APPENDIX A27

Environmental Pollution 268 (2021) 115723

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Persistent pollutants in Northern Gannet *Morus bassanus* eggs in Ireland: Levels and colony differences☆

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ARTICLE INFO

Article history:
Received 28 May 2020
Received in revised form
22 September 2020
Accepted 23 September 2020
Available online 5 October 2020

Keywords:
POPs
Metals
Stable isotope ratio analysis
Gannet
Seabird eggs

ABSTRACT

Seabird eggs are considered a favourable matrix for monitoring marine pollutants and are widely used as higher trophic level indicators. Persistent organic pollutants such as polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and other organochlorine compounds (OCs) as well as metals have been shown to have deleterious impacts on seabirds. The Northern Gannet *Morus bassanus* is an avian sentinel; the largest breeding seabird in Ireland and an obligate piscivore. Gannet eggs were collected from two island colonies off the east coast of Ireland in locations with divergent history of industrialisation. Contaminant levels were measured and differences in concentrations between colonies compared. Stable isotope ratios of carbon ($\delta_{13}C$) and nitrogen ($\delta_{15}N$) were measured in each egg to understand the influence of diet and trophic position on contaminant levels detected. Significantly higher levels of Σ14PCBs, Σ7PBDEs and total mercury were detected in Gannet eggs from Lambay Island near Dublin (Ireland's industrialised capital city) compared to Great Saltee Island. No differences were observed in levels of other OCs (HCB, ΣHCH, ΣCHL, ΣDDT) between the two colonies. Though Gannets travel significant distances when foraging for food, tracking studies have demonstrated that birds from proximal breeding colonies maintain exclusive feeding areas. Stable isotope ratio analysis in this study demonstrated that Gannets at both locations occupy similar dietary niches, indicating that dietary differences may not be the driver of differing contaminant levels between colonies. Levels of persistent pollutants in the Gannet eggs fall below most existing thresholds for adverse effects and are within internationally reported values. Recent population growth and range expansion of Gannets in Ireland suggest that persistent pollutants are not having an immediate impact on the Gannet population. This study will inform potential monitoring programmes that can help Ireland achieve good environmental status under the European Union's Marine Strategy Framework Directive.

1. Introduction

The majority of persistent pollutants such as polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and other organochlorine compounds (OCs) are resistant to natural degradation and are commonly found in both terrestrial and aquatic systems globally (Walker et al., 2012). Naturally occurring metals can occur in elevated levels as a result of anthropogenic processes and inputs (Dias and Edwards, 2003). A large number of organic compounds have been shown to exhibit a wide range of toxic properties in biota causing endocrine dysfunction, mutagenesis, or reproductive and behavioural disturbances (AMAP, 2003; Gore et al., 2015; Scheuhammer, 1987; Walker et al., 2012; WHO, 2012). Similarly, elevated levels of metals can cause both chronic and acute disorders in biota (Walsh, 1990).

Many persistent organic pollutants (POPs) and metals are lipophilic allowing them to bioaccumulate within the lipid of an

☆ This paper has been recommended for acceptance by Professor Christian Sonne.

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https://doi.org/10.1016/j.envpol.2020.115723
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organism and to biomagnify within food webs with subsequent consequences for higher trophic level organisms (Bustnes et al., 2003; Fisk et al., 2001; Morel et al., 1998). Many of these pollutants are not only persistent but can be mobilised 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 have been linked to localised inputs from heavily industrialised areas, and the marine environments act as the ultimate sink (Breivik et al., 2007; Jepson et al., 2016). Thus long-lived, high trophic level predators in marine environments with a lipid-rich prey sourced in high latitudes can be especially at risk from elevated concentrations of contaminants in marine environments.

As integral, conspicuous and long-lived components of aquatic ecosystems, seabirds have been used to infer diverse aspects of the health of marine environments (Furness and Camphuysen, 1997; Mallory et al., 2010). Seabird eggs are widely used for contaminant monitoring and inferring toxicological risk (Herzog et al., 2016). During the egg formation period, lipophilic persistent pollutants ingested by the adult female are passed into the developing oocyte with lipid reserves that are essential for the development of the embryo (Speake et al., 1998). Birds may primarily use recently acquired nutrients to form eggs (income breeding) or nutrients from stored reserves (capital breeding) or a combination of both (Bond and Diamond, 2010). Individuals from the same colony may adopt different strategies depending on the breeding condition of the bird (Bond and Diamond, 2010).

Through stable isotope ratio analysis it has been shown that many seabird species are income breeders (Bond and Diamond, 2010), making them suitable bio-indicators of contaminants present in marine environments within the foraging range of the adult female bird from the nesting site. Given the propensity for lipophilic contaminants to bioaccumulate and biomagnify, seabird eggs are a potentially easier matrix to assess contaminant concentration than in other matrices such as water – where such contaminants may be below the detection limit. In addition, it has been noted that other factors such as known sampling location, egg production date, and less inherent variability than other biotic or abiotic matrices result in a high statistical power to detect spatial and temporal variations (Dittmann et al., 2012). However, there are limitations to the use of seabird eggs as environmental indicators as only information on the contaminant burden of an adult female during the breeding season is provided for an income breeder. Female seabirds may have lower contaminant burdens than male conspecifics as they can excrete some of their contaminant burden via their eggs (Lewis et al., 1993), male seabirds may also have different diets than females which may also result in differences in contaminant burdens between males and females (Navarro et al., 2010). Seabirds may also metabolise contaminants at differential rates depending on the species, individual and the type of pollutant (Walker, 1990). Because many seabird species are long-lived with high annual adult survival rates (Wanless et al., 2006), collecting seabird eggs for contaminants analysis is less damaging than lethal sampling of adult birds.

Stable isotope ratio analysis of carbon ($\delta_{13}C$) and nitrogen ($\delta_{15}N$) can also provide information on the dietary niche and trophic level of 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.

The Northern Gannet *Morus bassanus* (hereafter Gannets) is native to the North Atlantic Ocean and is the largest pelagic seabird breeding in the region (Hamer et al., 2001). In Ireland there are approximately 48,000 breeding pairs of Gannets (Newton et al., 2015a), approximately 10% of the worldwide Gannet population (Wanless et al., 2005). Gannets have a broad diet and will eat small prey items such as sandeels *Ammodytes* spp. as well as larger pelagic fish such as mackerel *Scomber scombrus* (Lewis et al., 2003). Gannets, as top-level predators in marine ecosystems, are especially vulnerable to bioaccumulation of harmful contaminants. There have been numerous studies on levels and related effects of contaminants in Gannet eggs including long term studies, spanning several decades, in North America and Europe (Champoux et al., 2015; Crosse et al., 2012). The widespread use of DDT in the 1950s and 1960s negatively impacted the breeding performance of Gannets in Canada by causing eggshell thinning (Chapdelaine et al., 1987). Published data on seabird egg contaminant levels in Ireland is scant, with the most recent data published in a review by Walker (1990). There have been two studies on the levels of mercury in Gannet eggs from Little Skellig and Great Saltee Island from the 1970s and 1980s (Parslow and Jefferies, 1977; Walsh, 1993). No studies on organic pollutants in Gannets or their eggs sampled in Ireland have been published to date.

