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Long term trends in mercury and PCB congener concentrations in gannet (*Morus bassanus*) eggs in Britain.

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Abstract

Gannet (*Morus bassanus*) eggs from Bass Rock (North Sea) and Ailsa Craig (eastern Atlantic) were monitored for PCB congeners (1990-2004) and total mercury (1974-2004). Congener profiles for both colonies were dominated by PCBs 153, 138, 180, 118 and 170. All declined in concentration at Ailsa Craig but some (153, 170, 180) remained stable or increased slightly at Bass Rock. Egg congener concentrations at Bass Rock were typically 10-fold higher than at Ailsa Craig by 2002, and Principal Component Analysis indicated that colony differences were driven by the dominant congeners. Egg mercury concentrations were significantly lower at Bass Rock than Ailsa Craig and temporal trends differed, there being a significant decline at Ailsa Craig but a marginal increase at Bass Rock. Our results suggest there may be differences in contamination between the eastern Atlantic and North Sea and/or there are colony differences in prey selection and associated contaminant loads.

Keywords- Gannets, PCBs, mercury, Ailsa Craig, Bass Rock and temporal trends.

Capsule: *Monitoring of PCBs and Hg in gannet eggs reveals contrasting temporal patterns between colonies on the eastern Atlantic and North Sea coasts of Britain*.

1. Introduction

Polychlorinated biphenyls (PCBs) were produced commercially between the 1930s and 1970s in most industrialised countries and were widely used in industry and in

commercial products (Rice et al., 2003 and references therein). They are highly persistent and their use in open sources, release from damaged and degraded closed sources, and subsequent atmospheric transport has resulted in contamination of foodchains throughout the world. Similarly, mercury (Hg) frequently occurs in various environmental compartments, partly due to its natural occurrence but mainly because of widespread contamination from fossil fuel combustion, industry (such as chlor-alkali plants) and agriculture (Navarro et al., 1993; Carpi, 1997). PCBs and Hg are toxic, persistent, and are only slowly metabolised by vertebrates (Walker and Livingstone, 1992). Consequently, these contaminants progressively bioaccumulate in lipid rich organs in biota (Moriarty, 1990; Furness, 1993) and high levels of exposure have been related to various adverse effects (Giesy et al., 1994; Herzke et al., 2002), particularly on reproduction (Moriarty, 1990; Furness, 1993; Walker, 1994; Grassman et al., 1998).

Coastal birds are an important part of the marine and coastal ecosystems (Furness and Monaghan, 1987). They tend to occupy high trophic levels and, as a result, bioacummulate a wide range of contaminants via their diet. Because of this, contamination levels in their body tissues and eggs have been used as biomonitors of exposure to persistent pollutants (Fox et al., 1991; Bishop et al., 1995). Coastal birds are good bioindicators because they reveal current environmental exposure and respond relatively rapidly to contamination events (Lewis and Furness, 1991). Eggs are considered a particularly favourable matrix for monitoring because the contents are highly consistent in composition and reflect the exposure of a consistent part of the population (breeding females). Egg production results in the excretion of environmental contaminants, particularly hydrophilic pollutants, by females (Burger, 1994; Gochfeld and Burger, 1998; Connell et al., 2003; Lam et al., 2004) and several studies have shown that seabird eggs are

indicative of local contamination, reflecting pollutant intake by females foraging close to the colony prior to laying (Walker, 1994; Becker et al., 1998). Previous winter intake and age-dependent bioaccumulation are only minor contributors to the overall contaminant concentrations in eggs (Sell et al., 1974; Kambamanolidimou et al., 1991).

Air and soil concentrations of PCBs and Hg have fallen since PCB production ceased and emission levels of Hg in Europe decreased (Lead et al., 1997; Fowler et al., 2006). Although, PCB and Hg contaminant concentrations are reported to have fallen in some terrestrial and aquatic biota during the 1970s and 1980s, concentrations after 1990 appear to have levelled off or may even be increasing (Cifuentes et al., 2003; Simpson, 2007; Walker et al., 2007). One of the long term biomonitors of contaminant levels in marine systems around Britain has been gannet (*Morus bassanus*) eggs (Newton et al., 1990; Alcock et al., 2002; Shore et al., 2005). A previous study on eggs collected up until 1987 from colonies off both the Atlantic and North Sea coasts of Britain revealed a decline in Hg concentrations in two colonies (including Ailsa Craig) but a significant increase in four colonies (including Bass Rock) (Newton et al., 1990). This study also recorded a significant decrease in total PCB concentrations in eggs from four colonies (including Bass Rock and Ailsa Craig) and an increase for two colonies, but temporal trends of individual PCB congeners were not reported (Newton et al., 1990). A subsequent study of gannet eggs from a colony from the Atlantic coast of Britain found that there typically had been an order of magnitude decrease in PCB congeners between 1977 and 1998; PCB congener concentrations in eggs from North Sea colonies were not reported (Alcock et al., 2002).

