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Persistent pollutants in fresh and abandoned eggs of Common Tern (*Sterna hirundo*) and Arctic Tern (*Sterna paradisaea*) in Ireland

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ABSTRACT

Higher levels of persistent pollutants (Σ16PCB, Σ6PBDE, ΣHCH, ΣDDT, ΣCHL) were detected in fresh eggs of Common Terns *Sterna hirundo* from Rockabill Island near Dublin (Ireland's industrialised capital city) compared to Common and Arctic Terns *S. paradisaea* from Ireland's west coast. Intra-clutch variation of pollutant levels in Common Terns was shown to be low, providing further evidence that random sampling of one egg may be an appropriate sampling strategy. Significant differences in pollutant concentrations were detected between fresh and abandoned eggs on Rockabill. However, abandoned eggs can still provide a useful approximation of pollutants in bird eggs if non-destructive sampling is preferred. Levels of *p,p'* –DDE in tern eggs have decreased over time according to this study, in concurrence with worldwide trends. Results in this study fall below toxicological thresholds for birds and OSPARs EcoQO thresholds set for Common Tern eggs, except for mercury and HCH in the west coast.

1. Introduction

European Union (EU) Member States are mandated to monitor and assess the quality of their marine waters under the Marine Strategy Framework Directive (MSFD) (2008/56/EC), EU legislation that requires Member States achieve good environmental status (GES) of their marine waters (MSFD, 2008). Descriptor 8 of the MSFD concerns protection of marine waters against pollution by chemical contaminants. To achieve these aims, the MSFD foresees Member States and third countries cooperating on a regional basis through Regional Sea Conventions, such as the OSPAR Convention for Protection of the North East Atlantic (OSPAR, 2012). Seabird eggs are considered an effective matrix for monitoring trends of marine contaminants (OSPAR, 2007). OSPAR Ecological Quality Objectives (EcoQO) for persistent pollutants (mercury and organochlorines) in the eggs of the Common and Arctic Tern (*Sterna hirundo, S. paradisaea*), with recommended thresholds, were applied in assessments of the contamination status of the North Sea (Dittmann et al., 2011). EcoQOs function both as indicators (to provide specific variables for monitoring) and objectives (against which to measure progress) but are not thresholds of toxicological concern (Kroes et al., 2005; OSPAR, 2010).

Persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and other organochlorine compounds (OCs), as well as metals, have been shown to exhibit a wide range of toxic properties in birds (Burger and Gochfeld, 2001; Walker et al., 2012; Winter et al., 2013). These pollutants can be moved by long-range atmospheric transport and a process known as global distillation to locations distant from point of input (Fernández and Grimalt, 2003; Wania and MacKay, 1996). In western Europe, elevated concentrations of contaminants in marine environments have been linked to localised inputs from heavily industrialised areas (Breivik et al., 2007; Jepson et al., 2016).

Common and Arctic Terns are closely related, similar in size, occupy the same dietary niche and have similar foraging ranges (<30 km from breeding site) (Thaxter et al., 2012). In the report on the pilot study for the EcoQO for coastal bird eggs Dittmann et al. (2011) demonstrated

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that pollutants in the eggs of Arctic and Common Terns are similar, suggesting that Arctic Terns are a suitable alternative in areas where Common Terns are scarce. Both Common and Arctic Terns are migratory and have different wintering grounds (Cabot and Nisbet, 2013). However, contaminants found in the eggs of Common and Arctic Terns reflect the contaminant composition of the diet of an adult female during the egg formation period (income breeding) (Bond and Diamond, 2010). Both tern species feed predominantly on small fish (Green, 2017). During the egg formation period lipophilic persistent pollutants ingested by the adult female are passed into the developing egg with lipid reserves that are essential for the development of the embryo (Speake et al., 1998). Their eggs give an indication of levels in contaminant in the local marine environment, close to the breeding colony in a higher trophic level predator. This is advantageous for the detection of biomagnifying contaminants that are only found in small concentrations in the ecosystem (Dittmann et al., 2011). While Common and Arctic Terns are higher trophic level predators they occupy a lower trophic position than other piscivorous seabirds breeding in Ireland, such as the Northern Gannet *Morus Bossanus* and Guillemot *Uria aalge* that can feed on larger prey (Ainley et al., 2020; Lewis et al., 2003). There have been no published studies on contaminant levels in Common or Arctic Tern eggs in Ireland to date.

The clutch size of Common and Arctic Terns is typically 2–3 eggs (Cabot and Nisbet, 2013). OSPAR sampling guidelines recommend taking one fresh egg, randomly, from a clutch of 3 eggs (OSPAR, 2014) as intra-clutch variation of contaminants is low compared to interclutch (Becker et al., 1991). The use of fresh eggs is preferable in contaminant studies as embryo development may alter the concentration and composition of pollutants present in the egg (Drouillard et al., 2003; Klein et al., 2012). Abandoned eggs may contain embryos but can be unfertilized, addled, come from inexperienced breeders, or damaged physically which may also alter the makeup of pollutants present. However, there are few studies examining this and what changes may occur are unclear. Abandoned eggs can provide representative measurements of pollutants according to some investigations (Bustnes et al., 2015) and may be especially useful in determining pollutant levels in protected, endangered or at risk species where sampling may not be possible. Abandoned bird eggs have been used as indicators of pollutants in several studies and species, including seabirds and the Common Tern (Evers et al., 2003; Gochfeld and Burger, 1998; Ikemoto et al., 2005; Jaspers et al., 2005; Switzer et al., 1971). There have been few studies comparing fresh and abandoned eggs from the same species at the same time and location.

Intra-specific feeding strategies and diet of seabirds can vary between colonies as well as at an individual level which can create difficulties when comparing contaminant burdens between different individuals, sites and years. Stable isotope ratio analysis of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) can be used to determine the differences, if any, in dietary niche and trophic level of individual seabirds (Bond and Jones, 2009). These may be important considerations when interpreting the suitability of seabird eggs as a monitoring organism for the assessment of marine levels of contaminants given the high dispersal ability and wide dietary niche of many seabird species.

This research is part of a larger study to determine the feasibility of using seabirds as a higher trophic level indicator of contaminants in the Irish marine environment. The primary aims of this research are to: 1) provide and compare contemporary baseline data on the concentrations of persistent pollutants from tern colonies with divergent history of industrialization in Ireland, supported by stable isotope ratio analysis; 2) investigate intra-clutch variation of pollutants in tern eggs; 3) compare pollutant data from tern eggs against OSPAR EcoQO thresholds, known toxic thresholds and historical and contemporary concentrations found in tern eggs from other studies; 3) examine the feasibility of using abandoned tern eggs as a non-destructive alternative to the sampling of fresh eggs.

2. Methods

2.1. Sample collection

All samples in this study were collected under licence from the Irish National Parks and Wildlife Service, and in accordance with OSPAR Joint Assessment and Monitoring Programme guidelines (OSPAR, 2014). According to OSPAR requirements study sites should include both industrial areas as well as more pristine sites, less affected by industrialization (Dittmann et al., 2011). Rockabill Island (latitude: 53.597, longitude: −6.00354), located in the Irish Sea on the east coast of Ireland, eight km off the north coast of County Dublin (Fig. 1) was chosen as a sampling site due to its close proximity to Ireland's capital and largest city, Dublin which has an industrial history (Glennon et al., 2014). On the 30th of May 2017, 20 single Common Tern eggs were collected from 20 different three egg clutches on Rockabill Island. Three complete clutches of three eggs and 10 abandoned eggs were also collected from Rockabill on that date. Abandoned tern eggs on Rockabill Island are abundant and easy to find due to the large Common Tern population (> 2000 breeding pairs) and small breeding site (Nicholas et al., 2020). Abandoned eggs were chosen without any obvious damage to eggshell, or loss of weight where yolk and albumen had not desiccated.

In order to collect 20 eggs from areas less impacted by industrialization, multiple sites were sampled on the west coast of Ireland as colony sizes are much smaller than Rockabill Island and more vulnerable to the impacts of sampling. On the 1st and 3rd of June 2018, two Common Tern eggs were collected from Mutton Island (latitude: 53.25443, longitude: −9.05489) and on the 26th of June a further three eggs were collected from Rabbit Island (latitude: 53.26228, longitude: −9.00961) from Galway Bay on the west coast of Ireland. Mutton and Rabbit island are approximately 3 km apart and are referred to collectively as inner Galway Bay islands in this study. On the 6th and 12th of June 2018, 15 Arctic Tern eggs were collected from the Inishkea Islands (latitude: 53.13346, longitude: −10.19422), situated in the north-east Atlantic Ocean off the west coast of Ireland (Fig. 1).

To ensure that only fresh eggs were collected, with the exception of abandoned eggs, samples were placed in a small container of water (per OSPAR guidelines) to check if the eggs had been laid recently, as fresh eggs sink in fresh water (JAMP; OSPAR, 2014). Selected eggs were wrapped carefully in aluminium foil, placed in a sealed bag. Each individual egg was labelled and the time, date and exact location (GPS coordinates) were recorded. Eggs were immediately refrigerated on the day of sampling and frozen the following day at −20 °C.

