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First report of chlorinated and brominated hydrocarbon pollutants in marine bird eggs from an oceanic Indian Ocean island

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Highlights

- The first POPs data in marine bird eggs from an oceanic Indian Ocean island
- We sampled Sooty Tern and Common Noddy eggs
- POPs levels were very low, but Sooty Tern eggs overall had higher levels
- Very low levels of PBDEs were quantified
- Rodrigues is an excellent site to monitor changes in background POP levels

ABSTRACT

We report for the first time levels of persistent organic pollutants in marine bird eggs from an oceanic island in the Indian Ocean, the world's third largest ocean. Ten eggs each of the Common Noddy, also known as the Brown Noddy (*Anous stolidus*), and Sooty Tern (*Sterna fuscata*) were collected from Ile Cocos off the coast of the island of Rodrigues, located 560 km east of the island of Mauritius. ΣPCBs had the highest levels (2.2 and 2.6 ng/g wm, wet mass; 20 and 19 ng/g lm, lipid mass) for common Noddy and Sooty Tern, respectively (and following), then ΣDDT (1.9 and 3.1 ng/g wm; 17 and 23 ng/g lm), and mirex (0.96 and 0.69 ng/g wm; 8.7 and 5.0 ng/g lm). ΣChlordanes (0.094 and 0.15 ng/g wm; 0.48 and 0.73 ng/g lm) and Σtoxaphenes (0.26 and 0.61 ng/g wm; 2.4 and 5.9 ng/g lm) are rare data for these compounds from this ocean. Brominated flame retardants were low (0.08 and 0.07 ng/g wm; 0.7 and 0.7 ng/g lm). Multivariate analyses indicated different contamination patterns in the prey items as Sooty Terns had significantly higher levels of mean Σchlordanes and Σtoxaphenes, as well as CB105, -108 and -157. *p,p'*-DDE had an association with thinner eggshells in the Sooty Tern. Although the contaminant levels were in all respects low, industrialisation, development on the periphery, commercial exploitation of the marine environment, and pollutants transferred over long distances by marine debris is likely to add to chemical pressure in this region. Monitoring changes in background levels of pollutants in remote regions will indicate such trends, and marine bird eggs from Rodrigues would be an excellent site.

Key words:

Mauritius; Rodrigues; Common Noddy; Sooty Tern; PBDE; PCB; DDT; Eggshell thickness

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1. Introduction

One of the major characteristics of persistent organic pollutants (POPs, as defined by the Stockholm Convention) is the potential for long-range transport (LRT). Combined with being bioaccumulative and toxic, POPs pose a threat in isolated areas worldwide as it is found in many biotic and abiotic media (*inter alia* Bargagli, 2008; Braune et al., 2001; Chen and Hale, 2010; Savinova et al., 1995; Tanabe et al., 2004). It is therefore surprising that the world's third largest water body, the Indian Ocean (Costello et al., 2010), is so deficient in information regarding POP residues in the environment. What is available is about POPs in water and air (*inter alia* Chen and Hale, 2010; Tanabe et al., 1982; Tanabe et al., 2004; Würfl et al., 2006), coastal sediment (Barasa et al., 2007), plastic pellets (Ogata et al., 2009), dolphins (Mwevura et al., 2010), and pelagic fish (*inter alia* Ueno et al., 2004; Ueno et al., 2005). PCB residues were reported for fish and sediment from Saint Paul and Amsterdam Islands (Monod et al., 1995), and in plankton south-west of Africa (Joiris and Overloop, 1991). Even less is known about brominated flame retardant residues in the Indian Ocean (Chen and Hale, 2010; Li et al., 2011). Although not a POP, PAHs have been measured in oysters from Mayotte (Thomassin et al., 2011). Much more POP information is available from the adjacent Southern Ocean (*inter alia* Bustnes et al., 2007; Court et al., 1997; Guruge et al., 2001; Tanabe et al., 2004; van den Brink et al., 2011).

The only information we could find on POPs in marine bird eggs from any Indian Ocean island was by de Kock and Randall (1984) reporting on PCBs and organochlorine pesticides in Penguin, Gannet, and cormorant eggs from a South Africa coastal island at the far western part of the Indian Ocean. De Kock and Boshoff (1987) also reported on POPs in marine bird eggs from mainland colonies along the coast of South Africa, and POPs were reported for marine bird tissue from the coast of India (Kunisue et al., 2003). Given the global importance of POPs, the role of seabirds in marine ecology, and seabirds as indicator of contamination (Mallory et al., 2010), the lack of knowledge on POPs in marine bird eggs from any oceanic island in the Indian Ocean needs to be addressed. We report here on levels of POPs, including to our knowledge the very first reports of brominated flame-retardants, chlordanes, and toxaphenes, in any marine bird egg from the Indian Ocean.

2. Methods

2.1. Species and sampling site

We concentrated on colonial breeding species, avoiding those that are rare or endangered. Both species selected for this study are Terns (family Sternidae). The Common Noddy, also known as the Brown Noddy (*Anous stolidus*), weighs about 185 g and is 40 cm long (Gochfeld and Burger, 1996). Based on a study from the Seychelles (Catry et al., 2009), Common Noddies forage at intermediate distances from shore (50-300 km), and feed its chicks mainly on fish items (90% of induced regurgitated prey items) consisting mainly of Mullidae (40%), Carangidae (10%), Engraulidae (15%), Exocoetidae (3%), and fish larvae (17%). Cephalopods made up 11% of the items. The Sooty Tern (*Sterna fuscata*) weighs about 180 g and is 42 cm long, and usually forages more than 300 km offshore (Gochfeld and Burger, 1996). The Sooty Tern is also the most abundant tropical marine species, with 6 million pairs estimated to breed in the southwest Indian Ocean (Feare et al., 2007; Jaquemet et al., 2008). Its chicks regurgitated very much the same prey items as the Common Noddy; 82% of the fish items were made up of Mullidae (35%), Carangidae (10%), Engraulidae (9%),

Exocoetidae (19%), and fish larvae (1%) (Catry et al., 2009). Cephalopods contributed 18% of prey items. Mean prey lengths were almost the same; 70.0 mm and 70.7 mm for Common Noddy and Sooty Tern, respectively. The Morisita-Horn overlap index of dietary composition indicated a significant overlap of 0.851 between the two species (Catry et al., 2009). Another study on regurgitated prey items of Sooty Tern chicks from four different islands in the Mozambique Channel and Seychelles by the same team found that they were fed significantly different proportions of types of food, reflecting changes in prey availability and seasonality of breeding (Jaquemet et al., 2008). The foraging and breeding ecology of the Sooty Tern was concluded to be adaptive to season and prey availability. Sooty Terns breed exclusively on the ground in large colonies, and Common Noddies breed on the ground and on branches in somewhat looser aggregations (personal observations, Vikash Tatayah, and Mauritian Wildlife Foundation).

The island of Rodrigues (Fig. 1), 560 km east of Mauritius, is an autonomous region of the Republic of Mauritius, has a land area of 110 km², 40 000 inhabitants, and no industrial plants. Handcraft industry, agriculture, tourism, and fishing are the major economic activities. There are an estimated 5 000-10 000 and 5 000-6 500 annual breeding pairs of Common Noddy and Sooty Tern, respectively, on Ile Cocos and Ile aux Sable, an 'Open Nature Reserve' of Rodrigues (Fig. 1) (Jones et al., 2010). After an absence of almost a century, Sooty Terns re-colonised Ile Cocos and Ile aux Sable in the mid 1980s. The closest colony is on St Brandon's Rock (Cargados Carajos Shoals) 550 km to the northeast (estimated 300 000 pairs, unpublished data), and Ile aux Serpents offshore of Mauritius and 575 km away, with about 200 000 - 250 000 pairs (Feare et al., 2007). It is presumed that the colony on Ile Cocos and Ile aux Sable was re-established by birds from Bird Island in the Seychelles (1 950 km away) as birds ringed in the Seychelles in the 1990s were sighted on Ile Cocos in 2009 and 2012 (personal observations by Mauritian Wildlife Foundation personnel).