The aim of our present study was to examine how PCB congener and Hg concentrations in the eggs of gannets off the coast of Britain have changed over time. We have used long-term data that included new measurements of PCB congener and Hg

concentrations that cover the period 1987-2004. Furthermore, we aimed to determine if PCB congener profiles and temporal trends of major PCB congeners differed significantly between gannet eggs from the eastern Atlantic and North Sea coasts of Britain; such comparisons have not previously been carried out.

2. Materials and Methods

2.1. Egg collection

Gannet eggs were collected in two colonies, Ailsa Craig and Bass Rock. Ailsa Craig is a granite island situated in the outer part of the Firth of Clyde on the west coast of Scotland (Figure 1). It is a Site of Special Scientific Interest, being a nesting ground for seabirds, especially gannets, and it has been estimated to contain around 9 % of the world's gannet population during the breeding season (Mitchell et al., 2004). Bass Rock is an island in the outer part of the Firth of Forth on the east coast of Scotland in the North Sea (Figure 1). It is also a Site of Special Scientific Interest due to its gannet colony which holds approximately 11% of the world population of gannets during the breeding season (Mitchell et al., 2004). Breeding gannets feed locally but also forage up to 150 Km away from the colonies (Tasker et al., 1985; Hamer et al., 2001). However, gannets do not fly overland to feed and so the foraging areas of birds from Bass Rock and Ailsa Craig do not overlap; gannets from Bass Rock forage in the North Sea whereas birds from Ailsa Craig feed in waters to the west of Britain.

The eggs used in this study were collected under licence as part of the long-term monitoring programme of the Predatory Bird Monitoring Scheme (PBMS;

http://pbms.ceh.ac.uk). Eight to twelve fresh eggs were taken during laying or the early incubation period from separate nests from each colony. Sampling was carried out approximately every two years between 1971 and 2004. The length, breadth and weight of each egg were measured. The contents were collected either by "blowing" (removed through a small hole or opening in the egg) or, in more recent years, by cracking the eggs open. The contents were homogenised, kept at -20^{0}C for 6 to 9 months after collection, and then analysed.

2.2. Chemical analyses

A suite of 25 PCB congeners (8, 18, 28, 29, 31, 52, 77, 101, 105, 114, 118, 123, 126, 128, 138, 149, 153, 156, 157, 167, 169, 170, 180, 189 and 209) were determined in gannet eggs using gas chromatography with electron capture detection (GC-ECD). These analyses were only conducted from 1990 onwards and data are not available for eggs collected before this date. A sub- sample of each egg (1-2 g) was thawed, weighed accurately, ground with sand, dried with anhydrous sodium sulphate, and cold extracted in 50 ml of 1:1 acetone:hexane mixture. Half of the extract was evaporated to zero volume, the lipid content determined gravimetrically, and the extract was then re-dissolved in hexane. Lipids were removed from the extract using an alumina glass column packed with 1g of pre-treated alumina (4 h at 800°C) that had been deactivated with 5 % deionised water (w/w). Prior to analysis, dichlobenil was added as an internal standard. A 4 µl aliquot of each extract was injected into the gas chromtograph (Agilent, Wokingham, UK) using a splitless injector, and a 50 m HT8 column (0.22 mm internal diameter and 0.25 µm film thickness, SGE Milton Keynes, UK). The injector temperature was set at 250°C and the

carrier gas was helium (2.0 ml min^{-1}). The temperature programme was: isothermal at 50°C for 2 min, 45°C min^{-1} to 200°C, 1.5°C min^{-1} to 240°C, 2°C min^{-1} to 285°C, 50°C min^{-1} to 325°C and isothermal at 325°C for 10 min. The detector temperature was set at 335°C. Residues were quantified using the internal standard method and also using calibration curves of the standard PCBs (Greyhound Ltd, Birkenhead, UK).

Total Hg concentrations were measured in approximately 1 g sub-samples from each homogeneised egg. Samples were dried to constant weight at 80°C for 24 hours, solubilised at room temperature overnight in 2ml of (Analar) nitric acid, then heated at 90°C for 20 minutes followed by 120°C for 1 hour. To further digest the organic matter, 0.5 ml of 30 % hydrogen peroxide were added to the sample which was then heated at 120°C for 15 minutes. Samples were diluted with double-deionised water to known volume and a 10 % acid strength. Cold vapour atomic absorption spectrophotometry using 10% tin chloride in 20 % hydrochloric acid as a reducing agent, nitrogen as a carrier gas, and an absorbance wavelength of 253.7 nm was used to determine total Hg concentrations.