2.2. Sample preparation

All samples were later thawed so the contents of each egg could be homogenised and subsampled according to analysis type. Egg contents (yolk and albumen) were homogenised using an Ultra-Turrax® (IKA T25, Germany). Four sub-samples were taken from each egg and placed in two acid washed containers for metals and mercury analysis, respectively, and two solvent washed (*n*-hexane) jars for the analysis of POPs and stable isotope ratio analysis. Each egg was individually analysed for PCBs, PBDEs and OCs but pooled for metals and mercury analysis as the amount of homogenate was small for each egg (approximately 15 g). A pooled sample was created for eggs from Rockabill ($n = 20$), Inishkea Islands ($n = 15$) and inner Galway Bay islands ($n = 5$). Abandoned Common Tern eggs from Rockabill Island were also pooled ($n = 10$). Sample jars were then frozen at −20 °C until analysis. Individual egg sub-samples for metals and stable isotope ratio analysis were freeze dried (Labconco: Freeze Dryer – Model Freezone & Bulk Tray Drier (12*L*), USA).

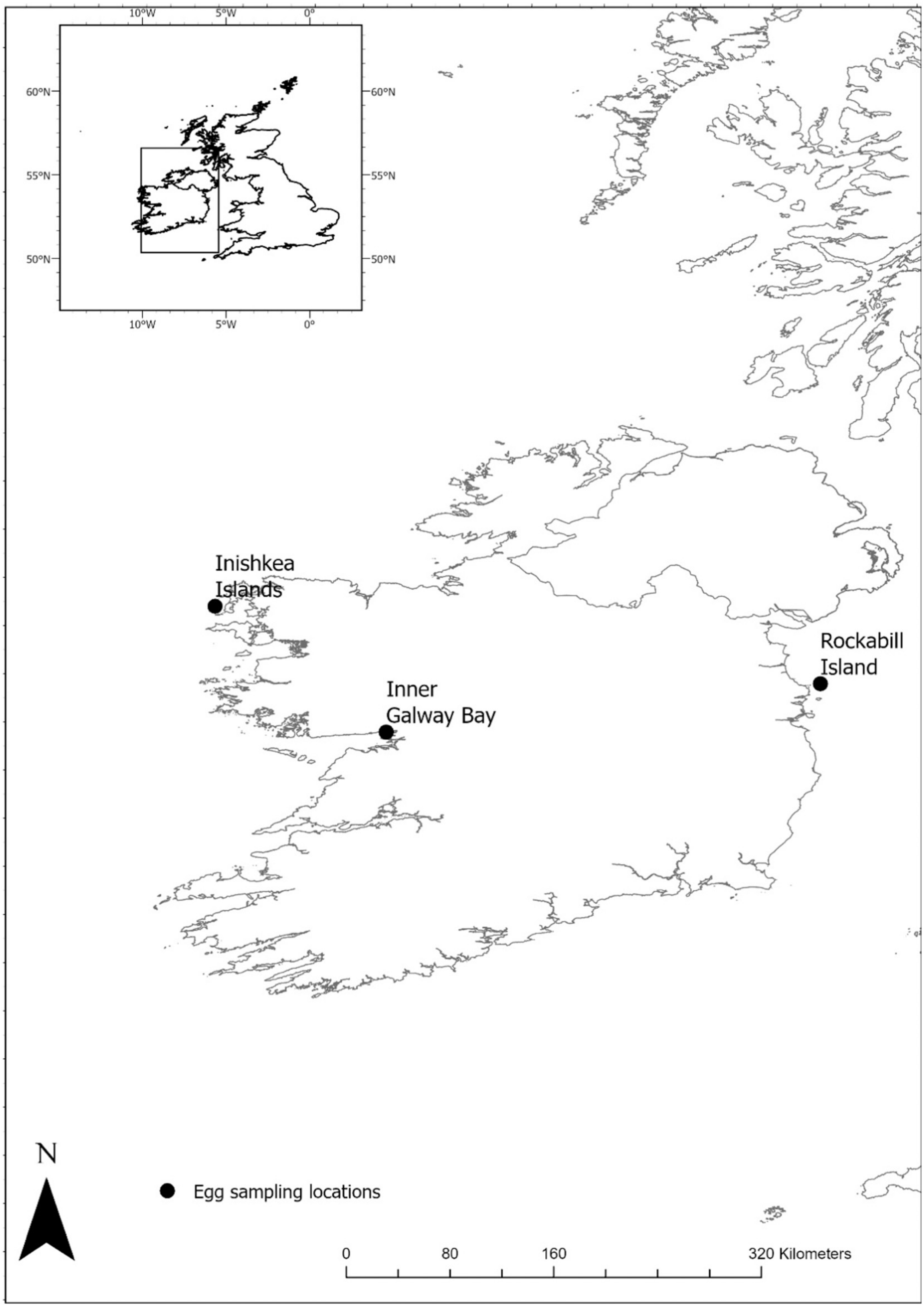

Fig. 1. Location of Rockabill Island, inner Galway Bay islands and Inishkea island tern colonies.

2.3. Contaminant analysis

2.3.1. Quality assurance

A comprehensive analytical quality assurance programme underpinned laboratory analyses. Reagent blanks, a certified reference material (CRM) and a laboratory reference material (LRM) were included in each batch of samples as quality controls. Egg homogenate from Great Black-backed Gull *Larus marinus*, that was collected from another study, was used as an LRM as no suitable seabird egg reference materials are currently commercially available. Fish tissue (NIST 1947, Lake Michigan Fish Tissue) was used as CRM for analysis of POPs and mussel tissue (Freeze-Dried, SRM 2976) for metals and mercury analysis.

As an additional quality control, a selection of samples were analysed in the Marine Institute laboratory (Co. Galway, Ireland), a state agency with a track record of successful participation in QUASIMEME (Quality Assurance of Information for Marine Environmental Monitoring in Europe) proficiency exercises for the analysis of POPs. This showed close agreement between the two laboratories.

2.3.2. PCBs, PBDEs and OCs analysis

Eggs were analysed for a suite of 16 PCB congeners (18, 28, 31, 52, 44, 101, 105, 149, 118, 105, 138, 153, 156, 170, 180, 209). A subset of these congeners, the Σ7PCB, also known as the ICES-7, refers to the sum of seven individual PCB congeners (28, 52, 101, 118, 138, 153, 180) that are widely used as indicators of PCB contamination in the marine environment (Webster et al., 2013). Eggs were analysed for 6 PBDE congeners (28, 47, 99, 100, 153, 154) and 17 OCs (HCBD, HCB, α-HCH, γ-HCH, β-HCH, heptachlor, heptachlor epoxide, oxychlordane, trans-chlordane, cis-chlordane, transnonachlor, *o, p*' –DDE, *p,p*' –DDE, *o,p*' –DDT, *o'p*' –DDD, *p,p*' –DDD, *p,p*' -DDT). ΣHCH refers to the sum of α-HCH, γ–HCH and β-HCH. ΣCHL refers to the sum of heptachlor, heptachlor epoxide, oxychlordane, trans-chlordane, cis-chlordane and transnonachlor. ΣDDT refers to the sum of *o,p*' –DDE, *p,p*' –DDE, *o,p*' –DDT, *o'p*' –DDD, *p,p*' –DDD and *p,p*' -DDT. Egg samples were extracted using the Smedes' lipid extraction techniques (i.e. 'Total' Lipid) (Smedes and Askland, 1999). All lipid concentrations were determined gravimetrically. Column chromatography, using alumina and silica to perform clean-up, was completed prior to analysis to remove lipids. All samples were spiked with internal standards (^{13}C isotopically labelled internal standards for PCBs, PBDEs and OCs). An Agilent 6890 gas chromatograph (GC) coupled to a 5973 N mass selective detector (MSD) with a 30 m DB5-MS column was used for these analyses. Oven temperature programming was used to achieve resolution of analyte peaks. Single ion monitoring (SIM) mode was used for the quantification of analytes. Electron ionisation (EI) methods were used with helium as a carrier gas. Analysis of all calibration standards and samples in SIM mode allowed for increased specificity and sensitivity. Further details on analytical methods used can be found in the supplementary materials. Where applicable, results were compared to OSPAR EcoQO thresholds for pollutants in tern eggs (OSPAR, 2010).

2.3.3. Metal analysis

Pooled samples were analysed for a suite of 14 metals (arsenic, cadmium, chromium, copper, lead, nickel, silver, zinc, aluminium, cobalt, iron, manganese, selenium and vanadium). Concentrated nitric acid (4 ml) and hydrogen peroxide (4 ml) were added to approximately 0.2 g freeze-dried egg homogenate, which was then digested in a laboratory microwave oven (CEM Mars Xpress). After cooling, samples were diluted to 50 ml with deionised water. Metal concentrations were determined by ICP-MS (Agilent 7700× with High Matrix Introduction (HMI) system).

