH range contain primary liable  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  in large amounts, plants would be expected to take up little  $^{137}\text{Cs}$ . However in modern agriculture three inorganic fertilizers (NPK) are added to soils. These are nitrogen, potassium and phosphorous. Schulz (1965) stated that the effect of fertilization on the liberation from caesium from soil is dependent on the chemical forms in which nitrogen, potassium and phosphorous are added to the soil. Nitrogen can be added as either the cation,  $\text{NH}_4^+$ , or as the anion,  $\text{NO}_3^-$ . If nitrogen was added as the  $\text{NH}_4^+$  ion, it would be expected to liberate a considerable amount of fixed  $^{137}\text{Cs}$  into a liable form. However if it were added as  $\text{Ca}(\text{NO}_3)_2$ , this fertilizer would be expected to have little effect on the fixed  $^{137}\text{Cs}$ . If potassium was added as  $\text{K}_2\text{SO}_4$  in the same amounts as  $\text{NH}_4^+$ , the added  $\text{K}^+$  would increase the uptake of  $\text{Cs}^+$  to about the same degree as  $\text{NH}_4^+$ . The authors also stated that the possible effects of microbiological oxidation of  $\text{NH}_4^+$  to  $\text{NO}_3^-$  and the

possibility of ion competition in plant uptake by the root should be taken into consideration. Schulz (1965) found from experiments that the effect of the potassium salts was much less than that of the  $\text{NH}_4^+$  and this result was not expected based on the complementary ion effects. The authors conclude that these observations may be due to three considerations; 1)  $\text{K}^+$  and  $\text{NH}_4^+$  are about equally effective in releasing fixed  $^{137}\text{Cs}$  to the soil solution, 2) Both  $\text{NH}_4^+$  and  $\text{K}^+$  in solution will compete with  $^{137}\text{Cs}$  for plant uptake, and 3) Microbiological oxidation of liable  $\text{NH}_4^+$  to  $\text{NO}_3^-$  will take place thus removing the  $\text{NH}_4^+$  from competition for  $^{137}\text{Cs}$  uptake. However the  $\text{NH}_4^+$ , which has substituted for the fixed  $^{137}\text{Cs}$  will not be readily oxidized and will remain as  $\text{NH}_4^+$  fixed as clay minerals thus preventing the  $^{137}\text{Cs}$  from reverting to the fixed state (Schulz, 1965).

Bunzl *et al* (2000) concluded that, from the very strong correlation between  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in the plant communities of the pastures analyzed, for a large variety of plant species present, radiocaesium and potassium do not necessarily behave in an analogous manner. McGee *et al* (1996) also found a highly significant negative correlation for two individual plant species growing on a peat land soil. They found that *Juncus squarrosus* accumulated significantly more  $^{40}\text{K}$  than did *Calluna vulgaris*, but the opposite was observed for the  $^{137}\text{Cs}$  content. Based on pot experiments, Sanchez *et al* (1988) concluded that the high plant uptake of radiocaesium in unimproved organic soils is partly due to substantial ammonium concentrations and low amounts of exchangeable potassium in the soil solution. von Fricks *et al* (2002) found that the uptake of  $^{137}\text{Cs}$  in *Salix* plants was affected by K availability. The  $^{137}\text{Cs}$  concentration in plants was higher in the 0 kg K treatment than in the 80 or 240 kg K treatments however very little difference was noted between the 80 kg K and 240 kg K treatments. This is in agreement with Rosén (1996) who found that  $^{137}\text{Cs}$  uptake by grass was significantly reduced at potassium fertilization rates of 100 kg K  $\text{ha}^{-1}$ . However this effect was much less pronounced as the fertilization rates were increased to 200 kg K  $\text{ha}^{-1}$ . Smolders *et al* (1996) also found that potassium fertilization reduced  $^{137}\text{Cs}$  availability at low potassium concentrations in soil, whereas at high potassium availability fertilization had little effect on  $^{137}\text{Cs}$  uptake. Frissel *et al* (2002) found from plotting exchangeable potassium against  $^{137}\text{Cs}$  TF values that potassium levels seem to respond to a threshold mechanism where below 0.5

mmol(+)/kg, exchangeable potassium levels exhibit a strong influence on TF values, while above that limit other factors determine the TF value.

Ciuffo *et al* (2002) determined soil to plant relationships for  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in a semi-natural grasslands of the Giulia Alps (Italy), and observed a negative correlation between  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in the plant community of the pasture. The authors concluded that in pastures with similar characteristics to that area,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  do not necessarily behave in an analogous way. In general they found that plants with low potassium content in its tissue tend to take up more radiocaesium from soil than plants with high tissue potassium content.

Tsukada and Nakamura (1999) found that the transfer factors of both  $^{137}\text{Cs}$  and stable Cs decreased in potato with increasing K concentration in the soil, which was mainly supplied as fertilizer. Gerzabek *et al* (1998) found significant negative correlations between  $^{137}\text{Cs}$  TF into agricultural crops with plant available potassium, total potassium, plant available phosphorous and the amount of EDTA-extractable zinc. The authors found positive correlation with the iron content of the soil and the amount of EDTA-extractable copper. Rigol *et al* (2002) studied the effect of organic matter on root uptake of radiocaesium and found that in organic soils the low clay content and high  $\text{NH}_4^+$  concentration in the soil solution are responsible for the high soil-plant transfer. Also the low  $\text{K}^+$  levels in the solid phase and in the soil solution in upland sites enhances root uptake of  $^{137}\text{Cs}$ . Robinson and Stone (1992) found that application of potassium to the soils of Bikini Atoll reduced the  $^{137}\text{Cs}$  content in coconuts with a 9-12 month period after single large applications and lasted for three years. A comparable but more short-lived reduction occurred in grass and herbaceous species beneath the palms.

Albers *et al* (1998) could not find a significant correlation between the caesium and potassium in soils or plants. Strebl *et al* (2002) concluded that neither the amount of exchangeable potassium nor  $^{40}\text{K}$  concentration in soil showed a significant correlation with  $^{137}\text{Cs}$  TF values. However the ratio of exchangeable potassium to  $^{40}\text{K}$  ( $\text{K}_{\text{exch}}/^{40}\text{K}$ ) showed a highly significant negative correlation with  $^{137}\text{Cs}$  TF values. The authors

suggested that the  $K_{\text{exch}}/^{40}\text{K}$  ratio indicates the fraction of total potassium available for plant uptake and therefore may possibly be a suitable descriptor for monovalent cation-binding conditions in the soils investigated. The authors also concluded that the behaviours of  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in meadow grass are not analogous over time.

Strebl *et al* (2002) suggest that in comparison with arable land, soil to plant transfer in meadows is higher and ecological half-times of radiocaesium in meadow ecosystems are greater than in intensively used agro ecosystems. Reasons given by the authors for the differences were cycling of the nutrients within the perennial plants and environmental conditions characterized by lower pH, higher content of organic matter in topsoil and a lower level of plant nutrient supply. Ehlken and Kirchner (1996) concluded that grazing of cattle and repeatable cutting of grass can enhance plant uptake of  $^{137}\text{Cs}$  by a factor of 2-3. They found that transfer factors for  $^{137}\text{Cs}$  was significantly higher if the grass was chopped after regrowth (simulate cattle grazing) than if it was cut once a year and the authors attributed this to the fact that grass plants which are defoliated repeatedly develop a more shallow root system resulting in preferential uptake from the uppermost soils layers where concentrations of radionuclides are highest.

Roca and Vallejo (1995) determined TF values ( $[^{134}\text{Cs}]_{\text{plant d.w.}} / [^{134}\text{Cs}]_{\text{total exch soil m}^{-2}}$ ) for  $^{134}\text{Cs}$  and K for vegetation growing on artificially contaminated sandy and sandy loam soil. Transfer factors for lettuce were calculated for seedlings, young plants and mature plants on both soil types. The TF values for seedlings growing on the sandy-loam and sandy soil were  $0.178 \pm 0.078$  and  $0.221 \pm 0.131$  respectively. The TF values for mature plants growing on sandy-loam and sandy soil were  $0.131 \pm 0.089$  and  $0.460 \pm 0.403$  respectively. Papastefanou *et al* (1999) determined soil to plant TF values ( $[^{137}\text{Cs}]_{\text{plant d.w.}} (\text{Bq kg}^{-1}) / [^{137}\text{Cs}]_{\text{soil d.w.}} (\text{Bq kg}^{-1})$ ) for grass over an eleven year period in the Thessaloniki area of northern Greece. The TF values for grassland ranged from 0.002 to 7.42, with an average value of 0.20 and TF for  $^{40}\text{K}$  ranged between 0.16 and 2.42 with an average value of 0.42. The authors concluded that the ecological half-life  $T_{\text{ec}}$ , for  $^{137}\text{Cs}$  in grassland of 3½ years is almost identical to that for  $^{40}\text{K}$  in grassland of 3½ years.

Rafferty *et al* (1994a) determined seasonal concentration ratios (CR = activity in grass (Bq/kg) d.w. / activity in 0-1 cm soil (Bq/kg) d.w.) for both  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in pasture from three grazing sites in Ireland. Overall the  $^{137}\text{Cs}$  values were much lower than those for  $^{40}\text{K}$  with the  $^{137}\text{Cs}$  values generally lower than 0.25 and the  $^{40}\text{K}$  values varying from 2- 6. The authors conclude that the uptake of  $^{137}\text{Cs}$  appears to occur during winter months whereas the  $^{40}\text{K}$  uptake follows the normal winter reduction expected for nutrient elements in perennial plants and a subsequent work carried out by Rafferty *et al* (1994b) concluded that that soil adhesion to grass surfaces contributes to  $^{137}\text{Cs}$  soil to plant transfer as levels of soil contamination increases during the winter months due to heavy rainfall and grazing. They further concluded that soil adhesion did not contribute significantly to the  $^{40}\text{K}$  content of the grass samples and differences between  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in this regard are due to the relative extent of plant uptake of the two radionuclides. The  $^{40}\text{K}$  uptake by grass in their study was higher than that of  $^{137}\text{Cs}$  so the contribution of soil adhesion in relation to  $^{40}\text{K}$  is relatively insignificant. Rafferty *et al* (1994a) also indicated that the seasonal fluctuations in the  $^{137}\text{Cs}$  content of the grasses are clearly reflected in the  $^{137}\text{Cs}$  of the faeces of grazing sheep on the sites. A six fold difference between winter and summer in the faecal  $^{137}\text{Cs}$  was noted indicating the importance of seasonality in the prediction of food chain contamination. The authors did not note a seasonal pattern in  $^{40}\text{K}$  in the faeces of the sheep and the authors suggested that one reason for this might be due to the fact that potassium is mostly excreted in urine, which would therefore reduce the faecal  $^{40}\text{K}$  content. Another possible reason given may be a difference in bioavailability of the ingested forms of  $^{137}\text{Cs}$  and  $^{40}\text{K}$ . Rafferty *et al* (1994c) analyzed the effect of soil contamination on levels of  $^{137}\text{Cs}$  in winter fodders taken from both mineral and organic soil. They found that the surface adhesion of soil is the most significant contributor to the total  $^{137}\text{Cs}$  content of the sample (grass, hay, silage and fodder beet). The authors even found this to be the case for organic soils where root uptake of radiocaesium is generally more significant than on mineral soils.

Frissel *et al* (2002) derived generic values for soil-to-plant TF values based on a conversion factor for systems compared to cereal crops. The TF definition used by the authors is the ratio between the units of radioactivity per unit of mass dry crop and the

units of radioactivity per unit of dry soil. The concentration in the crop refers to the concentration in the edible part of plant at harvest time where applicable and the concentration in the soil refers to the upper 20 cm of soil for all crops with the exception of grass for which 10 cm was used. The proposed TF values for cereals in all soils with high nutrient status (pH > 4.8) is 0.006, for clay and loam soils with medium nutrient status (pH > 4.8) is 0.03 and peat and sand soils the value is set at 0.05. The conversion factor for grass is proposed to be 4.5 based on a weight basis and if based on metered squared the conversion factor would be 9. The conversion factor for woody species leaves of trees and new wood is proposed to be 20.

Strebl *et al* (2002) determined both TF values and aggregated transfer factors ( $T_{agg}$ ) values for  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in meadow vegetation at grassland sites in Austria. The TF values were defined on a weight basis, dividing the radionuclide concentration in plant dry matter (Bq/kg) by the weighted average radionuclide concentration (0 - 5 and 5 - 10 depth increments = 0.75:0.25; Bq/kg). The  $T_{agg}$  value was calculated by dividing the  $^{137}\text{Cs}$  concentration in plant dry matter (Bq/kg) by the  $^{137}\text{Cs}$  soil inventory in the rooting depth of soil (i.e. sum of inventories in 0-5 and 5-10 cm layers; Bq/m<sup>2</sup>). The mean (geometric) TF values for  $^{137}\text{Cs}$  from eight pasture sites in Austria ranged from 0.03 to 1.06. The  $T_{agg}$  value for  $^{137}\text{Cs}$  ranged from 0.0005 to 0.018. The mean (arithmetic) mean TF value for  $^{40}\text{K}$  for the eight pasture sites ranged from 0.31 to 2.01.

Gastberger *et al* (2000) studied soil to plant transfer factors in Austrian pastures and yielded  $^{137}\text{Cs}$   $T_{agg}$  (m<sup>2</sup> kg<sup>-1</sup>) values of 0.043 and 0.013 for Alpine grassland and lower values of 0.0004 and 0.0006 for lowland pastures of the same geographical region.

Ehlken and Kirchner (1996) studied the seasonal variations in caesium and potassium soils to plant transfer in north German soils and found that differences in TF values between grass plants growing on different soils ranged up to a factor of about 100 for  $^{137}\text{Cs}$ . Caesium-137 TF values for grazed (repeatedly cut) and growth (cut once per year) grasses on the two mineral soils, namely Orthic Podsol and Eutirc Cambisol ranged between 0.05 – 0.35 and 0.01 and 0.2 respectively depending on time of cutting. The

transfer factors for grazed (repeatedly cut) and growth (cut once per year) grasses on the two organic soils, namely Eutric Histosol and Umbric Gleysol both ranged between 1 – 7 depending on time of cutting. The effect of organic matter on the transfer factor is evident in the previous results.

IUR (1994) published expected TF values (Bq per dry weight crop / Bq per dry weight soil) for grass grown on three different soil types. The expected TF values for grass grown on clay, loam (pH 6), sand (pH 5) and peat (pH 4) are  $1.1 \times 10^{-1}$ ,  $2.4 \times 10^{-1}$  and  $5.3 \times 10^{-1}$  respectively, reflecting the impact of both clay and organic matter on the transfer factor value.

Baeza *et al* (2001) determined transfer coefficient (TC: activity in plant (Bq/kg) d.w. per unit of activity in the soil (Bq/kg) d.w.) and effective transfer coefficients (ETC: activity in plant (Bq/kg) d.w. per unit of activity in available soil (Bq/kg) d.w.) for  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in aerial and below ground parts of pasture vegetation growing on a sandy loam soil in a Mediterranean grazing-land ecosystem. The annual mean TC and ETC values for  $^{137}\text{Cs}$  in aerial fractions was determined to be  $0.25 \pm 0.17$  and  $3.4 \pm 2.3$  respectively and the annual mean TC and ETC values for  $^{137}\text{Cs}$  in below ground fractions were determined to be  $0.43 \pm 0.17$  and  $6.0 \pm 2.8$  respectively. The annual mean and standard deviation TC and ETC for  $^{40}\text{K}$  in aerial fractions was determined to be  $0.3 \pm 0.37$  and  $18 \pm 20$  respectively and the annual mean and standard deviation TC and ETC for  $^{40}\text{K}$  in below ground fractions were determined to be  $0.25 \pm 0.12$  and  $15 \pm 7$  respectively. The authors pointed out that the standard deviations associated with the annual mean values are large; especially in the case of  $^{40}\text{K}$  as a consequence of a marked seasonal variation in their radioactive concentrations and that these variations are caused by the Mediterranean climate with its sharp seasonal contrasts in temperature and relative humidity which may affect radionuclide uptake. The authors found that during the time period November to February there was a net transfer of  $^{137}\text{Cs}$  and  $^{40}\text{K}$  from the available soil fraction to the roots and during the period May to November there was a continuous loss of biomass in the roots which in general had the consequence of provoking the return of the two radionuclides to the available soil fraction. The authors also found a net flux of

radionuclides from the root to the aerial part of the pasture during the November to February period due to the growth of the pasture and to the incorporation of radionuclides that this process carries with it. For the period May to November, the authors concluded that loss of biomass implies a negative flux for the radionuclides i.e. they leave the aerial part of the plants for the roots.

Velasco *et al* (2004) determined  $T_{agg}$  values (activity concentration in plant (Bq/kg) d.w. / activity superficial concentration on ground (Bq/m<sup>2</sup>)) for both <sup>137</sup>Cs and <sup>40</sup>K in grassland ecosystems over a period of time. Semi natural grassland were studied in an upland region of Italy where the dominant vegetation consists of herbaceous leguminous (*Edicago sativa*, *Trifolium repens*, *T. pratensis*) and gramineous species used for hay production and the soil type were of the sand or sandy-loam type derived from calcareous rocks. Results showed that the mean of  $T_{agg}$  (<sup>40</sup>K) value was more than 17 times higher than mean of  $T_{agg}$  (<sup>137</sup>Cs) value indicating the existence of a selection phenomenon between the two ions in favour of potassium by plant roots, The authors obtained a difference of two order of magnitude between minimum ( $4.45 \times 10^{-5}$ ) and maximum ( $9.53 \times 10^{-3}$ )  $T_{agg}$  (<sup>137</sup>Cs) value with a coefficient of variance of 159. The mean  $T_{agg}$  (<sup>137</sup>Cs) value was  $9.27 \cdot 10^{-4}$ . For  $T_{agg}$  (<sup>40</sup>K) value, the minimum and maximum values were  $4.19 \times 10^{-3}$  and  $9.19 \times 10^{-2}$  respectively with a mean value of  $1.55 \times 10^{-2}$ . Seasonal variation in  $T_{agg}$  values were found for both radionuclides with  $T_{agg}$  lower in late spring and increased in summer and late summer-autumn. This increase corresponds to an increase in nutrient requirements by the vegetation. The authors conclude that  $T_{agg}$  seasonal variability usually is attributed to two factors: the physiology and the development of the plant. Plants present different and distinctive activity patterns. Activity is low or null in winter and autumn, the seasons with adverse conditions for growth and reproduction. In spring and summer the activity level gradually increases reaching its highest point in summer.

Albers *et al* (1998) studied the spatial distribution of fallout <sup>137</sup>Cs and natural <sup>40</sup>K in soils and vegetation of alpine pastures growing on dystric cambisols. The mean  $T_{agg}$  value (activity concentration in plant (Bq/kg) d.w. / mean concentration of <sup>137</sup>Cs total

(Bq/m<sup>2</sup>) for two sites analyzed was  $1.3 \times 10^{-3} \text{ m}^2 \text{ kg}^{-1}$  and  $2.0 \times 10^{-3} \text{ m}^2 \text{ kg}^{-1}$  respectively with the latter site having a  $T_{\text{agg}}$  nearly twice that of the former. The authors conclude that this is due to the higher content of organic matter, higher CEC and pH at the latter site, which are known to fix <sup>137</sup>Cs less tenaciously, and also due to more intensive grazing at the latter sites, which is known to increase uptake of <sup>137</sup>Cs.

Bunzl and Kracke (1986) studied the accumulation from fallout (weapons resting) of <sup>137</sup>Cs in *Calluna vulgaris* (heather), *Vaccinium myrtillus* (bilberry), *Vaccinium uliginosus* (bog bilberry), *Vaccinium vitis-idaea* (mountain cranberry), *Vaccinium oxycoccos* (northern cranberry), *Trichoporum caespitosum* (grass) and *Molinia coeruleae* (grass) on a peatland in south Bavaria. The TF values ( Bq/kg dry vegetation /Bq/kg dry soil) for leaves, flowers, stem and roots of *Calluna vulgaris* (heather) were 7, 6, 0.6 and 2 respectively. The TF for leaves, flowers, berries, stem and roots of *Vaccinium myrtillus* (bilberry) were 3, 3, 2, 2, and 2 respectively. The TF for leaves, flowers, berries, stem and roots of *Vaccinium uliginosus* (bog bilberry) were 2, 3, 2, 0.9 and 1 respectively. The TF for leaves, flowers, berries, stem and roots of *Vaccinium vitis-idaea* (mountain cranberry) were 1, 4, 2, 1 and 1 respectively. The TF for leaves, flowers, berries and stem of *Vaccinium oxycoccos* (northern cranberry) were 2, 2, 1 and 0.9 respectively. The TF for old leaves (yellowed) and green leaves for both *Trichoporum caespitosum* (grass) and *Molinia coeruleae* (grass) were 0.3 and 2 respectively. The authors concluded that it is unlikely that foliar absorption has a significant effect on the transfer factors observed due to the low level of <sup>137</sup>Cs deposition fallout at the time of sampling with the possible exception of *Calluna vulgaris* and *V. vitis-idaea*, which are not deciduous plant and which can therefore accumulate fallout radionuclides for several years by direct contamination of the leaves.

Coughtrey *et al* (1989) studied the transfer of caesium and potassium to vegetation in upland grassland in England. Transfer factor values for pre-Chernobyl <sup>137</sup>Cs varied from 3.2 to 6.5 on wetter and less mineralized soil compared to TF values of 0.53 – 3.6 on drier more mineralized soil. Transfer factor values were based on bulk vegetation inhabiting the area which was a mixture of *Eriophorum augustifolium*, *E. vaginatum*

(cotton grass) and mosses such *Polytrichum commune* and *Sphagnum* sp. in wetter areas. Vegetation dominant in the dry, more mineralized area were *Nardus stricta* (mat grass), *Juncus squarrosus* (heath rush), *Festuca/Agrostis*, *Luzula campestris* (field woodrush), *Hypnum cupressiforme* and *Pleurozium schreberi*. Vegetation was separated into individual species and those, which showed the highest TF values, were *Sphagnum* sp followed by *Polytrichum commune*. Plant soil TF values for potassium at the wetter less mineralized sites were  $5.51 \pm 1.08$  and at the drier more mineralized sites were  $7.25 \pm 0.94$ . Coughtrey et al (1989) detected  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$  and pre-Chernobyl activities in *Juncus effuses* of 880 Bq/kg, 270 Bq/kg and 260 Bq/kg respectively and  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$  and pre-Chernobyl activities in *Eriophorum vaginatum* of 2100 Bq/kg, 770 Bq/kg and 340 Bq/kg respectively.

Bunzl and Kracke (1989) studied seasonal variations of soil to plant transfer of  $^{134,137}\text{Cs}$  and K into peatland vegetation in Bavaria. The authors calculated concentration ratios for two grass species (*Trichophorum caespitosum* and *Molinia caerulea*) dominant in the area. The CR (activity in the aboveground vegetation (Bq kg) d.w. / activity in the soil (Bq kg) d.w.) of  $^{137}\text{Cs}$  from global fallout decreased for *Trichophorum caespitosum* from 1.9 in the spring to 0.08 in the autumn and for Chernobyl derived  $^{137}\text{Cs}$  from 1.4 to 0.2. The CR for *Molinia caerulea* of  $^{137}\text{Cs}$  from global fallout decreased from 1.6 in the spring to 0.04 in the autumn and for Chernobyl derived  $^{137}\text{Cs}$  from 4.0 to 0.3. Possible reasons for this behaviour given by the authors are uptake of radiocaesium by different sections of the roots in time and or vertical transport of the radionuclide in the soil during the year. The authors also analyzed the evergreen plant *Calluna vulgaris* (heather) that was contaminated surfically by the Chernobyl fallout and found that the  $^{137}\text{Cs}$  concentrations remained relatively constant during 1987 (leaves and stems 10,000 and 5000 Bq kg<sup>-1</sup> respectively) even though the radiocaesium present was taken up by the leaves and transported within the plant. The radiocaesium present in the grass species was taken up by the roots and while they have perennial roots they were not sprouting during the time period of the Chernobyl fallout.

Bunzl and Kracke (1984) published TF values (Bq/kg plant d.w. / Bq/kg soil d.w. 0-15 cm) for  $^{137}\text{Cs}$  in *Calluna vulgaris* growing on a sandy soil growing on a Luxembourg heath. The TF values for leaves, flowers, stem and root of *Calluna vulgaris* were as follows 28.7, 35.3, 10.0 and 13.3 respectively. Bunzl and Kracke (1981) illustrated that *Calluna* flowers are important in honey production in some areas and demonstrated that the existence of another food-chain pathway for  $^{137}\text{Cs}$  to man is possible as they detected mean values of fallout caesium 78 times greater in honey derived from *Calluna* flowers than from the flowers of other species in samples collected throughout Germany.

Rafferty (1996) reported maximum observed  $^{137}\text{Cs}$  concentration in a range of peatland species. The  $^{137}\text{Cs}$  concentrations in *Calluna vulgaris* (ling heather), *Erica tetralix* (bog heather) and *Vaccinium myrtillus* (bilberry) were 3030 Bq kg<sup>-1</sup>, 514 Bq kg<sup>-1</sup> and 526 Bq kg<sup>-1</sup> respectively. The  $^{137}\text{Cs}$  concentration in *Scirpus cespitosus* (bulrush), *Eriophorum vaginatum* (bog cotton), and *Eriophorum angustifolium* (common cotton grass) were 231 Bq kg<sup>-1</sup>, 611 Bq kg<sup>-1</sup> and 625 Bq kg<sup>-1</sup> respectively. The  $^{137}\text{Cs}$  concentration found in *Sphagnum cuspidatum*, *Polytrichum commune* and *Cladonia impexa* (lichen) were 1137 Bq kg<sup>-1</sup>, 919 Bq kg<sup>-1</sup> and 1326 Bq kg<sup>-1</sup> respectively.

Dowdall *et al* (2005) studied the uptake of radionuclides by vegetation at a High Arctic and found that soil TF (Bq kg<sup>-1</sup> plant d.w. / Bq kg<sup>-1</sup> soil d.w.) values for  $^{137}\text{Cs}$  in vascular plants ranged between 19.1 and 0.16 and showed considerable variation within each species, while  $^{137}\text{Cs}$  T<sub>agg</sub> (Bq kg<sup>-1</sup> plant d.w. / Bq m<sup>-2</sup>) values for the vascular plants were in the range of 0.002 to 0.25 m<sup>2</sup> kg<sup>-1</sup>. They also found that uptake into mosses and lichens were higher than for vascular plants.

Green *et al* (1995) calculated transfer factors (Bq kg<sup>-1</sup> plant dry weight / Bq kg<sup>-1</sup> soil dry weight 0-20 cm) for a range of vegetation growing on an area of land reclaimed from the sea on the Lancashire coast in England. Mean TF values for the following vegetables: leafy green, root, tomato, onions, legumes and grain were  $2.7 \times 10^{-2}$ ,  $2.4 \times 10^{-2}$ ,  $2.5 \times 10^{-2}$ ,  $1.9 \times 10^{-2}$ ,  $2.1 \times 10^{-2}$  and  $1.1 \times 10^{-2}$  respectively. Transfer factor values for lettuce, radish, tomatoes, cucumber and marrow were  $2.9 \times 10^{-2}$ ,  $3.6 \times 10^{-2}$ ,  $2.5 \times 10^{-2}$ ,  $1.4 \times 10^{-2}$  and 3.5

$\times 10^{-2}$  respectively. These low TF values reflect the fact that in excess of 80% (> 90% in pasture land) of the  $^{137}\text{Cs}$  present in the soil is present in the residual (mineral) phase and therefore, less likely to be taken up by the plant inhabiting these areas. Nisbet *et al* (1999) published recommended soil to plant transfer factor for radiocaesium in arable systems and the recommended value for each crop grown on five different soil types is shown in the table below.

Soil	Cereals	Tubers	Green Vegetable	Brassicas	Root vegetables	Legumes	Onions
<i>Sand</i>	$2.1 \times 10^{-2}$	$1.1 \times 10^{-1}$	$2.1 \times 10^{-1}$	$1.2 \times 10^{-1}$	$5.4 \times 10^{-2}$	$7.4 \times 10^{-2}$	$1.3 \times 10^{-2}$
<i>Loam</i>	$1.4 \times 10^{-2}$	$2.9 \times 10^{-2}$	$1.2 \times 10^{-1}$	$2.8 \times 10^{-2}$	$3.7 \times 10^{-2}$	$1.1 \times 10^{-2}$	$8.5 \times 10^{-3}$
<i>Clay</i>	$1.1 \times 10^{-2}$	$2.9 \times 10^{-2}$	$6.6 \times 10^{-2}$	$4.4 \times 10^{-2}$	$2.2 \times 10^{-2}$	$3.8 \times 10^{-3}$	$5.6 \times 10^{-3}$
<i>Mineral<sup>a</sup></i>	$1.5 \times 10^{-2}$	$4.4 \times 10^{-2}$	$1.3 \times 10^{-1}$	$5.5 \times 10^{-2}$	$4.0 \times 10^{-2}$	$1.9 \times 10^{-2}$	$9.8 \times 10^{-2}$
<i>Organic</i>	$4.3 \times 10^{-2}$	$5.5 \times 10^{-2}$	$2.9 \times 10^{-1}$	$2.1 \times 10^{-1}$	$7.9 \times 10^{-2}$	$3.5 \times 10^{-2\ b}$	$6.7 \times 10^{-2}$

Notes:  
a) Data for sand loam and clay combined.  
b) Value extrapolated using scaling factor for loam.

Table 3: Recommended caesium TF value ( $\text{Cs}_{\text{plant activity d.w.}} / \text{Cs}_{\text{soil activity d.w.}}$ ) (Nisbet *et al*, 1999)

Tsukada and Nakamura (1999) studied the transfer of  $^{137}\text{Cs}$  and stable caesium from soil to potato in agricultural fields in Japan. Transfer factor values (concentration in potato: Bq/kg d.w. for  $^{137}\text{Cs}$  or mg/kg d.w. for caesium / Concentration in soil: Bq/kg d.w. for  $^{137}\text{Cs}$  or mg/kg d.w. for caesium) for  $^{137}\text{Cs}$  and stable caesium for potato were within the ranges 0.0037 – 0.16 and 0.00052 – 0.080 respectively with geometric means of 0.03 for  $^{137}\text{Cs}$  and 0.0075 for stable caesium. The mean geometric TF value for  $^{137}\text{Cs}$  is about four times higher than that of stable caesium implying that artificially added  $^{137}\text{Cs}$  is more mobile and more easily absorbed by plants than stable caesium.

Brambilla *et al* (2002) analyzed the uptake of  $^{134}\text{Cs}$  in processing tomato plants under experimental conditions in Italy. The average soil to fruit TF value (Bq/kg d.w. fruit / Bq/kg d.w. soil) for  $^{134}\text{Cs}$  was  $10 \pm 1.5$  which is nearly two orders of magnitude higher than the value of  $2.2 \times 10^{-1}$  derived for tomato fruit grown on a sandy loam soil (Frissel and van Bergeijk, 1989) and higher than the range  $4.6 \times 10^{-4} - 1.1 \times 10^{-1}$  reported for tomato by Ng *et al* (1982). The authors ascribe the differences to the peat growing substrate whose high organic matter (71.8%) responsible for a large part of the cation exchange capacity reduces  $^{134}\text{Cs}$  fixation on clay mineral leaving it more available for root uptake.

Copplestone *et al* (2001) study the transfer of  $^{137}\text{Cs}$  from soil to vegetation of a sand dune ecosystem receiving a continuing input of entrained radionuclides, principally via sea-to-land transfer. The CRs (Bq/kg d.w. plant / Bq/kg d.w. soil) for *Festuca rubra* (red fescue) and *Ammophila arenaria* (marram grass) ranged between 0.04 and 1.4.

Carini (2001) reviewed radionuclide transfer from soil to fruits and concluded that TF values for soil to fruit transfer of caesium show a wide range covering six orders of magnitude from  $10^{-4}$  to  $10^1$ . Generally speaking the TF values for caesium are higher in tropical rather than in temperate fruit. TF values for fruits of woody trees in temperate regions, grapevine included range from  $8.6 \times 10^{-4}$  to  $8.0 \times 10^{-2}$ . Transfer factor values for fruits of shrubs and herbaceous plants ranged from  $6.9 \times 10^{-4}$  to  $5.7 \times 10^{-3}$  and  $4.1 \times 10^{-4}$  to  $8.9 \times 10^{-3}$  respectively. Transfer factor values for citrus and olives range from  $7.6 \times 10^{-4}$  to  $3.5 \times 10^{-2}$  and TF values for subtropical and tropical fruits ranges from  $1.8 \times 10^{-3}$  to 3.8.

Yu *et al* (1998) assessed the transfer of  $^{137}\text{Cs}$  in three types of vegetables consumed in Hong Kong and derived TF values (Bq/kg d.w. plant / Bq/kg d.w. soil) for lettuce, Chinese white cabbage and celery (mean) of 0.12, 0.19 and 0.11 respectively.

Gerzabek *et al* (1998) studied the plant uptake of  $^{137}\text{Cs}$  in lysimeter experiments using a range of soils. The median TF values (Bq  $\text{kg}^{-1}$  plant d.m. / Bq  $\text{kg}^{-1}$  soil (0-20 cm) d.w.)

for  $^{137}\text{Cs}$  for mustard, potato leaves, potato tubers, maize straw and maize grains were 0.163, 0.156, 0.089, 0.041, 0.005 respectively. The TF value for *V. faba* leaves, *V. faba* beans, rye grass, wheat straw, wheat grains, sugar beet leaves and sugar beet root were 0.098, 0.050, 0.027, 0.021, 0.009, 0.312, and 0.046 respectively.

Livens *et al* (1991) studied the uptake of radiocaesium in a variety of plants and found that transfer to lower plants (bryophytes and lichens) is greater than that to higher plants by virtue of their different modes of nutrient uptake. Among the higher plants at any site the ericaceous plant species showed higher caesium uptake than others.

### 1.10 Summary of Literature

Uranium retention in the soil is increased in the presence of both clay and organic matter and in relation to organic matter the humic acid content plays a significant role in the accumulation of uranium. The redox potential (Eh) is thought to play a major role in the mobility of uranium with increasing accumulation of uranium within the soil as it becomes more reducing. The majority of studies indicate that uptake of uranium by vegetation is dependent on soil type and concentration ratios are lowest for clay soils followed by peat and highest for sandy soil. The distribution of uranium within the plant is inconclusive and is dependent on the individual plant and the nutrient status of the soil. Generally speaking uranium uptake is highest in the roots and leaves and lowest in fruits. The concentration factors (transfer factors) for all vegetation studied are low but vary significantly depending on the plant species and the soil substrate. The majority of concentration ratios are within the order  $10^{-3}$  to  $10^{-4}$  and a summary of the concentration ratios in grass can be seen in Table 4. Studies on uptake of natural radionuclides in bog ecosystems are rare and no CRs for uranium, radium, thorium or potassium in bog vegetation were found in the literature. The chemical composition of both soil and plant plays a significant role in the uptake of uranium with calcium and phosphorous being the elements of greatest importance. While conflicting information has been reported in general, calcium and phosphorous appears to have a significant positive effect on uranium uptake.

The behaviour of radium, like uranium, in soil is dependent on the clay and organic matter contents of the soil. Radium adsorption increases with increasing clay and organic matter. However unlike uranium, some authors conclude that radium is associated with the humin fraction of the organic matter present. The distribution of radium (in particular  $^{226}\text{Ra}$ ) in the soil is not just dependent on the chemistry of radium but also on the soil chemistries of  $^{238}\text{U}$ ,  $^{234}\text{U}$ ,  $^{230}\text{Th}$  and  $^{226}\text{Ra}$ . The translocation of radium within vegetation varies depending on the plant species and the nutrient status of both plant and soil. In general radium is found to be higher in the leaves and stems, than in the roots and lowest in grains and fruit. Concentration ratios for radium vary significantly between studies but in general the CRs are between  $10^{-3}$  and  $10^{-1}$  which is at least one order of magnitude greater than the uptake of uranium by vegetation. A summary of CRs for pasture vegetation can be seen in Table 4.

Thorium is generally considered to be quite immobile in soils with increased adsorption in the presence of clay particles. However, some studies has shown greater thorium mobility if present as organic complexes. The translocation of thorium within the plant varies with plant species and nutrient status of plant and soil but in general thorium accumulation appears to be higher in the roots than the above ground parts of the plant. CRs vary significantly between studies and in general the CRs are between  $10^{-3}$  and  $10^{-4}$ . The CRs for thorium in grass from a number of studies are shown in Table 4. Thorium CRs while lower than uranium in most cases are of the same order of magnitude. CR for both uranium and thorium are at least one order of magnitude lower than that of radium. The effect of chemical or physical properties on thorium uptake by plants is inconclusive from the literature.

The mobility of caesium in soil is highly dependent on the presence of clay. The clay content and more particularly illite clay plays a very significant role in rendering the caesium unavailable to vegetation due to the presence of RES and FES. The ability of organic matter to adsorb caesium is well known, however, the mode of interaction is less well understood than that of clay. Caesium appears to adsorb to organic matter in a non-specific form thus explaining the greater uptake of caesium from organic soils in

comparison to clay soils. Microbial content of the soil is thought to play a significant role in the accumulation of caesium within the upper horizons of the soil. Concentration ratios are generally highest for peat soil followed by sandy soils and lowest for clay soils.

Caesium TF values are affected by chemical status of both soil and plants and in particular by soil potassium, pH and possibly moisture content. Caesium and potassium while chemically similar do not behave in an analogous manner in the soil-plant system and caesium uptake is negatively correlated with potassium. Caesium CR are unaffected by soil pH unless pH drops below 4 – 5 where an increase in CR appears to result from a decrease in pH. Potassium uptake (including  $^{40}\text{K}$ ) in pasture vegetation is higher than caesium (Table 4) possibly due to the clay content of the soil. Caesium CRs are much higher for bog vegetation than pasture as seen in Table 4 and 5 respectively.

Author	CR <sub>dry</sub> for pasture vegetation				
	U 10 <sup>-4</sup>	Th 10 <sup>-4</sup>	Ra 10 <sup>-4</sup>	<sup>40</sup> K	<sup>137</sup> Cs
Linsalata <i>et al</i> (1989)	137 <sup>a</sup> 47 <sup>b</sup>		160-4000		
Ewers <i>et al</i> (2003)	60	30	300		
Pietrzak-Flis <i>et al</i> (1995)	45 <sup>c</sup> 27 <sup>d</sup>	37 <sup>e</sup> 23 <sup>d</sup>			
Moffett and Tellier (1977)	15 <sup>e</sup>	24	290		
Jenkins <i>et al</i> (1990)	105	73			
Vera Tomé <i>et al</i> (2003)	670 – 720 <sup>e</sup>	560 - 580	1700		
Ibrahim <i>et al</i> (1988)			252-1028 <sup>i</sup> 1582 <sup>e</sup>		
Marple (1980)			300 – 900 <sup>i</sup> 200 <sup>e</sup>		
Makrose <i>et al</i> (1993)			66 <sup>e</sup>		
Amaral <i>et al</i> (1988)			700 <sup>a</sup>		
Zararsiz <i>et al</i> (1997)		25 <sup>i</sup> 17 <sup>j</sup>			
Mahon and Mathewes (1983)	75 <sup>k</sup>				
Papastefanou <i>et al</i> (1999) <sup>f</sup>				0.16 – 2.42	0.002 – 7.42
Rafferty <i>et al</i> (1994a)				2 – 6	< 0.25
Strebl <i>et al</i> (2002)				0.31 – 2.01	0.03 – 1.06
Ehlken and Kirchner (1996)					0.01 – 7
IUR (1994)	230	110	800		.11 (clay) .24 (sand) .53 (peat)
Baeza <i>et al</i> (2001)				0.3 <sup>g</sup> 0.25 <sup>h</sup>	0.25 <sup>g</sup> 0.43 <sup>h</sup>
Notes: a) Unwashed grass or pasture b) Washed grass c) Pasture grown outdoors d) Pasture grown in a tent e) Uranium mine tailings f) <sup>134</sup> Cs Concentration ratios	Notes: g) Aerial fraction of pasture vegetation h) Below ground fraction of pasture vegetation i) Contaminated acidic soil j) Contaminated alkaline soil k) Naturally enhanced area l) Control plots				

Table 4: Summary of CR<sub>dry</sub> values for pasture vegetation (grass) from literature

Author	Flowers CR <sub>dry</sub>	Leaves CR <sub>dry</sub>	Stems CR <sub>dry</sub>	Roots CR <sub>dry</sub>
Bunzl and Kracke (1984)	28.7 <sup>a</sup>	35.3 <sup>a</sup>	10.0 <sup>a</sup>	13.3 <sup>a</sup>
Bunzl and Kracke (1986)	7 <sup>b</sup>	6 <sup>b</sup>	0.6 <sup>b</sup>	2 <sup>b</sup>
Notes: a) Sandy soil b) Peat soil				

Table 5: Summary of caesium CR<sub>dry</sub> values for *Calluna vulgaris* from literature.

## 2.0 METHODS

### 2.1 Ecosystem Selection

#### 2.1.1 Atlantic Blanket Bog: Cloonty Bog

The Atlantic blanket bog, located in Ballintrillick (approximately 20 km north-west of Sligo town, Grid Reference G 733 528: OSI Map Discovery Series 16) and is known as Cloonty bog. The site is easily identifiable as an Atlantic blanket bog and is indigenous to this region. The bog consists of both undisturbed peat and cutover bog with the undisturbed region being paramount to this study. The area is only accessible to machinery during the summer months and turf cutting activities in the cutover regions have diminished significantly in the last few years with no commercial turf cutting being carried out at present. The underlying bedrock geology in the area is lower and middle carboniferous, mostly limestone.

The bog covers an area of approximately 12 hectares with the cutover bog accounting for over 90% of this area. The undisturbed peat has an area of approximately 1 hectare and dominant vegetation in this region is a mixture of *Calluna vulgaris* (ling heather), *Eriophorum vaginatum* (bog cotton) and *Sphagnum* mosses. The area is mainly grazed by sheep. This study concerns itself mainly with the undisturbed region of the bog with very limited sampling occurring in the cutover region. The undisturbed region is of greater significance to this study as it shows the fate of radionuclides under natural conditions within this type of ecosystem.

#### 2.1.2 Upland Blanket Bog: Cronamuck Valley.

The upland blanket bog is located in the Cronamuck valley in Donegal along the Atlantic coast of Ireland. The region lies on the north-eastern part of the Barnesmore pluton, an isolated granite in area which forms the Bluestack mountain range in central Donegal

(Grid Ref H 022 928). The dominant vegetation within this upland blanket bog is a mixture of *Calluna vulgaris* (ling heather), *Eriophorum vaginatum* (bog cotton), *Myrica gale* (bog myrtle), *Erica tetralix* (cross-leaved heath), *Molinia caerulea* (purple moor-grass) and *Sphagnum* mosses.

An area of approximately 2 hectares between Clogher Burn and Cronamuck River at the juncture of two unnamed streams was chosen as the sampling site for this ecosystem. The region was chosen as elevated levels of natural radioactivity were found in the soil in the past (Dowdall, 2000). However, soil radioactivity levels within the region varied greatly. The sites were grazed mainly by sheep and the region was undisturbed as it is inaccessible for turf-cutting.

### 2.1.3 Wet Grassland: Doonally

The wet grassland ecosystem is located in Doonally which is approximately 5 km north of Sligo town at Grid reference G725 385 (OSI map Discovery series 16). The site is composed of a field of approximately 0.8 hectares being adjacent to two other fields. The field is inaccessible by machinery being surrounded by a deep depression. The deep depression has prevented fertilizer application in the past and therefore deeming the site suitable for use in the project. Fertilizer application increases the levels of both  $^{238}\text{U}$  and  $^{40}\text{K}$  in the soil and also affects the natural chemical reactions within the soil which is not desirable in this project.

The site is surrounded by fields of similar nature and is classified as a wet grassland habitat. The underlying bedrock geology in the area is lower and middle carboniferous, mostly limestone and the soil is of a loamy texture. The dominant vegetation on the site is a mixture of *Holcus lanatus* (Yorkshire fog), *Juncus effuses* (soft rush), *Urtica dioica* (stinging nettle), *Crataegus monogyna* (hawthorn tree), *Acer pseudoplatanus* (sycamore tree) and moss. The area is grazed by cattle and sheep and otherwise has remained unmodified by man.

## 2.2 Study Methodology

In order to fulfill the main objectives of this study, soil and vegetation was collected at each site for radiometric and chemical analysis. The ecosystems were sampled during the summer months with the Atlantic blanket bog and wet grassland sampled in 2002, and the upland blanket bog sampled in 2005. Vegetation dominant to each ecosystem was sampled. *Calluna vulgaris*, *Eriophorum vaginatum* and species of *Sphagnum* mosses were collected from the Atlantic blanket bog along with the underlying soil. *Calluna vulgaris*, *Eriophorum vaginatum*, *Myrica gale*, *Erica tetralix*, *Molinia caerulea* and *Sphagnum* mosses were collected from upland blanket bog along with the underlying soil. *Holcus lanatus*, *Juncus effuses*, *Urtica dioica*, *Crataegus monogyna*, *Acer pseudoplatanus* and moss were collected from the wet grassland along with the underlying soil.

All samples were analysed for  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$ , the first four being natural radionuclides in the terrestrial environment and the latter anthropogenic, resulting from weapons testing and nuclear releases. Uranium-238 and  $^{226}\text{Ra}$  are representatives of the  $^{238}\text{U}$  decay series, but both behave very differently in the soil-plant system. Radium-228 was used as an indicator for the  $^{232}\text{Th}$  series and, while radium and thorium may behave very differently, its use seems reasonable due to the short half-life of  $^{228}\text{Ra}$  compared to the time spans involved in the pedogenic processes.  $^{40}\text{K}$  was analyzed due to potassium's importance as an essential plant element and therefore the inevitable incorporation of  $^{40}\text{K}$  in the soil-plant system.  $^{137}\text{Cs}$  was analyzed to assess the transfer of this anthropogenic radionuclide within the terrestrial ecosystems. All samples were analyzed for total levels of the selected radionuclides.

A broad range of chemical parameters for both the soil and vegetation were analyzed in order to determine possible reasons for the enrichment or depletion of the radionuclides in the soil or to assess the impact of these parameters on the plant uptake of the radionuclides. Choice of these chemical parameters were informed by the findings of

research from previous literature and in the case of the soil included moisture content, organic matter, pH, cation exchange capacity, and exchangeable nutrients (potassium, calcium, magnesium and sodium). Chemical analysis of the vegetation involved determining the nutrient (calcium, magnesium and sodium) content present.

Transfer factor values were calculated for the bog and grassland vegetation. Transfer factor values in this study was defined as the radioactivity in the dry plant material divided by the average radioactivity in the upper 10 cm soil layer for grassland ecosystems and upper 20 cm soil layer for bog ecosystems (IUR, 1994).

$$\text{Transfer Factor (TF)} = \frac{\text{Activity concentration of nuclide per kg dry plant mass (Bq/kg)}}{\text{Activity concentration of nuclide in dry soil (Bq/kg)}}$$

Correlations were carried out between transfer factors (for each radionuclide) and the chemical parameters of both the soil and vegetation to assess what impact, if any, these parameters have on the uptake of radionuclides by vegetation.

### 2.3 Sampling Rationale

The sampling plan for the three ecosystems was relatively similar with the main focus on vegetation and the soil rooting zone. Soils were taken to a depth of 20 cm below each vegetation sample which is considered adequate for the rooting layer for each site sampled. One deeper layer (0 – 50 cm in bog or 0 – 40 cm in grassland) was taken at the Atlantic blanket bog and grassland in order to obtain a better understanding of the distribution of radionuclides within the soil. Only above ground vegetation was sampled at each ecosystem due to the difficulty in obtaining sufficient quantities of root for analysis. Vegetation was sampled 5 cm above the soil in order to minimize surface contamination present. Henceforth, sampling points within the ecosystems are referred to as sites from here on in and are labelled as outlined in Tables 6 – 8.

*Calluna vulgaris*, *Eriophorum vaginatum* and *Sphagnum* mosses are the dominant vegetation at the Atlantic blanket bog ecosystem and were therefore sampled. Three samples of *Calluna vulgaris*, two samples of *Eriophorum vaginatum* and two samples of *Sphagnum* mosses were collected along with the underlying soil. Vegetation was clipped with hand sheers 5 cm above the ground (minimize soil contamination), placed in sealed plastic bags and transferred to the laboratory. In the case of *Sphagnum* moss, collection of material 5 cm above the ground was not always practical. However, care was taken to minimize soil contamination. Soil samples of 1 m<sup>2</sup> in area to a depth of 20 cm were taken at 6 sites and to a depth of 50 cm at one site as outlined in Table 6. Soils were then sliced into 5 cm layers and analysed separately.

Soil and Vegetation	A1	A2	A3	A4	A5	A6	A7
<i>Calluna vulgaris</i>	√	√	√				
<i>Sphagnum</i> moss				√	√		
<i>Eriophorum vaginatum</i>						√	√
Soil: 0-5 cm	√	√	√	√	√	√	√
Soil: 5-10 cm	√	√	√	√	√	√	√
Soil: 10-15 cm	√	√	√	√	√	√	√
Soil: 15-20 cm	√	√	√	√	√	√	√
Soil: 20-25 cm			√				
Soil: 25-30 cm			√				
Soil: 30-35 cm			√				
Soil: 35-40 cm			√				
Soil: 40-45 cm			√				
Soil: 45-50 cm			√				

Table 6: Sampling sites for undisturbed Atlantic Blanket Bog, Cloonty Bog, Sligo.

*Calluna vulgaris* (ling heather), *Eriophorum vaginatum* (bog cotton), *Myrica gale* (bog myrtle), *Erica tetralix* (cross-leaved heath), *Molinia caerulea* (purple moor-grass) and

*Sphagnum* mosses were the dominant vegetation in the upland blanket bog and were therefore sampled at eight sites. Soil and vegetation were collected in a similar manner as outlined for the Atlantic blanket bog. Sites D1 – 4 and D 6 - 7 were all located within a closely defined area of approximately 1 km<sup>2</sup> and were in close proximity to Cronamuck River. Sites D 5 and D 8 were further away from the river and on higher ground. Soil and vegetation sampled at each site are outlined in Table 7

Soil & Vegetation	D1	D2	D3	D4	D5	D6	D7	D8
<i>Calluna vulgaris</i>		√		√	√		√	√
<i>Myrica gale</i>	√	√	√			√	√	√
<i>Erica tetralix</i>			√	√		√	√	√
<i>Eriophorum vaginatum</i>					√	√	√	
<i>Molinia caerulea</i>	√	√	√	√	√	√	√	
<i>Sphagnum</i> mosses	√		√			√	√	√
Soil: 0-5 cm	√	√	√	√	√	√	√	√
Soil: 5-10 cm	√	√	√	√	√	√	√	√
Soil: 10-20 cm	√	√	√	√	√	√	√	√

Table 7: Sampling sites for undisturbed Upland Blanket Bog, Cronamuck Valley, Donegal.

*Holcus lanatus*, *Juncus effuses*, *Urtica dioica*, *Crataegus monogyna*, *Acer pseudoplatanus* and *S.* moss were collected from the wet grassland along with the underlying soil. The wet grassland of interest consisted of approximately 0.8 hectares and 11 sites were sampled at random within this area. These sites are referred to in the text as Sites G and outlined in Table 8. Vegetation was clipped with hand sheers 5 cm above the ground, placed in sealed plastic bags and transferred to the laboratory. Soil samples of 1m<sup>2</sup> to a depth of 20 cm were taken at ten sites. One additional site was sampled to a depth of 40 cm (50 cm was not possible due to the presence of gravel and rock below 40 cm) as outlined in Table 8. Soils were then sliced into 5 cm layers and analyzed separately.

Soil & Vegetation	G1	G2	G3	G4	G5	G6	G7	G8	G9	G10	G11
<i>Holcus lanatus</i>	√	√	√	√	√	√	√	√			
<i>Juncus effuses</i>		√		√	√				√	√	√
<i>Urtica dioica</i>			√	√							
<i>Crataegus monogyna</i>	√										
<i>Acer pseudoplatanus</i>		√									
Moss		√				√					
Soil: 0-5 cm	√	√	√	√	√	√	√	√	√	√	√
Soil: 5-10 cm	√	√	√	√	√	√	√	√	√	√	√
Soil: 10-15 cm	√	√	√	√	√	√	√	√	√	√	√
Soil: 15-20 cm	√	√	√	√	√	√	√	√	√	√	√
Soil: 20-25 cm											√
Soil: 25-30 cm											√
Soil: 30-35 cm											√
Soil: 35-40 cm											√

Table 8: Sampling sites for Wet Grassland, Doonally, Sligo.

## 2.4 Sample Preparation

Samples were collected in the field and placed in sealed plastic bags in order to prevent contamination and minimize moisture loss and were then brought to the laboratory for radiochemical and chemical analysis. Soil moisture content and pH measurements were carried out on the wet soil and in all other cases pre-treatment as outlined below was required before any further chemical or radiometric analyses were carried out.

### 2.4.1 Soil

Samples were brought to the laboratory and dead plant material and rocks were removed. Soil was then dried at 35 °C (exchangeable nutrients unaffected by temperatures < 40°C)

until a constant mass was achieved (Tan, 1996), and then ground in a blender before being passed through a 2 mm sieve.

The soil was then packed into 1L marinelli beakers, sealed and analyzed for radionuclides by gamma spectrometry. Portions of the samples from the marinelli were removed for chemical analysis after the radiometric analysis.

#### **2.4.2 Vegetation**

Samples of vegetation which were collected at each site at approximately 5 cm above the ground were washed in distilled water in order to remove any debris or soil and dried immediately at 100 °C until a constant mass was achieved. The samples were then ground in a blender before being passed through a 2 mm sieve. *Calluna vulgaris* and *Myrica gale* were treated in a slightly different way because the leaves and stems had been separated before the washing and drying stages and these portions were analysed separately.

Samples were then packed into 200 ml tubs and sealed before radiometric analysis (gamma spectrometry) was carried out. Sub-samples from the tub were removed for chemical analysis after radiometric analyses. Vegetation samples were also analyzed for  $^{238}\text{U}$  and  $^{232}\text{Th}$  content by alpha spectrometry and ICP-MS using the methods outlined in Sections 2.5.2 and 2.5.3.

#### **2.5 Analytical Methods**

The main objective of determining radionuclides distribution in soil and plant systems was achieved via gamma spectrometry, alpha spectrometry and ICP-MS. Chemical analysis of soil and vegetation employed various standard techniques as outlined below.

## 2.5.1 Gamma Spectrometry

The primary radioanalytical technique employed throughout this project was high resolution gamma ray spectrometry. The system employed consisted of a high purity germanium detector coupled to an 8 k multi-channel analyzer. The software used for this analysis was GENIE-PC suite.

### 2.5.1.1 Germanium Detector Calibration

The detector was calibrated using a mixed standard containing radionuclides emitting gamma rays in the energy range 59.54 – 1332.49 keV. Calibrations using both the marinelli (1L) beaker and the 0.2 L tub were carried out. The calibration solution consisted of traceable radionuclides ( $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ ,  $^{139}\text{Ce}$ ,  $^{241}\text{Am}$ ,  $^{57}\text{Co}$  and  $^{109}\text{Cd}$ ) obtained from Physikalisch-Technische Bundesanstalt (Appendix 2) were mixed together and made up to volume in a 1L volumetric flask using 0.1M HCl as a diluent. The activities and gamma emission characteristic of the radionuclides used for the marinelli and tub calibrations are shown in Table 9.

Radionuclide	Half-life (days)	Energy (keV)	Emission Rate GPS/L
$^{241}\text{Am}$	$1.58 \times 10^3$	59.54	12.2
$^{109}\text{Cd}$	$4.62 \times 10^2$	88.03	4.1
$^{57}\text{Co}$	$2.71 \times 10^2$	122.06	76.3
		136.47	9.5
$^{139}\text{Ce}$	$1.38 \times 10^2$	165.86	66.3
$^{137}\text{Cs}$	$1.10 \times 10^4$	661.60	16.4
$^{60}\text{Co}$	$1.93 \times 10^3$	1173.23	28.6
		1332.48	28.6

Table 9: Calibration Solution for Gamma Detector.

The marinelli and tub calibration solutions were counted for a period long enough to ensure 10,000 counts after background correction in the smallest peak present. This ensured a 2 % error at a 95 % confidence level based on the following equation.

$$\% \sigma = (2/\sqrt{n}) \times 100$$

%  $\sigma$  = % error  
 n = no. of counts

The calibration used aqueous solution as outlined above and the density factors of soil and vegetation were corrected for by using density correction software (AEA Technology, 1998)

Efficiency and energy calibrations were checked using a range of soil and vegetation reference materials obtained from the IAEA. Table 10 outlines the soil and vegetation reference materials used (reference sheets for each reference material are shown in Appendix 1) and the radionuclides assessed in each of the reference materials. Efficiency and energy calibration were checked initially using these materials and periodically throughout the project and results were typically within the 95% confidence interval quoted on the reference sheets (Appendix 1).

Radionuclide	Radionuclides in Soil			Radionuclide in Clover
	IAEA-326	IAEA-375	IAEA-Soil-6	IAEA-156
<sup>238</sup> U	√			
<sup>232</sup> Th	√	√		
<sup>228</sup> Ra	√			
<sup>226</sup> Ra	√	√	√	
<sup>40</sup> K	√	√		√
<sup>137</sup> Cs		√	√	√

Table 10: Soil and Vegetation Reference Standards

### 2.5.1.2 Isotope Analysis

Analyses of  $^{238}\text{U}$  and  $^{232}\text{Th}$  were achieved via the characteristic emissions of daughters in secular equilibrium with them. Radium-226 was analysed using its characteristic gamma emission or via the emissions of its daughter nuclides. Potassium-40 and  $^{137}\text{Cs}$  were analysed via their characteristic strong well defined gamma emissions.

#### 2.5.1.2.1 $^{238}\text{U}$ Analysis

Direct determination of  $^{238}\text{U}$  by gamma spectrometry is impractical as the  $^{238}\text{U}$  isotope emits only a very weak gamma ray at 49.55 keV (0.064%). Therefore, quantification of  $^{238}\text{U}$  by gamma radiation is generally done by analyzing some of its daughter nuclides which have sufficiently strong gamma emissions, i.e.  $^{234}\text{Th}$  and  $^{234\text{m}}\text{Pa}$ . Uranium-235 activity may also be used to infer  $^{238}\text{U}$  activity. In order to determine  $^{238}\text{U}$  activity using the daughter nuclides it is essential that the daughter nuclide exists in secular equilibrium with the parent. This condition is fulfilled by any one of the first three daughters of  $^{238}\text{U}$ , i.e.  $^{234}\text{Th}$  (24.1 d),  $^{234\text{m}}\text{Pa}$  (1.17 m) and  $^{234}\text{Pa}$  (6.78 h) as each daughter has a relatively short half-lives in comparison to the parent. Problems arise with the use of each of the daughters for quantification.

Thorium-234 as mentioned above, is generally accepted to be in secular equilibrium in environmental samples with  $^{238}\text{U}$  having a half life of 24.1 days. Thorium-234 has two emissions, a singlet peak at 63 keV and a doublet peak at 92 keV. Problems arise from the doublet peak as samples with high levels of thorium may produce thorium (K) X-rays at 93 keV. Also, the 63 keV peak may be affected by  $^{231}\text{Th}$  (emission probability 0.023%) and  $^{232}\text{Th}$  (emission probability 0.255%) peaks. However, as  $^{235}\text{U}$  ( $^{231}\text{Th}$  parent) levels in environmental samples are generally very low then the contributions to the 63 keV peak are generally negligible.

The  $^{234\text{m}}\text{Pa}$  isotope is in secular equilibrium with its parent  $^{238}\text{U}$  in environmental samples having a half life of 1.17 minutes. This peak (1001 keV) is relatively free of interferences

from other peaks in the uranium spectrum even if samples contain high contents of thorium. The results obtained for this peak do not need any self-absorption corrections as it is a high-energy gamma ray. However, some confusion remains over the actual emission probability of the gamma ray. Korob and Blasiyh Nuño (2006) and Nzuruba (1999) published emission probabilities of  $0.0084 \pm 0.0001$  and  $0.00835 \pm 0.00004$  respectively. A major disadvantage of this peak is its low emission probability resulting in longer counting times.

Radium-226 is not necessarily in secular equilibrium with its parent due to its long half-life (1600 years) so therefore it cannot be directly used to determine  $^{238}\text{U}$  activities in environmental samples. Uranium-235 activity may be used to infer  $^{238}\text{U}$  activity. However, the emissions of  $^{235}\text{U}$  and  $^{226}\text{Ra}$  at 185.7 keV and 186.1 keV cannot be resolved by conventional gamma ray spectrometers. In environmental samples the contribution of  $^{226}\text{Ra}$  may be determined by hermetically sealing the sample container for a period of time (> 30 days) sufficient to allow secular equilibrium between  $^{226}\text{Ra}$  and its daughters,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$ .

The procedure adopted in this project was to quantify the  $^{238}\text{U}$  activity of the samples via the  $^{234}\text{Th}$  emissions.

#### 2.5.1.2.2 $^{226}\text{Ra}$ Analysis

Radium-226 has its own gamma emission at 186 keV but, as mentioned previously, the emissions of  $^{235}\text{U}$  and  $^{226}\text{Ra}$  cannot be resolved using conventional spectrometry.

Radium-226 activity in wet grassland soils was determined using its own peak at 186 keV and the contributions of  $^{235}\text{U}$  to the  $^{226}\text{Ra}$  peak were determined using the  $^{238}\text{U}/^{235}\text{U}$  activity ratio. In the case of peat and vegetation samples  $^{226}\text{Ra}$  activities were determined using the  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  peaks. Samples were sealed in marinelli beakers (peat) and in 200 ml tubs (vegetation) for in excess of 30 days in order to allow secular equilibrium to

occur. The  $^{226}\text{Ra}$  activity was then determined via the emissions of  $^{214}\text{Pb}$  (352 keV) and  $^{214}\text{Bi}$  (609 keV).

The ability of the marinelli beaker and the 200 ml tub to retain radon gas and allow secular equilibrium to occur between  $^{226}\text{Ra}$  and its daughters  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  was tested using a  $^{226}\text{Ra}$  standard solution and two internationally traceable soil reference materials: IAEA 326 and IAEA Soil 6 (Reference sheets, Appendix 1).

A solution of  $34.4 \text{ Bq L}^{-1}$  (degassed for 1 hour in an ultra sonic bath) was transferred to a 1L marinelli beaker which was sealed with thick PVC insulation tape. Lead-214 and  $^{214}\text{Bi}$  activities were recorded over a 40 day period. A solution of  $172 \text{ Bq L}^{-1}$  was prepared and transferred to the 0.2 L tub (previously degassed as above) and then was sealed with thick insulation tape. Lead-214 and  $^{214}\text{Bi}$  activities were analysed over a 40 day period. Figures 9 and 10 show the activities of  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  in the  $34.4 \text{ Bq L}^{-1}$  marinelli beakers and  $172 \text{ Bq L}^{-1}$  200 ml tub respectively.

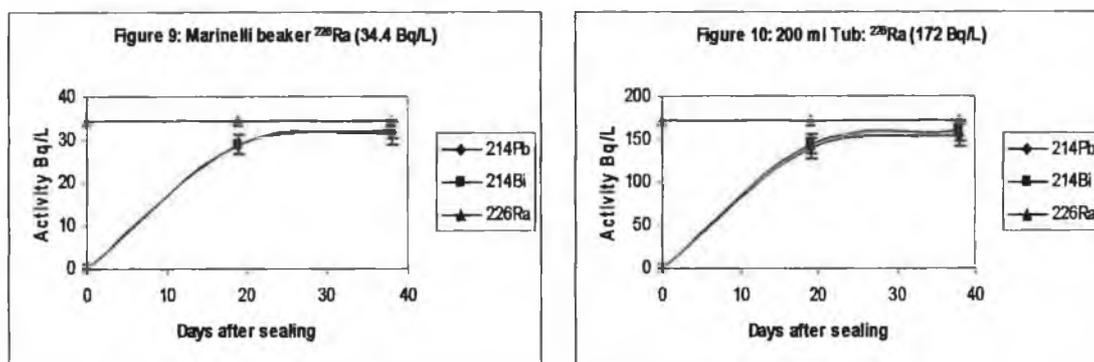
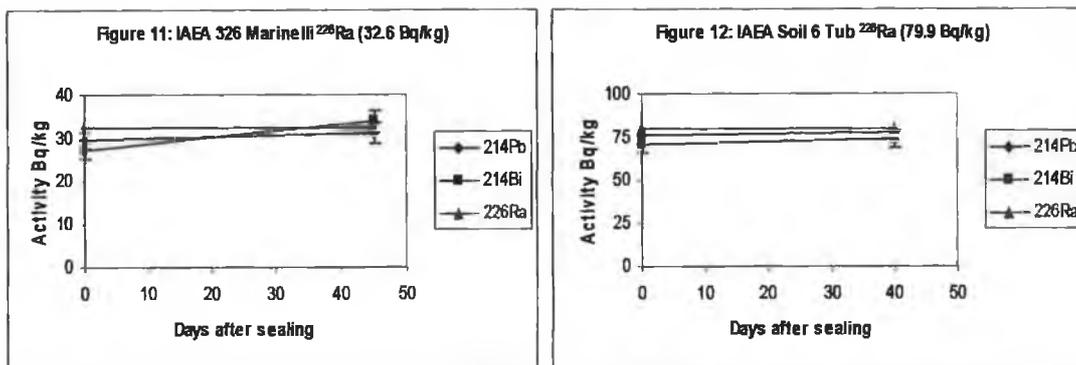


Figure 9 & 10: Plots of  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  activity in radium solution versus time for Marinelli beaker and 200 ml tub respectively.

Reference soil materials (IAEA 326 and IAEA Soil 6, Appendix 1) were packed into a 1L marinelli beaker and a 0.2 L tub and sealed with thick PVC insulation tape. Lead-214 and  $^{214}\text{Bi}$  activities were recorded over a 40 day period and radioactive equilibrium (> 90%) was achieved within a 40 day period as can be seen in Figures 11 and 12 below.



Figures 11 and 12: Plots of  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  activity versus time for soil reference material IAEA 326 and IAEA Soil 6 respectively.

Radioactive equilibrium (> 90%) was achieved in both containers for the radium solution and the soil within a 40 day period. This illustrates that the marinelli beaker and tub are suitable containers to use for radium analysis by  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  as secular equilibrium was established within a forty day period.

#### 2.5.1.2.3 $^{232}\text{Th}$ Analysis

Thorium-232 does not possess a gamma emission so therefore, it cannot be directly analyzed using gamma spectrometry. Radium-228 results are used as an indication of  $^{232}\text{Th}$  as  $^{228}\text{Ra}$  has a half-life of 6.7 years and is relatively immobile in the surficial environment. Therefore, it is possible to assume equilibrium between  $^{232}\text{Th}$  and  $^{228}\text{Ra}$ .  $^{228}\text{Ac}$  (half-life 6.13 hours) is in secular equilibrium with its parent  $^{228}\text{Ra}$  and the  $^{228}\text{Ac}$  peaks at 911 keV and 968 keV were used for analysis.

#### 2.5.1.2.4 $^{137}\text{Cs}$ Analysis

Determination of  $^{137}\text{Cs}$  was relatively straightforward via its well defined characteristic peak at 661 keV.

### 2.5.1.2.5 <sup>40</sup>K Analysis

Determination of <sup>40</sup>K was relatively straightforward via its well defined characteristic peak at 1460 keV.

### 2.5.1.3 Nuclear Data

The nuclear data (gamma energies, half lives, and emission probabilities) used in this study are shown in the Table 11.

Decay Series	Isotope	Half-life (years)	Principal $\gamma$ emissions (keV)	
			$\gamma$ emission	Emission prob.
Uranium - 238	<sup>234</sup> Th	$6.6 \times 10^{-2}$	63.3	4.80 %
			92.4/92.8	5.60 %
Uranium - 238	<sup>226</sup> Ra	$1.6 \times 10^3$	186.2	3.59 %
Uranium - 238	<sup>214</sup> Pb	$5.1 \times 10^{-3}$	351	37.60 %
Uranium - 238	<sup>214</sup> Bi	$3.79 \times 10^{-3}$	609.3	46.10 %
Uranium - 235	<sup>235</sup> U	$7.04 \times 10^8$	185.7	57.20 %
			205.3	5.01 %
Thorium - 232	<sup>228</sup> Ac	$7.02 \times 10^{-4}$	911.2	25.80 %
			968.9	15.80 %
	<sup>40</sup> K	$1.28 \times 10^9$	1461	10.67 %
	<sup>137</sup> Cs	$3.00 \times 10^4$	661.6	85.10 %

Table 11: Nuclear Data for  $\gamma$  emitting isotopes.

### 2.5.1.4 Counting

Samples were counted for a period sufficient to ensure a  $2\sigma$  error of less than 10% (15% for <sup>226</sup>Ra) after correction for laboratory background. Counting times varied from between 1 day and 2 weeks. Nuclides were identified using a library driven search routine and quantitative analyses were carried using the appropriate detector calibration

files (based on container specifications i.e. marinelli beaker or tub). Density correction of samples was made using Gamatool density correction software (version 2.1).

## 2.5.2 Alpha Spectrometry

The concentrations of  $^{238}\text{U}$  and  $^{232}\text{Th}$  in vegetation were below the limits of detection for gamma spectrometry using the methods outlined in Sections 2.5.1.2.1 and 2.5.1.2.3. Their levels in vegetation were determined by alpha spectrometry using the methods outlined in Sections 2.5.2.2 – 2.5.2.5.

The system employed consisted of an alpha analyst integrated alpha spectrometer with which consisted of two Passivated Implanted Planar Silicon (PIPS) detectors. The software package used for analysis was GENIE-2000 alpha acquisition and analysis software suite (Canberra Industries, 2001).

### 2.5.2.1 Alpha Spectrometry Detector Calibration

The detector was calibrated using a mixed alpha standard source which contained four alpha emitting radionuclides within the energy range 4100 – 5500 keV plated on a stainless steel disc. The plated disc contained a mixture of  $^{238}\text{U}$ ,  $^{234}\text{U}$ ,  $^{239}\text{Pu}$  and  $^{241}\text{Am}$  with known activity and was obtained from Analytix Inc. (Reference sheets, Appendix 2). The source was used for both the energy and efficiency calibration of the detector. The activities and alpha emissions of the radionuclides used for the calibration are shown in Table 12.

Radionuclide	Half-life (days)	Energy (keV)	Emission Rate GPS/Unit
$^{238}\text{U}$	$1.63 \times 10^{12}$	4184.4	1.7
$^{234}\text{U}$	$8.95 \times 10^7$	4761.5	1.7
$^{239}\text{Pu}$	$8.81 \times 10^6$	5147.7	1.7
$^{241}\text{Am}$	$1.58 \times 10^5$	5479.1	1.7

Table 12: Mixed Alpha Calibration Source

Efficiency and energy calibrations were checked using a range of soil reference materials obtained from the IAEA. Table 10 outlines the soil and vegetation reference materials used (Reference Sheets for each reference material are shown in Appendix 1) and the radionuclides assessed in each of the reference materials. Efficiency and energy calibration were checked initially using these materials and periodically throughout the project and results were typically within the 95% confidence interval quoted on the reference sheets (Appendix 1).

#### **2.5.2.2 Sample Preparation for Alpha Spectrometry.**

Sample preparation for alpha spectrometry was carried out using a method similar to León Vintó and Mitchell (2000). A known quantity (approx. 10 g) of dry vegetation was weighed into a crucible and ignited in a muffle furnace for a period of 24 hours while increasing the temperature from 50°C to 500°C by 50°C increments at regular time intervals. The resultant cooled ash was transferred into a beaker with 100 ml of aqua regia and 1 ml of both  $^{232}\text{U}$  and  $^{229}\text{Th}$ , uranium and thorium tracers (Reference Sheets, Appendix 2). The beaker was covered with a clock glass and digested for a period of 5 – 6 hours at a temperature of 200°C. The solution was then filtered through a Whatman filter paper no. 1. The filtrate was evaporated to dryness and converted to nitrate form by three successive additions of conc.  $\text{HNO}_3$ . The remaining residue was then re-dissolved in 70 ml of 8M  $\text{HNO}_3$ .