European Union (EU) Member States are required to monitor and assess the health of their marine waters under the EU's Marine Strategy Framework Directive (MSFD) (2008/56/EC), a major legislative instrument that helps Member States achieve or maintain good environmental status (GES) of their marine environments (EU, 2008; Lyons et al., 2017). Descriptor 8 of the MSFD describes protection against the pollution of marine waters by chemical contaminants. This present study is part of a larger, multi-species and multi-colony, investigation into the feasibility and suitability of using seabird eggs in a monitoring scheme as a higher trophic level indicator of contaminants in Irish marine environments. The objectives of this current study were: 1) to determine baseline levels of contaminants in Gannet eggs in Great Saltee Island and Lambay Island; 2) to assess differences in contaminant levels between breeding colonies supported by stable isotope ratio analysis and; 3) to compare levels observed with known toxic thresholds in other seabird studies to assess the potential impact of persistent pollutants on Gannets in Ireland.

2. Materials and methods

2.1. Sampling location

Lambay Island is a 250 ha island located in the Irish Sea, off the north coast of County Dublin, Ireland (Fig. 1) (Newton et al., 2015b). The island is home to approximately 700 pairs of Gannet (Newton et al., 2015a). Great Saltee Island is an 89 ha island located in the St. Georges Channel, off the southeast coast of County Wexford, Ireland (Fig. 1) (Newton et al., 2015b). The island is located approximately 158 km south of Lambay Island. The St. Georges Channel separates the Irish and Celtic Seas and is located off the southeast coast of Ireland and the southwest coast of Britain. The site is a Special Protection Area (SPA) under the EU Birds Directive. The island is home to approximately 5000 pairs of Gannets (Newton et al., 2015a).

2.2. Collection and preparation of egg samples

All samples in this study were collected under licence from the Irish National Parks and Wildlife Service, and in accordance with the Oslo and Paris commissions (OSPAR) Joint Assessment and Monitoring Programme guidelines (JAMP; OSPAR, 2014). On the May 7, 2017, 20 Gannet eggs were collected from Great Saltee Island. Only freshly laid eggs were collected in this study as the

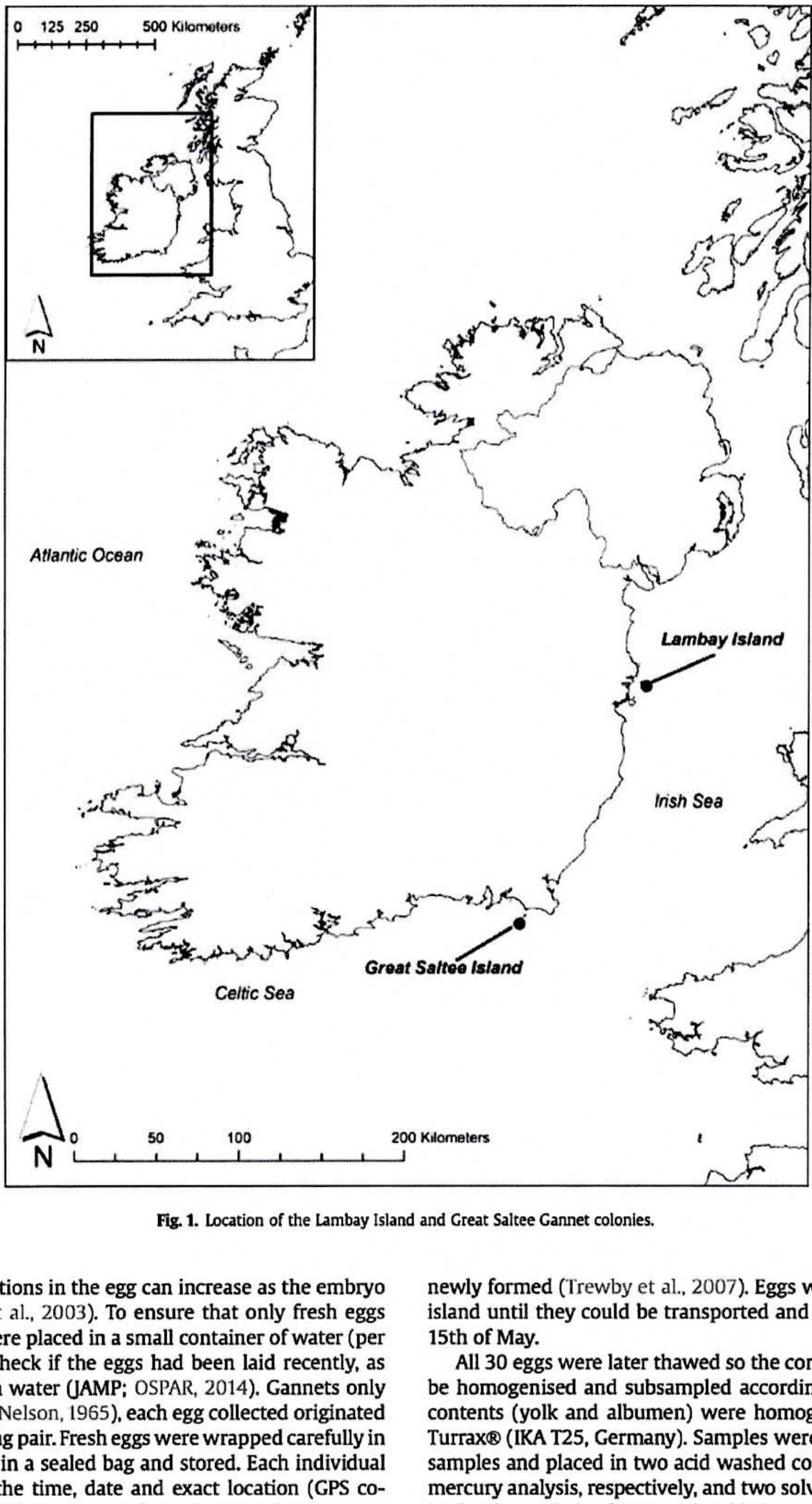

Fig. 1. Location of the Lambay Island and Great Saltee Gannet colonies.

contaminant concentrations in the egg can increase as the embryo develops (Drouillard et al., 2003). To ensure that only fresh eggs were collected, eggs 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). Gannets only lay one egg per clutch (Nelson, 1965), each egg collected originated from a different breeding pair. Fresh eggs were wrapped carefully in aluminium foil, placed in a sealed bag and stored. Each individual egg was labelled and the time, date and exact location (GPS coordinates) were recorded. On return from the island, eggs were immediately refrigerated on the day of sampling and frozen the following day at −20 °C. On the 13th and 14th of May, 10 Gannet eggs were collected from Lambay Island. Only 10 eggs were collected from Lambay Island as the colony is relatively small and newly formed (Trewby et al., 2007). Eggs were refrigerated on the island until they could be transported and frozen (−20 °C) on the 15th of May.