2.3.4. Total mercury analysis

Concentrated nitric acid (4 ml) was added to 0.6–0.8 g of egg homogenate, which was then digested in a laboratory microwave oven (CEM Mars Xpress). The microwave digestion procedure is based on a method developed by Hatch and Ott (1968). After cooling, potassium permanganate was added until the purple colour of the solution stabilised. Sufficient hydroxylamine sulphate / sodium chloride solution was added to neutralise the excess potassium permanganate and potassium dichromate was added as a preservative. The solution was diluted to 100 ml with deionised water. Following reduction of the samples with tin (II) chloride, mercury concentrations were determined by Cold Vapour Atomic Fluorescence Spectroscopy (CV-AFS) using a PSA Millennium Merlin Analyser. Results were compared to OSPAR EcoQO thresholds for mercury in tern eggs (OSPAR, 2010).

2.4. Stable isotope ratio analysis

Stable isotope ratio analysis of eggs was used in this study to investigate the trophic position and foraging niche of adult female terns (Fig. 3). Egg samples were homogenised and freeze dried. The isotopic composition of organic carbon and nitrogen was then measured in 50 tern egg samples (all samples excluding 9 eggs sampled for intra-clutch variation) by Iso-Analytical Limited (Crewe United Kingdom) using Elemental Analysis - Isotope Ratio Mass Spectrometry (EA-IRMS). Replicate samples of soy protein, l-alanine and tuna protein were also analysed as quality control. Variation in lipid content can confound interpretations of diet as lipids are depleted in δ^{13}C compared with protein (Elliott et al., 2014). δ^{13}C values were corrected using a lipid normalisation equation for aquatic bird egg (Elliott and Elliott, 2016).

2.5. Statistical analysis

Concentrations of POPs were calculated on a wet weight (ww) and lipid weight (lw) basis in ng/g. Organic pollutants concentrate more readily in the lipids of animal tissue and lw is advocated as superior to ww in the measurement of persistent lipophilic chemicals (Brown and Lawton, 1984). However, many studies report solely in ww (Gewurtz et al., 2011). Concentrations for metals were calculated in mg/kg ww and dry weight (dw). Concentrations below the limit of detection (LoD) and compounds not detected were assigned a value of half the LoD (Pereira et al., 2009). Statistical analysis was performed using Graphpad Prism 9. Shapiro-Wilk tests were used to assess normality, due to small sample sizes.

Differences in pollutant concentrations between colonies were assessed using ordinary one-way ANOVA with Tukey's multiple comparison post-hoc test for parametric data, and Kruskal Wallis test with Dunn's multiple comparison post-hoc test for nonparametric data. Direct comparisons between fresh and abandoned eggs from Rockabill Island were tested using Mann-Whitney *U* test for non-parametric data and unpaired *t*-test for parametric data.

Variation within complete clutches of Common Tern eggs was compared to the variation among separate clutches ($n = 20$). Intraclass correlation coefficient (ICC) was used as a measure of the repeatability of measurements within clutches (Lessells and Boag, 1987; Wolak et al., 2012). This is the ratio of the intra-clutch variance to the total variance. Intraclass correlation coefficients and their confidence intervals (CIs) were estimated using the R library ICC (Wolak et al., 2012), using the Searle (1971) method (Donner, 1986; Donner and Wells, 1986). Each variable was $\log_{10}$ transformed as estimates of variance can be sensitive to the normality assumption.

3. Results

3.1. Pollutant profiles (PCBs, PBDEs, OCs and metals)

All 16 PCB congeners analysed for in this study were detected in tern eggs (Table 1) with mean concentrations ranging from 186 ng/g ww in eggs from Rockabill Island (range: 102–381 ng/g ww) to 56.5 ng/g ww in eggs from Inishkea Islands (range: 23.4–175 ng/g ww) and 81.9 ng/g ww in eggs from inner Galway Bay islands (range: 20.7–145) ng/g ww). The mean concentration of Σ7PCB was 159 ng/g ww in eggs from Rockabill Island, 48.9 ng/g ww in eggs from Inishkea Islands and 71.9 ng/g ww in eggs from inner Galway Bay islands. PCB-153 was the most dominant congener and accounted for 34.4% of total Σ16PCB ww in Rockabill Island, 40.4% for Inishkea Islands and 43% for the inner Galway Bay islands. Significantly higher levels of Σ16PCB were detected in Rockabill Island when compared to the Inishkea Islands and inner Galway Bay islands for lw concentrations ($P < 0.0001$, $P = 0.0635$). No differences were observed in PCB concentrations between Inishkea Islands and inner Galway Bay islands. However, significantly higher levels of Σ16PCB were detected in inner Galway Bay Islands compared to the Inishkea Islands for both ww concentrations ($P = 0.0402$) and lw ($P = 0.0193$) when analysed independently of Rockabill Island.

All six PBDE congeners were detected in tern eggs (Table 2). The mean concentration of Σ6PBDE ww in eggs from Rockabill Island was 10.5 ng/g ww (range: 4.66–33.5 ng/g ww), 3.37 ng/g ww in eggs from Inishkea Islands (range: 1.9–4.7 ng/g ww) and 3.88 ng/g in eggs from inner Galway Bay islands (range: 3.4–5.2). The dominant congener in all sites was BDE-47, accounting for 44.6%, 58.9% and 51.6% of the

Table 1
Mean PCB concentrations, standard error of the mean (SEM) and median for tern eggs from Rockabill Island ($n = 20$), Inishkea Islands ($n = 15$) and inner Galway Bay islands ($n = 5$). Seven PCBs (Σ7) are −28, −52, −101, −118, −138,−153, and −180.

	Rockabill Island (n = 20)		Rockabill Island (abandoned) ($n = 10$)		Inishkea Islands (n = 15)		Inner Galway Bay islands (n = 5)	
	Mean ww (SEM) Median	Mean lw (SEM) Median	Mean ww (SEM) Median	Mean lw (SEM) Median	Mean ww (SEM) Median	Mean lw (SEM) Median	Mean ww (SEM) Median	Mean lw (SEM) Median
PCB-18	0.54 (0.06) 0.47	6.6 (0.76) 6.43	0.41 (0.05) 0.41	5.26 (0.76) 4.59	0.25 (0.04) 0.18	2.39 (0.41) 1.99	0.01 (0.004) 0	0.06 (0.06) 0
PCB-28	2.34 (0.12) 2.28	28.3 (1.63) 30.29	1.29 (0.14) 1.09	16 (1.65) 15.86	0.84 (0.07) 0.84	8.19 (0.78) 7.83	0.57 (0.04) 0.55	6.33 (0.42) 6.1
PCB-31	0.82 (0.05) 0.78	9.97 (0.62) 9.95	0.09 (0.02) 0.08	1.01 (0.17) 0.95	0.09 (0.02) 0.05	0.9 (0.26) 0.44	0.09 (0.01) 0.1	0.94 (0.15) 1.12
PCB-52	0.49 (0.04) 0.50	5.87 (0.48) 5.40	0.61 (0.17) 0.36	7.44 (1.9) 4.12	0.47 (0.09) 0.4	4.48 (0.83) 3.79	0.01 (0.00) 0	0.06 (0.04) 0
PCB-44	1.42 (0.1) 1.32	17.2 (1.23) 16.87	0.28 (0.05) 0.26	3.41 (0.59) 3.49	0.24 (0.09) 0.12	2.27 (0.77) 1.04	0.00 (0.00) 0	0.00 (0.00) 0
PCB-101	6.28 (0.70) 5.59	73.8 (7.35) 62.13	3.14 (0.54) 2.69	38.2 (5.67) 33.64	1 (0.2) 0.84	9.64 (1.82) 7.82	0.95 (0.11) 0.99	10.5 (0.93) 10.9
PCB-118	14.5 (1.39) 13.5	173 (15.1) 150.0	9.99 (1.74) 8.8	119 (17.1) 100.3	5.33 (0.5) 4.89	52.67 (6.11) 50.1	6.74 (0.98) 6.26	74.1 (8.75) 76.1
PCB-105	3.64 (0.49) 3.04	42.9 (5.22) 36.49	2.90 (0.51) 2.72	34.4 (5.03) 29.50	1.44 (0.16) 1.27	14.38 (1.94) 13.8	1.98 (0.29) 1.75	21.73 (2.62) 21.2
PCB-149	3.56 (0.46) 2.99	41.7 (4.87) 34.95	2.46 (0.73) 1.73	29.8 (7.89) 20.17	0.57 (0.14) 0.54	5.45 (1.24) 5.61	0.48 (0.05) 0.53	5.31 (0.55) 5.43
PCB-153	64.3 (5.80) 59	774 (68.3) 706	45.2 (8.84) 35	535 (90.6) 394	22.6 (3.27) 20	220 (31.4) 185	35.8 (8.13) 27	387 (73.1) 298
PCB-138	48.6 (4.58) 44	589 (56.4) 489	30.8 (5.14) 26	366 (51.2) 302	10.9 (1.23) 9.99	107 (13.2) 99	16.54 (3.21) 12.6	180 (28.5) 138
PCB-156	2.52 (0.32) 2.13	30 (3.47) 24.62	1.48 (0.3) 1.18	17.4 (3.01) 13.16	0.56 (0.07) 0.5	5.64 (0.89) 4.91	1.22 (0.19) 1.01	13.4 (1.66) 11.1
PCB-180	23 (2.34) 19.5	277 (28) 240.2	20.1 (4.38) 13.0	235 (46.3) 161.6	7.88 (2.44) 5.47	74.2 (21.3) 52.4	11.3 (2.41) 9.53	122 (21.8) 105
PCB-170	10.7 (1.25) 8.9	128 (14.27) 102.5	8.91 (1.91) 6.0	105 (20.4) 69.4	3.00 (0.79) 2.27	28.31 (6.81) 23.1	4.15 (0.97) 3.34	44.81 (8.78) 36.8
PCB-194	3.78 (0.44) 3.03	45.5 (4.96) 37.24	3.66 (0.78) 2.64	43.4 (8.54) 30.38	1.24 (0.46) 0.68	11.96 (4.19) 7.24	1.96 (0.39) 1.95	21.24 (3.58) 21.4
PCB-209	0.15 (0.02) 0.13	1.76 (0.29) 1.76	0.31 (0.03) 0.30	3.84 (0.26) 3.84	0.15 (0.03) 0.11	1.52 (0.35) 0.99	0.12 (0.02) 0.12	1.29 (0.2) 1.33
Σ16PCB	186 (16.8) 164.2	2245 (195) 1970.4	132 (24.1) 105.5	1560 (245) 1207.3	56.5 (8.97) 47.9	548 (83.9) 478	81.0 (16.55) 64.4	889 (1467) 710
Σ7PCB	159 (14.2) 140	1921 (167) 1694	111 (20.4) 89	1317 (206) 1020	48.9 (7.42) 42.3	475 (70.16) 417	71.9 (14.73) 55.8	780 (131) 614