2.2. Egg collection and preparation

Permission to collect eggs from Ile Cocos was obtained from two authorities: the National Parks and Conservation Service of the Ministry of Agro Industry, Food Production and Security, Mauritius, and the Commission for Agriculture, Natural Resources Rehabilitation and Water Resources, Rodrigues Regional Assembly. On 10 December 2008, ten eggs from ground nests of each species were collected from the breeding colonies on Ile Cocos, wrapped in foil, and frozen on the same day. Collections were done in partnership with local authorities and conservationists from the Mauritian Wildlife Foundation. Both species lay single eggs (Ricklefs and White, 1981) and Sooty Terns may lay a second egg when an egg is removed (Feare, 1976).

In Mauritius, the eggs were thawed and the egg contents homogenised with an ultrasonic homogeniser. The probe was thoroughly cleaned between samples by wiping with tissue paper, rinsing with soapy water, double distilled water, followed by pesticide grade acetone and hexane. Samples were individually stored frozen in HDPE containers and shipped to Norway where they arrived frozen. Membranes attached to the eggshells were removed with careful rinsing, placed in labelled individual foil containers, and dried in a desiccator for three months. Eggshell thickness at the equator was measured with an electronic calliper (Kroeplin B2R20S) accurate to 1 µm.

2.3. Extraction and analyses

The chemical analyses were performed in the accredited Laboratory of Environmental Toxicology at the Norwegian School of Veterinary Science (NS-EN ISO/IEC 17025 (TEST 137)). Because we expected low concentrations and to save costs, a pool of each species was analysed to determine target compounds for individual sample analyses. The levels of brominated flame retardants were so low that they were excluded from individual sample analyses, but levels obtained for pooled samples were included in Table 1. The extraction and clean-up procedures were based on Brevik (1978), modified as described elsewhere (Bouwman et al., 2008; Helgason et al., 2008; Polder et al., 2008b). Briefly, 2-3 g of the homogenized sample was weighed and the internal standards CB29, -112 and -207 (Ultra Scientific, RI, USA) added to all samples, as well as 10 ml grade 1 water, HPLC grade cyclohexane and glass-distilled acetone (3:2) (Rathburn Chemicals Ltd., Walkerburn, UK), and NaCl

(6%; Merck KGaA, Darmstadt, Germany). Additional internal standards BDE77, -119 and ^{13}C -209 (Cambridge Isotope Laboratories, Inc., Andover, MA, USA) were added only to the pooled samples. Liquid-liquid extraction was performed. The extracts were centrifuged (2095 g), the supernatant removed, concentrated and adjusted to 5 ml. Lipid determination was done gravimetrically; a 1-ml aliquot of the extract was placed in tarred glass containers and evaporated to dryness in a sand bath (40 °C) until the mass stabilised. Extracts were treated twice with concentrated sulphuric acid (Fluca Analytical, Sigma Aldrich, USA) to remove lipids. The lipid-free extract was then concentrated (evaporated) under a gentle flow of nitrogen gas (99.6%) to 0.4 ml on a sand bath at 40 °C, and transferred to amber GC-vials. A complete description of the GC analysis is provided elsewhere (Polder et al., 2008a). Integration and calculation was done using MSD Chemstation (G1701 version D.01.00; Agilent Technologies, Avondale, PA). The target ions for PBDEs and HBCD were m/z 79/81, for BDE209 m/z 485 and 487, and for ^{13}C -BDE209 at m/z 495 and 497. Limits of detection (LOD) were defined as three times the noise level. For all components, five- to eight-point linear calibration curves were used and calculations were done within the linear range for the component.

The method's quality control included three blanks per series of 15, a chicken egg blank sample, spiked recovery samples (also chicken egg) and an in-house reference sample. The results of the quality parameters were within acceptable ranges. The recoveries ranged between 75-146% (median 101%) for OCs and 91-139% (median 108%) for BFRs. The laboratory's accredited analytical quality has been approved by several international intercalibration tests. Acceptable results were obtained in 2008-2011 in EURL EUPT-AO 06; FAPAS test nr. 0580; MOE for NCP III phase 4 and 5; AMAP; CRL EUPT-AO 03, 04, 05, as well as the Interlaboratory Comparison on HBCD in Biological Samples by the Norwegian Institute of Public Health, Oslo, Norway.

2.4. Data treatment

Data were analysed using GraphPad Prism version 5.04 for Windows, GraphPad Software, San Diego California USA (www.graphpad.com). Most of the data were log-transformed, using only positive quantifications. T-tests were unpaired and two-tailed, and significance in all cases is $p < 0.05$. Multivariate analysis using MjM Software PC-ORD version 6.07 (www.pcord.com) was conducted to investigate patterns of associations between the species and pollutants, and influence of pollutants on eggshell thickness. We used Nonmetric Multidimensional Scaling (NMS) as it avoids the assumption of linear relationships among variables by using ranked distances to linearize the relationships between measured distances in ordination space. NMS deals much better with 0 for <LOQ than other ordination methods (McCune and Grace, 2002). The distance measure was relative Sørensen. Pollutant data were relativized for each egg to investigate pollution profiles using relative proportions of the pollutants. Random starting configurations were used with 250 runs of real data. Monte Carlo tests were done with 250 runs of randomised data. Final stress can be interpreted as follows: <5 excellent, 5–10 good, 10–20 general picture good, but not in detail, >20 not good (McCune and Grace, 2002). To investigate possible linear relationships between compounds and eggshell thickness, a PCA (principle component analysis) on untransformed data was conducted (999 runs) using variance/co-variance for the cross-product matrix.

PC-Ord was also used to conduct non-hierarchical indicator analysis for compounds between the two species to derive indicator values (IV), using the method of Dufrêne and Legendre (1997), with a Monte Carlo test of significance of the observed maximum indicator values with 4999 permutations. This test as used here combines the relative concentrations of 31 compounds (excluding the BDEs and CHB44) with its relative frequency of occurrence in the eggs of each species.

3. Results

3.1. Analyses

Analytical data are presented in Table 1 based on wet mass (wm) and lipid mass (lm). Wet mass was used for statistical analyses and most of the discussion because embryonic metabolism in the egg affects lipid content (Herzke et al., 2002; Speake et al., 1998). Lipid-mass based data are included for

comparisons with other sources. The compounds quantified are shown in Table 1. Compounds that were not detected were CB28 (LOQ 0.014 ng/g wm), BDEs -99, -153, -154, -183, -206, -207, -208, (LOQs 0.01 – 0.16 ng/g wm), PBT (pentabromotoluene), PBEB (pentabromoethylbenzene), DPTE (2,3-dibromopropyl-2,4,6-tribromophenyl ether), HBB (hexabromobenzene), and BTBPE (1,2-bis-(2,4,6-tribromophenoxy)ethane) (LOQs 0.005 – 0.025 ng/g wm). The toxaphene CHB-40 was present but could not be quantified. BDE209, although detected, was present in concentrations lower than the highest detected level in the procedural blanks and therefore not reported in Table 1. HBCD could not be quantified due to technical issues.