All 30 eggs 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). Samples were divided into four subsamples and placed in two acid washed containers for metals and mercury analysis, respectively, and two solvent washed (*n*-hexane) jar for the analysis of POPs and stable isotope ratio analysis. Sample jars were then frozen at −20 °C until chemical analysis. Subsamples for metals and stable isotope ratio analysis were freeze dried (Labconco: Freeze Dryer – Model Freezone & Bulk Tray Drier (12L), USA).

2.3. Contaminant analysis

2.3.1. Quality assurance

A comprehensive analytical quality assurance programme underpinned the sampling and laboratory analyses. This involved routine testing of quality control samples. 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. Limit of Detection (LoD), determined from replicate measurements of blank samples, ranged between 0.03 and 0.38 ng/g ww for POPs and between 0.0003 and 0.06 mg/kg ww for metals (including mercury); no mercury determinations in eggs were below the LoD. Limit of Quantitation (LoQ) ranged from 0.24 to 2.6 ng/g for POPs and 0.007–0.14 mg/kg ww for metals (including mercury). Recovery values, determined from CRM and LRM, varied between 85% and 115% across all analytes.

As an additional quality control, a selection of samples were cross checked 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.

2.3.2. PCBs, PBDEs and OCs analysis

Eggs were analysed for a suite of 14 PCBs (18, 28, 31, 44, 52, 101, 105, 118, 138, 153, 156, 170, 180, 209). The Σ7PCB, also known as the ICES-7, refers to the sum of seven individual PCB congeners (28, 52, 101, 118, 138, 153, 180). They are widely used as indicators of PCB contamination. Eggs were analysed for 7 PBDEs (28, 47, 99, 100, 153, 154, 183) and 17 OCs (HCBD, HCB, α-HCH, γ –HCH, β-HCH, heptachlor, heptachlor epoxide, oxychlordane, trans-chlordane, cis-chlordane, transnonachlor, op-DDE, pp – DDE, op – DDT, op – DDD, pp- DDD, pp – DDT). ΣHCH refers to the sum 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 op-DDE, pp – DDE, op – DDT, op – DDD, pp- DDD and pp – DDT. Egg samples were extracted by 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 lipid. All samples spiked with internal standards (100 mg of C13 isotopically labelled internal standards for PCBs, PBDEs and OCs). An Agilent 6890 gas chromatograph (GC) coupled to a 5973N mass spectrometric detector (MSD) with a 30 m DB5-MS column was used for this analysis. 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. The 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 information (SI).

2.3.3. Metal analysis

Eggs were analysed for 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 0.2g freeze-dried tissue, 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 7700x with High Matrix Introduction (HMI) system).

2.3.4. Mercury analysis

Concentrated nitric acid (4 ml) was added to 0.6–0.8 g of wet tissue, 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.

2.3.5. 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 Gannets. Egg samples were homogenised and freeze dried. Dehydration of samples is needed for stable isotope ration analysis (Carabel et al., 2006). The isotopic composition of organic carbon and nitrogen was then measured in all 30 gannet egg samples 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. All replicates passed quality controls and were within 0.74 standard deviations of expected value. Variation in lipid content can confound interpretations of diet as lipids are depleted in $\delta_{13}C$ compared with protein (Elliott et al., 2014). $\delta_{13}C$ values were corrected using a lipid normalisation equation for aquatic bird egg (Elliott et al., 2014).

2.3.6. Statistical analysis

Concentrations for POPs are presented on a wet weight (ww) and lipid weight (lw) basis in ng/g and concentrations for metals are presented in mg/kg ww and dry weight (dw). All eggs collected in this study were fresh and did not require a correction factor (Stickel et al., 1973). 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 8. Wilk-Shapiro tests were used to assess normality. Differences between colonies for PCBs, PBDEs, OCs, metals, stable isotopes ratios values were tested using a Mann-Whitney test for non-parametric data and an unpaired *t*-test was used for parametric data.

3. Results and discussion

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

In total, all 14 PCBs analysed for in this study were detected in Gannet eggs (Table 1) with mean concentrations ranging from 497 ng/g ww in eggs from Great Saltee Island (range 316–820 ng/g ww) to 842 ng/g ww in eggs from Lambay Island (range 510–1962 ng/g ww). The mean concentration of Σ7PCB was 439 ng/g ww in eggs from Great Saltee Island and 732 ng/g ww in eggs from Lambay Island. Σ7PCB contributed to 85.8 and 83.8% of Σ14PCB for Great Saltee Island and Lambay Island, respectively. PCB-153 was the most dominant congener and accounted for 39.3% of total Σ14PCB ww in Great Saltee Island and 37.3% for Lambay

Table 1
Mean PCB concentrations (ng/g ww and lw) and standard error of the mean (SEM) separated per congener for Gannet eggs from Great Saltee Island (n = 20) and Lambay Island (n = 10). Seven PCBs (Σ7) are −28, −52, −101, −118, −138,−153, and −180.