Table 2
Mean PBDE congener concentrations, standard error of the mean (SEM) and median for tern eggs from Rockabill Island (n = 20), Inishkea Islands (n = 15) and inner Galway Bay islands (n = 5).

	Rockabill Island (n = 20)		Rockabill Island (abandoned) (n = 10)		Inishkea Islands (n = 15)		Inner Galway Bay islands (n = 5)	
	Mean ww (SEM) Median	Mean lw (SEM) Median	Mean ww (SEM) Median	Mean lw (SEM) Median	Mean ww (SEM) Median	Mean lw (SEM) Median	Mean ww (SEM) Median	Mean lw (SEM) Median
BDE-28	0.3 (0.06) 0.17	3.31 (0.66) 2.35	0.31 (0.03) 0.31	4.11 (0.69) 3.54	0.11 (0.01) 0.13	1.17 (0.14) 1.48	0.07 (0.004) 0.07	0.73 (0.05) 0.73
BDE-47	4.95 (1) 3.5	58.1 (10.9) 38.6	5.83 (0.49) 5.6	70.8 (4.27) 73.3	2.02 (0.19) 1.83	20.1 (1.91) 19	1.99 (0.15) 1.88	22.29 (1.8) 22.8
BDE-100	2.02 (0.34) 1.3	24.2 (4.02) 16.9	2.27 (0.27) 2.3	28.5 (3.59) 26.1	0.42 (0.05) 0.40	3.99 (0.46) 3.92	0.54 (0.05) 0.51	5.97 (0.5) 5.59
BDE-99	1.6 (0.2) 1.51	19.7 (2.66) 17.1	1.5 (0.12) 1.47	18.7 (1.86) 16.83	0.32 (0.03) 0.26	3.10 (0.27) 2.90	0.56 (0.04) 0.57	6.27 (0.5) 6.32
BDE-154	1.21 (0.21) 0.95	14.2 (2.28) 10.73	1.04 (0.08) 1.11	13.5 (1.75) 12.56	0.12 (0.02) 0.1	1.21 (0.17) 1.11	0.49 (0.09) 0.37	5.41 (0.93) 4.3
BDE-153	0.45 (0.09) 0.31	5.29 (0.99) 3.31	0.49 (0.07) 0.50	6.19 (0.99) 5.95	0.37 (0.03) 0.36	3.57 (0.28) 3.42	0.23 (0.02) 0.22	2.62 (0.25) 2.62
Σ6PBDEs	10.5 (1.73) 7.9	125 (19.29) 88.8	11.5 (0.8) 11.1	142 (10.5) 142	3.37 (0.25) 3.32	33.1 (2.57) 36.8	3.88 (0.3) 3.44	43.4 (3.5) 41.2

Σ6PBDE concentration in Rockabill Island, Inishkea Islands and inner Galway Bay islands respectively. Significantly higher levels of Σ6PBDE were detected in Rockabill Island when compared to the Inishkea Islands and inner Galway Bay islands for lw concentrations ($P < 0.0001$, $P = 0.0201$) (Fig. 2). No differences were observed in PBDE concentrations between Inishkea Islands and inner Galway Bay islands.

All 17 OC compounds were detected in tern eggs (Table 3). Mean concentrations of HCBD in eggs from Rockabill Island was 0.29 ng/ww (range: 0.01–0.89) and 0.16 ng/ww (range: 0.07–0.3 ng/g ww) in eggs from Inishkea Islands and 0.15 ng/g ww in eggs from inner Galway Bay islands (range: 0.13–0.15 ng/g ww). There were no significant differences in HCBD (ww and lw) concentrations between the study sites. The mean concentrations of HCB in eggs from Rockabill Island was 11.4 ng/ww (range: 5.62–21.25), 14.1 ng/ww (range: 11.3–18.5) in Inishkea Islands and 7.12 ng/g ww in eggs from inner Galway Bay islands. There were no significant differences in HCB (ww and lw) concentrations

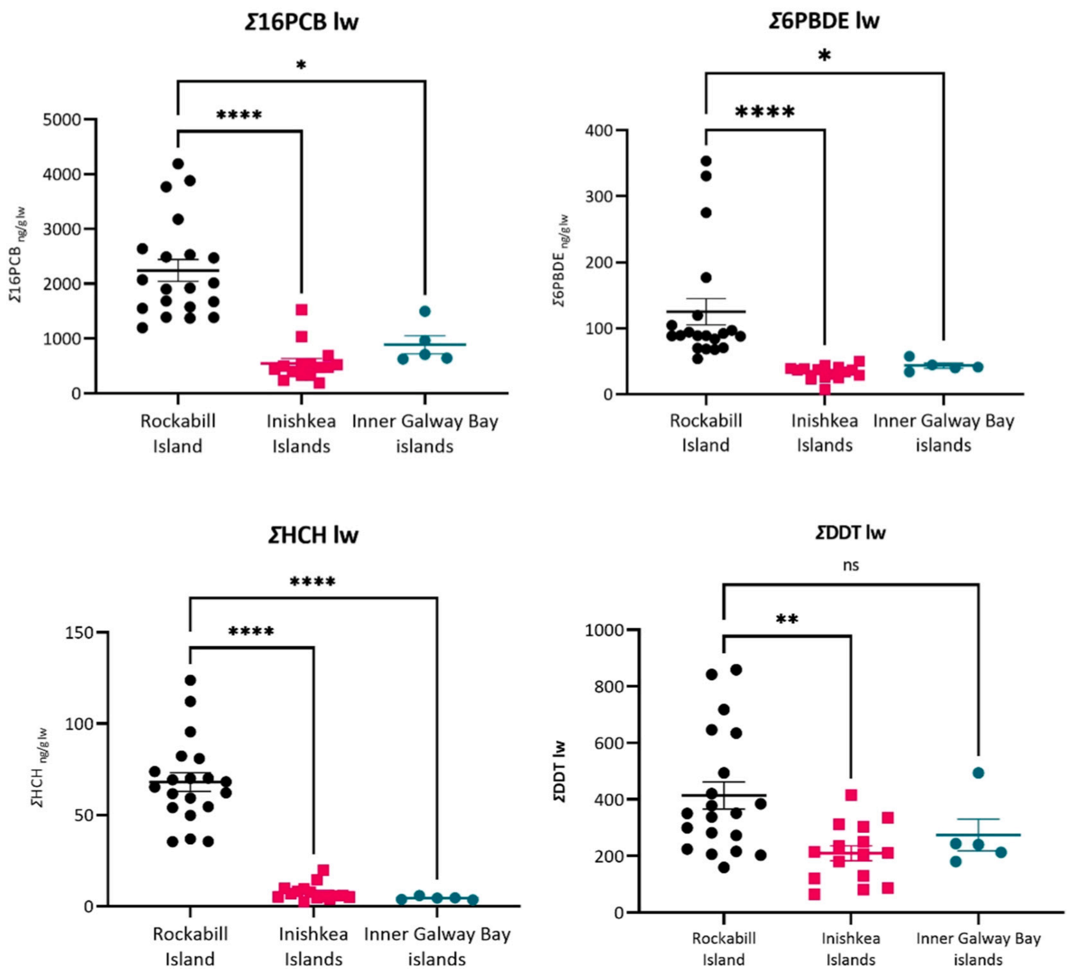

Fig. 2. Higher contaminant concentrations were observed in Common Tern eggs from Rockabill Island (n = 20) compared to Arctic Tern eggs from the Inishkea Islands (n = 15) and Common Tern eggs from inner Galway Bay islands (n = 5) for Σ16PCB, Σ6PBDE, ΣHCH, ΣDDT. Statistical significance was assessed using Kruskal Wallis test with Dunn's multiple comparison post-hoc test for nonparametric data. Error bars = SEM, * = P ≤ 0.05, ** = P ≤ 0.01, **** = P ≤ 0.0001, ns = not significant.