#### **2.5.2.3 Extraction Chromatography: Separation of $^{238}\text{U}$ and $^{232}\text{Th}$**

The digested sample was filtered through a 0.45  $\mu\text{m}$  filter and passes through an anion exchange column (DOWEX column, AG 1 x 8) which was previously conditioned with 50 ml of  $\text{HNO}_3$ . The solution was passed through a column at a rate of 1 - 2 ml/min. Uranium, iron and americium if present in the sample passed through the column as they do not form nitrate complexes. Thorium, plutonium and neptunium were retained due to their formation of nitrate complexes. The uranium, iron and americium fraction was collected for further purification. The thorium fraction was eluted off the column by

passing 70 ml of 12M HCL through the column. Thorium raffinate was then evaporated to dryness and converted to its chloride form by three successive additions of conc. HCL. The remaining residue was dissolved in 1: 8 ratio of 3M HCL and 1M ammonium chloride/.01M ammonium oxalate for electrodeposition.

Uranium, iron and americium raffinate from the first column was evaporated to dryness and dissolved in 9M HCL. The solution was passed through a second anion exchange column (DOWEX column conditioned with 9M HCL) at a rate of 1 – 2 ml/min to separate and purify uranium, and eliminate iron. Adsorbed iron and uranium are stripped from the column by the passage of 8M HNO<sub>3</sub> through the column and iron if present will elute off the column before uranium. In samples with high iron content, 8M HNO<sub>3</sub> was passed through the column until the eluate from the column was colourless indicating the elimination of most of the iron. Uranium raffinate was collected once the raffinate becomes clear and was purified to remove any residual iron by solvent extraction with isopropyl ether. Uranium raffinate was evaporated to dryness and converted to its chloride form and dissolved in 1: 8 ratio of 3M HCL and 1M ammonium chloride/.01M ammonium oxalate for electrodeposition.

#### **2.5.2.4 Electrodeposition**

The thorium and uranium samples were dissolved in 1: 8 ratio of 3M HCL and 1M ammonium chloride/.01M ammonium oxalate after conversion to their chloride form. Thymol blue indicator (2 – 3 drops) was added to the solution and the pH of the solution was adjusted to pH 2.0 using 1:1 (w/w) ammonia. The resultant solution was then electroplated onto a stainless steel disc in an electrolytic cell for a period of 45 – 60 minutes by the passage of a current of 1.0 Amp.

#### **2.5.2.5 Isotopic and Spectral Analysis.**

The analysis of uranium using <sup>232</sup>U as a monitor is straightforward in that the spectral peaks of <sup>238</sup>U, <sup>235</sup>U, <sup>234</sup>U and <sup>232</sup>U are all well resolved from one another. Sources should

be counted soon after sample purification, as short lived daughter nuclides ingrow quickly, yielding peaks some of which cannot be resolved from other peaks. In the case of the  $^{232}\text{U}$  tracers its daughter nuclide  $^{228}\text{Th}$  ingrows and  $^{228}\text{Th}$  peaks at 5340 keV (27 %) overlaps with  $^{232}\text{U}$  peak at 5320 keV (69 %). However, interference can be resolved by determining the  $^{228}\text{Th}$  contribution to  $^{232}\text{U}$  peaks by analyzing  $^{228}\text{Th}$  at the 5423 keV (73 %) peak.

The thorium spectra with the use of  $^{229}\text{Th}$  as a tracer is relatively uncomplicated. Spectral interferences arise from  $^{224}\text{Ra}$  in-growth from  $^{228}\text{Th}$ , as the  $^{224}\text{Ra}$  peak at 5449 keV (5 %) interferes with the  $^{228}\text{Th}$  peak at 5423 keV (73 %) and 5340 keV (27 %). This can be rectified by measuring the  $^{224}\text{Ra}$  peak at 5868 keV (95 %) and estimating the contribution from  $^{224}\text{Ra}$  peak at 5449 keV (5 %) in the  $^{228}\text{Th}$  window on the basis of the relative intensities of the two  $^{224}\text{Ra}$  peaks.

In the case of a mixed  $^{232}\text{U}/^{229}\text{Th}$  tracer, the contribution of  $^{228}\text{Th}$  from  $^{232}\text{U}$  tracers should be taken into consideration when measuring the  $^{228}\text{Th}$  levels in sample. The nuclear data for principal alpha emissions of uranium and thorium isotopes along with  $^{232}\text{U}$  and  $^{229}\text{Th}$  tracers are outlined in Table 13.

The radiochemical recovery for the tracers used was within the region of 60 – 80 % recovery for the methods utilised.

Decay Series	Isotope	Half-life (years)	Principal $\alpha$ emissions (keV)	
			$\alpha$ emission	Emission prob.
Uranium - 238	$^{238}\text{U}$	$4.47 \times 10^9$	4198	77 %
			4149	23 %
	$^{234}\text{U}$	$2.45 \times 10^5$	4773	72 %
			4721	27 %
	$^{230}\text{Th}$	$7.54 \times 10^4$	4688	76 %
			4621	23 %
	$^{226}\text{Ra}$	$1.60 \times 10^3$	4785	94 %
4602			6 %	
$^{222}\text{Rn}$	$1.05 \times 10^{-2}$	5490	100 %	
$^{218}\text{Po}$	$5.91 \times 10^{-6}$	6003	100 %	
$^{210}\text{Po}$	$3.79 \times 10^{-1}$	5297	100 %	
Uranium-232 Tracer	$^{232}\text{U}$	$72.0 \times 10^0$	5320	69 %
			5264	31 %
Thorium - 232	$^{232}\text{Th}$	$1.41 \times 10^{10}$	4010	77 %
			3952	23 %
	$^{228}\text{Th}$	$1.91 \times 10^0$	5423	73 %
			5340	27 %
	$^{224}\text{Ra}$	$1.00 \times 10^{-2}$	5686	95 %
			5449	5 %
$^{220}\text{Rn}$	$1.76 \times 10^{-6}$	6288	100 %	
$^{216}\text{Po}$	$4.75 \times 10^{-9}$	6779	100 %	
Thorium - 229 Tracer	$^{229}\text{Th}$	$7.34 \times 10^3$	6051	25 %
			6090	10 %
Thorium - 229 Tracer	$^{229}\text{Th}$	$7.34 \times 10^3$	4798	1.3 %
			4815	9.3 %
			4837	4.8 %
			4845	52 %
			4901	10.2 %
			4968	5.97 %
			4979	3.17 %
			5050	5.2 %
5052	1.6 %			

Table 13: Characteristics of the principal  $\alpha$  emitting isotopes of  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay series and tracers.

### **2.5.3 Inductively Coupled Plasma – Mass Spectrometry(ICP-MS): Sample preparation**

The concentration of  $^{238}\text{U}$  in vegetation was below the limits of detection for gamma spectrometry using the methods outlined in Sections 2.5.1.2.1 - 2.5.1.2.3 and in most cases below the limits of detection for alpha spectrometry using the methods outlined in Sections 2.5.2.1 – 2.5.2.5. The levels were determined by ICP-MS.

Sample preparation of vegetation for analysis by ICP-MS was via a published by León Vitró and Mitchell (2000). A known quantity (approx. 10 g) of dry vegetation was weighed in a crucible and ignited in a muffle furnace for a period of 24 hours while increasing the temperature from 50°C to 500°C by 50°C increments at regular time intervals. The resultant cooled ash was transferred into a beaker then covered with a clock glass and digested for 5 – 6 hours with repeated additions of conc. nitric acid, conc. hydrochloric acid and 30% hydrogen peroxide. The solution was then filtered through a Whatman filter paper no. 1 and diluted to a final volume of 100 ml. The sample was analysed for uranium by ICP-MS courtesy of Bord-na-Mona (accredited laboratory). Instrumental accuracy was ensured by the use of soil reference material (Table10) and the results obtained were typically within the 95% confidence interval quoted on the reference sheets (Appendix 1).

### **2.5.4 Chemical Analysis**

A range of chemical analysis was carried out on both the soil and vegetation during the course of this study, and the methods utilised are outlined below. All chemical analysis was carried out in triplicate and the errors associated with the chemical results presented in Section 3.0 of this thesis were lower than a 5% error based on % RSD.

#### **2.5.4.1 Soil pH**

Soil pH was recorded on samples at soil-to-water ratios of 1:2 (w: v) using a method reported by Mc Lean (1982). Approximately 5 g of sieved and air-dried soil was placed

in a beaker and 10 ml of distilled water was added. The contents were mixed vigorously on a mechanical stirrer for 30 seconds and allowed stand for 20 minutes. The pH was read on a calibrated pH meter (Orion Model 210A), whose electrodes were placed in the carefully stirred sample.

#### **2.5.4.2 Moisture Content**

Approximately 10- 15 g of soil (fresh) was weighed and dried at 110°C for 24 hours. Samples were then cooled for 1 hour and reweighed and the loss in mass corresponded to the moisture content.

#### **2.5.4.3 Organic Matter**

Organic matter was determined via loss on ignition for four hours. Approximately 10 g of dried soil (< 2 mm particle size) was placed in a crucible (conditioned at 900°C for 15 minutes and cooled in a desiccator) and heated in a muffle furnace at 550°C for four hours. The crucible was then cooled in a desiccator and weighed. Organic matter was calculated on a % basis from the formula below:

$$\% \text{ Organic Matter} = \frac{(A - B)}{M} \times \frac{100}{1}$$

A = Mass of crucible and soil before ignition (grams)

B = Mass of crucible and soil after ignition (grams)

M = Mass of soil (grams)

#### 2.5.4.4 Cation Exchange Capacity

Cation exchange capacity was measured using a variation of the sodium saturation method (Rhoades, 1982). Approximately 4.0 g of air dried soil (< 2 mm particle size) was placed in a round bottomed centrifuge tube. 33 ml of 1.0 N sodium acetate (NaOAc) solution (pH 7.5) was added and shaken mechanically shaker for 5 minutes to remove adsorbed cations. The suspension was then centrifuged at 10 krpm for 10 minutes and the supernatant was discarded. This procedure was repeated with three successive aliquots of ammonium acetate. Three 33 ml aliquots of propan-2-ol were then used to wash the soil and the same centrifuge procedure was used. Finally, three 33 ml aliquots of 1.0 M ammonium acetate (NH<sub>4</sub>OAc) were used to desorb the adsorbed sodium and the supernatants were collected. The supernatant was then filtered through a glass fibre filter paper and a 0.45 µm filter, combined and made up to volume with ultra pure water to 100 ml in a volumetric flask. The sodium content of the supernatant was analyzed using flame photometry and results were expressed as meq/100 g of soil.

#### 2.5.4.5 Exchangeable Calcium, Potassium, Sodium and Magnesium levels in Soil

Exchangeable cations (Ca<sup>2+</sup>, K<sup>+</sup>, Na<sup>+</sup> and Mg<sup>2+</sup>) were measured using an ammonium acetate extraction method of Knudsen *et al* (1982). Approximately 10 g of air dried soil (< 2 mm particle size) was placed in a 150 ml Erlenmeyer flask with 40 ml of 1N ammonium acetate (NH<sub>4</sub>OAc) and mixed mechanically for 30 minutes. The suspension was then pre-filtered through a glass fibre filter paper (Whatman no. 40) and then filtered through a 0.45 µm filter paper. The filtrate was then transferred to a 100 ml volumetric flask and made up to volume with 1N NH<sub>4</sub>OAc.

Potassium and sodium concentrations were analysed by flame emission photometry using a Corning 400 flame photometer. Calibration curves were prepared for both potassium

and sodium in the range 0 – 20 mg L<sup>-1</sup> and concentrations determined by extrapolation of emissions recorded.

Calcium and magnesium concentrations were determined by atomic absorption using a Perkin Elmer 2380 Atomic Absorption Spectrometer with an air/acetylene flame.

Calibration curves for calcium and magnesium were prepared in the ranges 0- 20 mg L<sup>-1</sup> and 0 – 1 mg L<sup>-1</sup> respectively. Lanthanum (1% w/v) was added to both sample and standards for both calcium and magnesium to eliminate chemical interferences.

#### **2.5.4.6 Total Calcium, Potassium, Sodium and Magnesium levels in Vegetation**

A known amount (approx. 1 g) of air dried soil (< 2mm particle size) was placed in a test-tube with 10 ml of conc. HNO<sub>3</sub>, covered by a watch glass and heated at 95°C for 3 – 4 hours. 10 ml of 30% H<sub>2</sub>O<sub>2</sub> was cautiously added in 1 ml aliquots and care was taken to ensure no sample loss during excessive effervescence of sample. The solution was heated at 95°C for a further 10 minutes and allowed to cool. The solution was then pre-filtered through a glass fibre filter paper and then filtered through a 0.45 µm filter. The filtrate was then transferred to a 100 ml volumetric flask and made up to volume with ultra pure water (USEPA, 1996).

Potassium and sodium concentrations were analyzed by flame emissions photometry using a Corning 400 flame photometer. Calibration curves were prepared for both potassium and sodium in the range 0 – 20 mg L<sup>-1</sup> and concentrations determined by extrapolation of emissions recorded.

Calcium and magnesium concentrations were recorded by atomic absorption using a Perkin Elmer 2380 Atomic Absorption Spectrometer with an air/acetylene flame.

Calibration curves for calcium and magnesium were prepared in the ranges 0- 20 mg L<sup>-1</sup> and 0 – 1 mg L<sup>-1</sup> respectively. Lanthanum (1% w/v) was added to both sample and standards for both calcium and magnesium to eliminate chemical interferences.

## **2.6 Statistical Analysis**

Statistical analysis was carried out on the results by the methods outlined below using Sigma Stat 2.0 for Windows software.

### **2.6.1 t-test**

The t-test is commonly used to evaluate if the differences in means between two groups is greater than what can be attributed to random sampling variation. The t-test is used on normally distributed sets of data and unpaired t-tests are used to test whether two populations have the same mean. Differences in mean values are regarded significant if  $p < 0.05$ , (Townsend, 2002).

### **2.6.2 One Way Analysis of Variance**

Analysis of variance (ANOVA) is used when differences between the means of several populations is required. Tukey comparisons are used to identify if the mean value of each population is significantly different from each other. Differences in mean values are regarded significant if  $p < 0.05$ . One-way ANOVA is like an extension of the t-test. While the t-test compares the mean of two populations one-way ANOVA compares the mean of three or more populations (Townsend, 2002).

### **2.6.3 Correlations**

Correlations and regressions are used to study the relationship between two types of measurements made on the same individuals. The term correlation is usually used to refer to Pearson's product moment correlation (other types exist though they are usually referred to by the full name to distinguish them from Pearson's product moments correlation). Pearson's product moment correlation gives a measure of how close the

relationship between the two types of measurement is to a straight line (Townsend, 2002). The Pearson's product moment correlation is used when the data is normally distributed while Spearman rank correlation is used when the data is not evenly distributed or where no assumptions are made about the shape of distributions (Townsend, 2002).

The correlation coefficient ( $r$ ) represents the proportion of common variation in the two variables (i.e. the strength of the relationship). The correlation coefficient ( $r$ ) varies between  $-1$  and  $+1$ . A correlation coefficient of  $-1$  indicates a perfect negative relationship between the variables. A correlation coefficient of  $+1$  indicates a perfect positive relationship between the variables. A correlation of  $0$  indicates no relationship between the variables. There is no convention as to what value of  $r$  can be described as a strong or weak correlation however the following guide given by Townsend (2002) was used in this study.

<b>Correlation</b>	<b>r</b>
Strong	$-1 \leftrightarrow -0.9$
Weak	$-0.9 \leftrightarrow -0.6$
Little or none	$-0.6 \leftrightarrow 0 \leftrightarrow 0.6$
Weak	$0.6 \leftrightarrow 0.9$
Strong	$0.9 \leftrightarrow 1.0$

### 3.0 RESULTS and DISCUSSION

The results and subsequent discussion of these results for the three chosen ecosystems are presented within this section of the thesis. The ecosystems are discussed in detail with regard to soil radionuclide concentrations, nutrient and radionuclide content of vegetation, transfer factors and correlations between these transfer factors and physiochemical properties of soil and vegetation. The Atlantic blanket bog is presented in section 3.1, followed by the upland blanket bog in section 3.2 and the wet grassland in section 3.3. The final part of the results and discussion section will encompass a summary of the three ecosystems and is presented in section 3.4.

#### 3.1 Atlantic Blanket Bog

Cloonty Bog defined as an Atlantic blanket bog is located in Ballintrillick, Sligo along the Atlantic coast of Ireland. The indigenous vegetation within the ecosystem was a mixture of *C. vulgaris* (Ling Heather), *E. vaginatum* (Bog cotton) and *S. moss*. Soil and vegetation were sampled from seven sites (A1 – A7), within the Atlantic blanket bog in order to assess the uptake of radionuclides by vegetation within this ecosystem and also to correlation TF values with soil physiochemical properties and vegetation nutrient status. Aboveground vegetation was sampled from each site and soil was sampled to a depth of 20 cm at each site in order to calculate TF values. Soil was also sampled to a depth of 50 cm at one of the sites (Site A3) in order to assess the distribution of radionuclides within the soil lower down in the soil profile.

The distribution of radionuclides within the soil and the soil physiochemical characteristics are discussed in section 3.1.1 and 3.1.2 respectively. Radionuclide and nutrient content of vegetation indigenous to this ecosystem are discussed in sections 3.1.3 and 3.1.4 respectively. Transfer factors were calculated for each radionuclide and are discussed in section 3.1.5 and correlations between these transfer factors and the chemical characteristics of the soil and vegetation are discussed in section 3.1.6. The discussion of Atlantic blanket bog results focuses mainly on TF values calculated and

significant correlations observed (sections 3.1.5 and 3.1.6) in keeping with the main objectives of this study. The results found within these sections are compared with findings published from similar studies. The soil radionuclide and physiochemical properties are tabulated and discussed but an extensive discussion and comparison of these results with other studies was not deemed necessary to fulfil the objectives of this study. Section 3.1.7 highlights the most significant finding from the investigation within this ecosystem.

### 3.1.1 Radionuclide distribution in Soil

The radionuclide distribution within the soil was determined by analysing the  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  content of four soil layers i.e. 0-5 cm, 5-10 cm, 10-15cm and 15-20 cm respectively at six sites (A1 –A2 and A4 – A7) and ten soil layers at one additional site (A3: 0-50 cm depth divided into 5 cm layers). Results are reported in sections 3.1.1.1 to 3.1.1.2 below and outlined in Table 14.

#### 3.1.1.1 $^{238}\text{U}$ , $^{226}\text{Ra}$ and $^{228}\text{Ra}$ distribution in soil

U-238 activities were measured in the soil using the  $^{234}\text{Th}$  method and results are tabulated in Table 14. The soil samples were counted for a period of 2 weeks however peak areas were not statistically sufficient, therefore the concentration of  $^{238}\text{U}$  in the soil was below the minimum detectable activity for the method used.

Ra-226 activities in the peat soil were below the minimum detectable activity using its own peak at 186 keV and the radon build-up method ( $^{214}\text{Bi}$  and  $^{214}\text{Pb}$ ) with the exception of two soils (Table 14). An activity of 1.5 Bq/kg was detected for the A1 5-10 cm layer with an associated error of 17 % and an activity of 1.3 Bq/kg was detected for the A5 5-10 cm layer with an associated error of 13 %. These soils had the highest density of all the peat soils analyzed thereby enabling shorter analysis times ( $> 0.5$  kg/L).

Site	Soil Layer	<sup>238</sup> U (Bq/kg) <sup>1</sup>	<sup>226</sup> Ra (Bq/kg) <sup>2</sup>	<sup>228</sup> Ra (Bq/kg) <sup>3</sup>	<sup>40</sup> K (Bq/kg)	<sup>137</sup> Cs (Bq/kg)
A 1	0-5 cm	< 3	< 4	< 2	33 ± 5	158 ± 10
	5-10 cm	< 2	< 4	< 1	26 ± 3	81 ± 5
	10-15 cm	< 3	< 4	< 1	26 ± 3	50 ± 3
	15-20 cm	< 3	< 3	< 2	18 ± 3	22 ± 1
A 2	0-5 cm	< 3	< 4	< 2	36 ± 4	199 ± 13
	5-10 cm	< 3	1.5 ± 0.34 <sup>4</sup>	< 1	30 ± 5	127 ± 8
	10-15 cm	< 3	< 4	< 1	31 ± 4	58 ± 4
	15-20 cm	< 3	< 5	< 2	19 ± 3	22 ± 1
A 3	0-5 cm	< 3	< 5	< 1	45 ± 6	149 ± 10
	5-10 cm	< 3	< 4	< 1	48 ± 7	98 ± 6
	10-15 cm	< 3	< 4	< 1	40 ± 5	75 ± 5
	15-20cm	< 2	< 3	< 1	47 ± 5	78 ± 5
	20-25 cm	< 3	< 4	< 1	30 ± 5	40 ± 3
	25-30 cm	< 3	< 5	< 2	21 ± 4	26 ± 2
	30-35 cm	< 3	< 5	< 1	13 ± 3	13 ± 1
	35-40 cm	< 3	< 4	< 1	13 ± 3	11 ± 1
	40-45 cm	< 3	< 4	< 1	14 ± 3	10 ± 1
	45-50 cm	< 2	< 3	< 1	13 ± 3	11 ± 1
A 4	0-5 cm	< 5	< 7	< 2	28 ± 4	230 ± 15
	5-10 cm	< 3	< 4	< 1	22 ± 3	120 ± 8
	10-15 cm	< 4	< 3	< 1	16 ± 4	43 ± 3
	15-20 cm	< 3	< 4	< 1	14 ± 3	22 ± 1
A 5	0-5 cm	< 3	< 4	< 1	30 ± 4	191 ± 13
	5-10 cm	< 3	1.3 ± 0.17 <sup>4</sup>	< 1	24 ± 3	144 ± 9
	10-15 cm	< 3	< 3	< 1	16 ± 4	47 ± 3
	15-20 cm	< 4	< 5	< 2	13 ± 3	23 ± 2
A 6	0-5 cm	< 4	< 5	< 11	29 ± 5	122 ± 8
	5-10 cm	< 3	< 4	< 9	24 ± 4	65 ± 4
	10-15cm	< 2	< 3	< 13	18 ± 3	32 ± 2
	15-20 cm	< 3	< 4	< 13	14 ± 3	16 ± 1
A 7	0-5 cm	< 4	< 6	< 11	25 ± 5	198 ± 12
	5-10cm	< 3	< 4	< 14	18 ± 3	49 ± 3
	10-15 cm	< 3	< 4	< 10	16 ± 4	26 ± 3
	15-20 cm	< 3	< 3	< 9	13 ± 3	13 ± 1
Notes: 1: Results calculated using the <sup>234</sup> Th peak (63 keV and 92 keV) 2: Results calculated using the <sup>226</sup> Ra peak (186 keV) 3: Results calculated using the <sup>228</sup> Ac peak (911 keV and 968 keV) 4: Results calculated using the <sup>214</sup> Pb and <sup>214</sup> Bi peak (351 keV and 609 keV)						

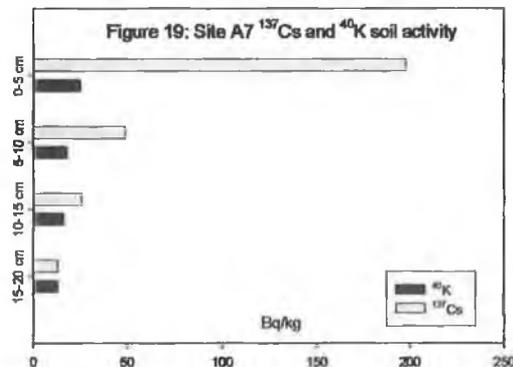
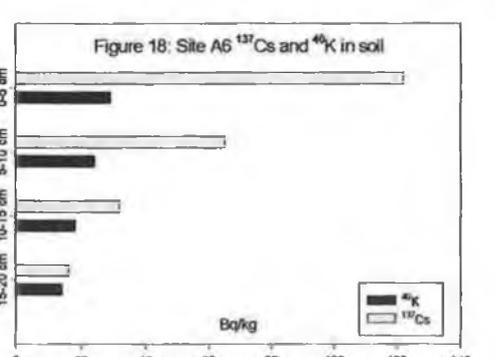
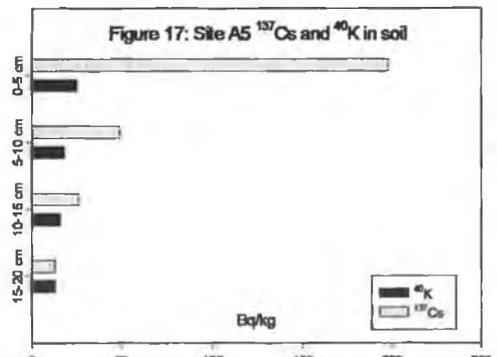
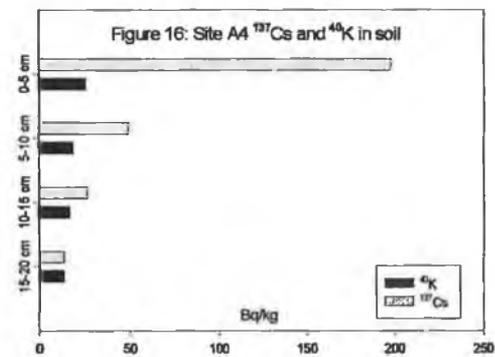
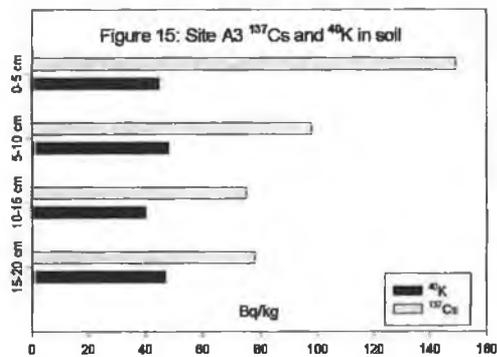
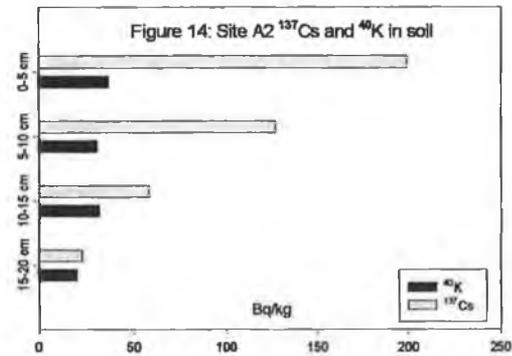
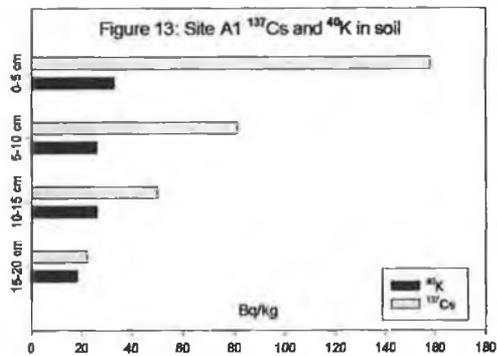
Table 14: <sup>238</sup>U, <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>40</sup>K and <sup>137</sup>Cs soil levels in soil at sites A1 – A7.

Ra-228 activities were below the limits of detection using the  $^{228}\text{Ac}$  method in all peat soils analyzed (Table 14). Radionuclide activities ( $^{238}\text{U}$ ,  $^{226}\text{Ra}$ , and  $^{228}\text{Ra}$ ) lower down in the soil profile at Site A3 (0 – 50 cm) were also below the limits of detection for the methods used (Table 14).

### 3.1.1.2 $^{40}\text{K}$ and $^{137}\text{Cs}$ distribution in Soil

The levels of  $^{40}\text{K}$  in peat soil ranged between 13 Bq/kg and 48 Bq/kg (Table 14), and were low at all seven sites analyzed as illustrated in Figures 13 – 19. This was also the case for concentrations of plant available potassium in the peat soil, with levels ranging between 70 mg/kg and 150 mg/kg (Table 15, Section 3.1.2). The results for potassium found within this study are in agreement with the low levels of plant available potassium found in peat soil by Sanchez *et al* (1999). The soil in the Atlantic blanket bog is acidic in nature (pH < 4.6) due to the high concentration of organic matter present (> 90%) and as reported previously acidic soils are generally nutrient deficient due to acid leaching (Tan, 1992).

Potassium - 40 levels at the seven sites were compared to assess if the levels are similar throughout the entire ecosystem. Potassium-40 activities on average ranged from 18 Bq/kg and 45 Bq/kg for the seven sites and the average concentration of  $^{40}\text{K}$  in the soil at site A3 was significantly higher (One Way ANOVA,  $p < 0.022$ ) than that found at any other site. Comparisons between the other six sites indicated that the mean  $^{40}\text{K}$  concentration for each site was statistically similar (One Way ANOVA,  $p > 0.05$ ).



Figures 13 –19: <sup>137</sup>Cs and <sup>40</sup>K activity in soil at Sites A1 – A7.

Potassium-40 concentrations were significantly higher in the 0-5 cm layer in comparison to the lower layers: 5-10 cm, 10-15 cm and 15 – 20 cm layer (One Way ANOVA,  $p < 0.03$ ). This is possibly due to recycling of potassium in the top layers of soil from decomposing plant material. Potassium-40 concentrations on average were also significantly higher in the 5-10 cm layer in comparison to the 15-20 cm layer (One Way ANOVA,  $p = 0.004$ ). The concentrations of plant available potassium in soil (Table 15, Section 3.1.2) followed the same pattern as  $^{40}\text{K}$  in soil. Plant available potassium levels were significantly higher in the 0-5 cm layer in comparison to the lower layers: 5-10 cm, 10-15 cm and 15 – 20 cm layer (One Way ANOVA,  $p < 0.036$ ).

Concentrations of  $^{137}\text{Cs}$  found in the peat soil were higher than  $^{40}\text{K}$  (t-test,  $p < 0.001$ ) and activities of both radionuclides decreased with increasing soil depth at all sites with the exception of  $^{137}\text{Cs}$  concentrations at Site A3 (Figures 13 - 19). The average concentration of  $^{137}\text{Cs}$  in the four soil layers (0-20 cm) was calculated for each site and results indicate that  $^{137}\text{Cs}$  levels in the peat soil between sites are not significantly different (One Way ANOVA,  $p > 0.05$ ).

Comparisons between soils layers were carried out and  $^{137}\text{Cs}$  mean concentrations was significantly higher in the 0-5 cm layer in comparison to the lower layers: 5-10 cm, 10-15cm and 15-20 cm (One Way ANOVA,  $p < 0.002$ ). Caesium-137 concentrations were also significantly higher in the 5-10 cm layer in comparison to both the 10-15 cm and 15-20 cm layer (One Way ANOVA,  $p < 0.016$ ). The decrease in  $^{137}\text{Cs}$  activity with increasing soil depth is best described using a log function as illustrated in Figure 20 where an inverse relationship was found to be present between log average  $^{137}\text{Cs}$  activity as a function of soil depth (linear regression:  $r^2 = 0.991$ ).

Figure 20: Log  $^{137}\text{Cs}$  versus soil depth

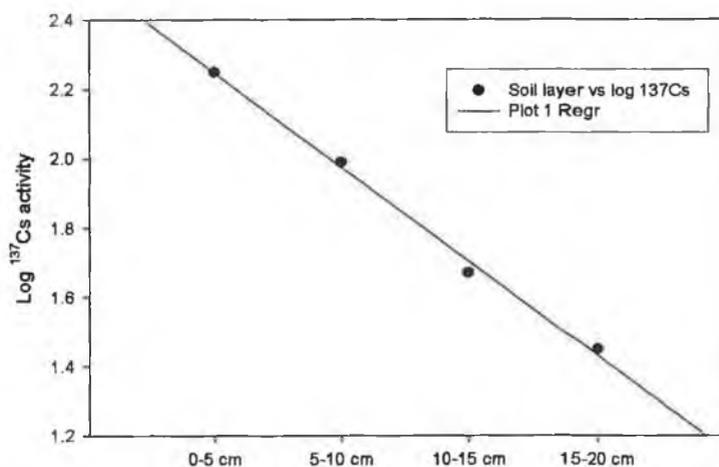


Figure 20: Log  $^{137}\text{Cs}$  activity versus soil depth for sites A1 – A7.

Concentrations of  $^{137}\text{Cs}$  were also found to be low further down in the soil profile with an activity of  $11 \pm 1$  Bq/kg observed in the bottom layer at site A3 (0-50 cm depth profile). The apparent low vertical mobility of  $^{137}\text{Cs}$  within the soil profile is possibly due to the high concentration of organic matter in the peat (> 90% in all peat soils). Many studies have shown that radiocaesium added to organic soil remains plant-available for extended periods of time and after uptake is recycled within the rooting zone of the vegetation in the form of decomposing plant material (Barber, 1964, Shand *et al*, 1994, Valcke and Cremers, 1994, Livens *et al*, 1991, Shaw and Bell, 2001 and Staunton *et al*, 2002).

### 3.1.2 Soil physiochemical properties

The physiochemical characteristics of the soil were extensively studied in order to ascertain if any of these parameters had a significant impact on the uptake of the radionuclide by vegetation. The physiochemical parameters chosen were moisture content, pH, organic matter content, available levels of nutrients (magnesium, sodium, calcium, potassium) and cation exchange capacity of the soil. The results for each of

these parameters are presented in Table 15 and are discussed in detail within this section. Correlations between these parameters and radionuclide uptake in vegetation are discussed in section 3.1.6.

The average moisture content of the soil varied between 86.4 % and 91.7 % for the sites sampled and the moisture content of the soil at sites A4 and A5 were significantly higher than the other sites sampled (One Way ANOVA,  $p < 0.001$ ). The vegetation found to be abundant at these two sites was *Sphagnum* moss and past studies have shown that moss is generally found abundant in the wetter regions of the bog (Jahns, 1983). Comparisons were carried out between the moisture content for the top four layers of the soils and results indicate there is no significant difference between the moisture content of each soil layer for the sites sampled (One Way ANOVA,  $p = 0.947$ ).

The average pH of the soil ranged from 3.4 and 4.5 and the pH of the soil at sites A4 and A5 were significantly higher than the other sites sampled (One Way ANOVA,  $p < 0.001$ ). The pH of the soil follows the same pattern as the moisture content of the soil at sites A4 and A5. Both parameters are significantly higher in comparison to the other sites sampled however no significant relationship was found between the moisture content and pH of the soil when tested (Pearson correlation coefficient of 0.60,  $p = 0.154$ ). Comparisons were carried out on the pH of the individual soil layers and results indicate that the pH of the soil is similar throughout the four soil layers at each site (One Way ANOVA,  $p = 0.967$ ).

The average organic matter content of the soil varied between 94.8 % and 96.7 % for the sites sampled and results indicate that there was no significant difference between the organic matter content of the soil at each site (One Way ANOVA,  $p = 0.309$ ). Similar observations were noted for the organic matter content in the individual soil layers at each site where results indicate that there is no significant difference in the organic matter content of each soil layer for the top four soil layers at each site (One Way ANOVA,  $p = 0.722$ ).

Site	Soil Layer	Moisture Content %	pH	Organic Matter %	Available Magnesium mg/kg	Available Sodium mg/kg	Available Calcium mg/kg	Available Potassium mg/kg	CEC of Soil meq/100g
A 1	0-5 cm	88.5	3.5	95.8	414	227	366	161	145
	5-10 cm	88.4	3.3	94.8	532	246	366	141	150
	10-15 cm	88.3	3.4	94.9	611	284	419	120	154
	15-20cm	88.8	3.4	95.5	419	223	295	72	147
A 2	0-5 cm	87.4	3.5	95.4	491	250	542	294	153
	5-10 cm	87.0	3.4	94.0	368	208	328	131	142
	10-15 cm	86.4	3.4	95.5	376	192	281	102	139
	15-20 cm	87.9	3.4	94.3	521	226	466	91	154
A 3	0-5 cm	85.6	4.0	96.5	317	126	253	138	132
	5-10 cm	86.5	3.7	94.9	381	207	340	160	138
	10-15 cm	87.5	3.6	95.3	209	142	175	90	145
	15-20cm	86.8	3.7	93.6	366	204	258	127	154
	20-25 cm	87.7	3.7	95.2	307	190	234	105	146
	25-30 cm	88.2	3.7	96.8	567	313	486	152	149
	30-35 cm	87.8	3.8	97.0	463	262	327	74	155
	35-40 cm	88.3	3.7	97.0	679	328	461	71	144
	40-45 cm	88.8	3.8	97.7	565	231	292	41	158
	45-50 cm	89.1	3.8	97.1	451	282	383	45	141
A 4	0-5 cm	91.2	4.5	97.0	436	199	401	230	144
	5-10 cm	91.5	4.5	98.6	433	238	333	84	166
	10-15 cm	91.6	4.5	96.0	693	285	603	100	156
	15-20cm	90.8	4.5	95.1	656	205	275	97	130
A 5	0-5 cm	90.8	3.9	96.8	726	244	632	307	116
	5-10 cm	92.6	4.0	96.3	471	186	386	167	125
	10-15 cm	92.8	4.0	95.7	400	209	367	109	151
	15-20 cm	90.5	3.9	94.3	607	335	570	140	117
A 6	0-5 cm	88.9	3.9	95.2	483	254	372	85	156
	5-10 cm	90.1	4.0	94.0	372	238	322	76	159
	10-15 cm	88.1	3.8	95.4	442	248	313	74	161
	15-20 cm	89.2	3.9	97.2	392	198	298	47	148
A 7	0-5 cm	85.4	3.9	94.5	389	191	370	160	148
	5-10cm	86.4	3.9	95.0	398	197	315	78	167
	10-15 cm	85.9	3.8	94.5	592	300	483	96	163
	15-20 cm	87.8	3.9	96.6	458	250	347	74	145

Table 15: Soil physiochemical properties: Sites A1 – A7

Plant available levels of calcium, magnesium, sodium and potassium were measured in the soil. The average levels of plant available calcium in the soil ranged from 257 mg/kg and 490 mg/kg and the average concentration of calcium at each site was statistically similar (One Way ANOVA,  $p = 0.083$ ). The concentration of plant available calcium in the individual soil layers at each site was found to be statistically similar (One Way ANOVA,  $p = 0.601$ ). The average levels of plant available magnesium in the soil ranged between 308 mg/kg and 555 mg/kg and levels of available magnesium in the soil were significantly higher at site A4 in comparison to site A3 (One Way ANOVA,  $p = 0.048$ ). The concentration of plant available magnesium remained relatively constant throughout the upper 20 cm of the soil as results for each of the four soil layers are relatively similar (One Way ANOVA,  $p = 0.601$ ).

Plant available sodium levels in the soil ranged on average from 170 mg/kg to 245 mg/kg. The average concentration of sodium at each site is statistically similar as is the case with the concentration of sodium in the individual soil layers indicating that the concentration of available sodium is relatively constant in the upper 20 cm of the soil (One Way ANOVA,  $p > 0.212$ ). The average levels of plant available potassium in the soil ranged from 71 mg/kg and 181 mg/kg and concentrations at each site were statistically similar (One Way ANOVA,  $p = 0.272$ ). The average concentration of plant available potassium in the soil was significantly higher in the 0-5 cm soil layer in comparison to the lower layers: 5-10 cm, 10-15 cm and 15-20 cm. These results also reflect the distribution of  $^{40}\text{K}$  within the soil as discussed previously in section 3.1.1.2.

The levels of plant available calcium, magnesium, sodium and potassium were compared and results indicate that their concentrations in soil decrease in the following order:

magnesium > calcium > sodium > potassium

The average concentration of magnesium, calcium, sodium and potassium were 462 mg/kg, 374 mg/kg, 226 mg/kg and 127 mg/kg respectively. The concentration of plant available magnesium in soil was significantly higher than levels of plant available calcium, sodium and potassium (One Way ANOVA,  $p < 0.045$ ). Concentrations of plant available calcium were significantly higher than levels of plant available sodium and potassium (One Way ANOVA,  $p < 0.001$ ). The levels of plant available potassium in the soil was significantly lower than the other three nutrients analysed (One Way ANOVA,  $p < 0.022$ ).

The cation exchange capacity of the soil ranged from 137 meq/100g to 156 meq/kg and the CEC of the soil at site A6 was significantly higher than that of the soil at site A5 (One Way ANOVA,  $p = 0.017$ ). The CEC of the soil remained relatively constant in the four soil layers indicating that CEC of the soil is similar through the upper 20 cm of the soil at each site sampled (One Way ANOVA,  $p = 0.34$ ).

In general the soil physiochemical characteristics remains relatively constant throughout the ecosystem based on the seven sites sampled. The exceptions to this are the pH,

moisture content, CEC and plant available magnesium levels in the soil. The moisture content and pH of the soil were significantly higher at two sites (A4 and A5) than the other five sites sampled. The CEC of the soil at Site A6 was significantly than the soil at site A5. Plant available magnesium levels in the soil at site A4 was significantly higher in comparison to levels in the soil at site A3. Table 16 compares the concentrations of the aforementioned soil properties at the seven sites sampled and show the results in descending however as stated previously the differences between results in all cases may not be statistically significant.

Soil Property	Sites
Moisture Content	A5 > A4 > A6 > A1 > A3 > A6 > A7
pH	A4 > A5 > A6 = A7 > A3 > A2 = A1
Organic Matter	A4 > A5 > A6 > A1 > A7 > A3 > A2
Available Magnesium	A4 > A5 > A1 > A7 > A2 > A6 > A3
Available Sodium	A1 > A5 > A6 = A7 > A4 > A2 > A3
Available Calcium	A5 > A2 > A4 > A7 > A1 > A6 > A3
Available Potassium	A5 > A2 > A3 > A4 > A1 > A7 > A6
CEC	A7 = A6 > A4 = A1 > A2 > A3 > A5

Table 16: Comparison of soil physiochemical properties at sites A1 – A7.

### 3.1.3 Radionuclide content of vegetation

Levels of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  were analyzed in the vegetation by gamma spectrometry and the results obtained are tabulated in Table 17. The concentration of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  in all vegetation were below the limits of detection for the methods used (Section 2.5.1.2) however the concentrations of  $^{40}\text{K}$  and  $^{137}\text{Cs}$  were detectable in all vegetation analyzed as outlined in Table 17.

Site	Sample	<sup>238</sup> U (Bq/kg)	<sup>226</sup> Ra (Bq/kg)	<sup>228</sup> Ra (Bq/kg)	<sup>40</sup> K (Bq/kg)	<sup>137</sup> Cs (Bq/kg)
A1	<i>Calluna vulgaris</i> - Leaves	< 11	< 14	< 7	123 ± 21	716 ± 50
A2	<i>Calluna vulgaris</i> - Leaves	< 10	< 10	< 8	100 ± 19	589 ± 40
A3	<i>Calluna vulgaris</i> - Leaves	< 9	< 11	< 8	86 ± 15	956 ± 65
A1	<i>Calluna vulgaris</i> - Stems	< 6	< 8	< 2	69 ± 13	302 ± 21
A2	<i>Calluna vulgaris</i> - Stems	< 6	< 8	< 2	73 ± 14	196 ± 13
A3	<i>Calluna vulgaris</i> - Stems	< 9	< 16	< 3	40 ± 7	282 ± 19
A4	<i>Sphagnum</i> moss	< 16	< 12	< 3	64 ± 13	142 ± 10
A5	<i>Sphagnum</i> moss	< 41	< 47	< 13	65 ± 13	325 ± 22
A6	<i>Eriophorum vaginatum</i>	< 14	< 15	< 3	293 ± 32	301 ± 20
A7	<i>Eriophorum vaginatum</i>	< 9	< 11	< 3	225 ± 25	192 ± 13

Table 17: Radionuclide concentrations in bog vegetation: Sites A1 – A7

Caesium-137 concentration in all the selected vegetation ranged between 192 Bq/kg and 956 Bq/kg (Table 17) and levels of <sup>137</sup>Cs in *C. vulgaris* leaves were significantly higher than in the stems of *C. vulgaris* (t-test,  $p < 0.05$ ). Concentrations of <sup>137</sup>Cs were also significantly higher in *C. vulgaris* leaves in comparison to both *E. vaginatum* and *Sphagnum* moss (One Way ANOVA,  $p < 0.05$ ). The concentration of <sup>137</sup>Cs in *C. vulgaris* stems, *E. vaginatum* and moss were statistically similar (One Way ANOVA,  $p > 0.05$ ).

Potassium-40 concentration in vegetation ranged from 40 Bq/kg to 293 Bq/kg (Table 17). The highest concentrations of <sup>40</sup>K were found to be present in *E. vaginatum*. Moss samples were found to contain the lowest concentration of <sup>40</sup>K and one of the moss samples also contained the lowest concentration of <sup>137</sup>Cs for the vegetation sampled. On the other hand the moss samples contained significantly higher concentrations of calcium and magnesium than any other plant species studied. This may be a reflection of the uptake mechanisms of mosses as the lack a well-developed root system and uptake of radionuclides occurs mainly through ion exchange processes from wet and dry deposition (Delfanti *et al*, 1991, Ugur *et al*, 2003). The uptake of calcium and magnesium by moss in this ecosystem may possibly be due to wet deposition.

The concentrations of  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in soil and vegetation were compared to assess if vegetation uptake was dependent on soil concentrations. Caesium-137 concentrations in vegetation appear to be independent of  $^{137}\text{Cs}$  concentration in soil (Spearman correlation coefficient of -0.24,  $p = 0.49$ ). The absence of a significant correlation between  $^{137}\text{Cs}$  in soil and vegetation was also noted by Bunzl *et al* (2000) and Ciuffo *et al* (2002). A similar observation was made when  $^{137}\text{Cs}$  concentration in vegetation when compared with  $^{40}\text{K}$  activities in soil where results indicate that  $^{137}\text{Cs}$  uptake is independent of  $^{40}\text{K}$  activity in soil. An interesting correlation was observed between  $^{40}\text{K}$  concentrations in vegetation and  $^{137}\text{Cs}$  concentration in soil where a Spearman correlation coefficient of -0.624,  $p = 0.048$ ) indicates an inverse relationship between these two parameters. These results appear to indicate that for the vegetation studied, the plants found to have the highest concentration of  $^{40}\text{K}$  present were growing on soil, which contained the lowest concentration of  $^{137}\text{Cs}$ . These results are discussed in greater detail with regard to their transfer factors in section 3.1.5.

#### 3.1.4 Nutrient content of vegetation

The calcium, magnesium and sodium concentrations in vegetation were determined to assess the impact of these nutrients on radionuclide uptake. The results for total concentrations of these nutrients in vegetation are outlined in Table 18 and correlations between these nutrients concentrations and radionuclide transfer factor values are discussed in section 3.1.6.

The concentrations of calcium in vegetation ranged from 1469 mg/kg in *Eriophorum vaginatum* to 8831 mg/kg in *Sphagnum* moss. The mean concentration of calcium in each of the plant species was calculated and results indicate that the concentration of calcium in *Sphagnum* moss was significantly higher in comparison to both *C. vulgaris* and *E. vaginatum* (One Way ANOVA,  $p < 0.05$ ). The concentration of calcium in *C. vulgaris* and *E. vaginatum* are statistically similar (One Way ANOVA,  $p > 0.05$ ). Comparisons of calcium concentration in plant portions with regard to *C. vulgaris* leaves indicate that the concentration (mean) of calcium found in the leaves of *C. vulgaris* was significantly

higher in comparison to the stems with mean concentrations of 1459 mg/kg and 4275 mg/kg for stems and leaves respectively (One Way ANOVA,  $p < 0.001$ ).

Site	Vegetation	Calcium mg/kg	Magnesium mg/kg	Sodium mg/kg
A1	<i>Calluna vulgaris</i> - Leaves	4595	1622	1235
A2	<i>Calluna vulgaris</i> - Leaves	3947	1586	1134
A3	<i>Calluna vulgaris</i> - Leaves	4282	1570	1129
A1	<i>Calluna vulgaris</i> - Stems	1708	485	759
A2	<i>Calluna vulgaris</i> - Stems	1130	511	1012
A3	<i>Calluna vulgaris</i> - Stems	1539	435	1248
A4	Sphagnum moss	8831	1350	1299
A5	Sphagnum moss	6257	1489	821
A6	<i>Eriophorum vaginatum</i>	1673	898	734
A7	<i>Eriophorum vaginatum</i>	1469	814	688

Table 18: Nutrient concentration of vegetation: Sites A1 – A7

The concentration of magnesium in vegetation ranged from 435 mg/kg and 1622 mg/kg and results indicate that the magnesium levels in *Sphagnum* moss are significantly higher than both *C. vulgaris*, and *E. vaginatum* (One Way ANOVA,  $p < 0.05$ ). Magnesium levels in *C. vulgaris*, and *E. vaginatum* were statistically similar (One Way ANOVA,  $p > 0.05$ ). Comparisons of magnesium concentrations in plant portions of *C. vulgaris* indicate that the concentration of magnesium was significantly higher in the leaves of *C. vulgaris* with regard to the stems (One Way ANOVA,  $p < 0.001$ ). The mean concentrations of magnesium in *C. vulgaris* stems and leaves were 477 mg/kg and 1593 mg/kg respectively.

The concentration of sodium in vegetation ranged from 734 mg/kg in *E. vaginatum* to 1234 mg/kg in *C. vulgaris*. Comparisons between species indicate that the mean concentration of sodium in the plant species is statistically similar for *C. vulgaris*, *E. vaginatum* and *Sphagnum* moss (One Way ANOVA,  $p = 0.333$ ). The sodium content of *C. vulgaris* stems and leaves are statistically similar with mean concentrations of 1006

mg/kg and 1166 mg/kg respectively (One Way ANOVA,  $p = 0.334$ ). The distribution of sodium within the plant portions of *C. vulgaris* does not reflect the pattern observed for both calcium and magnesium, which were both significantly higher in the leaves in comparison to the stems.

Calcium concentrations were significantly higher in *C. vulgaris* in comparison to both sodium and magnesium (One Way ANOVA,  $p < 0.001$ ) and sodium and magnesium levels in *C. vulgaris* are statistically similar (One Way ANOVA,  $p = 0.886$ ). Similar observation were made for both *E. vaginatum* and *Sphagnum* moss where the calcium concentration were significantly in each of the plant species in comparison to both sodium and magnesium levels (One Way ANOVA,  $p < 0.05$ ) and sodium and magnesium levels in plant species were statistically similar (One Way ANOVA,  $p < 0.941$ ).

Overall for the three plant species studied, the calcium levels were significantly higher in all vegetation sampled in comparison to both magnesium and sodium levels present (One Way ANOVA,  $p < 0.05$ ). The concentrations of sodium and magnesium in all three plant species were statistically similar (One Way ANOVA,  $p > 0.05$ ).

### 3.1.5 Transfer Factors for Atlantic Blanket Bog Vegetation

Transfer factors were calculated for each radionuclide based on the activity concentration of nuclide per kg of dry plant mass divided by the activity concentration of nuclide in top 20 cm of dry soil (Section 2.2). Transfer factors were not calculated for  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  as concentrations of these radionuclides were below the limits of detection in both the soil and vegetation as outlined in Table 14 and 17 respectively. Transfer factors values were calculated for  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in *C. vulgaris* (leaves and stems) and *E. vaginatum* but not for the moss species due to the lack of a well-developed root system (Dowdall *et al*, 2005). The results for  $^{137}\text{Cs}$  and  $^{40}\text{K}$  transfer factor values are outlined in Table 19.

Soil to vegetation TF values for  $^{137}\text{Cs}$  in the vascular plants studied ranged from 1.9 to 9.6 (Table 19). Transfer factor values for  $^{137}\text{Cs}$  were significantly higher in *C. vulgaris* leaves in comparison to the stems (t-test,  $p = 0.016$ ), comparisons between species, however show that  $^{137}\text{Cs}$  TF values for *C. vulgaris* and *E. vaginatum* are statistically similar (t-test,  $p > 0.05$ ). Transfer factor values for *C. vulgaris* reported within this study are similar to TF values reported in the literature of 1.8 – 3.3 (plant portion unspecified) and 0.6 and 6.0 for *C. vulgaris* stems and leaves respectively (Bunzl and Kracke, 1986, Livens *et al*, 1991).

Site	Sample	$^{40}\text{K}$ TF	$^{137}\text{Cs}$ TF
A1	<i>Calluna vulgaris</i> - Leaves	4.8 ± 1.4	9.2 ± 1.1
A2	<i>Calluna vulgaris</i> - Leaves	3.5 ± 1.1	5.8 ± 0.7
A3	<i>Calluna vulgaris</i> - Leaves	1.9 ± 0.5	9.6 ± 1.3
A1	<i>Calluna vulgaris</i> - Stems	2.7 ± 0.8	3.9 ± 0.5
A2	<i>Calluna vulgaris</i> - Stems	2.5 ± 0.8	1.9 ± 0.2
A3	<i>Calluna vulgaris</i> - Stems	0.9 ± 0.2	2.8 ± 0.4
A6	<i>Eriophorum vaginatum</i>	13.8 ± 4.0	5.1 ± 0.6
A7	<i>Eriophorum vaginatum</i>	12.3 ± 4.0	2.7 ± 0.4

Table 19: Transfer factor (TF) values for  $^{40}\text{K}$  and  $^{137}\text{Cs}$

The trend found within this study for  $^{137}\text{Cs}$  TF values in *C. vulgaris* leaves and stems (i.e. leaves > stems) is in agreement with the findings for  $^{137}\text{Cs}$  in *Salix viminalis* (basket willow) where the TF values decreased in the order: roots > leaves > cuttings > stems (von Fricks *et al*, 2002). Similar trends were also observed for *V. myrtilus* (bilberry), *V. uliginosus* (bog bilberry), *V. vitis-idaea* (mountain cranberry), *V. oxycoccos* (northern cranberry) and *C. vulgaris* where TF values in the plant portions decreased in the order: flowers > leaves = berries > roots > stems (Bunzl and Kracke, 1986). The above results for the distribution of  $^{137}\text{Cs}$  within *C. vulgaris* are in agreement with  $^{137}\text{Cs}$  distribution in tropical, spruce and pine trees where current knowledge indicates that  $^{137}\text{Cs}$  activity is highest in the youngest or growing part of the trees (Barci-Funel *et al*, 1995, McGee *et al*, 2000, Fogh and Anderson, 2001, Mosquera *et al*, 2006).

Potassium-40 TF values ranged from 0.9 to 13.8 (Table 19), and were significantly higher in *E. vaginatum* in comparison to both *C. vulgaris* leaves and stems (One Way ANOVA  $p < 0.001$ ). Comparisons between  $^{40}\text{K}$  TF values in *C. vulgaris* leaves and stems show that the differences are not statistically significant (One Way ANOVA,  $p = 0.42$ ).

Transfer factor values for  $^{137}\text{Cs}$  for the vegetation studied appear to be independent of the concentration of  $^{137}\text{Cs}$  in the soil (Spearman correlation coefficient of  $-0.0364$ ,  $p = 0.885$ ) as illustrated in Figure 21. A similar observation was made for  $^{137}\text{Cs}$  TF values for vegetation studied and  $^{40}\text{K}$  in soil where a Spearman correlation coefficient of  $0.230$  ( $p = 0.537$ ) indicates that  $^{137}\text{Cs}$  uptake by vegetation was independent on  $^{40}\text{K}$  activity in soil. The relationship (or lack of) between these parameters is illustrated in Figure 22. These results are in agreement with the findings of Bunzl *et al* (2000) and Ciuffo *et al* (2002) but are contradictory to the finding of Robinson and Stone (1992), Gerzabek *et al* (1998) and Tsukada and Nakamura (1999). However in the case of Robinson and Stone (1992) and Tsukada and Nakamura (1999) the potassium levels in the soils were continuously increased by the addition of artificial fertilizer which may possibly have a significant impact on the uptake of  $^{137}\text{Cs}$ .

Figure 21:  $^{137}\text{Cs}$  TF versus  $^{137}\text{Cs}$  activity in soil

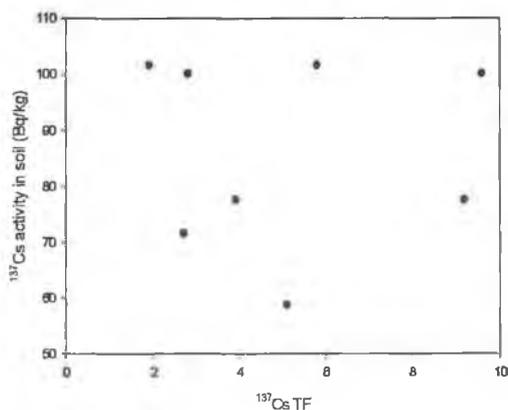


Figure 22:  $^{137}\text{Cs}$  TF versus  $^{40}\text{K}$  activity in soil.

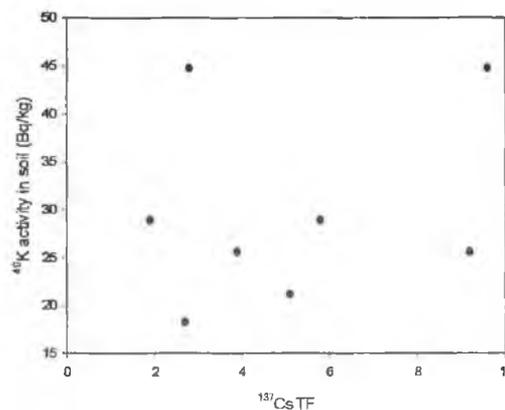


Figure 21 - 22: Correlation between  $^{137}\text{Cs}$  TF values and  $^{137}\text{Cs}$  and  $^{40}\text{K}$  activities in soil.

Figure 23:  $^{40}\text{K}$  TF versus  $^{137}\text{Cs}$  activity in soil

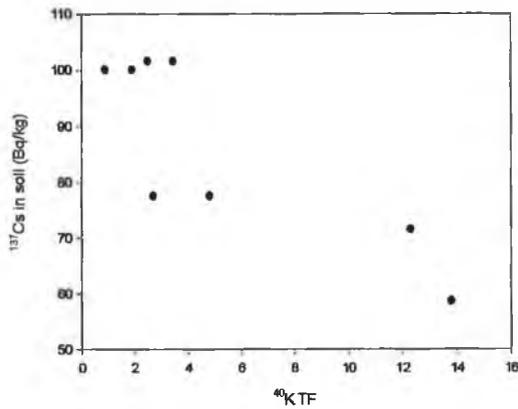
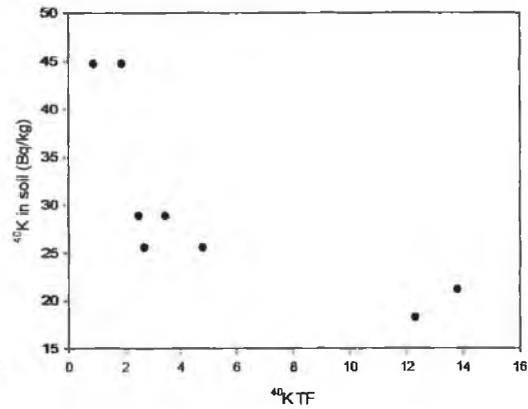


Figure 24:  $^{40}\text{K}$  TF versus  $^{40}\text{K}$  activity in soil



Figures 23 - 24: Correlation between  $^{40}\text{K}$  TF values and  $^{137}\text{Cs}$  and  $^{40}\text{K}$  activities in soil

Potassium-40 TF values were found to be significantly correlated with both the  $^{137}\text{Cs}$  concentration and  $^{40}\text{K}$  concentration in the soil as illustrated in Figures 23 and 24 respectively. A Spearman correlation of -0.691 ( $p = 0.047$ ) indicates an inverse relationship between uptake of  $^{40}\text{K}$  by vegetation and  $^{137}\text{Cs}$  in soil. A similar trend was also found for  $^{40}\text{K}$  activity in soil where a Spearman correlation coefficient of -0.909 indicates a very strong relationship between  $^{40}\text{K}$  TF values and  $^{40}\text{K}$  in soil. These results indicate that uptake of  $^{40}\text{K}$  by vegetation in the Atlantic blanket bog was highest on regions of soil which contained the lowest concentration of both  $^{137}\text{Cs}$  and  $^{40}\text{K}$ .

### 3.1.6 Correlations between vegetation transfer factors and chemical properties of soil and vegetation

Correlations were carried between transfer factor values ( $^{137}\text{Cs}$  and  $^{40}\text{K}$ ) and 1) the physiochemical parameters of the soil (Table 20) and 2) the nutrient status of the vegetation (Table 18). The purpose of these correlations was to determine if any of these parameters have a significant impact on the uptake of the aforementioned radionuclides. Table 20 represents the average concentration of each soil property studied over the

entire soil depth of 0-20 cm. The results are tabulated in this form to compare and correlate them with radionuclide transfer factor values, which are based on average activities over a soil depth of 0-20 cm. The full set of results for each individual soil layer (i.e. 0-5 cm, 5-10 cm, etc.) at all seven sites is presented in Table 15 (Section 3.1.2).

Site	Moisture %	pH	Organic Matter %	Available Potassium mg/kg	Available Calcium mg/kg	Available Sodium mg/kg	Available Magnesium mg/kg	CEC meq/100g
A 1	88.5	3.41	95.2	124	362	245	494	149
A 2	87.2	3.41	94.8	154	404	219	439	147
A 3	86.6	3.75	95.1	129	256	170	319	142
A 4	91.3	4.47	96.1	128	403	231	554	149
A 5	91.7	3.97	95.8	180	488	243	550	127
A 6	89.2	3.92	95.4	70	326	156	422	156
A 7	86.4	3.88	95.2	102	379	155	459	155

Table 20: Physicochemical properties of soil surface horizons at sites A1 – A7 (Average value for 0 - 20 cm)

The relationship between the radionuclide TF values and soil physicochemical properties and nutrient status of vegetation are outlined in Table 21 and any significant correlation are discussed in greater detail throughout this section.

Comparisons between  $^{137}\text{Cs}$  uptake in vegetation and soil physicochemical properties indicates that  $^{137}\text{Cs}$  TF values showed no significant correlation with any of the parameters outlined in Table 20. However correlations were found between  $^{137}\text{Cs}$  TF values and the nutrient status of the vegetation in particular the secondary nutrients, calcium and magnesium (Table 18).

Soil Properties	<sup>40</sup> K TF (All vegetation)	<sup>137</sup> Cs TF (All vegetation)	<sup>40</sup> K TF ( <i>C. vulgaris</i> )	<sup>137</sup> Cs TF ( <i>C. vulgaris</i> )
<sup>40</sup> K (Bq/kg)	X	X	X	X
<sup>137</sup> Cs (Bq/kg)	X	X	X	X
Moisture Content (%)	X	X	X	X
pH	X	X	X	X
Organic Matter (%)	X	X	X	X
Avail. Mg (mg/kg)	X	X	X	X
Avail. Na (mg/kg)	X	X	X	X
Avail. Ca (mg/kg)	X	X	X	X
Avail. K (mg/kg)	X	X	X	X
CEC	↑ <b>r<sup>2</sup> = 0.93</b> <b>p &lt; 0.001</b>	X	X	X
<b>Vegetation Nutrients</b>				
Calcium (mg/kg)	X	↑ <b>r<sup>2</sup> = 0.95</b> <b>p &lt; 0.001</b>	X	↑ <b>r<sup>2</sup> = 0.95</b> <b>p = 0.004</b>
Magnesium (mg/kg)	X	↑ <b>r<sup>2</sup> = 0.88</b> <b>p = 0.004</b>	X	↑ <b>r<sup>2</sup> = 0.89</b> <b>p = 0.017</b>
Sodium (mg/kg)	X	X	X	X
X: No correlation      ↑: Positive Correlation      ↓: Negative Correlation				

Table 21: Correlation between radionuclide transfer factor values and Atlantic blanket bog vegetation (Vegetation grouped together and *C. vulgaris*).

A Spearman correlation coefficient of 0.95 ( $p < 0.001$ ) indicates a strong correlation between the transfer of <sup>137</sup>Cs in vegetation and the calcium content of vegetation (Figure 25). A similar observation was noted between the magnesium content of vegetation and <sup>137</sup>Cs TF values (Pearson correlation coefficient of 0.88,  $p = 0.004$ ) as shown in Figure 26. No significant relationship was observed between the sodium content of vegetation and <sup>137</sup>Cs TF values (Spearman correlation coefficient of 0.43,  $p = 0.283$ ).

Figure 25: Plot of  $^{137}\text{Cs}$  TF values versus calcium content in vegetation

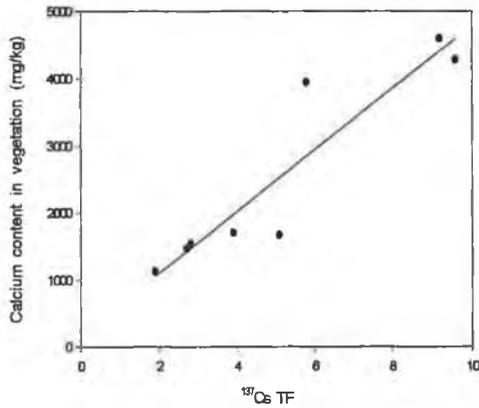
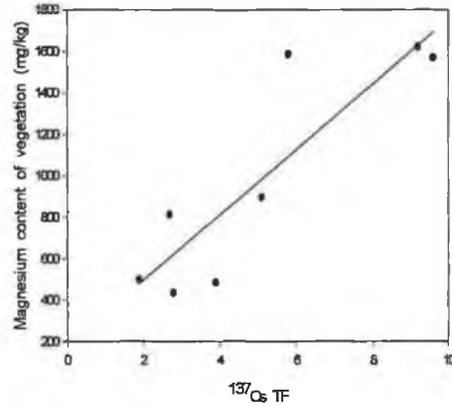


Figure 26: Plot of  $^{137}\text{Cs}$  TF values versus magnesium content of vegetation



Figures 25 - 26:  $^{137}\text{Cs}$  TF versus calcium and magnesium content of vegetation.

Figure 27: Plot of  $^{40}\text{K}$  TF values versus Soil CEC

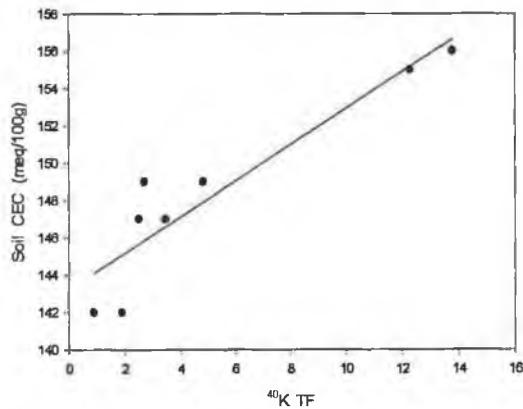


Figure 27:  $^{40}\text{K}$  TF versus soil CEC

Potassium-40 TF values showed no correlation with the nutritional status of the vegetation. Potassium-40 TF values showed little correlation with the soil physiochemical properties studied (Table 17) with the exception of soil cation exchange capacity. A Spearman correlation coefficient of 0.93 ( $p < 0.001$ ) indicates a strong relationship between  $^{40}\text{K}$  transfer to vegetation and the soil cation exchange capacity (Figure 27). As the soil CEC is a measure of the available cations present in the soil, then an increase in soil CEC would result in an increase in plant available nutrients in the soil (Ross, 1995).

### 3.1.7 Summary for Atlantic Blanket Bog.

The results of this study indicate that for the Atlantic blanket bog environment studied, uptake of  $^{137}\text{Cs}$  in vascular plants is not dependent on plant species studied as TF values for *C. vulgaris* and *E. vaginatum* are statistically similar. However in the case of *C. vulgaris* uptake of  $^{137}\text{Cs}$  is dependent on the plant compartment as the leaves have been found to accumulate significantly more  $^{137}\text{Cs}$  than the stems. This indicates that the highest concentration of  $^{137}\text{Cs}$  in *C. vulgaris* remains in the plant portions grazed by animals therefore possibly leading to human contamination through food consumption. Transfer of  $^{137}\text{Cs}$  to vegetation was also found to be positively correlated with the nutrient status of vegetation, in particular the calcium and magnesium content of vegetation.

Transfer factor values for  $^{40}\text{K}$  indicate that its uptake in vascular plants is dependent on plant species as significantly higher concentrations were found in *E. vaginatum* in comparison to *C. vulgaris*. However, unlike  $^{137}\text{Cs}$  its concentrations in *C. vulgaris* leaves and stems are not significantly different.

The concentration of both  $^{137}\text{Cs}$  and  $^{40}\text{K}$  found in moss samples were in general lower than those found in vascular plants probably reflecting the mode of uptake of radionuclides by moss.

## 3.2 Upland Blanket Bog

The upland blanket bog is located in the Cronamuck valley in Donegal along the Atlantic coast of Ireland. The region lies on the north-eastern part of the Barnesmore pluton, an isolated granite some 52 km<sup>2</sup> in area which forms the Bluestack mountain range in central Donegal. The indigenous vegetation is a mixture of *Calluna vulgaris* (ling heather), *Eriophorum vaginatum* (bog cotton), *Myrica gale* (bog myrtle), *Erica tetralix* (cross-leaved heath), *Molinia caerulea* (purple moor-grass) and *Sphagnum* mosses.

Soil and vegetation samples were collected from eight sites within the upland blanket bog. The distribution of radionuclides within the soil and the soil physiochemical characteristics are discussed in sections 3.2.1 and 3.2.2 respectively. Radionuclide and nutrient content of vegetation indigenous to this ecosystem are discussed in sections 3.2.3 and 3.2.4 respectively. Transfer factors were calculated for each radionuclide and are discussed in section 3.2.5 and correlations between these transfer factors and the chemical characteristics of the soil and vegetation are discussed in section 3.2.6. The main focus of the upland blanket bog discussion is based on the radionuclide TF values obtained and any significant correlations found between these and the chemical parameters studied in order to meet the main objectives of the this study. Section 3.2.7 highlights the most significant finding from the investigation within this ecosystem.

### 3.2.1 Radionuclide distribution in soil

The radionuclide distribution within the soil at sites D1 – D8 was determined by analysing the <sup>238</sup>U, <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>40</sup>K and <sup>137</sup>Cs content of three soil layers i.e. 0-5 cm, 5-10 cm 10-20 cm respectively at eight sites (D1 – D8). Results are reported in sections 3.1.1.1 to 3.1.1.2 below and outlined in Table 22.

### 3.2.1.1 $^{238}\text{U}$ , $^{226}\text{Ra}$ and $^{228}\text{Ra}$ distribution in soil

Uranium-238 activities in the upland blanket bog soil ranged from 5 Bq/kg to 36 Bq/kg with an average concentration of 19.1 Bq/kg for the eight sites sampled. The average  $^{238}\text{U}$  concentration of 33.3 Bq/kg at site D6 was significantly higher than its concentration at three other sites D4, D5 and D5 where the average concentration of  $^{238}\text{U}$  were 13.7 Bq/kg, 10.7 Bq/kg and 9.0 Bq/kg respectively (One Way ANOVA,  $p < 0.04$ ). Uranium-238 soil concentrations at all other sites were statistically similar (One Way ANOVA,  $p > 0.05$ ). The concentration of  $^{238}\text{U}$  in the three soil layers were not significantly different indicating that levels of this radioisotope remained relatively constant throughout the top 20 cm of soil (One Way ANOVA,  $p = 0.814$ ) as illustrated in Figures 28- 35.

Ra-226 concentration in the soil ranged from 4 Bq/kg to 31 Bq/kg with an average concentration for all sites sampled of 12.8 Bq/kg. The average concentration of  $^{226}\text{Ra}$  was statistically similar for all sites sampled, as is the case with the distribution of  $^{226}\text{Ra}$  within the top 20 cm of the soil. The concentration of  $^{226}\text{Ra}$  in the three soil layers 0-5 cm, 5-10 cm and 10-20 cm were statistically similar (ONE Way ANOVA,  $p = 0.593$ ).