	Great Saltee Island (n = 20)		Lambay Island (n = 10)	
	Mean ng/g ww (SEM)	Mean ng/g lw (SEM)	Mean ng/g ww (SEM)	Mean ng/g lw (SEM)
PCB-18	0.31 (0.03)	7.20 (0.57)	0.40 (0.06)	7.79 (0.57)
PCB-28	2.18 (0.07)	50.1 (3.48)	2.44 (0.22)	49.4 (3.75)
PCB-31	0.71 (0.02)	16.6 (1.52)	0.84 (0.03)	17.2 (0.93)
PCB-44	0.33 (0.07)	6.54 (1.25)	1.26 (0.24)	24.9 (3.50)
PCB-52	0.68 (0.04)	16.0 (1.67)	0.66 (0.08)	13.4 (1.37)
PCB-101	5.22 (0.37)	117 (8.84)	6.27 (0.86)	122 (7.63)
PCB-105	8.41 (0.64)	191 (16.6)	14.0 (1.39)	277 (13.9)
PCB-118	37.1 (2.19)	841 (60.2)	61.6 (7.12)	1219 (76.4)
PCB-138	92.2 (6.90)	2072 (155)	161 (34.2)	3094 (331)
PCB-153	201 (12.6)	4551 (340)	328 (57.2)	6326 (530)
PCB-156	7.50 (0.44)	170 (12.7)	14.8 (1.79)	294 (20.1)
PCB-170	39.1 (2.67)	882 (69.4)	75.8 (12.2)	1470 (115)
PCB-180	101 (7.07)	2270 (173)	172 (25.5)	3352 (251)
PCB-209	1.18 (26.4)	1.83 (0.07)	2.88 (0.32)	60.4 (7.67)
Σ14PCB	497 (31.6)	11,004 (800)	842 (139)	16,009 (1218)
Σ7PCB	439 (28.2)	9918 (720)	732 (123)	14,176 (1158)

Island (see supplementary material, Fig. S1). Significantly higher levels of Σ14PCB were detected in Lambay Island for both ww concentrations ($P \leq 0.01$) and when converted to lw ($P \leq 0.01$) (Fig. 2).

Seven PBDEs were detected in Gannet eggs (Table 2). The mean concentration of Σ7PBDEs ww in eggs from Great Saltee Island was 2.92 ng/g ww (range: 1.18–5.26 ng/g ww) and 5.45 ng/g ww in eggs from Lambay Island (range: 3.45–10.8 ng/g ww). The dominant congeners in both sites were BDE-154, 153 and 47, accounting for 67% and 68% of the Σ7PBDE concentration in Great Saltee and Lambay Island, respectively (Fig. S2). Significantly higher levels of Σ7PBDEs were detected in Lambay Island for both ww concentrations ($P \leq 0.001$) and when converted to lw ($P \leq 0.01$) (Fig. 3).

Seventeen OCs were detected in Gannet eggs (Table 3). Mean concentrations of HCB in eggs from Great Saltee Island was 14.3 ng/ww (range: 9.21–16.5) and 12.6 ng/ww (range: 9.13–21) in eggs from Lambay Island. Mean concentration of ΣHCHs ww in eggs from Great Saltee Island was 4.83 ng/g ww (range: 1.3–7.6 ng/g ww) and 6.96 ng/g ww (range: 2.8–12.5 ng/g ww) in eggs from Lambay Island. Mean concentration of ΣCHL ww in eggs from Great Saltee Island was 30.2 ng/g ww (range: 19.3–45.3 ng/g ww) and 28.9 ng/g ww in eggs from Lambay Island (range: 18.6–47.1 ng/g ww). Mean concentration of ΣDDTs (plus metabolites) ww in eggs from Great Saltee Island was 35.1 ng/g ww (range: 15.3–58.2 ng/g ww) and 52.6 ng/g ww in eggs from Lambay Island (range 19.8–202 ng/g ww). There were no significant differences in HCB, ΣHCHs, ΣCHL, ΣDDTs (plus metabolites) ww and lw concentrations between the two study sites. The major metabolite of DDT, pp-DDE, accounted for 90 and 92.8% of ΣDDTs in Great Saltee and Lambay Island respectively.

All fifteen metals analysed for were detected in Gannet eggs (Table 4). There was no significant difference between concentrations of metals in Gannet eggs between Great Saltee Island and Lambay Island, with the exception of mercury. Mean concentrations of total mercury in eggs from Great Saltee Island was 0.4 mg/kg ww (range: 0.25–0.82 mg/kg ww) and 0.62 mg/kg ww (range: 0.42–0.81 mg/kg ww) in eggs from Lambay Island. Significantly higher levels of total mercury were detected in Lambay Island for both ww concentrations ($P \leq 0.001$) and dw ($P \leq 0.001$) (Fig. 4).

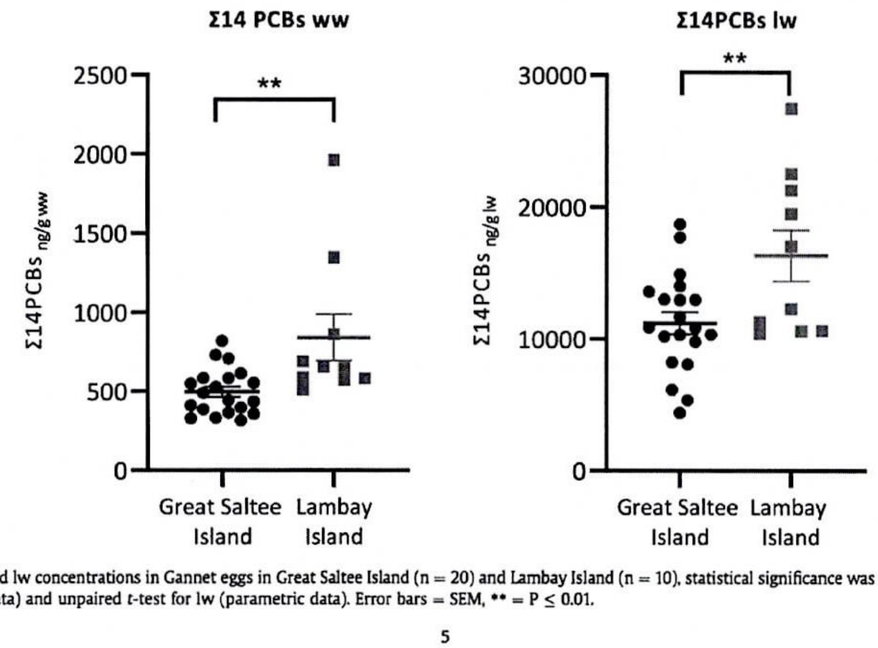

Fig. 2. Σ14PCB ng/g ww and lw concentrations in Gannet eggs in Great Saltee Island (n = 20) and Lambay Island (n = 10), statistical significance was tested by Mann Whitney test for ww (non parametric data) and unpaired *t*-test for lw (parametric data). Error bars = SEM, ** = $P \leq 0.01$.

Table 2
Mean Σ7PBDE concentrations (ng/g ww and lw) and standard error of the mean (SEM) separated per congener for Gannet eggs from Great Saltee Island (n = 20) and Lambay Island (n = 10).