between the study sites. Mean concentration of ΣHCH ww in eggs from Rockabill Island was 5.69 ng/g ww (range: 2.73–9.73 ng/g ww), 0.78 ng/g ww for Inishkea Islands (range: 0.32–1.75) and 0.4 n/g ww in eggs from inner Galway Bay islands (range: 0.32–0.48 ng/g ww). Significantly higher levels of ΣHCH were detected in Rockabill Island when compared to the Inishkea Islands and inner Galway Bay islands for both ww and lw concentrations (P < 0.0001 for both sites). No differences were observed in ΣHCH concentrations between Inishkea Islands and inner Galway Bay islands.

Mean concentration of ΣCHL ww in eggs from Rockabill Island was 9.11 ng/g ww (range: 0.65–52.2 ng/g ww) and 3.71 ng/g ww in eggs from Inishkea Islands (range 1.37–26.2 ng/g ww) and 1.8 ng/g ww in eggs from inner Galway Bay islands (range: 1.19–2.39). There were no significant differences in ΣCHL concentrations on Rockabill Island when compared to the Inishkea Islands and inner Galway Bay islands between the study sites for ww and lw concentrations.

Mean concentration of ΣDDT (plus metabolites) ww in eggs from Rockabill Island was 34.6 ng/g ww (range: 15.2–77.1 ng/g ww), 21.4 ng/g ww in eggs from Inishkea Islands (range: 6.6–47.5) and 24.3 ng/g ww in eggs from inner Galway Bay islands (range: 15.4–40.6). Significantly higher levels of ΣDDT (plus metabolites) were detected in Rockabill Island when compared to the Inishkea Islands but not inner Galway Bay islands for lw concentrations (P = 0.0032, P = 0.3641, respectively) (Fig. 2). No significant difference in ΣDDT concentrations were detected between the Inishkea Islands and inner Galway Bay islands.

The major metabolite of DDT, *p,p*' –DDE, accounted for 94, 98.6 and 98.4% of ΣDDT in Rockabill Island, Inishkea Islands and inner Galway Bay islands respectively. All fifteen metals analysed for were detected in tern eggs (Table 4). Individual metal levels (Al, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Ag, Cd, Hg) in pooled samples between all sited were broadly similar with concentrations from Rockabill generally a little higher than in west coast colonies. A greater difference was observed in lead levels in pooled tern eggs from Rockabill (0.13 mg/kg ww) compared to Inishkea Islands (0.001 mg/kg ww) and inner Galway Bay islands (0.002 mg/kg ww).

3.2. Intra-clutch variation

Reliability values range between 0 and 1, with values closer to 1 representing stronger reliability (i.e. intra-clutch variation is very low compared to interclutch variation). Fifteen of the 34 compounds (ww and lw) had intraclass correlation coefficients (ICC) above 0.9 which is an indication of excellent reliability. Eight values had intraclass correlation coefficients between 0.75 and 0.9 which is an indication of good reliability. Eleven values had intraclass correlation coefficients between 0.5 and 0.75 which is an indication of moderate reliability.

Table 3
Mean OC concentrations, standard error of the mean (SEM) and median (ng/g ww and lw) for tern eggs from Rockabill Island (n = 20), Inishkea Islands (n = 15) and inner Galway Bay islands (n = 5). ΣHCH = α-HCH, γ –HCH and β-HCH. ΣCHL = heptachlor, heptachlor epoxide, oxychlordane, trans-chlordane, cis-chlordane and transnonachlor. ΣDDT = *o,p'* –DDE, *p,p'* –DDE, *o,p'* –DDT, *o'p'* –DDD, *p,p'* –DDD, *p,p'* –DDT.

	Rockabill Island (*n* = 20)		Rockabill (abandoned) (n = 10)		Inishkea Islands (n = 15)		Inner Galway Bay islands (n = 5)	
	Mean ww (SEM) Median	Mean lw (SEM) Median	Mean ww (SEM) Median	Mean lw (SEM) Median	Mean ww (SEM) Median	Mean lw (SEM) Median	Mean ww (SEM) Median	Mean lw (SEM) Median
HCBD	0.29 (0.06) 0.24	3.41 (0.71) 2.86	0.69 (0.17) 0.53	8.68 (2.10) 6.53	0.16 (0.02) 0.14	1.6 (0.23) 1.24	0.15 (0.01) 0.14	1.64 (0.11) 1.5
HCB	11.4 (0.87) 10.5	139 (12.2) 130	7.54 (0.98) 7	93.2 (11.7) 80	14.1 (0.52) 13.2	137 (7.86) 146	7.12 (1.02) 5.6	80.2 (13.21) 63.2
α-HCH	0.38 (0.09) 0.24	4.23 (1.06) 2.81	1 (0.16) 0.86	12.4 (1.89) 11.56	0.08 (0.02) 0.06	0.81 (0.23) 0.57	0.02 (0.004) 0.01	0.18 (0.05) 0.14
β-HCH	4.85 (0.41) 4.55	58 (4.8) 56.3	3.42 (0.5) 3.4	46 (9.33) 39.1	0.62 (0.07) 0.58	6.14 (0.77) 5.28	0.38 (0.03) 0.41	4.27 (0.35) 4.44
γ-HCH	0.46 (0.08) 0.38	5.81 (1.11) 4.54	1.42 (0.18) 1.39	19 (3.79) 16.98	0.07 (0.02) 0.04	0.72 (0.24) 0.39	0.01 (0.002) 0.01	0.05 (0.02) 0.06
Heptachlor	0.49 (0.09) 0.34	5.94 (1.09) 4.99	0.21 (0.05) 0.17	3.01 (0.93) 2.34	0.08 (0.01) 0.08	0.84 (0.15) 0.71	0.00 (0.00) 0	0.00 (0.00) 0
Heptachlor Epoxide	0.75 (0.17) 0.48	9.28 (2.1) 5.17	1.47 (0.52) 0.67	21 (8.95) 9.07	2.45 (1.53) 0.87	26 (17.17) 9.55	0.93 (0.06) 0.94	10.39 (0.72) 10.5
Oxychlordane	7.07 (3.05) 1	84.6 (36.9) 12.85	4.27 (0.55) 4.04	56.6 (10.2) 49.1	1.04 (0.06) 1.04	10.4 (0.93) 10.2	0.77 (0.19) 0.47	8.52 (2.15) 5.2
trans-chlordane	0.05 (0.02) 0	0.51 (0.24) 0	0.06 (0.01) 0.05	0.92 (0.27) 0.48	0.04 (0.01) 0.03	0.38 (0.08) 0.29	0.08 (0.02) 0.08	0.9 (0.14) 0.92
cis-chlordane	0.12 (0.06) 0	1.32 (0.67) 0	0.13 (0.04) 0.1	1.62 (0.42) 1.18	0.05 (0.01) 0.04	0.52 (0.12) 0.45	0.00 (0.00) 0	0.05 (0.03) 0
Transnonachlor	0.63 (0.20) 0.25	7.27 (2.17) 3.15	0.1 (0.01) 0.09	1.4 (0.28) 1.17	0.05 (0.01) 0.04	0.52 (0.11) 0.41	0.02 (0.01) 0	0.18 (0.12) 0
o,p' -DDE	0.01 (0.01) 0	0.19 (0.18) 0	0.11 (0.04) 0.08	1.62 (0.68) 0.95	0.08 (0.02) 0.06	0.80 (0.16) 0.51	0.00 (0.00) 0	0.01 (0.01) 0
p,p' -DDE	32.4 (3.66) 29.9	388 (43.8) 330	16.8 (2.81) 12	200 (28) 174	21.1 (2.5) 20.7	2067 (25.6) 210	23.9 (3.94) 21.4	270 (50.7) 235
o,p' -DDT	0.53 (0.44) 0.02	5.92 (4.88) 0.25	0.61 (0.1) 0.61	8.09 (1.94) 6.96	0.01 (0.002) 0.004	0.07 (0.02) 0.03	0.005 (0.001) 0.004	0.05 (0.02) 0.04
o'p' -DDD	0.1 (0.03) 0.04	1.10 (0.35) 0.55	0.2 (0.05) 0.17	2.89 (1.13) 2.08	0.07 (0.01) 0.05	0.65 (0.12) 0.55	0.002 (0.002) 0.002	0.03 (0.01) 0.03
p,p' -DDD	0.85 (0.17) 0.61	9.78 (1.71) 7.50	0.59 (0.05) 0.6	7.84 (1.18) 6.96	0.01 (0.001) 0.01	0.12 (0.04) 0.07	0.28 (0.07) 0.28	3.11 (0.77) 2.94
p,p' -DDT	0.76 (0.19) 0.74	9.24 (2.24) 8.46	1.24 (0.15) 1.15	16.2 (2.82) 12.43	0.13 (0.02) 0.08	1.25 (0.24) 0.98	0.09 (0.01) 0.08	1.02 (0.16) 0.98
ΣHCH	5.69 (0.42) 5.57	68.1 (5.05) 66.7	5.85 (0.69) 5.7	77.4 (13.6) 67.9	0.78 (0.09) 0.66	7.68 (1.12) 5.93	0.4 (0.03) 0.42	4.5 (0.37) 4.57
ΣCHL	9.11 (3.59) 2.07	109 (43.2) 26.2	6.24 (1.18) 5.1	84.6 (21) 63.3	3.71 (1.63) 2.1	38.7 (18.6) 21.6	1.8 (0.28) 1.49	20 (3.16) 16.6
ΣDDT	34.6 (3.97) 32.4	414 (46.8) 351	19.6 (2.82) 15	236 (29) 223	21.4 (2.51) 21	210 (25.6) 211	24.3 (3.89) 21.8	274 (50.2) 240