3.2. Measurements and concentrations

Sooty Tern eggs (mean 26.5 g) were significantly ($p=0.00002$; t-test) lighter than Common Noddy eggs (mean 30.8 g). Fig. 2A shows the eggshell thickness and lipid content of the eggs. The Common Noddy had significantly thicker shells ($p=0.0095$) and marginally lower lipid content ($p=0.0507$; t-tests). Figs. 2B and 2C illustrates the levels and distribution of chlorinated compounds in the two Tern species. Sooty Terns had significantly higher mean Σ chlordane and Σ toxaphene, but lower mirex (Fig. 2B; t-tests). Although mean Σ DDT and Σ PCB were higher in Sooty Tern eggs (Fig 2C), the differences were not significant. For the individual PCB congeners, Sooty Terns had significantly higher CB105, -118, and -157 (Fig 2D). Although data for PBDEs were only available for pooled samples, Σ PBDE was slightly higher in Common Noddy (Table 1). Table 2 presents comparative data from other sources.

Fig. 3 shows the NMS ordination of the pollutant profiles of the individual eggs, in a bi-plot with the compounds indicated by vectors. Final stress was 6.46 for a two-dimensional solution. Axis 1 explained 86.4% of the ordination, and Axis 2 10.8%, for a cumulative of 97.2%. All Common Noddy eggs except CN9 had positive loadings on axis two, and all Sooty Tern eggs were negatively loaded. The two longest compound vectors were for p,p' -DDE and CHB26. p,p' -DDE (the longest vector) oriented parallel to Axis 1. Sooty Tern eggs are spread along Axis 1 (the first dimension), indicating a wider range of concentrations in Sooty Tern eggs (also shown in the wide spread in Fig. 2C). Sooty Tern eggs in the direction of the p,p' -DDE vector (to the left of the origin) have higher levels, and lower in the opposite direction. The vector for CHB26 (the 2nd longest vector) is oriented almost perpendicular to p,p' -DDE, indicating that the sources of these two compounds are unrelated, as well as CHB26 having higher levels in Common Noddy eggs.

3.3. Eggshell thickness

Eggshell thickness for the two species cannot be plotted as a descriptive variable on the same bi-plot (Fig. 3) as this variable differed significantly between the two species (Fig 2A). PCA plots of the individual species with eggshell thickness as a descriptive variable showed no pattern for Common Noddy, but the eggshell thickness and p,p' -DDE vectors for Sooty Tern (Fig. 4) were in opposing directions; increasing thickness is directly opposite increasing p,p' -DDE levels in dimensions 1 and 3. Axis 1 explained 98.9% of the variance and the plot was rotated 50°. This means that thinner eggshells were associated with higher levels of p,p' -DDE. All other compound vectors were perpendicular to the thickness and p,p' -DDE vectors and therefore had no influence on eggshell thickness. ST37, the thinnest eggshell at 0.258 mm was 8.3% thinner than the median thickness of 0.283 mm.

3.4. Indicator analysis

The indicator values are presented in Table 1. Mirex was the only significant indicator for the Common Noddy. For Sooty Tern eggs, γ -HCH, *cis*-nonachlor, CB-66, -105, -118, -157, and all toxaphenes were significant indicators. These compounds are therefore more likely to have higher levels in their respective species than the other.

4. Discussion

4.1. Species comparisons

The Sooty Tern and Common Noddy are of comparable size and mass and share many types of food (Catry et al., 2009), but the eggs differ in terms of eggshell thickness and %lipid (Table 1, Fig. 2A). The shells of Sooty Tern eggs were 13% thinner than the shells of Common Noddy eggs (Fig. 2A). The Sooty Tern eggs were also 14% lighter (see Section 3.2) but contained 17% more lipid than the Common Noddy eggs (Table 1, Fig. 2A). This suggests that Sooty Terns provide proportionally more lipids to their embryos and chicks than Common Noddies. This might be to accommodate for longer absences from the nests during incubation and chick development as Sooty Terns forage further from shore than Common Noddies. Incubation of the Sooty Tern and Common Noddy are 26-33 days and 28-37 days, respectively, and fledging periods are 56-70 and 40-56 days, respectively, followed by post-fledging dependency (Gochfeld and Burger, 1996). Such comparisons, and how pollutants may interact with incubation, fledging and post-fledging dependency, needs further investigation.

In one of the few studies on pollutants in marine birds from the western Indian Ocean, Catry et al. (2008) detected no differences in trophic feeding ecology based on stable isotopes in feathers or blood, nor any differences in mercury levels between the Sooty Tern and Common Noddy, despite the differences (though overlapping) in prey items and foraging ecology. On the other hand, Ramos and Tavares (2010) found that lower levels of mercury in feathers of inshore foraging species from the Seychelles compared with offshore feeding species such as the Sooty Tern. Unfortunately, Common Noddy was not sampled. Offshore feeding species, Ramos and Tavares (2010) explained, feed at higher trophic levels.

It must also be kept in mind that levels of pollutants in eggs reflect pre-breeding food intake by the female, while chick feathers reflect carry-over from the egg as well as prey items provided by both parents during fledging. Sooty Terns also start laying eggs from 6 years upwards, the longest juvenile (non-breeding) stage of any of the Terns (Gochfeld and Burger, 1996). The longer period before breeding may result in higher levels of accumulation of pollutants in the female Sooty Tern. Whatever the case may be, even if the lipid-based concentrations would be the same between species, a higher lipid content in the egg would result in more hydrophobic compounds in the egg reflected in wet-mass based concentrations.

4.2. Brominated compounds

The two species had comparable levels of the quantifiable BDE47 and BDE100 (near LOQs; Table 1). Compared with marine bird data from other regions, the Tern eggs from Rodrigues had levels of Σ PBDE an order of magnitude less (Tables 1 and 2). The absence of detectable levels of almost all brominated compounds analysed for, especially PBT, PBEB, DPTE, HBB, and BTBPE is notable as these compounds have been detected in remote regions such as in Arctic air (Möller et al., 2011). No results were listed in the global review on PBDEs in birds from the Indian Ocean (Chen and Hale, 2010). We could trace only two articles reporting brominated compounds from the Indian Ocean. A single sample of Skipjack Tuna muscle from Seychelles had no detectable PBDEs (LOQs 0.02-5 ng/g lw; Ueno et al., 2004). High concentrations of brominated compounds of biogenic origin were detected in the blubber of dolphins (210 000 ng/g lw in mature males) caught as by-catch between Zanzibar and mainland Tanzania (Mwevura et al., 2010), but anthropogenic brominated compounds were masked by organochlorine compounds and could not be determined.

4.3. Organochlorines

The organochlorine levels in the two Tern species are provided in Table 1, Figs 2B-D, and are compared with other data in Table 2. Although both species had low levels in all respects, Sooty Tern eggs had significantly higher Σ chlordanes and Σ toxaphenes, with mirex significantly higher in Common Noddy eggs (Fig. 2B). For Σ HCH, HCB, Σ DDT and Σ PCB, there were no differences (Figs. 2B and C), suggesting differences in their food and foraging behaviours. Mean levels of CB74, -99, 105, -118, -138, -153, -156, -157, -170, -180, -183, and -194 were higher in Sooty Terns than in Common

Noddy, but only CB-105, -118 and 157 were significantly higher. Offshore feeding species, Ramos and Tavares (2010) explain, feed at higher trophic levels, probably accounting for the generally higher levels of pollutants we found in Sooty Tern eggs.

Because the egg reflects accumulated and metabolised pollutants in food foraged by the female prior to egg laying, species-related differences in pollutants of prey items, foraging ecology, and breeding cycles is more likely to be reflected in egg contents during this period than in chick feathers during fledging. During fledging, adult foraging patterns change (Young et al., 2010), but changes in prey abundance might also be in effect (Catry et al., 2009). Mcleay et al. (2009) found that Crested Tern chicks were fed with smaller prey items than normally consumed by the adults, and Morrissey et al. (2010) found that female passerines utilised higher trophic level food prior to egg-laying. The significant differences in organochlorine content (Table 1 and Figs. 2B-D), although not large in absolute terms, probably reflect these differences and changes. Sooty Tern females therefore, probably consume prey with a slightly higher organochlorine content (except for mirex) before egg-laying than Common Noddy females.