The relationship between  $^{238}\text{U}$  and  $^{226}\text{Ra}$  in the soil was examined using the  $^{226}\text{Ra}/^{238}\text{U}$  equilibrium ratio outlined below.

$$^{226}\text{Ra}/^{238}\text{U} \text{ Equilibrium Ratio} = \frac{^{226}\text{Ra activity in soil (Bq/kg)}}{^{238}\text{U activity in soil (Bq/kg)}}$$

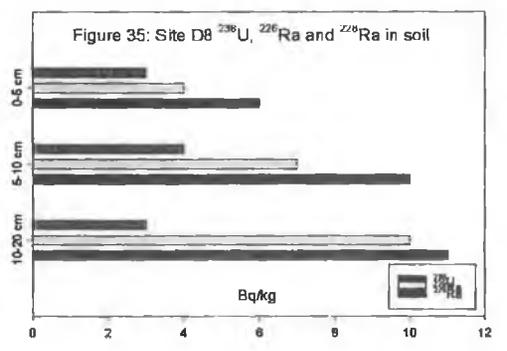
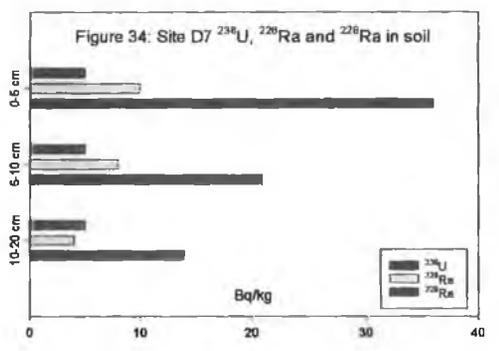
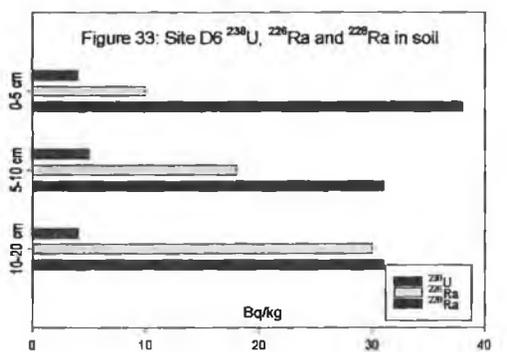
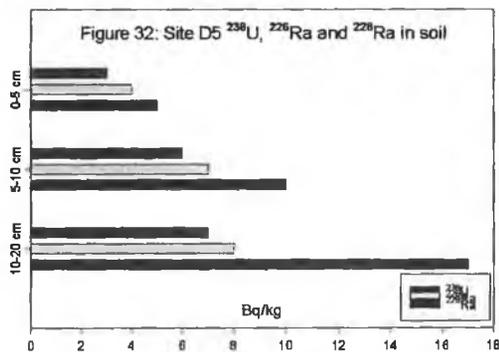
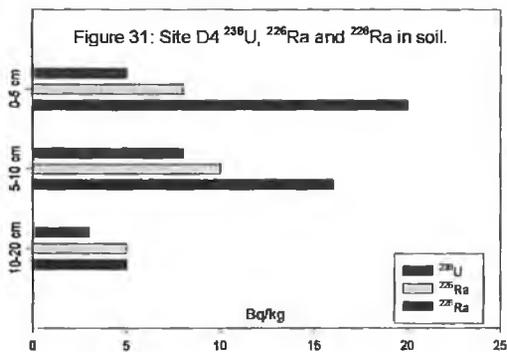
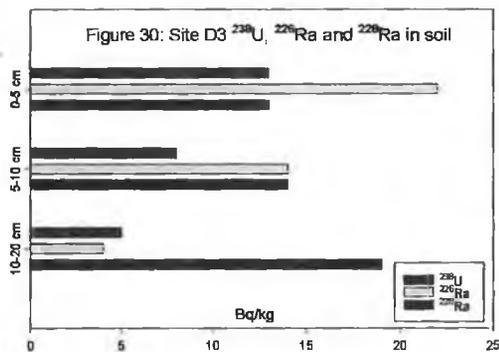
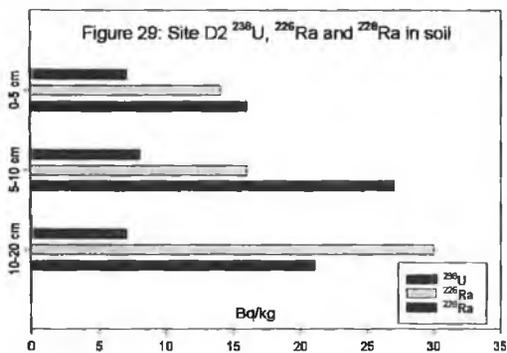
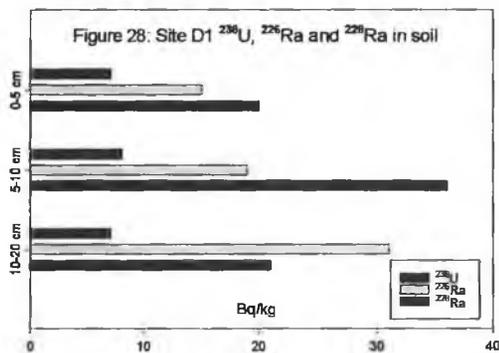
The purpose of this ratio is to ascertain the distribution of  $^{226}\text{Ra}$  in the soil in comparison to  $^{238}\text{U}$  due to the fact that  $^{226}\text{Ra}$  originated in the soil from the decay of  $^{238}\text{U}$ . The ratio for each site ranged from 0.31 to 0.94 indicating that the  $^{238}\text{U}$  concentration in the soil was higher than  $^{226}\text{Ra}$  at all sites however comparisons between the actual concentrations show that the differences were not statistically significant (One Way ANOVA,  $p = 0.97$ ). The fact that  $^{226}\text{Ra}$  is slightly lower in the soil at each site may indicate that  $^{226}\text{Ra}$  is being

leached to the lower layers of soil or that  $^{238}\text{U}$  is being preferentially retained in comparison to  $^{226}\text{Ra}$  in the upper layers of soil.

Site	Soil Layer	$^{238}\text{U}$ (Bq/kg)	$^{226}\text{Ra}$ (Bq/kg)	$^{228}\text{Ra}$ (Bq/kg)	$^{40}\text{K}$ (Bq/kg)	$^{137}\text{Cs}$ (Bq/kg)
D1	0-5 cm	20 ± 4	15 ± 1	7 ± 0.7	75 ± 6	262 ± 17
	5-10 cm	36 ± 4	19 ± 1	8 ± 0.7	67 ± 5	105 ± 7
	10-20 cm	21 ± 4	31 ± 2	7 ± 0.7	52 ± 3	16 ± 1
D2	0-5 cm	16 ± 6	14 ± 1	7 ± 0.7	90 ± 5	529 ± 35
	5-10 cm	27 ± 4	16 ± 1	8 ± 0.6	96 ± 7	166 ± 11
	10-20 cm	21 ± 3	30 ± 2	7 ± 0.6	54 ± 4	16 ± 1
D3	0-5 cm	13 ± 1	22 ± 2	13 ± 1	111 ± 9	186 ± 14
	5-10 cm	14 ± 1	14 ± 1	8 ± 0.6	56 ± 4	128 ± 10
	0-20 cm	19 ± 2	4 ± 0.5	5 ± 0.4	44 ± 4	52 ± 4
D4	0-5 cm	20 ± 2	8 ± 0.7	5 ± 0.4	40 ± 4	280 ± 22
	5-10 cm	16 ± 1	10 ± 1	8 ± 0.7	29 ± 3	110 ± 9
	10-20 cm	5 ± 0.5	5 ± 0.5	3 ± 0.4	17 ± 2	21 ± 2
D5	0-5 cm	5 ± 0.5	4 ± 0.5	3 ± 0.4	27 ± 3	344 ± 28
	5-10 cm	10 ± 1	7 ± 0.8	6 ± 0.5	25 ± 3	170 ± 14
	0-20 cm	17 ± 1	8 ± 0.8	7 ± 0.5	20 ± 2	30 ± 2
D6	0-5 cm	38 ± 3	10 ± 1	4 ± 0.4	40 ± 4	398 ± 32
	5-10 cm	31 ± 3	18 ± 2	5 ± 0.4	30 ± 3	133 ± 11
	10-20 cm	31 ± 3	30 ± 3	4 ± 0.4	29 ± 3	49 ± 4
D7	0-5 cm	36 ± 4	10 ± 1	5 ± 0.4	30 ± 3	307 ± 25
	5-10 cm	21 ± 2	8 ± 1	5 ± 0.4	25 ± 3	163 ± 13
	10-20 cm	14 ± 1	4 ± 0.5	5 ± 0.4	14 ± 1	43 ± 3
D8	0-5 cm	6 ± 0.7	4 ± 0.5	3 ± 0.4	28 ± 3	328 ± 26
	5-10 cm	10 ± 1	7 ± 0.6	4 ± 0.4	20 ± 2	184 ± 15
	10-20 cm	11 ± 1	10 ± 1	3 ± 0.4	17 ± 2	50 ± 4

Table 22:  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  levels in soil at sites D1 – D8

Ra-228 levels in the soil ranged from 3 Bq/kg to 13 Bq/kg at the eight sites sampled and these results are used as an indicator of the levels of  $^{232}\text{Th}$  within the soil. The average concentration of  $^{228}\text{Ra}$  at each of the eight sites was statistically similar (One Way ANOVA,  $p = 0.058$ ). A similar observation is made when comparing  $^{228}\text{Ra}$  in the soil layers as results indicate that the average concentration of the three soil layers is statistically similar (One Way ANOVA,  $p = 0.514$ ). These results indicate that the  $^{228}\text{Ra}$  levels remain relatively constant within the upper 20 cm of soil in the upland blanket bog based on the eight sites sampled.



Figures 28-35:  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in soil at sites D1 – D8

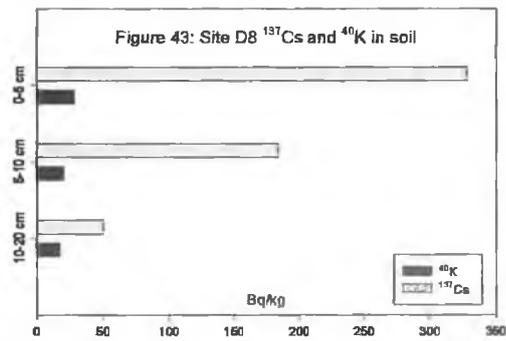
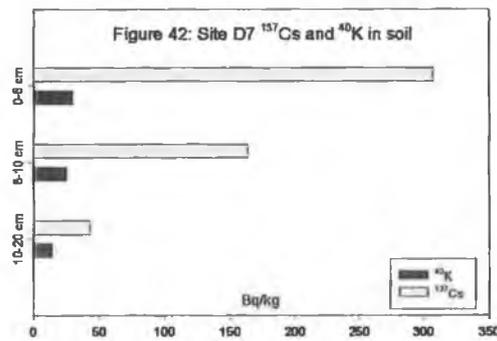
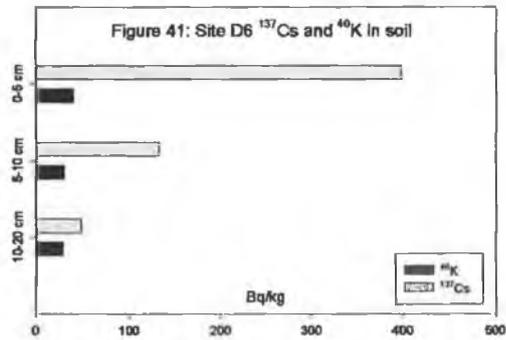
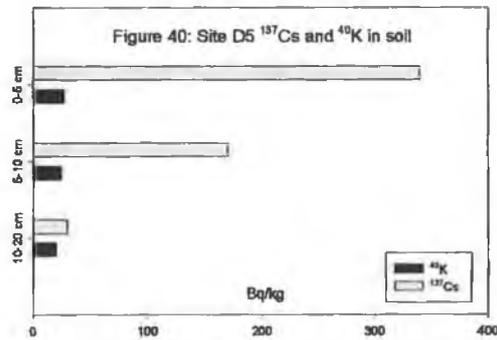
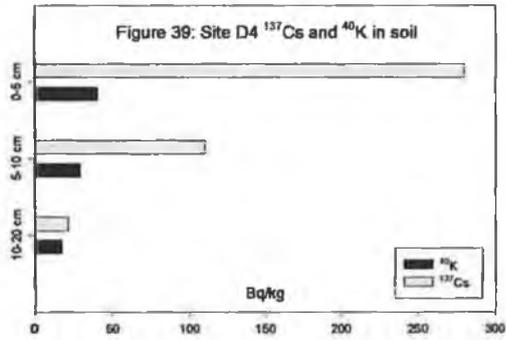
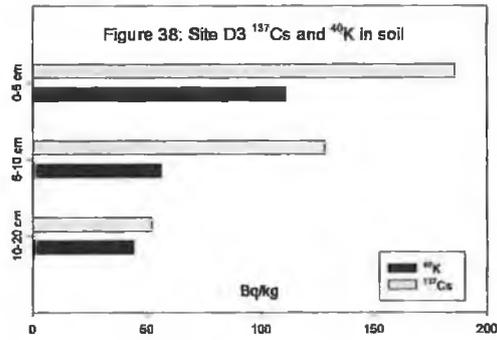
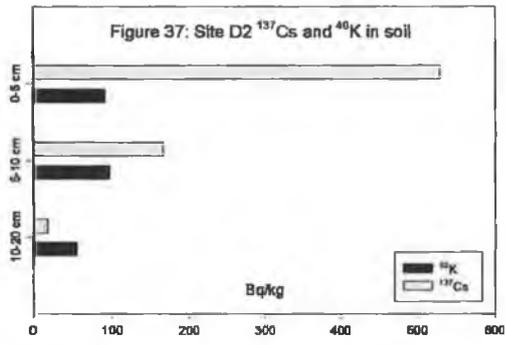
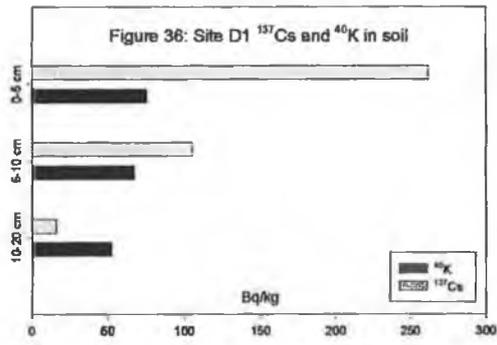
The levels of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in the soil at all sites were compared and results indicate that while  $^{238}\text{U}$  levels appear to be highest in the soil, followed by  $^{226}\text{Ra}$  and lowest for  $^{228}\text{Ra}$ , the average concentrations for each of the radionuclides are statistically similar (One Way ANOVA,  $p > 0.70$ ).

### 3.2.1.2 $^{40}\text{K}$ and $^{137}\text{Cs}$ distribution in soil

Levels of  $^{40}\text{K}$  in the soil ranged from 14 Bq/kg to 111 Bq/kg with an overall average concentration for the eight sites sampled of 43.2 Bq/kg. The average concentration of  $^{40}\text{K}$  in the soil at site D2 was 80.0 Bq/kg and was significantly higher than the concentration in the soil at sites D4, D5 and D7 of 28.7 Bq/kg, 24.0 Bq/kg and 23.0 Bq/kg respectively (One Way ANOVA,  $p < 0.027$ ). The average concentration of  $^{40}\text{K}$  in the soil at site D3 was also significantly higher than levels found at site D7, where average levels of  $^{40}\text{K}$  at both sites were 70.3 Bq/kg and 23.0 Bq/kg respectively. Potassium-40 concentrations at all other sites were statistically similar (One Way ANOVA,  $p > 0.05$ ).

Levels of  $^{40}\text{K}$  remained relatively constant throughout the upper 20 cm of the soil and while levels in the top 5 cm of the soil are higher than those in the 10-20 cm layer as illustrated in Figures 36 – 43, results indicate that the differences between these levels are not statistically significant (One Way ANOVA,  $p = 0.296$ ). The levels of plant available potassium were assessed in the soil and these results indicate that like  $^{40}\text{K}$ , concentrations of plant available potassium (Table 23) were higher in the 0-5 cm layer than the 10-20 cm layer however unlike  $^{40}\text{K}$  the differences between layers were statistically different. The levels of plant available potassium in the 0-5 cm soil layers were significantly higher than the two lower layers i.e. 5-10 cm and 10-20 cm (One Way ANOVA,  $p < 0.002$ ). The levels of plant available potassium in the two lower layers were found to be statistically similar (One Way ANOVA,  $p > 0.899$ ).

Caesium-137 levels in the soil ranged from 16 Bq/kg to 529 Bq/kg with an overall average concentration of 169.6 Bq/kg for the eight sites sampled. The average concentration of



Figures 36 ~ 43: <sup>137</sup>Cs and <sup>40</sup>K in soil at sites D1 – D8

$^{137}\text{Cs}$  in the three soil layers (0-20 cm) was calculated for each site and results indicate that  $^{137}\text{Cs}$  levels in the peat soil between sites are not significantly different (One Way ANOVA,  $p > 0.05$ ).

Caesium-137 levels are highest in the upper 5 cm of soil and levels decrease with increasing soil depth as illustrated in Figures 36 – 43. The average concentration of  $^{137}\text{Cs}$  in the 0-5 cm layer was 329.5 Bq/kg, which was significantly higher than  $^{137}\text{Cs}$  concentrations of 187.4 Bq/kg and 110.3 Bq/kg found in 5-10 cm and 10-20 cm layers respectively (One Way ANOVA,  $p < 0.001$ ). The average concentration of  $^{137}\text{Cs}$  found in the 5-10 cm layer was also significantly higher than levels found in 10-20 cm layer. These results indicate that  $^{137}\text{Cs}$  is being retained in the uppermost layers of the soil where root material is prominent. These results are in agreement with the results obtained in the Atlantic blanket bog mentioned previously and with previous studies which have shown that radiocaesium added to organic soil remains plant-available for extended periods of time and after uptake is recycled within the rooting zone of the vegetation in the form of decomposing plant material (Barber, 1964, Shand *et al*, 1994, Valcke and Cremers, 1994, Livens *et al*, 1991, Shaw and Bell, 2001 and Staunton *et al*, 2002).

The decrease in  $^{137}\text{Cs}$  activity with increasing soil depth is best described using a log function as illustrated in Figure 44 where an inverse relationship was found to be present between log average  $^{137}\text{Cs}$  activity as a function of soil depth (linear regression:  $r^2 = 0.977$ ). A similar relationship was noted for  $^{137}\text{Cs}$  activity in the Atlantic blanket bog soil (section 3.1.1.2), however results for the upland blanket bog appear to indicate that  $^{137}\text{Cs}$  is more preferentially retained in the upper 10 cm of the soil in comparison to the 10 - 20 cm layer when compared to the Atlantic blanket bog (Figures 20 and 44).

Figure 44: Log  $^{137}\text{Cs}$  activity versus soil depth for Sites D1- D8

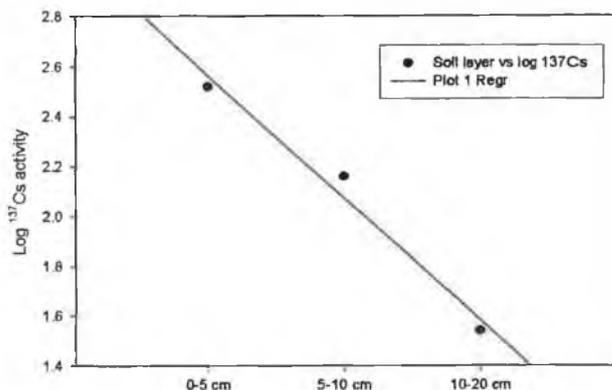


Figure 44: Log  $^{137}\text{Cs}$  activity versus soil depth for sites D1 – D8.

Comparisons between radionuclides levels in the upland blanket bog soils indicate that levels of  $^{137}\text{Cs}$  in the soil were significantly higher than levels of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{40}\text{K}$  respectively (One Way ANOVA,  $p < 0.001$ ). Levels of  $^{40}\text{K}$  in the soil were also significantly higher than  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  concentrations (One Way ANOVA,  $p < 0.01$ ). The average concentration of  $^{40}\text{K}$  (43.2 Bq/kg) appears to be higher than the average concentration of  $^{238}\text{U}$  (19.4 Bq/kg) however the differences were not statistically significant (One Way ANOVA,  $p = 0.19$ ). Levels of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  were all statistically similar (One Way ANOVA,  $p > 0.70$ ).

### 3.2.2 Soil Physiochemical properties

The physiochemical characteristics of the soil were extensively studied in order to ascertain if any of these parameters had a significant impact on the uptake of the radionuclide by vegetation. The physiochemical parameters chosen were the same as those chosen for the Atlantic blanket bog as outlined in section 3.1.2. The results for each of these parameters are presented in Table 23 and are discussed in detail within this section. Correlations between these parameters and radionuclide uptake in vegetation are discussed in section 3.2.6.

The moisture content of the soil varied between 84.0 % and 95.0% for the sites sampled with an average value of 90.4 % for the eight sites. The moisture content of the soil at site D3 was significantly higher than the soil at sites D1, D2, D4 and D6 (One Way ANOVA,  $p < 0.017$ ). The moisture content of the soil at site D8 was significantly higher than at sites D1, D2, D4 and D6 (One Way ANOVA,  $p < 0.025$ ). The moisture content of the soil at sites D5 and D7 was significantly greater than the soil at sites D1 and D2 (One Way ANOVA,  $p < 0.002$ ). The moisture content of the soil at site D6 was also significantly higher than the soil at site D1 (One Way ANOVA,  $p = 0.035$ ). While there appears to be significant difference between the moisture content of the soil at the majority of sites, the distribution of moisture within the top 20 cm of soil appears to remain relatively constant at each site. Comparisons between the three soil layers indicate that the differences between moisture of the individual soil layers are not statistically significant (One Way ANOVA,  $p = 0.853$ ).

Soil pH ranged from 4.1 to 4.7 with an overall average value of 4.4 for all sites sampled. Soil pH at sites D5 and D8 were significantly higher than the pH of the soil at sites D1, D2, D3, D4 and D6 respectively (One Way ANOVA,  $p < 0.023$ ). Soil pH at sites D3, D6 and D7 was also significantly higher than sites D2 and D4 and in the case of site D7 the soil pH is also significantly higher than the soil pH at site D1 (One Way ANOVA,  $p < 0.023$ ). The pH of the soil at all other sites was statistically similar (One Way ANOVA,  $p > 0.05$ ). The pH of the soil in the upper 20 cm of the soil at all sites sampled remained relatively constant as results indicate the differences between the average values for the three soil layers are not statistically significant (One Way ANOVA,  $p = 0.845$ ).

The organic matter content of the peat soil ranged from 80.3 % to 98.2 % (Table 21) with an overall average value for the organic matter content of the soils at the eight sites sampled of 93.1 %. The organic matter content of the soil at sites D3, D4, D5, D7 and D8 was significantly higher than the organic matter content of the soil at sites D1 and D7 (One Way ANOVA,  $p < 0.007$ ). This organic matter content of the soil at sites D4, D7 and D8 was also significantly higher than that of the soil at site D6 (One Way ANOVA,  $p < 0.049$ ). The organic matter content of the soil at all other sites were statistically similar

(One Way ANOVA,  $p > 0.05$ ). The levels of organic matter in the upper 20 cm of the soil appears to be relatively constant as the differences between the overall mean values for the three soil layers are not significantly different (One Way ANOVA,  $p = 0.716$ ).

Site	Site	Moisture Content %	pH	Organic Matter %	Available Magnesium mg/kg	Available Sodium mg/kg	Available Calcium mg/kg	Available Potassium mg/kg	CEC of Soil meq/100g
D 1	0-5 cm	87.3	4.3	80.3	184	216	127	266	117
	5-10 cm	86.8	4.2	86.8	178	143	194	134	99
	10-20 cm	84.0	4.1	86.4	258	144	178	46	112
D 2	0-5 cm	84.7	4.2	88.7	256	208	228	273	127
	5-10 cm	88.7	4.1	82.6	196	148	147	154	106
	10-20 cm	88.3	4.1	88.9	247	150	171	98	116
D 3	0-5 cm	93.5	4.4	94.9	355	151	323	107	172
	5-10 cm	94.4	4.4	96.4	262	112	262	37	154
	10-20 cm	93.5	4.4	96.8	470	177	311	111	147
D 4	0-5 cm	88.2	4.1	97.0	410	150	392	139	110
	5-10 cm	89.7	4.2	97.1	370	153	393	127	142
	10-20 cm	88.7	4.2	97.3	550	199	271	111	152
D 5	0-5 cm	91.8	4.6	96.8	483	187	406	180	174
	5-10 cm	91.6	4.6	97.1	521	226	369	126	116
	10-20 cm	92.2	4.7	95.9	428	253	344	104	147
D 6	0-5 cm	90.9	4.5	89.5	259	203	240	272	124
	5-10 cm	89.0	4.3	85.3	178	135	99	72	101
	10-20 cm	89.3	4.4	94.5	339	267	144	115	107
D 7	0-5 cm	92.9	4.7	96.8	367	190	476	198	115
	5-10 cm	92.3	4.5	95.8	459	181	386	116	142
	10-20 cm	91.8	4.5	98.2	420	167	264	99	152
D 8	0-5 cm	92.2	4.7	98.2	654	226	1041	153	146
	5-10 cm	95.0	4.7	96.4	284	124	484	67	155
	10-20 cm	93.6	4.7	97.4	423	155	632	67	177

Table 23: Soil physiochemical properties: Sites D1 – D8

Levels of plant available nutrients, magnesium, sodium, calcium and potassium were examined in the soil, in order to ascertain the nutrient status of the soil and to determine if levels of these nutrients present in the soil have an impact on the uptake of radionuclides by vegetation within this ecosystem. This was also the case for the CEC of the soil, which is a measure of the soils ability to retain and exchange cations within the soil.

The average levels of plant available calcium in the soil ranged from 161 mg/kg to 719 mg/kg and the average concentration of calcium in the soil at site D8 (719 mg/kg) was statistically higher than the concentrations in the soil at all other sites sampled (One Way ANOVA,  $p < 0.039$ ). The levels of plant available calcium in the soil at the other seven sites sampled were all statistically similar (One Way ANOVA,  $p > 0.05$ ). The concentration of calcium in the individual soil layers at each site was found to be

statistically similar indicating that the concentration of calcium remained constant in the upper 20 cm of soil in the bog ecosystem (One Way ANOVA,  $p = 0.443$ ). Average levels of plant available magnesium in the soil ranged from 207 mg/kg to 477 mg/kg for the sites sampled and the average level of magnesium in the soil at site D5 of 477 mg/kg was statistically higher than the level of 207 mg/kg found in the soil at site D1 (One Way ANOVA,  $p = 0.04$ ). The levels of magnesium at all other sites were statistically similar (One Way ANOVA,  $p > 0.05$ ). The concentration of magnesium in the individual soil layers at each site was found to be statistically similar (One Way ANOVA,  $p = 0.40$ ).

Average levels of plant available sodium in the soil ranged 147 mg/kg to 222 mg/kg and the average concentration at each site was statistically similar (One Way ANOVA,  $p = 0.458$ ). The concentration of sodium in the individual soil layers were found to be statistically similar (One Way ANOVA,  $p = 0.098$ ), indicating that that the concentration of sodium in the upper 20 cm of the bog soil remains relatively constant. The average levels of plant available potassium in the soil ranged from 85 mg/kg to 175 mg/kg and the average concentration in the sites at each site was statistically similar (One Way ANOVA,  $p = 0.80$ ). The levels of plant available potassium were assessed in the soil layers and results indicate the levels of plant available potassium in the 0-5 cm soil layers were significantly higher than the two lower layers i.e. 5-10 cm and 10-20 cm (One Way ANOVA,  $p < 0.002$ ). The levels of plant available potassium in the two lower layers were found to be statistically similar (One Way ANOVA,  $p > 0.899$ ).

The levels of plant available calcium, magnesium, sodium and potassium were compared and results indicate that their concentrations in the soil decrease in the following order:

$$\text{magnesium} > \text{calcium} > \text{sodium} > \text{potassium}$$

The average concentration of magnesium, calcium, sodium and potassium in the soil were 356 mg/kg, 328 mg/kg, 176 mg/kg and 132 mg/kg respectively. The trend found in the soil in this ecosystem is in agreement with the trend found for available nutrients in the Atlantic blanket bog soil (section 3.1.2). The concentration of plant available magnesium in soil was significantly higher than levels of plant available sodium and potassium (One Way ANOVA,  $p < 0.012$ ). Concentrations of plant available calcium

were significantly higher than levels of plant available sodium and potassium (One Way ANOVA,  $p < 0.04$ ). The levels of plant available potassium and sodium were statistically similar as is the case with calcium and magnesium levels in the in the soil (One Way ANOVA,  $p > 0.84$ ).

The cation exchange capacity of the soil ranged from 109 meq/100g to 159 meq/100g and the CEC of the soil at site D8 was significantly higher than that of the soil at site D1 (One Way ANOVA,  $p = 0.047$ ). The CEC of the soil at all other sites were statistically similar (One Way ANOVA,  $p > 0.05$ ). The CEC of the soil remained relatively constant in the four soil layers indicating that the CEC of the soil is similar throughout the upper 20 cm of the soil at each site sampled (One Way ANOVA,  $p = 0.61$ ).

In summary the soil physiochemical properties varied throughout the sites studied within the ecosystem. The moisture content of the soil varied throughout the ecosystem for the sites sampled and soils with the highest levels of moisture were found at site D3, D8, D7 and D5 respectively. The pH of the soil was also found to vary significantly as is the case with the organic matter content of soil. However in the case of the three aforementioned parameters the levels remained relatively constant for the three soil layers at each site. In the case of available nutrients levels in the soil, potassium and sodium levels remained relatively constant for the eight sites sampled. Levels of available calcium remained constant between sites with the exception of site D8 which contained levels that were significantly higher than all other sites sampled. The levels of available magnesium also remained relatively constant between sites with the exception of site D5 that had levels significantly higher than site D1. The concentration of calcium, magnesium and sodium are relatively constant in the upper 20 cm of soil whereas the levels of potassium in the upper 5 cm is significantly greater than the levels found in the lower 15 cm of soil. The CEC of the soil was significantly higher at site D8 in comparison to site D1 and remained relatively constant throughout the layers of the soil for all sites sampled. Table 24 compares the concentrations of the aforementioned soil properties at the eight sites sampled and shows the results in descending however as

stated previously the differences between results may not be statistically significant in all cases.

Soil Property	Sites
Moisture Content	D3 > D8 > D7 > D5 > D6 > D4 > D2 > D1
pH	D8 > D5 = D7 > D6 = D3 > D4 = D1 > D2
Organic Matter	D8 > D4 > D7 > D5 > D3 > D6 > D2 > D1
Available Magnesium	D5 > D8 > D4 > D7 > D3 > D6 > D2 > D1
Available Sodium	D5 > D6 > D7 > D2 > D8 = D1 > D4 > D3
Available Calcium	D8 > D7 > D5 > D4 > D3 > D2 > D1 > D6
Available Potassium	D2 > D6 > D1 > D7 > D5 > D4 > D8 > D3
CEC	D8 > D3 > D5 > D7 > D4 > D2 > D6 > D1

Table 24: Comparison of soil physiochemical properties at sites D1 – D8.

The impact of the soil physiochemical properties studied on radionuclide transfer factors and any significant correlation found between parameters is discussed in detail in section 3.2.6.

### 3.2.3 Radionuclide content of vegetation

Levels of  $^{226}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in the vegetation were analyzed by gamma spectrometry,  $^{238}\text{U}$  was analyzed by gamma spectrometry, alpha spectrometry and ICP-MS while  $^{228}\text{Ra}$  ( $^{232}\text{Th}$ ) was analyzed using both gamma spectrometry and alpha spectrometry. The results for all radionuclides are outlined in Table 25. The concentrations of  $^{238}\text{U}$  and  $^{228}\text{Ra}$  ( $^{232}\text{Th}$ ) in vegetation were below the limits of detection for the gamma and alpha spectrometry methods outlined in section 2.5.1 – 2.5.2. In the case of alpha spectrometry only a selection of the vegetation samples were analysed (Table 25), and the levels of both radionuclides were below the limits of detection. The levels of  $^{238}\text{U}$  in all vegetation samples were also below the limits of detection for the ICP-MS method outlined in

section 2.5.3. Radium-226 was detectable in the majority of vegetation using gamma spectrometry and  $^{40}\text{K}$  and  $^{137}\text{Cs}$  were detectable in all vegetation as outlined in Table 25.

Radium-226 levels in the vegetation ranged from 1 Bq/kg to 26 Bq/kg, however the levels in the leaves of *Myrica gale* were below the limits of detection for the gamma spectrometry methods used (with the exception of one sample at site D6). Radium-226 concentration was highest in *Erica tetralix* species (8 – 26 Bq/kg) and levels found in this species of vegetation were higher than any other vegetation species sampled (One Way ANOVA,  $p < 0.004$ ). Radium-226 levels in *C. vulgaris*, *E. vaginatum*, *M. caerulae*, *S. moss* and *M. gale* (stems) were all statistically similar (One Way ANOVA,  $p > 0.05$ ). In relation to plant compartments the levels of  $^{226}\text{Ra}$  in *C. vulgaris* leaves and stems were found to be statistically similar. In the case of *M. gale* leaves and stems the concentrations of  $^{226}\text{Ra}$  in the stems ranged from 1 Bq/kg to 4.9 Bq/kg and the levels in the leaves were below the limits of detection with the exception of one plant sampled at site D6 (concentration of 3.2 Bq/kg  $^{226}\text{Ra}$  in *M. gale* leaves). While statistical analysis could not be carried out on this set of data (*M. gale* leaves data set too small), the results indicate that levels in the stems of *M. gale* are higher than those found in the leaves. The uptake of radium by these species of vegetation will be discussed in greater detail in relation to their TF values in section 3.2.5.

Potassium-40 concentrations in vegetation ranged from 14 Bq/kg to 410 Bq/kg (Table 25) and on average the lowest concentration was found in the *S. moss* species (34 Bq/kg) and the highest concentration found in *M. caerulae* species (180 Bq/kg). The average levels of  $^{40}\text{K}$  in *M. caerulae*, *M. gale*, *S. moss*, *E. tetralix*, *C. vulgaris* and *E. vaginatum* were all statistically similar (One Way ANOVA,  $p > 0.05$ ). The concentrations of  $^{40}\text{K}$  in the leaves and stems of both *C. vulgaris* and *M. gale* are not significantly different (One Way ANOVA,  $p > 0.05$ ) indicating that uptake of  $^{40}\text{K}$  by these plant species in this ecosystem is not dependent on plant compartment.

Site	Vegetation	<sup>238</sup> U		<sup>232</sup> Rn	<sup>226</sup> Ra		<sup>40</sup> K	<sup>137</sup> Cs
		(Bq/kg)		(Bq/kg)	(Bq/kg)		(Bq/kg)	(Bq/kg)
D 1	<i>Molinia caerulea</i>	< 0.19		<b>1.8 ± 0.4</b>	< 1.5		<b>234 ± 21</b>	<b>205 ± 14</b>
	<i>Myrica gale</i> leaves	< 0.12	< 0.20*	< 1.5	< 3.5	< 0.50*	<b>149 ± 16</b>	<b>101 ± 7</b>
	<i>Myrica gale</i> stems	< 0.16	< 0.18*	<b>1.5 ± 0.3</b>	< 2.0	< 0.80*	<b>87 ± 7</b>	<b>80 ± 5</b>
	Moss	< 0.10		<b>3.2 ± 0.4</b>	< 1.0		<b>14 ± 3</b>	<b>227 ± 15</b>
D 2	<i>Molinia caerulea</i>	< 0.24	< 0.51*	<b>7.1 ± 0.7</b>	< 1.5	< 0.65*	<b>156 ± 12</b>	<b>148 ± 10</b>
	<i>Calluna vulgaris</i> leaves	< 0.11		<b>5.9 ± 0.9</b>	< 2.8		<b>33 ± 8</b>	<b>632 ± 43</b>
	<i>Myrica gale</i> leaves	< 0.12		< 1.5	< 3.9		<b>177 ± 18</b>	<b>68 ± 4</b>
	<i>Myrica gale</i> stems	< 0.21		<b>3.7 ± 0.4</b>	< 0.8		<b>50 ± 3</b>	<b>44 ± 3</b>
D 3	<i>Molinia caerulea</i>	< 0.21		<b>3.9 ± 1</b>	< 4.6		<b>336 ± 27</b>	<b>222 ± 15</b>
	<i>Myrica gale</i> leaves	< 0.53	< 1.0*	< 2.1	< 4.4	< 0.95*	<b>40 ± 4</b>	<b>208 ± 17</b>
	<i>Myrica gale</i> stems	< 0.25	< 0.80*	<b>4.9 ± 0.5</b>	< 14	< 1.3*	<b>153 ± 12</b>	<b>52 ± 4</b>
	<i>Erica tetralix</i>	< 0.23		<b>14 ± 1</b>	< 5.0		<b>410 ± 33</b>	<b>119 ± 10</b>
D 4	<i>Molinia caerulea</i>	< 0.22		<b>1.9 ± 0.4</b>	< 2.7		<b>209 ± 14</b>	<b>18 ± 1</b>
	<i>Calluna vulgaris</i> leaves	< 0.20		<b>5.3 ± 0.9</b>	< 2.0		<b>90 ± 11</b>	<b>857 ± 55</b>
	<i>Calluna vulgaris</i> stems	< 0.17		<b>8.8 ± 0.8</b>	< 2.5		<b>39 ± 4</b>	<b>204 ± 17</b>
	<i>Erica tetralix</i>	< 0.20		<b>18 ± 2</b>	< 4.2		<b>141 ± 17</b>	<b>94 ± 6</b>
D 5	<i>Molinia caerulea</i>	< 0.19		<b>5.2 ± 0.5</b>	< 4.0		<b>205 ± 16</b>	<b>101 ± 8</b>
	<i>Calluna vulgaris</i> leaves	< 0.14	< 0.15*	<b>9.9 ± 1</b>	< 6.0	< 0.65*	<b>57 ± 5</b>	<b>553 ± 42</b>
	<i>Calluna vulgaris</i> stems	< 0.14	< 0.19*	<b>3.0 ± 0.4</b>	< 8.8	< 1.4*	<b>11 ± 1</b>	<b>212 ± 16</b>
	<i>Eriophorum vaginatum</i>	< 0.17		<b>3.2 ± 0.4</b>	< 3.5		<b>198 ± 23</b>	<b>439 ± 29</b>
D6	<i>Molinia caerulea</i>	< 0.20		<b>6.6 ± 0.8</b>	< 2		<b>29 ± 3</b>	<b>23 ± 2</b>
	<i>Myrica gale</i> leaves	< 0.58		<b>3.2 ± 0.4</b>	< 7		<b>94 ± 9</b>	<b>33 ± 3</b>
	<i>Myrica gale</i> stems	< 0.20		<b>3.9 ± 0.6</b>	< 5		<b>54 ± 9</b>	<b>34 ± 4</b>
	<i>Erica tetralix</i>	< 0.25	< 0.40*	<b>26.1 ± 2</b>	< 24	< 0.85*	<b>14 ± 3</b>	<b>55 ± 4</b>
	<i>Eriophorum vaginatum</i>	< 0.24		<b>2.5 ± 0.6</b>	< 7		<b>49 ± 8</b>	<b>143 ± 11</b>
	Moss	< 0.41		<b>6.1 ± 1</b>	< 7		<b>59 ± 12</b>	<b>70 ± 5</b>
D 7	<i>Molinia caerulea</i>	< 0.39	< 0.50*	<b>1.8 ± 0.4</b>	< 13	< 1.1*	<b>92 ± 9</b>	<b>53 ± 4</b>
	<i>Calluna vulgaris</i> leaves	< 0.16		<b>5.9 ± 0.8</b>	< 10		<b>47 ± 5</b>	<b>434 ± 33</b>
	<i>Calluna vulgaris</i> stems	< 0.27		<b>4.1 ± 0.6</b>	< 17		<b>20 ± 4</b>	<b>300 ± 23</b>
	<i>Myrica gale</i> leaves	< 0.26		< 3.5	< 8		<b>67 ± 13</b>	<b>29 ± 3</b>
	<i>Myrica gale</i> stems	< 0.27		<b>1.0 ± 0.2</b>	< 9		<b>56 ± 5</b>	<b>42 ± 4</b>
	<i>Erica tetralix</i>	< 0.44		<b>8.4 ± 1</b>	< 14		<b>28 ± 4</b>	<b>64 ± 5</b>
	<i>Eriophorum vaginatum</i>	< 0.26	< 0.69*	<b>3.9 ± 0.4</b>	< 10	< 1.0*	<b>126 ± 11</b>	<b>209 ± 20</b>
	Moss	< 0.43		< 2	< 24		<b>37 ± 4</b>	<b>242 ± 20</b>
D 8	<i>Calluna vulgaris</i> leaves	< 0.42	< 0.75*	<b>3.1 ± 0.4</b>	< 8	< 1.2*	<b>59 ± 6</b>	<b>496 ± 38</b>
	<i>Calluna vulgaris</i> stems	< 0.64	< 0.91*	<b>2.7 ± 0.3</b>	< 9	< 1.5*	<b>60 ± 6</b>	<b>435 ± 33</b>
	<i>Myrica gale</i> leaves	< 0.18		< 1.4	< 9		<b>243 ± 51</b>	<b>44 ± 4</b>
	<i>Myrica gale</i> stems	< 0.15		<b>1.4 ± 0.3</b>	< 10		<b>94 ± 8</b>	<b>36 ± 3</b>
	<i>Erica tetralix</i>	< 0.24		<b>9.8 ± 1</b>	< 9		<b>247 ± 20</b>	<b>81 ± 6</b>
	Moss	< 0.24		<b>2.4 ± 0.3</b>	< 14		<b>25 ± 2</b>	<b>84 ± 7</b>

\*<sup>238</sup>U and <sup>232</sup>Th results from alpha spectrometry, all other <sup>238</sup>U and <sup>232</sup>Th (<sup>226</sup>Ra) results were obtained using ICP-MS and gamma spectrometry respectively.

Table 25: Radionuclide concentrations in upland bog vegetation: Sites D1 – D8

The levels of  $^{137}\text{Cs}$  in vegetation ranged from 18 Bq/kg to 632 Bq/kg for the variety of species sampled and lowest concentrations of  $^{137}\text{Cs}$  were found in *M. gale* (average concentration of 64 Bq/kg) and the highest concentration found in *C. vulgaris* (average concentration of 441 Bq/kg). Caesium-137 levels in the leaves of *C. vulgaris* were higher than in all other plant species sampled and were also higher than levels found in the stems of *C. vulgaris* indicating that uptake of  $^{137}\text{Cs}$  in *C. vulgaris* is dependent on the portion of the plant (One Way ANOVA,  $p < 0.001$ ). The levels of  $^{137}\text{Cs}$  in the stems of *C. vulgaris* were also significantly higher than those found in *E. tetralix*, and in both the leaves and stems of *M. gale* (One Way ANOVA,  $p < 0.05$ ). Caesium-137 concentrations in *E. tetralix*, *E. vaginatum*, *M. caerulea*, *M. gale* and *S. moss* were not significantly different (One Way ANOVA,  $p > 0.05$ ). The average levels of  $^{137}\text{Cs}$  found in the leaves and stems of *M. gale* were 81 Bq/kg and 48 Bq/kg respectively and the differences between the levels of  $^{137}\text{Cs}$  were not significantly different (One Way ANOVA,  $p = 0.999$ ), indicating that for *M. gale* uptake of  $^{137}\text{Cs}$  within this ecosystem is not dependent on plant compartment.

The  $^{40}\text{K}$  and  $^{137}\text{Cs}$  content of *M. caerulea* were significantly higher than the  $^{226}\text{Ra}$  content (One Way ANOVA,  $p < 0.04$ ), while the  $^{40}\text{K}$  and  $^{137}\text{Cs}$  content were statistically similar (One Way ANOVA,  $p = 0.22$ ). The  $^{40}\text{K}$  and  $^{137}\text{Cs}$  content of *M. gale* were significantly higher than the  $^{226}\text{Ra}$  content (One Way ANOVA,  $p < 0.013$ ), while the  $^{40}\text{K}$  and  $^{137}\text{Cs}$  content were statistically similar (One Way ANOVA,  $p = 0.08$ ). The  $^{40}\text{K}$  content of *M. gale* leaves was significantly higher than the  $^{226}\text{Ra}$  concentration (One Way ANOVA,  $p < 0.05$ ). The concentration of  $^{137}\text{Cs}$  in *C. vulgaris* was significantly higher than  $^{226}\text{Ra}$  and  $^{40}\text{K}$  (One Way ANOVA,  $p < 0.001$ ), while the concentration of the latter two radionuclides were statistically similar (One Way ANOVA,  $p = 0.239$ ). Further investigation into the accumulation of radionuclides in plant compartments of *C. vulgaris* indicates that the concentration of  $^{137}\text{Cs}$  in the leaves of *C. vulgaris* was significantly higher than the concentration of both  $^{226}\text{Ra}$  and  $^{40}\text{K}$  in the leaves and stems and also higher than the concentration of  $^{137}\text{Cs}$  in the stems (One Way ANOVA,  $p < 0.001$ ). Results also indicate that  $^{137}\text{Cs}$  concentration in the stems of *C. vulgaris* was higher than the concentration of both  $^{40}\text{K}$  and  $^{226}\text{Ra}$  in the leaves and stems of *C. vulgaris* (One Way

ANOVA,  $p < 0.006$ ). The concentration of  $^{40}\text{K}$  and  $^{226}\text{Ra}$  in the leaves and stems of *C. vulgaris* were both statistically similar (One Way ANOVA,  $p = 0.41$ ). The concentration of  $^{40}\text{K}$  in *E. tetralix* was significantly higher than both  $^{226}\text{Ra}$  and  $^{137}\text{Cs}$  (One Way ANOVA,  $p = 0.04$ ), while the concentration of the latter two radionuclides were statistically similar (One Way ANOVA,  $p > 0.28$ ). The concentration of  $^{137}\text{Cs}$  in *E. vaginatum* was significantly higher than  $^{226}\text{Ra}$  (One Way ANOVA,  $p = 0.04$ ), while the concentration of  $^{137}\text{Cs}$  and  $^{40}\text{K}$  are statistically similar as is the case with  $^{40}\text{K}$  and  $^{226}\text{Ra}$  concentration in *E. vaginatum* (One Way ANOVA,  $p > 0.28$ ).

In general for all upland blanket bog vegetation sampled, the average concentration of  $^{137}\text{Cs}$  was statistically higher than the levels of  $^{226}\text{Ra}$  found present (One Way ANOVA,  $p = 0.008$ ) but was not significantly different than the levels of  $^{40}\text{K}$  found in the vegetation (One Way ANOVA,  $p > 0.19$ ). Potassium-40 levels as mentioned previously were statistically similar to the levels of  $^{137}\text{Cs}$  in vegetation and were also statistically similar to  $^{226}\text{Ra}$  levels found in the vegetation (One Way ANOVA,  $p > 0.19$ ).

The concentrations of  $^{137}\text{Cs}$ ,  $^{40}\text{K}$  and  $^{226}\text{Ra}$  in soil and vegetation were compared to assess if vegetation uptake was dependent on soil concentrations. Radium-226 concentrations in all vegetation appear to be independent of  $^{226}\text{Ra}$  in soil (Spearman correlation coefficient of 0.14,  $p = 0.44$ ). Potassium-40 concentrations in all vegetation appear to be independent of  $^{40}\text{K}$  levels in the soil and are also independent of the  $^{137}\text{Cs}$  levels in the soil (Spearman correlation coefficient of 0.10,  $p = 0.53$ ). Similar observations were also made for  $^{137}\text{Cs}$  levels in vegetation and soil where a Spearman correlation coefficient of -0.18 ( $p = 0.25$ ) indicates the absence of correlation between these two parameters as shown in Figure 47. No correlation appears to be evident between  $^{137}\text{Cs}$  levels in vegetation and  $^{40}\text{K}$  levels in the soil (Spearman correlation coefficient of -0.03,  $p = 0.87$ ). The concentration of the three radionuclides in upland vegetation and their corresponding concentration in the underlying soil are illustrated in Figures 45 – 47.

Figure 45: Plot of  $^{226}\text{Ra}$  in vegetation versus  $^{226}\text{Ra}$  in soil (upland blanket bog vegetation)

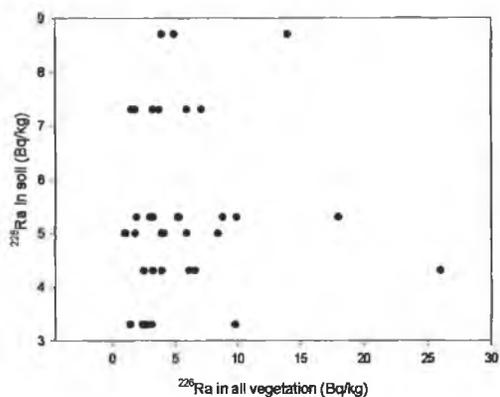


Figure 46: Plot of  $^{40}\text{K}$  in vegetation versus  $^{40}\text{K}$  in soil (upland blanket bog vegetation)

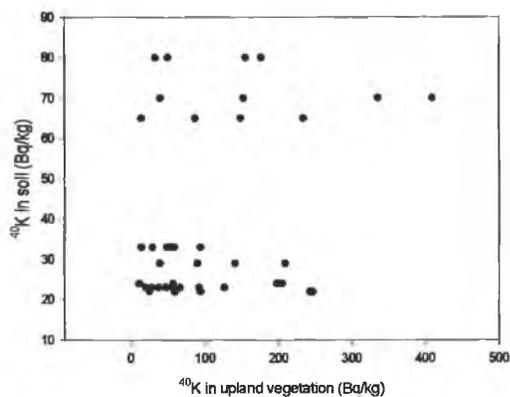
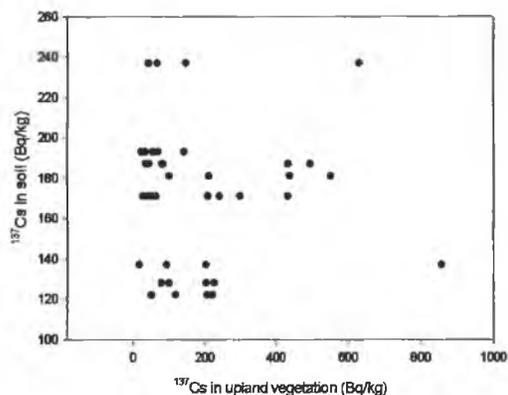


Figure 47: Plot of  $^{137}\text{Cs}$  in vegetation versus  $^{137}\text{Cs}$  in soil (upland blanket bog vegetation)



Figures 45 – 47: Correlation between radionuclide activity in soil and vegetation for upland blanket bog vegetation.

The relationship between soil and vegetation radionuclides is discussed in greater detail with regard to their TF values in section 3.2.5.

### 3.2.4 Nutrient content of vegetation

The calcium, magnesium and sodium content of the vegetation were analyzed for all vegetation sampled at sites D1 - D8 and results are outlined in Table 26. Correlations between these nutrients and radionuclide transfer factor values for all vegetation are discussed in detail in section 3.2.5.

Site	Vegetation	Calcium (mg/kg)	Magnesium (mg/kg)	Sodium (mg/kg)
<b>D 1</b>	<i>Molinia caerulea</i>	1700	1952	1005
	<i>Myrica gale</i> leaves	2636	2185	1390
	<i>Myrica gale</i> stems	2516	1759	735
	Moss	1618	1265	345
<b>D 2</b>	<i>Molinia caerulea</i>	1484	1929	2096
	<i>Calluna vulgaris</i> leaves	2771	2512	1178
	<i>Calluna vulgaris</i> stems	1314	1491	419
	<i>Myrica gale</i> leaves	2242	2432	999
	<i>Myrica gale</i> stems	1554	1045	331
<b>D 3</b>	<i>Molinia caerulea</i>	1088	1371	940
	<i>Myrica gale</i> leaves	1482	2561	1245
	<i>Myrica gale</i> stems	1915	1332	333
	<i>Erica tetralix</i>	1752	1294	957
	Moss	4815	2549	293
<b>D 4</b>	<i>Molinia caerulea</i>	1625	1214	415
	<i>Calluna vulgaris</i> leaves	2777	2185	1061
	<i>Calluna vulgaris</i> stems	870	855	439
	<i>Erica tetralix</i>	1595	1088	1049
<b>D 5</b>	<i>Molinia caerulea</i>	1280	1699	832
	<i>Calluna vulgaris</i> leaves	2912	2223	618
	<i>Calluna vulgaris</i> stems	1667	1291	536
	<i>Eriophorum vaginatum</i>	1483	1797	1297
	Moss	4744	2193	387
<b>D6</b>	<i>Molinia caerulea</i>	1253	797	202
	<i>Myrica gale</i> leaves	3265	1197	732
	<i>Myrica gale</i> stems	2532	1324	132
	<i>Erica tetralix</i>	2432	1023	455
	<i>Eriophorum vaginatum</i>	2421	1530	331
	Moss	3497	2262	448
<b>D 7</b>	<i>Molinia caerulea</i>	2195	819	214
	<i>Calluna vulgaris</i> leaves	4762	1559	231
	<i>Calluna vulgaris</i> stems	2012	1000	348
	<i>Myrica gale</i> leaves	3582	1140	661
	<i>Myrica gale</i> stems	2799	1051	378
	<i>Erica tetralix</i>	2682	973	526
	<i>Eriophorum vaginatum</i>	1991	985	363
	Moss	4525	2250	436
<b>D 8</b>	<i>Calluna vulgaris</i> leaves	4202	1658	452
	<i>Calluna vulgaris</i> stems	1838	1079	304
	<i>Myrica gale</i> leaves	3023	1617	1916
	<i>Myrica gale</i> stems	3966	1093	425
	<i>Erica tetralix</i>	2521	883	431
	Moss	5543	2349	422

Table 26: Nutrient concentration of vegetation: Sites D1 – D8

The concentration of calcium in vegetation ranged from 870 mg/kg in *C. vulgaris* (stems) to 4744 mg/kg in *S. moss*. The average concentration of calcium in each species of vegetation was calculated and results indicate that the highest concentration was found in *S. moss* species (4123 mg/kg), while the lowest concentration was found in *M. caerulae* (1518 mg/kg). The concentration of calcium in *S. moss* is significantly higher than that found in *M. caerulae*, *C. vulgaris* stems, *E. vaginatum*, *E. tetralix* and *M. gale* stems (One Way ANOVA,  $p < 0.014$ ). The concentration of calcium found in *C. vulgaris* is significantly higher than that found in *M. caerulae*. The concentration of calcium found in the leaves of *C. vulgaris* is significantly higher than the stems, indicating that calcium uptake by *C. vulgaris* is dependent on plant compartment (One Way ANOVA,  $p < 0.013$ ). In the case of *M. gale* the concentration of calcium in the leaves and stems are not significantly different indicating that unlike *C. vulgaris*, calcium uptake by this plant species is not dependent on the plant compartments sampled (One Way ANOVA,  $p = 1.0$ ). The concentration of calcium found in all other plant species was statistically similar (One Way ANOVA,  $p > 0.05$ ).

The concentration of magnesium in vegetation ranged from 855 mg/kg in *C. vulgaris* (stems) to 2549 mg/kg in *S. moss*. The average concentration of magnesium in each species of vegetation was calculated and results indicate that the highest concentration was found in *S. moss* species (2145 mg/kg), while the lowest concentration was found in *E. tetralix* (1060 mg/kg). The concentration of magnesium in *S. moss* was significantly higher than concentrations in *E. tetralix*, *C. vulgaris* stems and *M. gale* stems (One Way ANOVA,  $p < 0.02$ ) but was statistically similar to the concentrations found in *M. caerulae*, *E. vaginatum*, *M. gale* leaves and *C. vulgaris* leaves (One Way ANOVA,  $p > 0.05$ ). The concentration of magnesium in *C. vulgaris* leaves was significantly higher than concentrations found in *E. tetralix* and *C. vulgaris* stems and in the latter case results indicate that magnesium uptake is dependent on plant compartment (One Way ANOVA  $p < 0.04$ ). The concentration of magnesium in *M. gale* leaves and stems were statistically similar indicating that for this species uptake is not dependent on the plant compartments sampled (One Way ANOVA,  $p > 0.05$ ). The magnesium concentration in all other plant species was statistically similar (ONE Way ANOVA,  $p > 0.05$ ).

The concentration of sodium in vegetation ranged from 132 mg/kg in *M. gale* stems to 2096 mg/kg in *M. caerulea*. The average concentration of sodium in each species of vegetation was calculated and results indicate that the highest concentration was found in *M. gale* leaves (1157 mg/kg), while the lowest concentration was found in *S. moss* (381 mg/kg). The levels of sodium found in all species of vegetation sampled were not significantly different (One Way ANOVA,  $p > 0.05$ ). The concentration of sodium in the leaves and stems of both *C. vulgaris* and *M. gale* were both statistically similar indicating that for both species the uptake of sodium is not dependent on plant compartments sampled (ONE Way ANOVA,  $p > 0.05$ ).

The concentrations of the three nutrients were compared for each plant species sampled to assess if nutrient accumulation is dependent in plant species. The calcium, magnesium and sodium concentration in all *M. caerulea* samples was found to be statistically similar indicating that this species of grass does not appear to preferentially uptake any of the nutrients analyzed (One Way ANOVA  $p > 0.05$ ). The calcium and magnesium concentration of *M. gale* were both significantly higher than sodium levels present in this species and in the case of calcium the levels found in *M. gale* were also significantly higher than magnesium concentration (One Way ANOVA,  $p < 0.037$ ). Further investigation into the nutrient concentration of *M. gale* with regard to the leaves and stems indicates that the calcium concentration of the leaves of *M. gale* was significantly higher than the sodium concentration of both leaves and stems and the magnesium concentration of the leaves (One Way ANOVA,  $p < 0.002$ ). Results also indicate that the calcium concentration of the stems was significantly higher than the sodium concentration of both leaves and stems and the magnesium concentration of the stems (One Way ANOVA,  $p < 0.007$ ). The magnesium concentration of the leaves of *M. gale* was also found to be significantly higher than the sodium concentration found in the stems (One Way ANOVA,  $p < 0.002$ ).

The calcium and magnesium concentration of *S. moss* was significantly higher than sodium levels present and in the case of calcium, the levels present in the moss samples

were significantly higher than magnesium concentrations (One Way ANOVA,  $p < 0.007$ ). The calcium and magnesium levels found in *C. vulgaris* (stems and leaves together) was significantly higher than sodium levels present and calcium levels in this plant species were also significantly higher than magnesium levels found (One Way ANOVA,  $p < 0.008$ ). Further investigation into nutrient accumulation in *C. vulgaris* with regard to plant compartments (leaves and stems) indicates that the calcium concentration of the leaves is significantly higher than the sodium and magnesium concentration of both leaves and stems and also significantly higher than the calcium concentration of the stems (One Way ANOVA,  $p < 0.001$ ). The magnesium concentration of the leaves was found to be significantly higher than the sodium concentration of the leaves and stems (One Way ANOVA,  $p < 0.016$ ). The calcium concentration in the stems was found to be significantly higher than the sodium concentration in the stems (One Way ANOVA,  $p < 0.016$ ). The calcium concentration of *E. tetralix* was significantly higher than both the magnesium and sodium levels present (One Way ANOVA,  $p < 0.007$ ), while the concentration of the latter two nutrients were statistically similar (One Way ANOVA,  $p = 0.48$ ). The calcium concentration of *E. vaginatum* was significantly higher than sodium levels present (One Way ANOVA,  $p = 0.037$ ) but not significantly different than magnesium levels present (One Way ANOVA,  $p = 0.42$ ). The magnesium and sodium levels found present in *E. vaginatum* were not significantly different (One Way ANOVA,  $p = 0.20$ ).

Overall for the plant species studied, the calcium levels were significantly higher in all vegetation sampled in comparison to both magnesium and sodium levels present (One Way ANOVA,  $p < 0.012$ ). The magnesium levels were also significantly higher sodium levels when comparing all plant species (One Way ANOVA,  $p = 0.02$ ).

### 3.2.5 Transfer Factors for Upland Blanket Bog Vegetation

Transfer factor values were calculated for each radionuclide based on the concentration of radionuclide in vegetation in comparison to the average concentration in the upper 20

cm of underlying soil (section 2.2). Transfer factor values are the method used within this study to assess the uptake of radionuclides by vegetation within each ecosystem and as this method is used extensively within the literature the values found within this study are compared to similar published studies. One of the primary objectives of this study is to assess the uptake of radionuclide within vegetation indigenous to each ecosystem which is therefore accomplished through the TF method. Another primary objective is to correlate these TF values with soil physiochemical properties and the nutrient status of the vegetation and these results are discussed in detail in the next section and the results found within this study is compared to previous finding from similar studies.

Transfer factor values for  $^{226}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in vegetation are outlined in Table 27, however transfer factor values could not be calculated for  $^{238}\text{U}$  and  $^{228}\text{Ra}$  as the concentration of these radionuclides in vegetation were below the limits of detection for the methods outlined in sections 2.5.1 – 2.5.3. Transfer factor values were not calculated for *S. moss* as it lacks a well-developed root system.

Radium-226 transfer factor values on average range from 0.07 for *M. gale* (stems) to 2.4 for *E. tetralix* (Table 24). In the case of *M. gale*, the leaves were found to have the lowest TF value and  $^{226}\text{Ra}$  levels was only detected in one sample as the levels in the other five samples was below the limits of detection for the methods used. Transfer factor values for  $^{226}\text{Ra}$  were significantly higher in *E. tetralix* than *M. gale* (leaves and stems), *M. caerulea*, *C. vulgaris* (leaves and stems) and *E. vaginatum* (One Way ANOVA,  $p < 0.05$ ). Transfer factor values for all other species of vegetation sampled were statistically similar (One Way ANOVA,  $p > 0.05$ ). In the case of *C. vulgaris* and *M. gale* results indicate that  $^{226}\text{Ra}$  uptake is not dependent on plant compartment as TF values were not significantly different for the leaves and stems however in the latter case only one TF value was recorded for *M. gale* leaves (One Way ANOVA,  $p = 1.0$ ).

Site	Vegetation	<sup>238</sup> U TF	<sup>226</sup> Ra TF	<sup>228</sup> Ra TF	<sup>40</sup> K TF	<sup>137</sup> Cs TF
D 1	<i>Molinia caerulea</i>	< 0.007	<b>0.08 ± 0.02</b>	< 0.2	<b>3.60 ± 0.58</b>	<b>1.60 ± 0.21</b>
	<i>Myrica gale</i> leaves	< 0.005	< 0.08	< 0.07	<b>2.30 ± 0.41</b>	<b>0.80 ± 0.10</b>
	<i>Myrica gale</i> stems	< 0.006	<b>0.07 ± 0.02</b>	< 0.11	<b>0.90 ± 0.13</b>	<b>0.60 ± 0.07</b>
D 2	<i>Molinia caerulea</i>	< 0.011	<b>0.35 ± 0.06</b>	< 0.2	<b>1.90 ± 0.27</b>	<b>0.60 ± 0.08</b>
	<i>Calluna vulgaris</i> leaves	< 0.005	<b>0.29 ± 0.06</b>	< 0.09	<b>0.40 ± 0.12</b>	<b>2.70 ± 0.38</b>
	<i>Myrica gale</i> leaves	< 0.006	< 0.08	< 0.6	<b>2.20 ± 0.35</b>	<b>0.30 ± 0.04</b>
	<i>Myrica gale</i> stems	< 0.010	<b>0.18 ± 0.03</b>	< 0.1	<b>0.60 ± 0.07</b>	<b>0.20 ± 0.03</b>
D 3	<i>Molinia caerulea</i>	< 0.014	<b>0.29 ± 0.09</b>	< 0.5	<b>4.78 ± 0.72</b>	<b>1.88 ± 0.28</b>
	<i>Myrica gale</i> leaves	< 0.035	< 0.16	< 0.11	<b>0.57 ± 0.09</b>	<b>1.70 ± 0.27</b>
	<i>Myrica gale</i> stems	< 0.016	<b>0.37 ± 0.07</b>	< 0.15	<b>2.18 ± 0.33</b>	<b>0.43 ± 0.07</b>
	<i>Erica tetralix</i>	< 0.015	<b>1.05 ± 0.17</b>	< 0.6	<b>5.83 ± 0.87</b>	<b>0.98 ± 0.16</b>
D 4	<i>Molinia caerulea</i>	< 0.016	<b>0.25 ± 0.07</b>	< 0.5	<b>7.29 ± 1.3</b>	<b>0.13 ± 0.02</b>
	<i>Calluna vulgaris</i> leaves	< 0.015	<b>0.69 ± 0.18</b>	< 0.4	<b>3.14 ± 0.72</b>	<b>6.26 ± 0.94</b>
	<i>Calluna vulgaris</i> stems	< 0.001	<b>1.15 ± 0.21</b>	< 0.5	<b>1.36 ± 0.29</b>	<b>1.49 ± 0.25</b>
	<i>Erica tetralix</i>	< 0.015	<b>2.35 ± 0.47</b>	< 0.8	<b>4.92 ± 1.1</b>	<b>0.69 ± 0.10</b>
D 5	<i>Molinia caerulea</i>	< 0.018	<b>0.82 ± 0.17</b>	< 0.8	<b>8.54 ± 1.6</b>	<b>0.56 ± 0.09</b>
	<i>Calluna vulgaris</i> leaves	< 0.013	<b>1.56 ± 0.33</b>	< 0.12	<b>2.38 ± 0.48</b>	<b>3.05 ± 0.49</b>
	<i>Calluna vulgaris</i> stems	< 0.013	<b>0.47 ± 0.11</b>	< 0.26	<b>0.46 ± 0.09</b>	<b>1.17 ± 0.19</b>
	<i>Eriophorum vaginatum</i>	< 0.016	<b>0.51 ± 0.12</b>	< 0.7	<b>8.25 ± 1.9</b>	<b>2.42 ± 0.36</b>
D6	<i>Molinia caerulea</i>	< 0.006	<b>0.34 ± 0.04</b>	< 0.5	<b>0.88 ± 0.17</b>	<b>0.12 ± 0.02</b>
	<i>Myrica gale</i> leaves	< 0.017	<b>0.17 ± 0.04</b>	< 1.6	<b>2.85 ± 0.51</b>	<b>0.17 ± 0.03</b>
	<i>Myrica gale</i> stems	< 0.006	<b>0.20 ± 0.05</b>	< 1.2	<b>1.64 ± 0.43</b>	<b>0.18 ± 0.03</b>
	<i>Erica tetralix</i>	< 0.008	<b>1.34 ± 0.24</b>	< 0.20	<b>0.42 ± 0.13</b>	<b>0.28 ± 0.04</b>
	<i>Eriophorum vaginatum</i>	< 0.007	<b>0.32 ± 0.11</b>	< 1.6	<b>1.79 ± 0.48</b>	<b>0.36 ± 0.05</b>
D 7	<i>Molinia caerulea</i>	< 0.016	<b>0.25 ± 0.08</b>	< 0.22	<b>4.00 ± 0.80</b>	<b>0.31 ± 0.04</b>
	<i>Calluna vulgaris</i> leaves	< 0.007	<b>0.80 ± 0.20</b>	< 2.0	<b>2.04 ± 0.45</b>	<b>2.54 ± 0.38</b>
	<i>Calluna vulgaris</i> stems	< 0.011	<b>0.56 ± 0.14</b>	< 3.4	<b>0.87 ± 0.18</b>	<b>1.75 ± 0.26</b>
	<i>Myrica gale</i> leaves	< 0.011	< 0.48	< 1.6	<b>2.91 ± 0.87</b>	<b>0.17 ± 0.03</b>
	<i>Myrica gale</i> stems	< 0.011	<b>0.14 ± 0.04</b>	< 1.8	<b>2.43 ± 0.49</b>	<b>0.25 ± 0.04</b>
	<i>Erica tetralix</i>	< 0.019	<b>1.15 ± 0.28</b>	< 2.8	<b>1.22 ± 0.30</b>	<b>0.37 ± 0.06</b>
	<i>Eriophorum vaginatum</i>	< 0.011	<b>0.53 ± 0.11</b>	< 0.20	<b>5.48 ± 1.1</b>	<b>1.22 ± 0.21</b>
D 8	<i>Calluna vulgaris</i> leaves	< 0.047	<b>0.44 ± 0.10</b>	< 0.36	<b>2.72 ± 0.57</b>	<b>2.65 ± 0.39</b>
	<i>Calluna vulgaris</i> stems	< 0.071	<b>0.39 ± 0.08</b>	< 0.45	<b>2.77 ± 0.58</b>	<b>2.32 ± 0.37</b>
	<i>Myrica gale</i> leaves	< 0.020	< 0.2	< 2.7	<b>11.22 ± 3.6</b>	<b>0.23 ± 0.04</b>
	<i>Myrica gale</i> stems	< 0.017	<b>0.20 ± 0.04</b>	< 3.0	<b>4.34 ± 0.87</b>	<b>0.19 ± 0.03</b>
	<i>Erica tetralix</i>	< 0.027	<b>1.4 ± 0.28</b>	< 2.7	<b>11.4 ± 2.1</b>	<b>0.43 ± 0.06</b>

Table 27: Transfer factor (TF) values for <sup>238</sup>U, <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>40</sup>K and <sup>137</sup>Cs at sites D1 – D8

The TF values for  $^{226}\text{Ra}$  for the vegetation studied were compared with previous findings from literature however the availability of TF values for natural radionuclides within these species of vegetation is very limited. Therefore the results from this study are compared to TF values from similar vegetation in other previous published studies.

Mahon and Mathewes (1983) reported TF values in the range of 0.01 – 0.24 for  $^{226}\text{Ra}$  uptake in four sample groups, which were forbs and grasses, shrubs, deciduous trees and coniferous and these results are in close agreement to the range found within this study. The results obtained for *M. caerulea* (average TF value of 0.34) are in agreement with the results reported by Linsalata *et al* (1989), Vera Tomé *et al* (2003) and Ibrahim and Whicker (1988) of 0.016 – 0.40, 0.170 and 0.0252 - 0.1028 respectively for  $^{226}\text{Ra}$  uptake in pasture vegetation. The results from this study are however in general higher and in some cases an order of magnitude higher than the results reported by Ewers *et al* (2003), Moffett and Tellier (1977), Makrose *et al* (1993), Amaral *et al* (1988) and IUR (1994) for  $^{226}\text{Ra}$  in pasture vegetation of 0.03, 0.029, 0.0066, 0.07 and 0.08 respectively. The results from this study indicate that  $^{226}\text{Ra}$  uptake in *C. vulgaris* and *M. gale* is not dependent on plant compartment (leaves and stems) however in the latter case as stated previously the data set is very limited. These results are in disagreement with the findings of Makrose *et al* (1993) where uptake of  $^{226}\text{Ra}$  in tomato plants decreased in the order: leaves > root > stems > fruit. The range of  $^{226}\text{Ra}$  TF values (0.07 – 2.4) for the vegetation within this study were higher than those reported by Radhakrishna *et al* (1996) for  $^{226}\text{Ra}$  in vegetables (not specified) and rice of 0.004 and 0.007 respectively. They were also higher than the TF value of 0.008 reported by McDowell-Boyer *et al* (1980) for  $^{226}\text{Ra}$  in vegetables, and by Watson *et al* (1984) who reported TF values of 0.003 and 0.012 for fruits and vegetables respectively.

The uptake of  $^{226}\text{Ra}$  in the vegetation sampled within this ecosystem was higher than that for the isotopes of uranium and thorium and these results are in agreement with the findings of Linsalata *et al* (1989) who stated that the TF values for elements in the oxidation 2+ (Ra) state are always greater than for those elements in 4+ (Th) oxidation state.

Potassium-40 transfer factor values in the vascular plants studied ranged from 0.4 for *C. vulgaris* (leaves) to 11.2 for *M. gale* (leaves), however on average TF values ranged from 1.4 for *C. vulgaris* (stems) to 5.2 for *E. vaginatum*. Transfer factor values were compared based on plant species and results indicate that  $^{40}\text{K}$  transfer from soil to vegetation in this ecosystem is similar for all species of vegetation (One Way ANOVA,  $p > 0.05$ ). In the case of *C. vulgaris* and *M. gale* results indicate that  $^{40}\text{K}$  uptake is not dependent on plant compartment as TF values were not significantly different for the leaves and stems (One Way ANOVA,  $p > 0.05$ ).

Limited information is available in the literature with regard to  $^{40}\text{K}$  uptake by these species of vegetation however where possible comparison between the findings of this study and published findings from similar studies were made.