Table 4
Metal concentrations (mg/kg ww and dw) for pooled samples from Rockabill Island, Inishkea Islands and inner Galway Bay islands.

	Rockabill Island (n = 20)		Rockabill (abandoned) (n = 10)		Inishkea Islands (n = 15)		Inner Galway Bay islands (n = 5)	
	mg/kg ww	mg/kg dw	mg/kg ww	mg/kg dw	mg/kg ww	mg/kg dw	mg/kg ww	mg/kg dw
Al	0.21	0.86	0.20	0.91	0.15	0.77	0.21	0.96
V	0.001	0.01	0.001	0.01	0.001	0.003	0.001	0.003
Cr	0.01	0.04	0.01	0.04	0.02	0.11	0.04	0.20
Mn	0.48	1.97	0.38	1.71	0.29	1.48	0.33	1.5
Fe	15.5	64.1	12.1	55.2	12.1	61.2	10.1	45.8
Co	0.01	0.02	0.01	0.03	0.01	0.03	0.01	0.03
Ni	0.01	0.02	0.002	0.01	0.02	0.09	0.04	0.19
Cu	0.78	3.23	0.60	2.74	0.58	2.93	0.52	2.36
Zn	15.5	64.2	12.1	55.2	12.1	61.2	10.1	45.8
As	0.12	0.52	0.13	0.57	0.06	0.33	0.1	0.46
Se	0.78	3.22	0.72	3.31	0.72	3.65	0.59	2.67
Ag	0.005	0.02	0.005	0.02	0.002	0.01	0.003	0.01
Cd	0.0005	0.002	0.001	0.003	0.001	0.003	0.0003	0.001
Pb	0.13	0.53	0.01	0.06	0.001	0.01	0.002	0.01
Hg	0.15	0.62	0.18	0.84	0.13	0.68	0.16	0.74

3.3. Abandoned eggs

Mercury concentrations in the pooled sample of fresh eggs were similar to that of pooled sample of abandoned eggs (0.15 and 0.18 mg/kg ww respectively), additionally levels of metals (SI) within the same colony were similar. There was no difference was observed in concentrations of ΣHCH and ΣCHL between fresh and abandoned eggs. However, significantly higher levels of Σ16PCBs, Σ6PBDE and ΣDDT were detected in fresh eggs compared to abandoned eggs. Σ16PCB, Σ7PCB, HCB, ΣDDT and ΣCHL were approximately 20–40% lower in abandoned eggs than in fresh eggs from Rockabill Island and $<$ 10% difference for Σ6PBDE. Contaminant profiles are similar between fresh and abandoned eggs (Fig. S1, S2 and S3). PCB-153 was the most dominant congener and accounted for 34.4% of total Σ16PCB ww in fresh eggs and 34% in abandoned eggs (Fig. S1). Substituting results from abandoned eggs for fresh eggs in this study for comparison with EcoQO and ecotoxicological thresholds (Table 5) gives the same outcome. Comparing abandoned and fresh eggs from Rockabill Island with Arctic Tern eggs from the Inishkea Islands also yields similar results. Significant differences between fresh and abandoned compared to eggs from Insihkea Islands can be detected for Σ16PCB, Σ6PBDE, ΣHCH and ΣCHL. Similarly, both fresh and abandoned eggs showed no difference when HCB concentrations compared. ΣDDT was the only exception to this, no differences were observed in ΣDDTs between abandoned eggs on Rockabill Island and west coast colonies. ΣDDT concentrations in fresh eggs from Rockabill Island were significantly higher than Inishkea Islands.

3.4. OSPAR EcoQO

OSPAR EcoQO recommend that ΣPCBs should not exceed 20 ng/g ww in Common Tern eggs (OSPAR, 2010). Even allowing for the fact that the EcoQO is based on the sum of 62 PCBs, the results from this study (Σ16PCB mean concentration of 186 and 62.9 ng/g ww in tern eggs from Rockabill Island and the west coast respectively) are higher than this threshold. The OSPAR commission state that concentrations should not exceed 10 ng/g ww for DDT and metabolites (exceeded in both areas, 34.6 ng/g ww and 22.2 ng/g ww in eggs from Rockabill Island and the west coast respectively), and 2 ng/g for HCB (exceeded in both areas, 11.4 and 12.4). The EcoQO for HCH (2 ng/g ww) was met in eggs the west coast (0.68) but not in Rockabill Island (5.69). OSPAR guidelines state that the average concentrations of mercury in the fresh mass of ten eggs from separate clutches of the Common Tern breeding adjacent to industrialised areas (in this study represented by Rockbill Island) should not significantly exceed concentrations in the fresh mass of ten eggs from separate clutches of the same species breeding in similar, but not industrial, habitats (i.e. west coast colonies). The EcoQO is this study has been met as the levels of mercury in both areas were very similar (0.15 mg/kg ww) (Table 4).

4. Discussion

4.1. Site differences and stable isotope ratio analysis

Care must be taken when interpreting differences in contaminant burdens in higher trophic level predators between multiple sites, especially without interannual data. Differing feeding strategies, diet and in the case of this study, different (but closely related) species, are important considerations when interpreting concentrations of pollutants found in tern eggs. Significantly higher concentrations of pollutants (Σ16PCB, Σ6PBDE, ΣHCH, ΣDDT, ΣCHL) were found in Common Tern eggs from Rockabill compared to the colonies on the west coast of Ireland, Arctic Tern eggs from Inishkea Islands and Common Tern eggs from inner Galway Bay islands (Fig. 2). Higher concentrations of POPs in tern eggs from Rockabill Island compared to the west coast are likely a result of the former's proximity to Ireland's largest city, Dublin. Rockabill Island is located in the Irish Sea, a semi-enclosed sea located between the east coast of Ireland and the west coast of Britain. Fluvial transport of contaminants into the Irish Sea and the presence of two large cities with some industrial history, Dublin and Liverpool, are likely to be significant contributors to contamination in the region. Elevated levels of persistent pollutants (OCs, PCBs, metals) have also been found in mussels *Mytilus edulis* and shellfish in Dublin Bay compared to other sites along the Irish coast (Glynn et al., 2004; Widdows et al., 2002). Pollutant levels in the eggs of Cormorant *Phalacrocorax carbo* and Shag *P. aristotelis* eggs from Dublin Bay were higher than other parts of Ireland (Wilson and Earley, 1986).

Stable isotope ratio analysis showed significant differences between Rockabill Island and west coast colonies. Arctic Terns from the Inishkea islands in this study fed at a lower trophic level ($\delta^{15}N$) than Common Terns from Rockabill Island and inner Galway Bay islands (Fig. 3). $\delta^{13}C$ values in this study indicate that birds from Inishkea Islands are feeding on less pelagic prey items than Common Terns, inshore organisms show higher $\delta^{13}C$ (less negative than) values than pelagic organisms (Fig. 3)

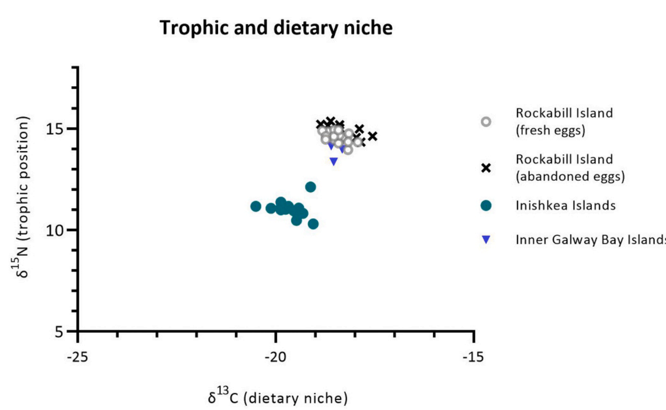

Fig. 3. Stable isotopes ratios of carbon and nitrogen (‰) in fresh and abandoned tern eggs from Rockabill Island ($n = 20$), Insihkea Islands ($n = 15$) and inner Galway Bay islands ($n = 5$) showing the dietary niche of adult birds. $\delta^{13}C$ corrected for lipid using method from (Elliott and Elliott, 2016).