4.4. Multivariate analyses

Fig. 3 shows the results of an NMS analysis of the eggs and compound profile. Axis 1 was oriented in the directions of p,p' -DDE and p,p' -DDT (opposing), and Axis 2 in the direction of CHB-26. Common Noddy eggs were mostly associated with HCHs, PCBs, mirex, and some of the chlordanes, while Sooty Tern was more associated with CHB-26 and p,p' -DDD. Interestingly, the two species separated distinctly, again indicating females of the two species utilising food with different compound profiles. Multivariate indicator analyses (Table 1) showed that mirex was significantly more likely to have higher levels in Common Noddy than in Sooty Tern eggs, while most all of the toxaphenes, as well as the pesticides γ -HCH and cis-nonachlor were more likely to be significantly higher in Sooty Tern eggs. Again, this analysis shows differences in pollutants in the prey of the female Tern species.

Sooty Terns had higher mean and median p,p' -DDE than Common Noddy (Table 1), but the difference was not significant ($p=0.2132$; t-test). Sooty Tern eggs also had a larger variation in compound profile, mainly in the direction of p,p' -DDE (Fig. 3). Fig 4 investigated this association with eggshell thickness. Although weak, a maximum of 8% thinner shells (compared with the median) was associated with higher p,p' -DDE levels. Although p,p' -DDE is associated with eggshell thinning in many species (Bowerman et al., 1995; Fry, 1995), the low levels detected (max 9.3 ng/g wm in Sooty Tern egg ST37) is far below levels expected to induce such effects. However, the perpendicularity of all other compound vectors on the thickness (Fig. 4) variable is remarkable as it shows no association whatsoever with eggshell thickness and p,p' -DDE. Levels of DDE in other species and colonies in association with eggshell thickness need more attention before firmer conclusions can be drawn.

4.5. Possible sources

Measured in air from the Indian Ocean, BDE47, -49, -99, and -100 were the most prominent, with BDE47 contributing more than 60% (BDE209 was not measured) (Li et al., 2011). The pattern was ascribed to air sourced from the Indian sub-continent and presumed to originate from e-waste recovery activities from the West and South coasts of India. In the absence of any other data from the region and the very low levels of BDE in the eggs, aerial long-range transport seems a contributing source. However, a single Kelp Gull egg from the Atlantic coast of South Africa (Velddrif, near the city of Cape Town) contained 55% BDE47 out of a Σ PBDE of 0.9 ng/g wm (Table 2, and Polder et al., 2008b), suggesting other possible sources or processes.

Temporal air data of organochlorines over the Indian Ocean was interpreted by Würl et al. (2006) for surveys done in 1976, 1989, 1990, 2004/2005. The sampling cluster closest to Mauritius was about 1 500 km away to the north, near the Chagos Atoll and Maldives. From 1976 to 2005, Σ DDT and Σ HCH decreased by up to two orders of magnitude. PCBs in air were not measured prior to 2005, but further to the east near Sri Lanka, a similar reduction was found. However, back trajectories showed different source areas for the air; in 1976, it was from the Bay of Bengal, in 2005

it was mainly oceanic from the east. Würl et al. (2006) attributed the main sources of PCBs as from open burning and military activities on some islands.

The only HCH isomer with insecticidal effect is γ -HCH, which when purified is marketed as lindane, while the environmentally most persistent and bioaccumulative isomer is β -HCH (Walker et al., 1999). When produced the technical HCH mixture consists of 55-80% α -HCH, 5-14% β -HCH, 8-15% γ -HCH, 2-16% δ -HCH, and 1-5% ϵ -HCH (Breivik et al., 1999). Historically, the technical mixture was replaced in insecticide formulation by pure lindane in Europe and North America in the early 1980s (Breivik et al., 1999), but the technical mixture could still be used in developing countries. The absence of detectable levels of α -HCH however, indicates no recent use of the technical mixture in regions that affect the foraging areas of the studied bird populations. The low levels of γ -HCH indicates that the use of lindane has probably ceased in that same area, while the concentrations of β -HCH is a consequence of its higher environmental persistence and bioaccumulation potential relative to the other isomers (Walker et al., 1999).

It should be considered that PCBs and other organochlorines in the eggs were derived from Rodrigues itself. Power generation is by a fuel-oil driven generator (11 MWh installed capacity) at Port Mathurin (Fig. 1). Wind turbines contribute about 4% of electricity generation. However, all transformers on Rodrigues were manufactured after 1990, and the National Implementation Plan of Mauritius for the Stockholm Convention indicated no PCB contaminated oils in the transformers. Open burning and cooking does occur, and the small agricultural activities may have used lindane and chlordanes. A small supply of DDT was kept on the island as insurance against malaria outbreaks but was never needed, as there are no anopheline mosquitoes on Rodrigues. Tern eggs from Rodrigues had the lowest HCB, Σ HCH, Σ DDT and Σ PCB of all marine birds listed in Table 2, and mirex levels was only slightly higher than the lowest level from the South African Kelp Gull egg. The very low levels of all compounds compared with data from elsewhere (Table 2), as well as the feeding patterns of the birds suggest that if the island itself was the source, it contributed very little, and then only indirectly via marine food webs.

Plastics production is increasing globally (five-fold increase 1976-2008) and the amount of debris in the oceans will likely increase due to lack of waste management (Law et al., 2010). Many plastics are manufactured with added chemicals for polymerisation, plasticizers, and flame retardants. Plastics can also accumulate pollutants such as PCBs and DDTs from the water (Ogata et al., 2009), concentrating them within weeks to orders of magnitude higher than in the surrounding water (Mato et al., 2001; Teuten et al., 2007; Teuten et al., 2009). The long-range transport potential of floating plastics is well documented, and plastics with inherent and accumulated chemicals, can be ingested by seabirds (Ryan, 2008) and also end up on distant shores. Our experience on Ile Cocos and other beaches of Rodrigues indicated very little plastics. This was confirmed by Barnes (2004) who quantified marine debris on beaches of the southern Indian Ocean islands and found an accumulation rate of 4.41 (items/km)/year for Rodrigues, which is quite low. With so little plastic washing ashore, it is unlikely that marine debris is a significant vector of organic pollutants towards Rodrigues, but it may well be elsewhere (Heskett et al., 2012).

Ramos and Tavares (2010) showed significant reductions in mercury levels between 1996 and 2005 in marine bird eggs from the Seychelles. Because of its isolated nature, marine bird eggs from Rodrigues would therefore serve as excellent monitors of presence and changes in background levels of pollutants for the region.

5. Conclusions

The Indian Ocean is surrounded by 37 countries inhabited by a third of the world's population. One half of the global shipping container traffic and 70% of petroleum products crosses the Indian Ocean. Resources from Africa and the Middle East have to cross the ocean to satisfy the demands of the growing economies of China, India, and others. Conflict in some parts of the Indian Ocean remains a chronic problem, emphasising the strategic role this ocean will play in the future (Kaplan, 2010). Economic, political, military, and social conditions in and around the Indian Ocean are changing

rapidly which may well result an increased release of pollutants. Although the classic chlorinated compounds such as PCBs, DDT, and HCHs will gradually decrease due to Stockholm Convention interventions, other pollutants such as PAHs, plastics and pharmaceuticals associated with increased industry, transport, and consumerism may well see an increase. We report, for the first time to our knowledge, brominated flame retardants (near LOQ) and toxaphenes in marine bird eggs from an island in the Indian Ocean. Levels of organic pollutants in Tern eggs from Rodrigues were very low compared with data from the Antarctic and other regions. Although we found indications of eggshell thinning associated with *p,p'*-DDE in Sooty Terns, it should be treated with caution as the levels were low. Since Rodrigues is distant from any notable sources of organic pollutants as witnessed by the low levels we found, it could serve as a site for monitoring temporal changes in background levels of POPs and other pollutants.