For quality control and assurance purposes, a sample blank, a sample of an uncontaminated chicken egg and second sample of chicken egg spiked with a known concentration of PCB congeners or Hg were analysed with each batch of samples. Recovery values determined from the spike samples varied between 75% and 110% for the PCB congeners and between 70 % and 114 % for Hg. Limits of detection (LoD), determined from the calibration curve and based on the mean weight of the egg contents that were analysed, varied between 0.029 and 0.157 ng g^{-1} wet weight (wet wt.) for PCB congeners and between 0.79 and 2.09 µg/g dry weight (dry wt.) for different years for Hg; Hg determinations in eggs were below the LoD. Recoveries and limits of detection did not

vary systematically over time for PCB congeners or Hg and data were not recovery corrected.

2.3. Statistical analysis

Individual PCB congeners and Hg concentrations are presented on a wet wt. and dry wt. basis, respectively. Wet weight concentrations were not corrected for fresh weight (Stickel et al., 1973) as all eggs were fresh on collection. For statistical purposes, non-detected concentrations were assigned a value of half the limit of detection. Residue data had skewed distributions and so average annual concentrations are presented as geometric means for each contaminant for each year in which eggs were analysed. These geometric mean values were the data used in the statistical models used to test the significance of variation in concentrations over time and between colonies.

The variation over time in PCB congener and Hg concentrations in eggs were analysed separately for each colony by linear regression. To determine if overall absolute values and trends over time in contaminant concentrations varied significantly between Ailsa Craig and Bass Rock, data for both colonies were analysed together using a general linear model (GLM) in which colony was a factor and year a covariate. Inspection of the variances in the contaminant concentration data and the residuals from the regressions and GLMs confirmed that the underlying assumptions of the statistical models were not violated. We used Principal Component Analysis (PCA, Umetrics, Simca-P) to discriminate patterns of spatial variation in the PCB congeners. PCA was performed using all individual congener concentrations as input variables. The PCA is not sensitive to covariance and uneven variance among the variables and does not require the analytical

error in the predictor variables to be negligible. Before analysis, all variables were mean centered and scaled to unit variance.

3. Results

3.1. PCB congener profile

Some PCB congeners (8, 18, 28, 29, 77, 123, 126, 157, 189, 209) had annual mean concentrations that were always or mostly below detection limits and are not reported further. The LoDs of these congeners were similar (within the same range) to those that had higher mean concentrations. The remaining congeners (31, 52, 101, 105, 114, 118, 128, 138, 149, 153, 156, 167, 169, 170 and 180) were detected more frequently and the geometric mean and range of their concentrations are given in Table 1. These congeners were detected in 15.4 to 100% of the samples (Table 1). Four congeners (PCB 31, 52, 101 and 169) were detected in <50% of the samples and five congeners (118, 138, 153, 170 and 180) were detected in > 95% of the samples.

PCBs in the eggs from both colonies were dominated by the same five congeners (Figure 2 and Table 1). PCB 153 was the most dominant, occurring in concentrations of up to 0.58 µg g^{1} wet wt. and comprising approximately 30-40% of the congener summed total PCB concentration. PCBs 138 and 180 between them comprised another 30-40% of the congener sum total PCBs and, like PCB 153, occurred in concentrations around 10 times higher than those of other congeners. PCBs 118 and 170 together comprised a further 15-20% of the total PCB concentration and were present in concentrations up to 0.12 µg g^{-1} wet wt. A sixth congener, PCB 128, contributed less on average to the sum total PCB

concentration than the other dominant congeners (Figure 2), but also occurred in concentrations of up to 0.12 µg g^{-1} wet wt. when present. Congeners 31, 52, 101, 105, 114, 149, 156, 167 and 169 were present in (geometric mean) concentrations of up to 0.05 µg g^{-1} wet wt.