Table 5
Summary statistics comparing abandoned eggs (n = 10) to fresh eggs from Rockabill Island (n = 20), west coast colonies (n = 20) and EcoQO threshold where applicable (ng/g ww). Statistical significance was tested by Mann Whitney U test (non-parametric data). * = $P \leq 0.05$, ** ≤ 0.01, **** = $P \leq 0.0001$, ns = not significant, na = not applicable.

	Rockabill eggs	Mean (SEM)	Median	Fresh v abandoned	EcoQO	v Inishkea Islands
Σ16PCB	Fresh	2245 (195)	1970	*	Surpassed	****
	Abandoned	1560 (245)	1207		Surpassed	****
Σ6PBDE	Fresh	125 (19.3)	88.8	*	na	****
	Abandoned	142 (10.5)	142		na	****
HCB	Fresh	139 (12.2)	130	**	Surpassed	*
	Abandoned	93.2 (11.7)	80		Surpassed	**
ΣHCH	Fresh	68.1 (5.05)	66.7	ns	Surpassed	****
	Abandoned	77.4 (13.6)	67.9		Surpassed	****
ΣCHL	Fresh	109 (43.2)	39.4	ns	na	*
	Abandoned	84.6 (21)	69.7		na	***
ΣDDT	Fresh	414 (46.8)	351	*	Surpassed	**
	Abandoned	236 (29)	223		Surpassed	ns

(France 1995). Clupeids (mostly sprat *Sprattus sprattus*) and sandeels (*Ammodytes* sp.) were the main prey items provisioned to Common Tern chicks on Rockabill Island in 2017 (53% and 42% respectively) (McKeon et al., 2017) which is likely to be similar to the diet of adult females during the egg formation period. As contaminants can biomagnify as trophic level increases this could exacerbate differences between Rockabill Island, where contaminants are expected to be higher, and the Inishkea Islands. $\delta^{15}N$ values were more similar between Common Tern eggs from Rockabill Island and inner Galway Bay islands and significant differences in pollutant levels were detected regardless, although the sample size from inner Galway Bay islands is small ($n = 5$).

Higher Σ16PCB concentrations of pollutants in eggs from inner Galway Bay islands compared to Inishkea Islands is likely a result of their respective locations. Inner Galway Bay islands are located within a semi-enclosed bay that is partially shielded from the Atlantic Ocean by the Aran Islands, Mutton Island itself is connected to Galway city via a causeway. In contrast, the Inishkeas are offshore islands in a relatively pristine location in the Atlantic Ocean. However, the sample size from the inner Galway Bay islands is very small (n = 5) and the stable isotope signatures are markedly different between two sites. It is likely that with a larger sample size more differences in contaminant burden would become apparent.

Dittmann et al. (2011) showed that Arctic Tern eggs had similar contaminant levels to Common Tern eggs but more research in needed to determine the suitability of using Arctic Tern eggs in place of Common Tern eggs where the latter is scarce. Stable isotope ratio analysis is an important tool when interpreting contaminant concentration in higher trophic level organisms, combined with long-term data from the same colony and species it can provide a clearer indication of differences in contaminants between colonies and species.

4.2. Intra-clutch variation

OSPARs JAMP guidelines recommend taking one egg randomly from each clutch (OSPAR, 2014), this sampling strategy has been adopted as intra-clutch variation is low compared to interclutch variation (Becker et al., 1991). An earlier study of intra-clutch variation in Common Terns showed that later laid eggs generally have higher organochlorine concentrations (approximately 20%) than the first-laid egg (Nisbet, 1982). The ICC value for most compounds in this study was high (ww and lw) indicating that in general, intra-clutch variation in POPs is low compared to interclutch variability in accordance with Becker et al. (1991) and taking one egg randomly from a full clutch is an appropriate sampling strategy. Early eggs of Common and Arctic Terns have been shown to have significantly higher levels of mercury than late eggs (Akearok et al., 2010; Becker, 1992) which may need to be investigated further in relation to monitoring.

4.3. Utility of abandoned eggs

Stable isotope ratio analysis showed no significant difference between fresh and abandoned eggs on Rockabill Island indicating that adult female birds between the two groups had a similar diet (Fig. 3). Metals results between fresh and abandoned eggs were similar. Contrasting results were determined for POPs with some pollutants also showing similarities (ΣHCH and ΣCHL) while other pollutants were significantly higher in fresh eggs (Σ16PCBs, Σ6PBDE and ΣDDT). Metals and lipophilic organic pollutants are transferred into the egg in different ways which may explain contrasting patterns, mercury binds with proteins primarily found in albumen while lipophilic POPs are found primarily in the yolk (Blundell and Jenkins, 1977; Drouillard et al., 2003). A similar study comparing fresh and failed eggs in Common Terns from the same colony in Germany found differences in PCBs and ΣDDT between fresh and failed eggs but no difference is mercury between the two groups (Becker et al., 1993). It is possible the albumen is less impacted by the impacts of abandonment and that intact eggs may provide reliable measurements of metals. However, this study used pooled samples for mercury and metals analysis and making direct comparisons is somewhat limited.

Despite having significantly lower concentrations of Σ16PCBs, Σ6PBDE and ΣDDT, abandoned egg samples were still within the lower end of the range of pollutant concentrations found in fresh eggs. Substituting results from abandoned eggs for fresh eggs in this study for EcoQO and ecotoxicological thresholds (Table 5) gives the same outcome, despite differences detected in pollutant concentration between fresh and abandoned eggs. Similarly, comparing abandoned eggs from Rockabill Island with Arctic Tern eggs from the Inishkea Islands also yields similar results, suggesting that spatial differences in pollutant concentrations can be determined in abandoned eggs. In Becker et al. (1993) a comparison of fresh and failed eggs in Common Terns from the same colony in Germany found significantly higher levels of PCBs and ΣDDT in failed eggs, in contrast to this study. Contamination levels in eggs can change throughout the breeding season, a variable that was accounted for in Becker et al. (1993) but not in this study which may explain contrasting results. This study shows that there are differences in contaminant concentrations in organic pollutants between fresh and abandoned eggs but similar levels of metals. This supports previous studies that recommend fresh eggs for the monitoring of contaminants (Klein et al., 2012). However, abandoned eggs may provide a useful approximation of contaminants in Common Tern eggs if non-destructive sampling methods are preferred, perhaps in areas where the conservation status of Common and Artic Terns is a concern. The use of abandoned eggs may be the only possible means to analyse eggs of vulnerable species for pollutants (Eriksson et al., 2016). Caution is advised when interpreting results from abandoned eggs where the recorded levels are close to a proposed threshold. In these instances, the use of fresh eggs is more advisable. This approach may not be possible for many colonies as only eggs in good physical condition were retrieved for this study, made possible by the large Common Tern population size and presence of full-time wardens on Rockabill Island.

4.4. Comparisons with other studies and worldwide trends

This is the first published study of pollutants in Common or Arctic Terns eggs in Ireland. However, the authors have obtained historical, unpublished data of pollutants (*p,p*' –DDE and PCBs) in Common Tern eggs from Ireland. Historical data were obtained from Dr. David Cabot who carried out studies on organochlorine contamination of Irish seabird eggs during the 1960s and 1970s. Levels of *p,p*' -DDE from Common Tern eggs on Rockabill Island from 1965 and 1966 had an average concentration of 218 ng/g ww ($n = 15$, range: 100 to 300 ng/g ww) which is higher than levels detected in this study (32.4 ng/g ww, range: 15.2–77.1 ng/g ww). This analysis was carried out by the Laboratory of the Government Chemist (now LGC Group), London. Caution is advised when interpreting historical measurements of DDT as PCBs could be confused for DDT-type compounds. However, analysis took place in 1967 when analytical methods had improved sufficiently to isolate DDT from PCB compounds (Shea, 1973). Historical pollutant data of a single Common Tern egg collected from Rockabill Island in 1978 were also obtained, analysed by the University of Reading. *p,p*' –DDE levels were reported as <50 ng/g ww in this egg (mean of 32.4 reported in this study). PCB levels (without individual congener information) were reported as <5000 ng/g ww, considerably higher than Σ16PCB levels reported from Rockabill in this study (mean = 186 ng/g ww). Peak DDT usage occurred during the 1950s (Rapaport et al., 1985), while peak PCB inputs occurred in the 1960/70s (Alcock et al., 2000). Lower present-day levels of the pollutants reflect bans and restrictions placed on certain contaminants in the intervening years. Due to their high toxicity and their persistent and bioaccumulative (PBT) properties, the production and use of many POPs have been phased out through various worldwide legislative instruments such as the Stockholm Convention on POPs, a legally binding international environmental treaty, that aims to ban, eliminate or restrict the production and use of POPs. These instruments have been successful, leading to broadly downward trends of pollutants in birds in marine environments (Braune et al., 2005; Helgason et al., 2008; Nyberg et al., 2015; Rigét et al., 2019). Similar declines have also been recorded in Common Terns for POPs (Becker et al., 2001) and metals (Burger and Gochfeld, 2004). While the use of PCBs and OCs has been either banned or restricted in Europe, including Ireland, since the 1970s and 80s and PBDEs, more recently, since the 2000s, several POPs are still in use outside of Europe (Gioia et al., 2011; WHO, 2011).