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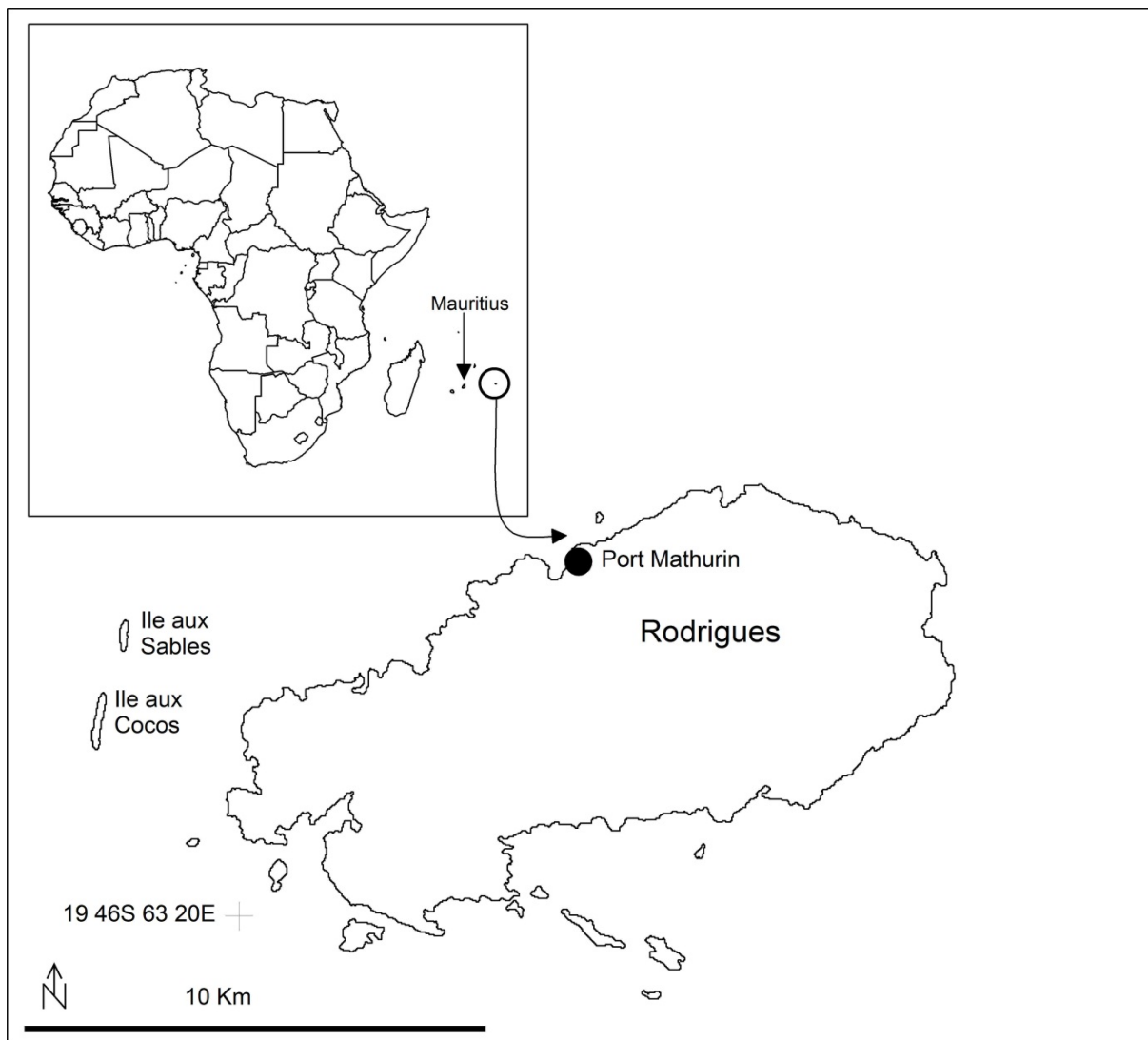


Fig.1. Location of Rodrigues and Ile Cocos in the Indian Ocean.

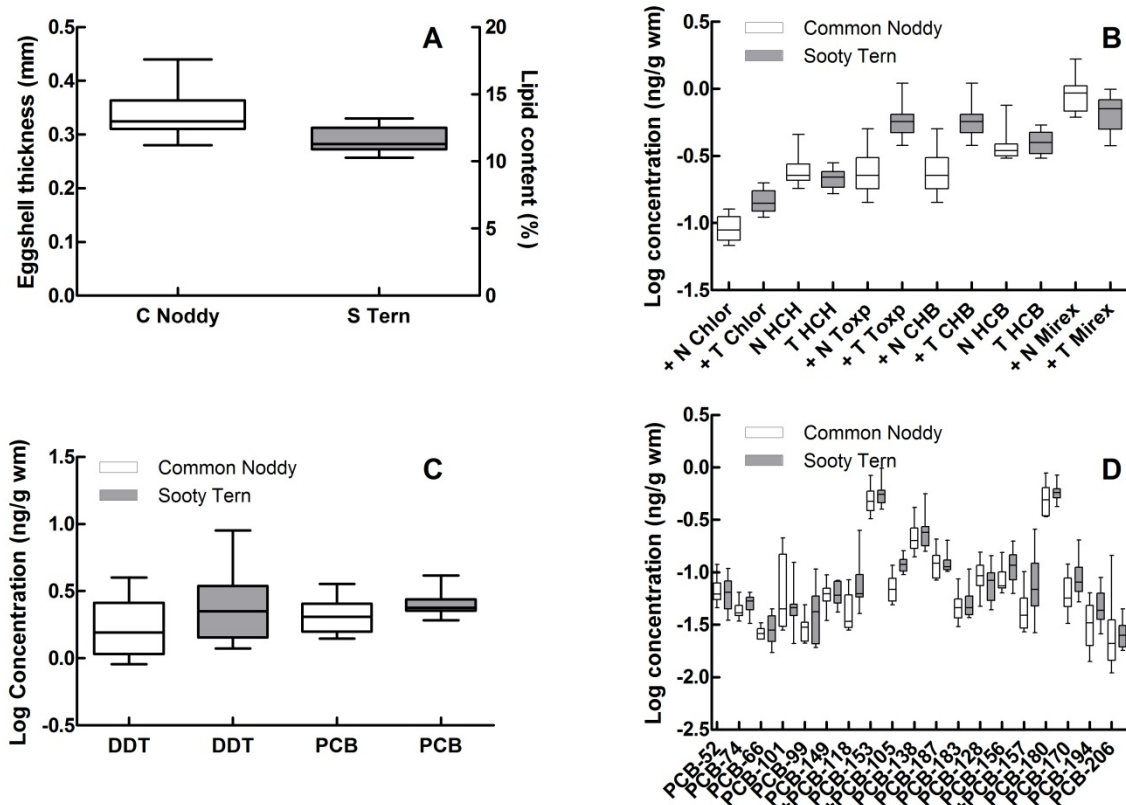


Fig. 2. Box and whisker plots (horizontal lines are medians, 25 and 75% confidence intervals, and minima and maxima) of: (A) Eggshell thickness (left axis) and egg lipid content (right axis) for Common Noddy (C Noddy) and Sooty Tern (S Tern). (B) of log concentrations of Σ chlordanes, Σ HCHs, Σ toxaphenes (Σ CHB), and HCB and mirex. Compounds with a + prefix indicates significant differences between species. (C) Log concentrations of Σ DDT and Σ PCBs. (D) Log concentrations of individual PCB congeners. Congeners with a + prefix indicates significant differences between species.