Potassium-40 TF values for *M. caerulea* ranged from 0.88 – 7.29 and were higher than TF values reported by Bunzl and Kracke (1989) for the same vegetation growing within a similar ecosystem. Transfer factors values for *M. caerulea* from this study were also higher than those reported by Papastefanou *et al* (1999), Rafferty *et al* (1994a), Strebl *et al* (2002), and Baeza *et al* (2001) of 0.16 – 2.42, 2 – 6, 0.31 – 2.01 and 0.3 respectively for pasture vegetation growing in grassland ecosystems. These results presented within this study provide an interesting comparison to grassland ecosystems given that upland blanket bog soils are generally considered nutrient deficient and while the levels of  $^{40}\text{K}$  were low in the soil ( $< 112 \text{ Bq/kg}$ ), the TF values presented are higher than for the grassland ecosystems reported by the aforementioned authors.

Potassium-40 TF values for *E. vaginatum* ranged from 1.8 – 8.3 and were lower but not significantly lower (t-test,  $p > 0.05$ ), than those found for  $^{40}\text{K}$  in the same vegetation in the Atlantic blanket bog ecosystem. The results found for *E. vaginatum* in both ecosystems are in agreement with TF values reported by Coughtrey *et al* (1989) of  $5.51 \pm 1.08$  for *E. vaginatum* and *E. augustifolium* (combined together) growing on wetter, less mineralized regions of an upland grassland in England.

Potassium-40 TF values for *C. vulgaris* ranged from 0.4 – 2.8 and were lower but not significantly lower than TF values for  $^{40}\text{K}$  in the same vegetation species in the Atlantic blanket bog (section 3.1) as there is no statistical difference between the TF values in the leaves from each ecosystem and the stems in each ecosystem (One Way ANOVA,  $p > 0.05$ ). The trend found within this study for  $^{40}\text{K}$  TF values in *C. vulgaris* where the TF values for the leaves and stems were not significantly different is also in agreement with the findings for  $^{40}\text{K}$  TF values in this vegetation in the Atlantic blanket bog ecosystem.

Transfer factor values for  $^{137}\text{Cs}$  in vascular plants ranged from 0.12 for *M. caerulea* to 6.26 for *C. vulgaris* (leaves) as shown in Table 24, however on average TF values ranged from 0.29 for *E. tetralix* to 1.6 for *C. vulgaris* (leaves). Caesium-137 TF values were significantly higher in the leaves of *C. vulgaris* than *M. gale* (leaves and stems), *E. tetralix*, *M. caerulea* and *E. vaginatum* (One Way ANOVA,  $p < 0.017$ ). Transfer factor values were also significantly higher in the leaves of *C. vulgaris* in comparison to the stems indicating that  $^{137}\text{Cs}$  uptake in this plant species is dependent on plant compartment (One Way ANOVA,  $p = 0.037$ ). Transfer factor values for the leaves and stems of *M. gale* indicate that uptake of  $^{137}\text{Cs}$  is not dependent on plant compartment as the TF values are statistically similar (One Way ANOVA,  $p = 0.998$ ) Transfer factor values for all other plant species are statistically similar (One Way ANOVA,  $p > 0.05$ ).

Caesium-137 TF values for *M. caerulea* ranged from 0.12 – 1.9 (average  $^{137}\text{Cs}$  TF value of 0.74) and these results are in agreement with the findings of Papastefanou *et al* (1999), Strebl *et al* (2002), Ehlken and Kirchner (1996) and Baeza *et al* (2001) for  $^{137}\text{Cs}$  in pasture vegetation of 0.002 – 7.42, 0.03 – 1.06, 0.01 – 7 and 0.25 respectively. The results found in this study are also in close agreement with  $^{137}\text{Cs}$  TF value reported by IUR (1994) of 0.53 for grass growing on an organic soil.

The TF values for  $^{137}\text{Cs}$  in *C. vulgaris* were similar to the values found for this radionuclide in the same plant species growing in the Atlantic blanket bog (section 3.1), as results indicate that there is no statistical difference between the TF values in the leaves from each ecosystem and the stems in each ecosystem (One Way ANOVA,  $p >$

0.05). The trend found within this study for  $^{137}\text{Cs}$  TF values in *C. vulgaris* leaves and stems (leaves > stems) are also in agreement with the findings for  $^{137}\text{Cs}$  TF values in this vegetation in the Atlantic blanket bog ecosystem. As stated previously (section 3.1.5), these findings are in agreement with the findings of Bunzl and Kracke (1986), Barci-Funel *et al* (1995), McGee *et al* (2000), Fogh and Anderson (2001), von Fricks *et al* (2002) and Mosquera *et al* (2006).

The trend found for  $^{137}\text{Cs}$  uptake in the leaves and stems of *M. gale* (leaves = stems) does not follow the trend found for *C. vulgaris* in either the Atlantic blanket bog or upland blanket bog ecosystem or the trend found for  $^{137}\text{Cs}$  uptake in tropical, spruce and pine trees (Barci-Funel *et al*, 1995, McGee *et al*, 2000, Fogh and Anderson, 2001, Mosquera *et al*, 2006).

Caesium-137 TF values in *E. vaginatum* ranged from 0.36 – 2.42 and while lower than those found for  $^{137}\text{Cs}$  in the same vegetation in the Atlantic blanket bog ecosystem (2.7 – 5.1), however the difference between the two sets of data was not significantly different (t-test,  $p > 0.05$ ). The results found within both these ecosystems are in agreement with  $^{137}\text{Cs}$  TF values reported by Coughtrey *et al* (1989) of 3.2 – 6.5 for *E. vaginatum* and *E. angustifolium* (combined together) growing on wetter, less mineralized regions of an upland grassland in England.

Caesium-137 TF values for *E. tetralix* ranged from 0.28 – 0.98 and were significantly lower than those found for *C. vulgaris* and as both are heather species found in the same ecosystem these results would indicate that  $^{137}\text{Cs}$  uptake in heather is dependent on species type.

Radium-226,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  TF values were compared for all vegetation in order to ascertain which isotopes were being accumulated by the different species of vegetation in this ecosystem. Potassium-40 TF values were significantly higher in the grass species, *M. caerulea*, in comparison to both  $^{226}\text{Ra}$  and  $^{137}\text{Cs}$  (One Way ANOVA,  $p < 0.002$ ) and TF values for  $^{137}\text{Cs}$  and  $^{226}\text{Ra}$  were statistically similar (One Way ANOVA,  $p = 0.89$ ).

Potassium-40 TF values for *M. gale* were found to be significantly higher than both  $^{226}\text{Ra}$  and  $^{137}\text{Cs}$  when looking at the plant species as a whole (One Way ANOVA,  $p < 0.001$ ), and  $^{226}\text{Ra}$  and  $^{137}\text{Cs}$  TF values not significantly different (One Way ANOVA,  $p = 0.61$ ). However when the individual plant compartments (leaves and stems) were studied the TF values for all three radioisotopes appeared to be statistically similar (One Way ANOVA,  $p > 0.05$ ). In the case of *C. vulgaris* when comparing the leaves and stems together, TF values for  $^{137}\text{Cs}$  and  $^{40}\text{K}$  were found to be significantly higher than  $^{226}\text{Ra}$  (One Way ANOVA,  $p < 0.05$ ) and TF values for  $^{137}\text{Cs}$  and  $^{40}\text{K}$  were not significantly different (One Way ANOVA,  $p = 0.18$ ). When comparing the leaves and stems of *C. vulgaris* separately results indicate that  $^{137}\text{Cs}$  TF values for the leaves were significantly higher than  $^{226}\text{Ra}$  TF values for both leaves and stems and  $^{40}\text{K}$  TF values for the stems (One Way ANOVA,  $p < 0.05$ ). Potassium-40 TF values for *E. tetralix* were significantly higher than  $^{137}\text{Cs}$  (One Way ANOVA,  $p < 0.05$ ) but statistically similar to  $^{226}\text{Ra}$  TF values (One Way ANOVA,  $p > 0.05$ ). Transfer factor values for  $^{226}\text{Ra}$  and  $^{137}\text{Cs}$  in this species of vegetation were not significantly different (One Way ANOVA,  $p > 0.05$ ). In the case of *E. vaginatum*, TF values for all three isotopes,  $^{226}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  were statistically similar (One Way ANOVA,  $p > 0.05$ ).

Transfer factor values for the three isotopes ( $^{226}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$ ) were compared based on all plant species,  $^{226}\text{Ra}$  TF values were found to be the lowest with an average value of 0.56, followed by  $^{137}\text{Cs}$  TF (average value of 1.23) and the highest TF values were found to be for  $^{40}\text{K}$  with an average value of 3.1. Potassium-40 TF values were found to be significantly higher than both  $^{226}\text{Ra}$  and  $^{137}\text{Cs}$  (One Way ANOVA,  $p < 0.016$ ), and comparisons between  $^{226}\text{Ra}$  and  $^{137}\text{Cs}$  indicate that the TF values are not significantly different (One Way ANOVA,  $p = 0.53$ ).

Transfer factor values for  $^{226}\text{Ra}$  in vascular plants studied indicate that uptake is independent of  $^{226}\text{Ra}$  in the soil (Spearman correlation coefficient of -0.080,  $p = 0.67$ ) as illustrated in Figure 48. These results are in agreement with the findings of Vasconellas *et al* (1987) and Kohler *et al* (2002) but are in contrast to the findings of Blanco Rodríguez *et al* (2002) and Makrose *et al* (1993).

Potassium-40 TF values for all vascular vegetation appear to be dependent of the concentration of  $^{40}\text{K}$  in the soil as shown in Figure 49. A Spearman correlation coefficient of  $-0.41$ , ( $p = 0.015$ ) indicate an inverse relationship between the concentration of  $^{40}\text{K}$  in the soil and transfer of this radionuclide transfer into vegetation. A similar but much stronger correlation is present between the uptake of  $^{40}\text{K}$  in *M. gale* and the concentration of  $^{40}\text{K}$  in soil as shown in Figure 53 (Spearman correlation coefficient of  $-0.81$ ,  $p = 0.00$ ). The data for *M. gale* was removed from the original correlation for all vegetation and the relationship between  $^{40}\text{K}$  in the soil and in all other vegetation was studied and results then indicate that there is no significant relationship between  $^{40}\text{K}$  in all vegetation (except *M. gale*) and the concentration of  $^{40}\text{K}$  in the soil (Spearman correlation coefficient of  $-0.20$ ,  $p = 0.33$ ). The relationship between  $^{40}\text{K}$  transfer into vegetation and  $^{137}\text{Cs}$  levels in the soil was studied and results indicate that there is no significant relationship between these two parameters as shown in Figure 51 (Spearman correlation coefficient of  $-0.237$ ,  $p = 0.16$ ). These results indicate that levels of  $^{137}\text{Cs}$  in the soil have no impact on the levels of  $^{40}\text{K}$  in the vegetation studied.

Figure 48: Plot of  $^{226}\text{Ra}$  TF versus  $^{226}\text{Ra}$  in soil (upland blanket bog vegetation)

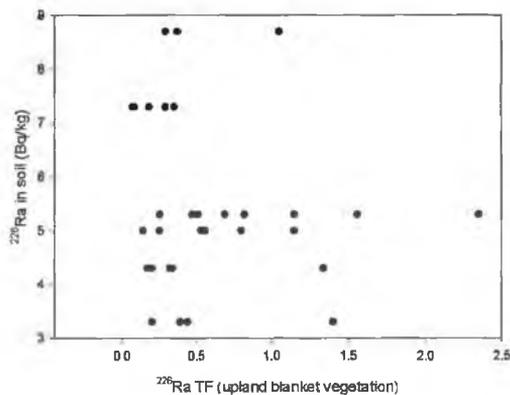


Figure 50: Plot of  $^{137}\text{Cs}$  TF values versus  $^{137}\text{Cs}$  in soil (upland blanket bog vegetation)

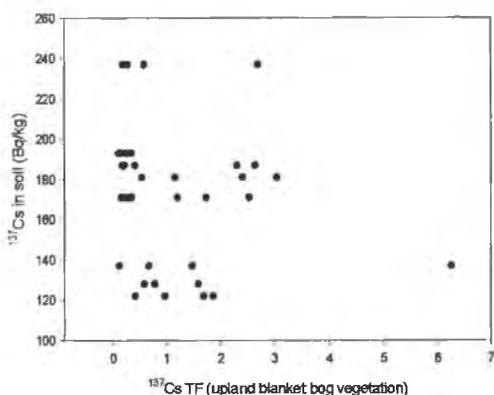


Figure 52: Plot of  $^{137}\text{Cs}$  TF versus  $^{40}\text{K}$  in soil (upland blanket bog vegetation)

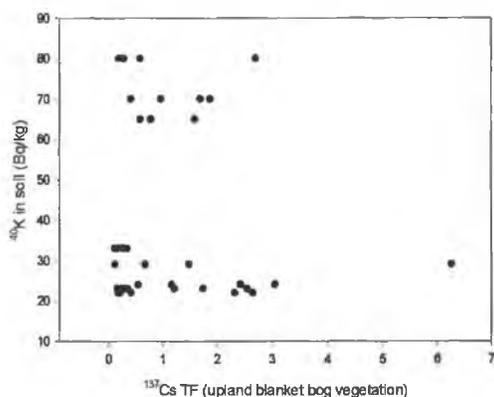


Figure 51: Plot of  $^{40}\text{K}$  TF versus  $^{137}\text{Cs}$  in soil (upland blanket bog vegetation)

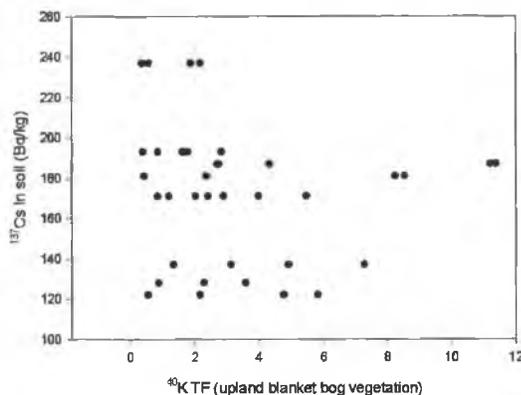
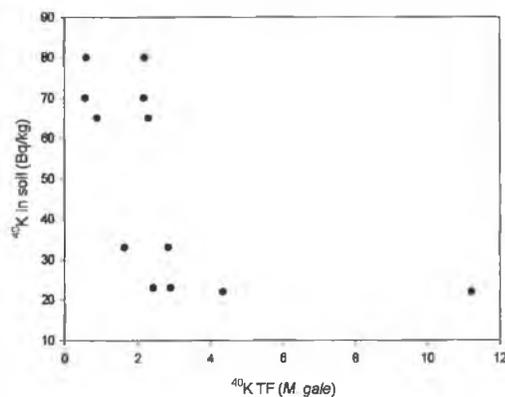


Figure 53: Plot of  $^{40}\text{K}$  TF values versus  $^{40}\text{K}$  in soil (*M. gale*)



Transfer factor values for  $^{137}\text{Cs}$  for the vegetation studied appear to be independent of the concentration of  $^{137}\text{Cs}$  in the soil (Spearman correlation coefficient of  $-0.305$ ,  $p = 0.07$ ) as illustrated in Figure 50. A similar observation was made for  $^{137}\text{Cs}$  TF values for vegetation studied and  $^{40}\text{K}$  in soil where a Spearman correlation coefficient of  $-0.051$  ( $p = 0.812$ ) indicates that  $^{137}\text{Cs}$  uptake by vegetation was independent on  $^{40}\text{K}$  activity in soil. The relationship (or lack of) between these parameters is illustrated in Figure 52. These results are in agreement with the findings of Albers *et al* (1998), Bunzl *et al* (2000) and Strebl *et al* (2002) but are contradictory to the finding of Robinson and Stone (1992), Gerzabek *et al* (1998), Tsukada and Nakamura (1999), Sanchez *et al* (1999) and Frissel *et al* (2002). However when studying plant species on their own, the uptake of  $^{137}\text{Cs}$  in both *E. tetralix* and *M. gale* was found to be inversely proportional to the concentration of  $^{137}\text{Cs}$  in the soil as illustrated in Figures 54 and 55 respectively.

Figure 54: Plot of  $^{137}\text{Cs}$  TF values versus  $^{137}\text{Cs}$  in soil (*E. tetralix*)

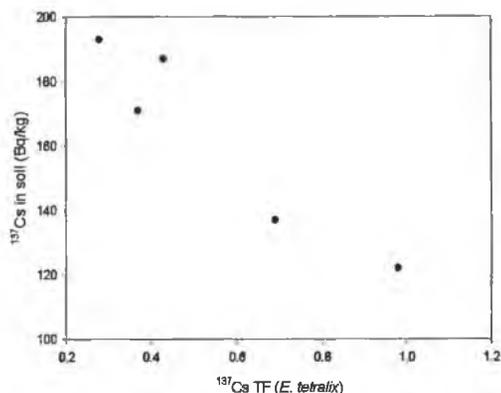
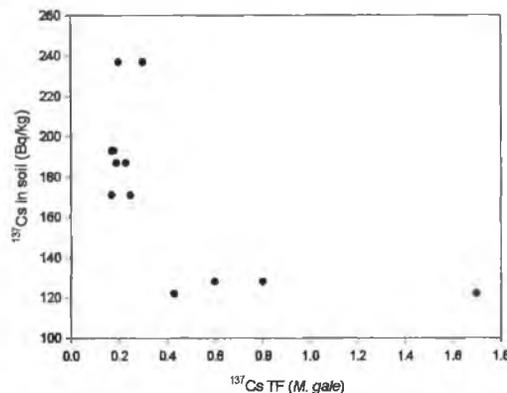


Figure 55: Plot of  $^{137}\text{Cs}$  TF values versus  $^{137}\text{Cs}$  in soil (*M. gale*)



### 3.2.6 Correlations between vegetation transfer factors and chemical properties of soil and vegetation

Correlations were carried out between transfer factor values for  $^{226}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  for all vegetation sampled in upland blanket bog (Table 27) and both the soil physiochemical parameters (Table 23) and the nutrient status of the vegetation (Table 26). The purpose of these correlations was to determine if any of these parameters have a significant impact on the uptake of the aforementioned radionuclides. The TF values are based on the average radionuclide concentration in the upper 20 cm of soil therefore it was necessary to correlate these results with the average concentration of each soil property as outlined in Table 28. The full set of results for each individual soil layer (i.e. 0-5 cm, 5-10 cm and 10-20 cm) at all eight sites is presented in Table 23 (Section 3.2.2).

Site	Moisture %	pH	Organic Matter %	Available Magnesium mg/kg	Available Sodium mg/kg	Available Calcium mg/kg	Available Potassium mg/kg	CEC meq/100g
D 1	86.0	4.2	84.5	207	168	166	149	109
D 2	87.2	4.1	86.7	233	169	182	175	116
D 3	93.8	4.4	96.0	362	147	299	85	158
D 4	88.9	4.2	97.1	443	167	352	126	135
D 5	91.9	4.6	96.6	477	222	373	137	146
D 6	89.7	4.4	89.8	259	202	161	153	111
D 7	92.3	4.6	96.9	415	179	375	138	136
D 8	93.6	4.7	97.3	454	168	719	96	159

Table 28: Physicochemical properties of soil surface horizons at sites D1 – D8 (Average value for 0 - 20 cm)

Correlations were carried out initially for each radionuclide with the soil properties and nutrient status of vegetation for all vegetation species sampled to assess if there is an overall effect of any particular parameter affecting uptake within this ecosystem. Similar correlations were then carried on the individual plant species for each radionuclide. The relationship between the radionuclide TF values and soil physicochemical properties and nutrient status of vegetation are outlined in Tables 29 and 30 respectively and any significant correlations are discussed in greater detail throughout this section.

Comparisons between  $^{226}\text{Ra}$  TF values and soil physicochemical properties indicate that  $^{226}\text{Ra}$  uptake in vegetation is significantly affected by organic matter, available magnesium, available potassium and CEC of the soil. A spearman correlation coefficient of 0.49 ( $p = 0.01$ ) indicates a relatively strong positive correlation between the uptake of  $^{226}\text{Ra}$  in all vegetation studied and the organic matter content of the soil as shown in Figure 56. A similar observation was noted between  $^{226}\text{Ra}$  TF values and available magnesium levels in the soil (Spearman rank coefficient of 0.53,  $p = 0.002$ ) as shown in Figure 57 however a Spearman correlation coefficient of -0.37 ( $p = 0.04$ ) indicates a negative correlation between  $^{226}\text{Ra}$  TF and available potassium levels in the soil (Figure 58). The uptake of  $^{226}\text{Ra}$  by all vegetation also appears to be affected by the CEC of the

soil where a Spearman correlation coefficient of 0.37 ( $p = 0.04$ ) indicates a moderately strong positive correlation between  $^{226}\text{Ra}$  TF values and the CEC of the soil (Figure 59). Radium-226 TF values (all vegetation species) appear to be unaffected by the nutrient status of the vegetation. These results indicate that as the organic matter, available magnesium, and CEC of the soil increase, the transfer of radium into vegetation is increased while the opposite in the case for the levels of available potassium in the soil.

Soil Properties	$^{226}\text{Ra}$ TF (All veg.)	$^{40}\text{K}$ TF (All veg.)	$^{137}\text{Cs}$ TF (All veg.)	$^{226}\text{Ra}$ TF ( <i>M. caeruleae</i> )	$^{40}\text{K}$ TF ( <i>M. caeruleae</i> )	$^{137}\text{Cs}$ TF ( <i>M. caeruleae</i> )	$^{226}\text{Ra}$ TF ( <i>E. tetralix</i> )	$^{40}\text{K}$ TF ( <i>E. tetralix</i> )	$^{137}\text{Cs}$ TF ( <i>E. tetralix</i> )
$^{238}\text{U}$ (Bq/kg)	X	X	X	X	X	X	X	X	X
$^{226}\text{Ra}$ (Bq/kg)	X	X	X	X	X	X	X	X	X
$^{228}\text{Ra}$ (Bq/kg)	X	X	X	X	X	X	X	X	X
$^{40}\text{K}$ (Bq/kg)	X	↓ $r^2 = -0.41$ $p = 0.015$	X	X	X	X	X	X	X
$^{137}\text{Cs}$ (Bq/kg)	X	X	X	X	X	X	X	X	↓ $r^2 = -0.95$ $p = 0.014$
Moisture Content (%)	X	X	X	X	X	X	X	X	X
pH	X	↑ $r^2 = 0.36$ $p = 0.03$	X	X	X	X	X	X	X
Organic Matter (%)	↑ $r^2 = 0.49$ $p = 0.01$	↑ $r^2 = 0.46$ $p = 0.006$	X	X	X	X	X	X	X
Avail. Mg (mg/kg)	↑ $r^2 = 0.53$ $p = 0.002$	↑ $r^2 = 0.47$ $p = 0.004$	X	X	↑ $r^2 = 0.84$ $p = 0.02$	X	X	X	X
Avail. Na (mg/kg)	X	X	X	X	X	X	X	X	↓ $r^2 = -0.89$ $p = 0.04$
Avail. Ca (mg/kg)	X	↑ $r^2 = 0.44$ $p = 0.007$	X	X	↑ $r^2 = 0.81$ $p = 0.03$	X	X	X	X
Avail. K (mg/kg)	↓ $r^2 = -0.37$ $p = 0.04$	↓ $r^2 = -0.49$ $p = 0.002$	X	X	X	X	X	X	X
CEC	↑ $r^2 = 0.37$ $p = 0.04$	↑ $r^2 = 0.43$ $p = 0.009$	X	X	X	X	X	X	X
Vegetation Nutrients									
Calcium (mg/kg)	X	X	X	X	X	X	X	X	X
Magnesium (mg/kg)	X	X	↑ $r^2 = 0.44$ $p = 0.009$	X	X	X	X	X	↑ $r^2 = 0.95$ $p = 0.04$
Sodium (mg/kg)	X	X	X	X	X	X	X	X	X
X: no correlation      †: positive correlation      ‡: negative correlation									

Table 29: Correlation between radionuclide transfer factors and upland blanket bog vegetation (All vegetation, *M. caeruleae* and *E. tetralix*).

Soil Properties	<sup>226</sup> Ra TF <i>E. vaginatum</i>	<sup>40</sup> K TF <i>E. vaginatum</i>	<sup>137</sup> Cs TF <i>E. vaginatum</i>	<sup>226</sup> Ra TF <i>C. vulgaris</i>	<sup>40</sup> K TF <i>(C. vulgaris)</i>	<sup>137</sup> Cs TF <i>C. vulgaris</i>	<sup>226</sup> Ra TF <i>(M. gale)</i>	<sup>40</sup> K TF <i>(M. gale)</i>	<sup>137</sup> Cs TF <i>(M. gale)</i>
<sup>238</sup> U (Bq/kg)	X	X	X	X	X	X	X	X	X
<sup>226</sup> Ra (Bq/kg)	X	X	X	X	X	X	X	X	X
<sup>228</sup> Ra (Bq/kg)	X	X	X	X	X	X	X	X	X
<sup>40</sup> K (Bq/kg)	X	X	X	X	X	X	X	X	X
<sup>137</sup> Cs (Bq/kg)	X	X	X	X	X	X	X	X	$r^2 = -0.62$ $p = 0.029$
Moisture Content (%)	↑ $r^2 = 0.99$ $p = 0.04$	X	X	X	X	X	X	X	X
pH	X	X	X	X	X	X	X	↑ $r^2 = 0.71$ $p = 0.008$	X
Organic Matter (%)	↑ $r^2 = 0.999$ $p = 0.03$	X	X	X	↑ $r^2 = 0.77$ $p = 0.012$	X	X	↑ $r^2 = 0.62$ $p = 0.029$	X
Avail. Mg (mg/kg)	X	X	X	X	X	X	X	↑ $r^2 = 0.62$ $p = 0.029$	X
Avail. Na (mg/kg)	X	X	X	X	X	X	X	X	↓ $r^2 = -0.76$ $p = 0.003$
Avail. Ca (mg/kg)	↑ $r^2 = 0.997$ $p = 0.049$	X	X	X	X	X	X	X	X
Avail. K (mg/kg)	X	X	X	X	↓ $r^2 = -0.71$ $p = 0.034$	X	X	X	X
CEC	X	X	X	X	X	X	X	X	X
Vegetation Nutrients									
Calcium (mg/kg)	X	X	↓ $r^2 = -0.999$ $p = 0.03$	X	X	↑ $r^2 = 0.68$ $p = 0.036$	X	↑ $r^2 = 0.90$ $p = 0.000$	↓ $r^2 = -0.64$ $p = 0.022$
Magnesium (mg/kg)	X	X	X	X	X	↑ $r^2 = 0.83$ $p = 0.002$	X	X	↑ $r^2 = 0.69$ $p = 0.011$
Sodium (mg/kg)	X	X	X	X	X	X	X	X	X

X: no correlation      ↑: positive correlation      ↓: negative correlation

Table 30: Correlation between radionuclide transfer factors and upland blanket bog vegetation (*E. vaginatum*, *C. vulgaris* and *M. gale*)

Potassium-40 TF values for all vegetation species were compared with the soil properties outlined in Table 28 and significant correlation were found between these TF values and the following soil properties: soil pH, organic matter, available magnesium, available calcium, available potassium and CEC of the soil.

Figure 56: Plot of  $^{226}\text{Ra}$  TF versus organic matter (upland blanket bog vegetation)

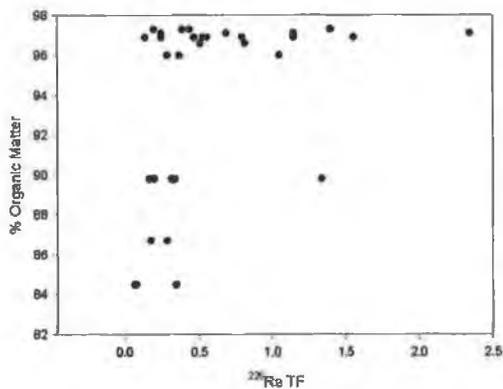


Figure 57: Plot of  $^{226}\text{Ra}$  TF versus Avail Mg in soil (upland blanket bog vegetation)

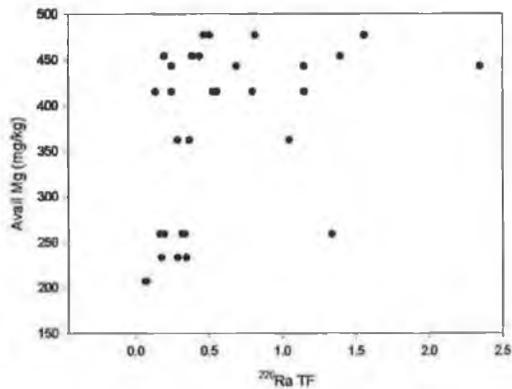


Figure 58: Plot of  $^{226}\text{Ra}$  TF versus Avail. K in soil (upland blanket bog vegetation)

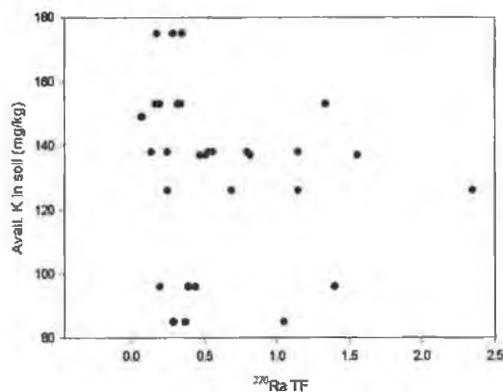


Figure 59: Plot of  $^{226}\text{Ra}$  TF versus CEC of soil (upland blanket bog vegetation)

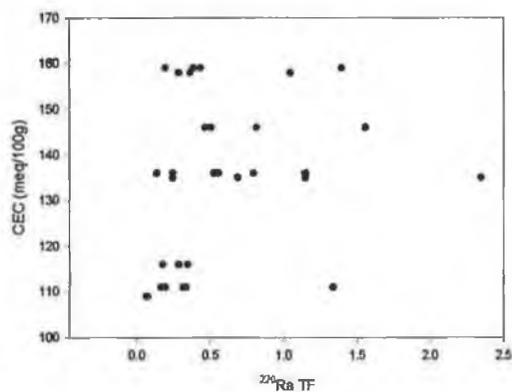


Figure 60: Plot of  $^{40}\text{K}$  TF versus soil pH (upland blanket bog vegetation)

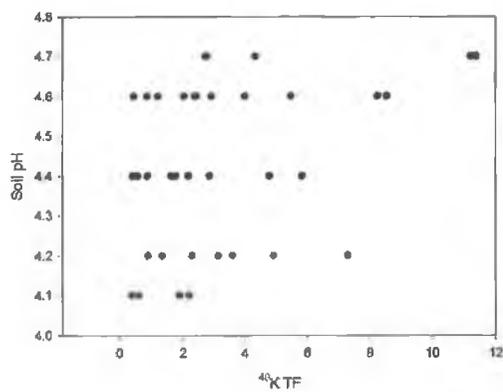
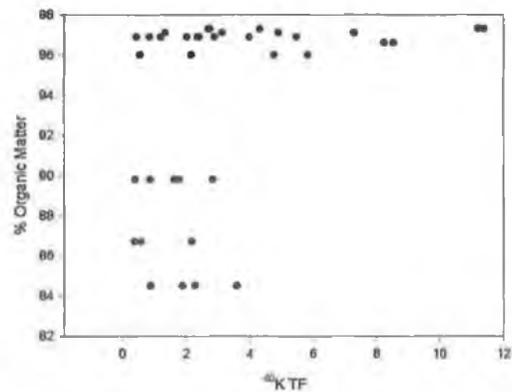


Figure 61: Plot of  $^{40}\text{K}$  TF versus % organic matter in soil (upland blanket bog vegetation)



A Spearman correlation coefficient of 0.36 ( $p = 0.03$ ) indicates a significant positive correlation between the uptake of  $^{40}\text{K}$  by all vegetation and the pH of the soil (Figure 60) as is also the case with  $^{40}\text{K}$  uptake and organic matter content of the soil where a Spearman correlation coefficient of 0.46 ( $p = 0.006$ ) indicates a significant positive correlation between these two parameters as shown in Figure 61. The uptake of  $^{40}\text{K}$  was found to be positively correlated with the available magnesium levels in the soil (Spearman correlation coefficient of 0.47,  $p = 0.004$ ) as shown in Figure 62. A similar observation was noted between  $^{40}\text{K}$  uptake and available calcium levels in the soil as shown in Figure 63 (Spearman correlation coefficient of 0.44,  $p = 0.007$ ).

Figure 62: Plot of  $^{40}\text{K}$  TF versus Avail. Mg in soil (upland blanket bog vegetation)

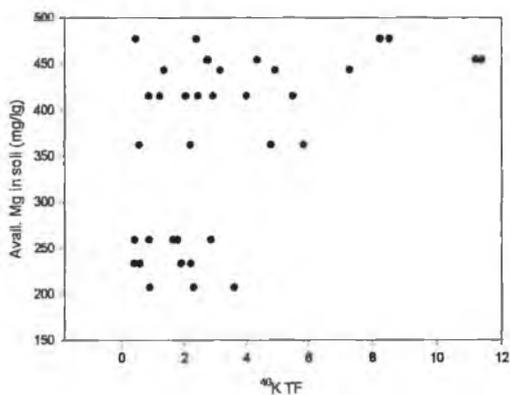


Figure 63: Plot of  $^{40}\text{K}$  TF versus Avail. Ca in soil (upland blanket bog vegetation)

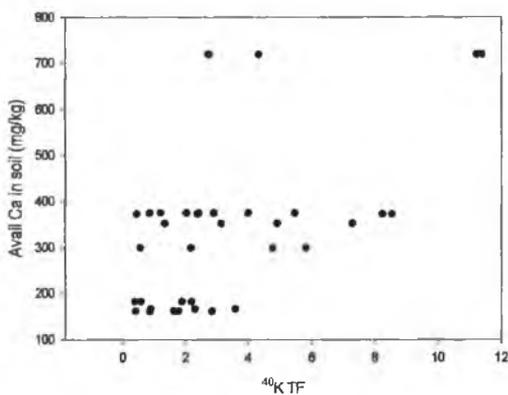


Figure 64: Plot of  $^{40}\text{K}$  TF versus Avail. K in soil (upland blanket bog vegetation)

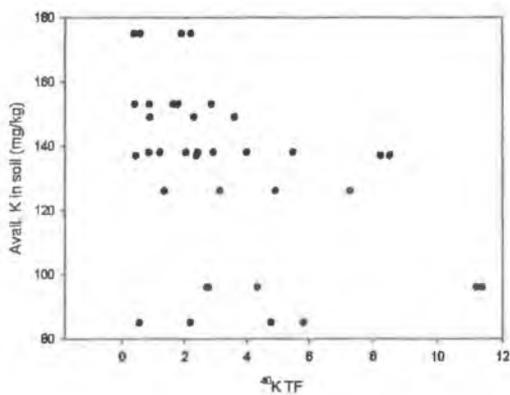
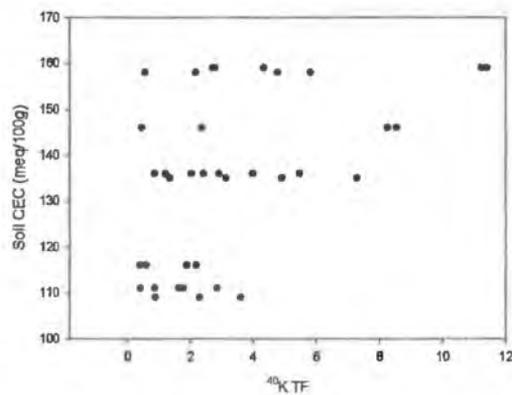


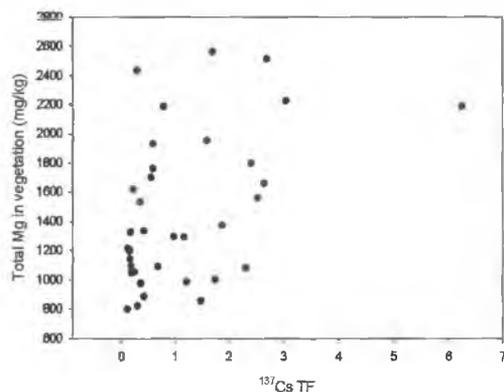
Figure 65: Plot of  $^{40}\text{K}$  TF versus soil CEC (upland blanket bog vegetation)



The uptake of  $^{40}\text{K}$  by all vegetation was also found to be diminished in soil with higher available potassium levels as shown in Figure 64 where a Spearman correlation coefficient of  $-0.49$  ( $p = 0.002$ ) indicates a negative correlation between these two parameters. A similar observation was made between  $^{40}\text{K}$  TF for all vegetation and the levels of  $^{40}\text{K}$  in the soil (Figure 49), however as stated previously this correlation was heavily dependent on the *M. gale* plant species. The uptake of  $^{40}\text{K}$  by all vegetation was found to be positively correlated with the CEC of the soil as shown in Figure 65 (Spearman correlation coefficient of  $0.43$ ,  $p = 0.009$ ). The latter correlation is in agreement with the findings for  $^{40}\text{K}$  uptake in the Atlantic blanket bog where the  $^{40}\text{K}$  TF values increased as the CEC of the soil increased. Potassium-40 TF values (for all vegetation species) appear to be unaffected by the nutrient status of the vegetation which is also in agreement with the findings for the vegetation in the Atlantic blanket bog.

Caesium-137 TF values showed no correlation with the soil physiochemical properties (Table 25) however a significant correlation was noted between the uptake of  $^{137}\text{Cs}$  by all vegetation and the magnesium content of the vegetation. A Spearman correlation coefficient of  $0.44$  ( $p = 0.009$ ) indicates a significant relationship between the uptake of  $^{137}\text{Cs}$  by all vegetation and magnesium content of vegetation as shown in Figure 66. No significant relationship was observed between both the calcium and sodium content of vegetation and  $^{137}\text{Cs}$  TF values. A similar correlation was also present between the magnesium levels in Atlantic blanket bog vegetation and TF values for  $^{137}\text{Cs}$ .

Figure 66: Plot of  $^{137}\text{Cs}$  TF versus Total Mg content of vegetation (upland blanket bog vegetation)



Correlation for each radionuclide was then carried out for each of the individual plant species to ascertain if any of the soil properties or vegetation nutrient status affect uptake of the radionuclide based on the individual plant species.

Radium-226 uptake by *M. caerulea* appears to be independent of both the soil properties studied and nutrient status of vegetation as is the case with  $^{137}\text{Cs}$  uptake by this species of grass. Potassium-40 uptake by *M. caerulea* was found to be positively correlated with the levels of available magnesium in the soil (Pearson correlation coefficient of 0.84,  $p = 0.02$ ) as shown in Figure 67. A similar observation was noted between uptake of  $^{40}\text{K}$  by this grass species and the available calcium levels in the soil where a Pearson correlation coefficient of 0.81 ( $p = 0.03$ ) indicates that uptake of  $^{40}\text{K}$  by grass was highest on soils with high levels of calcium (Figure 68). No significant correlation was noted between  $^{40}\text{K}$  TF values for *M. caerulea* and the nutrient status of this vegetation.

Figure 67: Plot of  $^{40}\text{K}$  TF versus Avail. Mg in soil (*M. caerulea*)

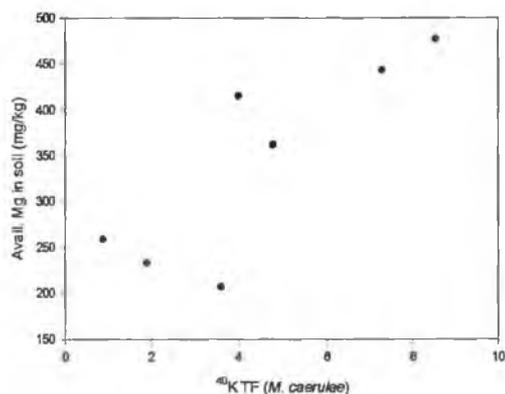
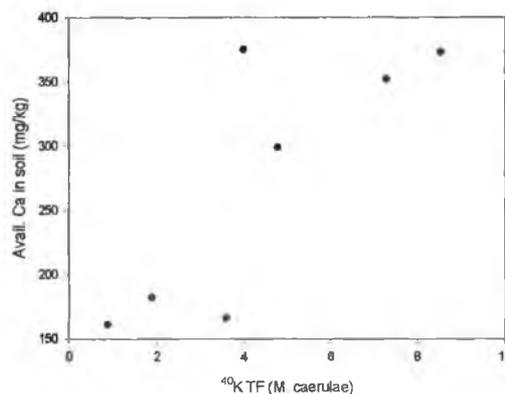


Figure 68: Plot of  $^{40}\text{K}$  TF versus Avail. Ca in soil (*M. caerulea*)



Radium-226 uptake by *E. tetralix* appears to be independent of both the soil properties studied and nutrient status of vegetation as is the case with  $^{40}\text{K}$  uptake with this species of heather. Caesium-137 uptake by *E. tetralix* was found to be significantly affected by the levels of available sodium in the soil where a Pearson correlation coefficient of -0.89 ( $p =$

0.04) indicates a strong negative relationship between these two parameters as shown in Figure 69. Caesium and sodium are both Group 1 elements in the periodic table and high levels of sodium in the soil appear to be diminishing the uptake of  $^{137}\text{Cs}$  by this species of vegetation. Numerous studies have highlighted the relationship between caesium levels in vegetation and potassium (another Group 1 element) in the soil, however, limited information is available in the literature with regard to the relationship between caesium and sodium (Robinson and Stone, 1992, Gerzabek *et al*, 1998, Tsukada and Nakamura, 1999, Sanchez *et al*, 1999 and Frissel *et al*, 2002). Caesium-137 uptake by *E. tetralix* was also found to be positively correlated with the total levels of magnesium in this vegetation as shown in Figure 70, indicating that uptake of this radionuclide was highest in the vegetation samples which contained highest levels of magnesium (Pearson correlation coefficient of 0.95,  $p = 0.04$ ). A similar observation was noted between  $^{137}\text{Cs}$  TF values for all vegetation and total magnesium levels in all vegetation (Figure 66).

Figure 69: Plot of  $^{137}\text{Cs}$  TF versus Avail. Na in soil (*E. tetralix*)

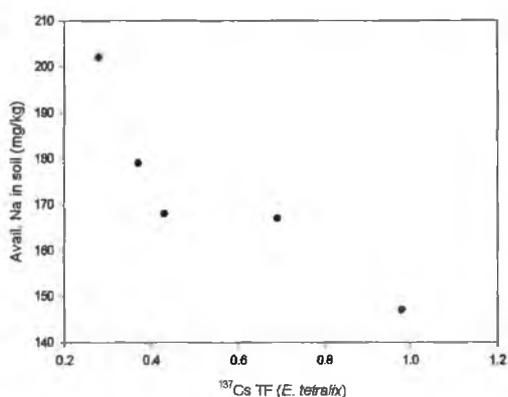
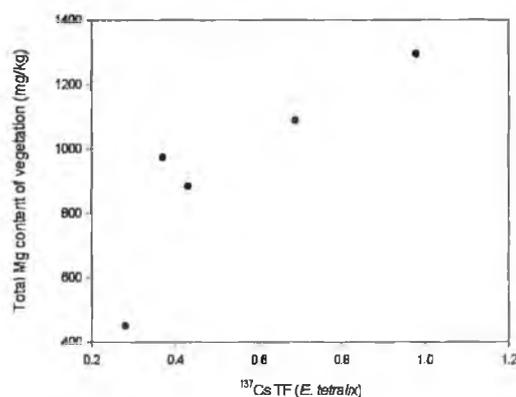


Figure 70: Plot of  $^{137}\text{Cs}$  TF versus Total Mg in vegetation (*E. tetralix*)



Radium-226 uptake by *E. vaginatum* was found to be positively correlated with the moisture levels in the soil as shown in Figure 71 (Pearson correlation coefficient of 0.99,  $p = 0.04$ ). A similarly strong correlation was also noted between  $^{226}\text{Ra}$  TF values and the organic matter content of the soil (Pearson correlation coefficient of 0.999,  $p = 0.03$ ) as shown in Figure 72. Radium-226 uptake was also found to be affected by the levels of available calcium in the soil where a Pearson correlation coefficient of 0.997 ( $p = 0.049$ )

indicates an uptake of  $^{226}\text{Ra}$  by *E. vaginatum* on soil with higher levels of available calcium (Figure 73) which is in contrast to the findings of Arkhipov *et al* (1984), Makrose *et al* (1993) and Blanco Rodríguez *et al* (2002).

Figure 71: Plot of  $^{226}\text{Ra}$  TF versus % soil moisture (*E. vaginatum*)

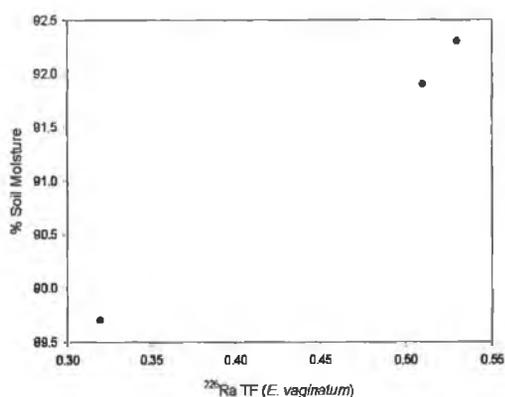


Figure 72: Plot of  $^{226}\text{Ra}$  TF versus soil organic matter (*E. vaginatum*)

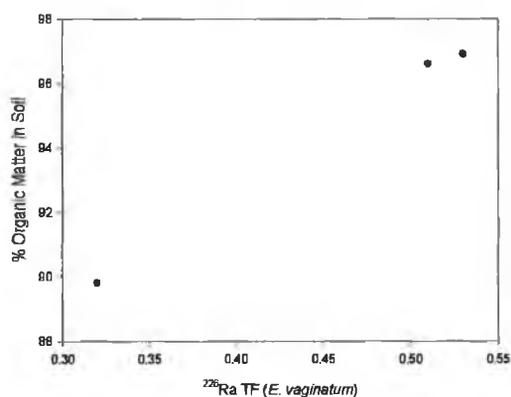


Figure 73: Plot of  $^{226}\text{Ra}$  TF versus Avail. Ca in soil (*E. vaginatum*)

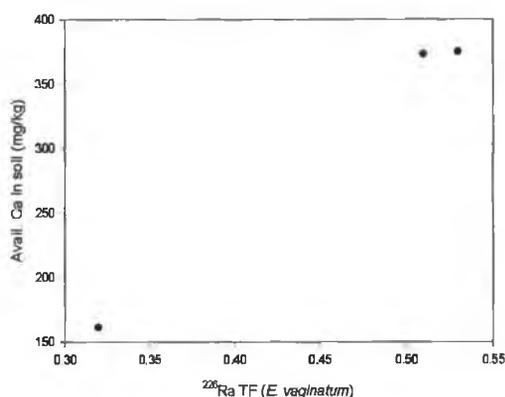
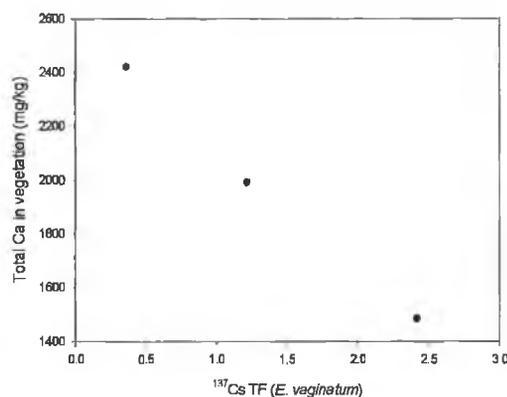


Figure 74: Plot of  $^{137}\text{Cs}$  TF versus Total Ca in vegetation (*E. vaginatum*)



Potassium-40 uptake by *E. vaginatum* appears to be independent of both the soil properties studied and nutrient status of vegetation. Caesium-137 uptake by *E. vaginatum* was found to be independent of the soil physiochemical properties studied (Table 28) however a significant correlation appears to be present between  $^{137}\text{Cs}$  uptake by *E. vaginatum* and the total calcium present in the vegetation as shown in Figure 74 (Pearson correlation coefficient of -0.999 ( $p = 0.30$ )). These results indicate that uptake of  $^{137}\text{Cs}$  by

*E. vaginatum* is adversely affected by the calcium levels present in the vegetation. These results are contrary to the findings for the Atlantic blanket bog vegetation and also for the findings for *C. vulgaris* within this ecosystem. These results appear to indicate that the effect of calcium levels in the plant on  $^{137}\text{Cs}$  TF values is highly dependent on plant species.

Radium-226 uptake by *C. vulgaris* appears to be independent of both the soil properties studied and nutrient status of vegetation. Potassium-40 uptake by *C. vulgaris* was found to be positively correlated with the organic matter content of the soil where a Spearman correlation coefficient of 0.77, ( $p = 0.012$ ), indicates that  $^{40}\text{K}$  uptake by *C. vulgaris* was highest on soil with higher soil organic matter (Figure 75). An inverse relationship was noted between  $^{40}\text{K}$  TF values and the available levels of K in the soil as shown in Figure 76 indicating that the highest uptake of  $^{40}\text{K}$  by *C. vulgaris* was from soils with the lowest concentration of available potassium (Pearson correlation coefficient of -0.71,  $p = 0.034$ ). No significant relationship was noted between the nutrient status of the vegetation and uptake of  $^{40}\text{K}$  by *C. vulgaris*

Figure 75: Plot of  $^{40}\text{K}$  TF versus soil organic matter (*C. vulgaris*)

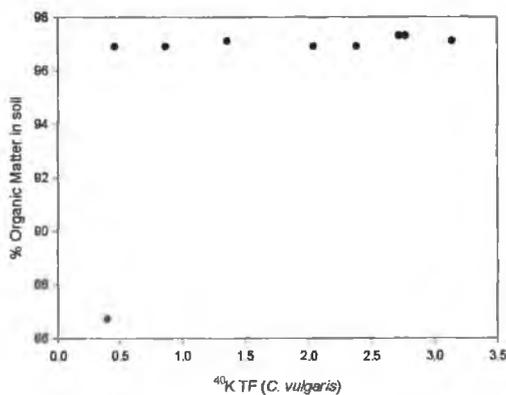
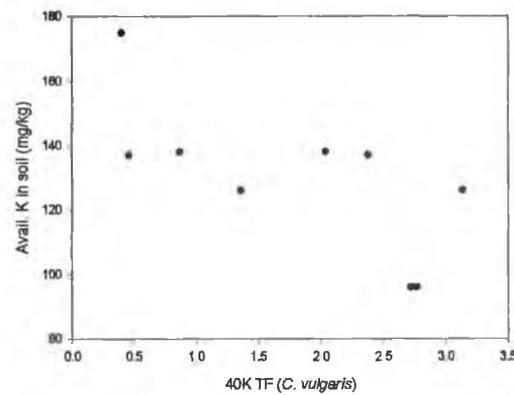


Figure 76: Plot of  $^{40}\text{K}$  TF versus Avail. K in soil (*C. vulgaris*)



Caesium-137 uptake by *C. vulgaris* appears to be independent of the soil physiochemical parameters studied (Table 28), however significant relationships were noted between  $^{137}\text{Cs}$  uptake and both the calcium and magnesium content of the vegetation. A Spearman correlation coefficient of 0.68 ( $p = 0.036$ ) indicates a significant positive relationship

between  $^{137}\text{Cs}$  uptake and calcium content of this vegetation (Figure 77) and a Spearman correlation coefficient of 0.83 ( $p = 0.002$ ) also indicates a significant positive correlation between  $^{137}\text{Cs}$  uptake and magnesium content of this vegetation (Figure 78). These results indicate that for *C. vulgaris* species sampled uptake of  $^{137}\text{Cs}$  is increased when the concentration of calcium and magnesium is at its highest. These results are similar to the findings for  $^{137}\text{Cs}$  uptake by *C. vulgaris* in the Atlantic blanket bog (section 3.1.6).

Figure 77: Plot of  $^{137}\text{Cs}$  TF versus total Ca content of vegetation (*C. vulgaris*)

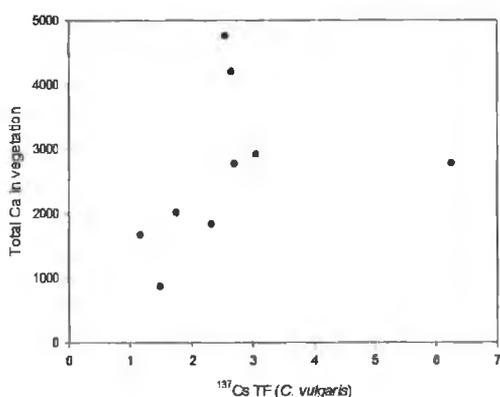
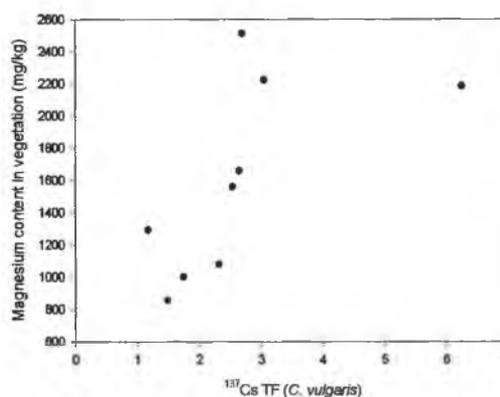


Figure 78: Plot of  $^{137}\text{Cs}$  TF versus total Mg content of vegetation (*C. vulgaris*)



Further investigation were carried out on *C. vulgaris* with regard to how the radionuclides were taken up by the different plant compartments (leaves and stems) in order to assess if the soil and nutrients status have any impact on the uptake of these radionuclides within the plant compartments.

Radium-226 uptake in the leaves of *C. vulgaris* was found to be positively correlated with the levels of available sodium in the soil as shown in Figure 79 (Pearson correlation coefficient of 0.943,  $p = 0.016$ ). Radium-226 uptake in the stems of *C. vulgaris* was found to be negatively correlated with the moisture content of the soil as shown in Figure 80 (Pearson correlation coefficient of -0.96,  $p = 0.036$ ). A similar observation was noted between  $^{226}\text{Ra}$  TF values for *C. vulgaris* stems and the pH of the soil where the results indicate that uptake by the stems decreased as the soil pH increased as shown in Figure 81 (Pearson correlation coefficient of -0.993,  $p = 0.006$ ). An inverse relationship was also noted between  $^{226}\text{Ra}$  TF values and the calcium content of the stems (Figure 82) where

the results indicate that  $^{226}\text{Ra}$  was highest in the stems which contained the lowest concentration of calcium (Pearson correlation coefficient of -0.91,  $p = 0.089$ ). This result is in agreement with the findings of Arkhipov *et al* (1984), Makrose *et al* (1993) and Blanco Rodríguez *et al* (2002) who reported diminished uptake of  $^{226}\text{Ra}$  in the presence of high levels of calcium.

Figure 79: Plot of  $^{226}\text{Ra}$  TF versus Avail Na in soil (*C. vulgaris* leaves)

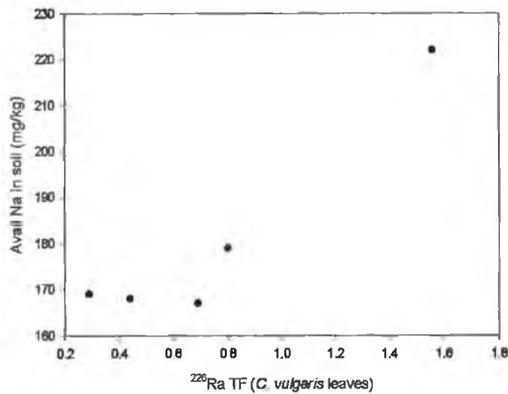


Figure 80: Plot of  $^{226}\text{Ra}$  TF versus % Moisture in soil (*C. vulgaris* stems)

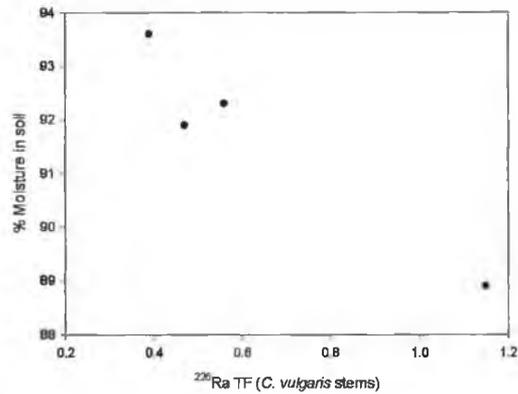


Figure 81: Plot of  $^{226}\text{Ra}$  TF versus Soil pH (*C. vulgaris* stems)

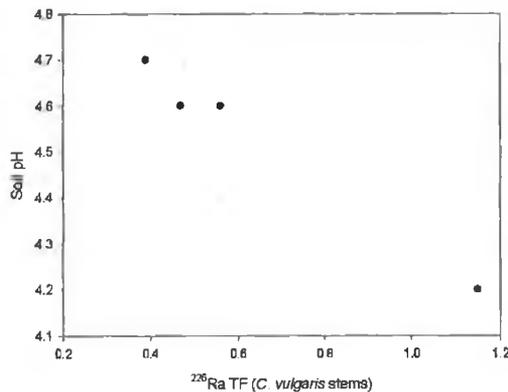
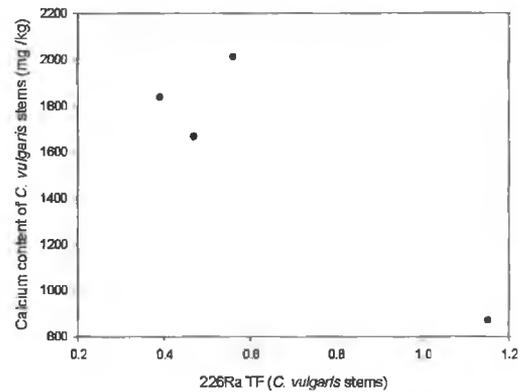


Figure 82: Plot of  $^{226}\text{Ra}$  TF versus calcium content of stems (*C. vulgaris* stems)



Uptake of  $^{40}\text{K}$  by the leaves of *C. vulgaris* was found to be affected by the available magnesium levels in the soil where a Pearson correlation coefficient of 0.922 ( $p = 0.026$ ) indicates a strong positive correlation between  $^{40}\text{K}$  up the leaves and the available levels of magnesium in the soil (Figure 83). However the opposite was seen for  $^{40}\text{K}$  uptake in the stems of *C. vulgaris* and the available levels of potassium in the soil where a Pearson

correlation coefficient of -0.98 ( $p = 0.019$ ) indicates that the uptake of  $^{40}\text{K}$  in the stems of *C. vulgaris* is lower on soils with higher concentrations of available potassium present (Figure 84). No significant correlation was found to be present between the leaves and stems (separately) and the nutrient status of the vegetation.

Figure 83: Plot of  $^{40}\text{K}$  TF versus Avail. Mg in soil (*C. vulgaris* leaves)

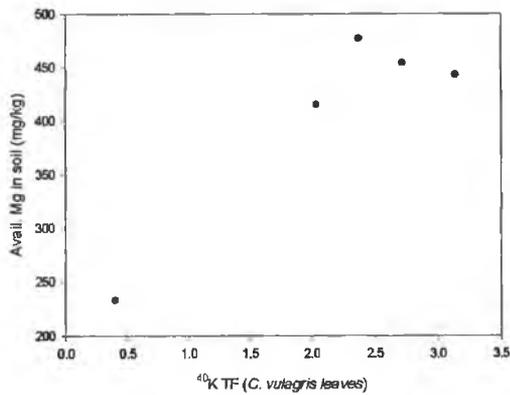
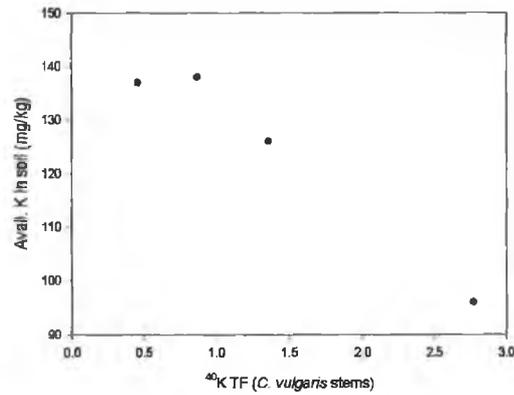


Figure 84: Plot of  $^{40}\text{K}$  TF versus Avail. K in soil (*C. vulgaris* stems)



Caesium-137 uptake by the leaves and stems of *C. vulgaris* (separately) appear to be independent of both the soil properties studied and nutrient status of vegetation.

Radium-226 uptake by *M. gale* appears to be independent of both the soil properties studied and nutrient status of vegetation. Potassium-40 uptake by *M. gale* was found to be significantly correlated with the pH, organic matter content, available magnesium content and available calcium content of the soil. A Spearman correlation coefficient of 0.71 ( $p = 0.008$ ) indicates a significant positive correlation between the uptake of  $^{40}\text{K}$  by *M. gale* and the pH of the soil as illustrated in Figure 85. Potassium-40 uptake by *M. gale* was found to increase as the organic matter content of the soil increased as shown in Figure 86 (Spearman correlation coefficient of 0.62,  $p = 0.029$ ).

Figure 85: Plot of  $^{40}\text{K}$  TF versus soil pH (*M. gale*)

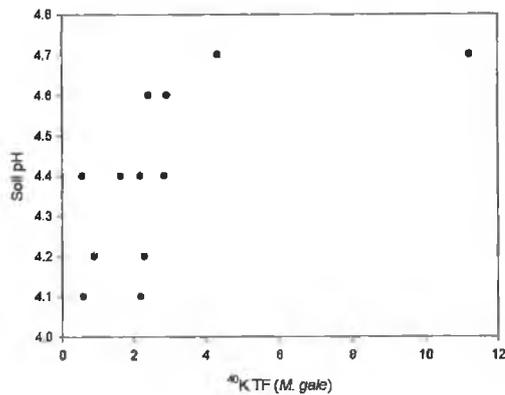
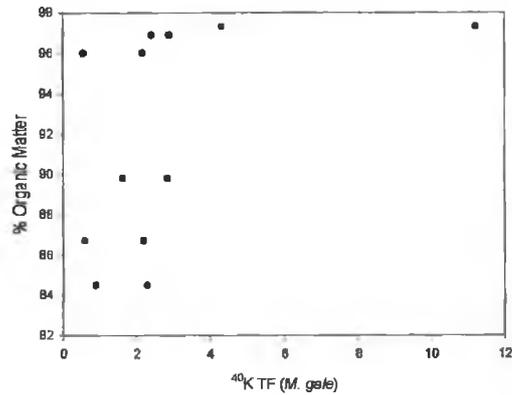


Figure 86: Plot of  $^{40}\text{K}$  TF versus soil organic matter (*M. gale*)



A Spearman correlation coefficient of 0.62 ( $p = 0.029$ ) indicates a relatively strong positive correlation between the uptake of  $^{40}\text{K}$  by *M. gale* and the levels of available magnesium in the soil as shown in Figure 87. Potassium-40 uptake by this vegetation was also found to be positively correlated with the levels of calcium in the vegetation as illustrated in Figure 88 (Spearman correlation coefficient of 0.90 ( $p = 0.000$ )).

Figure 87: Plot of  $^{40}\text{K}$  TF versus Avail. Mg in soil (*M. gale*)

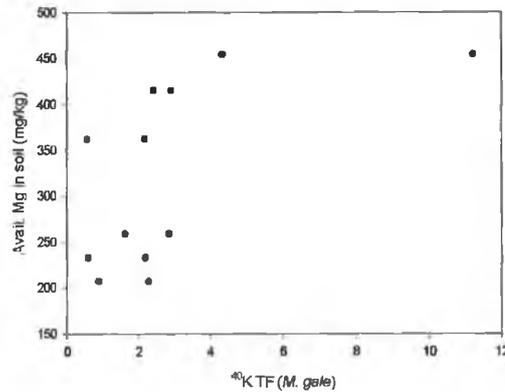
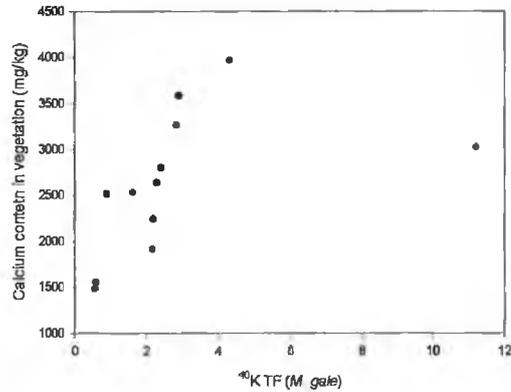


Figure 88: Plot of  $^{40}\text{K}$  TF versus Ca content of vegetation (*M. gale*)



Caesium-137 TF values for *M. gale* showed little correlation with soil properties with the exception of available sodium in the soil where TF values decrease as the available levels of sodium in the soil increased as shown in Figure 89 (Spearman correlation coefficient of -0.76 ( $p = 0.003$ )). This result is in agreement with the uptake of  $^{137}\text{Cs}$  by *E. tetralix* within this ecosystem. The uptake of  $^{137}\text{Cs}$  by *M. gale* was correlated with the nutrient status of this vegetation and results indicate that TF values increased as the calcium

content of the vegetation decreased as shown in Figure 90 (Spearman correlation coefficient of -0.64,  $p = 0.022$ ). The opposite was observed between  $^{137}\text{Cs}$  uptake and magnesium levels where a Spearman correlation coefficient of 0.69 ( $p = 0.011$ ) indicates that the transfer of  $^{137}\text{Cs}$  was higher in vegetation with high levels of calcium (Figure 91).

Figure 89: Plot of  $^{137}\text{Cs}$  TF versus Avail. Na in soil (*M. gale*)

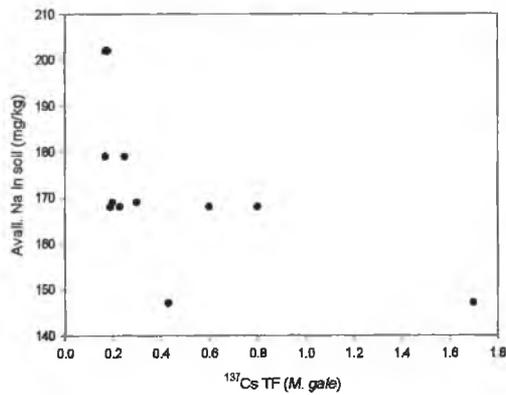


Figure 90: Plot of  $^{137}\text{Cs}$  TF versus calcium content in vegetation (*M. gale*)

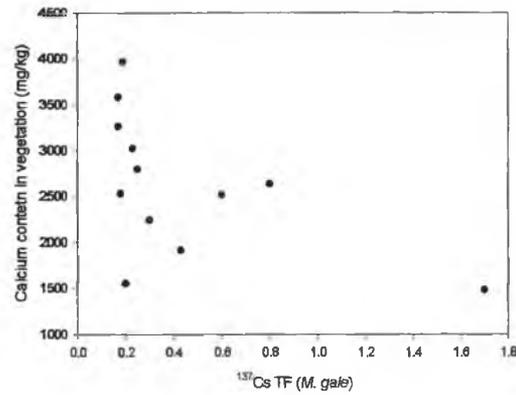
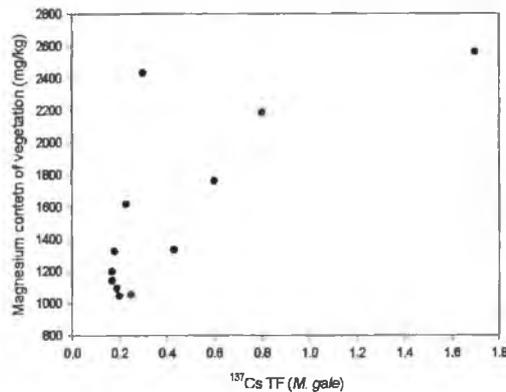


Figure 91: Plot of  $^{137}\text{Cs}$  TF versus magnesium content of vegetation (*M. gale*)

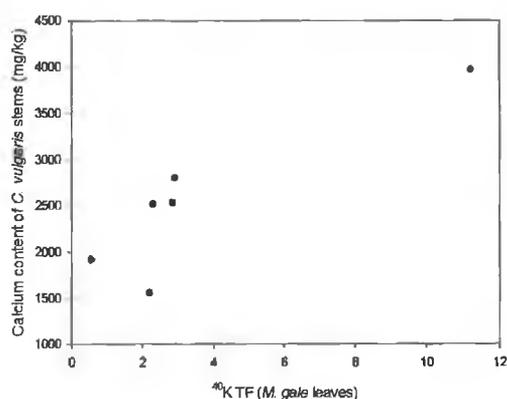


Further investigation were carried out on *M. gale* with regard to how the radionuclides were taken up by the different plant compartments (leaves and stems) in order to assess if the soil and nutrients status have any impact on the uptake of these radionuclides within the plant compartments.

Radium-226 uptake in the leaves and stems were compared independently with the nutrient status of vegetation and soil physiochemical characteristics in order to ascertain if uptake of the particular radionuclides in the different compartments is affected by any of the aforementioned properties. Radium-226 uptake in either the leaves or stems when assessed separately showed little correlation with the nutrient status or soil properties.

Potassium-40 uptake by the leaves of *M. gale* showed little correlation with any of the aforementioned soil properties, however,  $^{40}\text{K}$  uptake by the leaves was found to be positively correlated with the calcium content of the stems as shown in Figure 92 (Spearman correlation coefficient of 0.94,  $p = 0.017$ ).

Figure 92: Plot of  $^{40}\text{K}$  TF versus calcium content of *M. gale* stems (*M. gale* leaves)



Potassium-40 uptake by the stems of *M. gale* appear to be influenced by the moisture content of the soil and results indicate, as illustrated in Figure 93, that as the moisture content of the soil increases the uptake of  $^{40}\text{K}$  in the stems was increased (Pearson correlation coefficient of 0.83,  $p = 0.04$ ). The soil pH was observed to have a positive impact on the transfer of  $^{40}\text{K}$  in *M. gale* stems as shown in Figure 94 (Pearson correlation coefficient of 0.93,  $p = 0.006$ ). A similar observation was made for the organic matter content of soil where a Pearson correlation coefficient of 0.84 ( $p = 0.03$ ) indicate a strong positive correlation between the organic matter in the soil and the transfer of  $^{40}\text{K}$  to the stems of *M. gale* (Figure 95).

Figure 93: Plot of  $^{40}\text{K}$  TF versus moisture content of soil (*M. gale* stems)

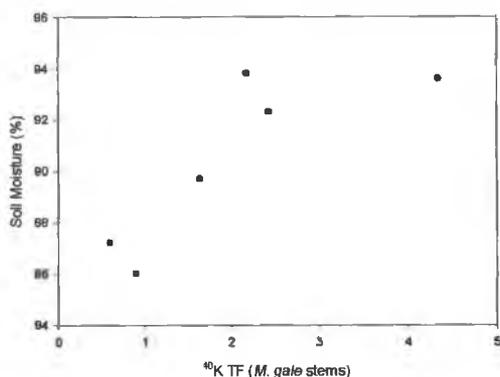


Figure 94: Plot of  $^{40}\text{K}$  TF versus soil pH (*M. gale* stems)

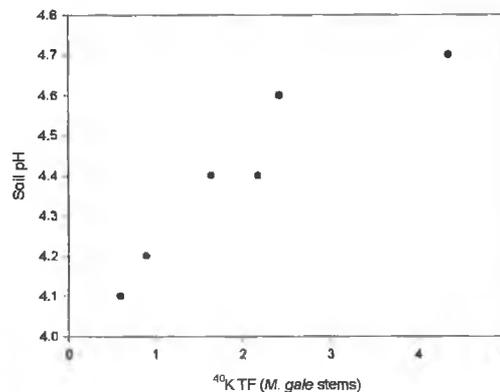
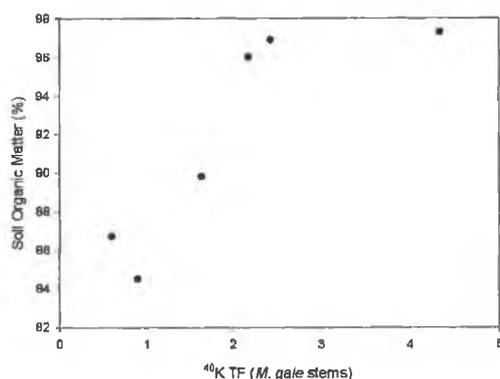


Figure 95: Plot of  $^{40}\text{K}$  TF versus organic matter content of soil (*M. gale* stems)



The uptake of  $^{40}\text{K}$  by the stems of *M. gale* was also found to be positively correlated with both the available magnesium and calcium levels in the soil. A Pearson correlation coefficient of 0.91 ( $p = 0.011$ ) indicates that as the available magnesium levels of the soil increased so did the uptake of  $^{40}\text{K}$  by the stems of *M. gale* (Figure 96). A similar observation was made between the available calcium levels in the soil and the uptake of  $^{40}\text{K}$  by this vegetation as shown in Figure 97 (Pearson correlation coefficient of 0.95,  $p = 0.003$ ). The uptake of  $^{40}\text{K}$  by the stems showed little correlation with the nutrient status of the vegetation with exception to the calcium content of the stems (Figure 98), where results indicate that stems with high levels of calcium also contained high levels of  $^{40}\text{K}$  (Pearson correlation coefficient of 0.85,  $p = 0.03$ ).