Pollutant concentrations in this study are generally lower or within the lower end of the range of contaminant concentrations reported in terns eggs from the North Sea, spanning seven countries (Dittmann et al., 2012; Soerensen and Faxneld, 2020). Using PCB-153 as a marker for ΣPCB, mean levels in this study from Rockabill Island (64.3 ng/g) were much lower than concentrations determined from eight colonies in the Wadden Sea in 2010 (Netherlands, Germany and Denmark) where mean concentrations ranged from 164 to 683 ng/g ww (Becker et al., 2001). Similarly, mean *p,p'* –DDE concentrations in eggs from Rockabill Island (32.4 ng/g ww) were lower than all colonies in the Wadden Sea (mean concentrations ranging from 46 ng/g to 345 ng/g ww). Total mercury concentrations in Rockabill Island were also lower than concentrations detected in the Wadden Sea. This gives an indication of the relatively low levels of pollutants in Ireland's marine waters compared to the North Sea, especially given that Rockabill Island had significantly higher pollutant concentrations than eggs from the Inishkeas and inner Galway Bay islands. Other OC compounds (HCB, HCH, chlordanes) were within the lower end of the range found in Wadden Sea colonies. Few data on PBDEs in tern eggs are available, mean levels of Σ6PBDE in tern eggs from Rockabill (10.52 ng/g ww) were lower than levels recorded in Common Tern eggs from Massachusetts, USA (Σ9PBDE, congeners unknown, 30–180 ng/g in 1994–2005) (reported in Arnold et al., 2020). Mean BDE-47 concentrations of 10.6 (range: 5.57–20.3) in Common Tern eggs from Chesapeake Bay, Maryland, USA (Rattner et al., 2011) were higher than concentrations determined in eggs from Rockabill Island (mean: 4.9 ng/g), the Inishkea Islands (3.02 ng/g ww) and inner Galway Bay islands (1.99 ng/g ww). Levels of metals (As, Cr, Mn, Se) were similar to levels reported in Common Tern eggs in Barnegat Bay, USA from 2014 and broadly similar to internationally reported levels for terns eggs (As, Cr, Cu, Se, Zn) (summarised in U.S. Fish and Wildlife Service, 2001). Pollutant concentrations (ΣDDT, ΣCHL, ΣHCH) in Arctic Tern eggs from high Arctic Canada (1998–2005) were generally lower than concentrations reported in this study (Mallory and Braune, 2012). Median ΣPCB concentrations (63.4 ng ww) reported in Mallory and Braune (2012) are similar to concentrations recorded in Arctic Tern eggs from the Inishkea islands (47.9 ng/ww) and Common Tern eggs from inner Galway Bay islands (64.4 ng/ww).

Pollutant levels in Northern Gannet eggs from Lambay Island, approximately 12 km from Rockabill Island, from the same year were generally higher (Σ14PCB, ΣCHL, ΣDDT, mercury) than tern eggs from Rockabill (Power et al., 2021). Biomagnification of contaminants may explain differences in pollutant levels between species as Gannets are the largest breeding seabird in the region (Hamer et al., 2001) and feed on larger prey items than Common Terns. Mean HCB levels (ng/g ww) between Gannet eggs on Lambay Island compared with tern eggs from Rockabill Island and west coast colonies were very similar (12.6, 11.4, 12.4 ng/g ww). The biomagnification process of HCB in biota is highly variable and less predictable that other pollutants (Moermond and Verbruggen, 2013).

4.5. OSPAR EcoQO and thresholds

In summary, the EcoQO was not met for PCBs, DDT and metabolites, HCB in all sites and HCH in Rockabill Island. The EcoQO was met for HCH in the west coast and for mercury in both sites. The EcoQO for ΣPCB, DDT and metabolites and HCB was also exceeded in all sites and countries in a large scale study of tern eggs within the EcoQO programme, encompassing seven countries in the North Sea between 2008 and 2010, as well as a separate study applying the tern egg EcoQO to Sweden (Dittmann et al., 2012; Soerensen and Faxneld, 2020). The EcoQO for HCH, which was not met on Rockabill Island, was fulfilled in all sites in Sweden (Soerensen and Faxneld, 2020) and for most sites in the aforementioned study by Dittmann et al. (2012). HCH levels in tern eggs from Middlesbrough, United Kingdom exceeded the threshold by a similar margin to tern eggs from Rockabill (Dittmann et al., 2012). In contrast to this study, the EcoQO for mercury was exceeded in all sites and countries in both Soerensen and Faxneld (2020) and Dittmann et al. (2012).

EcoQOs function both as indicators and objectives but are not thresholds for adverse physiological effects. Ecotoxicological thresholds specific to Common Terns have been established for mercury (Shore et al., 2011), and concentrations reported in this study (0.15 mg/kg) are associated with no adverse effects (levels of 1 mg/kg are associated with no adverse effects). Similarly, Braune et al. (2012) suggest concentrations of 1.1 mg/kg in the eggs of Arctic Tern can have harmful effects based on dosing eggs with graded concentrations of mercury. Common Terns in Lake Michigan, USA with mean PCB concentration of approximately 12,000 ng/g ww in their eggs were associated with low reproductive success, and chick malformations (Ward et al., 2010). Similarly, ΣPCB concentrations of 5000 to 9000 ng/g ww in Common Tern eggs from the Elbe Estuary, Germany were associated with low hatching success (Becker et al., 1993). Mean PCB concentration in tern eggs from Rockabill Island, the most contaminated site in this study, was much lower than both these studies (186 ng/g ww). The Common Tern is one of the most sensitive bird species to the negative effects of DDE (Arnold et al., 2020). Mean DDE levels in Common Tern egg of approximately 4000 ng/g ww have been associated with severe impacts on breeding success (Fox, 1976; Nisbet and Fox, 2009) and concentrations of 1000 ng/g ww have been associated with hatching failures (Nisbet and Reynolds, 1984), mean ΣDDT concentration in tern eggs from Rockabill Island and west coast colonies was relatively low in comparison (34.6 and 22.2 ng/g ww respectively). Dosing Common Tern eggs with DE-71 (PBDE mixture consisting primarily of the congeners BDE-47, 99, 100, 153, 154) had no impact on chick survival or hatching success (Rattner et al., 2013). The highest dosage administered to a Common Tern egg in Rattner et al. (2013) was estimated to be approximately 4800–7600 ng/g ww (actual uptake into egg) which is 3 to 4 orders of magnitude higher than concentrations detected in Common Tern eggs in this study. Rattner et al. (2013) Suggest that Common Terns may be less sensitive to DE-71 than some other bird species such as American Kestrels *Falco sparverius*.

5. Conclusion

The North Sea EcoQO system for coastal bird eggs was successfully applied to Ireland in this study. Elevated levels of POPs were detected in tern eggs from Rockabill Island, illustrating the usefulness of having relatively pristine areas as well as potentially more impacted sites in the EcoQO system. This study has also provided further, more recent, supporting evidence that random sampling of one egg from a full clutch is an appropriate sampling strategy. This study supports previous research indicating fresh eggs are preferable when monitoring contaminants. The EcoQO in this study was only met for mercury (in both areas) and HCH in the west coast. The EcoQO was exceeded for PCBs, DDT and metabolites, HCB in all sites and HCH in Rockabill Island. However, the levels of legacy pollutants in this study did not surpass any published toxic thresholds established for birds.

CRediT authorship contribution statement

Philip White: conceptualization, methodology, review & editing, supervision **Brendan McHugh:** conceptualization, methodology, review & editing, supervision **Simon Berrow:** conceptualization, review & editing, supervision **Moira Schlingermann:** investigation **Aaron McKeown:** investigation **David Cabot:** investigation **Marissa Tannian:** investigation **Stephen Newton:** conceptualization, review & editing, supervision **Evin McGovern:** conceptualization, review & editing, supervision **Sinéad Murphy:** conceptualization, review & editing, supervision **Denis Crowley:** validation, investigation **Linda O'Hea:** investigation **Brian Boyle:** investigation **Ian O'Connor:** conceptualization, review & editing, supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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