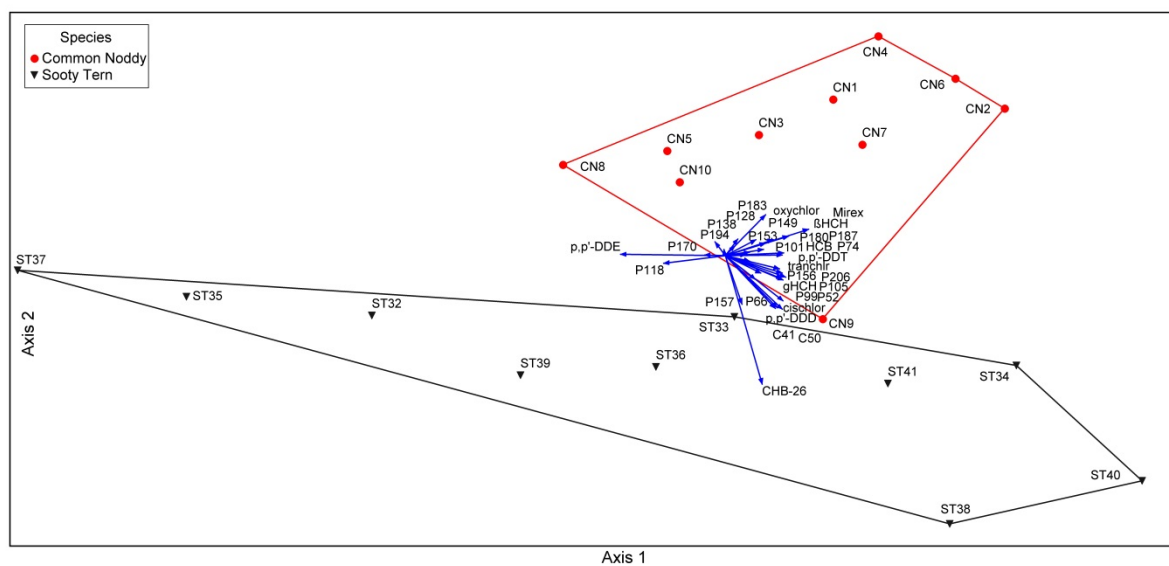


Fig. 3. NMS biplot of relativized compounds and individual eggs. Final stress was 6.46 for a two-dimensional solution. Axis 1 explained 86.4% of the ordination, and Axis 2 10.8%, for a cumulative of 97.2%.

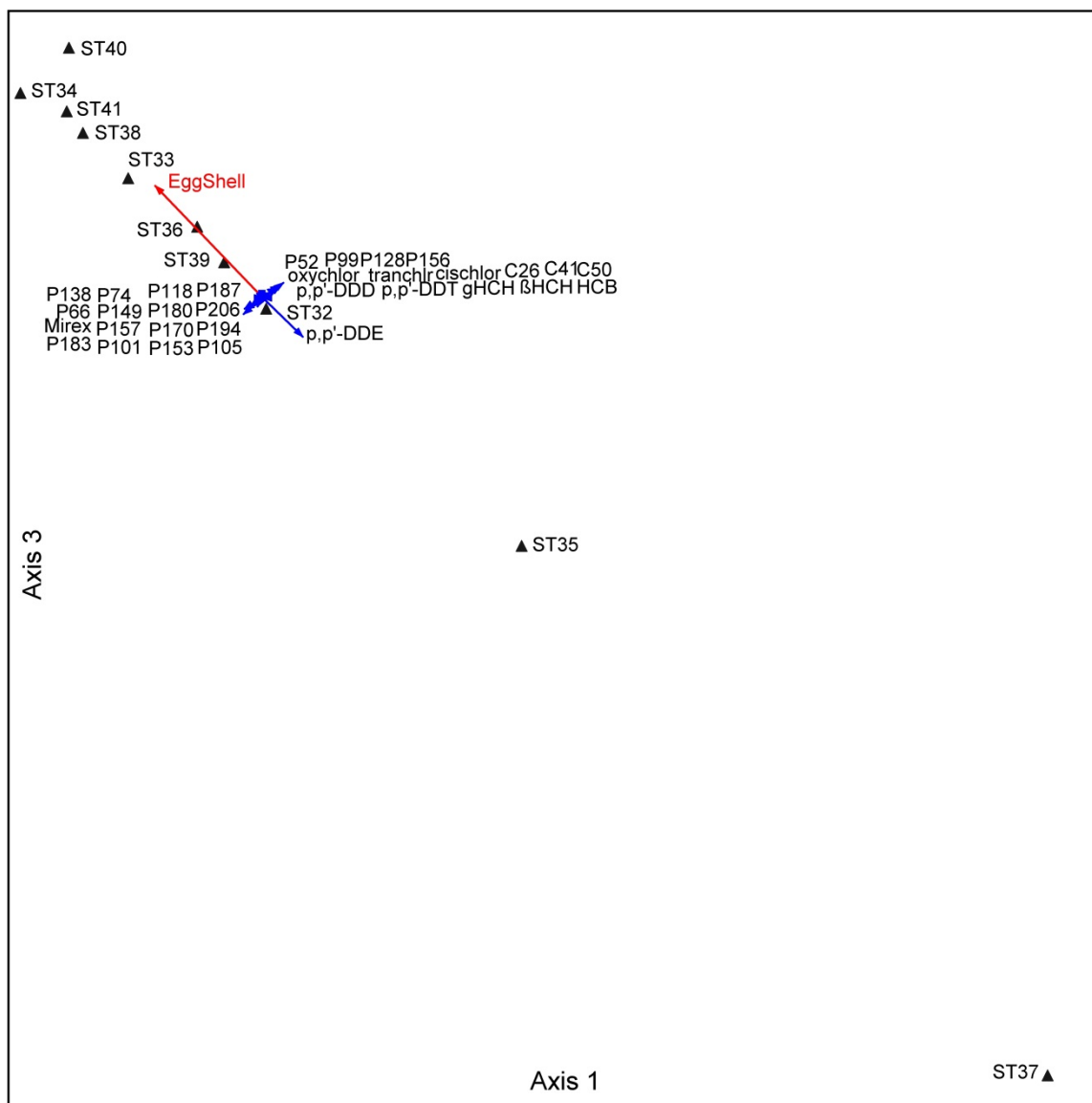


Fig. 4. PCA biplot of untransformed compounds in Sooty Tern eggs and eggshell thickness. Axis 1 explained 98.9% of the variance and the plot was rotated 50°.

Table 1. Concentrations of persistent compounds, eggshell thickness, and lipid content in Common Noddy (CN) and Sooty Tern (ST) eggs. Data are presented based on either wet mass or lipid mass. Indicator values are presented on the right.