	Great Saltee Island (n = 20)		Lambay Island (n = 10)	
	Mean ng/g ww (SEM)	Mean ng/g lw (SEM)	Mean ng/g ww (SEM)	Mean ng/g lw (SEM)
BDE-28	0.11 (0.02)	2.45 (0.51)	0.47 (0.11)	9.20 (1.40)
BDE-47	0.59 (0.07)	14.1 (1.96)	0.87 (0.14)	16.7 (1.45)
BDE-99	0.34 (0.04)	7.94 (1.14)	0.38 (0.03)	7.59 (0.42)
BDE-100	0.35 (0.05)	7.61 (1.09)	0.85 (0.10)	17.1 (1.49)
BDE-153	0.54 (0.04)	12.7 (1.36)	0.98 (0.17)	19.2 (1.78)
BDE-154	0.84 (0.07)	20.8 (2.94)	1.86 (0.24)	37.3 (3.17)
BDE-183	0.14 (0.05)	3.15 (1.13)	0.05 (0.03)	1.27 (0.72)
ΣPBDE	2.92 (0.22)	68.7 (7.31)	5.45 (0.65)	108 (7.71)

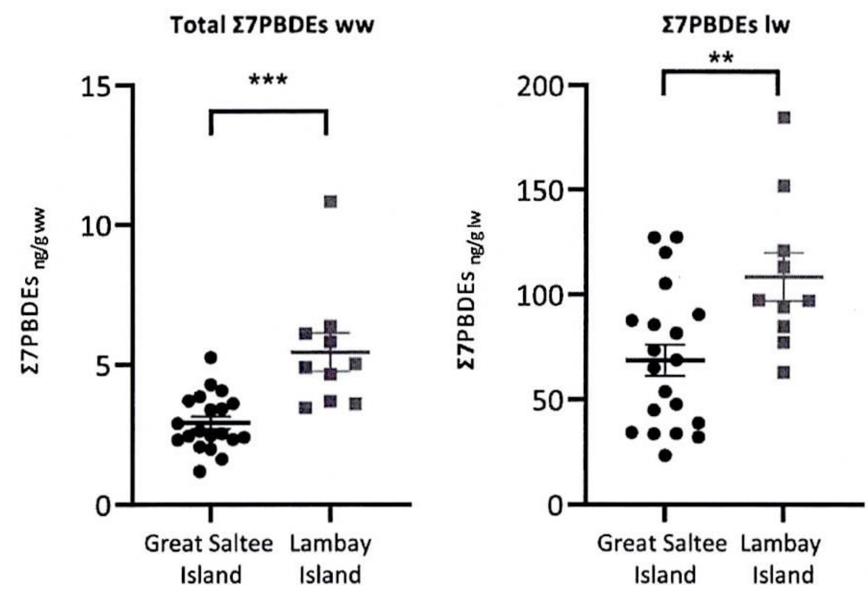

Fig. 3. Σ7PBDE ng/g ww and lw concentrations in Gannet eggs in Great Saltee Island (n = 20) and Lambay Island (n = 10), statistical significance was tested by Mann Whitney test for ww (non parametric data) and by unpaired *t*-test for lw (parametric). Error bars = SEM, ** = $P \leq 0.01$ *** = $P \leq 0.001$.

Table 3
Mean OC concentrations (ng/g ww and lw) and standard error of the mean (SEM) separated per compound for Great Saltee Island and Lambay Island.

	Great Saltee Island (n = 20)		Lambay Island (n = 10)	
	Mean ng/g ww (SEM)	Mean ng/g lw (SEM)	Mean ng/g ww (SEM)	Mean ng/g lw (SEM)
HCBD	0.48 (0.06)	10.8 (1.47)	0.33 (0.05)	7.20 (1.33)
HCB	14.3 (0.72)	326 (23.5)	12.60 (0.52)	252.1 (8.47)
α-HCH	0.54 (0.07)	13.2 (2.48)	0.75 (0.09)	16.4 (2.83)
β −HCH	2.80 (0.29)	66.6 (9.64)	4.19 (0.31)	84.8 (6.40)
γ −HCH	1.48 (0.25)	31.7 (4.76)	2.01 (0.45)	47.6 (13.9)
Heptachlor	0.63 (0.11)	14.5 (2.77)	1.16 (0.09)	23.8 (2.32)
Heptachlor Epoxide	4.10 (0.27)	95.6 (9.84)	4.21 (0.32)	86.3 (7.70)
Oxychlordane	10.2 (0.56)	232 (16.8)	9.05 (0.75)	179 (15.8)
trans-chlordane	0.24 (0.03)	5.55 (0.67)	0.21 (0.02)	4.0 (0.31)
cis-chlordane	1.68 (0.13)	38.3 (3.44)	1.91 (0.18)	36.9 (2.63)
Transnonachlor	13.3 (0.92)	300 (22.1)	12.4 (1.13)	238 (11.5)
op-DDE	0.07 (0.02)	1.55 (0.42)	0.06 (0.01)	1.35 (0.38)
op-DDT	0.24 (0.03)	5.50 (0.62)	0.19 (0.02)	3.85 (0.31)
op-DDD	0.17 (0.06)	3.60 (1.28)	0.23 (0.05)	4.18 (0.82)
pp-DDD	1.23 (0.09)	27.4 (2.17)	1.30 (0.16)	24.74 (1.97)
pp-DDE	32.0 (2.36)	724 (61.1)	49.4 (11.0)	895 (142)
pp-DDT	1.43 (0.12)	32.2 (2.94)	1.37 (0.17)	26.2 (2.32)
ΣHCH	4.83 (0.29)	112 (11.9)	6.96 (0.59)	149 (19.5)
ΣCHL	30.2 (2.01)	686 (55.6)	28.9 (2.5)	569 (40.2)
ΣDDT	35.1 (2.53)	794 (65.5)	52.6 (11.4)	955 (146)

Table 4
Mean metal concentrations (mg/kg ww and dw) and standard error of the mean (SEM) for Gannet eggs from Great Saltee Island (n = 20) and Lambay Island (n = 10).