3.2. Temporal trends in PCBs

Although the dominant congeners in eggs at Ailsa Craig and Bass Rock were similar, temporal trends in congener concentrations were not always consistent between the two colonies. Concentrations of four congeners (PCBs 138, 128, 118 and 170) declined significantly over time in eggs from Ailsa Craig (R^2 = 0.674, $F_{(1,4)}$ = 9.82, P= 0.05 in all cases; Figure 3). At Bass Rock, PCBs 138 and 128 likewise decreased significantly during the monitoring period (R^2 = 0.653 $F_{(1,4)}$ = 8.52, P=0.05; Figure 3) and rates of decline did not differ significantly from those at Ailsa Craig ($F_{(1,,8)}$ = 2.69, P = 0.05). The half-lives for these congeners, determined using linear regression analyses for data for the two colonies combined, were 4.7 years (PCB 138) and 2.5 years (PCB 128). Concentrations of PCB 118 in Bass Rock eggs also decreased (Figure 3). Although this decline was not statistically significant (R^2 = 0.141, $F_{(1,5)}$ = 0.84, P = 0.402), it did not differ ($F_{(1,10)}$ = 0.18, P = 0.684) from the significant decline observed at Ailsa Craig. The overall half-life for PCB 118 (data for both colonies combined) was 11.7 years.

In contrast, the temporal trend for PCB 170 did differ significantly between the two colonies ($F_{1,10}$=4.96, P=0.05). In eggs from Ailsa Craig, PCB 170 decreased with a half-life of 4.4 years whereas, in Bass Rock, the concentration of this congener appeared to be increasing around 14 % (Figure 3), although not significantly (R^2 = 0.06, $F_{(1,5)}$ = 0.32, P = 0.598). The lack of any decline for PCB 170 in Bass Rock eggs precluded the calculation of

a half-life for this congener at this location. The concentrations of the remaining dominant congeners (153 and 180), appeared to remain constant or decline marginally (although not significantly; $R^2 = 0.313$, $F_{(1,5)} = 2.27$, $P>0.05$ in all cases) at both colonies. The half-lives for PCBs 153 and 180, calculated using data pooled for the two colonies, were 47.8 and 42.4 years, respectively.

Overall, differences between colonies in temporal trends were reflected by the fact that the concentrations of most congeners were typically at most two-fold higher in Bass Rock eggs than in Ailsa Craig eggs in 1992, but were typically 10-fold higher by 2002 (Table 1).

3.3. Spatial variability in PCBs

Most of the spatial variation in PCB congeners was explained by the first two principal components in our PCA model (PC1: 83.0%; PC2: 12.4%). There was a certain degree of separation along PC1, with the majority of eggs from Ailsa Craig grouped on the negative side and most of the Bass Rock eggs spreading across the negative and positive sides (Figure 4A). The more evident clustering of the Ailsa Craig data suggests a higher similarity in PCB concentrations in eggs from this colony compared with those from Bass Rock. PC2 also highlighted some separation between colonies, with most of Ailsa Craig's eggs having a positive score and Bass Rock eggs having negative and positive scores. The PCA loadings showed that the separation between Ailsa Craig and Bass Rock was driven mainly by the dominant congeners, with the main cluster of data for Ailsa Craig eggs appearing to be less influenced by these dominant congeners (Figure 4B). Samples with positive PC1 were highly influenced by PCBs 138, 153 and 180 and had high concentration of these PCBs, which included some eggs from Bass Rock and from Ailsa Craig. Samples

with positive scores in PC2 were mostly influenced by congener 138 and those with negative loadings were dominated mostly by congeners 180, 153, 118 and 170 (Figure 4B). The dominant congeners driving the observed separation pattern between the two colonies were also some of the heavier congeners (higher level of chlorination) that were analysed.

3.4.TEQs

The toxicity of individual congeners was assessed using the toxic equivalent approach (Van den Berg et al., 1998). We determined the concentrations of the TCDD toxic equivalents (TEQs) for each individual egg analysed between the years of 1998 to 2004. TEQs for eggs from previous years were not calculated because the suite of relevant congeners that were analysed was incomplete. The minimum and maximum concentrations of the TCDD toxic equivalents (TEQs) in gannet eggs from Ailsa Craig varied between 0.079 and 166.5 pg g^{-1} wet wt. and those from Bass Rock varied between 1.99 and 46.7 pg g^{-1} wet wt..

3.5. Hg

Geometric mean Hg concentrations varied between 1.5 and 5.1 µg g^{-1} dry wt. in eggs from Ailsa Craig, with the highest mean concentration in 1971 and the lowest in 1997. Hg concentrations in eggs from Bass Rock varied between 0.8 µg g^{-1} dry wt. (in 1977) and 2.7 µg g^{-1} dry wt. (1981) and, over the whole period of monitoring, were significantly lower than in eggs from Ailsa Craig ($F_{1,33}$=24.1, P<0.005). The temporal trends for Hg differed for the two colonies. Hg concentrations in Ailsa Craig eggs declined significantly from the 1970s to 2004 (R^2=0.661 $F_{(1,17)}$= 33.2, P<0.001), whereas Hg concentrations in eggs from Bass Rock increased marginally over the same period, although the increase was not

statistically significant (R^2=0.073 $F_{(1,16)}$ = 1.27, P>0.05; Figure 5). The difference in temporal trends between the two cobnies was highly significant ($F_{(1,,33)}$ =23.8, P<0.001).