	Wet mass ng/g												Lipid mass ng/g												Indicator values			
	Common Noddy (n=10)						Sooty Tern (n=10)						Common Noddy (n=10)						Sooty Tern (n=10)									
	Mean	Median	SD	Min	Max	Pos	Mean	Median	SD	Min	Max	Pos	Mean	Median	SD	Min	Max	Pos	Mean	Median	SD	Min	Max	Pos	Sp	IV	p	
HCB	0.39	0.35	0.15	0.30	0.80	10	0.41	0.40	0.079	0.30	0.54	10	3.6	3.1	1.2	2.4	6.1	10	3.0	3.0	0.54	2.2	3.9	10	ST	51	0.8332	
β-HCH	0.25	0.22	0.085	0.18	0.47	10	0.20	0.21	0.034	0.15	0.27	10	2.3	2.1	0.91	1.5	4.5	10	1.5	1.5	0.28	1.1	1.9	10	CN	54.8	0.1374	
γ-HCH	0.013	0.012	0.002	<LOQ	0.017	8	0.015	0.015	0.003	0.012	0.021	10	0.12	0.10	0.05	<LOQ	0.24	8	0.11	0.11	0.018	0.09	0.15	10	ST	60.1	0.0140	
Σ-HCH	0.26	0.23	0.086	0.18	0.48	10	0.22	0.22	0.036	0.16	0.28	10	2.4	2.2	0.95	1.5	4.7	10	1.6	1.6	0.29	1.2	2.0	10				
Oxychlordane	0.040	0.038	0.012	0.028	0.068	10	0.044	0.044	0.018	<LOQ	0.073	9	0.38	0.32	0.15	0.25	0.68	10	0.33	0.34	0.13	<LOQ	0.52	9	CN	47.8	0.9730	
Cis-chlordane	0.036	0.033	0.009	0.023	0.053	10	0.076	0.067	0.03	0.040	0.14	10	0.34	0.31	0.13	0.19	0.66	10	0.55	0.50	0.19	0.26	0.85	10	ST	67.9	0.0002	
Trans-nonachlor	0.022	0.019	0.007	<LOQ	0.033	8	0.025	0.024	0.008	0.015	0.039	10	0.18	0.17	0.04	<LOQ	0.27	8	0.18	0.18	0.05	0.11	0.28	10	ST	58.8	0.0944	
Σ-Chlordanes	0.094	0.089	0.021	0.068	0.13	10	0.15	0.14	0.03	0.11	0.20	10	0.48	0.46	0.08	0.37	0.66	10	0.73	0.71	0.20	0.45	1.0	10				
p,p'-DDE	1.67	1.32	0.97	0.79	3.64	10	2.80	1.98	2.58	0.91	9.32	10	15	13	6.1	7.0	26.0	10	21	15	21	6.5	74.2	10	ST	62.6	0.2260	
p,p'-DDD	0.052	0.030	0.067	0.013	0.24	10	0.070	0.072	0.041	0.017	0.14	10	0.41	0.27	0.40	0.20	1.5	10	0.50	0.52	0.29	0.13	0.84	10	ST	57.0	0.5469	
p,p'-DDT	0.21	0.22	0.10	0.080	0.39	10	0.18	0.18	0.036	0.136	0.23	10	1.8	1.7	0.60	0.68	2.5	10	1.3	1.4	0.28	0.91	1.9	10	CN	53.4	0.4701	
Σ-DDT	1.9	1.57	1.1	0.89	4.1	10	3.0	2.3	2.6	1.2	9.5	10	17	15	6.5	8.7	29	10	23	17	21	8.4	75.7	10				
Mirex	0.96	0.93	0.33	0.61	1.74	10	0.69	0.71	0.20	0.37	1.00	10	8.7	8.4	2.8	5.3	14	10	5.0	5.3	1.4	2.8	7.5	10	CN	58.2	0.0234	
PCB-52	0.070	0.062	0.023	<LOQ	0.12	8	0.068	0.065	0.025	0.034	0.11	10	0.63	0.59	0.17	<LOQ	0.90	8	0.50	0.46	0.20	0.27	0.88	10	ST	54.9	0.2651	
PCB-74	0.044	0.041	0.009	0.034	0.066	10	0.051	0.053	0.010	0.032	0.065	10	0.41	0.40	0.13	0.27	0.74	10	0.37	0.37	0.071	0.24	0.51	10	ST	53.7	0.1248	
PCB-66	0.026	0.026	0.004	<LOQ	0.033	7	0.030	0.028	0.009	0.017	0.045	10	0.26	0.20	0.11	<LOQ	0.46	7	0.22	0.20	0.066	0.12	0.32	10	ST	61.9	0.0342	
PCB-101	0.088	0.045	0.069	<LOQ	0.21	9	0.050	0.046	0.029	<LOQ	0.12	9	0.71	0.60	0.40	<LOQ	1.4	9	0.36	0.35	0.16	<LOQ	0.74	9	CN	57.1	0.2927	
PCB-99	0.030	0.030	0.009	0.021	0.051	10	0.046	0.042	0.028	0.019	0.12	10	0.28	0.27	0.10	0.13	0.44	10	0.33	0.32	0.17	0.14	0.68	10	ST	60.0	0.0860	
PCB-149	0.063	0.063	0.017	0.033	0.10	10	0.063	0.061	0.016	0.041	0.084	10	0.56	0.57	0.08	0.43	0.66	10	0.47	0.45	0.15	0.30	0.69	10	ST	50.2	0.9510	
PCB-118	0.047	0.034	0.022	0.028	0.085	10	0.088	0.063	0.068	0.039	0.27	10	0.42	0.41	0.18	0.21	0.71	10	0.66	0.48	0.55	0.27	2.2	10	ST	65.4	0.0250	
PCB-153	0.51	0.48	0.17	0.321	0.85	10	0.58	0.56	0.18	0.40	1.02	10	4.7	4.5	1.7	2.7	8.4	10	4.3	4.1	1.5	2.8	8.1	10	ST	53.1	0.4091	
PCB-105	0.072	0.069	0.023	0.049	0.12	10	0.12	0.12	0.021	0.095	0.16	10	0.68	0.61	0.30	0.31	1.4	10	0.89	0.90	0.18	0.63	1.3	10	ST	62.9	0.0004	
PCB-138	0.23	0.20	0.088	0.14	0.43	10	0.26	0.24	0.13	0.16	0.59	10	2.1	2.0	0.72	1.2	3.4	10	2.0	1.7	1.1	1.1	4.7	10	ST	53.4	0.5377	
PCB-187	0.12	0.12	0.040	0.084	0.22	10	0.13	0.11	0.033	0.10	0.21	10	1.1	1.2	0.32	0.72	1.8	10	0.93	0.85	0.30	0.64	1.7	10	ST	50.1	0.9898	
PCB-183	0.049	0.046	0.017	0.030	0.09	10	0.054	0.046	0.023	0.037	0.11	10	0.45	0.43	0.15	0.28	0.75	10	0.40	0.35	0.20	0.22	0.89	10	ST	52.3	0.6291	
PCB-128	0.10	0.09	0.033	0.046	0.16	10	0.082	0.084	0.031	0.043	0.15	10	1.00	0.75	0.68	0.3	2.7	10	0.62	0.64	0.26	0.32	1.2	10	CN	54.2	0.3101	
PCB-156	0.087	0.074	0.030	0.063	0.16	10	0.12	0.12	0.042	0.061	0.20	10	0.80	0.70	0.25	0.61	1.4	10	0.88	0.84	0.32	0.41	1.4	10	ST	57.9	0.0614	
PCB-157	0.047	0.039	0.024	0.027	0.11	10	0.10	0.070	0.074	0.025	0.27	10	0.41	0.36	0.15	0.24	0.69	10	0.68	0.52	0.44	0.18	1.6	10	ST	67.5	0.0338	
PCB-180	0.53	0.50	0.20	0.34	0.90	10	0.59	0.58	0.12	0.42	0.87	10	4.8	4.8	1.4	2.9	6.9	10	4.3	4.3	1.1	3.0	6.9	10	ST	52.5	0.4575	
PCB-170	0.067	0.057	0.030	0.032	0.12	10	0.10	0.081	0.047	0.051	0.21	10	0.61	0.60	0.25	0.27	0.98	10	0.72	0.60	0.39	0.36	1.7	10	ST	59.3	0.0938	
PCB-194	0.035	0.034	0.017	0.014	0.065	10	0.050	0.044	0.020	0.025	0.09	10	0.31	0.29	0.12	0.14	0.55	10	0.37	0.31	0.16	0.18	0.74	10	ST	58.6	0.0902	
PCB-206	0.036	0.021	0.042	<LOQ	0.15	9	0.026	0.025	0.009	<LOQ	0.045	9	0.29	0.26	0.25	<LOQ	0.92	9	0.20	0.19	0.071	<LOQ	0.33	9	CN	52.0	0.6971	
Σ-PCB	2.2	2.1	0.72	1.4	3.6	10	2.6	2.4	0.67	1.9	4.3	10	20	19	6.4	12	33	10	19	18	5.9	14	34	10				
CHB-26	0.075	0.075	0.011	<LOQ	0.083	2	0.21	0.20	0.042	0.14	0.28	10	0.48	0.48	0.07	0.43	0.53	2	1.5	1.6	0.28	0.95	2.0	10	ST	93.2	0.0002	
CHB-41	0.049	0.041	0.023	<LOQ	0.10	8	0.072	0.068	0.016	0.055	0.11	10	0.38	0.34	0.12	0.29	0.65	8	0.53	0.52	0.12	0.36	0.76	10	ST	65.0	0.0046	
CHB-44						0	0.078					1						0	0.56					1				
CHB-50	0.22	0.19	0.10	0.14	0.41	10	0.50	0.50	0.11	0.32	0.69	10	2.0	1.8	0.54	1.3	3.0	10	3.7	3.8	0.79	2.1	5.0	10	ST	69.1	0.0004	
CHB-62						0	0.27					1						0	1.9					1				
Σ-CHB	0.26	0.23	0.12	0.141	0.51	10	0.61	0.57	0.22	0.37	1.1	10	2.4	2.1	0.73	1.6	3.8	10	5.9	6.0	1.8	3.4	10	10				
BDE-47	0.06					Pool	0.04					Pool	0.5					Pool	0.4						Pool			
BDE-100	0.02					Pool	0.03					Pool	0.2					Pool	0.3						Pool			
Σ-BDE	0.08					Pool	0.07					Pool	0.7					Pool	0.7						Pool			
Eggshell thickness mm	0.338	0.325	0.046	0.280	0.440		0.291	0.283	0.025	0.258	0.330	Lipid %	11.4	11.6	3.2	5.0	15.7		13.7	13.4	1.30	11.9	16.8					