	Great Saltee Island (n = 20)		Lambay Island (n = 10)	
	Mean mg/kg ww (SEM)	Mean mg/kg dw (SEM)	Mean mg/kg ww (SEM)	Mean mg/kg dw (SEM)
Al	0.13 (0.03)	0.71 (0.16)	0.09 (0.01)	0.49 (0.07)
V	0.001 (0.0003)	0.01 (0.001)	0.001 (0.0001)	0.01 (0.00)
Cr	0.01 (0.001)	0.04 (0.003)	0.01 (0.001)	0.03 (0.01)
Mn	0.20 (0.01)	0.15 (0.03)	0.19 (0.02)	0.10 (0.02)
Fe	18.2 (1.24)	98 (4.95)	18.7 (2.53)	106 (13.97)
Co	0.003 (0.0002)	0.01 (0.001)	0.002 (0.0002)	0.01 (0.001)
Ni	0.01 (0.001)	0.04 (0.001)	0.01 (0.001)	0.03 (0.005)
Cu	0.59 (0.02)	3.22 (0.08)	0.56 (0.01)	3.17 (0.05)
Zn	7.96 (0.39)	43.3 (1.38)	7.73 (0.32)	43.9 (1.45)
As	0.12 (0.01)	0.63 (0.03)	0.12 (0.01)	0.70 (0.03)
Se	0.59 (0.02)	3.23 (0.06)	0.60 (0.02)	3.43 (0.07)
Ag	0.001 (0.0002)	0.01 (0.001)	0.001 (0.0002)	0.01 (0.001)
Cd	0.001 (0.0003)	0.01 (0.002)	0.003 (0.0001)	0.02 (0.0002)
Pb	0.01 (0.001)	0.03 (0.01)	0.01 (0.001)	0.03 (0.01)
Hg	0.40 (0.03)	2.18 (0.16)	0.62 (0.03)	3.54 (0.21)

3.2. Contaminant sources and stable isotope ratio analysis

The Irish Sea is 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. Higher concentrations of Σ14PCBs, Σ7PBDEs and total mercury in Gannet eggs from Lambay Island compared to Great Saltee Island are likely a result of the former's proximity to Ireland's largest city, Dublin. Elevated levels of persistent pollutants (OCs, PCBs, metals) have also been found in Mussels *Mytilus edulis* in Dublin Bay compared to other sites along the Irish coast, including Wexford (Widdows et al., 2002). Although Dublin has had a significant industrial history (Glennon et al., 2014), it is not to the same extent as more heavily industrialised British or European counterparts. Elevated levels of contaminants have been found in higher-trophic level organisms in Liverpool Bay (Law et al., 1992), sediments in the bay have accumulated considerable levels of mercury from a variety of sources (Dickson and Boelens, 1988).

Gannets are wide-ranging birds and can travel significant distances to forage for food from the colony during the breeding season (100s–1000s km) (Wakefield et al., 2013). Lambay Island and Great Saltee Island are only 158 km apart, which is within the dispersion range of Gannets. Tracking studies of foraging Gannets in Europe have shown that individuals from neighboring colonies (as close as 27 km) have mutually exclusive foraging areas, where one might expect them to overlap (Wakefield et al., 2013). It is also possible the female Gannets stay closer to their respective breeding colonies during the egg formation period. It is proposed that the adult females from different colonies that laid the eggs in this study foraged predominantly in different areas during the egg formation period. The egg formation period for seabirds can be up to 30 days (Goldstein, 1987). Gannets start to return to breeding colonies as

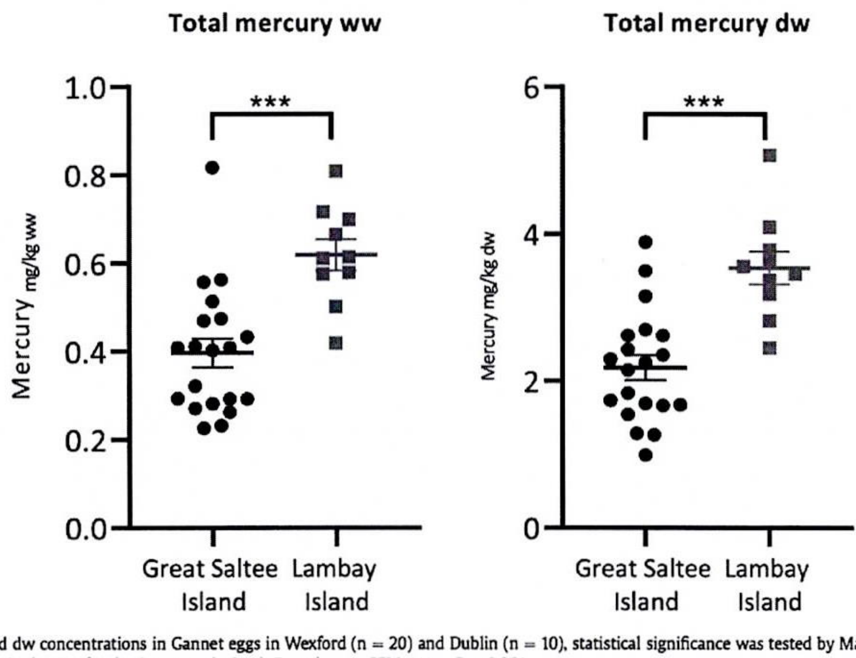

Fig. 4. Total mercury ww and dw concentrations in Gannet eggs in Wexford (n = 20) and Dublin (n = 10), statistical significance was tested by Mann Whitney test for ww (non parametric data) and by unpaired *t*-test for dw (parametric data). Error bars = SEM, *** = $P \leq 0.001$.

Table 5
Mean values of % lipid and stable isotopes ratios of carbon and nitrogen (‰) in Great Saltee Island and Lambay Island, δ13C corrected for lipid using method from Elliott et al. (2014). Statistical significance was tested by unpaired *t*-test (parametric data), ns = not significant.

Location	% lipid Mean (SEM)	C:N ratio Mean (SEM)	$\delta_{15}N$ Mean (SEM)	$\delta_{13}C_{corrected}$ Mean (SE)
Great Saltee Island	4.71 (0.28)	6.82 (0.15)	14.5 (0.1)	−18.01 (0.09)
Lambay Island	5.09 (0.34)	6.58 (0.16)	14.8 (0.16)	−18.05 (0.12)
Comparison of sites	ns	ns	ns	ns

early as January and begin egg-laying in mid-April (Nelson, 1965). Stable isotope ratio analysis showed no significant difference between the two sites indicating that Gannets from both sites have similar dietary composition ($\delta_{13}C$) and they fed at similar trophic levels ($\delta_{15}N$) (Table 5). It is possible that different prey items have similar isotopic signatures but with different contaminant burdens. However, given the overlap, with little variation, of isotopic signatures between sites and the proximity of the two colonies it is likely that Gannets are feeding on the same, or very similar, prey items. Therefore, differences in contaminant levels between the two sites is not expected to be driven by dietary composition but by differences in feeding locations. Gannets from Lambay Island are in closer proximity to Dublin Bay and more exposed to elevated levels of contaminants. Tracking studies indicate that Gannets from Lambay ($n = 3$) stay close to Dublin Bay during foraging trips. Birds from Great Saltee Island ($n = 35$) travelled primarily south-west of the colony with only a small number of birds travelling north into Irish Sea (Wakefield et al., 2013).