Figure 96: Plot of  $^{40}\text{K}$  TF versus Avail. Mg in soil  
(*M. gale* stems)

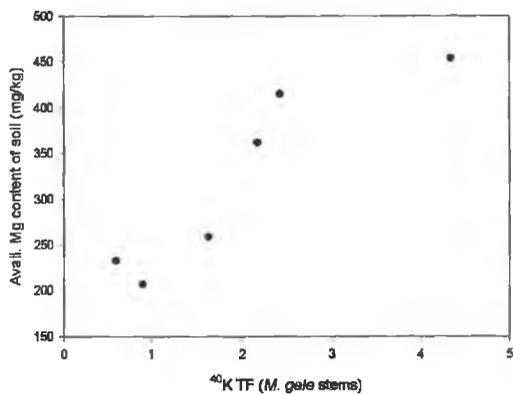


Figure 97: Plot of  $^{40}\text{K}$  TF versus Avail. Ca in soil  
(*M. gale* stems)

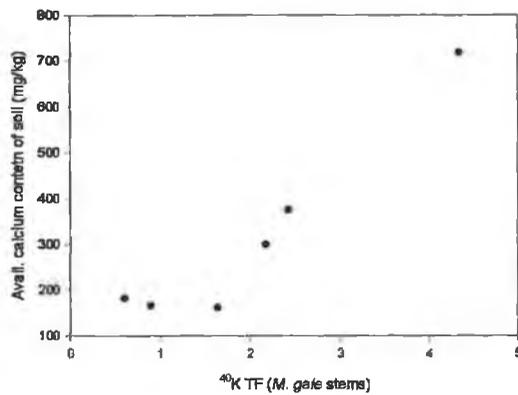
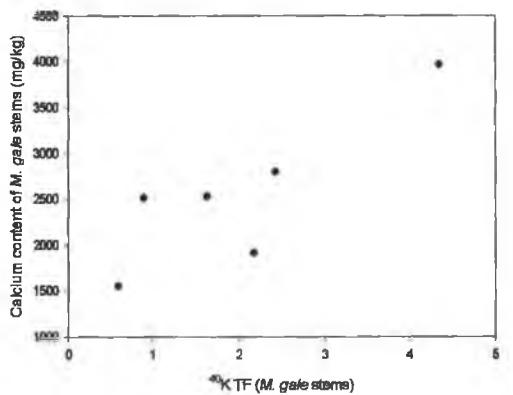


Figure 98: Plot of  $^{40}\text{K}$  TF versus calcium content of *M. gale* stems  
(*M. gale* stems)



### 3.2.7 Summary for Upland Blanket Bog

The results of this study indicate that for the upland blanket bog ecosystem under investigation, uptake of  $^{226}\text{Ra}$  in vascular plants was significantly higher for *E. tetralix* species in comparison to all other vegetation. Radium-226 TF values for all other vegetation were statistically similar however in the case of *M. gale* the concentration of  $^{226}\text{Ra}$  in the leaves was below the limit of detection for methods used with the exception of one sample. The transfer of  $^{226}\text{Ra}$  into vegetation (based on all vegetation) was found to be significantly increased where vegetation is growing on soils with higher concentration of organic matter, plant available magnesium and high CEC, however, was diminished in the presence of high levels of plant available potassium. In relation to the individual plant species and  $^{226}\text{Ra}$  TF value, results for *M. gale* indicate that the uptake of  $^{226}\text{Ra}$  is diminished where the plant is growing on soils with higher concentration of  $^{40}\text{K}$ . Uptake of  $^{226}\text{Ra}$  by *E. vaginatum* in relation to its TF values were found to be significantly increased when growing on soils with higher levels of soil moisture, organic matter content and plant available calcium. Radium-226 TF values for *M. caerulea*, *C. vulgaris* and *E. tetralix* were independent of the soil physiochemical properties and vegetation nutrients studied. However, when assessing  $^{226}\text{Ra}$  TF values of *C. vulgaris* based on its plant compartments, the transfer of  $^{226}\text{Ra}$  into the leaves of *C. vulgaris* was higher when growing on soil with high levels of plant available sodium. In relation to the stems of *C. vulgaris*,  $^{226}\text{Ra}$  TF values were significantly lower on soils with higher levels of soil moisture and soil pH. Radium-226 transfer in the stems of this vegetation was also significantly lower in samples which contained high concentrations of calcium.

Potassium-40 TF values were found to be statistically similar for all vegetation species which was also the case when comparing leaves and stems of both *C. vulgaris* and *M. gale*. These results indicate that uptake of this radionuclide by the vegetation was not dependent on plant species and was also not influenced by the plant compartments. Potassium-40 TF values (based on all vegetation) were found to increase when growing on soil with higher concentrations of organic matter, plant available calcium and magnesium, high soil pH and CEC. However the TF values for vegetation (all vegetation

grouped together) were diminished when growing on soil with high levels of available potassium. A similar observation was made between the levels of  $^{40}\text{K}$  in vegetation and its concentration in soil. Transfer of  $^{40}\text{K}$  into *M. caerulea* was found to be dependent on the levels of plant available calcium and magnesium where TF values increased for this species of grass when it was found growing on soils with higher levels of both of these nutrients. The uptake of  $^{40}\text{K}$  by *M. gale* as stated previously was negatively influenced by high concentration of  $^{40}\text{K}$  in the soil. In relation to  $^{40}\text{K}$  TF values for this vegetation they were found to be positively influenced by high soil pH, high levels of organic matter and plant available magnesium and also by high levels of calcium in the vegetation. Potassium-40 TF values for the leaves of *M. gale* were also found to increase as the calcium levels in the stems decreased. Uptake by the stems of *M. gale* with regard to its TF values was found to be positively correlated with soil moisture, pH and organic matter content of the soil. Transfer of  $^{40}\text{K}$  into *C. vulgaris* was found to increase when growing on soil with high levels of organic matter but was diminished in the presence of high levels of plant available potassium in the soil which was also the case when correlating  $^{40}\text{K}$  TF values for the stems of *C. vulgaris* and plant available potassium in the soil. Transfer of  $^{40}\text{K}$  in to the leaves of *C. vulgaris* was also found to be positively correlated with levels of plant available magnesium in the soil.

Caesium-137 TF values based on the vegetation grouped together were increased in vegetation with high concentrations of magnesium present. Transfer of  $^{137}\text{Cs}$  into *E. tetralix* was higher in the presence of high levels of magnesium in the plants but was adversely affected by levels of both  $^{137}\text{Cs}$  and plant available sodium in the soil. Transfer of  $^{137}\text{Cs}$  into *E. vaginatum* was found to decrease in the presence of high levels of calcium in the vegetation. Caesium-137 TF values for *C. vulgaris* were positively correlated with the levels of both calcium and magnesium in the vegetation whereas TF values for the leaves and stems of this vegetation when assessed separately appear to be independent of both the soil properties and vegetation nutrients examined. Uptake of  $^{137}\text{Cs}$  by *M. gale* in relation to its TF values was increased in vegetation with high concentrations of magnesium present but the opposite was evident in vegetation with high levels of calcium present. Transfer factors values were also diminished in the

presence of high levels of both  $^{137}\text{Cs}$  and plant available sodium in the soil. Transfer factors for this vegetation was also compared based on its plant compartments sampled (leaves and stems) and results indicate that  $^{137}\text{Cs}$  TF values for the individual plant compartment were independent of the soil parameters and vegetation nutrients examined.

Transfers factors values for  $^{226}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  for all vegetation were found to be independent of the concentrations of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in the soil. Radium-226 and  $^{137}\text{Cs}$  TF values were also independent of  $^{40}\text{K}$  concentration whereas  $^{40}\text{K}$  TF for all vegetation grouped together was shown to be adversely affected by the concentration of  $^{40}\text{K}$  in the soil. Caesium-137 TF for *E. tetralix* and *M. gale* were found to be negatively correlated with the concentration of  $^{137}\text{Cs}$  in the soil.

Soil moisture appears to have little impact on radionuclide TF values with the exception of  $^{226}\text{Ra}$  TF values for *E. vaginatum* as these two parameters were found to be positively correlated with each other. Soil pH was found to be significantly important in the transfer of  $^{40}\text{K}$  into vegetation as results indicate that TF values for all vegetation and in particular *M. gale* were higher on soils with high pH. Organic matter content of the soil appears to have a positive impact on both  $^{226}\text{Ra}$  and  $^{40}\text{K}$  TF values based on all vegetation grouped together. This was also the case for  $^{226}\text{Ra}$  TF values for *E. vaginatum* and  $^{40}\text{K}$  TF values for *C. vulgaris* and *M. gale*.

Plant available levels of magnesium in the soil was found to have a positive impact on both  $^{226}\text{Ra}$  and  $^{40}\text{K}$  TF values for the vegetation as a whole and also on  $^{40}\text{K}$  TF values for *M. caeruleae* and *M. gale*. Plant available levels of sodium in the soil were found to have an adverse impact on  $^{137}\text{Cs}$  TF values for both *E. tetralix* and *M. gale*. Levels of plant available calcium were found to be positively correlated with  $^{40}\text{K}$  TF values for the vegetation group as a whole and in particular *M. caeruleae* and also on  $^{226}\text{Ra}$  TF values for *E. vaginatum*. Levels of plant available potassium in the soil was found to have an adverse impact on both  $^{226}\text{Ra}$  and  $^{40}\text{K}$  TF values for all vegetation grouped together and also for  $^{40}\text{K}$  TF values for *C. vulgaris*. The CEC of the soil was found to be positively correlated with both  $^{226}\text{Ra}$  and  $^{40}\text{K}$  TF values for all vegetation grouped together.

The magnesium content of the vegetation was found to be positively correlated with  $^{137}\text{Cs}$  TF values for all vegetation grouped together and in particular, *E. tetralix*, *C. vulgaris* and *M. gale*. The calcium content of the vegetation was found to have a positive impact on  $^{40}\text{K}$  TF values for *M. gale*. It was also found to have a conflicting impact on  $^{137}\text{Cs}$  TF values as results indicate that it was positively correlated with TF values for *C. vulgaris* but was negatively correlated with TF values for both *E. vaginatum* and *M. gale*. Transfer factor values for  $^{226}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  appear to be independent of the sodium content of the vegetation for all species sampled.

In summary the parameters which were found to have a significantly positive impact on radionuclide TF values were: moisture content, soil pH, soil organic matter, plant available levels of calcium and magnesium in soil, soil CEC and magnesium content of vegetation. Levels of plant available sodium and potassium,  $^{40}\text{K}$  in soil and  $^{137}\text{Cs}$  in soil were all found to have a significant negative affect on some TF values and the impact of the calcium content of vegetation was dependent on both radionuclide and vegetation species.

Transfer factor values for the three isotopes ( $^{226}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$ ) were compared based on all plant species and results indicate that on average  $^{226}\text{Ra}$  TF values were found to be the lowest followed by  $^{137}\text{Cs}$  TF and the highest TF values were found to be for  $^{40}\text{K}$ . Potassium-40 TF values were found to be significantly higher than both  $^{226}\text{Ra}$  and  $^{137}\text{Cs}$ , and comparisons between  $^{226}\text{Ra}$  and  $^{137}\text{Cs}$  indicate that the TF values are not significantly different.

### 3.3 Wet Grassland

The grassland site classified as semi-natural grassland is situated in Doonally, Sligo along the west coast of Ireland. The dominant vegetation in this area was *Holcus lanatus* (Yorkshire fog), *Juncus effuses* (soft rushes), *Urtica dioica* (Stinging Nettle), *Crataegus monogyna* (Hawthorn tree), *Acer pseudoplatanus* (Sycamore tree) and moss.

Soil and vegetation samples were collected from eleven sites within the wet grassland. The distribution of radionuclides and physiochemical characteristics within the soil are discussed in sections 3.3.1 and 3.3.2 respectively. Radionuclide and nutrient content of vegetation indigenous to this ecosystem are discussed in sections 3.3.3 and 3.3.4 respectively. Transfer factors were calculated for each radionuclide and are discussed in section 3.3.5 and correlations between these transfer factors and the chemical characteristics of the soil and vegetation are discussed in section 3.3.6. Section 3.3.7 highlights the most significant finding from the investigation within this ecosystem.

#### 3.3.1 Radionuclide distribution in soil

The radionuclide distribution within the soil was determined by analysing the  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  content of four soil layers i.e. 0-5 cm, 5-10 cm 10-15 cm and 15-20 cm respectively at eleven sites (G1 – G11). Results are reported in sections 3.3.1.1 to 3.3.1.2 below and outlined in Table 31.

##### 3.3.1.1 $^{238}\text{U}$ , $^{226}\text{Ra}$ and $^{228}\text{Ra}$ distribution in soil

Uranium-238 activities in the wet grassland soil ranged from 18.5 Bq/kg to 24.8 Bq/kg at the eleven sites sampled and statistical analysis of the data indicate that the average concentration of  $^{238}\text{U}$  at each site was statistically similar (One Way ANOVA,  $p = 0.15$ ). The concentration of  $^{238}\text{U}$  in the four soil layers at each site was not significantly different indicating that levels of this radioisotope remained relatively constant

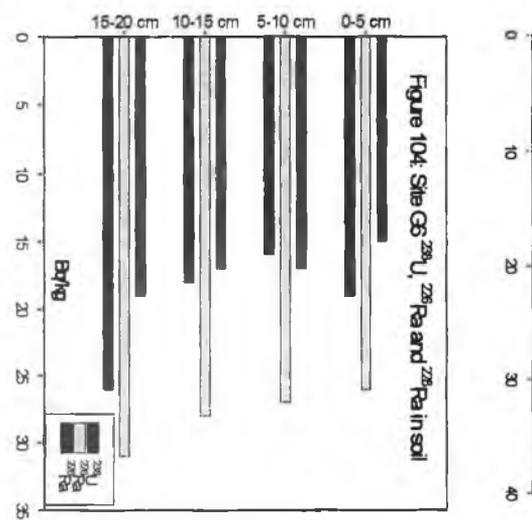
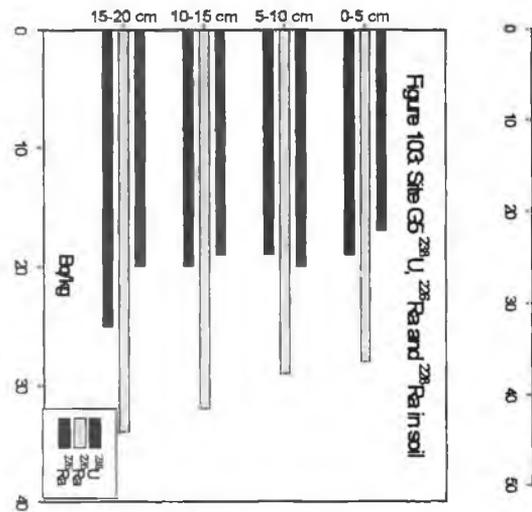
throughout the top 20 cm of soil (One Way ANOVA,  $p > 0.05$ ) as illustrated in Figures 99 - 109.

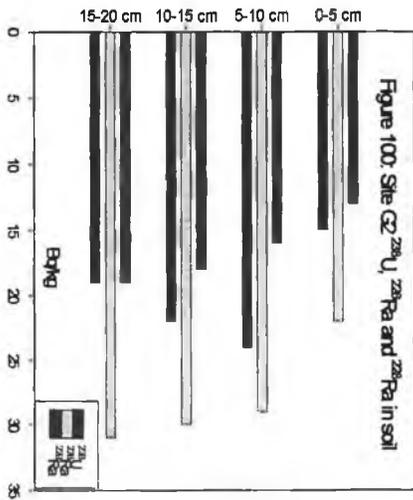
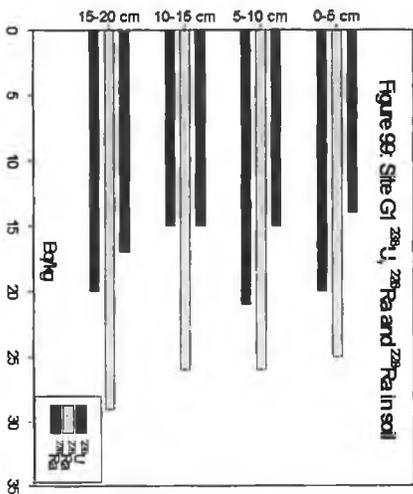
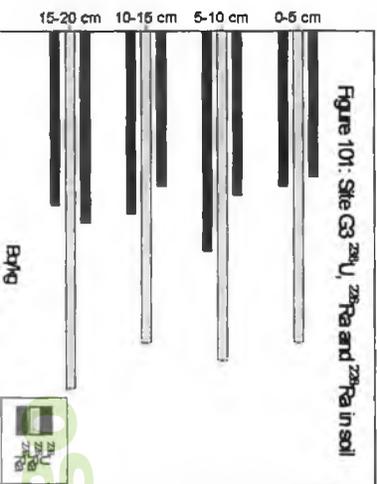
Radium-226 levels in the soil ranged from 25 Bq/kg to 45.8 Bq/kg and the average levels of  $^{226}\text{Ra}$  found in the soil at site G10 was significantly higher than all other sites with the exception of G7, G9 and G11 (One Way ANOVA,  $p < 0.05$ ). The concentration of  $^{226}\text{Ra}$  in the soil at G7 was significantly higher than all other sites with the exception of G9, G10 and G11 (One Way ANOVA,  $p < 0.05$ ). The concentration of  $^{226}\text{Ra}$  in the soil at G11 was significantly higher than the levels found at sites G1 – G6. The concentration of  $^{226}\text{Ra}$  in the soil at G11 was significantly higher than the levels found at the following sites: G1, G2, G4, G5, G6 and G8. The concentration of  $^{226}\text{Ra}$  in the soil at G3 was significantly higher than the levels found at G1, G2 and G6. The concentration of  $^{226}\text{Ra}$  in the soil at G4 was significantly higher than the levels found at G1 and G8. The concentration of  $^{226}\text{Ra}$  in the four soil layers were not significantly different indicating that levels of this radioisotope remained relatively constant throughout the top 20 cm of soil (One Way ANOVA,  $p = 0.74$ ) as illustrated in Figures 99- 109.

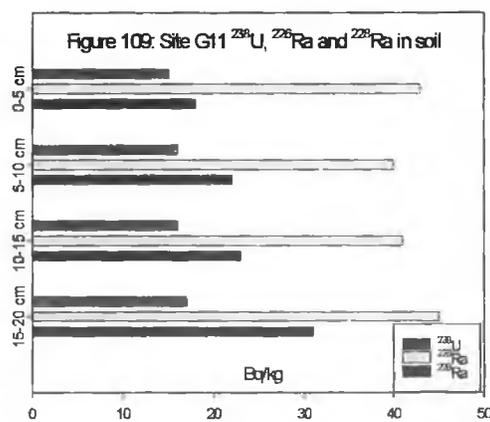
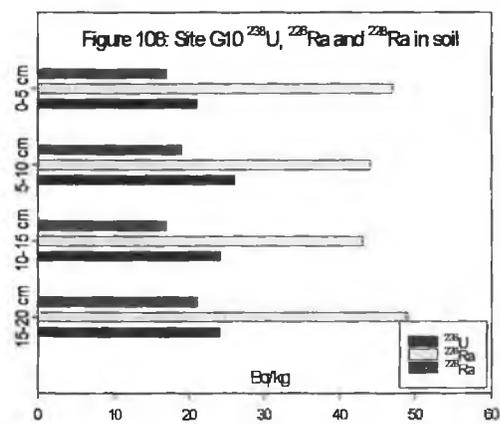
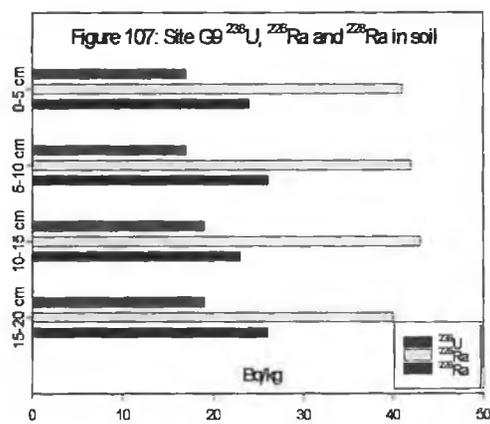
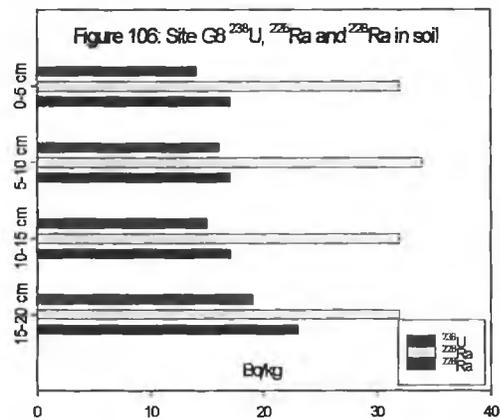
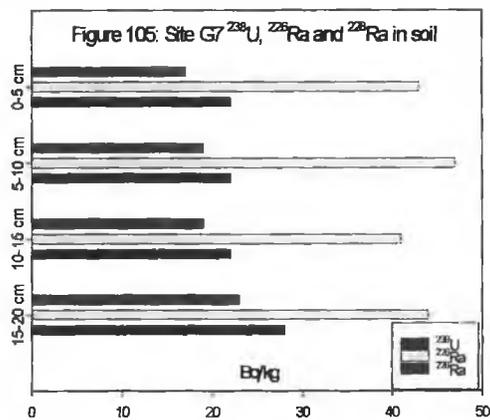
The relationship between  $^{238}\text{U}$  and  $^{226}\text{Ra}$  in the soil was examined using the  $^{226}\text{Ra}/^{238}\text{U}$  equilibrium ratio. The ratio for each site ranged from 1.4 to 1.9 indicating that the  $^{238}\text{U}$  concentration in the soil was lower than  $^{226}\text{Ra}$  at all sites, however, comparisons between the actual concentrations of  $^{238}\text{U}$  and  $^{226}\text{Ra}$  show that the differences were statistically significant (t-test,  $p < 0.001$ ). The ratio of  $^{226}\text{Ra}$  to  $^{238}\text{U}$  in the soil at each site was statistically similar as is the case for the individual soil layers (One Way ANOVA,  $p > 0.05$ ). The fact that  $^{238}\text{U}$  is lower in the soil at each site may indicate that  $^{238}\text{U}$  is being leached to the lower layers of soil or that  $^{226}\text{Ra}$  is being preferentially retained in comparison to  $^{238}\text{U}$  in the upper layers of soil.

Site	Soil Layer	<sup>238</sup> U (Bq/kg)	<sup>226</sup> Ra (Bq/kg)	<sup>228</sup> Ra (Bq/kg)	<sup>40</sup> K (Bq/kg)	<sup>137</sup> Cs (Bq/kg)
G 1	0-5 cm	20 ± 2	25 ± 2	14 ± 1	277 ± 18	107 ± 8
	5-10 cm	21 ± 2	26 ± 2	15 ± 1	289 ± 22	72 ± 5
	10-15 cm	15 ± 2	26 ± 2	15 ± 1	287 ± 23	46 ± 3
	15-20 cm	20 ± 2	29 ± 3	17 ± 2	287 ± 22	7 ± 0.5
G 2	0-5 cm	15 ± 2	22 ± 2	13 ± 1	240 ± 18	79 ± 5
	5-10 cm	24 ± 2	29 ± 3	16 ± 1	324 ± 25	67 ± 4
	10-15 cm	22 ± 2	30 ± 3	18 ± 2	332 ± 25	36 ± 3
	15-20 cm	19 ± 2	31 ± 3	19 ± 2	338 ± 29	12 ± 1
G 3	0-5 cm	17 ± 2	34 ± 3	16 ± 2	263 ± 21	73 ± 5
	5-10 cm	24 ± 2	36 ± 3	18 ± 2	320 ± 35	69 ± 5
	10-15 cm	20 ± 2	34 ± 3	17 ± 2	320 ± 35	30 ± 2
	15-20 cm	19 ± 2	39 ± 3	21 ± 3	327 ± 26	3 ± 0.4
G 4	0-5 cm	26 ± 3	30 ± 3	16 ± 1	271 ± 27	68 ± 6
	5-10 cm	18 ± 2	34 ± 2	16 ± 1	280 ± 20	64 ± 4
	10-15 cm	15 ± 1	33 ± 2	16 ± 1	275 ± 21	32 ± 2
	15-20 cm	20 ± 2	34 ± 2	16 ± 1	302 ± 23	7 ± 0.5
G 5	0-5 cm	19 ± 2	28 ± 2	17 ± 1	290 ± 25	119 ± 7
	5-10 cm	19 ± 2	29 ± 2	20 ± 2	290 ± 22	98 ± 6
	10-15 cm	20 ± 2	32 ± 2	19 ± 2	302 ± 23	28 ± 2
	15-20 cm	25 ± 3	34 ± 3	20 ± 2	318 ± 24	12 ± 1
G 6	0-5 cm	19 ± 2	26 ± 2	15 ± 1	309 ± 23	116 ± 7
	5-10 cm	16 ± 2	27 ± 2	17 ± 2	315 ± 25	95 ± 6
	10-15 cm	18 ± 2	28 ± 2	17 ± 1	316 ± 23	34 ± 2
	15-20 cm	26 ± 2	31 ± 2	19 ± 2	335 ± 26	9 ± 0.6
G 7	0-5 cm	22 ± 2	43 ± 3	17 ± 1	316 ± 23	73 ± 5
	5-10 cm	22 ± 2	47 ± 4	19 ± 2	337 ± 23	30 ± 2
	10-15 cm	22 ± 2	41 ± 3	19 ± 2	344 ± 25	23 ± 1
	15-20 cm	28 ± 2	44 ± 3	23 ± 2	353 ± 26	16 ± 1
G 8	0-5 cm	17 ± 1	32 ± 2	14 ± 1	266 ± 19	116 ± 7
	5-10 cm	17 ± 1	34 ± 2	16 ± 1	281 ± 19	98 ± 8
	10-15 cm	17 ± 1	32 ± 2	15 ± 1	263 ± 18	46 ± 3
	15-20 cm	23 ± 2	32 ± 2	19 ± 2	288 ± 20	20 ± 1
G 9	0-5 cm	24 ± 2	41 ± 3	17 ± 1	325 ± 23	84 ± 5
	5-10 cm	26 ± 2	42 ± 3	17 ± 1	354 ± 25	70 ± 4
	10-15 cm	23 ± 2	43 ± 3	19 ± 2	352 ± 25	33 ± 2
	15-20 cm	26 ± 2	40 ± 3	19 ± 2	349 ± 24	12 ± 0.8
G 10	0-5 cm	21 ± 2	47 ± 4	17 ± 1	302 ± 21	93 ± 6
	5-10 cm	26 ± 2	44 ± 3	19 ± 2	316 ± 22	74 ± 5
	10-15 cm	24 ± 2	43 ± 3	17 ± 1	314 ± 22	25 ± 1
	15-20 cm	24 ± 2	49 ± 4	21 ± 2	346 ± 24	4 ± 0.3
G 11	0-5 cm	18 ± 2	43 ± 3	15 ± 1	281 ± 20	93 ± 6
	5-10 cm	22 ± 2	40 ± 3	16 ± 1	294 ± 21	86 ± 6
	10-15 cm	23 ± 2	41 ± 3	16 ± 1	290 ± 21	25 ± 2
	15-20 cm	31 ± 3	45 ± 3	17 ± 1	294 ± 21	10 ± 0.6
	20-25 cm	31 ± 3	43 ± 3	17 ± 1	286 ± 20	8 ± 0.6
	25-30 cm	23 ± 2	42 ± 3	17 ± 1	293 ± 21	6 ± 0.4
	30-35 cm	20 ± 2	53 ± 4	19 ± 2	337 ± 24	4 ± 0.3
35-40 cm	20 ± 2	66 ± 5	18 ± 2	300 ± 22	4 ± 0.3	

Table 31: <sup>238</sup>U, <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>40</sup>K and <sup>137</sup>Cs levels in soils at sites G1 – G11





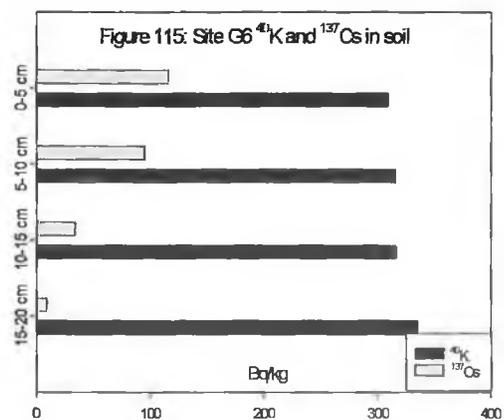
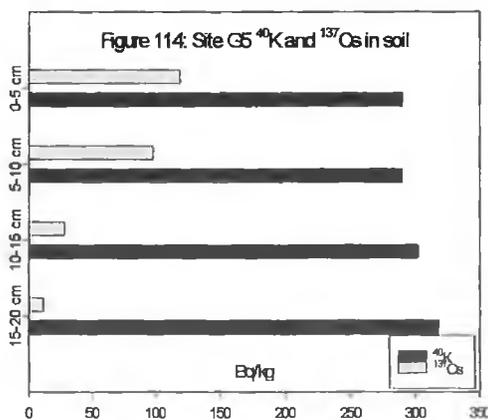
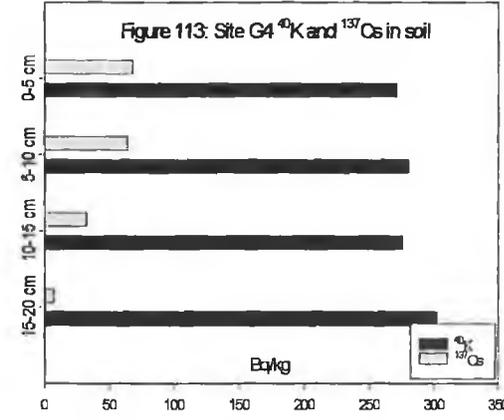
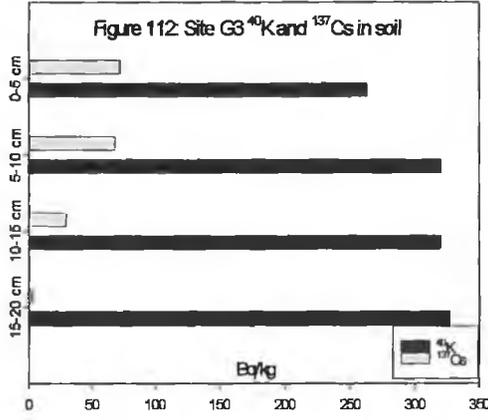
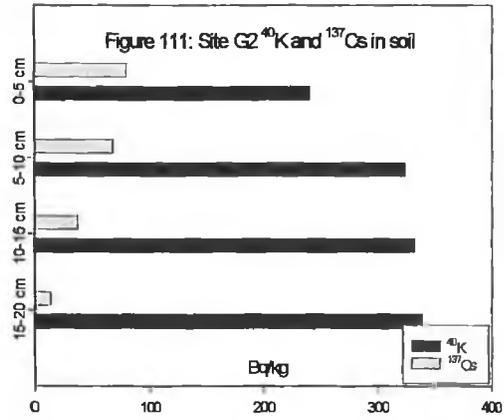
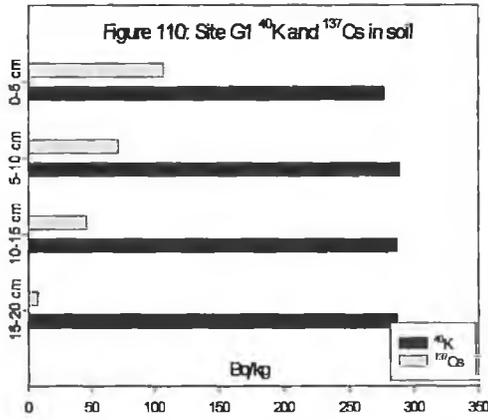


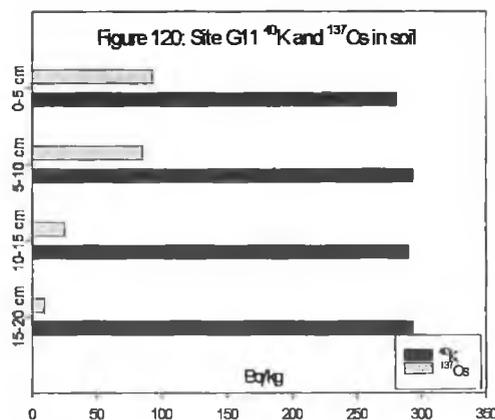
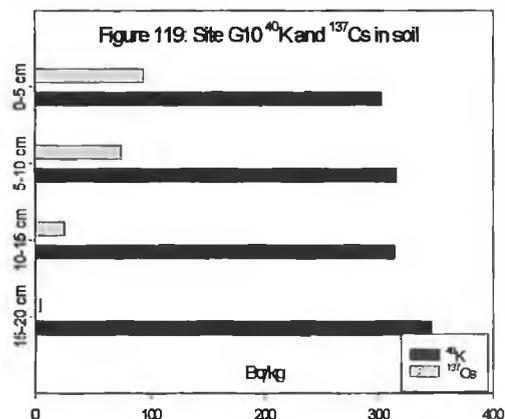
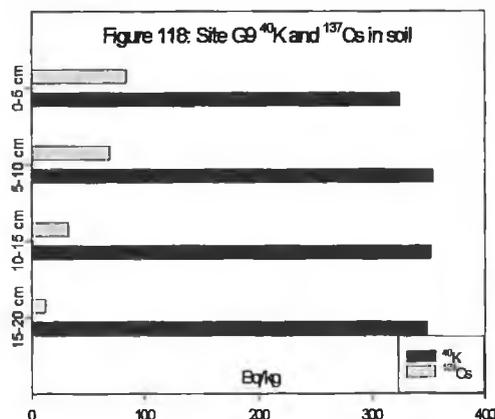
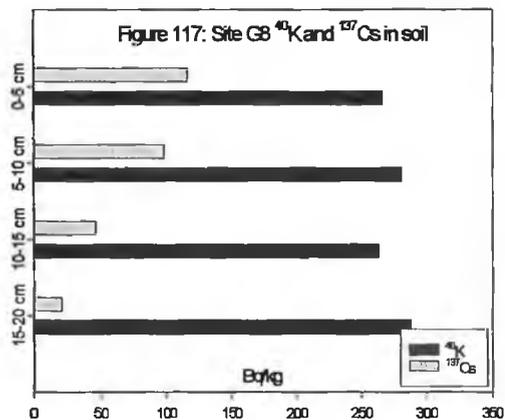
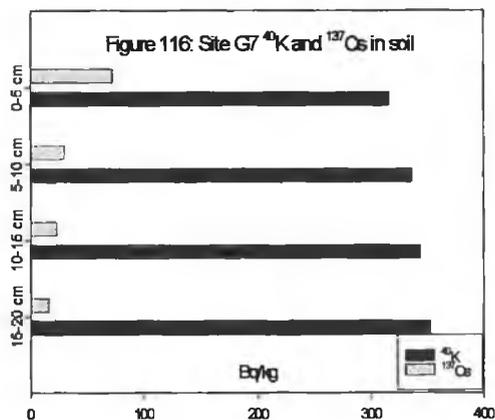
Figures 99 –109:  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  activity in soil at Sites G1 – G11.

Radium-228 levels in the grassland soil ranged from 15.3 Bq/kg to 19.5 Bq/kg and the average concentration of this radionuclide in the soil at all eleven sites was statistically similar (One Way ANOVA,  $p > 0.05$ ). The concentration in the top four soil layers were compared for each site and results indicate that the concentration of  $^{228}\text{Ra}$  in the 15-20 cm layer was significantly higher than levels found in the 0-5 cm layer (One Way ANOVA,  $p < 0.05$ ) indicating that  $^{228}\text{Ra}$  levels are higher lower down in the soil profile in comparison to the top layers of soil. The concentrations of  $^{226}\text{Ra}$  in all other soil layers were statistically similar (One Way ANOVA,  $p > 0.05$ ).

### 3.3.1.2 $^{40}\text{K}$ and $^{137}\text{Cs}$ distribution in soil

Levels of  $^{40}\text{K}$  in the soil ranged from 275 Bq/kg to 345 Bq/kg with an overall average concentration for the eleven sites sampled of 307 Bq/kg. The average concentration of  $^{40}\text{K}$  in the soil at site G9 was significantly higher than levels found at G1, G4, G8 and G11 (one Way ANOVA,  $p < 0.05$ ). The average levels of  $^{40}\text{K}$  found in the soil at site G7 was also significantly higher than concentration found in the soil at sites G1, G4 and G8 respectively. The concentration of  $^{40}\text{K}$  was significantly higher in the lower layers of soil as results indicate that levels in the 15-20 cm layer was significantly higher than those found in the 0-5 cm layer (One Way ANOVA,  $p = 0.011$ ). The concentrations of  $^{40}\text{K}$  in the other layers were statistically similar (One Way ANOVA,  $p > 0.05$ ).





Figures 110 –120:  $^{137}\text{Cs}$  and  $^{40}\text{K}$  activity in soil at Sites G1 – G11.

Caesium-137 levels in the soil ranged from 3 Bq/kg to 119 Bq/kg with an overall average concentration of 52.6 Bq/kg for the eleven sites sampled. The average concentration of  $^{137}\text{Cs}$  in the three soil layers (0-20 cm) was calculated for each site and results indicate that  $^{137}\text{Cs}$  levels in the soil between sites are not significantly different (One Way ANOVA,  $p = 0.98$ ). Caesium-137 levels appear to be highest in the upper 5 cm of soil and levels are decreasing with increasing soil depth as illustrated in Figures 110 – 120. The average concentration of  $^{137}\text{Cs}$  in the 0-5 cm layer was 92.8 Bq/kg which was significantly higher than levels found in the 5-10 cm, 10-15 cm and 15-20 cm layers respectively. The average concentration of  $^{137}\text{Cs}$  in the 5-10 cm layers was also significantly higher than levels found in both the 10-15 cm layers and 15-20 cm layer respectively. The concentration of  $^{137}\text{Cs}$  in the 10-15 cm layer was also significantly higher than levels in the 15-20 cm layer (One Way ANOVA,  $p = 0.004$ )

The decrease in  $^{137}\text{Cs}$  activity with increasing soil depth is best described using a log function as illustrated in Figure 121 where an inverse relationship was found to be present between log average  $^{137}\text{Cs}$  activity as a function of soil depth (linear regression:  $r^2 = 0.92$ ). Concentrations of  $^{137}\text{Cs}$  were also found to be lower further down in the soil profile with an activity of  $4 \pm 0.3$  Bq/kg observed in the bottom layer at site G11 (0-40 cm depth profile).

Figure 121: Log of  $^{137}\text{Cs}$  activity versus soil depth for Sites G1 - G11

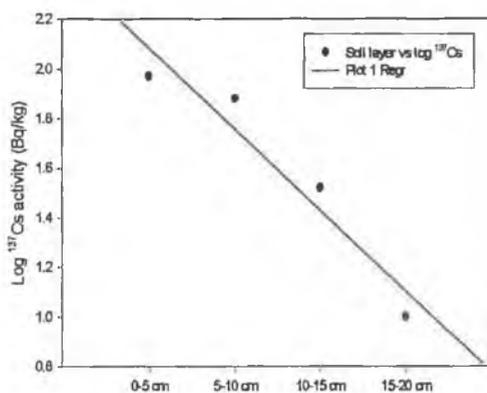


Figure 121: Log  $^{137}\text{Cs}$  activity versus soil depth for sites G1 – G11

Comparisons between radionuclide levels in the grassland soils indicate that levels of  $^{40}\text{K}$  in the soil were significantly higher than  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{137}\text{Cs}$  respectively (One Way ANOVA,  $p < 0.001$ ). Results also indicate that  $^{137}\text{Cs}$  in the soil was significantly higher than levels of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in the soil (One Way ANOVA,  $p < 0.008$ ). Uranium-238,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  levels in the soil were all statistically similar (One Way ANOVA,  $p > 0.05$ ).

### 3.3.2 Soil physiochemical properties

The physiochemical characteristics of the soil were extensively studied in order to ascertain if any of these parameters had a significant impact on the uptake of the radionuclide by vegetation. The physiochemical parameters chosen were the same as those chosen for the Atlantic blanket bog as outlined in section 3.1.2. The results for each of these parameters are presented in Table 32 and are discussed in detail within this section. Correlations between these parameters and radionuclide uptake in vegetation are discussed in section 3.3.6.

The moisture of the soil ranged from 19% to 62% with an average value of 37.7% for the eleven sites sampled. The average moisture content at each site was statistically similar (One Way ANOVA,  $p = 0.84$ ). The moisture content of the soil varied within the upper 20 cm of the soil and results indicate that the level of moisture decreases with depth. The moisture content in the 0-5 cm layer was significantly higher than the 5-10 cm, 10-15 cm and 15-20 cm layer (One Way ANOVA,  $p < 0.001$ ). The moisture content in the 5-10 cm layer was significantly higher than the 10-15 cm and 15-20 cm layer (One Way ANOVA,  $p < 0.001$ ).

Site	Soil Layer	Moisture Content %	pH	Organic Matter %	Available Magnesium mg/kg	Available Sodium mg/kg	Available Calcium mg/kg	Available Potassium mg/kg	CEC of Soil meq/100g
G 1	0-5 cm	42.3	4.9	16.0	153	48	429	154	39
	5-10 cm	35.1	5.0	11.0	100	45	275	109	42
	10-15 cm	29.9	5.0	9.4	67	38	96	87	52
	15-20 cm	24.9	5.1	5.7	50	39	256	61	40
G 2	0-5 cm	53.8	5.1	16.1	204	50	765	249	52
	5-10 cm	45.7	5.1	12.1	123	41	462	265	46
	10-15 cm	39.2	5.1	7.9	120	37	315	129	45
	15-20 cm	32.3	5.1	5.2	91	27	318	93	36
G 3	0-5 cm	58.4	5.3	17.0	229	62	1038	258	53
	5-10 cm	45.9	5.2	10.4	138	53	618	183	44
	10-15 cm	32.9	5.4	7.0	114	34	661	109	43
	15-20 cm	27.5	5.6	4.7	124	34	750	95	39
G 4	0-5 cm	49.8	5.7	17.9	244	55	1692	592	41
	5-10 cm	45.9	5.6	10.7	193	46	1256	476	57
	10-15 cm	36.4	5.4	7.4	166	34	965	288	42
	15-20 cm	27.0	5.2	4.9	140	29	777	132	33
G 5	0-5 cm	40.8	5.3	15.2	200	51	855	152	47
	5-10 cm	33.6	5.3	10.5	113	50	334	92	42
	10-15 cm	26.1	5.2	7.4	81	40	297	82	48
	15-20 cm	23.8	5.2	5.9	100	36	384	72	38
G 6	0-5 cm	43.5	4.9	16.4	141	58	287	186	57
	5-10 cm	37.2	5.0	12.6	131	54	215	153	40
	10-15 cm	28.1	4.8	9.3	103	50	202	106	36
	15-20 cm	32.9	4.8	6.8	61	38	138	66	43
G 7	0-5 cm	50.5	5.4	14.6	217	77	858	214	37
	5-10 cm	36.3	5.2	8.7	133	59	475	120	41
	10-15 cm	30.0	5.2	6.6	120	51	459	102	43
	15-20 cm	29.5	4.9	5.6	134	48	443	121	42
G 8	0-5 cm	56.9	5.3	18.0	188	64	677	125	52
	5-10 cm	47.4	5.0	14.1	132	56	505	97	43
	10-15 cm	30.6	5.0	6.4	101	52	385	69	39
	15-20 cm	24.8	5.0	4.5	88	50	311	60	40
G 9	0-5 cm	55.5	5.3	13.2	215	63	649	303	50
	5-10 cm	43.1	5.3	9.2	160	56	418	282	44
	10-15 cm	40.8	5.3	8.8	119	48	483	248	39
	15-20 cm	33.2	5.1	5.1	105	38	377	174	41
G 10	0-5 cm	62.0	5.0	14.7	168	63	844	144	49
	5-10 cm	53.4	5.1	12.8	157	53	685	196	45
	10-15 cm	34.4	5.4	7.4	148	45	615	161	44
	15-20 cm	27.2	5.6	5.7	141	40	612	83	34
G 11	0-5 cm	59.0	5.6	17.7	232	66	1121	252	40
	5-10 cm	42.6	5.3	9.0	156	59	771	167	42
	10-15 cm	29.7	5.5	5.1	120	53	573	127	39
	15-20 cm	24.1	5.2	3.7	93	53	456	67	37
	20-25 cm	25.5	5.3	4.0	74	43	456	57	29
	25-30 cm	33.3	5.3	4.1	95	42	511	70	32
	30-35 cm	27.3	5.5	4.1	102	40	515	56	35
	35-40 cm	19.9	5.4	2.3	104	42	588	46	30

Table 32: Soil physiochemical properties: Sites G1 – G11

Soil pH ranged from 4.8 to 5.7 with an average value for all eleven sites of 5.2. The pH of the soil at site G4 was significantly higher than the soil at sites G1, G6 and G8 respectively. (One Way ANOVA,  $p < 0.04$ ). Soil pH at site G11 was significantly higher than the pH of the soil at sites G1 and G6 respectively (One Way ANOVA,  $p < 0.04$ ). The pH of the soil at site G3 was also significantly higher than the soil at site G6 (One Way ANOVA,  $p = 0.004$ ). The pH of the soil in the upper 20 cm of the soil at all sites remained relatively constant as results indicate that the differences between the average value for the four soil layers are not statistically significant (one Way ANOVA,  $p = 0.82$ ).

The organic matter content of the soil at each site ranged from 2.3% to 18.0% with an average value of 9.4% for all sites sampled. The average organic matter content of the soil at each site was statistically similar (One Way ANOVA,  $p = 0.98$ ). The organic matter content of the soil at each site decreased with increasing soil depth and result indicate that the levels present in the 0-5 cm layer were significantly higher than those present in the 5-10 cm, 10-15 cm and 15-20 cm layer respectively (One Way ANOVA,  $p < 0.001$ ). Similar observations were made between the 5-10 cm layer and subsequent lower layers as results indicate that levels present in the 5-10 cm layer were significantly higher than those found in both the 10-15 cm and 15-20 cm layer (One Way ANOVA,  $p < 0.001$ ). The levels present in the 10-15 cm layer were also significantly higher than those found in the 15-20 cm layer (One Way ANOVA,  $p = 0.003$ ).

The levels of plant available magnesium in the soil ranged from 50 mg/kg to 244 mg/kg with an average value of 135.2 mg/kg for all sites sampled. The levels of plant available magnesium were on average statistically similar at each site sampled (One Way ANOVA,  $p = 0.33$ ). The available magnesium levels in the soil at each site appear to decrease with increasing soil depth. Available magnesium concentration in the 0-5 cm layer were significantly higher than levels found in the 5-10 cm, 10-15 cm, and 15-20 cm layer respectively (One Way ANOVA,  $p < 0.001$ ). A similar observation was made for the 5-10 cm layer where the concentration in this layer was significantly higher than the 15-20 cm layer (One Way ANOVA,  $p = 0.025$ ).

The levels of plant available sodium in the soil ranged from 27 mg/kg to 77 mg/kg with an average value of 48.1 mg/kg for all sites sampled. The levels of plant available sodium were on average statistically similar at each site sampled (One Way ANOVA,  $p = 0.07$ ). The levels of available sodium in the soil at each site appear to decrease with increasing soil depth. Available sodium concentrations in the 0-5 cm were significantly higher than both the 10-15 cm and 15-20 cm layer (One Way ANOVA,  $p < 0.001$ ). Similar observations were made for the 5-10 cm and 15-20 cm layer where levels of available sodium in the former were significantly higher than in the latter at each site as shown in Table 32 (One Way ANOVA,  $p = 0.002$ ).

The average levels of plant available calcium in the soil ranged from 96 mg/kg to 1692 mg/kg with an average value of 566.1 mg/kg for all sites sampled. The levels of available calcium in the soil at site G4 was significantly higher than those found in the soil at the following sites: G1, G2, G5 – G9 (One Way ANOVA,  $p < 0.01$ ). The levels of available calcium in the soil at site G3 was also significantly higher than in the soil at G6 (One Way ANOVA,  $p = 0.03$ ). The levels of available calcium in the soil at each site appear to decrease with increasing soil depth. Available calcium concentrations in the 0-5 cm were significantly higher than both the 10-15 cm and 15-20 cm layer (One Way ANOVA,  $p < 0.01$ ).

The average levels of plant available potassium in the soil ranged from 46 mg/kg to 592 mg/kg with an average value of 156.9 mg/kg for all sites sampled. The levels of available potassium in the soil at site G4 was significantly higher than levels found present in the soil at the following sites: G1, G3, G5 – G8, G10 and G11 (One Way ANOVA,  $p < 0.04$ ). The levels of available potassium in the soil at each site appear to decrease with increasing soil depth. Available calcium concentrations in the 0-5 cm were significantly higher than those found present in the 15-20 cm layer (One Way ANOVA,  $p = 0.005$ ).

The levels of plant available calcium, magnesium, sodium and potassium in the upper 20 cm of the soil at each site was compared and results indicate that their concentrations in the soil decrease in the following order:

calcium > potassium > magnesium > sodium

The average levels of calcium, magnesium, sodium and potassium in the soil were 571 mg/kg, 166 mg/kg, 139 mg/kg and 49 mg/kg respectively. The concentration of available calcium in the soil was significantly higher than levels of available sodium, magnesium and potassium respectively (One Way ANOVA,  $p < 0.001$ ). The concentrations of available sodium, magnesium and potassium were statistically similar (One Way ANOVA,  $p > 0.05$ ).

The CEC of the soil ranged from 29 meq/100g to 57 meq/100g with an average CEC of 42.1 meq/100g for all sites sampled. The CEC of the soil were on average statistically similar at each site sampled (One Way ANOVA,  $p = 0.99$ ). The CEC of the soil at each site appears to decrease with increasing soil depth. On average the CEC of the 0-5 cm layer of the soil was significantly higher than the 15-20 cm layer, as was the case with the 5-10 cm layer which was significantly higher than the 15-20 cm layer (One Way ANOVA,  $p < 0.05$ ).

In summary the soil physiochemical properties varied throughout the sites studied within the ecosystem. The moisture content of the soil at each site remained relatively constant however the levels decreased with increasing depth. The pH of the soil was highest at sites G3, G4 and G11 and was relatively similar at all other sites. Soil pH remained relatively constant within the upper 20 cm of the soil. The organic matter content of the soil at each site was relatively constant however the levels decreased with soil depth for each site. The available magnesium and sodium levels were relatively similar for each site and levels for both nutrients decreased with increasing soil depth. The levels of available calcium and potassium in the soil was significantly higher at site G4 than most other sites sampled and levels of both nutrients decreased with increasing soil depth. The CEC of the soil at each site were relatively similar and as in the case of all available nutrients the CEC decreased with increasing soil depth. The CEC of the soil is its ability to retain cations ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^{+}$  and  $\text{Na}^{+}$ ) and therefore as the CEC of the soil decreases it would also be expected that the levels of available cations would decrease as was seen in this study. A summary of the average levels of soil physiochemical properties are outlined below and results indicate that there is no overall trend with regard to these

properties for the eleven sites sampled. In general the soil nutrients appear to decrease in concentration with increasing soil depth possibly due to recycling of nutrients by roots of vegetation.

Soil Property	Sites
Moisture Content	G10 > G9 > G2 > G3 > G8 > G4 > G11 > G7 > G6 > G1 > G5
pH	G4 > G11 > G3 > G10 > G9 = G5 > G7 > G2 > G8 > G1 > G6
Organic Matter	G6 > G8 > G1 > G2 > G4 > G10 > G3 > G5 > G9 > G7 = G11
Available Magnesium	G4 > G3 > G10 > G7 > G11 > G9 > G2 > G8 > G5 > G6 > G1
Available Sodium	G7 > G11 > G8 > G9 > G10 > G6 > G3 > G5 > G1 > G4 > G2
Available Calcium	G4 > G3 > G11 > G10 > G7 > G9 > G8 > G5 > G2 > G1 > G6
Available Potassium	G4 > G9 > G2 > G3 > G11 > G10 > G7 > G6 > G1 > G5 > G8
CEC	G3 = G2 > G6 > G5 > G9 = G8 > G1 = G4 > G10 > G7 > G11

Table 33: Comparison of soil physiochemical properties at sites G1 – G11

### 3.3.3 Radionuclide content of vegetation

Levels of  $^{226}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  were analysed in the vegetation by gamma spectrometry,  $^{238}\text{U}$  was analysed in the vegetation by gamma spectrometry, alpha spectrometry and ICP-MS while  $^{228}\text{Ra}$  ( $^{232}\text{Th}$ ) was analysed in the vegetation using both gamma spectrometry and alpha spectrometry. The results for all radionuclides are outlined in Table 34. The concentration of  $^{238}\text{U}$  and  $^{228}\text{Ra}$  ( $^{232}\text{Th}$ ) in vegetation were below the limits of detection for the gamma and alpha spectrometry methods outlined in section 2.5.1 – 2.5.2. In the case of alpha spectrometry only a selection of vegetation was analysed (Table 34) and the levels of both radionuclides were below the limits of detection. The levels of  $^{238}\text{U}$  in all vegetation samples were also below the limits of detection for the ICP-MS method outlined in section 2.5.3. Radium-226 and  $^{137}\text{Cs}$  were only detectable in some of vegetation while  $^{40}\text{K}$  was detectable in all vegetation as outlined in Table 34.

Radium-226 levels in vegetation were below the limits of detection for the method used with the exception of two samples of *H. lanatus* at sites G3 and G7 where the levels were

found to be 4.6 Bq/kg and 3.0 Bq/kg respectively. Radium-226 levels were also detectable in *C. monogyna* and *A. pseudoplatanus* and the levels in the stems of *C. monogyna* were higher than the levels in the leaves of 9.6 Bq/kg and 5.8 Bq/kg respectively. The concentration of  $^{226}\text{Ra}$  found in the leaves of *A. pseudoplatanus* was 1.2 Bq/kg which was lower than those found in *C. monogyna*.

Potassium-40 concentrations in vegetation ranged from 96 Bq/kg to 1036 Bq/kg for the variety of species sampled. The concentration of  $^{40}\text{K}$  on average appears to be highest in nettles and lowest in the stems of *C. monogyna* however results indicate that there is no significant difference in the average concentration of  $^{40}\text{K}$  in all vegetation species.

Site	Vegetation	$^{238}\text{U}$		$^{226}\text{Ra}$	$^{228}\text{Ra}$		$^{40}\text{K}$	$^{137}\text{Cs}$
		(Bq/kg)		(Bq/kg)	(Bq/kg)		(Bq/kg)	(Bq/kg)
G 1	<i>Holcus lanatus</i>	< 0.23	< 0.20*	< 3.6	< 7	< 0.50*	440 ± 35	< 1.8
G 2	<i>Holcus lanatus</i>	< 0.24		< 3.0	< 6		423 ± 30	< 1.2
	<i>Juncus effuses</i>	< 0.34	< 0.30*	< 3.5	< 9	< 0.68*	356 ± 42	< 1.6
	Moss	< 0.22		< 2.8	< 6		138 ± 24	1.9 ± 0.3
G 3	<i>Holcus lanatus</i>	< 0.22	< 0.30*	4.6 ± 1.0	< 6	< 0.55*	690 ± 54	1 ± 0.2
	<i>Urtica dioica</i>	< 0.20		< 3.0	< 7		588 ± 50	< 1.2
G 4	<i>Holcus lanatus</i>	< 0.27		< 5.5	< 14		292 ± 29	< 2.7
	<i>Juncus effuses</i>	< 0.24		< 3.4	< 7		510 ± 41	< 1.5
	<i>Urtica dioica</i>	< 0.38	< 0.30*	< 6.5	< 13	< 0.8*	1036 ± 90	2.3 ± 0.4
G 5	<i>Holcus lanatus</i>	< 0.35		< 2.0	< 5		245 ± 24	< 1.6
	<i>Juncus effuses</i>	< 0.23		< 2.0	< 5		203 ± 23	< 0.9
G 6	<i>Holcus lanatus</i>	< 0.21		< 4.8	< 11		675 ± 65	< 2.0
	Moss	< 0.32		< 2.9	< 6		96 ± 25	3.2 ± 0.4
G 7	<i>Holcus lanatus</i>	< 0.34	< 0.40*	3.0 ± 0.4	< 7	< 0.67*	857 ± 69	1.5 ± 0.3
G 8	<i>Holcus lanatus</i>	< 0.33		< 2.0	< 8		750 ± 60	< 2.0
G 9	<i>Juncus effuses</i>	< 0.27		< 3.5	< 7		412 ± 40	< 1.5
G 10	<i>Juncus effuses</i>	< 0.44	< 0.35*	< 2.5	< 6	< 0.55*	490 ± 51	< 1.8
G 11	<i>Juncus effuses</i>	< 0.48		< 4.0	< 6		730 ± 57	1.5 ± 0.3
	<i>Crataegus monogyna</i> leaves	< 0.17		5.8 ± 1.0	< 3		603 ± 45	< 0.5
	<i>Crataegus monogyna</i> stems	< 0.16		9.6 ± 1.3	< 11		98 ± 10	< 3.1
	<i>Acer pseudoplatanus</i> leaves	< 0.87		1.2 ± 0.3	< 2		648 ± 47	< 0.3

\*  $^{238}\text{U}$  and  $^{232}\text{Th}$  ( $^{228}\text{Ra}$ ) results from alpha spectrometry, all other  $^{238}\text{U}$  and  $^{232}\text{Th}$  ( $^{228}\text{Ra}$ ) results were obtained using ICP-MS and gamma spectrometry respectively.

Table 34: Radionuclide concentrations in wet grassland vegetation: Site G1 – G11

Caesium-137 in grassland vegetation was only detected in six of the vegetation species sampled as shown in Table 34. The concentration of  $^{137}\text{Cs}$  in *S. moss* at sites G2 and G6 was found to be 1.9 Bq/kg and 3.2 Bq/kg respectively. Caesium-137 levels in *U. diocia* at site G4 was found to be 2.3 Bq/kg and levels in *H. lanatus* at sites G3 and G7 was found to be 1.0 Bq/kg and 1.5 Bq/kg respectively. The concentration of  $^{137}\text{Cs}$  in *J. effuses* at site G11 was found to be 1.5 Bq/kg. Caesium-137 levels in all other vegetation were below the limits of detection for the methods used and in general the levels detectable in vegetation were very low in comparison to levels found in vegetation in both the Atlantic and upland blanket bogs. Caesium-137 levels in grassland vegetation are in general very low due to the nature of the soil where the clay content within the soil binds caesium in an irreversible manner thereby preventing the uptake of it by vegetation.

In general for all grassland vegetation sampled, the average concentration of  $^{40}\text{K}$  in vegetation was statistically higher than the levels of  $^{226}\text{Ra}$  and  $^{137}\text{Cs}$  present (One Way ANOVA,  $p < 0.001$ ) and the concentration of  $^{226}\text{Ra}$  and  $^{137}\text{Cs}$  detectable in the samples were statistically similar (One Way ANOVA,  $p = 1.0$ ).

The concentration of  $^{40}\text{K}$  in vegetation was compared within levels of  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in soil to assess if vegetation uptake was dependent on levels of these radionuclides present in the soil. Potassium-40 levels in all vegetation appear to be independent of the levels of both  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in the soil as shown in Figures 122 and 123 respectively. (Spearman rank correlation coefficient,  $p > 0.05$ ). Similar results are obtained when comparing the  $^{40}\text{K}$  levels within the plant species (i.e. *H. lanatus*, *J. effuses*, *S. moss* and *U. diocia*) with the levels of both these radionuclides in the soil.

Figure 122: Plot of  $^{40}\text{K}$  in vegetation versus  $^{40}\text{K}$  in soil (grassland vegetation)

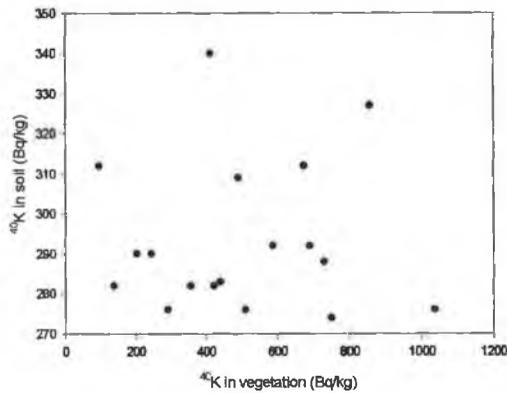
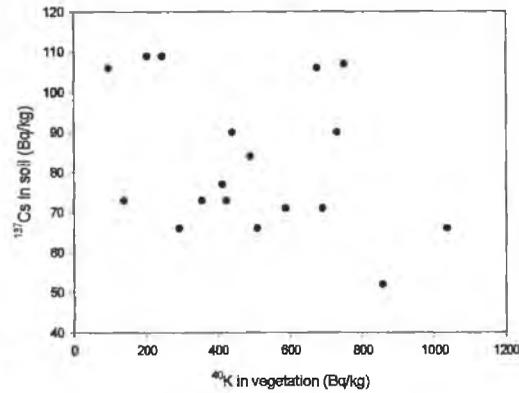


Figure 123: Plot of  $^{40}\text{K}$  in vegetation versus  $^{137}\text{Cs}$  in soil (grassland vegetation)



### 3.3.4 Nutrient content of vegetation

The calcium, magnesium and sodium content of the vegetation were analysed for all vegetation sampled at sites G1 – G11 and results are outlined in Table 35. Correlations between these nutrients and radionuclide transfer factor values for all vegetation are discussed in detail in section 3.3.5.

The concentration of calcium in vegetation ranged from 958 mg/kg in *J. effuses* to 32,766 mg/kg in *U. diocia*. The average concentration of calcium in each species of vegetation was calculated and results indicate that the highest concentration on average was found in *U. diocia* (30448 mg/kg), while the lowest concentration was found in *J. effuses* (1242 mg/kg). Based on the results obtained the levels of calcium in the vegetation decreased in the following order:

$$U. diocia > S. moss > A. pseudoplatanus > C. monogyna > H. lanatus > J. effuses$$

The concentration of calcium in *U. diocia* is significantly higher than levels present in *J. effuses*, *H. lanatus*, *S. moss*, *C. monogyna* and *A. pseudoplatanus* (One Way ANOVA,  $p < 0.001$ ). The concentrations of calcium in all other species of vegetation were statistically similar (One Way ANOVA,  $p > 0.05$ ).

Site	Vegetation	Calcium (mg/kg)	Magnesium (mg/kg)	Sodium (mg/kg)
G 1	<i>Holcus lanatus</i>	2512	2131	1146
G 2	<i>Holcus lanatus</i>	2369	2103	1170
	<i>Juncus effuses</i>	958	1632	2142
	Moss	2930	1541	714
G3	<i>Holcus lanatus</i>	3580	2583	1251
	<i>Urtica dioica</i>	28130	4163	715
G 4	<i>Holcus lanatus</i>	2216	1602	569
	<i>Juncus effuses</i>	1366	2255	2960
	<i>Urtica dioica</i>	32766	5967	1005
G 5	<i>Holcus lanatus</i>	2435	1812	481
	<i>Juncus effuses</i>	1708	1332	1418
G 6	<i>Holcus lanatus</i>	2137	3303	2220
	Moss	4122	1837	405
G 7	<i>Holcus lanatus</i>	2291	1396	1569
G 8	<i>Holcus lanatus</i>	2231	993	1726
G 9	<i>Juncus effuses</i>	988	610	1710
G10	<i>Juncus effuses</i>	1249	841	1702
G 11	<i>Juncus effuses</i>	1186	763	1400
	<i>Crataegus monogyna</i> leaves	4898	3069	1284
	<i>Crataegus monogyna</i> stems	1412	505	777
	<i>Acer pseudoplatanus</i> leaves	3439	2451	502

Table 35: Nutrient content of vegetation at sites G1 – G11

The concentration of magnesium in vegetation ranged from 505 mg/kg in *C. monogyna* (stems) to 5967 mg/kg in *U. dioica*. The average concentration of magnesium in each species of vegetation was calculated and results indicate that the highest concentration on average was found in *U. dioica* (5065 mg/kg), while the lowest concentration was found in *J. effuses* (1238 mg/kg). Based on the results obtained the levels of magnesium in the vegetation decreased in the following order:

*U. dioica* > *A. pseudoplatanus* > *H. lanatus* > *C. monogyna* > *S. moss* > *J. effuses*

The concentration of magnesium in *U. diocia* is significantly higher than levels present in *J. effuses*, *H. lanatus*, *S. moss*, *C. monogyna* (One Way ANOVA,  $p < 0.02$ ) but is statistically similar that levels found in *A. pseudoplatanus* (One Way ANOVA,  $p > 0.05$ ). The concentrations of magnesium found present in all other species of vegetation were statistically similar (One Way ANOVA,  $p > 0.05$ ).

The concentration of sodium in vegetation ranged from 502 mg/kg in *A. pseudoplatanus* to 2960 mg/kg in *J. effuses*. The average concentration of sodium in each species of vegetation was calculated and results indicate that the highest concentration on average was found in *J. effuses* (1889 mg/kg), while the lowest concentration was found in *A. pseudoplatanus* (502 mg/kg). Based on the results obtained the levels of sodium in the vegetation decreased in the following order:

*J. effuses* > *H. lanatus* > *C. monogyna* > *U. diocia* > *S. moss* > *A. pseudoplatanus*

The concentration of sodium found present in all species of vegetation was statistically similar. (One Way ANOVA,  $p > 0.05$ ).

Calcium concentrations were significantly higher in *H. lanatus* in comparison to the sodium levels (One Way ANOVA,  $p = 0.002$ ), while on average the levels of calcium and magnesium present are statistically similar which is also the case when comparing magnesium and sodium levels in this species of grass (One Way ANOVA,  $p > 0.05$ ). The concentration of all three nutrients in *J. effuses* was statistically similar (One Way ANOVA,  $p > 0.05$ ). The concentration of calcium found in *U. diocia* was significantly higher than levels of both magnesium and sodium present (One Way ANOVA,  $p < 0.002$ ), while the levels of magnesium and sodium are statistically similar (One Way ANOVA,  $p = 0.24$ ). Calcium concentrations were significantly higher in *S. moss* in comparison to the sodium levels (One Way ANOVA,  $p = 0.02$ ), while on average the levels of calcium and magnesium present are statistically similar which is also the case when comparing magnesium and sodium levels in this species of grass (One Way ANOVA,  $p > 0.05$ ).

Overall for the plant species studied, the calcium levels were significantly higher in all vegetation sampled in comparison to the sodium levels present (One Way ANOVA,  $p < 0.05$ ). The concentrations of sodium and magnesium in all three plant species were statistically similar as is the case when comparing calcium and magnesium levels present (One Way ANOVA,  $p > 0.05$ ).

### 3.3.5 Transfer Factors for Wet Grassland Vegetation

Transfer factor values for radionuclide uptake by vegetation were calculated based on the activity of radionuclide in vegetation in comparison to the activity in upper 10 cm of soil as outlined in section 2.2. Transfer factor values for  $^{40}\text{K}$  in vegetation are outlined in Table 36, however, transfer factor values could not be calculated for  $^{238}\text{U}$  and  $^{228}\text{Ra}$  as the concentration of these radionuclides in all vegetation were below the limits of detection for the methods outlined in sections 2.5.1 – 2.5.3. Transfer factors values for  $^{226}\text{Ra}$  and  $^{137}\text{Cs}$  could not be calculated for the majority of vegetation sampled as the levels of these radionuclides were below the limits of detection for the methods used in the majority of vegetation sampled. Transfer factor values were not calculated for *S. moss*, *Crataegus monogyna* and *Acer pseudoplatanu*. *S. moss* lacks a well-developed root system and uptake by this vegetation generally occurs by aerial deposition. *Crataegus monogyna* and *Acer pseudoplatanu* are both trees that have extensive rooting systems that extend beyond the sampling depth carried out in this study.

Radium-226 transfer factor values were calculated for *H. lanatus* at two sites sampled as levels of  $^{226}\text{Ra}$  are below the limits of detection in all other vegetation. Transfer factor values for  $^{226}\text{Ra}$  in *H. lanatus* at the two sites were 0.07 and 0.13 respectively. These results are in agreement with the findings for  $^{226}\text{Ra}$  in pasture vegetation of Vera Tomé *et al* (2003), Amaral *et al* (1988), Marple *et al* (1980), Ibrahim and Whicker (1988), Linsalata *et al* (1989) and IUR (1994) who reported TF values of 0.17, 0.07, 0.03 – 0.09, 0.025 – 0.10, 0.016 – 0.4 and 0.08 respectively. In the latter case the IUR recommend a TF value of 0.08 for pasture vegetation growing on grassland soil which is in close

agreement with the findings of this project. The TF values found within this study are higher but not significantly than the findings of Ewers *et al* (2003), Moffett and Tellier (1977), and Makrose *et al* (1993) for  $^{226}\text{Ra}$  in pasture vegetation of 0.03, 0.029 and 0.0066 respectively.

Site	Vegetation	$^{238}\text{U}$ TF	$^{226}\text{Ra}$ TF	$^{228}\text{Ra}$ TF	$^{40}\text{K}$ TF	$^{137}\text{Cs}$ TF
G 1	<i>Holcus lanatus</i>	< 0.01	< 0.14	< 0.03	<b>1.6 ± 0.2</b>	< 0.02
G 2	<i>Holcus lanatus</i>	< 0.01	< 0.12	< 0.44	<b>1.5 ± 0.2</b>	< 0.02
	<i>Juncus effuses</i>	< 0.01	< 0.14	< 0.03	<b>1.3 ± 0.2</b>	< 0.02
G 3	<i>Holcus lanatus</i>	< 0.01	<b>0.13 ± 0.03</b>	< 0.03	<b>2.4 ± 0.4</b>	<b>0.01 ± 0.002</b>
	<i>Urtica dioica</i>	< 0.01	< 0.09	< 0.42	<b>2.0 ± 0.3</b>	< 0.02
G 4	<i>Holcus lanatus</i>	< 0.07	< 0.17	< 0.91	<b>1.1 ± 0.2</b>	< 0.04
	<i>Juncus effuses</i>	< 0.01	< 0.11	< 0.46	<b>1.9 ± 0.3</b>	< 0.02
	<i>Urtica dioica</i>	< 0.01	< 0.20	< 0.05	<b>3.8 ± 0.6</b>	<b>0.03 ± 0.007</b>
G 5	<i>Holcus lanatus</i>	< 0.02	< 0.07	< 0.26	<b>0.8 ± 0.1</b>	< 0.01
	<i>Juncus effuses</i>	< 0.01	< 0.07	< 0.26	<b>0.7 ± 0.1</b>	< 0.01
G 6	<i>Holcus lanatus</i>	< 0.01	< 0.18	< 0.69	<b>2.2 ± 0.3</b>	< 0.02
G 7	<i>Holcus lanatus</i>	< 0.02	<b>0.07 ± 0.01</b>	< 0.04	<b>2.6 ± 0.4</b>	<b>0.03 ± 0.008</b>
G 8	<i>Holcus lanatus</i>	< 0.02	< 0.06	< 0.53	<b>2.7 ± 0.4</b>	< 0.06
G 9	<i>Juncus effuses</i>	< 0.01	< 0.08	< 0.41	<b>1.2 ± 0.2</b>	< 0.02
G 10	<i>Juncus effuses</i>	< 0.01	< 0.05	< 0.03	<b>1.6 ± 0.2</b>	< 0.02
G 11	<i>Juncus effuses</i>	< 0.02	< 0.09	< 0.39	<b>2.5 ± 0.4</b>	<b>0.02 ± 0.005</b>

Table 36: Transfer factor (TF) values for  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  at sites G1 – G11

Potassium-40 transfer factor values in the vascular plants studied ranged from 0.7 to 3.8 (Table 36). The transfer factors values for *H. lanatus*, *J. effuses*, and *U. dioica* were on average 1.86, 1.53 and 2.9 respectively and there is no statistical difference between the average TF values for these species of vegetation. These results indicate that transfer of  $^{40}\text{K}$  to vegetation within this ecosystem is not dependent on plant species. The results for  $^{40}\text{K}$  TF factors values are in broad agreement with the findings of Papastefanou *et al*

(1999), Rafferty *et al* (1994a), Strebl *et al* (2002) and Baeza *et al* (2001) for  $^{40}\text{K}$  in pasture vegetation of 0.16 – 2.42, 2.0 – 6.0, 0.31 – 2.01 and 0.3 respectively.