Table 2. Reports with comparable data.

	n	Locality	Collected	HCB	ΣHCH	ΣChlord	p,p'-DDE	p,p'-DDT	ΣDDT	Mirex	PCB-153	ΣPCB	PCB ^a	BDE-47	ΣBDE	PBDE ^a	Reference
Common Noddy	10	Rodrigues	2008	0.39	0.26	0.09	1.7	0.21	1.9	0.96	0.51	2.2	20	0.06	0.08	9	this study
Sooty Tern	10	Rodrigues	2008	0.41	0.22	0.15	2.8	0.18	3.1	0.69	0.58	2.6	20	0.02	0.07	9	this study
Sooty Tern		Hawaii	1980						15								Ohlendorf & Harrison (1986)
Sooty Tern		Midway	1980						12								Ohlendorf & Harrison (1986)
Roseate Tern	3	South Africa (St Croix)	1979				40					340					de Kock and Randall (1984)
Common Tern	10	Netherlands (Griend)	2000	4.8	3.2	0.6	46	0.3	47.5		215	864	51				Becker et al. (2001)
Common Tern	10	USA (Lake Michigan)										10 963					Ward et al. (2010)
Common Tern ^{b,c}	9	China (Yellow River)	2008		24				302			17			6.3		Gao et al. (2009)
Black Tern	3	USA (Lake Michigan)	1995			9			86			58					Wayland et al. (2000)
Caspian Tern	3	USA (Lake Michigan)	1996			36			1781			786					Wayland et al. (2000)
Arctic Tern ^b	6	Iceland	2002/4	5.7	0.65	0.82 ^d	28.2				16.1	56	12				Jörundsdóttir et al. (2010)
Adélie Penguin	6	Antarctica	1995/96	18.7	0.59	1.83	20.7	0.93	23		2.7	25	56	0.17	0.29	7	Corselini et al. (2006)
Adélie Penguin	37	Antarctica (East)	2003/5	4.7	na		4.3					22	54				Corselini et al. (2011)
Adélie Penguin	9	Antarctica (West)	not clear	7.6	1.8		19	0.49				12	54				Corselini et al. (2011)
Adélie Penguin	27	Antarctica (Ross Sea)	1988/90	5.6			6.3		7.1			9					Court et al. (1997)
Gentoo Penguin	9	Antarctica	2004/5	3.7					15			5	43				Schiavone et al. (2009)
Cape Gannet	4	South Africa (St Croix)	1979				40					50					de Kock and Randall (1984)
Brown Booby	6	Mexico	2006				53										Mellink et al. (2009)
African Penguin	21	South Africa (St Croix)	1979				60					240					de Kock and Randall (1984)
Atlantic Puffin ^b	5	Norway (Røst)	2003	57.5	2.03	52.5	123.0	28.2	151	4.07	89.5	347	20				Helgason et al. (2008)
Atlantic Puffin	5	Norway (North)	2003											5	7	8	Helgason et al. (2009)
Black-legged Kittiwake ^b	6	Norway (Røst)	2003	47.8	1.95	59.3	145.9	7.19	153	8.09	225	778	20				Helgason et al. (2008)
Black-legged Kittiwake	5	Norway (North)	2003											18.7	26.1	8	Helgason et al. (2009)
Black-legged Kittiwake	12	Canada (Nunavut)	2003	15.7	7.7	45.3	43.5		43.5	2.1		177	59				Braune et al. (2001)
Herring Gull	10	Sweden	2008					19.5				205		2.58	8.3	11	Carlsson et al. (2011)
Herring Gull ^b	6	Norway (Røst)	2003	31.7	1.55	116	416.5	14.4	431	6.20	328	1099	20				Helgason et al. (2008)
Herring Gull	5	Norway, North	2003										20	37.9	60.5	8	Helgason et al. (2009)
Kelp Gull	1	South Africa (Veldrif)	2004	5.2	1.3	0.77	88	<LOQ	88	0.81	30	100	20	0.5	0.9	8	Polder et al. (2008b)
Brown-hooded Gull	7	Chile	1996	6.7	0.1			137				16	7				Cifuentes et al. (2003)
Northern Fulmar	15	Canada (Nunavut)	2003	13.2	5.7	112	112	10.9	124	3.7		167	59				Braune et al. (2001)
South Polar Skua	46	Antarctica (Ross Sea)	1988/91	21.1			96.9		99.7			214					Court et al. (1997)
Thick-billed Murre	15	Canada (Nunavut)	2003	27.6	11.2	30.5	103		103	1.4		120	59				Braune et al. (2001)
Common Eider	10	Sweden	2008					20.1				108		0.28	0.6	11	Carlsson et al. (2011)
Oystercatcher	10	Netherlands (Griend)	2000	2.5	1.7	1.5	21.7	0.3	22.5		168.2	623	51				Becker et al. (2001)
African black Oystercatcher	5	South Africa (St Croix)	1979				30					630					de Kock and Randall (1984)

^a Number of congeners used in quantification

^b Calculated

^c Median

^d *Trans*-nonachlor only