It is unknown to what extent the Northern Gannet uses recently derived nutrients to form their eggs (Champoux et al., 2015). The closely related Cape Gannet *M. capensis* has been reported as an income breeder (Rishworth et al., 2014) and the majority of aquatic birds studied have been shown to be income breeders (Bond and Diamond, 2010; Hobson et al., 1997). Differing levels of contaminants between colonies with similar stable isotope ratios in this study suggest that Northern Gannets are using recently derived nutrients to form their eggs. Stable isotope ratio analysis of blood from adult female Gannets from Great Saltee Island during the same breeding season had similar $\delta_{13}C$ (mean of −18.27) and $\delta_{15}N$ (mean of 14.77) values to this study (−18.01 and 14.05) (Malvat et al., 2020). This provides further support than Gannets are using recently derived nutrients to form eggs as blood isotope values represent local nutrient sources (Bond and Diamond, 2010).

3.3. Comparisons with thresholds

There are no thresholds for any contaminants specifically set for Gannet eggs. Toxic thresholds for other bird species are presented in Table S3. Thresholds for aquatic species or seabirds were selected where possible. ΣPCB concentrations in Gannet eggs from both colonies were far below critical thresholds for adverse effects for PCBs in other seabird species' eggs such as Herring Gull *L. argentatus* and Forster's Tern *Sterna forsteri*. Levels of PBDEs fell far below critical thresholds for adverse effects for two birds of prey, American Kestrel *Falco sparverius* (Fernie et al., 2009) and Osprey *Pandion haliaetus* (Henny et al., 2009). The levels of HCB and heptachlor epoxide were also far lower that toxic thresholds for Japanese Quail *Coturnix coturnix japonica* (Schwetz et al., 1974) and American Kestrel (Henny et al., 1983). Levels of DDE were lower than toxic thresholds for the Brown Pelican *Pelecanus occidentalis*, a species particularly sensitive to the impacts of DDE (Blus, 1982). Levels of selenium also fell below suggested toxic thresholds and are low enough to suggest that they be deficient in this metal (Ohlendorf and Heinz, 2011). Shore et al. (2011) suggest that reproductive success may be impaired in most bird species with mercury concentrations of ≥0.6 mg/kg in the egg, a threshold exceeded by six out of 10 eggs from Lambay Island and one out of 20 eggs from Great Saltee Island in this study. However, Gannets with higher levels of mercury in their eggs than this threshold in other studies have had favourable breeding success (Eisler, 2010). OSPAR's EcoQO for ΣPCBs, HCB, HCH, DDT and metabolites and mercury for Common Tern *S. hirundo* and Oystercatcher *Haematopus ostralegus* are all exceeded in this study (OSPAR, 2010).

3.4. Persistent organic pollutants - comparisons with other studies and worldwide trends

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 Persistent Organic Pollutants (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 marine environments (Environmental Protection Agency, 2018, 2008; European Parliament, 2003). While the use of PCBs and OCs has been either banned or restricted in Europe since the 1970s and 80s and PBDEs, more recently, since the 2000s, several POPs are still in use outside of Europe such as DDT which is used in malaria vector control (WHO, 2011) amongst other applications. Other major emissions of POPs into marine environments are still prevalent as a result of illegal dumping and burning (Gioia et al., 2011).

Long term declines of PCBs (1990–2004) have been detected in Gannet eggs from colonies in Britain, with the exception of congeners 153, 170, 180 that remained stable (Pereira et al., 2009), and in Canada (1969–2009) (Champoux et al., 2015). PCB levels and profiles in this study are similar to contemporary, internationally reported values for Gannet eggs in both Scotland and Canada (Champoux et al., 2015; Pereira et al., 2009). PCB-153 > 180 > 138 > 118 > 170 dominated PCB profile for contaminants in Gannet eggs, with PCB-153 alone contributing to approximately 40% of ΣPCB in both Great Saltee and Lambay Island (Fig. S1). These highly persistent congeners contribute significantly to PCB burden in other seabird species however PCB profiles can differ (Borgå et al., 2012). It should be noted that different studies of PCBs, and other contaminant groups, in seabird eggs report different suites of congeners. Therefore, ΣPCB values reported may include different PCB congeners depending on the study. Direct comparison of PCB loadings in eggs between studies is difficult to achieve without individual congener information.

PBDEs in Britain have also decreased in Gannet eggs between 1977 and 2007 (Crosse et al., 2012). The mean ww ΣPBDE concentrations in this study (Great Saltee Island: 2.92 ng/g, 1.18–5.26 ng/g, Lambay Island: 5.45 ng/g, 3.45–10.8 ng/g) fell within values reported internationally for Gannets eggs in Scotland (BDE-47, 99, 100, 153, 154) and Canada (BDE-47) (Champoux et al., 2017; Crosse et al., 2012) with similar values detected in a range of European seabirds (Helgason et al., 2009).

The concentrations of other OCs has also declined in Gannet eggs in Canada (Champoux et al., 2015), including a dramatic decrease in levels of pp-DDE. Concentrations of other OCs detected in this study were generally lower than in Gannet eggs from Bonaventure Island in Canada, ΣHCHs were similar in both studies while the ΣDDTs found in Gannet eggs in Canada were more than double the levels found in those from Ireland (Champoux et al., 2015). It is likely that the long-term trend in concentrations of POPs in Ireland is similar to the aforementioned studies.