Transfer factor values for  $^{137}\text{Cs}$  in vascular plants were calculated in four of the vegetation sampled and were below the limits of detection for all other vegetation sampled (Table 36). Transfer factors for  $^{137}\text{Cs}$  in *H. lanatus* were obtained for two samples at sites G3 and G7 of 0.01 and 0.03 respectively. Caesium-137 was detectable in *U. dioica* at site G4, with a TF value of 0.03 and detectable in *J. effuses* at site G11 with a TF of 0.02. The results found within were lower than the results reported by Baeza *et al* (2001) for  $^{137}\text{Cs}$  in the aerial fraction of pasture vegetation of 0.25 and were also lower than the recommended TF value for  $^{137}\text{Cs}$  in pasture vegetation growing on clay-loam of 0.11, reported by IUR (1994). The results are however in agreement with the range of results reported by the following authors: Papastefanou *et al* (1999), Strebl *et al* (2002) and Ehlken and Kirchner (1996) of 0.002 – 7.42, 0.03 – 1.06 and 0.01 – 7.0 respectively. Rafferty *et al* (1994a) studied  $^{137}\text{Cs}$  transfer into pasture vegetation growing in grassland ecosystems in Ireland and reported TF values of < 0.25 which are in agreement with the findings of this study as  $^{137}\text{Cs}$  was below the limit of detection in the majority of vegetation and where detectable the TF values for the of vegetation were < 0.25 (Table 36). Transfer factors for  $^{137}\text{Cs}$  within this ecosystem were very low in comparison to both the Atlantic and upland blanket bog. This is possibly due to the fact that caesium is bound in an irreversible manner within the clay fraction of the loam soil and therefore not available for uptake.

Potassium-40 TF values were compared with the levels of both  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in the soil to assess if uptake of  $^{40}\text{K}$  is dependent on the concentration of either of these radionuclides. A Spearman correlation coefficient of -0.01 ( $p = 0.71$ ) indicates that the transfer of  $^{40}\text{K}$  into all vegetation is independent of the concentration of  $^{40}\text{K}$  in the soil as shown in Figure 124. Similar observation was made between  $^{40}\text{K}$  TF values and  $^{137}\text{Cs}$  in the soil as shown in Figure 125 where a Pearson correlation coefficient of -0.30 ( $p = 0.26$ ) indicates that the TF values are independent of  $^{137}\text{Cs}$  soil concentration.

Figure 124: Plot of  $^{40}\text{K}$  TF values versus  $^{40}\text{K}$  in soil (grassland vegetation)

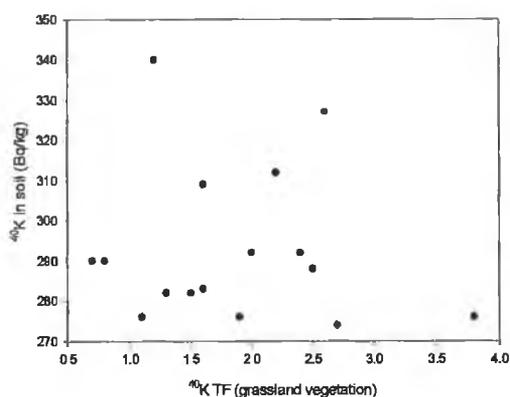
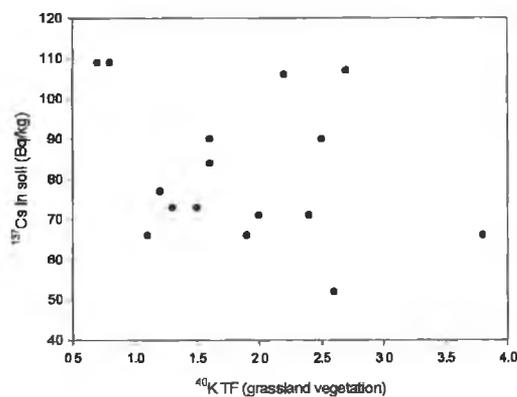


Figure 125: Plot of  $^{40}\text{K}$  TF values versus  $^{137}\text{Cs}$  in soil (grassland vegetation)



### 3.3.6 Correlations between vegetation transfer factors and chemical properties of soil and vegetation

Correlations were carried out between transfer factor values for  $^{40}\text{K}$  and  $^{137}\text{Cs}$  for all vegetation sampled in wet grassland (Table 36) and both the soil physiochemical parameters (Table 32) and the nutrient status of the vegetation (Table 35). The purpose of these correlations was to determine if any of these parameters have a significant impact on the uptake of the aforementioned radionuclides. The TF values are based on the average radionuclide concentration in the upper 10 cm of soil. Therefore it was necessary to correlate these results with the average concentration of each soil property as outlined in Table 37. The full set of results for each individual soil layer (i.e. 0-5 cm, 5-10 cm and 10-20 cm) at all eight sites is presented in Table 32 (Section 3.3.2).

Site	Moisture %	pH	Organic Matter %	Available Magnesium mg/kg	Available Sodium mg/kg	Available Calcium mg/kg	Available Potassium mg/kg	CEC of soil meq/100g
G 1	38.7	5.0	13.5	127	47	352	132	41
G 2	49.8	5.1	14.1	164	46	614	257	49
G 3	52.1	5.3	13.7	184	58	828	221	49
G 4	47.9	5.7	14.3	219	51	1474	534	49
G 5	37.2	5.3	12.9	157	51	595	122	45
G 6	40.4	5.0	14.5	136	56	251	170	49
G 7	43.4	5.3	11.7	175	68	667	167	39
G 8	52.2	5.2	16.1	160	60	591	111	48
G 9	49.3	5.3	11.2	188	60	534	293	47
G10	57.7	5.1	13.8	163	58	765	170	47
G11	50.8	5.5	13.4	194	63	946	210	41

Table 37: Physiochemical properties of soil surface horizon at sites G1 – G11  
(Average value for 0-10 cm)

Correlations were carried out initially for  $^{40}\text{K}$  and  $^{137}\text{Cs}$  with the soil properties and nutrient status of vegetation for all vegetation species sampled to assess if there is an overall effect of any particular parameter affecting uptake within this ecosystem. In the case of  $^{40}\text{K}$ , correlations were then carried on the individual plant species for each radionuclide. A summary of the results from the correlation between the radionuclides and the aforementioned parameters are tabulated in Table 38 and any significant findings are shown in graphical format in Figures 126 – 129 respectively.

Soil/Vegetation Properties	<sup>40</sup> K TF (All vegetation)	<sup>137</sup> Cs TF (All vegetation)*	<sup>40</sup> K TF ( <i>H. lanatus</i> )	<sup>40</sup> K TF ( <i>J. effuses</i> )
<sup>238</sup> U (Bq/kg)	X	X	X	X
<sup>226</sup> Ra (Bq/kg)	X	X	X	X
<sup>228</sup> Ra (Bq/kg)	X	X	X	X
<sup>40</sup> K (Bq/kg)	X	X	X	X
<sup>137</sup> Cs (Bq/kg)	X	X	X	X
Moisture Content (%)	X	X	X	X
pH	X	X	X	X
Organic Matter (%)	X	X	X	X
Avail. Mg (mg/kg)	X	X	X	X
Avail. Na (mg/kg)	↑ <b>r<sup>2</sup> = 0.50</b> <b>p = 0.04</b>	X	↑ <b>r<sup>2</sup> = 0.77</b> <b>p = 0.02</b>	X
Avail. Ca (mg/kg)	X	X	X	X
Avail. K (mg/kg)	X	X	X	X
CEC	X	X	X	X
<b>Vegetation Nutrients</b>				
Calcium (mg/kg)	X	X	X	X
Magnesium (mg/kg)	↑ <b>r<sup>2</sup> = 0.54</b> <b>p = 0.03</b>	X	X	X
Sodium (mg/kg)	X	X	↑ <b>r<sup>2</sup> = 0.82</b> <b>p = 0.01</b>	X
X: no significant correlation present between the two variables.				
* Correlation carried out on vegetation which contained detectable concentrations of <sup>137</sup> Cs.				

Table 38: Summary of correlations between TF values and soil/vegetation properties.

Correlations between <sup>40</sup>K TF values and soil physiochemical properties indicate that <sup>40</sup>K uptake in vegetation is significantly affected by the available sodium levels in the soil. A Spearman correlation coefficient of 0.50 (p = 0.04) indicates a linear relationship between the transfer of <sup>40</sup>K from soil to wet grassland vegetation and the available sodium levels in the soil as shown in Figure 126. A Spearman correlation coefficient of 0.54 (p = 0.03) indicates a significant relationship between the uptake of <sup>40</sup>K by all vegetation and magnesium content of vegetation as shown in Figure 127 however no significant relationships appears to be present between <sup>40</sup>K TF values and either the

calcium or sodium content of the vegetation.

Figure 126: Plot of  $^{40}\text{K}$  TF values versus Avail Na in soil (grassland vegetation)

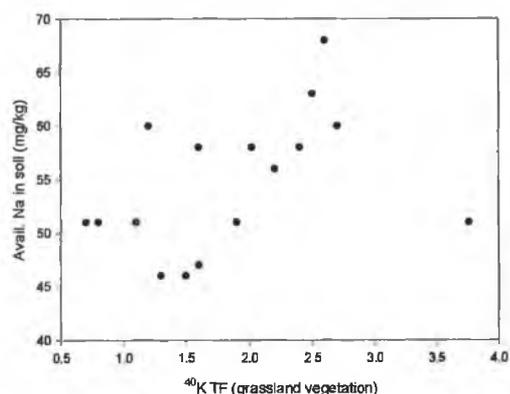
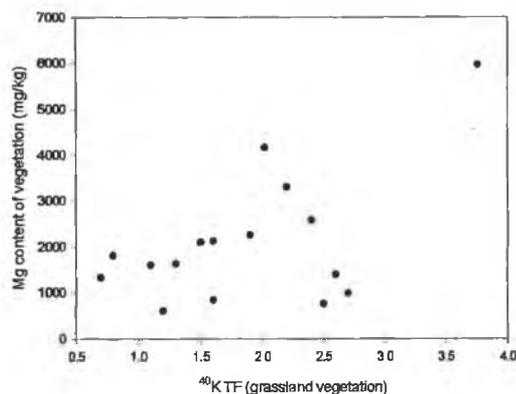


Figure 127: Plot of  $^{40}\text{K}$  TF versus Mg content of vegetation (grassland vegetation)



Correlation for  $^{40}\text{K}$  TF values was then carried out for *H. lanatus* and *J. effuses* separately (*U. dioica* data set too small) to ascertain if any of the soil properties or vegetation nutrient status affect uptake of the radionuclide based on the individual plant species. Potassium-40 TF values for *H. lanatus* showed little correlation the soil physiochemical properties with the exception of the available sodium levels in the soil. A Spearman correlation coefficient of 0.77 ( $p = 0.02$ ) indicates a significant positive correlation between the uptake of  $^{40}\text{K}$  by this vegetation and the levels of available sodium in the soil as shown in Figure 128. A similar observation was made for the uptake of  $^{40}\text{K}$  by all vegetation and available sodium in the soil (Figure 126) however in the case of *H. lanatus* the correlation appears to be much stronger. The uptake of  $^{40}\text{K}$  by *H. lanatus* with regard to TF values also appears to be significantly impacted by the levels of total sodium in the vegetation as shown in Figure 129. A Spearman correlation coefficient of 0.82 ( $p = 0.01$ ) indicates a strong positive between the  $^{40}\text{K}$  TF values for *H. lanatus* and the concentration of sodium within the vegetation.

Figure 128: Plot of  $^{40}\text{K}$  TF versus Avail. Na in soil (*H. lanatus*)

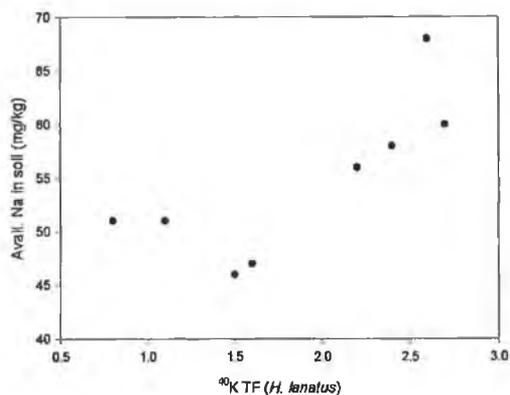
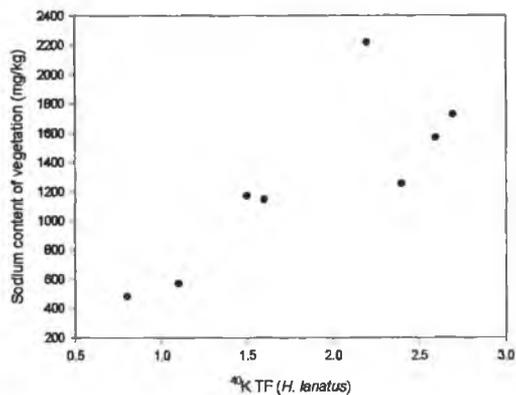


Figure 129: Plot of  $^{40}\text{K}$  TF versus Na content of vegetation (*H. lanatus*)



Potassium-40 uptake by *J. effuses* appears to be independent of both the soil properties studied and nutrient status of vegetation.

Caesium-137 TF values for wet grassland vegetation appears to be independent of both the soil properties studied and nutrient status of vegetation as shown in Table 38.

### 3.3.7 Summary for Wet Grassland

The results for this ecosystem indicate that uptake of  $^{226}\text{Ra}$  and  $^{137}\text{Cs}$  by vegetation were significantly lower than for  $^{40}\text{K}$  and levels of both  $^{238}\text{U}$  and  $^{228}\text{Ra}$  in vegetation were below the limits of detection for the methods used. Radium-266 was detectable in *H. lanatus*, *C. monogyna* and *A. pseudoplatanus* but was below the limits of detection for *J. effuses* and *U. diocia*. In the case of *H. lanatus*  $^{226}\text{Ra}$  was only detectable in two of the eight samples collected and TF values were 0.07 and 0.13 respectively.

Potassium-40 TF values in vascular plants ranged from 0.7 to 3.8 and on average the TF values for the three types of vegetation, *H. lanatus*, *J. effuses* and *U. diocia* were all statistically similar. These results indicate that transfer of  $^{40}\text{K}$  into vegetation within this ecosystem is not dependent on the species of vegetation. Potassium-40 TF values for all vegetation grouped together appears to be positively correlated with the levels of

available sodium in the soil and a similar but much stronger correlation was present when comparing the available levels of sodium in the soil with  $^{40}\text{K}$  TF values for *H. lanatus*. The uptake of  $^{40}\text{K}$  by *H. lanatus* in relation to its TF values was also found to be positively correlated with the levels of sodium within the vegetation. The uptake of  $^{40}\text{K}$  by *J. effuses* appears to be independent of both the soil properties and nutrient status of the vegetation. The uptake of  $^{40}\text{K}$  by all vegetation was also found to be positively influenced by the levels of magnesium in the vegetation. This correlation was evident between  $^{137}\text{Cs}$  TF values and magnesium in vegetation in the two other ecosystem studied.

Caesium-137 levels in the majority of vegetation were below the limits of detection which may be linked to the nature of the soil within this ecosystem. Transfer factors values were calculated in four of the vegetation sampled (two *H. lanatus*, one *J. effuses* and one *U. diocia*) and were in general very low ranging from 0.01 to 0.03. Transfer factor values for  $^{137}\text{Cs}$  in this vegetation appear to indicate that its uptake is not dependent on plant species, soil physiochemical properties or nutrient status of vegetation.

### 3.4 Comparison of Ecosystems

The primary objective of this study was to assess the uptake of natural and anthropogenic radionuclides by vegetation within three natural ecosystems of Ireland. The three ecosystems were an Atlantic blanket bog, upland blanket bog and wet grassland and were chosen based on the fact that they were either natural or semi-natural ecosystems in Ireland and they cover a significant proportion of the landmass of Ireland. The chosen ecosystems differ in soil radionuclide concentrations, soil physiochemical properties, vegetation species and nutrient status of vegetation. An in-depth discussion (section 3.1 – 3.3) has been carried out on radionuclide TF values within each of the individual ecosystems to ascertain what impacts the above properties have on the uptake of the radionuclides by vegetation within these ecosystems. The remainder of this discussion will focus on the comparison between the three ecosystems in order to ascertain if there are any impacting factors affecting radionuclides common to all three ecosystems and also to highlight vegetation which appear to accumulate radionuclides to a greater extent than any other vegetation sampled.

The scope of this project focuses mainly on the fate of these radionuclides within the chosen ecosystems as radiological protection internationally has shifted from the protection of humans to the protection of the environment as a whole. This is based on the recommendations from the International Commission on Radiological Protection (ICRP) that the levels outlined in current radiation protection standards which are based on the protection of humans may be too high for other natural organisms and vegetation present in these ecosystems. It therefore has become important to investigate the impact of these radionuclides within natural ecosystems. While the focus of this project is on the vegetation within these ecosystems, some consideration needs to be given to the potential transfer of these radionuclides into the human food-chain from animal grazing. The radionuclide levels in the vegetation from this study will be compared with the limits specified in Title 10, Code of Federal Regulations (U.S. Nuclear Regulatory Commission) to assess the potential risk to humans. This document outlines the

requirements on all persons and organizations who receive a license from U. S. Nuclear Regulatory Commission (NRC) to use nuclear materials or operate nuclear facilities. The limits are based on occupational exposure to radioactive material and are categorised based on mode of entry into the body. The category of interest for this study is the annual limit of intake (ALI) of radioactive material by oral ingestion. These ALIs are based on the annual intake of a given radionuclide by “Reference Man” which would result in either (1) a committed effective dose equivalent of 5 rems (stochastic ALI) or (2) a committed dose equivalent of 50 rems to an organ or tissue (non- stochastic ALI). It is important to remember though that there is no safe level of exposure to ionising radiation.

### 3.4.1 Soil Radionuclide Concentrations

Radionuclide ( $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$ ) concentrations in the soil from the three ecosystems were compared to ascertain which soil contained the highest concentration of each radionuclide. Uranium-238 levels in the Atlantic blanket bog soil were below the limits of detection for the methods used in all soils sampled. The average concentration of  $^{238}\text{U}$  in the soil at the upland blanket bog and the grassland were 19.4 Bq/kg and 21.2 Bq/kg and statistics show that there is no significant difference between the average concentrations at the two sites (t-test,  $p = 0.50$ ).

Radium-226 levels were also below the limits of detection for methods used in all Atlantic blanket bog soils sampled. The average levels in the upland blanket bog and grassland were 12.8 Bq/kg and 35.1 Bq/kg and results indicate that the average concentration of this radionuclide in the grassland soil is significantly higher than the upland blanket bog soil (t-test,  $p < 0.001$ ). Radium-228 levels were also below the limits of detection for methods used in all Atlantic blanket bog soils sampled. The average levels in the upland blanket bog and grassland were 5.8 Bq/kg and 17.3 Bq/kg and like  $^{226}\text{Ra}$ , results indicate that the average concentration of this radionuclide in the grassland soil is significantly higher than the upland blanket bog soil (t-test,  $p < 0.001$ ).

Soil Property	Ecosystem		Comments
<sup>238</sup> U (Bq/kg)	Grassland > Upland bog (21.2)      (19.4)		Atlantic blanket bog soil below limits of detection. Grassland and upland bog statistically similar
<sup>226</sup> Ra (Bq/kg)	Grassland > Upland bog (35.1)      (12.8)		Atlantic blanket bog soil below limits of detection. Grassland and upland bog significantly different.
<sup>228</sup> Ra (Bq/kg)	Grassland > Upland bog (17.3)      (5.8)		Atlantic blanket bog soil below limits of detection. Grassland and upland bog significantly different.
<sup>40</sup> K (Bq/kg)	Grassland > Upland bog > Atlantic bog (306.2)      (43.2)      (25.7)		
<sup>137</sup> Cs (Bq/kg)	Upland bog > Atlantic bog > Grassland (169.6)      (137.8)      (52.6)		
			Grassland soil levels significantly higher than both bog soils. Upland blanket bog and Atlantic blanket bog statistically similar
			Grassland soil levels significantly lower than both bog soils. Upland blanket bog and Atlantic blanket bog statistically similar

Table 39: Comparison of soil radionuclide concentrations for three ecosystems

Potassium-40 and <sup>137</sup>Cs levels were detectable in all soils sampled from the three ecosystems. The average levels of <sup>40</sup>K found in the soils of the Atlantic blanket bog, upland blanket bog and grassland were 25.7 Bq/kg, 43.2 Bq/kg and 306.2 Bq/kg respectively. The average concentration of <sup>40</sup>K in the grassland soil was significantly higher than in both the Atlantic blanket bog and upland blanket bog soil (One Way ANOVA,  $p < 0.001$ ). The concentration of <sup>40</sup>K in the soil in the two bog ecosystems were statistically similar (One Way ANOVA,  $p = 0.25$ ).

The average concentration of <sup>137</sup>Cs in the soils of the Atlantic blanket bog, upland blanket bog and grassland were 137.8 Bq/kg, 169.6 Bq/kg and 52.6 Bq/kg respectively. The average concentration of <sup>137</sup>Cs in the soil in the Atlantic blanket and upland blanket bogs were both higher than levels found in the wet grassland (One Way ANOVA,  $p < 0.001$ ).

Average levels of  $^{137}\text{Cs}$  in the two bog ecosystems were statistically similar (One Way ANOVA,  $p = 0.09$ )

### **3.4.2 Soil physiochemical properties**

Comparisons were carried out between the soil physiochemical properties for the three ecosystems in order to identify which ecosystem is nutrient deficient and to compare the differences between the soils.

The average moisture content for the soil in the three ecosystems, Atlantic blanket bog, upland blanket bog and grassland was 88.7 %, 90.4 % and 37.7 % respectively. The highest values were found in the two blanket bog soils and the average values for these two soils were statistically similar (One Way ANOVA,  $p = 0.64$ ). The moisture content of the grassland soil was significantly lower than the two bogland soils as would be expected as bogs are characteristically wet acidic soils (One Way ANOVA,  $p < 0.001$ ).

The pH of the grassland soil (average value of 5.2) was significantly higher than the pH of the soil in both the Atlantic blanket and upland blanket bog soil of 3.8 and 4.4 respectively (One Way ANOVA,  $p < 0.001$ ). The pH of the upland blanket bog soil was also significantly higher than the Atlantic blanket bog soil (One Way ANOVA,  $p < 0.001$ ).

The average organic matter content of the soil in the three ecosystems, Atlantic blanket bog, upland blanket bog and grassland was 95.7 %, 93.1 % and 9.4 % respectively. The highest values were found as shown in the two blanket bog soils and average values were statistically similar (One Way ANOVA,  $p = 0.051$ ). The content of organic matter was significantly lower in fact at least ten times lower in the grassland soil in comparison to the two other ecosystems (One Way ANOVA,  $p < 0.001$ ).

Soil Property	Ecosystem	Comments
Moisture Content (%)	Upland bog > Atlantic bog > Grassland (90.4) (88.7) (37.7)	Upland blanket bog and Atlantic blanket bog statistically similar. Grassland soil levels significantly lower than both bog soils.
pH	Grassland > Upland bog > Atlantic bog (5.2) (4.4) (3.8)	All significantly different.
Organic Matter (%)	Atlantic bog > Upland bog > Grassland (95.7) (93.1) (9.4)	Upland blanket bog and Atlantic blanket bog statistically similar. Grassland soil levels significantly lower than both bog soils.
Available Magnesium (mg/kg)	Atlantic bog > Upland bog > Grassland (470) (356) (135)	All significantly different.
Available Sodium (mg/kg)	Atlantic bog > Upland bog > Grassland (232) (177) (48)	All significantly different.
Available Calcium (mg/kg)	Grassland > Atlantic bog > Upland bog (566) (372) (328)	Grassland soil levels significantly higher than both bog soils. Upland blanket bog and Atlantic blanket bog statistically similar
Available Potassium (mg/kg)	Grassland > Upland bog > Atlantic bog (157) (132) (118)	All statistically similar
CEC (meq/100g)	Atlantic bog > Upland bog > Grassland (147) (133) (42)	All significantly different

Table 40: Comparison of soil physiochemical properties for three ecosystems

The concentrations of four plant available nutrients in the soil were compared for the three ecosystems and results are outlined in Table 40. The concentration of available magnesium and sodium were significantly higher in the Atlantic blanket bog soil in comparison to the two other ecosystems and also significantly higher in the upland blanket bog soil in comparison to the grassland (One Way ANOVA,  $p < 0.001$ ). Levels of plant available calcium in the grassland soil was significantly higher than in the soil of both bog ecosystems (One Way ANOVA,  $p < 0.001$ ), but the concentrations in the soil of both bog ecosystems was statistically similar (One Way ANOVA,  $p = 0.762$ ). Levels of available potassium in the soil was found to be highest in the grassland soil and lowest in

the Atlantic blanket bog soil but the concentration in the soil of all three ecosystems was statistically similar (One Way ANOVA,  $p = 0.134$ ). The CEC of the soil in both bog ecosystems was significantly higher than the soil in the grassland ecosystem (One Way ANOVA,  $p < 0.001$ ) and the CEC of the soil in the Atlantic blanket bog was also significantly higher than the soil in the upland blanket bog (One Way ANOVA,  $p = 0.002$ ). In general the nutrient status of the peat soil was significantly higher or similar to the grassland soil with the exception of the calcium content. The ability of both peat soils to exchange cations was also higher than the grassland soil indicating that this may have a positive impact on plant uptake of nutrients and radionuclides.

### 3.4.3 Nutrient status of vegetation

The nutrient content (calcium, magnesium and sodium) of all vegetation from the three ecosystems were compared to assess which vegetation contained the highest concentrations.

The calcium content of *U. diocia* from the wet grassland was significantly higher than all other vegetation sampled in all three ecosystems (One Way ANOVA,  $p < 0.001$ ). The calcium content of *S. moss* from Atlantic blanket bog was also significantly higher than the concentrations found in *E. vaginatum* and *C. vulgaris* (leaves and stems) from the same ecosystem (One Way ANOVA,  $p < 0.011$ ). The calcium content of *S. moss* from Atlantic blanket bog was also significantly higher than the levels found in the following upland blanket bog vegetation: *M. caerulea*, *E. tetralix*, *E. vaginatum*, *M. gale* (leaves and stems) and *C. vulgaris* (leaves and stems) and *S. moss* (One Way ANOVA,  $p < 0.001$ ).

The calcium content of *S. moss* from Atlantic blanket bog was also significantly higher than the levels found in the following grassland vegetation: *H. lanatus*, *J. effuses*, *S. moss*, *A. pseudoplatanus* and *C. monogyna* (One Way ANOVA,  $p < 0.025$ ).

The calcium content of the leaves of *C. vulgaris* from Atlantic blanket bog was also significantly higher than the levels found in *J. effuses*, *M. caerulea*, and the stems of *C. vulgaris* from both bog ecosystems (One Way ANOVA,  $p < 0.018$ ).

The calcium content of *S. moss* from upland blanket bog was also significantly higher than the levels found in *J. effuses*, *M. caerulea*, and the stems of *C. vulgaris* from both bog ecosystems (One Way ANOVA,  $p < 0.006$ ). The calcium content in all other vegetation was statistically similar.

The magnesium content of *U. dioica* from the wet grassland was significantly higher than all other vegetation sampled in all three ecosystems (One Way ANOVA,  $p < 0.001$ ) while the levels present in all other vegetation was statistically similar.

The sodium content of *J. effuses* from the grassland ecosystem was significantly higher than the levels present in *S. moss*, and the stems of both *M. gale* and *C. vulgaris* from upland blanket bog (One Way ANOVA,  $p < 0.001$ ). The levels present in all other vegetation are statistically similar.

#### 3.4.4 Transfer Factors and Correlations

Transfer factor values for all vegetation from the three ecosystems were compared to assess if species of vegetation had a significant impact on the uptake of the particular radionuclides. Transfer factors values were not calculated for *S. moss*, *A. pseudoplatanus* and *C. monogyna* as in the former case the vegetation lacks a well developed root system and the latter two cases the sampling depth in this ecosystem would not have been sufficient to cover the roots of these trees. Radionuclide TF values were then compared with the soil physiochemical properties and nutrient status of vegetation to ascertain if any of these properties have a significant impact on the uptake of the selected radionuclides. The finding from the individual ecosystems have been discussed in detailed within the previous sections (3.1 – 3.3) and the remainder of this discussion will

focus on identifying soil and vegetation properties which affect radionuclide uptake that are common to all three ecosystems and also to identify species of vegetation which have the ability to accumulate radionuclides to a greater extent than all other vegetation.

The levels of both  $^{238}\text{U}$  and  $^{228}\text{Ra}$  ( $^{232}\text{Th}$ ) were detectable in the soil from the upland blanket bog and grassland but they below the limits of detection in the soil for the Atlantic blanket bog and for all vegetation from the three ecosystems therefore no actual transfer factors could be calculated for these radioisotopes. This would indicate that there is little concern with regard to transfer of these radionuclides into the vegetation or potential transfer into the human food chain through animal grazing. This assumption is supported by comparing the levels found in this study with the threshold levels for occupational annual limits of intake (oral ingestion) outlined by U.S. NRC of 370 kBq and 74 kBq for  $^{238}\text{U}$  and  $^{228}\text{Ra}$  respectively.

Radium-226 TF values were calculated for all vegetation in the upland blanket bog and for *H. lanatus* (two samples) in the wet grassland. It was not possible to calculate TF values for  $^{226}\text{Ra}$  in the Atlantic blanket bog as the concentration of  $^{226}\text{Ra}$  was below the limits of detection in both the soil and vegetation of this ecosystem.

Comparisons of  $^{226}\text{Ra}$  TF values indicate that TF values for *E. tetralix* were significantly higher than all other vegetation in the upland blanket bog and also significantly higher than values for *H. lanatus* from the wet grassland (One Way ANOVA,  $p < 0.048$ ).

**$^{226}\text{Ra}$  TF values *E. tetralix* >  $^{226}\text{Ra}$  TF values for all vegetation**

Radium-226 TF values in all other vegetation were statistically similar thereby indicating that *E. tetralix* appears to have the ability to accumulate  $^{226}\text{Ra}$  to greater extent than all other vegetation analysed. It is important to consider the potential transfer of  $^{226}\text{Ra}$  into the human food through animal grazing as sheep are commonly found grazing on heather species within this type of ecosystem. However, the levels found in this study ( $< 0.03$  kBq for all vegetation) do not warrant significant concern to humans when compared

with the occupational annual limit of intake for oral ingestion specified by U.S. NRC of 74 kBq. However, further work should concern itself with assessing the uptake of  $^{226}\text{Ra}$  in regions of elevated natural radioactivity. The uptake of  $^{226}\text{Ra}$  by this species of vegetation in other ecosystems also warrants further investigation in order to ascertain if this vegetation has the ability to bioaccumulate  $^{226}\text{Ra}$  while growing on soils with varying physiochemical properties.

Radium-226 TF values were found to be positively impacted by the levels of organic matter, available calcium in soil, available magnesium and the CEC of the soil. The uptake of  $^{226}\text{Ra}$  appears to be diminished in the presence of high levels of available potassium in the soil and calcium within the vegetation. Uptake of  $^{226}\text{Ra}$  within the upland blanket bog was diminished in vegetation growing on soils with high pH. This appears also to be the case when comparing  $^{226}\text{Ra}$  TF values for the upland blanket bog and grassland ecosystem as the grassland soil has a significantly higher pH and  $^{226}\text{Ra}$  was only detectable in two vegetation samples from this ecosystem; however the fact that the vegetation species within these two ecosystems are very different cannot be ignored. The moisture content of the soil appears to have a conflicting impact on the uptake of  $^{226}\text{Ra}$  by vegetation within the upland blanket bog (Table 41) and this would appear to indicate that the impact of soil moisture on  $^{226}\text{Ra}$  uptake is species dependent.

Potassium-40 TF values were calculated for all vegetation in the three ecosystems and comparisons between these TF values indicate that transfer of  $^{40}\text{K}$  into *E. vaginatum* from Atlantic blanket bog was significantly higher than all other vegetation analysed from these ecosystems (One Way ANOVA,  $p < 0.003$ ).

**$^{40}\text{K}$  TF value *E. vaginatum* (Atlantic blanket bog) >  $^{40}\text{K}$  TF values for all vegetation**

Transfer factor values for  $^{40}\text{K}$  in all other vegetation from the three ecosystems were found to be statistically similar therefore indicating that *E. vaginatum* has the ability to accumulate  $^{40}\text{K}$  to a greater extent than any other vegetation sampled. However *E. vaginatum* was also sampled in the upland blanket bog and  $^{40}\text{K}$  TF values for these

samples were significantly lower than its counterpart in the Atlantic blanket bog and statistically similar to all other vegetation indicating that while *E. vaginatum* may demonstrate the ability to accumulate  $^{40}\text{K}$  over all other vegetation within the Atlantic blanket bog sampled it does not necessarily behave in this manner in all ecosystems. The annual limit of intake by oral ingestion for  $^{40}\text{K}$  specified by U.S. NRC is 11,100 kBq, therefore the levels found within this study (< 1 kBq for all vegetation) do not possess a substantial risk in relation to human contamination through the food-chain.

TF value	Ecosystem	Positive Correlation	Negative Correlation
$^{226}\text{Ra}$ TF	Upland Bog	Soil Organic Matter Soil Moisture Content Avail. Ca in soil Avail. Mg in soil Soil CEC	Soil Moisture Content Soil pH Avail. K in soil Ca in vegetation
$^{40}\text{K}$ TF	Atlantic Bog	Soil CEC	None
	Upland Bog	Soil Organic Matter Soil Moisture Content Soil pH Avail. Ca in soil Avail. Mg in soil Soil CEC Ca in vegetation Mg in vegetation	$^{40}\text{K}$ in soil Avail. K in soil Ca in vegetation
	Grassland	Avail. Na in soil Mg in vegetation Na in vegetation	None
$^{137}\text{Cs}$ TF	Atlantic Bog	Ca in vegetation Mg in vegetation	None
	Upland Bog	Ca in vegetation Mg in vegetation	$^{137}\text{Cs}$ in soil Avail. Na in soil Ca in vegetation
	Grassland	None	None

Table 41: Correlations between TF values and physiochemical properties of soils and vegetation for all three ecosystems.

Potassium-40 TF values for vegetation within the two bog ecosystems were found to be positively impacted by soils with high cation capacity and in the case of the upland blanket bog TF values increased in vegetation growing on soils with high levels of calcium and magnesium present. The pH of the soil, levels of organic matter, moisture content,  $^{40}\text{K}$  and available potassium in the soil appear to have impacted the uptake of  $^{40}\text{K}$  by upland blanket bog vegetation but do not appear to significantly impact the uptake of  $^{40}\text{K}$  within the other two ecosystems. These parameters may be specific to this type of ecosystem or the vegetation within it.

The levels of magnesium in the vegetation from the upland blanket appear to have a positive affect on the uptake of  $^{40}\text{K}$  by the vegetation. A similar correlation were evident between  $^{40}\text{K}$  uptake in grassland vegetation and the levels of this nutrient present however this was not evident in the Atlantic blanket bog ecosystem. The effect of calcium within the vegetation on  $^{40}\text{K}$  is less clear as conflicting correlations were evident within the upland blanket bog vegetation indicating that the impact of this nutrient on the uptake of  $^{40}\text{K}$  is highly dependent on the species of vegetation. The levels of both available sodium in the soil and sodium in the vegetation appear to have a positive impact on the uptake of  $^{40}\text{K}$  by grassland vegetation however these correlations do not appear to be present for either the Atlantic blanket or upland blanket bog vegetation.

Caesium-137 TF values were calculated for all vegetation in both the Atlantic blanket and upland blanket bog but was only calculated for a limited number of vegetation samples within the wet grassland ( $\frac{1}{2}$  the *U. dioica* samples,  $\frac{1}{4}$  of the *H. lanatus* and  $\frac{1}{5}$  of the *J. effuses* samples) as levels of  $^{137}\text{Cs}$  in the other vegetation samples were below the limits of detection for the methods used.

Comparisons between TF values indicate that  $^{137}\text{Cs}$  transfer into the leaves of *C. vulgaris* from the Atlantic blanket bog was significantly higher than all other vegetation sampled in the three ecosystems (One Way ANOVA,  $p < 0.001$ ).

**$^{137}\text{Cs}$  TF value *C. vulgaris* leaves (Atl. blan. bog) >  $^{137}\text{Cs}$  TF values for all vegetation**

Similar results were also evident for  $^{137}\text{Cs}$  uptake by *C. vulgaris* in the upland blanket bog (UBB). Caesium-137 TF values were significantly higher in the leaves of *C. vulgaris* from the upland blanket bog than for all other vegetation from the same ecosystem (One Way ANOVA,  $p < 0.017$ ). Transfer factor values were also significantly higher in the leaves of *C. vulgaris* in comparison to the stems indicating that  $^{137}\text{Cs}$  uptake in this plant species is dependent on plant compartment (One Way ANOVA,  $p = 0.037$ ).

$^{137}\text{Cs}$  TF value *C. vulgaris* leaves (UBB) >  $^{137}\text{Cs}$  TF value *C. vulgaris* stems (UBB)

$^{137}\text{Cs}$  TF value *C. vulgaris* leaves (UBB) >  $^{137}\text{Cs}$  TF value *M. gale* (UBB)

$^{137}\text{Cs}$  TF value *C. vulgaris* leaves (UBB) >  $^{137}\text{Cs}$  TF value *E. tetralix* (UBB)

$^{137}\text{Cs}$  TF value *C. vulgaris* leaves (UBB) >  $^{137}\text{Cs}$  TF value *M. caerulea* (UBB)

$^{137}\text{Cs}$  TF value *C. vulgaris* leaves (UBB) >  $^{137}\text{Cs}$  TF value *E. vaginatum* (UBB)

These results appear to indicate that *C. vulgaris* or more importantly the leaves of this vegetation demonstrate the ability to accumulate  $^{137}\text{Cs}$  to a greater extent than the majority of vegetation assessed. The effect appears to be more pronounced within the Atlantic blanket bog ecosystem as results indicate that  $^{137}\text{Cs}$  uptake within the leaves of *C. vulgaris* within this ecosystem was significantly higher than all other vegetation but was also evident to a lesser extent in the leaves of the same vegetation within the upland blanket bog. This species of vegetation has in previous studies been shown to significantly accumulate  $^{137}\text{Cs}$  and is of concern due to the potential transfer of this radionuclide into the human food chain from grazing sheep. This potential transfer into sheep led to a ban on the consumption of sheep found grazing in contaminated areas of the United Kingdom following the Chernobyl accident in 1986. The fact that the leaves of this vegetation hold significantly higher levels than the stems also increases the potential transfer into the human food chain. However, levels found within the vegetation from this study ( $< 1$  kBq) are low in comparison to the threshold values specified by U.S. NRC for annual limit of intake (oral ingestion) of 3,700 kBq.

The Atlantic blanket bog vegetation appears to demonstrate a greater ability to accumulate  $^{137}\text{Cs}$  in comparison to their counterparts within the upland blanket as shown

in *C. vulgaris* and also in the case of *E. vaginatum*. Caesium-137 TF values for *E. vaginatum* in the Atlantic blanket bog was significantly higher than TF values for *M. caerulea* (One Way ANOVA,  $p < 0.014$ ).

**$^{137}\text{Cs}$  TF value *E. vaginatum* (Atl. blan. bog) >  $^{137}\text{Cs}$  TF value *M. caerulea***

However this does not appear to be specific for this species of vegetation as TF values for *E. vaginatum* from upland blanket bog were statistically similar to all other vegetation.

The uptake of  $^{137}\text{Cs}$  within the vegetation from the Atlantic blanket appears to be positively impacted by the levels of magnesium present in the vegetation and a similar observation was apparent for the upland blanket bog vegetation. This correlation does not appear to be present in the grassland ecosystem however uptake of  $^{137}\text{Cs}$  was very low in this vegetation and the data set available is limited. The impact of calcium in vegetation on  $^{137}\text{Cs}$  TF values is less clear as it appears initially to have a positive impact on uptake of  $^{137}\text{Cs}$  in vegetation from both ecosystems but in the case of the upland blanket bog the results are contradictory leading to the conclusion that the effect of vegetation calcium levels on  $^{137}\text{Cs}$  uptake is species dependent.

The impact of magnesium and calcium levels in vegetation on  $^{40}\text{K}$  and  $^{137}\text{Cs}$  are very similar. Magnesium levels in vegetation was shown to have a positive impact on uptake of both radionuclides however the effect of calcium levels in vegetation on  $^{40}\text{K}$  and  $^{137}\text{Cs}$  uptake is contradictory leading to the conclusion in both cases that the impact of calcium levels on uptake of these radionuclides is dependent on the species of vegetation.

The uptake of  $^{137}\text{Cs}$  by the upland blanket bog was found to be negatively impacted by the levels  $^{137}\text{Cs}$  and available sodium in the soil, however, these correlations only appear to be evident for the vegetation from this ecosystem as the uptake of  $^{137}\text{Cs}$  by the vegetation from the two other ecosystems does not appear to be affected by these parameters.

In summary, uptake of  $^{238}\text{U}$  and  $^{228}\text{Ra}$  ( $^{232}\text{Th}$ ) was very low in all vegetation and was below the limits of detection in all vegetation studied. Uptake of  $^{226}\text{Ra}$  was very low in some vegetation and was below the limits of detection for the Atlantic blanket bog vegetation and the majority of grassland vegetation. Radium-226 was detectable in upland blanket bog and some grassland vegetation and TF values for all vegetation were statistically similar with the exception of *E. tetralix* from the upland blanket bog. *E. tetralix* is a species of heather found within boglands of Ireland and in particular the upland blanket bogs where it was evident in this study and  $^{226}\text{Ra}$  TF values for this species were statistically higher than all other species of vegetation sampled indicating that this heather may have the ability to accumulate  $^{226}\text{Ra}$ . Potassium-40 TF values were calculated for all vegetation in the three ecosystems and were significantly higher for *E. vaginatum* from the Atlantic blanket bog than all other vegetation however TF values for this vegetation from the upland blanket bog were similar to all other vegetation. This indicates that while *E. vaginatum* may have the ability to accumulate  $^{40}\text{K}$  within the Atlantic blanket bog it does not necessarily behave in an analogous manner in all ecosystems. Caesium-137 TF values were in general higher in the Atlantic blanket bog vegetation and in particular in the leaves of the heather species *C. vulgaris* which were found to accumulate  $^{137}\text{Cs}$  to a greater extent than any other vegetation assessed. A similar observation was apparent for the same vegetation in the upland blanket bog ecosystem.

The effect of soil physiochemical properties and nutrient status of vegetation appears to be contradictory in nature and are highly dependent on the radionuclide of interest, species of vegetation and the ecosystem of choice, however, in the case of magnesium levels in vegetation the results are more apparent. The levels of magnesium in vegetation appear to have a positive impact on the uptake of both  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in the majority of vegetation for the three ecosystems studied.

## 4.0 CONCLUSIONS

The primary objective of this study was to study the transfer of selected radionuclides ( $^{238}\text{U}$ ,  $^{228}\text{Ra}$ ,  $^{226}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$ ) from soil to vegetation in three Irish ecosystems and to ascertain if soil physiochemical properties, nutrient status of vegetation or species of vegetation have a significant affect on radionuclide uptake by vegetation. The results obtained outline the levels of these radionuclides within the soil and vegetation and also demonstrate the physiochemical properties of the soil and nutrient status of the vegetation for the three ecosystems. However, the central focus of this project is on radionuclide TF values and significant correlations which are evident between the TF values and soil properties or nutrient status of vegetation. While a detailed account of all the findings is discussed in detail within Section 3 of this thesis, a summary of the most significant findings are outlined within this section. The nature of this study is relatively new in Ireland in that while previous authors have studied the levels of radionuclides in the soil and to a limited extent within some vegetation, a comprehensive study on the comparison of radionuclide uptake within internationally recognised ecosystems in Ireland has not been done previous to this. The ecosystems chosen were categorised based on the guidelines outlined in the Interpretation Manual of European Union Habitats published by the European Commission (1999). The results within this thesis focuses mainly on the uptake of the radionuclides within the vegetation from each ecosystem in-line with recommendations from the ICRP and in addition to this, some consideration is also given to the potential transfer of these radionuclides into the human food-chain.

The results indicate that the levels of  $^{238}\text{U}$ ,  $^{228}\text{Ra}$  ( $^{232}\text{Th}$ ) and  $^{226}\text{Ra}$  were low in all soils and below the limits of detection for the Atlantic blanket bog soils. Uranium-238 and  $^{228}\text{Ra}$  concentrations in the vegetation from the three ecosystems were below the limits of detection for the methods used within this study. Radium-226 was detectable within the majority of vegetation from the upland blanket bog and also in the grass species, *H. lanatus* found in the grassland however, levels in the Atlantic blanket bog vegetation were below the limits of detection for the methods used in this study. Radium - 226 TF values in upland blanket bog vegetation ranged from 0.07 to 2.4 and transfer factors

values indicate that uptake of  $^{226}\text{Ra}$  was significantly higher for *E. tetralix* than for all other species of vegetation from the upland bog and than *H. lanatus* from the grassland. Radium-226 TF values in all other vegetation were statistically similar thereby indicating that *E. tetralix* appears to have the ability to accumulate  $^{226}\text{Ra}$  to greater extent than all other vegetation analysed.

Potassium-40 levels in the soil were significantly higher in the grassland soil in comparison to both the Atlantic blanket bog and upland blanket bog. The levels in the latter two ecosystems were found to be statistically similar. Potassium-40 was detectable in all vegetation from the three ecosystems and TF values ranged from 0.9 to 13.8 for Atlantic blanket bog vegetation and were significantly higher in *E. vaginatum* in comparison to both *C. vulgaris* leaves and stems. Uptake of  $^{40}\text{K}$  was detectable in all upland blanket bog vegetation and  $^{40}\text{K}$  TF values in the vascular plants ranged from 0.4 for *C. vulgaris* (leaves) to 11.2 for *M. gale* (leaves), however on average TF values ranged from 1.4 for *C. vulgaris* (stems) to 5.2 for *E. vaginatum*. Transfer factor values were compared based on plant species and results indicate that  $^{40}\text{K}$  transfer from soil to vegetation in this ecosystem is similar for all species of vegetation. In the case of *C. vulgaris* and *M. gale*, results indicate that  $^{40}\text{K}$  uptake is not dependent on plant compartment as TF values were not significantly different for the leaves and stems. Potassium-40 was detectable in all grassland vegetation and TF values ranged from 0.7 to 3.8 and were statistically similar for all species of vegetation indicating that transfer of  $^{40}\text{K}$  to vegetation within this ecosystem is not dependent on these plant species. Comparisons of  $^{40}\text{K}$  TF values for all vegetation in the three ecosystems indicate that uptake in *E. vaginatum* from the Atlantic blanket bog was significantly higher than all other vegetation analysed. This appears to indicate that *E. vaginatum* has the ability to accumulate  $^{40}\text{K}$  to a greater extent than all other vegetation, however, the TF values for the same vegetation in the upland blanket bog were significantly lower than its counterpart in the Atlantic blanket bog and statistically similar to all other vegetation.

Caesium-137 levels were found to be significantly higher in the two bogland soils in comparison to the grassland soil and levels of  $^{137}\text{Cs}$  decreased with an increasing soil

depth in all three ecosystems indicating that the majority of  $^{137}\text{Cs}$  is remaining within the rooting zone of the majority of vegetation. Caesium-137 was detectable in all Atlantic blanket bog vegetation and TF values ranged from 1.9 to 9.6 and TF values were significantly higher in the leaves of *C. vulgaris* in comparison to the stems. Caesium-137 was detectable in all upland blanket bog vegetation and TF values in vascular plants ranged from 0.12 for *M. caeruleae* to 6.26 for *C. vulgaris* (leaves), however, on average TF values ranged from 0.29 for *E. tetralix* to 1.6 for *C. vulgaris* (leaves). Caesium-137 TF values were significantly higher in the leaves of *C. vulgaris* than *M. gale* (leaves and stems), *E. tetralix*, *M. caeruleae* and *E. vaginatum*. Transfer factor values were also significantly higher in the leaves of *C. vulgaris* in comparison to the stems indicating that  $^{137}\text{Cs}$  uptake in this plant species appears to be dependent on plant compartment. A similar observation was made in the Atlantic blanket bog. Transfer factor values for the leaves and stems of *M. gale* indicate that uptake of  $^{137}\text{Cs}$  is not dependent on plant compartment as the TF values are statistically similar. Transfer factor values for all other plant species are statistically similar. Caesium-137 was only detectable in a four samples of vegetation from the grassland ecosystem and TF values in these cases ranged from 0.01 to 0.03. The uptake of  $^{137}\text{Cs}$  within grassland vegetation was found to be very low and TF values were in general lower than 0.02.

Comparisons of  $^{137}\text{Cs}$  TF values for all vegetation in the three ecosystems indicate that that uptake of  $^{137}\text{Cs}$  was highest in the leaves of *C. vulgaris*. This observation was apparent for both blanket bogs. Caesium-137 TF values also indicate that uptake of  $^{137}\text{Cs}$  is higher in the Atlantic blanket bog vegetation in comparison to the vegetation from the other two ecosystems.

The effect of soil physiochemical properties and the nutrient status of the vegetation on radionuclide uptake by vegetation appear to be highly dependent on the radionuclide of interest, species of vegetation and ecosystem of choice. Radium-226 TF values were only calculated for upland blanket bog vegetation and one species of grassland vegetation as levels in all other vegetation were below the limits of detection. Radium-226 TF values for upland blanket bog vegetation were found to be positively influenced by high levels

of organic matter, available calcium and available magnesium in the soil and with soil of high CEC. Transfer factors values for  $^{226}\text{Ra}$  were found to be adversely affected by high levels of available potassium in the soil and calcium in the vegetation. Radium-226 TF values were also significantly lower for vegetation growing on soils with high pH. The effect of soil moisture on the uptake of  $^{226}\text{Ra}$  appears to be contradictory and is dependent on the species of vegetation as both negative and positive correlation was found between these two parameters.

The impact of soil properties and nutrient status of the vegetation on  $^{40}\text{K}$  and  $^{137}\text{Cs}$  uptake by vegetation is less clear and in some cases contradictory. Potassium-40 TF values for Atlantic blanket bog were found to be positively influenced with the CEC of the soil and this observation was also evident for upland blanket bog vegetation. Potassium-40 TF values were also positively influenced by the following soil physiochemical parameters: organic matter, soil moisture content, pH of soil, available levels of calcium and magnesium in the soil. The nutrient status of the vegetation also appears to have enhanced the uptake of  $^{40}\text{K}$ , in particular, the levels of magnesium in vegetation whereas the levels of calcium in vegetation appears to have a contradictory impact on the uptake of  $^{40}\text{K}$  and is species dependent with this ecosystem. The uptake of  $^{40}\text{K}$  in grassland vegetation was higher in vegetation with high levels of magnesium as was evident in the upland blanket bog vegetation. Uptake of  $^{40}\text{K}$  was also enhanced in grassland vegetation with high level of sodium and in vegetation growing on soils with high levels of available sodium. Results indicate that for the Atlantic blanket bog and grassland vegetation the uptake of  $^{40}\text{K}$  is not adversely affected by any of the soil parameters or vegetation nutrients studied, whereas the uptake of  $^{40}\text{K}$  was lower for upland blanket bog vegetation growing on soil with high levels of  $^{40}\text{K}$  and available potassium and in vegetation with high levels of calcium present.

The nutrient status of vegetation appears to have a significant impact on the uptake of  $^{137}\text{Cs}$  on both the Atlantic blanket bog and upland blanket bog vegetation. High levels of magnesium and calcium both appear to enhance the uptake of  $^{137}\text{Cs}$  in the vegetation from the two ecosystems however the effect of calcium in vegetation is less clear and has

a negative influence within some vegetation. This appears to indicate that the effect of calcium on  $^{137}\text{Cs}$  uptake by upland blanket bog vegetation is species dependent. The uptake of  $^{137}\text{Cs}$  by grassland vegetation was very low and undetectable in the majority of cases. The vegetation with detectable levels of  $^{137}\text{Cs}$  present appears to be unaffected by the soil parameters studied and vegetation nutrients studied. Results indicate that for the Atlantic blanket bog and grassland vegetation the uptake of  $^{137}\text{Cs}$  is not affected by any of the soil parameters or vegetation nutrients, whereas the uptake of  $^{137}\text{Cs}$  was lower for upland blanket bog vegetation growing on soil with high levels of  $^{137}\text{Cs}$  and available sodium. The most apparent observation from the data is the effect of magnesium levels on the uptake of both of  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in bog vegetation. The levels of magnesium in vegetation from these ecosystems appear to have a positive influence on the uptake of both  $^{40}\text{K}$  and  $^{137}\text{Cs}$ .

The influence of soil radionuclide levels on vegetation TF values are clear and results indicate that uptake of  $^{226}\text{Ra}$  by vascular plants is independent of levels of  $^{226}\text{Ra}$  in the soil. Potassium-40 TF values indicate that uptake of this radionuclides was adversely impacted by the levels of  $^{40}\text{K}$  in the soil. This observation was evident for the vegetation from the two bog ecosystems, however, the uptake of  $^{40}\text{K}$  by grassland vegetation appears to be independent of the levels present in the soil. Caesium-137 TF values were compared with the levels in the soil for the three ecosystems and results indicate that uptake of  $^{137}\text{Cs}$  by these vegetation is independent of the levels in the soil with the exception of *E. tetralix* and *M. gale*.

In summary, this study highlights the behaviour of  $^{238}\text{U}$ ,  $^{228}\text{Ra}$  ( $^{232}\text{Th}$ ),  $^{226}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  within three Irish ecosystems with respect to their transfer into indigenous vegetation. Based on the results obtained, the uptake of  $^{238}\text{U}$  and  $^{228}\text{Ra}$  by vegetation from the three ecosystems was undetectable in all vegetation while levels in the soil from the wet grassland and upland bog were detectable. It is reasonable to assume from these findings that the vegetation from these particular ecosystems do not portray the ability to accumulate either of these radionuclides to any significant extent. However, this

assumption cannot be upheld in the case of the Atlantic Blanket bog vegetation as the levels of these radionuclides were undetectable in the soil of this ecosystem.

The behaviour of  $^{226}\text{Ra}$  in the Atlantic blanket bog and grassland ecosystems is very similar to  $^{238}\text{U}$  and  $^{228}\text{Ra}$  as the levels in the vegetation from both ecosystems were below the limits of detection with the exception of *H. lanatus* from the grassland which demonstrated limited ability to accumulate  $^{226}\text{Ra}$ . The behaviour of  $^{226}\text{Ra}$  within the upland blanket bog is quite different as uptake of this radionuclide was detectable in the majority of the indigenous vegetation. The uptake of  $^{226}\text{Ra}$  was in general low in comparison to both  $^{40}\text{K}$  and  $^{137}\text{Cs}$  and TF values indicate that the uptake by all species of vegetation was similar with the exception of *E. tetralix*. This species of heather demonstrated the ability to accumulate  $^{226}\text{Ra}$  to a greater extent than all other vegetation from all three ecosystems. The results also suggest that the uptake of  $^{226}\text{Ra}$  by vegetation is significantly enhanced when growing on soils with higher levels of organic matter, plant available magnesium and CEC whereas it is diminished on soils with high level of plant available potassium. This may possibly be a factor when considering the diminished uptake by grassland vegetation as the soil within this ecosystem has significantly lower levels of organic matter, plant available magnesium and CEC in comparison to the upland bog soils and higher levels of plant available potassium.

Potassium-40 TF values were in general significantly higher than  $^{226}\text{Ra}$  and it was the only radionuclide detectable in all vegetation from the three ecosystems. The uptake of this radionuclide was significantly dependent on the species of vegetation as *E. vaginatum* was found to accumulate  $^{40}\text{K}$  to a greater extent than all other vegetation. This observation is dependent on the origin of the vegetation as higher uptake by *E. vaginatum* was evident in the Atlantic blanket bog in comparison to the upland blanket bog.

Caesium-137 TF values were detectable in all vegetation from the two bog ecosystems but only to a very limited extent in the grassland ecosystem. The most apparent observation from this data was the ability of *C. vulgaris* to accumulate  $^{137}\text{Cs}$  and in particular the leaves of this species of vegetation. This observation was noted in the two

bog ecosystems but was more pronounced in the Atlantic blanket bog. The impact of soil physiochemical properties and nutrient status of vegetation on both  $^{40}\text{K}$  and  $^{137}\text{Cs}$  uptake was both complex and contradictory and the most apparent observation from this data was the positive influence of magnesium levels in vegetation on both radionuclides.

The results within this thesis have demonstrated the behaviour of these radionuclides within natural ecosystems in Ireland and have focused mainly on the impact of these radionuclides within the vegetation indigenous to these ecosystems. In addition to this it is important to consider the potential transfer of these radionuclides into the human food-chain with respect to the levels present in vegetation in comparison to recommended threshold levels for human exposure outlined in legislation. The results have been compared with annual limits of intake (ALIs) specified in Title 10, Code of Federal Regulations (U.S. Nuclear Regulatory Commission) and are discussed within section 3.4 of this thesis. It was found that radionuclide levels in the vegetation within this study were much lower than the ALIs outlined in legislation and were generally of little concern in relation to human contamination. In the case of  $^{226}\text{Ra}$ , the heather species *E. tetralix* demonstrated the ability to accumulate significant quantities of this radionuclide. This is of concern as sheep are commonly found grazing on heather within this type of ecosystem (upland blanket bog), however, the levels found in this vegetation and all vegetation studied were at least 2,500 times lower than those outlined in the ALIs for oral ingestion. Therefore, the potential risk to humans is substantially reduced. This is also the case with vegetation which has demonstrated the ability to accumulate significant quantities of  $^{40}\text{K}$  and  $^{137}\text{Cs}$ . The levels of  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in all vegetation were 11,000 and 3,700 lower respectively than those specified in the ALIs for oral ingestion. Therefore, by comparison, the levels of radionuclides in the vegetation of this study do not possess a significant threat to the human food-chain, however, it is important to remember when dealing with radionuclides that there is no safe level of exposure to ionising radiation.

The approach of this study is novel in the Irish context as it is the first study to compare the uptake of these radionuclides by vegetation from the perspective of three internationally recognised ecosystems. The results indicate that the uptake of  $^{226}\text{Ra}$ ,  $^{40}\text{K}$

and  $^{137}\text{Cs}$  by the chosen vegetation is varied and complex and is significantly dependent on the species of vegetation, soil radionuclide concentration, soil physiochemical properties and the nutrient status of the vegetation. These results provide a foundation for understanding the fate of these radionuclides within the chosen ecosystems, however, further exploration is required to expand on the body of knowledge contained in this thesis. Such work should focus upon the species of vegetation which have demonstrated the ability to accumulate significant levels of the individual radionuclides. The influence of soil radionuclide levels on vegetation uptake could be assessed by the use of pot trials as the levels in the soils of this study were low for all three ecosystems. The integration of pot trials into this type of study could also be expanded to investigate the importance of soil physiochemical properties on radionuclide uptake by each vegetation species. This could be achieved by significantly varying the soil composition upon which the vegetation is growing. Additional research within other internationally recognised ecosystems in Ireland would also be very beneficial and particularly within ecosystems with naturally elevated levels of radioactivity. Such future work could build on the knowledge derived from this study and thus enhance our understanding on the behaviour of these radionuclides within other internationally recognised ecosystems.

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# APPENDIX 1



# REFERENCE SHEET

## REFERENCE MATERIAL

### IAEA-375

#### RADIONUCLIDES AND TRACE ELEMENTS IN SOIL

Date of issue: January 2000<sup>Ⓟ</sup>

Recommended Values  
(Based on dry weight)

Reference Date for decay correction: 31<sup>st</sup> December 1991

Radionuclide	Recommended Value Bq/kg	95% Confidence Interval Bq/kg	N*
<sup>40</sup> K	424	417 – 432	84
<sup>90</sup> Sr	108	101 – 114	43
<sup>106</sup> Ru	56	53 – 58	26
<sup>125</sup> Sb	77	74 – 79	38
<sup>129</sup> I	0.0017	0.0013 – 0.0021	10
<sup>134</sup> Cs	463	454 – 472	87
<sup>137</sup> Cs	5280	5200 – 5360	91
<sup>226</sup> Ra	20	18 – 22	37
<sup>232</sup> Th	20.5	19.2 – 21.9	11

Recommended Values  
(Based on dry weight)

Element	Recommended Value mg/kg	95% Confidence Interval mg/kg	N*
Th	5.2	5.0 – 5.4	26
U	1.86	1.66 – 2.05	30

Number of accepted laboratory means which were used to calculate the recommended values and confidence intervals.

Revision of the reference sheet dated December 1997; original report date: August 1994.

Note: Some evidence has been presented to suggest that this material may be contaminated with "hot particles" resulting from the Chernobyl accident. The frequency of the occurrence of these "hot particles" is unknown and consequently, it is possible that significantly elevated activities may be observed for anthropogenic radionuclides in some sub-samples.

**Information Values**  
(Based on dry weight)

Reference Date for decay correction: 31<sup>st</sup> December 1991

Radionuclide	Information Value Bq/kg	95% Confidence Interval Bq/kg	N*
<sup>228</sup> Th	21	17 – 25	6
<sup>234</sup> U	25	17 – 32	5
<sup>238</sup> U	24.4	19.0 – 29.8	7
<sup>238</sup> Pu	0.071	0.056 – 0.085	10
<sup>239+240</sup> Pu	0.30	0.26 – 0.34	20
<sup>241</sup> Am	0.13	0.11 – 0.15	6

**Information Values**  
(Based on dry weight)

Element	Information Value mg/kg	95% Confidence Interval mg/kg	N*
As	2.56	2.24 – 2.88	6
Ba	335	316 – 354	6
La	17.6	15.3 – 19.9	5
Ni	9.7	7.9 – 11.6	5
Rb	48	44 – 52	8
Yb	1.73	1.24 – 2.22	5

*Number of accepted laboratory means which were used to calculate the information values and confidence intervals.*

The values listed above were established on the basis of statistically valid results submitted by laboratories which had participated in an international intercomparison exercise organized in 1992. The details concerning the criteria for allocation as a recommended value can be found in the report (IAEA/AL/075) "Report on the Intercomparison Run EA-375: Radionuclides in Soil" [1]. This report is available free of charge upon request.

**Intended Use**

This sample is intended to be used as a reference material for the measurement of radionuclides in soil samples. It can also be used as a quality control material for the assessment of a laboratory's analytical work, for the validation of analytical methods and for quality assurance within a laboratory.

**Dry weight determination**

All recommended values are expressed on a dry weight basis. Therefore the dry weight must be determined at the time of analysis, using separate sub-samples of at least 500 mg dried to constant weight in a drying oven set to 105 °C. Subsequent weighings should differ by less than 5 mg.

**Instructions for use**

The recommended minimum intake masses for analysis of trace elements and radionuclides are 0.2 g and 6g respectively.

Analysts are reminded to take appropriate precautions in order to avoid contamination of the material during handling. Special precautions are required for the storage of this material.

**References**

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# REFERENCE SHEET

## REFERENCE MATERIAL

### IAEA-326

## RADIONUCLIDES IN SOIL

Date of issue: April 2001<sup>⊕</sup>

Recommended Values  
(Based on dry weight)

Reference Date for decay correction: 31<sup>st</sup> December 1994

Radionuclide	Recommended Value Bq/kg	95% Confidence Interval Bq/kg	N*
$^{40}\text{K}$	580	571 – 589	148
$^{90}\text{Sr}$	10.2	9.7 – 10.7	37
$^{210}\text{Pb}$	53.3	48.8 – 57.8	24
$^{226}\text{Ra}$	32.6	31.0 – 34.2	58
$^{228}\text{Ra}$	40.1	39.3 – 40.9	108
$^{228}\text{Th}$	39.1	38.2 – 40.0	55
$^{230}\text{Th}$	34.1	31.8 – 36.4	10
$^{232}\text{Th}$	39.4	37.6 – 41.2	21
$^{234}\text{U}^\#$	29.3	26.5 – 29.3	31
$^{238}\text{U}$	29.4	28.1 – 30.7	31
$^{238}\text{Pu}$	0.019	0.017 – 0.021	14
$^{239+240}\text{Pu}$	0.50	0.48 – 0.52	44

\* Number of accepted laboratory means which were used to calculate the recommended values and confidence intervals.

# The recommended value for  $^{234}\text{U}$  was calculated from the  $^{238}\text{U}$  recommended activity concentration in combination with the median of the  $^{234}\text{U}/^{238}\text{U}$  activity ratio calculated from participants results.

⊕ Revision of the original reference sheet dated May 2000.

**Information Values**  
(Based on dry weight)

Reference Date for decay correction: 31<sup>st</sup> December 1994

Radionuclide	Information Value	95% Confidence Interval	N*
	Bq/kg	Bq/kg	
<sup>106</sup> Ru	< 3	—	44
<sup>125</sup> Sb	< 1.5	—	52
<sup>154</sup> Eu	< 3	—	39
<sup>155</sup> Eu	< 1.8	—	4
<sup>235</sup> U	1.48	1.21 – 1.75	11
<sup>241</sup> Am <sup>##</sup>	0.19	0.16 – 0.22	12

\* Number of accepted laboratory means which were used to calculate the information values and confidence intervals.

## NB: The <sup>241</sup>Am value quoted is only valid for the reference date, as decay correction to alternated dates requires knowledge of the <sup>241</sup>Pu concentration which is not currently known.

The values listed above were established on the basis of statistically valid results submitted by laboratories which had participated in an international intercomparison exercise organized in 1994. The details concerning the criteria for qualification as a recommended value can be found in the report (IAEA/AL/100) "Report on the Intercomparison Run IAEA-326/327: Radionuclides in Soil" [1]. This report is available free of charge upon request.

#### Intended Use

This sample is intended to be used as a reference material for the measurement of radionuclides in soil samples. It can also be used as a quality control material for the assessment of a laboratory's analytical work, for the validation of analytical methods and for quality assurance within a laboratory.

#### Dry weight determination

All recommended values are expressed on a dry weight basis. Therefore the dry weight must be determined at the time of analysis, using separate sub-samples of at least 500 mg dried to constant weight in a drying oven set to 105 °C. Subsequent weighings should differ by less than 5 mg.

#### Instructions for use

The recommended minimum sample masses for analysis of  $\alpha$ -emitters,  $\beta$ -emitters and  $\gamma$ -emitters are 0.5g, 0.5g and 30g respectively.

Analysts are reminded to take appropriate precautions in order to avoid contamination of the material during handling. No special precautions are required for the storage of this material.

#### References

- [1] Bojanowski R., Radecki Z., Campbell M.J., Burns K.I., and Trinkl A., Report on the Intercomparison Run for the Determination of Radionuclides in Soils IAEA-326 and IAEA-327. IAEA/AL/100, IAEA, Vienna, Austria 2001.



International Atomic Energy Agency  
Analytical Quality Control Services  
Wagramer Strasse 5, P.O.Box 100, A-1400 Vienna, Austria

# REFERENCE SHEET

REFERENCE MATERIAL

**IAEA-SOIL-6**

RADIONUCLIDES IN SOIL

Date of issue: September 1999<sup>Ⓢ</sup>

Recommended Values  
(Based on dry weight)

Reference Date for decay correction: 30<sup>th</sup> January 1983

Element	Recommended Value Bq/kg	95% Confidence Interval Bq/kg	N*
<sup>90</sup> Sr	30.34	24.20 - 31.67	20
<sup>137</sup> Cs	53.65	51.43 - 57.91	21
<sup>226</sup> Ra	79.9	69.6 - 93.4	23
<sup>239+240</sup> Pu	1.04	0.96 - 1.11	6

*Number of accepted laboratory means which were used to calculate the recommended values and confidence intervals about the median value.*

*Update of the revised reference sheet dated September 1997*

The values listed above were established on the basis of statistically valid results submitted by laboratories which had participated in an international intercomparison exercise conducted between 1983-1984. The details concerning the criteria for qualification as a recommended value can be found in the report (IAEA/RL/111) "Report on the Intercomparison Run IAEA-Soil-6: Radionuclides in soil" [1]. This report is available free of charge upon request.

## Intended Use

This sample is intended to be used as a reference material for the measurement of radionuclides in soil samples. It can also be used as a quality control material for the assessment of a laboratory's analytical work, for the validation of analytical methods and for quality assurance within a laboratory.

## Homogeneity

The homogeneity of the material was assessed non-destructively by measuring the  $^{40}\text{K}$  and  $^{137}\text{Cs}$  activities in twenty bottles, chosen at random, using gamma-spectrometry. Taking into consideration the activity concentrations measured for  $^{137}\text{Cs}$  and  $^{40}\text{K}$ , the material could be considered homogeneous for sample sizes of 250 g.

Note: Experience with this material at the Agency's Laboratories, Seibersdorf, has shown that 250 g is an acceptable intake mass for  $^{239+240}\text{Pu}$  and  $^{90}\text{Sr}$  determination [2].

## Dry weight determination

All recommended values are expressed on a dry weight basis. Therefore the dry weight must be determined at the time of analysis, using separate sub-samples of at least 500 mg dried to constant weight in a drying oven set to 105 °C. Subsequent weighings should differ by less than 5 mg.

## Instructions for use

The recommended sample size for analysis is 250 g. Analysts are reminded to take appropriate precautions in order to avoid contamination of the material during handling. No special precautions are required for the storage of this material.

## References

- Pszonicki L., Hanna A. N., Reichl F. and Suschny O., Report on the Intercomparison Run IAEA-Soil-6: Radionuclides in Soil. IAEA/RL/111, IAEA, Vienna, Austria 1984.
- Fajgelj A., Radecki Z., Burns K.I., Moreno Bermudez J., De Regge P.P., Danesi P.R., Bojanowski R. and La Rosa J., Intended Use of the IAEA Reference Materials Part I: Examples on Reference Materials for the Determination of Radionuclides or Trace Elements in "The Use of Matrix Reference Materials in Environmental Analytical Processes", Eds. Fajgelj A., Parkany M., The Royal Society of Chemistry, Cambridge, UK, 1999.

### Summary

This report contains the results of the intercomparison IAEA-156 on the determination of radionuclides in clover. Initially participants were requested to determine the levels of  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{40}\text{K}$ ,  $^{90}\text{Sr}$  and invited to provide data for other radionuclides. The participants included 46 laboratories located in 25 countries, and statistical evaluation of their data yield recommended values for these four radionuclides. Additional radionuclides reported were  $^{210}\text{Pb}$ ,  $^{239}\text{Pu}$  and  $^{125}\text{Sb}$ ; however, insufficient data exists to statistically determine recommended values for these radionuclides.