3.5. *Metals - comparisons with other studies and worldwide trends*

Zinc, iron, copper, manganese, chromium, vanadium, nickel, selenium and cobalt are biologically essential metals (Metcheva et al., 2011). Essential metals such as iron and selenium can be toxic at high levels (O'Hara et al., 2011). Zinc levels at both sites were lower than levels recorded in Gannet eggs from Scotland (Parslow and Jefferies, 1977). Few data exist on levels of iron in seabirds, or their eggs, and it is not considered a toxic threat in the same way as other metals are (Walsh, 1990). The levels of chromium, manganese, and nickel in seabird eggs were also low in this study but information on their levels elsewhere and their impact on seabirds is limited (Walsh, 1990). Cadmium, lead, arsenic, aluminium, silver and mercury are toxic trace metals (Metcheva et al., 2011). Individual levels of cadmium, lead and silver were low in this study (<0.04 mg/kg). Cadmium levels in birds are generally very low and eggs are not considered to be an important source of cadmium elimination (Wayland and Scheuhammer, 2011). Seabirds are less exposed to lead as other bird groups such as birds of prey, scavengers and terrestrial birds in areas with spent ammunition containing lead (Franson and Pain, 2011). The levels of silver and arsenic in Gannet eggs in this study were low (<1 mg/kg ww). Mean mercury concentrations in Great Saltee Island (0.4 mg/kg ww) are similar to levels from the same site from 1987 to 1988 (0.3, 0.38 respectively) (Walsh, 1993). Gannet eggs from Little Skellig, Ireland's largest Gannet colony situated in the Atlantic Ocean off the southwest coast of Ireland, in 1973 and 1987 had similar mean mercury concentrations to contemporary levels in Lambay Island (0.62 mg/kg ww) (Parslow and Jefferies, 1977; Walsh, 1993). Mercury concentrations in this study were also higher than levels detected in Gannet eggs in Bonaventure Island, Canada (0.2 mg/kg) (Champoux et al., 2015). Mercury concentrations in Gannet eggs from Canada declined between 1969 and 2009 similarly to those at Ailsa Craig, Scotland between 1974 and 2004 but increased marginally at Bass Rock in the North Sea (Champoux et al., 2015; Pereira et al., 2009). Global fluctuation (both upward and downward) in mercury levels has been reported over time in seabird eggs (Burgess et al., 2013; Weseloh et al., 2011). Contrasting trends may be a result of differences in mercury emissions between Europe and North America, where mercury emissions are more tightly controlled and are generally decreasing, and other regions, such as Asia, where emissions are reported to be increasing (UNEP, 2013). Location specific and dietary differences between sites and populations may also explain these differing trends. It is also possible for levels of mercury to increase in higher trophic level predators despite less mercury being biologically available. Simulations suggest that warming seawater can increase bioaccumulation rates of mercury in organisms (Schartup et al., 2019) and dietary changes in pelagic fish in response to overfishing can also lead to changes bioaccumulation rates (Schartup et al., 2019).

3.6. *Potential impact of persistent pollutants on Gannets in Ireland*

While levels of contaminants including DDT were high in both Europe and North America during the 1950s and 60s, the population of Gannets in Ireland and Europe have increased and their range has expanded (Lloyd et al., 2010). This is the opposite of Quebec, Canada where populations and productivity declined during this period (Chapdelaine et al., 1987) as a result of DDT causing eggshell thinning. More recently, the Irish Gannet population increased by over 30% in a 10-year period (2004–2014) (Newton et al., 2015a). Gannet productivity in 2007 from the Ireland's Eye colony (<10 km from Lambay Island) was 0.69 (chicks fledged per pair) (Trewby et al., 2007). This is very similar to productivity of 0.72 for Gannets in Britain between 1986 and 2010 (JNCC, 2011). The breeding success in North Sea Gannet colonies also appears to be stable (Hamer et al., 2007). Gannet population increases in Europe are likely to be a result of their high adult survival rates (>90% annual adult survival rate) combined with their dietary behaviour (Wanless et al., 2006). Gannets are adaptable and their large size and their ability to dive up to depths of 20m (Brierley and Fernandes, 2001) allow them to hunt a wide variety of prey items. Gannets also feed on fisheries discards (JNCC, 2011), giving them access to a range of prey items not normally available to them (Grémillet et al., 2008). Changes to the EU Common Fisheries Policy in relation to fisheries discard may have an impact on the population growth of Gannets. As Gannets have great dietary flexibility, they are likely to be able to adapt their diet in response to declines in discard (Bicknell et al., 2013). However, if pelagic fish populations are not sufficient it may negatively impact the health of Gannet populations (Bicknell et al., 2013). The population growth and range expansion of Gannets in Europe and Ireland coupled with high productivity suggest that the levels of persistent pollutants in Gannet eggs are not having an immediate impact on the breeding success and population of Gannets on Lambay or Great Saltee Island. However, it is possible the Gannets highly successful feeding strategy masks the negative impact of contaminants. A study of a top predator seabird in the north-eastern Atlantic suggests that the negative impacts of contaminants may be mitigated by improved feeding conditions (Bustnes et al., 2015). During adverse circumstances even low levels of contaminants may have negative ecological consequences on seabirds (Bustnes et al., 2015). Gannets in poor breeding conditions may be more vulnerable to the negative impacts of persistent pollutants. The possibility that the cumulative impact of legacy pollutants as well as untested emerging pollutants is having a negative impact on Gannets cannot be ruled out.

4. Conclusions

This study provides a comprehensive overview of POPs and metals in Gannet eggs in two island colonies on the east coast of Ireland. Elevated levels of Σ14PCB, Σ7PBDEs and total mercury were determined in Lambay Island compared to Great Saltee Island, most likely due Lambay Island's greater proximity to Dublin city. There was no significant different in levels of other OCs and metals between the two colonies. Differences in some contaminant burden between two colonies is most likely a result of mutually exclusive feeding areas of Gannets from different sites. This is supported by stable isotope ratio analysis which shows that Gannets from both sites occupy a similar dietary niche. There are no thresholds for any contaminants specifically set for Gannet eggs. Levels of persistent pollutants in Gannet eggs fall below most existing thresholds for adverse effects in other species, with the exception of mercury. However, thresholds set for mercury are not specific to Gannets and may not impact their reproductive success. Persistent pollutant levels are within internationally reported values for Gannets and other seabirds and it is likely that the long-term trend in concentrations of contaminants in Ireland is similar to other studies. Few data exist on contaminants in seabird eggs in Ireland and this study may help inform future contaminant monitoring programmes for seabirds.

Author contributions

Philip White: Conceptualization, Methodology, review & editing, Supervision, Andrew Power: Conceptualization, Methodology, Investigation, Writing - original draft, Visualization. Brendan McHugh: Conceptualization, Methodology, review & editing, Supervision, Simon Berrow: Conceptualization, review & editing, Supervision, Moira Schlingermann: 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.

Acknowledgements

We thank the Neale and Baring family for permission to work on Great Saltee and Lambay Island. The National Parks and Wildlife Service, particularity David Tierney and Alyn Walsh, for generously sharing their knowledge on Gannets. We are grateful to Heidi Acampora, Niall Keogh, Peter Cutler, Brian Burke, Emma Cartuyvels, Vincent Hyland and Kristina Steinmetz for valuable assistance in the field. We thank Cillian Gately and Dorota Bielesza for helping with laboratory work. This Cullen Fellowship (Grant-Aid Agreement No.CF/16/01) is carried out with the support of the Marine Institute, and funded under the Marine Research Programme 2014-2020 by the Irish Government.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2020.115723.

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