The radionuclides, their recommended values and confidence intervals are listed below:

Cs-134:	132.1 Bq/kg	(126.4 - 137.7)
Cs-137:	264 Bq/kg	(254 - 274 )
K-40 :	657 Bq/kg	(637 - 676 )
Sr-90 :	14.8 Bq/kg	(13.4 - 16.3 )

Reference date: 1 August 1986

Table C

Recommended Values and Confidence Intervals for  
Radionuclides in IAEA-156 Clover

Radionuclide	Recommended value, Bq/kg	Confidence interval[*]
$^{134}\text{Cs}$	132	126 - 138
$^{137}\text{Cs}$	264	254 - 274
$^{40}\text{K}$	657	637 - 676
$^{90}\text{Sr}$	14.8	13.4 - 16.3

[\*] Confidence intervals are for significance level 0.05

Reference date: 1 August 1986

## **APPENDIX 2**



# Kalibrierschein

*Calibration certificate*

Gegenstand:  
*Object:* Aktivitätsnormal

Hersteller:  
*Manufacturer:* Physikalisch-Technische Bundesanstalt  
Fachlaboratorium 6.11

Typ:  
*Type:* Wässrige Lösung in Glasampulle

Kenn-Nummer:  
*Serial number:* 2001-1258

Auftraggeber:  
*Applicant:* Statens Strålevern  
Norwegian Radiation Protection Authority  
Grini næringspark 13  
  
N-1332 Østerås

Anzahl der Seiten:  
*Number of pages:* 3

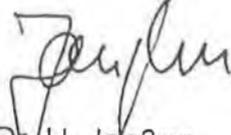
Geschäftszeichen:  
*Reference number:* PTB-6.11-210/30.2001

Kalibrierzeichen:  
*Calibration mark:* PTB-6.11-2001-1258

Datum der Kalibrierung:  
*Date of calibration:* 10.04.2001

Im Auftrag  
*By order* Braunschweig, 10.04.2001

Bearbeiter:  
*Examiner:*

  
Dr. H. Janßen

Siegel  
*Seal*



  
M. Ehlers

## Empfohlene Radionukliddaten

Stand: 01.08.1999

Radionuklid: Co-60

Halbwertszeit:  $T_{1/2} = 1925,3(4)$  Tage [5,2712(11) Jahre]Gammastrahlen-Energien  $E$  und –Emissionswahrscheinlichkeiten  $p$  pro Zerfall:

Strahlungs- typ	$E$ in keV	$p$	Kommentar
$\gamma$	347,14	0,000075(4)	
$\gamma$	826,10	0,000076(8)	
$\gamma$	1173,23	0,9985(3)	
$\gamma$	1332,49	0,999826(6)	
$\gamma$	2158,57	0,000012(2)	
$\gamma$	2505,69	0,000000020(4)	

**Meßunsicherheiten:**

Die Ziffern in Klammern hinter dem Zahlenwert einer Meßgröße geben die Meßunsicherheit in der letzten Stelle des Zahlenwertes an. Die Meßunsicherheiten entsprechen der einfachen Standardabweichung.

**Literatur:**

Schötzig, U. und Schrader, H.:

Halbwertszeiten und Photonen-Emissionswahrscheinlichkeiten von häufig verwendeten Radionukliden; PTB-Bericht PTB-Ra-16/5, Braunschweig 1998, ISBN 3-89701-279-0

Gegenstand: <i>Object</i>	Aktivitätsnormal
Typ: <i>Type</i>	Wässrige Lösung in Glasampulle
Kennzeichnung: <i>Serial number</i>	2001-1258
Radionuklid: <i>Radionuclide</i>	Cobalt-60
Chemische Zusammensetzung der wässrigen Lösung: <i>Chemical composition of the aqueous solution</i>	HCl            0,1mol/L CoCl <sub>2</sub> 50 mg/L
Kalibrierverfahren: <i>Method of calibration</i>	Die spezifische Aktivität der Lösung wurde durch Messung der Gammastrahlung mit einer 4π-Ionisationskammer bestimmt. Die 4π-Ionisationskammer ist eine Sekundärnormal-Messeinrichtung, kalibriert mit Aktivitätsnormalen der PTB.
Lösungsmasse: <i>Mass of solution</i>	$m = (2,0232 \pm 0,0017) \text{ g}$
Spezifische Aktivität: <i>Specific activity</i>	$a = (4,55 \pm 0,05) \text{ kBq/g}$
Bezugszeitpunkt: <i>Reference date</i>	01.03.2001, 00:00 Uhr MEZ
Messunsicherheit: <i>Uncertainty of measurement</i>	Angegeben ist die erweiterte Messunsicherheit, die sich aus der Standardmessunsicherheit durch Multiplikation mit dem Erweiterungsfaktor $k = 2$ ergibt. Sie wurde gemäß dem "Guide to the Expression of Uncertainty in Measurement" (ISO, 1995) ermittelt. Der Wert der Messgröße liegt im Regelfall mit einer Wahrscheinlichkeit von annähernd 95 % im zugeordneten Werteintervall.



Sligo  
An Institut für Teilchenphysik, Silgach

PREPARERING AV TRACER-LØSNING

Løpenr. Stamløsning: 2001-1258

Prosedyre benyttet:

Nuklide: Co-60

Halvt.,  $T_{1/2}$  dager: 1925.3

Ref. dato,  $t_0$ : 01.03.2001

Total mengde st.løsning, (g) 2.0232 ± 0.0007

Mengde st.løsning veld ut, (g): 1.9757

Tetthet st.løsning g/cm<sup>3</sup> (se sertifikat) 1.00

Aktivitet / ampulle, Bq (se sertifikat) 9205.56

D<sub>0</sub> Aktivitet tatt ut av ampulle, Bq: 8989.4 #DIV/0!

Prepareringsdato: 18.09.2001

Aktivitet ved prep.dato, Bq:

1. fortynning

Vekt, tom flaske/kolbe, (g):

Total vekt flaske, med løsn. etter fortynning (g):

Mengde løsning tilsatt st.løsning, (g):

Sp.akt. i tracer løsn. etter 1. fortynning, A, (Bq/g): 4.494 #VERD!!

Sp.akt. i tracer løsn. etter 1. fortynning, A, (Bq/ml): 4.494 #VERD!!

Nytt løpenr.

Merknad: Kolbe benyttet?

Tracer preparert av (dato, sign.): 18/09-01 Bjørn Lind

Vitne til preparering. (dato, sign.): De Dordall. 18.09.2001



Kalibrierschein  
Calibration certificate

Gegenstand:  
*Object:* Aktivitätsnormal

Hersteller:  
*Manufacturer:* Physikalisch-Technische Bundesanstalt  
Fachlaboratorium 6.11

Typ:  
*Type:* Wässrige Lösung in Glasampulle

Kenn-Nummer:  
*Serial number:* 10850

Auftraggeber:  
*Applicant:* Statens Strålevern  
Norwegian Radiation Protection Authority  
Grini næringspark 13  
  
N-1332 Østerås

Anzahl der Seiten:  
*Number of pages:* 3

Geschäftszeichen:  
*Reference number:* PTB-6.11-210/30.2001

Kalibrierzeichen:  
*Calibration mark:* PTB-6.11-10850

Datum der Kalibrierung:  
*Date of calibration:* 21.03.2001

Im Auftrag  
*By order* Braunschweig, 21.03.2001

Bearbeiter:  
*Examiner:*

Dr. H. Janßen

Siegel  
*Seal*



M. Ehlers

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Sligo  
An Institiúid Teicneolaíochta, Sligeach

Gegenstand:  
*Object*

Aktivitätsnormal

Typ:  
*Type*

Wässrige Lösung in Glasampulle

Kennzeichnung:  
*Serial number*

10850

Radionuklid:  
*Radionuclide*

Americium-241

Chemische Zusammensetzung  
der wässrigen Lösung:  
*Chemical composition of the  
aqueous solution*

HNO<sub>3</sub> 0,1 mol/L  
La(NO<sub>3</sub>)<sub>3</sub> 35 mg/L

Kalibrierverfahren:  
*Method of calibration*

Die spezifische Aktivität der Lösung wurde durch Messung der Gammastrahlung mit einer 4 $\pi$ -Ionisationskammer bestimmt. Die 4 $\pi$ -Ionisationskammer ist eine Sekundärnormal-Messeinrichtung, kalibriert mit Aktivitätsnormalen der PTB.

Lösungsmasse:  
*Mass of solution*

$m = (2,0226 \pm 0,0017) \text{ g}$

Spezifische Aktivität:  
*Specific activity*

$a = (4,86 \pm 0,07) \text{ kBq/g}$

Bezugszeitpunkt:  
*Reference date*

01.03.2001, 00:00 Uhr MEZ

Messunsicherheit:  
*Uncertainty of measurement*

Angegeben ist die erweiterte Messunsicherheit, die sich aus der Standardmessunsicherheit durch Multiplikation mit dem Erweiterungsfaktor  $k = 2$  ergibt. Sie wurde gemäß dem "Guide to the Expression of Uncertainty in Measurement" (ISO, 1995) ermittelt. Der Wert der Messgröße liegt im Regelfall mit einer Wahrscheinlichkeit von annähernd 95 % im zugeordneten Werteintervall.



## Empfohlene Radionuklidaten

Stand: 01.08.1999

Radionuklid: Am-241

Halbwertszeit:  $T_{1/2} = 1,5785(24) \cdot 10^5$  Tage [432,2(7) Jahre]

Photonen-Energien  $E$  und  $\alpha$ -Emissionswahrscheinlichkeiten  $p$  pro Zerfall:

Strahlungs- typ	$E$ in keV	$p$	Kommentar
Np-L <sub>1</sub>	11,87	0,00837(10)	
Np-L <sub>α2</sub>	13,76	0,0108(5)	
Np-L <sub>α1</sub>	13,95	0,1193(11)	
Np-L <sub>η</sub>	15,86	0,00377(15)	
Np-L <sub>β</sub>	17,54	0,1861(15)	
Np-L <sub>γ</sub>	21,01	0,0482(4)	
γ	26,34	0,02400(20)	
γ	32,18	0,000174(4)	
γ	33,20	0,00126(3)	
γ	42,73	0,000055(11)	
γ	43,42	0,00073(8)	
γ	55,56	0,000181(18)	
γ	57,85	0,000052(15)	
γ	59,54	0,359(4)	
γ	64,83	0,00000145(18)	
γ	67,45	0,0000042(10)	
γ	69,76	0,000029(4)	
γ	75,80	0,0000059(18)	
γ	98,97	0,000203(4)	
γ	102,98	0,000195(4)	
γ	123,01	0,0000100(3)	
γ	125,30	0,0000408(9)	
γ	146,55	0,00000461(11)	
γ	169,56	0,00000173(4)	
γ	208,01	0,00000791(17)	

PREPARERING AV TRACER-LØSNING

Løpenr. Stam-løsning 10850

Prosedyre benyttet:

Nuklide: Am-241

Halvt.  $T_{1/2}$  dager:  $1.5785 \times 10^5$

Ref. dato  $t_0$ : ~~21.03.2001~~

01.03.2001

Total mengde st.løsning, (g) 2.0226 ± 0.0017

Mengde st.løsning veld ut, (g) 1.9501.

Tetthet st.løsning  $g/cm^3$  (se sertifikat) 1.00

Aktivitet i ampulle, Bq (se sertifikat) 9829.83.

D<sub>0</sub> Aktivitet tatt ut av ampulle, Bq: 9477.4 #DIV/0!

Prepareringsdato: 18.09.2001

Aktivitet ved prep.dato, Bq

1. fortytning

Vekt, tom flaske/kolbe, (g)

Total vekt flaske, med løsn. etter fortytning (g)

Mengde løsning tilsatt st.løsning, (g)

Sp.akt. i tracer løsn. etter 1. fortytning, A, (Bq/g) 4.738 #VERDI!!

Sp.akt. i tracer løsn. etter 1. fortytning, A, (Bq/ml) 4.738 #VERDI!!

Nytt løpenr.:

Merknad: Kolbe benyttet?

Tracer preparert av (dato, sign.):

18/9-01 Bjørn Lind

Vitne til preparering (dato, sign.):

M. Dowdell, 18.09.2001



## Kalibrierschein

*Calibration certificate*

Gegenstand:  
*Object:* Aktivitätsnormal

Hersteller:  
*Manufacturer:* Physikalisch-Technische Bundesanstalt  
Fachlaboratorium 6.11

Typ:  
*Type:* Wässrige Lösung in Glasampulle

Kenn-Nummer:  
*Serial number:* 2001-1177

Auftraggeber:  
*Applicant:* Statens Strålevern  
Norwegian Radiation Protection Authority  
Grini næringspark 13  
  
N-1332 Østerås

Anzahl der Seiten:  
*Number of pages:* 3

Geschäftszeichen:  
*Reference number:* PTB-6.11-210/30.2001

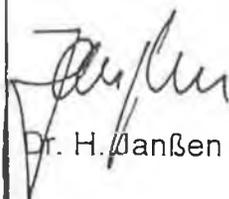
Kalibrierzeichen:  
*Calibration mark:* PTB-6.11-2001-1177

Datum der Kalibrierung:  
*Date of calibration:* 03.04.2001

Im Auftrag  
*By order*

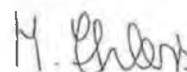
Braunschweig, 03.04.2001

Bearbeiter:  
*Examiner:*

  
Dr. H. Janßen

Siegel  
*Seal*





M. Ehlers

Gegenstand:  
*Object*

Aktivitätsnormal

Typ:  
*Type*

Wässrige Lösung in Glasampulle

Kennzeichnung:  
*Serial number*

2001-1177

Radionuklid:  
*Radionuclide*

Cobalt-57

Chemische Zusammensetzung  
 der wässrigen Lösung:  
*Chemical composition of the  
 aqueous solution*

HCl            0,1 mol/L  
 CoCl<sub>2</sub>        50 mg/L

Kalibrierverfahren:  
*Method of calibration*

Die spezifische Aktivität der Lösung wurde durch Messung der Gammastrahlung mit einer 4π-Ionisationskammer bestimmt. Die 4π-Ionisationskammer ist eine Sekundärnormal-Messeinrichtung, kalibriert mit Aktivitätsnormalen der PTB.

Lösungsmasse:  
*Mass of solution*

$m = (1,9787 \pm 0,0017) \text{ g}$

Spezifische Aktivität:  
*Specific activity*

$a = (23,8 \pm 0,4) \text{ kBq/g}$

Bezugszeitpunkt:  
*Reference date*

01.03.2001, 00:00 Uhr MEZ

Messunsicherheit:  
*Uncertainty of measurement*

Angegeben ist die erweiterte Messunsicherheit, die sich aus der Standardmessunsicherheit durch Multiplikation mit dem Erweiterungsfaktor  $k = 2$  ergibt. Sie wurde gemäß dem "Guide to the Expression of Uncertainty in Measurement" (ISO, 1995) ermittelt. Der Wert der Messgröße liegt im Regelfall mit einer Wahrscheinlichkeit von annähernd 95 % im zugeordneten Werteintervall.

**Sligo**  
 An Institut für Teilchenphysik, Sligo



## Empfohlene Radionuklidaten

Stand: 01.08.1999

Radionuklid: Co-57

Halbwertszeit:  $T_{1/2} = 271,83(8)$  TagePhotonen-Energien  $E$  und  $\gamma$ -Emissionswahrscheinlichkeiten  $p$  pro Zerfall:

Strahlungs- typ	$E$ in keV	$p$	Kommentar
Fe-L	0,71	0,0127(21)	
Fe-K $_{\alpha}$	6,40	0,510(7)	
Fe-K $_{\beta}$	7,06	0,069(1)	
$\gamma$	14,41	0,0916(15)	
$\gamma$	122,06	0,8560(17)	
$\gamma$	136,47	0,1068(8)	
$\gamma$	230,40	0,0000040(12)	
$\gamma$	339,69	0,000037(11)	
$\gamma$	352,33	0,000030(9)	
$\gamma$	366,80	0,000012(4)	
$\gamma$	570,09	0,00016(5)	
$\gamma$	692,41	0,00149(10)	
$\gamma$	706,54	0,000050(15)	

**Meßunsicherheiten:**

Die Ziffern in Klammern hinter dem Zahlenwert einer Meßgröße geben die Meßunsicherheit in der(n) letzten Stelle(n) des Zahlenwertes an. Die Meßunsicherheiten entsprechen der einfachen Standardabweichung.

**Literatur:**

Schötzig, U. und Schrader, H.:

Halbwertszeiten und Photonen-Emissionswahrscheinlichkeiten von häufig verwendeten Radionukliden; PTB-Bericht PTB-Ra-16/5, Braunschweig 1998, ISBN 3-89701-279-0

PREPARERING AV TRACER LØSNING

Løpenr. Stamløsning: 2001-1177

Prosedyre benyttet:

Nuklide:

Halvt.  $T_{1/2}$  i dager:

Ref. dato,  $t_0$ :

Total mengde st.løsning, (g)	1,9787 ± 0.0017
Mengde st.løsning veld ut, (g)	1.9411.
Tetthet st.løsning g/cm <sup>3</sup> (se sertifikat)	1.00
Aktivitet i ampulle, Bq (se sertifikat)	47093.06
D <sub>0</sub> Aktivitet tatt ut av ampulle, Bq	46198.18 #DIV/0!

Prepareringsdato:

Aktivitet ved prep dato, Bq:

1. fortykning:

Vekt, tom flaske/kolbe (g)	<input type="text"/>
Total vekt flaske, med løsn. etter fortykning (g)	<input type="text"/>
Mengde løsning tilsatt st.løsning, (g)	<input type="text"/>
Sp.akt. i tracer løsn. etter 1. fortykning, A, (Bq/g)	23.099 #VERDI!
Sp.akt. i tracer løsn. etter 1. fortykning, A, (Bq/ml)	23.099. #VERDI!

Nytt løpenr.

Merknad: Kolbe benyttet?

Tracer preparert av (dato, sign.):

Vitne til preparering. (dato, sign.):



139 C<sub>2</sub>



## Kalibrierschein

*Calibration certificate*

Gegenstand:  
*Object:* Aktivitätsnormal

Hersteller:  
*Manufacturer:* Physikalisch-Technische Bundesanstalt  
Fachlaboratorium 6.11

Typ:  
*Type:* Wässrige Lösung in Glasampulle

Kenn-Nummer:  
*Serial number:* 2001-1190

Auftraggeber:  
*Applicant:* Statens Strålevern  
Norwegian Radiation Protection Authority  
Grini næringspark 13  
  
N-1332 Østerås

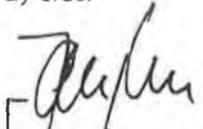
Anzahl der Seiten:  
*Number of pages:* 3

Geschäftszeichen:  
*Reference number:* PTB-6.11-210/30.2001

Kalibrierzeichen:  
*Calibration mark:* PTB-6.11-2001-1190

Datum der Kalibrierung:  
*Date of calibration:* 03.04.2001

Im Auftrag  
*By order*

  
Dr. H. Janßen

Braunschweig, 03.04.2001

Siegel  
*Seal*



Bearbeiter:  
*Examiner:*

  
M. Ehlers

## Physikalisch-Technische Bundesanstalt

Seite 2 zum Kalibrierschein vom 03.04.2001, Kalibrierzeichen: PTB-6.11-2001-1190  
 Page 2 of calibration certificate of 03.04.2001, calibration mark: PTB-6.11-2001-1190

Gegenstand: <i>Object</i>	Aktivitätsnormal
Typ: <i>Type</i>	Wässrige Lösung in Glasampulle
Kennzeichnung: <i>Serial number</i>	2001-1 190
Radionuklid: <i>Radionuclide</i>	Cer-139
Chemische Zusammensetzung der wässrigen Lösung: <i>Chemical composition of the aqueous solution</i>	HCl            1 mol/L CeCl <sub>3</sub> 35 mg/L
Kalibrierverfahren: <i>Method of calibration</i>	Die spezifische Aktivität der Lösung wurde durch Messung der Gammastrahlung mit einer 4π-Ionisationskammer bestimmt. Die 4π-Ionisationskammer ist eine Sekundärnormal-Messeinrichtung, kalibriert mit Aktivitätsnormalen der PTB.
Lösungsmasse: <i>Mass of solution</i>	$m = (2,0612 \pm 0,0017) \text{ g}$
Spezifische Aktivität: <i>Specific activity</i>	$a = (125,0 \pm 1,3) \text{ kBq/g}$
Bezugszeitpunkt: <i>Reference date</i>	01.03.2001, 00:00 Uhr MEZ
Messunsicherheit: <i>Uncertainty of measurement</i>	Angegeben ist die erweiterte Messunsicherheit, die sich aus der Standardmessunsicherheit durch Multiplikation mit dem Erweiterungsfaktor $k = 2$ ergibt. Sie wurde gemäß dem "Guide to the Expression of Uncertainty in Measurement" (ISO, 1995) ermittelt. Der Wert der Messgröße liegt im Regelfall mit einer Wahrscheinlichkeit von annähernd 95 % im zugeordneten Werteintervall.



## Empfohlene Radionukliddaten

Stand: 01.08.1999

Radionuklid: Ce-139

Halbwertszeit:  $T_{1/2} = 137,66(6)$  Tage

### Photonen-Energien $E$ und –Emissionswahrscheinlichkeiten $p$ pro Zerfall:

Strahlungs- typ	$E$ in keV	$p$	Kommentar
La-L	5,0	0,120(6)	
La-K $_{\alpha_2}$	33,03	0,2247(22)	
La-K $_{\alpha_1}$	33,44	0,413(4)	
La-K $_{\beta_1}$	37,8	0,1229(18)	
La-K $_{\beta_2}$	38,7	0,0311(8)	
$\gamma$	165,86	0,7990(4)	

### Meßunsicherheiten:

Die Ziffern in Klammern hinter dem Zahlenwert einer Meßgröße geben die Meßunsicherheit in der(n) letzten Stelle(n) des Zahlenwertes an. Die Meßunsicherheiten entsprechen der einfachen Standardabweichung.

### Literatur:

Schötzig, U. und Schrader, H.:

Halbwertszeiten und Photonen-Emissionswahrscheinlichkeiten von häufig verwendeten Radionukliden; PTB-Bericht PTB-Ra-16/5, Braunschweig 1998, ISBN 3-89701-279-0

PREPARERING AV TRACER-LØSNING

Løpenr. Stam-løsning: 2001-1190

Prosedyre benyttet:

Nuklide:

Halvt.  $T_{1/2}$  i dager:

Ref. dato,  $t_0$ :

Total mengde st.løsning, (g)

Mengde st.løsning veld ut, (g)

Tetthet st.løsning  $g/cm^3$  (se sertifikat):

Aktivitet i ampulle, Bq (se sertifikat):

$D_0$  Aktivitet tatt ut av ampulle, Bq:

Prepareringsdato:

Aktivitet ved prep.dato, Bq:

1. fortynning:

Vekt, tom flaske/kolbe, (g)

Total vekt flaske, med løsn etter fortynning (g)

Mengde løsning tilsatt st.løsning, (g)

Sp.akt. i tracer løsn. etter 1. fortynning, A, (Bq/g):

Sp.akt. i tracer løsn. etter 1. fortynning, A, (Bq/ml):

Nytt løpenr:

Merknad: Kolbe benyttet?

Tracer preparert av (dato, sign.):

Vitne til preparering (dato, sign.):

# PREPARERING AV TRACER-LØSNING

Løpenr. Stam-løsning: 2001-1190

Prosedyre benyttet:

Nuklide:

Halvt.  $T_{1/2}$  dager:

Ref. dato,  $t_0$ :

Total mengde st.løsning, (g)

Mengde st.løsning veid ut, (g):

Tetthet st.løsning  $g/cm^3$  (se sertifikat):

Aktivitet i ampulle, Bq (se sertifikat):

D<sub>0</sub> Aktivitet tatt ut av ampulle, Bq:

Prepareringsdato:

Aktivitet ved prep.dato, Bq:

## 1. fortytning:

Vekt, tom flaske/kolbe, (g):

Total vekt flaske, med løsn. etter fortytning (g):

Mengde løsning tilsatt st.løsning, (g):

Sp.akt. i tracer løsn. etter 1. fortytning, A, (Bq/g):

Sp.akt. i tracer løsn. etter 1. fortytning, A, (Bq/ml):

Nytt løpenr:

Merknad: Kolbe benyttet?

Tracer preparert av (dato, sign.):

Vitne til preparering. (dato, sign.):



## Kalibrierschein

*Calibration certificate*

**Gegenstand:** Aktivitätsnormal  
*Object:*

**Hersteller:** Physikalisch-Technische Bundesanstalt  
*Manufacturer:* Fachlaboratorium 6.11

**Typ:** Wässrige Lösung in Glasampulle  
*Type:*

**Kenn-Nummer:** 12032  
*Serial number:*

**Auftraggeber:** Statens Strålevern  
*Applicant:* Norwegian Radiation Protection Authority  
Grini næringspark 13  
N-1332 Østerås

**Anzahl der Seiten:** 3  
*Number of pages:*

**Geschäftszeichen:** PTB-6.11-210/30.2001  
*Reference number:*

**Kalibrierzeichen:** PTB-6.11-12032  
*Calibration mark:*

**Datum der Kalibrierung:** 21.03.2001  
*Date of calibration:*

**Im Auftrag**  
*By order*

  
Dr. H. Janßen

Braunschweig, 21.03.2001

**Siegel**  
*Seal*



**Bearbeiter:**  
*Examiner:*

  
M. Ehlers

## Physikalisch-Technische Bundesanstalt

Seite 2 zum Kalibrierschein vom 21.03.2001, Kalibrierzeichen: PTB-6.11-12032  
 Page 2 of calibration certificate of 21.03.2001, calibration mark: PTB-6.11-12032

Gegenstand: <i>Object</i>	Aktivitätsnormal
Typ: <i>Type</i>	Wässrige Lösung in Glasampulle
Kennzeichnung: <i>Serial number</i>	12032
Radionuklid: <i>Radionuclide</i>	Caesium-137
Chemische Zusammensetzung der wässrigen Lösung: <i>Chemical composition of the aqueous solution</i>	HCl      0,1 mol/L CsCl     50 mg/L
Kalibrierverfahren: <i>Method of calibration</i>	Die spezifische Aktivität der Lösung wurde durch Messung der Gammastrahlung mit einer $4\pi$ -Ionisationskammer bestimmt. Die $4\pi$ -Ionisationskammer ist eine Sekundärnormal-Messeinrichtung, kalibriert mit Aktivitätsnormalen der PTB.
Lösungsmasse: <i>Mass of solution</i>	$m = (2,0306 \pm 0,0017) \text{ g}$
Spezifische Aktivität: <i>Specific activity</i>	$a = (4,06 \pm 0,06) \text{ kBq/g}$
Bezugszeitpunkt: <i>Reference date</i>	01.03.2001, 00:00 Uhr MEZ
Messunsicherheit: <i>Uncertainty of measurement</i>	Angegeben ist die erweiterte Messunsicherheit, die sich aus der Standardmessunsicherheit durch Multiplikation mit dem Erweiterungsfaktor $k = 2$ ergibt. Sie wurde gemäß dem "Guide to the Expression of Uncertainty in Measurement" (ISO, 1995) ermittelt. Der Wert der Messgröße liegt im Regelfall mit einer Wahrscheinlichkeit von annähernd 95 % im zugeordneten Werteintervall.



## Empfohlene Radionukliddaten

Stand: 01.08.1999

**Radionuklid:** Cs-137

**Halbwertszeit:**  $T_{1/2} = 1,100(9) \cdot 10^4$  Tage [30,13(24) Jahre]

**Photonen-Energien  $E$  und –Emissionswahrscheinlichkeiten  $p$  pro Zerfall:**

Strahlungs- typ	$E$ in keV	$p$	Kommentar
Ba-L	4,7	0,0090(5)	
Ba-K $_{\alpha_2}$	31,82	0,0195(4)	
Ba-K $_{\alpha_1}$	32,19	0,0359(7)	
Ba-K $_{\beta_1}$	36,4	0,01055(22)	
Ba-K $_{\beta_2}$	37,3	0,00266(8)	
Ba-137m	661,66	0,8500(20)	

### Meßunsicherheiten:

Die Ziffern in Klammern hinter dem Zahlenwert einer Meßgröße geben die Meßunsicherheit in der(n) letzten Stelle(n) des Zahlenwertes an. Die Meßunsicherheiten entsprechen der einfachen Standardabweichung.

### Literatur:

Schötzig, U. und Schrader, H.:

Halbwertszeiten und Photonen-Emissionswahrscheinlichkeiten von häufig verwendeten Radionukliden; PTB-Bericht PTB-Ra-16/5, Braunschweig 1998, ISBN 3-89701-279-0



PREPARERING AV TRACER-LØSNING

Løpenr. Stam-løsning: 12032

Prosedyre benyttet:

Nuklide: Cs 137

Halvt.,  $T_{1/2}$  dager:  $1,100 \times 10^4$

Ref. dato,  $t_0$ : 21.03.2001

Total mengde st.løsning, (g)	2,0306 ± 0,0017
Mengde st.løsning veid ut, (g)	1,9668
Tetthet st.løsning g/cm <sup>3</sup> (se sertifikat)	1,00
Aktivitet i ampulle, Bq (se sertifikat)	8244,23
D <sub>0</sub> Aktivitet tatt ut av ampulle, Bq	7985,20 #DIV/0!

Prepareringsdato: 18.04.2001

Aktivitet ved prep dato, Bq

1. fortytning:

Vekt, tom flaske/kolbe, (g)	<input type="text"/>
Total vekt flaske, med løsn etter fortytning (g)	<input type="text"/>
Mengde løsning tilsatt st.løsning, (g)	<input type="text"/>

Sp.akt. i tracer løsn. etter 1. fortytning, A, (Bq/g)	3,992	#VERDI!!
Sp.akt. i tracer løsn. etter 1. fortytning, A, (Bq/ml)	3,992	#VERDI!!

Nytt løpenr:

Merknad: Kolbe benyttet?

Tracer preparert av (dato, sign.):

18/4-01 Bjørn Lind

Vitne til preparering, (dato, sign.):

H. Dordalle - 18.04.2001





## Kalibrierschein

Calibration certificate

Gegenstand:  
*Object:* Aktivitätsnormal

Hersteller:  
*Manufacturer:* Physikalisch-Technische Bundesanstalt  
Fachlaboratorium 6.11

Typ:  
*Type:* Wässrige Lösung in Glasampulle

Kenn-Nummer:  
*Serial number:* 2001-1185

Auftraggeber:  
*Applicant:* Statens Strålevern  
Norwegian Radiation Protection Authority  
Grini næringspark 13  
  
N-1332 Østerås

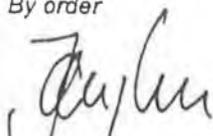
Anzahl der Seiten:  
*Number of pages:* 3

Geschäftszeichen:  
*Reference number:* PTB-6.11-210/30.2001

Kalibrierzeichen:  
*Calibration mark:* PTB-6.11-2001-1185

Datum der Kalibrierung:  
*Date of calibration:* 05.04.2001

Im Auftrag  
*By order*

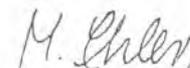
  
Dr. H. Janßen

Braunschweig, 05.04.2001

Siegel  
*Seal*



Bearbeiter:  
*Examiner:*

  
M. Ehlers

Gegenstand:  
*Object*

Aktivitätsnormal

Typ:  
*Type*

Wässrige Lösung in Glasampulle

Kennzeichnung:  
*Serial number*

2001-1185

Radionuklid:  
*Radionuclide*

Cadmium-109

Chemische Zusammensetzung  
der wässrigen Lösung:  
*Chemical composition of the  
aqueous solution*

HCl      0,1 mol/L  
CdCl<sub>2</sub>    45 mg/L

Kalibrierverfahren:  
*Method of calibration*

Die spezifische Aktivität der Lösung wurde durch Messung der Gammastrahlung mit einer 4 $\pi$ -Ionisationskammer bestimmt. Die 4 $\pi$ -Ionisationskammer ist eine Sekundärnormal-Messeinrichtung, kalibriert mit Aktivitätsnormalen der PTB.

Lösungsmasse:  
*Mass of solution*

$m = (2,0016 \pm 0,0017) \text{ g}$

Spezifische Aktivität:  
*Specific activity*

$a = (17,2 \pm 0,3) \text{ kBq/g}$

Bezugszeitpunkt:  
*Reference date*

01.03.2001, 00:00 Uhr MEZ

Messunsicherheit:  
*Uncertainty of measurement*

Angegeben ist die erweiterte Messunsicherheit, die sich aus der Standardmessunsicherheit durch Multiplikation mit dem Erweiterungsfaktor  $k = 2$  ergibt. Sie wurde gemäß dem "Guide to the Expression of Uncertainty in Measurement" (ISO, 1995) ermittelt. Der Wert der Messgröße liegt im Regelfall mit einer Wahrscheinlichkeit von annähernd 95 % im zugeordneten Werteintervall.



## Empfohlene Radionukliddaten

Stand: 01.08.1999

Radionuklid: Cd-109

Halbwertszeit:  $T_{1/2} = 462,1(14)$  Tage

Photonen-Energien  $E$  und –Emissionswahrscheinlichkeiten  $p$  pro Zerfall:

Strahlungs- typ	$E$ in keV	$p$	Kommentar
Ag-L	3,1	0,1034(26)	
Ag-K $_{\alpha_2}$	21,99	0,2899(25)	
Ag-K $_{\alpha_1}$	22,16	0,547(4)	
Ag-K $_{\beta_1}$	24,9	0,1514(18)	
Ag-K $_{\beta_2}$	25,5	0,0263(10)	
$\gamma$	88,03	0,03626(20)	

### Meßunsicherheiten:

Die Ziffern in Klammern hinter dem Zahlenwert einer Meßgröße geben die Meßunsicherheit in der(n) letzten Stelle(n) des Zahlenwertes an. Die Meßunsicherheiten entsprechen der einfachen Standardabweichung.

### Literatur:

Schötzig, U. und Schrader, H.:

Halbwertszeiten und Photonen-Emissionswahrscheinlichkeiten von häufig verwendeten Radionukliden; PTB-Bericht PTB-Ra-16/5, Braunschweig 1998, ISBN 3-89701-279-0

PREPARERING AV TRACER-LØSNING

Løpenr. Stamløsning: 2001-1185

Prosedyre benyttet:

Nuklide:

Halvt.  $T_{1/2}$  dager:

Ref. dato,  $t_0$ :

Total mengde st.løsning, (g)

Mengde st.løsning veid ut, (g)

Tetthet st.løsning g/cm<sup>3</sup> (se sertifikat):

Aktivitet i ampulle, Bq (se sertifikat):

D<sub>0</sub> Aktivitet tatt ut av ampulle, Bq:

Prepareringsdato:

Aktivitet ved prep.dato, Bq:

1. fortynning:

Vekt, tom flaske/kolbe, (g):

Total vekt flaske, med løsn. etter fortynning (g):

Mengde løsning tilsatt st.løsning, (g):

Sp.akt. i tracer løsn. etter 1. fortynning, A, (Bq/g):

Sp.akt. i tracer løsn. etter 1. fortynning, A, (Bq/ml):

Nytt løpenr:

Merknad: Kolbe benyttet?

Tracer preparert av (dato, sign.):

Vitne til preparering (dato, sign.):

# Appendix 1

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## Standard Reference Material Certificates

---

1. NIST supplied Standard Reference Material 4324A: Uranium-232 Radioactivity Standard
2. NIST supplied Standard Reference Material 4328B: Thorium-229 Radioactivity Standard



# National Institute of Standards & Technology Certificate

## Standard Reference Material 4324A Uranium-232 Radioactivity Standard

This Standard Reference Material (SRM) consists of radioactive uranium-232 nitrate and nitric acid dissolved in 5 mL of distilled water. The solution is contained in a flame-sealed NIST borosilicate-glass ampoule. The SRM is intended for the calibration of alpha-particle counting instruments and for the monitoring of radiochemical procedures.

### Radiological Hazard

The SRM ampoule contains uranium-232 with a total activity of approximately 200 Bq. Uranium-232 decays by alpha-particle emission. The progeny of uranium-232 have a total activity of approximately 1400 Bq and decay by alpha- and beta-particle emission. None of the alpha or beta particles escape from the SRM ampoule. During the decay process X-rays and gamma rays with energies from 10 keV to 4.5 MeV are also emitted. Most of these photons escape from the SRM ampoule but their intensities are so small that they do not represent a radiation hazard. Approximate unshielded dose rates at several distances (as of the reference time) are given in note [a]\*. The SRM should be used only by persons qualified to handle radioactive material.

### Chemical Hazard

The SRM ampoule contains nitric acid ( $\text{HNO}_3$ ) with a concentration of 2 moles per liter of water. The solution is corrosive and represents a health hazard if it comes in contact with eyes or skin. If the ampoule is to be opened to transfer the solution, the recommended procedure is given on page 2. The ampoule should be opened only by persons qualified to handle both radioactive material and strong acid solution.

### Storage and Handling

The SRM should be stored and used at a temperature between 5 and 65 °C. The solution in an unopened ampoule should remain stable and homogeneous until at least November 2003.

The ampoule (or any subsequent container) should always be clearly marked as containing radioactive material. If the ampoule is transported it should be packed, marked, labeled, and shipped in accordance with the applicable national, international, and carrier regulations. The solution in the ampoule is a dangerous good (hazardous material) both because of the radioactivity and because of the strong acid.

### Preparation

This Standard Reference Material was prepared in the Physics Laboratory, Ionizing Radiation Division, Radioactivity Group, J.M.R. Hutchinson, Group Leader. The overall technical direction and physical measurements leading to certification were provided by L.L. Lucas of the Radioactivity Group.

The support aspects involved in the preparation, certification, and issuance of this SRM were coordinated through the Standard Reference Materials Program by N.M. Trahey.

Gaithersburg, Maryland 20899  
June 1995

Thomas E. Gills, Chief  
Standard Reference Materials Program

### Recommended Procedure for Opening the SRM Ampoule

- 1) If the SRM solution is to be diluted, it is recommended that the diluting solution have a composition comparable to that of the SRM solution.
- 2) Wear eye protection, gloves, and protective clothing and work over a tray with absorbent paper in it. Work in a fume hood. In addition to the radioactive material, the solution contains strong acid and is corrosive.
- 3) Shake the ampoule to wet all of the inside surface of the ampoule. Return the ampoule to the upright position.
- 4) Check that all of the liquid has drained out of the neck of the ampoule. If necessary, gently tap the neck to speed the process.
- 5) Holding the ampoule upright, score the narrowest part of the neck with a scribe or diamond pencil.
- 6) Lightly wet the scored line. This reduces the crack propagation velocity makes for a cleaner break.
- 7) Hold the ampoule upright with a paper towel, a wiper, or a support jig. Position the scored line away from you. Using a paper towel or wiper to avoid contamination, snap off the top of the ampoule by pressing the narrowest part of the neck away from you while pulling the tip of the ampoule towards you.
- 8) Transfer the solution from the ampoule using a pycnometer or a pipet with dispenser handle. NEVER PIPETTE BY MOUTH.
- 9) Seal any unused SRM solution in a flame-sealed glass ampoule, if possible, to minimize the evaporation loss

See also reference [4]\*.

PROPERTIES OF SRM 4324A  
(Certified values are shown in bold type)

Source identification number	NIST SRM 4324A		
Physical Properties:			
Source description	Liquid in flame-sealed NIST borosilicate-glass ampoule		
Ampoule specifications	Body outside diameter	(16.5 ± 0.5) mm	
	Wall thickness	(0.60 ± 0.04) mm	
	Barium content	Less than 2.5 %	
	Lead-oxide content	Less than 0.02 %	
	Other heavy elements	Trace quantities	
<b>Solution density</b>	<b>(1.062 ± 0.002) g·mL<sup>-1</sup> at 22.5 °C [b]*</b>		
Solution mass	Approximately 5.3 g		
Chemical Properties:			
Solution composition	Chemical Formula	Concentration (mol·L <sup>-1</sup> )	Mass Fraction (g·g <sup>-1</sup> )
	H <sub>2</sub> O	52	0.88
	HNO <sub>3</sub>	2.0	0.12
	UO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub>	2 × 10 <sup>-10</sup>	8 × 10 <sup>-11</sup>
Radiological Properties:			
<b>Radionuclide</b>	<b>Uranium-232</b>		
<b>Reference time (Separation time)</b>	<b>1330 EST, 6 November 1993</b>		
<b>Massic activity of the solution [c]</b>	<b>39.23 Bq·g<sup>-1</sup></b>		
<b>Relative expanded uncertainty (k=2)</b>	<b>0.50% [d]</b>		
Alpha-particle-emitting impurities	None detected [e]		
Photon-emitting impurities	None detected [f]		
Half lives used	Uranium 232: (68.9 ± 0.4) a [g] Thorium-228: (1.9131 ± 0.0009) a Radium-224: (3.66 ± 0.04) d Radon-220: (55.6 ± 0.1) s Polonium-226: (0.145 ± 0.002) s Lead-212: (10.64 ± 0.01) h Bismuth-212: (60.55 ± 0.06) m Thallium-208: (3.053 ± 0.004) m		
Measuring instruments	Two 4π(α+β) liquid-scintillation counting systems		

## EVALUATION OF THE UNCERTAINTY OF THE MASSIC ACTIVITY [d]\*

Input Quantity $x_i$ , the source of uncertainty	Method Used To Evaluate $u(x_i)$ , the standard uncertainty of $x_i$	Relative Uncertainty Of Input Quantity, $u(x_i)/x_i$ , (%) [h]	Relative Sensitivity Factor, $ \partial y/\partial x_i  \cdot$ ( $x_i/y$ ) [i]	Relative Uncertainty of Output Quantity, $u_i(y)/y$ , (%) [j]
Uncertainties Evaluated By Statistical Methods				
Massic alpha-particle emission rate	Standard deviation of the mean for 10 sets of $4\pi(\alpha+\beta)$ liquid-scintillation measurements	0.02	1.0	0.02
Background	Standard deviation of the mean for 10 sets of measurements	3.5	[k] 0.002	0.01
Uncertainties Evaluated By Other Means				
Mass calibration of the balance	Estimated from manufacturer's data	0.05	1.0	0.05
Decay correction for uranium-232	Standard uncertainty of the half life	0.58 [m]	0.0006 [n]	0.0004
Decay-scheme data	Standard uncertainty of the probability of decay by alpha-particle emission	0.01	1.0	0.01
Correction for ingrowth of daughter radionuclides	Estimated	0.02	1.0	0.02
Live-time [p]	Estimated	0.10	1.0	0.10
Detection efficiency of the liquid-scintillation counting systems	Estimated	0.20	1.0	0.20
Alpha-particle-emitting impurities	Limit of detection [q]	100.	0.001	0.10
Photon-emitting impurities	Limit of detection [q]	100.	0.0001	0.01
Relative Combined Standard Uncertainty of the Output Quantity, $u_c(y)/y$ , (%)				0.25
Coverage Factor, $k$				$\times 2$
Relative Expanded Uncertainty of the Output Quantity, $U/y$ , (%)				0.50

## NOTES

- [a] The Sievert is the SI unit for dose equivalent. See reference [1]. One  $\mu\text{Sv}$  is equal to 0.1 mrem.

Distance from Ampoule (cm):	1	30	100
Approximate Dose Rate ( $\mu\text{Sv/h}$ ):	< 0.1	-	-

- [b] The stated uncertainty is two times the standard uncertainty.

- [c] **Massic activity** is the preferred name for the quantity activity per unit mass. See reference [1].

- [d] The reported value,  $y$ , of massic activity (activity per unit mass) at the reference time was not measured directly but was derived from measurements and calculations of other quantities. This can be expressed as  $y = f(x_1, x_2, x_3, \dots, x_n)$ , where  $f$  is a mathematical function derived from the assumed model of the measurement process.

The value,  $x_i$ , used for each input quantity  $i$  has a **standard uncertainty**,  $u(x_i)$ , that generates a corresponding uncertainty in  $y$ ,  $u_i(y) = |\partial y / \partial x_i| \cdot u(x_i)$ , called a **component of combined standard uncertainty** of  $y$ .

The **combined standard uncertainty** of  $y$ ,  $u_c(y)$ , is the positive square root of the sum of the squares of the components of combined standard uncertainty.

The combined standard uncertainty is multiplied by a **coverage factor** of  $k = 2$  to obtain  $U$ , the **expanded uncertainty** of  $y$ .

Since it can be assumed that the possible estimated values of the massic activity are approximately normally distributed with approximate standard deviation  $u_c(y)$ , the unknown value of the massic activity is believed to lie in the interval  $y \pm U$  with a level of confidence of approximately 95 percent.

For further information on the expression of uncertainties, see references [2] and [3].

- [e] Estimated limits of detection for alpha-particle-emitting impurities are:

0.04	$\alpha \cdot \text{s}^{-1} \cdot \text{g}^{-1}$ for energies less than 3.9 MeV,
0.4	$\alpha \cdot \text{s}^{-1} \cdot \text{g}^{-1}$ for energies between 3.9 MeV and 4.9 MeV, and
0.04	$\alpha \cdot \text{s}^{-1} \cdot \text{g}^{-1}$ for energies greater than 5.6 MeV.

- [f] Estimated limits of detection for photon-emitting impurities are:

0.013	$\gamma \cdot \text{s}^{-1} \cdot \text{g}^{-1}$ for energies between 9 and 125 keV,
0.009	$\gamma \cdot \text{s}^{-1} \cdot \text{g}^{-1}$ for energies between 135 and 234 keV,
0.004	$\gamma \cdot \text{s}^{-1} \cdot \text{g}^{-1}$ for energies between 243 and 579 keV,
0.002	$\gamma \cdot \text{s}^{-1} \cdot \text{g}^{-1}$ for energies between 588 and 1616 keV, and
0.001	$\gamma \cdot \text{s}^{-1} \cdot \text{g}^{-1}$ for energies between 1624 and 1900 keV,

provided that the photons are separated in energy by 4 keV or more from photons emitted in the decay of uranium-232 and progeny.

- [g] Evaluated Nuclear Structure Data File (ENSDF), June 1995. The stated uncertainty is the standard uncertainty.

- [h] Relative standard uncertainty of the input quantity  $x_i$ .

- [i] The relative change in the output quantity  $y$  divided by the relative change in input quantity  $x_i$ . If  $|\partial y/\partial x_i| \cdot (x_i/y) = 1.0$ , then a 1% change in  $x_i$  results in a 1% change in  $y$ . If  $|\partial y/\partial x_i| \cdot (x_i/y) = 0.05$ , then a 1% change in  $x_i$  results in a 0.05% change in  $y$ .
- [j] Relative component of combined standard uncertainty of output quantity  $y$ , rounded to two significant figures or less. The relative component of combined standard uncertainty of  $y$  is given by  $u_i(y)/y = |\partial y/\partial x_i| \cdot (x_i/y) = |\partial y/\partial x_i| \cdot (x_i/y) \cdot u(x_i)/x_i$ . The numerical values of  $u(x_i)/x_i$ ,  $|\partial y/\partial x_i| \cdot (x_i/y)$ , and  $u_i(y)/y$ , all dimensionless quantities, are listed in columns 3, 4, and 5, respectively. Thus, the value in column 5 is equal to the value in column 4 multiplied by the value in column 3. The input quantities are independent, or very nearly so. Hence the covariances are zero or negligible.
- [k]  $|\partial y/\partial x_i| \cdot (x_i/y) = (\text{average background count rate})/(\text{average net sample count rate})$ .
- [m] The relative standard uncertainty of  $\lambda \cdot t$  is determined by the relative standard uncertainty of  $\lambda$  (i.e., of the half life). The relative standard uncertainty of  $t$  is negligible.
- [n]  $|\partial y/\partial x_i| \cdot (x_i/y) = |\lambda \cdot t|$
- [p] The live time is determined by counting the pulses from a gated oscillator.
- [q] The standard uncertainty for each undetected impurity that might reasonably be expected to be present is estimated to be equal to the estimated limit of detection for that impurity, i.e.  $u(x_i)/x_i = 100\%$ .  $|\partial y/\partial x_i| \cdot (x_i/y) = \{(\text{response per Bq of impurity})/(\text{response per Bq of U-232})\} \cdot \{(\text{Bq of impurity})/(\text{Bq of U-232})\}$ . Thus  $u_i(y)/y$  is the relative change in  $y$  if the impurity were present with a massic activity equal to the estimated limit of detection.

#### REFERENCES

- [1] International Organization for Standardization (ISO), *ISO Standards Handbook - Quantities and Units*, 1993. Available from the American National Standards Institute, 11 West 42nd Street, New York, NY 10036, U.S.A. 1-212-642-4900.
- [2] International Organization for Standardization (ISO), *Guide to the Expression of Uncertainty in Measurement*, 1993. Available from the American National Standards Institute, 11 West 42nd Street, New York, NY 10036, U.S.A. 1-212-642-4900. (Listed under ISO miscellaneous publications as "ISO Guide to the Expression 1993".)
- [3] B. N. Taylor and C. E. Kuyatt, *Guidelines for Evaluating and Expressing the Uncertainty of NIST Measurement Results*, NIST Technical Note 1297, 1993. Available from the Superintendent of Documents, U.S. Government Printing Office, Washington, DC 20407, U.S.A.
- [4] National Council on Radiation Protection and Measurements Report No. 58, *A Handbook of Radioactivity Measurements Procedures*, Second Edition, 1985. Available from the National Council on Radiation Protection and Measurements, 7910 Woodmont Avenue, Bethesda, MD 20814 U.S.A.



# National Institute of Standards & Technology Certificate

## Standard Reference Material 4328B Thorium-229 Radioactivity Standard

This Standard Reference Material (SRM) consists of radioactive thorium-229 nitrate and nitric acid dissolved in 5 mL of distilled water. The solution is contained in a flame-sealed NIST borosilicate-glass ampoule. The SRM is intended for the calibration of alpha-particle counting instruments and for the monitoring of radiochemical procedures.

### Radiological Hazard

The SRM ampoule contains thorium-229 with a total activity of approximately 170 Bq. Thorium-229 decays by alpha-particle emission. The progeny of thorium-229 have a total activity of approximately 1200 Bq and decay by alpha- and beta-particle emission. None of the alpha or beta particles escape from the SRM ampoule. During the decay process X-rays and gamma rays with energies from 10 keV to 2 MeV are also emitted. Most of these photons escape from the SRM ampoule but their intensities are so small that they do not represent a radiation hazard. Approximate unshielded dose rates at several distances (as of the reference time) are given in note [a]\*. The SRM should be used only by persons qualified to handle radioactive material.

### Chemical Hazard

The SRM ampoule contains nitric acid ( $\text{HNO}_3$ ) with a concentration of 1 mole per liter of water. The solution is corrosive and represents a health hazard if it comes in contact with eyes or skin. If the ampoule is to be opened to transfer the solution, the recommended procedure is given on page 2. The ampoule should be opened only by persons qualified to handle both radioactive material and strong acid solution.

### Storage and Handling

The SRM should be stored and used at a temperature between 5 and 65 °C. The solution in an unopened ampoule should remain stable and homogeneous until at least July 2005.

The ampoule (or any subsequent container) should always be clearly marked as containing radioactive material. If the ampoule is transported it should be packed, marked, labeled, and shipped in accordance with the applicable national, international, and carrier regulations. The solution in the ampoule is a dangerous good (hazardous material) both because of the radioactivity and because of the strong acid.

### Preparation

This Standard Reference Material was prepared in the Physics Laboratory, Ionizing Radiation Division, Radioactivity Group, J.M.R. Hutchinson, Group Leader. The overall technical direction and physical measurements leading to certification were provided by L.L. Lucas of the Radioactivity Group.

The support aspects involved in the preparation, certification, and issuance of this SRM were coordinated through the Standard Reference Materials Program by N.M. Trahey.

Gaithersburg, Maryland 20899  
July 1996

Thomas E. Gills, Chief  
Standard Reference Materials Program

### Recommended Procedure for Opening the SRM Ampoule

- 10) If the SRM solution is to be diluted, it is recommended that the diluting solution have a composition comparable to that of the SRM solution.
- 11) Wear eye protection, gloves, and protective clothing and work over a tray with absorbent paper in it. Work in a fume hood. In addition to the radioactive material, the solution contains strong acid and is corrosive.
- 12) Shake the ampoule to wet all of the inside surface of the ampoule. Return the ampoule to the upright position.
- 13) Check that all of the liquid has drained out of the neck of the ampoule. If necessary, gently tap the neck to speed the process.
- 14) Holding the ampoule upright, score the narrowest part of the neck with a scribe or diamond pencil.
- 15) Lightly wet the scored line. This reduces the crack propagation velocity makes for a cleaner break.
- 16) Hold the ampoule upright with a paper towel, a wiper, or a support jig. Position the scored line away from you. Using a paper towel or wiper to avoid contamination, snap off the top of the ampoule by pressing the narrowest part of the neck away from you while pulling the tip of the ampoule towards you.
- 17) Transfer the solution from the ampoule using a pycnometer or a pipet with dispenser handle. NEVER PIPETTE BY MOUTH.
- 18) Seal any unused SRM solution in a flame-sealed glass ampoule, if possible, to minimize the evaporation loss

See also reference [4]\*.

PROPERTIES OF SRM 4328B  
(Certified values are shown in bold type)

Source identification number	NIST SRM 4328B		
Physical Properties:			
Source description	Liquid in flame-sealed NIST borosilicate-glass ampoule		
Ampoule specifications	Body outside diameter	(16.5 ± 0.5) mm	
	Wall thickness	(0.60 ± 0.04) mm	
	Barium content	Less than 2.5 %	
	Lead-oxide content	Less than 0.02 %	
	Other heavy elements	Trace quantities	
Solution density	<b>(1.036 ± 0.002) g·mL<sup>-1</sup> at 20 °C [b]*</b>		
Solution mass	<b>(5.174 ± 0.002) g [b]</b>		
Chemical Properties:			
Solution composition	Chemical Formula	Concentration (mol·L <sup>-1</sup> )	Mass Fraction (g·g <sup>-1</sup> )
	H <sub>2</sub> O	54	0.93
	HNO <sub>3</sub>	1.1	0.07
	<sup>229</sup> Th(NO <sub>3</sub> ) <sub>4</sub>	2 × 10 <sup>-8</sup>	1 × 10 <sup>-8</sup>
Radiological Properties:			
Radionuclide	Thorium-229		
Reference time (Separation time)	1200 EST, 1 July 1996 [c]		
Massic activity of the solution [d]	<b>33.36 Bq·g<sup>-1</sup></b>		
Relative expanded uncertainty ( <i>k</i> = 2)	<b>0.66% [e] [f]</b>		
Daughter radionuclides	Approximately in equilibrium [c]		
Alpha-particle-emitting impurities	None detected [g]		
Photon-emitting impurities	None detected [h]		
Half lives used in the decay correction	Thorium-229: (7880 ± 120) a [i]		
Measuring instruments	NIST "0.8π" α defined-solid-angle counter with scintillation detector plus silicon surface-barrier alpha-spectrometer system and a 4π(α+β) liquid-scintillation counting system		

## EVALUATION OF THE UNCERTAINTY OF THE MASSIC ACTIVITY [e] [f]\*

Input Quantity $x_i$ , the source of uncertainty  (and individual uncertainty components where appropriate)	Method Used To Evaluate $u(x_i)$ , the standard uncertainty of $x_i$  (A) denotes evaluation by statistical methods (B) denotes evaluation by other methods	Relative Uncertainty Of Input Quantity, $u(x_i)/x_i$ , (%) [j]	Relative Sensitivity Factor, $ \partial y/\partial x_i  \cdot$ $(x_i/y)$ [k]	Relative Uncertainty of Output Quantity, $u_i(y)/y$ , (%) [m]
Massic alpha-particle emission rate, corrected for background and decay	Standard deviation of the mean for one "0.8π" α plus alpha-spectrometer measurement and four liquid-scintillation measurements (A)	0.06	1.0	0.06
Decay correction for thorium-229	Standard uncertainty of the half life (A)	1.52 [n]	0.001 [p]	0.002
Decay-scheme data	Standard uncertainty of the probability of decay by alpha-particle emission (A)	0.01	1.0	0.01
Extrapolation of alpha-particle-count-rate-versus-energy to zero energy	Estimated (B)	0.25	1.0	0.25
Gravimetric measurements	Estimated (B)	0.10	1.0	0.10
Live-time [q]	Estimated (B)	0.10	1.0	0.10
Alpha-particle detection efficiency of scintillators	Estimated (B)	0.10	1.0	0.10
Geometry of 0.8π α and surface-barrier detectors	Estimated (B)	0.25	0.2	0.05
Alpha-particle-emitting impurities	Limit of detection (B) [r]	100.	0.001	0.10
Photon-emitting impurities	Limit of detection (B) [r]	100.	0.0001	0.01
Relative Combined Standard Uncertainty of the Output Quantity, $u_c(y)/y$ , (%)				0.33
Coverage Factor, $k$				$\times 2$
Relative Expanded Uncertainty of the Output Quantity, $U/y$ , (%)				0.66

## NOTES

- [l] The Sievert is the SI unit for dose equivalent. See reference [1]. One  $\mu\text{Sv}$  is equal to 0.1 mrem.  
 Distance from Ampoule (cm):           1       30       100  
 Approximate Dose Rate ( $\mu\text{Sv/h}$ ):       < 0.1       -       -
- [m] The stated uncertainty is two times the standard uncertainty.
- [n] The thorium-229 master solution was chemically purified on 7 May 1984. The daughter radionuclides have been growing in since that time.
- [o] **Massic activity** is the preferred name for the quantity activity divided by the total mass of the sample. See reference [1].
- [p] The reported value,  $y$ , of massic activity (activity per unit mass) at the reference time was not measured directly but was derived from measurements and calculations of other quantities. This can be expressed as  $y = f(x_1, x_2, x_3, \dots, x_n)$ , where  $f$  is a mathematical function derived from the assumed model of the measurement process.

The value,  $x_i$ , used for each input quantity  $i$  has a **standard uncertainty**,  $u(x_i)$ , that generates a corresponding uncertainty in  $y$ ,  $u_i(y) = |\partial y / \partial x_i| \cdot u(x_i)$ , called a **component of combined standard uncertainty** of  $y$ .

The **combined standard uncertainty** of  $y$ ,  $u_c(y)$ , is the positive square root of the sum of the squares of the components of combined standard uncertainty.

The combined standard uncertainty is multiplied by a **coverage factor** of  $k = 2$  to obtain  $U$ , the **expanded uncertainty** of  $y$ .

Since it can be assumed that the possible estimated values of the massic activity are approximately normally distributed with approximate standard deviation  $u_c(y)$ , the unknown value of the massic activity is believed to lie in the interval  $y \pm U$  with a level of confidence of approximately 95 percent.

For further information on the expression of uncertainties, see references [2] and [3].

- [q] The value of each standard uncertainty component, and hence the value of the expanded uncertainty itself, is a best estimate based upon all available information, but is only approximately known. That is to say, the "uncertainty of the uncertainty" is large and not well known. This is true for uncertainties evaluated by statistical methods (e.g., the relative standard deviation of the standard deviation of the mean for the massic count rate is approximately 50%) and for the uncertainties evaluated by other methods (which could easily be over estimated or under estimated by substantial amounts). The unknown value of the expanded uncertainty is believed to lie in the interval  $U/2$  to  $2U$  (i.e., within a factor of 2 of the estimated value).
- [r] Estimated limits of detection for alpha-particle-emitting impurities are:  
 0.03  $\alpha \cdot \text{s}^{-1} \cdot \text{g}^{-1}$  for energies less than 3.5 MeV,  
 0.3  $\alpha \cdot \text{s}^{-1} \cdot \text{g}^{-1}$  for energies between 3.5 MeV and 4.5 MeV, and  
 0.03  $\alpha \cdot \text{s}^{-1} \cdot \text{g}^{-1}$  for energies greater than 5.1 MeV.  
 In particular, the massic activity of thorium-228 is less than  $0.001 \text{ Bq} \cdot \text{g}^{-1}$ .
- [s] Estimated limits of detection for photon-emitting impurities are:  
 0.003  $\gamma \cdot \text{s}^{-1} \cdot \text{g}^{-1}$  for energies between 90 and 506 keV,  
 0.0003  $\gamma \cdot \text{s}^{-1} \cdot \text{g}^{-1}$  for energies between 516 and 1900 keV,  
 provided that the photons are separated in energy by 4 keV or more from photons emitted in the decay of thorium-229 and progeny.

- [t] The stated uncertainty is the standard uncertainty. See reference [5].
- [u] Relative standard uncertainty of the input quantity  $x_i$ .
- [v] The relative change in the output quantity  $y$  divided by the relative change in input quantity  $x_i$ . If  $|\partial y/\partial x_i| \cdot (x_i/y) = 1.0$ , then a 1% change in  $x_i$  results in a 1% change in  $y$ . If  $|\partial y/\partial x_i| \cdot (x_i/y) = 0.05$ , then a 1% change in  $x_i$  results in a 0.05% change in  $y$ .
- [m] Relative component of combined standard uncertainty of output quantity  $y$ , rounded to two significant figures or less. The relative component of combined standard uncertainty of  $y$  is given by  $u_i(y)/y = |\partial y/\partial x_i| \cdot u(x_i)/y = |\partial y/\partial x_i| \cdot (x_i/y) \cdot u(x_i)/x_i$ . The numerical values of  $u(x_i)/x_i$ ,  $|\partial y/\partial x_i| \cdot (x_i/y)$ , and  $u_i(y)/y$ , all dimensionless quantities, are listed in columns 3, 4, and 5, respectively. Thus, the value in column 5 is equal to the value in column 4 multiplied by the value in column 3. The input quantities are independent, or very nearly so. Hence the covariances are zero or negligible.
- [n] The relative standard uncertainty of  $\lambda \cdot t$  is determined by the relative standard uncertainty of  $\lambda$  (i.e., of the half life). The relative standard uncertainty of  $t$  is negligible.
- [o]  $|\partial y/\partial x_i| \cdot (x_i/y) = |\lambda \cdot t|$
- [r] The live time is determined by counting the pulses from a gated oscillator.
- [s] The standard uncertainty for each undetected impurity that might reasonably be expected to be present is estimated to be equal to the estimated limit of detection for that impurity, i.e.  $u(x_i)/x_i = 100\% \cdot |\partial y/\partial x_i| \cdot (x_i/y) = \{(\text{response per Bq of impurity})/(\text{response per Bq of Th-229})\} \cdot \{(\text{Bq of impurity})/(\text{Bq of Th-229})\}$ . Thus  $u_i(y)/y$  is the relative change in  $y$  if the impurity were present with a massic activity equal to the estimated limit of detection.

#### REFERENCES

- [5] International Organization for Standardization (ISO), *ISO Standards Handbook - Quantities and Units*, 1993. Available from the American National Standards Institute, 11 West 42nd Street, New York, NY 10036, U.S.A. 1-212-642-4900.
- [6] International Organization for Standardization (ISO), *Guide to the Expression of Uncertainty in Measurement*, 1993. Available from the American National Standards Institute, 11 West 42nd Street, New York, NY 10036, U.S.A. 1-212-642-4900. (Listed under ISO miscellaneous publications as "ISO Guide to the Expression 1993".)
- [7] B. N. Taylor and C. E. Kuyatt, *Guidelines for Evaluating and Expressing the Uncertainty of NIST Measurement Results*, NIST Technical Note 1297, 1993. Available from the Superintendent of Documents, U.S. Government Printing Office, Washington, DC 20407, U.S.A.
- [8] National Council on Radiation Protection and Measurements Report No. 58, *A Handbook of Radioactivity Measurements Procedures*, Second Edition, 1985. Available from the National Council on Radiation Protection and Measurements, 7910 Woodmont Avenue, Bethesda, MD 20814 U.S.A.
- [9] S.J. Goldstein, M.T. Murrell, and R.W. Williams, *Physical Review C*, 40 (1989) 6.

# CERTIFICATE OF CALIBRATION

## Standard Radionuclide Source

67990-121

24.1 mm Diameter x 0.65 mm Thick Stainless Steel Disk

This standard radionuclide source was prepared by electrodeposition of a mixture of alpha emitters onto a stainless steel disk. Total alpha activity was determined with a ZnS scintillation detector. Radionuclide activities were calculated from the total activity and the fraction of activity for each radionuclide determined by alpha spectroscopy.

Analytics maintains traceability to the National Institute of Standards and Technology through Measurements Assurance Programs as described in USNRC Regulatory Guide 4.15, Rev. 1.

CALIBRATION DATE: May 1, 2004 12:00 EST

TOTAL ACTIVITY (dpm): 411

RELATIVE EXPANDED UNCERTAINTY (dpm [k=2]): 12

ENERGY RANGE (keV): 3700-7950

ISOTOPE	ACTIVITY (dpm)	HALF-LIFE	RELATIVE EXPANDED UNCERTAINTY (dpm [k=2])	ENERGY RANGE (keV)
U-238	101	4.468 E9 y	3.0	3900-4290
U-234	100	2.455 E5 y	3.0	4580-4860
Pu-239	101	2.41 E4 y	3.0	4950-5240
Am-241	101	4.322 E2 y	3.0	5275-6690

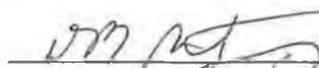
Impurities: U-235 ≈4.2 dpm

Diameter of active area: 24.1 mm

CAUTION: Active material deposited on the unmarked surface. Handle carefully to prevent scratching or damaging the active surface of this source (i.e., use Teflon coated forceps). Store in the container provided when not in use.

PO NUMBER 201327, Item 1 (7400-SRC)

SOURCE CALIBRATED BY:

  
 Daniel M. Montgomery, Radiochemist

Q A APPROVED:

 06-09-04

## **APPENDIX 3**



## **APPENDIX 4**

## Soil-to-plant transfer of $^{137}\text{Cs}$ and $^{40}\text{K}$ in an Atlantic blanket bog ecosystem

C. Moran-Hunter, J. O'Dea

Institute of Technology Sligo, Ballinade, Sligo, Ireland

(Received June 19, 2007)

The transfer of  $^{137}\text{Cs}$  and  $^{40}\text{K}$  from soil to vegetation was studied in an Atlantic blanket bog ecosystem along the Atlantic coast of Ireland where the dominant vegetation is a mixture of *Calluna vulgaris*, *Eriophorum vaginatum* and *Sphagnum* mosses. The impact of soil chemistry and nutritional status of vegetation on the uptake of both radionuclides was also examined. Cesium-137 transfer factors values ranged from 1.9 to 9.6 and accumulation of  $^{137}\text{Cs}$  was higher in the leaves of *C. vulgaris* than in the stems. Transfer factors values for  $^{137}\text{Cs}$  in both *C. vulgaris* and *E. vaginatum* were similar indicating that for the vegetation studied, uptake is not dependent on plant species. The uptake of  $^{137}\text{Cs}$  in bog vegetation was found to be positively correlated with the nutrient status of vegetation, in particular the secondary nutrients, calcium and magnesium. Potassium-40 transfer factors ranged from 0.9 to 13.8 and uptake was higher in *E. vaginatum* than in *C. vulgaris*, however, unlike  $^{137}\text{Cs}$ , the concentrations of  $^{40}\text{K}$  within the leaves and stems of *C. vulgaris* were similar. The concentration of both  $^{137}\text{Cs}$  and  $^{40}\text{K}$  found in moss samples were in general lower than those found in vascular plants.

### Introduction

The long term behavior of natural and anthropogenic radionuclides in terrestrial ecosystems is of major concern due to their chemical and radiological toxicities and the potential for the entry and accumulation of these toxicities into the human food chain. A focus of environmental research post Chernobyl has been on the fate of radionuclides, particularly  $^{137}\text{Cs}$ , in terrestrial ecosystems of Europe. Cesium-137 is an anthropogenic radionuclide which was introduced into the environment in the form of atmospheric deposition from nuclear weapons testing and nuclear accidents. Environmental concerns have arisen due to  $^{137}\text{Cs}$  relatively long half-life (30 years) and the ease with which cesium, which has no known biological role, enters biological systems.<sup>1</sup>

Radiocesium bioavailability within terrestrial ecosystems is dependent upon soil properties and in particular upon the clay content, organic matter content and the K status of the soil.<sup>2,3</sup> Cesium forms a specific and almost irreversible attachment to clay minerals, notably the 2:1 mineral illite,<sup>4</sup> resulting in diminished soil-to-plant transfer of  $^{137}\text{Cs}$  in soils with high clay content which is well documented within the literature.<sup>5–7</sup> On the other hand, organic matter only forms a weak bond with  $^{137}\text{Cs}$  in the soil, thereby leaving it available for uptake by vegetation and it also reduces the affinity of clay minerals for  $^{137}\text{Cs}$  thus further increasing its plant availability.<sup>2,5,8,9</sup> The nutrient status of soil, in particular the potassium content, has a significant effect on plant uptake of  $^{137}\text{Cs}$  as both ions are chemically similar, however they do not necessarily behave in an analogous manner.<sup>6,10,11</sup>

The bogs and heathlands of Europe were significantly affected by Chernobyl radioactivity as

deposition tended to be higher in upland bogs compared to lowland agricultural areas and the soil characteristics of peatlands (high organic matter content and low nutrient status) has resulted in  $^{137}\text{Cs}$  remaining persistently available for plant uptake. High levels of  $^{137}\text{Cs}$  has been recorded for vegetation growing in upland bogs,<sup>12,13</sup> but limited information is available on the fate of  $^{137}\text{Cs}$  within other internationally recognized peatland ecosystems in particular the Atlantic blanket bogs (lowland blanket bog) and raised bogs of Europe. These three ecosystems (upland blanket bog, Atlantic blanket bog and raised bog) are commonly found in Northern Europe and in particular along the western seaboard of Scotland, England, Wales and Ireland. In Ireland, these ecosystems cover approximately 1/6th the landmass of the country. The vegetation indicative of these ecosystems are also found in other regions across the world. For example *Calluna vulgaris* commonly known as ling heather is found abundantly within upland blanket bogs and Atlantic blanket bogs in the British Isles, but is also native over the greater part of Europe and along the Atlantic coast of North America.<sup>14</sup> High levels of  $^{137}\text{Cs}$  in *Calluna vulgaris* has been documented in literature for both pre- and post-Chernobyl.<sup>12,13,15–17</sup> As *C. vulgaris* forms part of the dietary intake of sheep and hill cattle the  $^{137}\text{Cs}$  it contains, can easily be transferred into the human food chain.

The present study was undertaken in order to examine soil-to-plant transfer of  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in an Atlantic blanket bog. A secondary objective was to assess the influence of soil characteristics and the nutritional status of vegetation on the uptake of both radionuclides as limited information is available on the fate of these radionuclides within this particular ecosystem and in organic soils in general.

\* E-mail: moran.carmel@itsligo.ie

## Experimental

### Site selection

The sampling site defined as an Atlantic blanket bog<sup>18,19</sup> is located north west of Sligo town on the western seaboard of Ireland (54° 24.9' N, 08° 24.7' W, 50 m above sea level). The underlying geology of the region is lower and middle carboniferous, mostly limestone. The bog consists of both undisturbed peat and cutover bog (area used for turf extraction) and the undisturbed peat, which is the region of interest within this study, covers an area of approximately 1 hectare. Dominant vegetation within this region is a mixture of *Calluna vulgaris* (Ling Heather), *Eriophorum vaginatum* (Bog Cotton) and *Sphagnum* mosses.

### Sampling and sample preparation

Seven sites (1–7) were sampled within the undisturbed region of the bog during the summer months of 2002. At each site, an area of 1 m<sup>2</sup> was marked out and portions of above ground vegetation (*C. vulgaris* leaves and stems, *E. vaginatum* and *Sphagnum* mosses) were collected approximately 5 cm above the ground to minimize soil adhesion.

Vegetation samples were sealed in polythene bags before being transported to the laboratory. Soil samples were taken at each site directly below the sampled vegetation using a stainless steel shovel from a defined area, typically 60 cm × 60 cm<sup>2</sup> and down to a depth of 20 cm. These soil blocks were then divided into four layers (0–5 cm, 5–10 cm, 10–15 cm and 15–20 cm) and placed in sealed polythene bags to minimize moisture loss before being transported to the laboratory. At Site 3, soil was sampled to a depth of 50 cm and then sectioned into ten layers, each 5 cm in depth, to obtain information on the vertical distribution of  $^{137}\text{Cs}$  in the soil profile.

In the laboratory, vegetation was thoroughly washed in distilled water to remove adhering soil and debris and dried at 105° C for 24 hours.<sup>20</sup> *Calluna vulgaris* was separated into leaves and stems before washing and drying and both plant compartments were analyzed separately. Soils were dried at 35° C (exchangeable nutrients unaffected at temperatures <40° C) until a constant weight was achieved.<sup>21</sup> Root material was removed from the dried soils before grinding in a stainless steel blender and passing through a sieve of 2 mm aperture. Soil and vegetation samples were packed into 1-liter Marinelli and 200-ml tubs for gamma-spectrometric analysis.

### Radiometric analysis

Cesium-137 and  $^{40}\text{K}$  were determined by direct gamma-spectrometry using a high purity germanium

detector linked to an 8K multi-channel analyzer incorporating the GENIE-PC software suite from Canberra. The system was calibrated using traceable isotope solutions [Physikalisch-Technische Bundesanstalt (PTB)] and all results were corrected for matrix interferences using density correction factors calculated from Gamatool software suite.<sup>22</sup> Validity of efficiency and energy calibrations was determined using a range of soil and vegetation reference materials (IAEA – 326, 375, Soil-6 and 156) and results were within the 95% confidence interval quoted for the reference materials.

Cesium-137 was quantified using its 661 keV emission and  $^{40}\text{K}$  was quantified using its 1460 keV emission. Samples were counted for a period sufficient to ensure a 2 $\sigma$  error of less than 10% for  $^{137}\text{Cs}$  and 25% for  $^{40}\text{K}$  after correction for laboratory background. Counting times varied from between 1 day and 2 weeks. Nuclides were identified using a library driven search routine and quantitative analyses were carried out using the appropriate detector calibration. Radionuclide results were reported in Bq/kg on a dry weight basis.

### Chemical analysis

Soil pH was measured using soil to water ratios of 1:2 (w/v).<sup>23</sup> Soil moisture content was determined by loss in weight after drying at 105° C for 24 hours and organic matter was determined on dry soils by loss on ignition (550° C, 4 hours). Soil CEC was measured using a sodium saturation method.<sup>24</sup> Exchangeable potassium, calcium, sodium and magnesium were determined in the soil using an ammonium acetate fraction method.<sup>25</sup> Total calcium, sodium and magnesium in the vegetation were determined using a strong nitric acid digestion and analyzed using flame photometry (sodium) and atomic absorption (calcium and magnesium).

### Data analysis

For each site, individual plant transfer factors (TF) were calculated for all vegetation as a ratio of activity concentrations of plant material and underlying soil (0–20 cm) as given by:

$$\text{TF} = \text{Bq/kg plant material (d.w.)} / \text{Bq/kg soil (d.w.)}$$

Statistical analyses for differences between TFs or radionuclide concentrations were carried out using *t*-test (parametric test), or One Way ANOVA (parametric test). Correlations were carried out using either Pearson Product Moment correlation (normally distributed data) or Spearman's rank-correlation test (for non parametric data). Statistical analysis was carried out using SigmaStat 2.03 software suite.<sup>26</sup>

### Results and discussion

Concentrations of  $^{137}\text{Cs}$  found in the peat soils were higher than  $^{40}\text{K}$  ( $t$ -test,  $p < 0.001$ ) and activities of both radionuclides decreased with increasing soil depth at all sites (Table 1). Cesium-137 mean concentrations were significantly higher in the 0–5 cm layer (One Way ANOVA,  $p < 0.002$ ) than the subsequent lower layers (Fig. 1). The decrease in  $^{137}\text{Cs}$  activity with increasing soil depth is best described using a log function where an inverse relationship was found to be present between log average  $^{137}\text{Cs}$  activity as a function of soil depth ( $r^2 = 0.991$ ). Concentrations of  $^{137}\text{Cs}$  were also found to be low further down in the soil profile with an activity of  $11 \pm 1$  Bq/kg observed in the 45–50 cm layer at Site 3 (0–50 cm depth profile). The apparent low vertical mobility of  $^{137}\text{Cs}$  within the soil profile is possibly due to the high concentration of organic matter in the peat (>90% in all peat soils). Research has shown that radiocesium added to organic soil remains plant-available for extended periods of time and after uptake is recycled within the rooting zone of the vegetation in the form of decomposing plant material.<sup>1,2,4,27–29</sup>

Potassium-40 mean concentrations were significantly higher in the 0–5 cm layer (One Way ANOVA,  $p < 0.05$ ) in comparison to the 15–20 cm layer, which is possibly resulting from the recycling of  $^{40}\text{K}$  in the top layers of the soil from decomposing plant material. Potassium-40 activities were low in all peat soils (<50 Bq/kg), which is indicative of organic soils, where the acidic nature of the soil results in leaching of essential nutrients.<sup>2</sup>

Table 1. Activity concentrations of radionuclides in soils (in Bq/kg)\*

Site	Soil layer, cm	$^{137}\text{Cs}$	$^{40}\text{K}$
1	0–5	$158 \pm 10$	$33 \pm 5$
	5–10	$81 \pm 5$	$26 \pm 3$
	10–15	$50 \pm 3$	$26 \pm 3$
	15–20	$22 \pm 1$	$18 \pm 3$
2	0–5	$199 \pm 13$	$36 \pm 4$
	5–10	$127 \pm 8$	$30 \pm 5$
	10–15	$58 \pm 4$	$31 \pm 4$
	15–20	$22 \pm 1$	$19 \pm 3$
3	0–5	$149 \pm 10$	$45 \pm 6$
	5–10	$98 \pm 6$	$48 \pm 7$
	10–15	$75 \pm 5$	$40 \pm 5$
	15–20	$78 \pm 5$	$47 \pm 5$
	20–25	$40 \pm 3$	$30 \pm 5$
	25–30	$26 \pm 2$	$21 \pm 4$
	30–35	$13 \pm 1$	$13 \pm 3$
	35–40	$11 \pm 1$	$13 \pm 3$
	40–45	$10 \pm 1$	$14 \pm 3$
45–50	$11 \pm 1$	$13 \pm 3$	
4	0–5	$230 \pm 15$	$28 \pm 4$
	5–10	$120 \pm 8$	$22 \pm 3$
	10–15	$43 \pm 3$	$16 \pm 4$
	15–20	$22 \pm 1$	$14 \pm 3$
5	0–5	$191 \pm 13$	$30 \pm 4$
	5–10	$144 \pm 9$	$24 \pm 3$
	10–15	$47 \pm 3$	$16 \pm 4$
	15–20	$23 \pm 2$	$13 \pm 3$
6	0–5	$122 \pm 8$	$29 \pm 5$
	5–10	$65 \pm 4$	$24 \pm 4$
	10–15	$32 \pm 2$	$18 \pm 3$
	15–20	$16 \pm 1$	$14 \pm 3$
7	0–5	$198 \pm 12$	$25 \pm 5$
	5–10	$49 \pm 3$	$18 \pm 3$
	10–15	$26 \pm 3$	$16 \pm 4$
	15–20	$13 \pm 1$	$13 \pm 3$

\* Concentration of radionuclides present in soil  $\pm$  instrument error in the measurement.

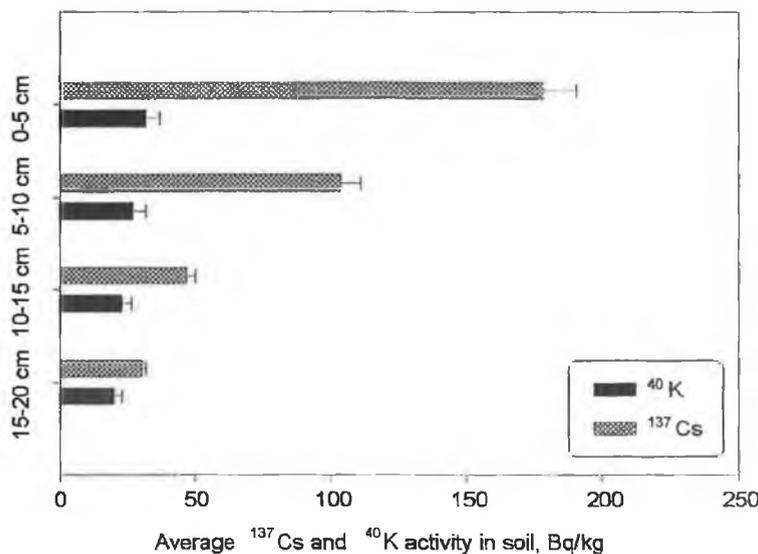


Fig. 1. Plot of average  $^{137}\text{Cs}$  and  $^{40}\text{K}$  activity in soil profile

Levels of <sup>137</sup>Cs found in vegetation were higher in *C. vulgaris* leaves than in all other vegetation and activities in *C. vulgaris* stems, *E. vaginatum* and moss were relatively similar (Table 2). The highest concentration of <sup>40</sup>K was found to be present in *E. vaginatum*. Moss samples were found to contain the lowest concentration of <sup>40</sup>K and one of the moss samples also contained the lowest concentration of <sup>137</sup>Cs for the vegetation sampled. This may be a reflection of the uptake mechanisms of mosses as the lack a well-developed root system and uptake of radionuclides occurs mainly through ion exchange processes from wet and dry deposition.<sup>30,31</sup>

Transfer factors values were calculated for <sup>137</sup>Cs and <sup>40</sup>K in *C. vulgaris* leaves and stems and *E. vaginatum*. Transfer factors were not calculated for moss species due to the lack of a well-developed root system.<sup>20</sup>

Soil TF values for <sup>137</sup>Cs in the vascular plants studied ranged from 1.9 to 9.6 (Table 3). Transfer factors values for <sup>137</sup>Cs were significantly higher in *C. vulgaris* leaves in comparison to the stems (*t*-test, *p*=0.016), however, comparisons between species show that <sup>137</sup>Cs TFs for *C. vulgaris* and *E. vaginatum* are statistically similar (*t*-test, *p*>0.05). Transfer factors for *C. vulgaris* reported within this study are similar to TF values reported in the literature of 1.8–3.3 (plant portion unspecified)<sup>2</sup> and 6 and 0.6 for *C. vulgaris* leaves and stems, respectively.<sup>16</sup> The trend found within this study for <sup>137</sup>Cs TF values in *C. vulgaris* leaves and stems (i.e., leaves > stems) is in agreement with the findings for <sup>137</sup>Cs in *Salix viminalis* (basket willow) where the TF values decreased in the order: roots > leaves > cuttings > stems.<sup>32</sup> Similar trends were also observed for *V. myrtillus* (bilberry), *V. uliginosus* (bog bilberry), *V. vitis-idaea* (mountain cranberry), *Vaccinium oxycoccos* (northern cranberry) and *Calluna vulgaris* where TF values in the plant portions decreased in the order: flowers > leaves = berries > roots > stems.<sup>16</sup> The above results for the distribution of <sup>137</sup>Cs within *C. vulgaris* are in agreement with <sup>137</sup>Cs distribution in tropical, spruce and pine trees where current knowledge indicates that <sup>137</sup>Cs activity is highest in the youngest or growing part of the trees.<sup>33–36</sup>

Potassium-40 TF values ranged from 0.9 to 13.8 and were significantly higher in *E. vaginatum* in comparison to both *C. vulgaris* leaves and stems (One Way ANOVA *p*<0.001). Comparisons between <sup>40</sup>K TF values in *C. vulgaris* leaves and stems show that the differences are not statistically significant (One Way ANOVA, *p*=0.42).

Further investigation into <sup>137</sup>Cs uptake in vegetation indicates that <sup>137</sup>Cs TF values showed little correlation with soil physiochemical properties (Table 4). However, correlations were found between <sup>137</sup>Cs TF values and the nutrient status of the vegetation (Table 5), in particular the secondary nutrients, calcium and magnesium (Table 5). A Spearman correlation coefficient of 0.95 (*p*<0.001) indicates a strong correlation between the transfer of <sup>137</sup>Cs in vegetation and the calcium content of vegetation (Fig. 2). A similar observation was noted between the magnesium content of vegetation and <sup>137</sup>Cs TF values (Pearson correlation coefficient of 0.88 (*p*=0.004)) as shown in Fig. 3. However, no relationship was observed between the sodium content of vegetation and <sup>137</sup>Cs TF values (Spearman correlation coefficient of 0.43 (*p*=0.283)).

On the other hand <sup>40</sup>K TF values showed no correlation with the nutritional status of the vegetation. Potassium-40 TF values showed little correlation with the soil physiochemical properties studied (Table 4) with the exception of soil cation exchange capacity. A Spearman correlation coefficient of 0.93 (*p*<0.001) indicates a strong relationship between <sup>40</sup>K transfer to vegetation and the soil cation exchange capacity (Fig. 4). As the soil CEC is a measure of the available cations present in the soil, then an increase in soil CEC would result in an increase in plant available nutrients in the soil.<sup>37</sup>

Table 2. Activity concentrations of radionuclides in bog vegetation (in Bq/kg)\*

Site	Sample	<sup>137</sup> Cs	<sup>40</sup> K
1	<i>Calluna vulgaris</i> – leaves	716 ± 50	123 ± 21
2	<i>Calluna vulgaris</i> – leaves	589 ± 40	100 ± 19
3	<i>Calluna vulgaris</i> – leaves	956 ± 65	86 ± 15
1	<i>Calluna vulgaris</i> – stems	302 ± 21	69 ± 13
2	<i>Calluna vulgaris</i> – stems	196 ± 13	73 ± 14
3	<i>Calluna vulgaris</i> – stems	282 ± 19	40 ± 7
4	<i>Sphagnum</i> moss	142 ± 10	64 ± 13
5	<i>Sphagnum</i> moss	325 ± 22	65 ± 13
6	<i>Eriophorum vaginatum</i>	301 ± 20	293 ± 32
7	<i>Eriophorum vaginatum</i>	192 ± 13	225 ± 25

\* Concentration of radionuclides present in vegetation ± instrument error in the measurement.

Table 3. Transfer factor (TF) values for <sup>137</sup>Cs and <sup>40</sup>K

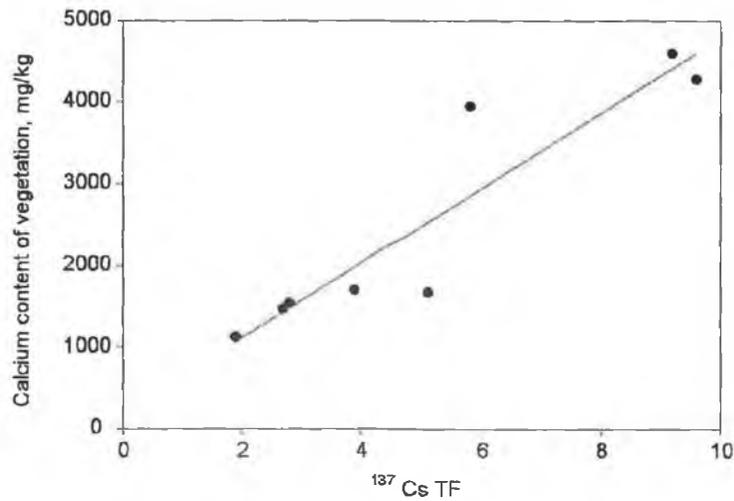
Site	Sample	<sup>137</sup> Cs	<sup>40</sup> K
1	<i>Calluna vulgaris</i> – leaves	9.2	4.8
2	<i>Calluna vulgaris</i> – leaves	5.8	3.5
3	<i>Calluna vulgaris</i> – leaves	9.6	1.9
1	<i>Calluna vulgaris</i> – stems	3.9	2.7
2	<i>Calluna vulgaris</i> – stems	1.9	2.5
3	<i>Calluna vulgaris</i> – stems	2.8	0.9
6	<i>Eriophorum vaginatum</i>	5.1	13.8
7	<i>Eriophorum vaginatum</i>	2.7	12.3

Table 4. Physicochemical properties of soil surface horizons (0–20 cm)

Site	Moisture, %	pH	Organic matter, %	Available potassium, mg/kg	Available calcium, mg/kg	Available sodium, mg/kg	Available magnesium, mg/kg	CEC, meq/100 g
1	88.5	3.41	95.2	124	362	245	494	149
2	87.2	3.41	94.8	154	404	219	439	147
3	86.6	3.75	95.1	129	256	170	319	142
4	91.3	4.47	96.1	128	403	231	554	149
5	91.7	3.97	95.8	180	488	243	550	127
6	89.2	3.92	95.4	70	326	156	422	156
7	86.4	3.88	95.2	102	379	155	459	155

Table 5. Nutrient concentration of vegetation

Site	Vegetation	Calcium, mg/kg	Sodium, mg/kg	Magnesium, mg/kg
1	<i>Calluna vulgaris</i> – leaves	4595	1235	1622
2	<i>Calluna vulgaris</i> – leaves	3947	1134	1586
3	<i>Calluna vulgaris</i> – leaves	4282	1129	1570
1	<i>Calluna vulgaris</i> – stems	1708	759	485
2	<i>Calluna vulgaris</i> – stems	1130	1012	511
3	<i>Calluna vulgaris</i> – stems	1539	1248	435
4	<i>Sphagnum</i> moss	8831	1299	1350
5	<i>Sphagnum</i> moss	6257	821	1489
6	<i>Eriophorum vaginatum</i>	1673	734	898
7	<i>Eriophorum vaginatum</i>	1469	688	814

Fig. 2. Plot of  $^{137}\text{Cs}$  TF values versus calcium content of vegetation

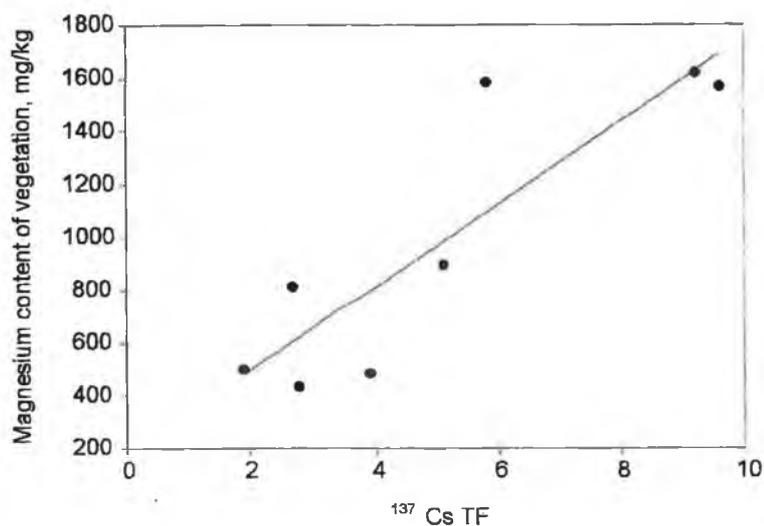


Fig. 3. Plot of  $^{137}\text{Cs}$  TF values versus magnesium content of vegetation

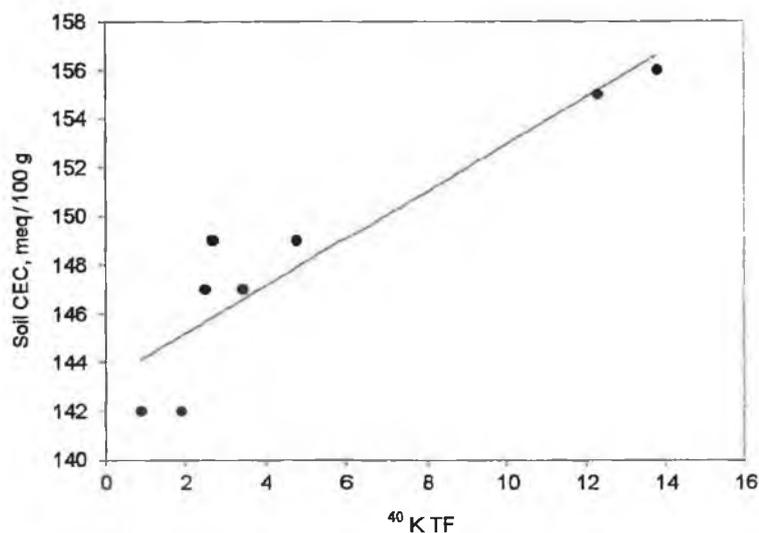


Fig. 4. Plot of  $^{40}\text{K}$  TF values versus soil CEC

### Conclusions

The results of this study indicate that for the Atlantic blanket bog environment studied, the uptake of  $^{137}\text{Cs}$  in vascular plants is not dependent on the plant species studied as TF values for *C. vulgaris* and *E. vaginatum* are statistically similar. However, in the case of, *C. vulgaris* the uptake of  $^{137}\text{Cs}$  is dependent on the plant compartment as the leaves have been found to accumulate significantly more  $^{137}\text{Cs}$  than the stems. This indicates that the highest concentration of  $^{137}\text{Cs}$  in *C. vulgaris* remains in the plant portions grazed by animals, therefore, possibly leading to human contamination through food consumption. The transfer

of  $^{137}\text{Cs}$  to vegetation was also found to be positively correlated with the nutrient status of vegetation in particular the calcium and magnesium content of vegetation.

Transfer factor values for  $^{40}\text{K}$  indicate that its uptake in vascular plants is dependent on plant species as significantly higher concentrations were found in *E. vaginatum* in comparison to *C. vulgaris*. However, unlike  $^{137}\text{Cs}$ , its concentrations in *C. vulgaris* leaves and stems are not significantly different. The concentration of both  $^{137}\text{Cs}$  and  $^{40}\text{K}$  found in moss samples were in general lower than those found in vascular plants probably reflecting the mode of uptake of radionuclides by moss.

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The authors would like to thank the Higher Education Authority for providing funding for this research project. The authors would also like to thank Dr. Mark DOWDALL for his helpful advice and technical assistance.

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## Organic soil as a radionuclide sink in a High Arctic environment

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(Received October 12, 2004)

The role of organic soil as a sink for radioactive contaminants in a High Arctic environment was studied. Samples were obtained from an area of organic soil located on the Arctic archipelago of Svalbard (79° N) and from a non-organic control site in the same region. Samples were differentiated into organic layers and the underlying material and measured for a suite of anthropogenic and natural radionuclides and for a variety of soil chemistry parameters. Results indicated that the organic components of the soil constitute a sink term for a number of radionuclides. Values for Pu isotopes, <sup>137</sup>Cs and <sup>238</sup>U were appreciably higher at the study site than at the control site, by up to a factor of 40 for Pu and 20 for <sup>137</sup>Cs and <sup>238</sup>U. The source of <sup>238</sup>U to the site appears to be enrichment of this isotope from surface or melt water via adsorption to either iron hydroxides or organic matter although the situation pertaining to Pu and <sup>137</sup>Cs remains less clear.

### Introduction

The role of organic soils and their constituents in the retention and accumulation of radionuclides, both natural and anthropogenic, has been well documented by a number of papers over the years.<sup>1–5</sup> This ability to concentrate low levels of radionuclides from waters has implications for the potential of organic soils to act as sinks for such radionuclides leading to localized areas of elevated radioactivity. Although often considered as a region where radionuclides exhibit low mobility in the surficial terrestrial environment due to the lack of liquid water for much of the year and the low biological activity, this study details the occurrence of elevated radionuclides in a young organic soil at a location in the High Arctic and discusses the radiometric results with respect to the chemical properties of the soil at the site in question. The results of the study are used to ascertain whether the organic soil in the High Arctic environment can constitute a sink term for radionuclides.

Organic soil and peat can be found in the High Arctic as thin horizons typically between 5 and 20 cm deep,<sup>6</sup> although thicker deposits have been found at higher latitudes on Spitsbergen,<sup>7</sup> Novaya Zemlya<sup>6</sup> and in the Canadian Arctic archipelago.<sup>8</sup> Climatic conditions prevailing in the High Arctic (>75°N) are generally not conducive to the formation of organic soil, but such soils can occur where there is a lateral infiltration of water or where inputs of nutrients can result in increased vegetative growth. Such conditions may be found where soils are saturated by snow melt or where significant nutrient input occurs via, for example, the presence of large numbers of birds or animals at a certain location for periods of time.

The organic soil for which results are reported in this paper lies on Brøggerhalvøya (Brøgger peninsula), Kongsfjorden, on the island of Spitsbergen, located in the Svalbard Archipelago (Fig. 1). The prevailing climate on the west coast of Spitsbergen is influenced by the North Atlantic Current which brings warm water to the region, with an average temperature of –14 °C in February and 5 °C in July. Precipitation is low, approx. 400 mm per annum,<sup>9</sup> and falls mainly as snow during the winter months. The geology of the region consists of sedimentary rocks of Middle and Upper Carboniferous and Permian ages with sandstones and limestones at various locations.<sup>10</sup> Mountainous slopes are scree covered and the lowland areas of the peninsula are partly covered by marine deposits. Post-glacial terraces are present, consisting of gravel and sand with low organic material content.<sup>11</sup> Soils of the area vary with depth and are generally low in humic materials and are of ages between 9,000 and 12,000 years.<sup>12</sup> Vegetation of the region is typical of the Middle Arctic Tundra Zone<sup>13</sup> with low lying water saturated areas being dominated by mosses. DOWDALL et al.<sup>14</sup> surveyed the soils of Brøggerhalvøya for levels of anthropogenic (<sup>137</sup>Cs) and natural gamma-emitting radionuclides and reported relatively low level uniform distributions of both with <sup>226</sup>Ra/<sup>238</sup>U secular equilibrium being reported for the majority of the samples taken. However, the same study also reported an area of organic soil at the base of a seabird colony exhibiting elevated levels of <sup>137</sup>Cs and natural nuclides with a marked disequilibrium in the <sup>238</sup>U series. This report details investigation of the radionuclide burden of this soil and chemical parameters of pertinence to the premise that organic soils can constitute a radionuclide sink in this High Arctic environment.

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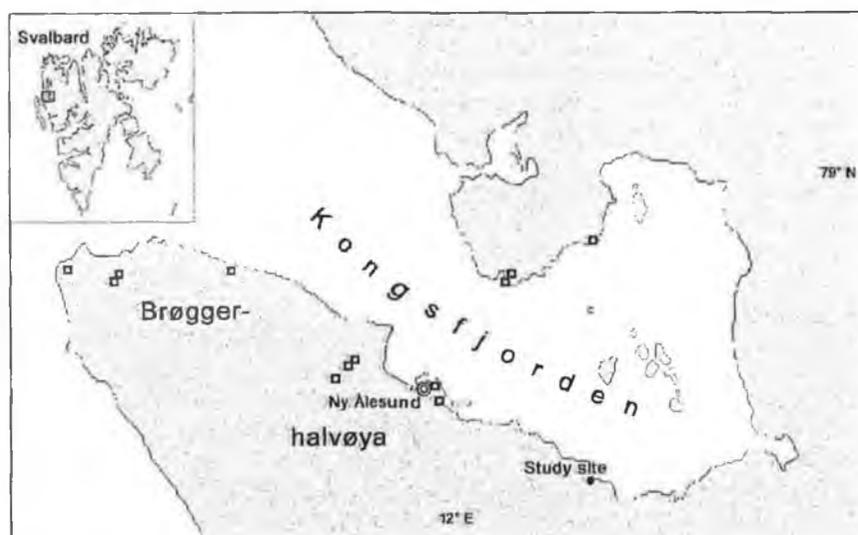


Fig. 1. Location of study site and control soil sampling sites (open squares) within the Kongsfjorden area

Although the Arctic region, in general, and Svalbard in particular, can be characterized as exhibiting low levels of radioactive contamination, the Eurasian arctic regions remain acutely vulnerable to radioactive contamination from a wide variety of sources which have been well described elsewhere.<sup>15</sup> The significance attached to this potential contamination is increased substantially by a number of factors including the uniqueness and vulnerability of Arctic ecosystems, the lack of knowledge pertaining to the long term behavior of radionuclides in the Arctic environment and the increasing global emphasis on the concept of protection of the environment. Therefore, the identification and elucidation of processes that can lead to the occurrence of localized regions of elevated radioactivity is of some importance to Arctic radioecology. The occurrence of elevated radionuclide levels associated with High Arctic organic soils is imbued with extra significance when viewed within the context of the overall nutrient status of such locations, which generally tend to be nutrient poor. Deposits of organic material in the High Arctic are often related to enhanced nutrient input from the accumulation of deposited faecal material below seabird colonies. This enhanced nutrient input can promote a greater abundance and diversity of vegetation to grow at these locations<sup>16</sup> which may then provide important grazing sites for herbivores (e.g., reindeer). Therefore, vegetation growing in soils exhibiting significantly elevated radionuclide levels could potentially lead to an increased radionuclide intake to High Arctic herbivores. Given current predictions for climate change and the impact those changes may have on the Arctic, it is important to understand the behavior of radionuclides in Arctic terrestrial ecosystems.

## Experimental

### *Sampling and methods*

The area of organic soil studied in this paper is located at the base of an accumulation of deposited faecal material below a kittiwake seabird colony of some 1000 breeding pairs. The soil at the base of this cliff receives run-off from the cliff and seasonal melt water from snow. The area of organic soil extends for ~50 m along the cliff and ~20 m out from the cliff base, which is ~50 m from the sea. The soil at the location consists of a basement mineral till (~4 cm deep), a thin well defined layer of humified organic material (<5 cm) and a surface mat of moss (*Sanionia uncinata*).

In 2002, a series of transects were established across the area of organic soil, running parallel and at right angles to the base of the cliff. At predetermined locations, samples of the surficial material present were taken using stainless steel implements, in plots of ~10 cm<sup>2</sup>. The samples were put in polyethylene bags and transferred to the laboratory. The living material was removed from the samples and the gross samples were divided into sub-samples of humified organic material and the underlying till. The subdivided samples were then dried at 105 °C to constant weight, homogenized in a stainless steel blender and passed through a 2 mm sieve. The prepared samples were then subjected to chemical and radiometric analysis. For comparison, a series of soil samples exhibiting no accumulation of organic material, was taken from within the Kongsfjorden area (Fig. 1). These soils mostly consisted of well drained, ahumic soils typical of Polar

Desert soils<sup>17</sup> and were sampled and treated in the same way as the organic soils, but with no post-collection sub-sampling or differentiation.

#### *Radiometric analysis*

Samples were analyzed by both high resolution gamma-spectrometry for gamma-emitting natural and anthropogenic radionuclides and by alpha-spectrometry for isotopes of Am and Pu.

**Gamma-spectrometry:** Dried and homogenized samples were packed into plastic bags (14 to 200 ml) and counted on an electrically cooled, 8k channel, high-resolution gamma-spectrometer (HpGe) with a nominal resolution of 1.8 keV at 1332 keV and a relative efficiency of 40%. Spectra were collected for periods of between 24 and 48 hours. All spectra were corrected for background and the effects of varying sample density and chemical composition. <sup>137</sup>Cs was determined via its characteristic emission at 661 keV, <sup>40</sup>K by its emission at 1460 keV. Thorium-232 activities were inferred from the activity of <sup>228</sup>Ac which can be assumed to be in secular equilibrium in terrestrial matrices. <sup>238</sup>U was determined via the emissions of its immediate daughter <sup>234</sup>Th at 63 keV. <sup>226</sup>Ra was assayed by correcting the peak at 186 keV for the <sup>235</sup>U contribution utilizing an activity ratio of 21.4 between this isotope and <sup>238</sup>U. The detector was calibrated with standards using IAEA reference soils and vegetation samples. A 5% loading of duplicates indicated reproducibility of better than 10%.

**Alpha-spectrometry:** Activities of <sup>238</sup>Pu, <sup>239+240</sup>Pu and <sup>241</sup>Am were determined on 10 g aliquots of dried sample material. <sup>242</sup>Pu and <sup>243</sup>Am were used as yield determinants. Samples were leached with nitric acid before being subjected to a separation and purification sequence involving ion-exchange, co-precipitation and solvent extraction before being electroplated to stainless steel disks and counted using silicon implanted passive detectors.

#### *Chemical analysis*

The pH of selected samples was measured using 5 g aliquots of homogenized material in distilled water (1:2 w/v). Organic material was determined by the loss of ignition (550 °C, 4 hours), organic carbon by using the rapid dichromate oxidation technique<sup>18</sup> involving oxidation of approximately 0.3 g of material with 1N K<sub>2</sub>CrO<sub>7</sub> and H<sub>2</sub>SO<sub>4</sub> followed by titration with 0.2N ferrous ammonium sulphate. Water soluble nitrate, phosphate and sulphate were assayed using ion chromatography. Sodium saturation method<sup>19</sup> was used to determine cation-exchange capacity.

## Results

Radionuclide data and statistics for fractionated samples from the study site and for control soil samples is presented in Table 1. Chemical parameters are presented in Table 2, concentration and activity ratios of actinides for some samples in Table 3.

## Discussion

The High Arctic region is usually considered as exhibiting relatively low levels of radionuclide contamination. However, the Svalbard Archipelago, lying in close proximity to sites of previous atmospheric weapon tests and in the path of continued and potential oceanic long-range exposure to sources of radionuclides from the European mainland, remains acutely vulnerable to such contamination. The historical radioactive contamination of the archipelago can be gleaned from a limited number of previous studies and estimates. Within the 70 to 80°N latitude band, deposition estimates of <sup>137</sup>Cs, <sup>239+240</sup>Pu and <sup>238</sup>Pu from atmospheric weapon testing have been calculated at 1–5 kBq/m<sup>2</sup>, 13.3 Bq/m<sup>2</sup> and 0.3 Bq/m<sup>2</sup>, respectively.<sup>15,20,21</sup> Comparable measurements of deposition on Svalbard to the estimated values have been reported in the order of 0.2 to 2.2 kBq/m<sup>2</sup> for <sup>137</sup>Cs<sup>22,23,24</sup> and 14 to 26 Bq/m<sup>2</sup> for <sup>239+240</sup>Pu.<sup>25,26</sup> Svalbard is considered to have been relatively unaffected by fallout from the Chernobyl accident in 1986. PINGLOT et al.<sup>24</sup> reported Chernobyl fallout levels for <sup>137</sup>Cs of only 20 Bq/m<sup>2</sup>, while NEGOITA<sup>27</sup> reported typical post-Chernobyl <sup>137</sup>Cs levels in soils of between <1.5 and 35.8 Bq/kg, with one measurement of 213 Bq/kg.

Values of <sup>137</sup>Cs in control surface soil samples of this study were in broad agreement with previously reported values<sup>27,28</sup> in a range of 1.3 to 34.6 Bq/kg, while average levels of <sup>238</sup>U (29 Bq/kg), <sup>226</sup>Ra (28 Bq/kg), <sup>232</sup>Th (28 Bq/kg) and <sup>40</sup>K (624 Bq/kg) were similar to global averages for soil.<sup>21</sup> Conversion to depositional values (utilizing sample dimensions and an average bulk density of 0.78) resulted in areal activities in the order of 0.1 to 2.6 kBq/m<sup>2</sup> (mean 1.1 kBq/m<sup>2</sup>) for control samples, which are in good agreement with previous measurements. The activity concentrations of <sup>238</sup>Pu, <sup>239+240</sup>Pu and <sup>241</sup>Am in a control soil sample were in broad agreement with previously reported estimated and observed integrated deposition values.<sup>20,25,27</sup> <sup>238</sup>Pu/<sup>239+240</sup>Pu and <sup>241</sup>Am/<sup>239+240</sup>Pu activity ratios for the control soil were similar to the published global fallout ratios for Svalbard of 0.025 and 0.37, respectively,<sup>20,25</sup> indicating that the source term for the exhibited levels was the fallout from atmospheric weapons tests in the 1960's and 1970's.

Table 1. Activity statistics for overlying organic and underlying mineral materials from the study site and control soil samples. All values are in Bq/kg dry weight

	$^{137}\text{Cs}$	$^{40}\text{K}$	$^{238}\text{U}$	$^{226}\text{Ra}$	$^{232}\text{Th}$
Overlying organic material					
<i>n</i>	16	16	16	16	16
Mean	151.0	499.3	94.9	43.8	30.0
Std. dev.	56.1	91.0	54.1	24.0	5.7
Min	65.0	340.0	33.0	7.0	22.3
Median	153.5	512.5	84.0	41.0	28.5
Max	250.0	626.0	244.0	94.0	38.4
Underlying mineral material					
<i>n</i>	16	16	16	16	16
Mean	35.8	624.3	29.3	28.9	28.2
Std. dev.	23.6	31.6	7.4	7.4	1.6
Min	3.6	562.0	20.0	19.0	25.1
Median	34.5	629.0	29.0	28.0	28.0
Max	91.6	684.0	50.0	45.0	31.2
Control soils					
<i>n</i>	12	12	12	12	12
Mean	14.9	375.9	27.9	38.0	24.8
Std. dev.	12.2	186.8	8.6	10.7	9.4
Min	1.3	22.0	17.0	21.0	10.3
Median	10.6	434.5	25.5	40.5	28.3
Max	39.0	564.0	47.0	51.3	35.6

Table 2. Statistics of chemical properties for overlying organic material and underlying mineral material samples from the study site

	pH	Organic matter, %	Organic carbon, %	Nitrate, mg/kg	Phosphate, mg/kg	Sulphate, mg/kg	CEC, meq/100 g
Overlying organic material							
<i>n</i>	16	16	16	16	16	16	16
Mean	6.2	31.6	11.9	51.4	91.9	61.4	75.5
Std. dev.	0.3	9.1	4.1	167.1	60.6	37.2	35.6
Min	5.8	14.3	5.1	< 1	< 2	25.9	27.4
Median	6.3	33.2	11.6	< 1	73.6	44.7	73.1
Max	6.6	46.5	19.6	672.9	219.3	132.7	139.7
Underlying mineral material							
<i>n</i>	16	16	16	16	16	16	16
Mean	6.8	1.9	0.7	1319	23.4	21.5	5.6
Std. dev.	0.5	1.5	0.4	1666	33.6	19.3	4.6
Min	5.8	0.6	0.3	32.9	< 0.5	< 0.5	1.5
Median	6.9	1.1	0.5	354.4	10.8	16.1	3.2
Max	7.5	5.1	1.7	6260.5	123.7	75.8	15.9

Table 3. Average activity concentrations and activity ratios of actinides in overlying organic material from the study site and in a control soil sample. All values are in Bq/kg dry weight

	$^{238}\text{Pu}$	$^{239+240}\text{Pu}$	$^{241}\text{Am}$	$^{238}\text{Pu}/^{239+240}\text{Pu}$	$^{241}\text{Am}/^{239+240}\text{Pu}$	$^{239+240}\text{Pu}/^{137}\text{Cs}$
Control soil						
<i>n</i> = 1	0.01 ± 0.007	0.2 ± 0.03	0.05 ± 0.02	0.056	0.27	0.035
Overlying organic material						
<i>n</i> = 6	0.25 ± 0.05	6.9 ± 2.1	2.8 ± 0.8	0.038 ± 0.01	0.42 ± 0.03	0.033 ± 0.01

Levels of the natural nuclides in samples taken from the control areas are in good agreement with previous studies<sup>14</sup> although  $^{238}\text{U}/^{226}\text{Ra}$  ratios are slightly lower than the values reported in the relevant study.  $^{238}\text{U}/^{226}\text{Ra}$  ratios in the control samples are indicative of low mobility of these radionuclides, most probably due to the suppression of the segregative processes

(precipitation, selective dissolution, etc.) most often responsible for this disequilibrium due to the low levels of liquid water in the areas from which these samples were drawn.

Activity concentrations of  $^{137}\text{Cs}$  and  $^{238}\text{U}$  exhibited by the organic layer of soils at the study site were up to 20 fold higher than levels in the control soils, with

average levels of 165 Bq/kg and 101 Bq/kg, respectively. As for  $^{137}\text{Cs}$ , elevated activity concentrations of  $^{238}\text{Pu}$ ,  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  were observed in the organic layer up to 73 fold higher than the levels of these radionuclides in the control soil, with average levels of  $0.25\pm 0.05$  Bq/kg,  $6.9\pm 2.1$  Bq/kg and  $2.8\pm 0.8$  Bq/kg, respectively. However, despite these elevated levels, the average  $^{238}\text{Pu}/^{239+240}\text{Pu}$  and  $^{241}\text{Am}/^{239+240}\text{Pu}$  activity ratios for the organic layer were similar to the control soil ratios reflecting a global fallout signature. Furthermore, the average  $^{239+240}\text{Pu}/^{137}\text{Cs}$  activity ratio ( $0.033\pm 0.01$ ) in the organic layer was similar to both the control soil  $^{239+240}\text{Pu}/^{137}\text{Cs}$  activity ratio (0.035) and to an estimated global fallout  $^{239+240}\text{Pu}/^{137}\text{Cs}$  activity ratio between 70 and 80°N of 0.03,<sup>20,21</sup> when allowing for the decay of  $^{137}\text{Cs}$ , suggesting a similar enrichment process for  $^{137}\text{Cs}$  and Pu and Am isotopes.

The mineral materials underlying the organic layer at the study site reflect the average levels for the control samples. No significant difference was determined between  $^{238}\text{U}$  in the mineral layers at the study site and those exhibited by the control samples ( $t$ -test,  $p=0.651$ ) nor  $^{232}\text{Th}$  (Mann-Whitney,  $p=0.981$ ).  $^{226}\text{Ra}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$  were all significantly greater in the mineral layers than in the control samples. Strong significant (Pearson, 0.05 level) correlations were observed between  $^{137}\text{Cs}$ ,  $^{226}\text{Ra}$  and  $^{238}\text{U}$  and the organic carbon content of the mineral samples (Pearson coefficients of 0.626, 0.726 and 0.618, respectively), indicating that the content of these nuclides in the mineral layers is due to ingress of nuclide bearing organic materials (Fig. 2). No strong relationship was observed between radiometric parameters and other chemical factors in the mineral

layers. Within the organic layers at the study site,  $^{40}\text{K}$  is negatively correlated with organic carbon (Pearson coefficient  $-0.73$ ) indicating that this nuclide is not enriched within the organic material, its presence being largely accounted for by whatever mineral material is present in the organic layers. Compared to the control samples, both  $^{137}\text{Cs}$  and  $^{238}\text{U}$  display significantly higher levels in the organic material from the study site (Mann-Whitney  $P<0.01$ ).

The three species exhibiting the greatest levels of enrichment within the organic material of the studied soil are  $^{238}\text{U}$ ,  $^{137}\text{Cs}$  and the Pu isotopes. The most likely source of these nuclides at the study site is via lateral ingress of water (primarily direct in flux of snow and ice melt waters or stream/soil waters) at the site. Thermodynamic data for uranium<sup>29</sup> suggest that uranium may be found in surficial waters under oxidizing conditions as complexes of sulphate, phosphate, fluoride and organic materials. Chemical results for the organic materials taken in this study indicate significant amounts of soluble sulphate and phosphate which it must be assumed were carried to the site by ingress of water. The most probable source of these species is from the accumulated faecal deposits at the site. Although thermodynamic data for uranium at temperatures less than 20 °C is not commonly available, it would appear that conditions at the site are conducive to the transport of uranium with water to the location. Although the content of uranium in snow melt water is low ( $1.9\text{ Bq/m}^3$ )<sup>30</sup> the relatively large amounts of water that enter the site could constitute a source of uranium of the soil by adsorption of uranium from the solution.

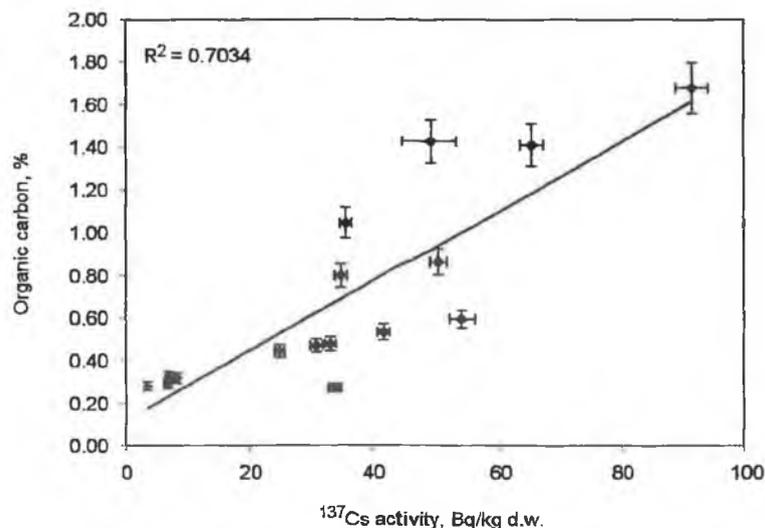


Fig. 2. Relationship between  $^{137}\text{Cs}$  activities in the mineral layer of the study site and the organic carbon content of samples

The concept that uranium is transferred to the site via water is supported by the fact that neither  $^{226}\text{Ra}$  nor  $^{232}\text{Th}$  are enriched to the same extent relative to  $^{238}\text{U}$ . Neither of these nuclides is considered mobile in the surficial environment and their levels in the organic matrix are more reflective to the parent material than  $^{238}\text{U}$ . A second possibility is that the uranium bearing stream/soil water is responsible for the uranium influx. The differentiation between these waters and those previously discussed is that the stream/soil waters may have accumulated uranium via weathering of rocks as opposed to receiving it via atmospheric deposition. The ability of organic material (typically humic acids) to adsorb uranium from solution is due to some extent to the shape of the divalent uranyl ion which is easily adsorbed between layers of carbonaceous material and the role of organic soil constituents in adsorption of uranium. MEGUMI and MAMURO<sup>31</sup> have also identified the role of iron and manganese oxides in the retardation of uranium mobility via a reduction process and given the large amounts of both iron and organic material at the study site, it would appear reasonable to hypothesize that accumulation of uranium at the study site is due to a process involving either adsorption to organic materials or immobilization with iron oxides.

Unlike uranium, neither Pu isotopes nor  $^{137}\text{Cs}$  have natural sources and cursory consideration indicates that the most probable sources for the excess levels observed at the site are seawater and glacial melt water. Seawater could possibly be transferred to the site via the actions of the resident seabird population although it would appear improbable that an equivalent amount of seawater could be transferred as the amount of water arriving at the site via melt water. A further complication is that neither Pu or  $^{137}\text{Cs}$  inputs to the site via meltwater could be steady state as maximum inputs would have occurred during the periods after the peak depositions (late 1960's for both Pu and  $^{137}\text{Cs}$  and again some time after 1986 for  $^{137}\text{Cs}$ ). Based on the data to hand, therefore, it remains difficult to assign a mode of ingress for these nuclides to the site.

### Conclusions

Levels of anthropogenic and natural radionuclides associated with an organic soil in a High Arctic environment as compared with corresponding levels in control soils indicate that the organic soil constitutes a significant sink for radionuclides in this environment. Activities of  $^{137}\text{Cs}$  and  $^{238}\text{U}$  are greater within the organic soil by a factor 20 and up to 40 times greater for Pu isotopes. Isotope ratios indicate that the source of much of the anthropogenic activity in these organic soils derives from global weapons test fallout. The ratios also

suggest that only  $^{238}\text{U}$ ,  $^{137}\text{Cs}$  and Pu isotopes are enriched within the organic soil, no evidence for enrichment of  $^{226}\text{Ra}$  or  $^{232}\text{Th}$  series nuclides being observed.

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## Uptake of radionuclides by vegetation at a High Arctic location

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Received 9 February 2004; accepted 31 May 2004

*Uptake of the anthropogenic radionuclide <sup>137</sup>Cs is highest for moss species.*

### Abstract

Radionuclide levels in vegetation from a High Arctic location were studied and compared to in situ soil concentrations. Levels of the anthropogenic radionuclide <sup>137</sup>Cs and the natural radionuclides <sup>40</sup>K, <sup>238</sup>U, <sup>226</sup>Ra and <sup>232</sup>Th are discussed and transfer factor (TF) values and aggregated transfer (Tag) values are calculated for vascular plants. Levels of <sup>137</sup>Cs in vegetation generally followed the order mosses > lichen > vascular plants. The uptake of <sup>137</sup>Cs in vascular plants showed an inverse relationship with the uptake of <sup>40</sup>K, with <sup>137</sup>Cs TF and Tag values generally higher than <sup>40</sup>K TF and Tag values. <sup>40</sup>K activity concentrations in all vegetation showed little correlation to associated soil concentrations, while the uptake of <sup>238</sup>U, <sup>226</sup>Ra and <sup>232</sup>Th by vascular and non-vascular plants was generally low.

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**Keywords:** Arctic; Vegetation; Radionuclides; Uptake; <sup>137</sup>Cs; Soil

### 1. Introduction

In terms of the radioecology of the terrestrial Arctic ecosystem, the uptake of radionuclides by vegetation has probably received the most attention, due to the potential food chain transfer to man, the relationship between Arctic inhabitants and specific indigenous herbivores (i.e. reindeer and caribou) and the impact of the Chernobyl Accident on areas within the Arctic (e.g. AMAP, 1998; Strand et al., 2002). Certain characteristics of the Arctic terrestrial environment and the vegetation growing in this region allow the accumulation of radionuclide contaminants within the resident vegetation. In the High Arctic, where the growing season is short and annual production, decomposition and overall turnover rates are low, the growth strategy

of Arctic plants is geared towards survival with modest annual growth (Bliss et al., 1973). Due to slow growth rates, Arctic plants have evolved longer life spans, so that the plants can accumulate enough energy to reach sexual maturity. However, longer life spans also provide greater opportunities for the uptake of contaminant species. Furthermore, Arctic plants tend to keep their leaves over winter, so no accumulated contaminants are lost through leaf fall, but are rather stored within the tissues from season to season, while the physical characteristics of certain Arctic plants (e.g. mosses and lichens), such as the lack of wax cuticles and root systems, permit accumulation of contaminants directly from the atmosphere and surface water.

Due to the lack of available water, slower rates of decomposition and the relative shallow depth of the active layer within Arctic soils, the redistribution of radionuclides in the terrestrial environment is retarded compared with more active temperate ecosystems. As

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a consequence, radionuclides in the High Arctic are subject only to localized transfers and generally decay in situ, increasing the ecological half-lives of radionuclides within the ecosystem (Taylor et al., 1985). Within the Arctic, attention has focused on the uptake of radionuclides by vegetation in the Low Arctic, principally on the transfer of  $^{137}\text{Cs}$  through the Lichen–Reindeer/Caribou–Human food chain (e.g. Liden, 1961; Hanson et al., 1975), with few studies concerned with vegetation in the High Arctic. This study reports the results of investigations conducted into the uptake of radionuclides by plant species in the Kongsfjorden area, Svalbard (78°55' N, 11°56' E).

Contamination of the Svalbard area by anthropogenic radionuclides is generally low, particularly in relation to more temperate latitudes. UNSCEAR (2000) report  $^{137}\text{Cs}$  deposition values of only 1.04 and 0.39 kBq/m<sup>2</sup> in the 70–80° and 80–90° latitude bands, respectively, compared to 4.8 kBq/m<sup>2</sup> in 40–50° latitude band. On Svalbard itself,  $^{137}\text{Cs}$  deposition from atmospheric weapon testing has been estimated, on the basis of soil/lichen sampling, at  $2.2 \pm 0.3$  kBq/m<sup>2</sup> (Hallstadius et al., 1982), though lower estimates exist between 0.2 and 0.54 kBq/m<sup>2</sup> from a survey of glacial ice cores, which showed higher deposition on the eastern side of Svalbard compared to the western side (Pinglot et al., 1994). Other studies report pre-Chernobyl soil  $^{137}\text{Cs}$  values of 1.51 and 1.59 kBq/m<sup>2</sup> (Kjos-Hanssen and Toerresdal, 1982), close to the higher global fallout estimate of Hallstadius et al. (1982), while Negoita (1997, 1999) reported typical post-Chernobyl  $^{137}\text{Cs}$  levels in soils between <1.5 and 35.8 Bq/kg, with one measurement of 213 Bq/kg. Svalbard is generally considered to have been relatively unaffected by fallout from the Chernobyl Accident, with the ice core survey of Pinglot et al. (1994) showing Chernobyl fallout levels for  $^{137}\text{Cs}$  of only 0.02 kBq/m<sup>2</sup>, a factor of 10 lower than levels from fallout due to atmospheric weapon testing. Levels of the natural radionuclides  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in soils in the Svalbard area display little variation (Dowdall et al., 2003) and are in broad agreement with average global concentrations (UNSCEAR, 2000).

In Svalbard vegetation, levels of  $^{137}\text{Cs}$  in Polar Willow (*Salix polaris*) and moss (unknown sp.) have been reported at 27 and 230 Bq/kg (Kjos-Hanssen and Toerresdal, 1982; Aarkrog et al., 1984), respectively, with levels of  $^{137}\text{Cs}$  in lichen (*Cladonia* sp.) of  $580 \pm 20$  Bq/kg (Holm et al., 1983). No data have been previously reported for U or Th series radionuclides in vegetation from the region.

## 2. Materials and methods

During summer field campaigns in 2001 and 2002 in Kongsfjorden, Svalbard, samples of flowering plants

(*Dryas octopetala*, *Cassiope tetragona* and *Silene acaulis*), grasses (*Deschampsia alpine* and *Carex nardina*), mosses (*Racomitrium ericoides*, *Sanonia uncinata*, *Amphidium lapponum* and *Bryum* sp.) and lichen (*Cetraria nivalis*) were taken together with soil samples from the same locations from which the vegetative samples were abstracted (Fig. 1). The surface parts of the plants were sampled using scissors and placed in polythene bags, frozen and returned to the laboratory. In all cases, care was taken to ensure that each sample consisted of only one plant species. Soil samples were collected with a clean steel shovel by removing the soil from a defined area, typically 20 × 20 cm and down to the underlying parent material, which in no cases exceeded 15 cm. Soil samples were placed in polythene bags, frozen and transported to the laboratory.

In the laboratory, vegetation was thoroughly rinsed in distilled water to remove any adhering detritus and remaining soil. Soil and vegetation samples were dried at 105 °C for 24 h, homogenised in a stainless steel laboratory blender and passed through a sieve of 2 mm aperture. Samples were then packed into standard plastic containers of volumes between 14 and 500 ml for gamma analysis. This analysis was conducted using a low background HPGe detector (Canberra Ind.) and an 8 k channel analyser. The system was calibrated using traceable standard isotope solutions and all results were corrected for matrix and spectral interferences.  $^{137}\text{Cs}$  was quantified using its 661 keV emission,  $^{40}\text{K}$  via its 1460 keV emission and U and Th series nuclides were quantified using methods described in Dowdall and O'Dea (1999). All samples were analysed according to current laboratory QA/QC procedures involving blanks, international reference materials (IAEA-156, 375, 368) and spikes.

At one site, selected as part of an ancillary study, soil chemistry parameters were measured for a series of soil samples taken below an extensive area of the moss *S. uncinata*. Soil pH was measured using 5 g aliquots of homogenised material in distilled water (1:2 w/v). Organic material was determined by loss on ignition (550 °C, 4 h), organic carbon being determined using the rapid dichromate oxidation technique of Nelson and Sommers (1982) involving oxidation of approximately 0.3 g of material with 1 N  $\text{K}_2\text{CrO}_7$  and  $\text{H}_2\text{SO}_4$  followed by titration with 0.2 N ferrous ammonium sulphate. Water soluble nitrate, phosphate, chloride and sulphate were assayed using ion chromatography. A sodium saturation method (Rhoades, 1982) was used to determine cation exchange capacity.

## 3. Results and discussion

Tabulated results of the activity concentrations of  $^{137}\text{Cs}$ ,  $^{40}\text{K}$ ,  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  in soils, vascular

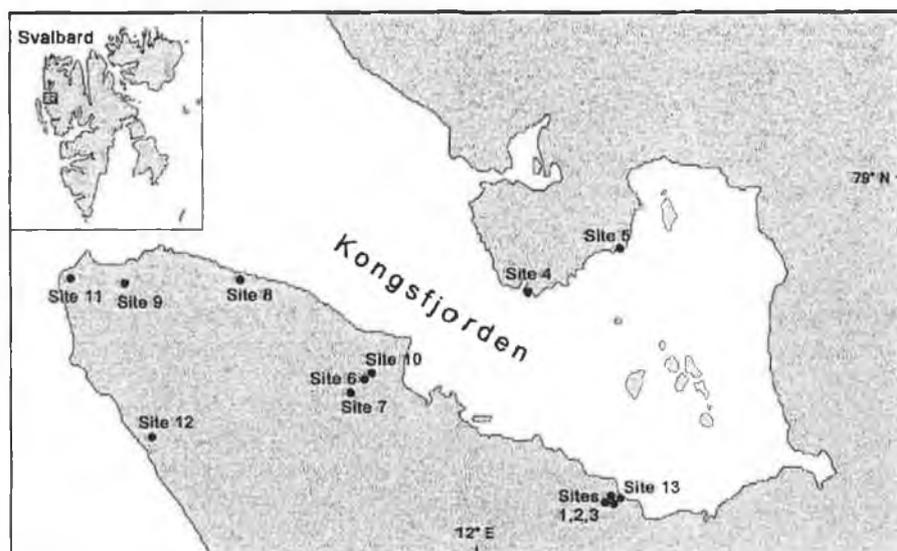


Fig. 1. Location of sampling sites within Kongsfjorden.

plants, mosses and lichen are provided in Tables 1, 2 and 3. Uptake factors and chemical parameters of relevance are presented in Tables 4 and 5.

For the anthropogenic radionuclide  $^{137}\text{Cs}$ , the exhibited activities in the samples displayed broad agreement with previously published values for the Svalbard region although the paucity of previous data makes comparison somewhat difficult. Levels in mosses and lichens were, as expected, significantly higher than values reported for vegetation in the Antarctic region (Mietelski et al., 2000) where the impact of atmospheric weapons testing and the Chernobyl incident were much less than for the Arctic. Levels of  $^{137}\text{Cs}$  in vegetation generally followed the order mosses > lichen > vascular plants, which is probably a reflection of different uptake mechanisms of this radionuclide. As mosses and lichens

lack a well-developed root system, uptake of radionuclides occurs mainly through ion-exchange processes directly from wet and dry deposition (Delfanti et al., 1999; Ugur et al., 2003). In contrast, vascular plants tend to absorb radionuclides from the soil upon which the plant is growing (Bell et al., 1988).  $^{137}\text{Cs}$  (along with U and Th series nuclides) has been shown to

Table 1  
Activity concentrations of radionuclides in soils at sites where vegetation samples were taken

Site	$^{137}\text{Cs}$	$^{40}\text{K}$	$^{238}\text{U}$	$^{226}\text{Ra}$	$^{232}\text{Th}$
1	58 ± 2	319 ± 11	163 ± 10	133 ± 15	23 ± 1
2	48 ± 2	441 ± 33	185 ± 10	248 ± 20	27 ± 1
3	123 ± 4	380 ± 31	134 ± 7	137 ± 12	23 ± 1
4	6 ± 2	542 ± 17	26 ± 7	38 ± 8	31 ± 1
5	39 ± 1	499 ± 37	35 ± 6	22 ± 10	31 ± 1
6	12 ± 1	413 ± 31	47 ± 2	35 ± 4	34 ± 1
7	15 ± 1	558 ± 21	34 ± 4	43 ± 5	35 ± 1
8	24 ± 1	431 ± 17	21 ± 4	47 ± 6	26 ± 1
9	10 ± 1	205 ± 15	38 ± 4	49 ± 6	14 ± 1
10	35 ± 1	220 ± 8	20 ± 3	32 ± 4	15 ± 1
11	5 ± 1	215 ± 16	24 ± 4	45 ± 7	15 ± 1
12	56 ± 2	78 ± 13	75 ± 9	24 ± 12	10 ± 1
13 (a)	163 ± 2	492 ± 7	92 ± 3	48 ± 3	30 ± 1

All values in Bq/kg dry weight and for all samples  $n = 1$ , except (a) where  $n = 12$  and average values are stated.

Table 2  
Activity concentrations of radionuclides in vascular plants

Site	Sample	$^{137}\text{Cs}$	$^{40}\text{K}$	$^{238}\text{U}$	$^{226}\text{Ra}$	$^{232}\text{Th}$
1	<i>Deschampsia alpine</i>	19 ± 1	350 ± 27	25 ± 6	42 ± 8	8 ± 1
2	<i>Deschampsia alpine</i>	14 ± 1	306 ± 24	40 ± 6	43 ± 9	4 ± 1
3	<i>Deschampsia alpine</i>	20 ± 1	256 ± 33	743 ± 40	164 ± 50	15 ± 2
4	<i>Dryas octopetala</i>	101 ± 3	46 ± 5	<12	<14	2 ± 1
	<i>Cassiope tetragona</i>	109 ± 3	52 ± 8	<17	<5	3 ± 1
	<i>Silene acaulis</i>	19 ± 1	88 ± 8	<11	<13	5 ± 1
	<i>Carex nardina</i>	64 ± 2	71 ± 27	<18	<7	10 ± 3
5	<i>Dryas octopetala</i>	31 ± 1	200 ± 11	<11	<25	10 ± 1
6	<i>Dryas octopetala</i>	62 ± 16	66 ± 30	<40	<56	<15
	<i>Silene acaulis</i>	47 ± 2	75 ± 18	<28	<20	<11
7	<i>Dryas octopetala</i>	77 ± 8	180 ± 31	24 ± 4	19 ± 5	20 ± 2
8	<i>Dryas octopetala</i>	34 ± 1	162 ± 11	<15	<20	11 ± 1
9	<i>Dryas octopetala</i>	64 ± 11	58 ± 19	<23	<31	<11

All values in Bq/kg dry weight and for all samples  $n = 1$ .

Table 3  
Radionuclide activities for samples of mosses and lichen

Soil	Sample	<sup>137</sup> Cs	<sup>40</sup> K	<sup>238</sup> U	<sup>226</sup> Ra	<sup>232</sup> Th
Moss						
3	<i>Bryum</i> sp.	11 ± 3	267 ± 62	<25	<31	<12
4	<i>Amphidium lapponum</i>	29 ± 1	47 ± 4	<7	<6	<4
7	<i>Sanonia uncinata</i>	117 ± 4	288 ± 17	<25	<30	18 ± 2
8	<i>Bryum</i> sp.	216 ± 36	104 ± 23	<24	<36	<14
10	<i>Racomitrium ericoides</i>	292 ± 70	<92	<14	<18	<10
11	<i>Bryum</i> sp.	166 ± 32	369 ± 19	<29	<36	<14
12	<i>Bryum</i> sp.	124 ± 29	42 ± 24	<32	<39	<12
13	<i>Sanonia uncinata</i> (a)	37 ± 1	337 ± 7	72 ± 3	42 ± 3	21 ± 1
Lichen						
4	<i>Cetraria nivalis</i>	140 ± 4	60 ± 8	<17	<20	4 ± 1
5	<i>Cetraria nivalis</i>	75 ± 2	57 ± 11	<18	<20	20 ± 1

All values in Bq/kg dry weight and for all samples  $n = 1$ , except (a) where  $n = 12$  and average values stated.

demonstrate a low tendency towards mobility in this High Arctic environment (Dowdall et al., 2003) which may reflect the lower uptake exhibited by vascular plants.

Transfer factor (TF) values and aggregated transfer (Tag) values are commonly encountered means of quantifying and expressing plant uptake of radionuclides. Dimensionless TF values can be conveniently defined as the ratio of the activity concentrations of plant material and the soil upon which the plant has grown

$$TF = \text{Bq/kg plant material (d.w.)} / \text{Bq/kg soil (d.w.)}$$

Tag values are defined as the ratio of the radionuclide activity concentration in an organism (Bq/kg d.w. or

w.w.) to the areal deposition of the nuclide in soil (Bq/m<sup>2</sup>) and are expressed in units of m<sup>2</sup>/kg

$$\text{Tag (m}^2/\text{kg)} =$$

$$\text{Bq/kg plant material (d.w. or w.w.)} / \text{Bq/m}^2$$

In an attempt to compare values, the TF values obtained in this work were converted to aggregated transfer (Tag) values by taking the average soil sampling depth to be 10 cm and the soil bulk density to be 0.78 g/cm<sup>3</sup> (Batjes, 1995). <sup>137</sup>Cs is predominantly contained within the top 10 cm of soils in Svalbard, while <sup>40</sup>K, <sup>238</sup>U, <sup>226</sup>Ra and <sup>232</sup>Th display relatively uniform vertical soil distributions (Dowdall et al., 2003).

Soil TF values for <sup>137</sup>Cs in all vascular plants ranged between 19.1 and 0.16 and showed considerable variation within the same species, sampled from different locations, while <sup>137</sup>Cs Tag values for the vascular plants in this study were in the range of 0.002 to 0.25 m<sup>2</sup>/kg (Table 4), which is in good agreement with a previously published range of 0.006 to 0.18 m<sup>2</sup>/kg for comparable vegetation in the Low Arctic (IRNEG, 2002). It should be noted however that the effect of age differences between the samples has not been used in assessing uptake of this isotope. A Spearman correlation coefficient of -0.619 ( $p = 0.24$ ) indicates a weak inverse relationship between levels of <sup>137</sup>Cs in soil and vascular plants in this study, whereas it is generally assumed that plant:soil relationships for metallic species can be described by an increase in plant concentrations with an increase in soil concentrations. However, the uptake of cesium by vascular plants can be influenced by a number of factors and this observation may be an artefact of a secondary mechanism.

Further investigation of uptake in vascular plants is facilitated by a plot of <sup>137</sup>Cs Tag values against <sup>40</sup>K Tag values (Fig. 2) which shows a clear inverse relationship

Table 4  
Transfer factor (TF) values and aggregated transfer (Tag; m<sup>2</sup>/kg d.w.) values for <sup>137</sup>Cs, <sup>40</sup>K, <sup>238</sup>U, <sup>226</sup>Ra and <sup>232</sup>Th in vascular plants

Site	Sample	<sup>137</sup> Cs		<sup>40</sup> K		<sup>238</sup> U		<sup>226</sup> Ra		<sup>232</sup> Th	
		TF	Tag	TF	Tag	TF	Tag	TF	Tag	TF	Tag
1	<i>Deschampsia alpine</i>	0.32	0.0042	1.10	0.014	0.2	0.0020	0.32	0.0041	0.33	0.0043
2	<i>Deschampsia alpine</i>	0.29	0.0038	0.69	0.0090	0.2	0.0028	0.17	0.0023	0.14	0.0018
3	<i>Deschampsia alpine</i>	0.16	0.0021	0.67	0.0088	5.5	0.072	1.20	0.016	0.64	0.0084
4	<i>Dryas octopetala</i>	17.7	0.23	0.08	0.0011	<0.4	<0.0060	<0.4	<0.0048	0.07	0.0010
	<i>Cassiope tetragona</i>	19.1	0.25	0.10	0.0012	<0.6	<0.0085	<0.2	<0.0017	0.09	0.0012
	<i>Silene acaulis</i>	3.26	0.042	0.16	0.0021	<0.4	<0.0055	<0.4	<0.0045	0.17	0.0023
	<i>Carex nardina</i>	11.3	0.15	0.13	0.0017	<0.7	<0.0090	<0.2	<0.0024	0.33	0.0043
5	<i>Dryas octopetala</i>	0.79	0.010	0.40	0.0052	<0.3	<0.0042	<1.2	<0.0149	0.32	0.0041
6	<i>Dryas octopetala</i>	5.39	0.070	0.16	0.0021	<0.8	<0.011	<1.6	<0.0207	<0.5	<0.0058
	<i>Silene acaulis</i>	4.10	0.053	0.18	0.0024	<0.6	<0.0077	<0.6	<0.0074	<0.3	<0.0043
7	<i>Dryas octopetala</i>	5.31	0.069	0.32	0.0042	0.7	<0.0092	0.45	<0.0058	0.57	0.0074
8	<i>Dryas octopetala</i>	1.45	0.019	0.38	0.0049	<0.7	<0.0093	<0.4	<0.0055	0.43	0.0056
9	<i>Dryas octopetala</i>	6.55	0.085	0.28	0.0037	<0.6	<0.0079	<0.6	<0.0083	<0.8	<0.0102

For all samples  $n = 1$ .

Table 5  
Soil chemistry parameters of soil samples associated with the moss *S. uncinata* ( $n = 12$ )

	pH	Organic matter (%)	Organic carbon (%)	Nitrate (mg/kg)	Phosphate (mg/kg)	CEC (meq/100 g)
Average	6.26	33.4	12.5	11.7	118.3	79.2
Max	6.60	40.7	16.5	83	320.1	139.7
Min	5.82	22.5	7.1	0	0	40.7

(Spearman correlation coefficient of  $-0.9$  ( $p < 0.01$ )) suggesting that the uptake of  $^{137}\text{Cs}$  is controlled to some extent by the uptake of potassium. Indeed, similar observations have been made for various plant species from other climatic regions (e.g. Nishita et al., 1961; Strebl et al., 2002; Frissel et al., 2002). It is interesting to note however, that except for the three samples of *D. alpine*, a grass species,  $^{137}\text{Cs}$  Tag and TF values were higher than  $^{40}\text{K}$  Tag and TF values in all cases. This would suggest that  $^{137}\text{Cs}$  is perhaps more readily available for uptake in this High Arctic location than  $^{40}\text{K}$ , although the lack of High Arctic specific data pertaining to uptake of  $^{137}\text{Cs}$  in the vascular plant species studied makes meaningful comparison of the results of this study with accepted values difficult.

A lower correlation between levels of  $^{137}\text{Cs}$  in mosses and lichen and soil (Spearman correlation coefficient of  $-0.431$  ( $p = 0.058$ )) emphasizes the different uptake mechanisms of these plants. Furthermore, levels of  $^{137}\text{Cs}$  in samples of the moss *S. uncinata* showed little correlation with a range of soil chemistry parameters of the underlying soil (Table 5), with the exception of a weak correlation with the soil's cation exchange capacity (Spearman correlation coefficient of  $0.608$  ( $p = 0.036$ )). Only a very low correlation (Spearman correlation coefficient of  $-0.243$  ( $p = 0.4$ )) was determined for

$^{40}\text{K}$  in vascular plants and associated soil, suggesting that the uptake of this nuclide is not related to the soil concentration, although this may be due to the fact that  $^{40}\text{K}$  and stable potassium are probably in excess in the surrounding soil.

The uptake of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  by vascular plants is generally low, with the exception of anomalously high values for  $^{238}\text{U}$  and  $^{226}\text{Ra}$  for a grass species growing on a soil exhibiting relatively high levels of these isotopes. Due to the low level of uptake of these nuclides, the majority of TF and Tag values for  $^{238}\text{U}$  and  $^{226}\text{Ra}$  are "less than" values, TF values typically being  $<0.7$  and  $<0.6$ , respectively, with Tag values two orders of magnitude lower. TF values for detectable levels of  $^{232}\text{Th}$  ranged from 0.07 to 0.64, with Tag values ranging from 0.001 to 0.0084. Overall, these TF values are in good agreement with values reported for other Arctic vascular plants (Verhovskaya, 1972; Litver et al., 1976) and for vascular plants from more temperate regions (Linsalata et al., 1989; Vera Tome et al., 2003).

The levels of  $^{40}\text{K}$  in mosses and lichen were comparable to levels observed in the vascular plants and showed no correlation to soil concentrations of this nuclide, nor to concentrations of  $^{137}\text{Cs}$  within the mosses and lichen.  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  activity concentrations in mosses and lichen were generally below the level of detection, although detectable levels of these nuclides were found in samples of the moss *S. uncinata* at one site. The lack of detectable levels of these nuclides in mosses and lichen is probably due to the way in which these plants absorb radionuclide contaminants; i.e. via contact with water rather than from the underlying substrate. However, the detectable activity concentrations of  $^{238}\text{U}$  in *S. uncinata* showed good correlation with the levels of  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  within the moss and strong correlation with the distribution of this nuclide in the underlying soil (Spearman correlation coefficient  $0.873$  ( $p < 0.01$ )). Where sufficient data existed, no correlation was determined between the levels of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  in the moss *S. uncinata* and the underlying soil chemistry parameters.

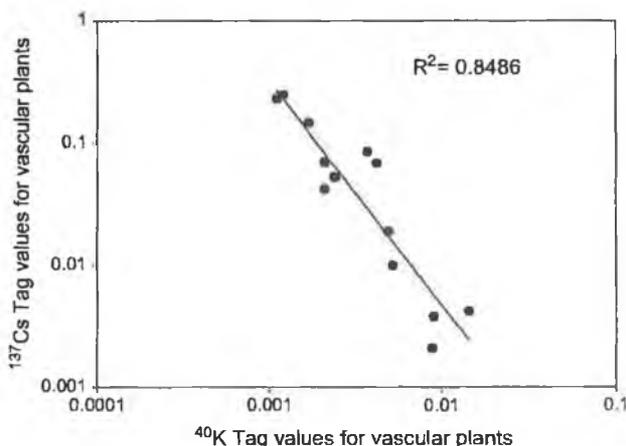


Fig. 2. Plot of  $^{137}\text{Cs}$  versus  $^{40}\text{K}$  aggregated transfer (Tag) values for vascular plants.

#### 4. Conclusion

The results of this study indicate that for the High Arctic environment studied, uptake of the anthropogenic radionuclide  $^{137}\text{Cs}$  is highest for moss species and lowest for the vascular species, this being most probably due to the differing modes of uptake exhibited by these species. The difference in uptake is further exemplified by the observed inverse relationships between the uptake of  $^{137}\text{Cs}$  and  $^{40}\text{K}$  for vascular species which are absent for mosses and lichens. Uptake of the natural

radionuclides of the U and Th series is very low in all species.

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