4. Discussion

4.1. PCB congener profile

The PCB congeners profiles (Figure 2) suggest a marked similarity in contamination in eggs between Ailsa Craig and Bass Rock. In both colonies, PCBs 153, 138 and 180 predominated and accounted for more than 70% of the sum of total congeners. The same dominant congeners were detected in gannet eggs from Ailsa Craig by Alcock *et al.* (2002), and other studies have shown that these same congeners account for most (70-90%) of the PCBs in the eggs of other raptors (Herzke et al., 2002). PCB contamination in the eggs of white-tailed sea eagles (*Haliaeetus albicilla*), another marine fish eating bird, is also dominated by PCB 138 (Herzke et al., 2002). The similarity in egg PCB congener profiles across species with different feeding habits and trophic status most probably reflect selective bioaccumulation of each congener across several trophic levels (Norstrom, 1988; Borlakoglu et al., 1990a). Studies that examined congener patterns in birds relative to their prey (Braune and Norstrom, 1989; Guruge and Tanabe, 1997) or to the original congener composition in Aroclor source mixtures (Borlakoglu et al., 1990b) revealed that PCBs can be broadly classified into persistent or readily degraded congener groups. The more rapidly eliminated compounds possess unsubstituted vicinal *ortho-meta* positions and the most persistent lack unsubstituted carbons in the *meta-para* positions (Walker, 2001). In our study all dominant congeners belonged to the latter group. Their recalcitrant nature is

therefore, likely to account for the observed congener profile and the similarity between colonies.

4.2. Spatio-temporal trends in PCB congeners and Hg

Some individual PCB congeners in eggs from Ailsa Craig and Bass Rock were declining, continuing a trend observed since the 1970s (Alcock et al., 2002). An overall decrease in PCB residues in gannet eggs was expected as rapid declines in organochlorinated compounds were observed in the 1970s and 1980s following their ban or reduced usage in the USA, Western Europe and Canada (Newton et al., 1990; Newton et al., 1999). However, while the calculated half-life of congener 138 for Ailsa Craig (3.2 years; this study) was approximately two-fold lower than that estimated for the period 1977-1998 (Alcock et al., 2002), the half-life of congener 118 was similar in both studies (5 years in this study and 5.9 years in Alcock et al., 2002). In contrast, the half-life of congener 153, estimated at 10.1 years by Alcock et al. (2002), almost doubled (18 years). This increase in half-lives is not unique to gannets; other studies have also shown that the rate of decrease of PCBs has slowed over the last decade (Becker and Cifuentes, 2004; Braune et al., 2005). Variance in the temporal trends of half-lives for congeners may reflect differences in initial environmental inputs and remobilisation processes.

The temporal trends for some congeners varied spatially. The most marked was PCB 170, concentrations declining with a half-life of 4.4 years in eggs from Ailsa Craig but increasing marginally in eggs from Bass Rock. The differences between colonies in temporal trends for individual congeners were reflected by PCA, the more homogenous pattern of decline across congeners in Ailsa Craig eggs being indicated by tighter clustering

in the PCA compared with eggs from Bass Rock (Figure 4A). The dominant congeners, which were some of the heavier congeners studied, appear to be the main drivers of the spatial distinction between colonies. The eggs from Bass Rock with higher concentrations of these congeners had positive scores in PC1 and the Ailsa Craig eggs appeared to be less influenced by these congeners (Figure 4B). The presence of higher concentrations of recalcitrant congeners in Bass Rock eggs suggest that gannets in this colony could be affected by these contaminants for longer than those from Ailsa Craig. The lack of degradation of these congeners can account for their relatively high concentrations, but cannot explain spatial differences in temporal trends.

There was also spatial variation in both the absolute concentrations and temporal trends for Hg residues in gannet eggs. Mercury concentrations were higher in eggs from Ailsa Craig (west coast) than in those from Bass Rock (east coast), as found in earlier years (Newton et al., 1990). An east-west gradient was observed in Hg concentrations in common loon (*Gavia immer*) eggs across North America that was consistent with Hg deposition patterns (Evers et al., 2003). However, air Hg concentrations are currently higher in eastern that western Scotland (Fowler et al., 2006), the opposite of the west-east difference in Hg that we observed in the gannet eggs. Furthermore, temporal trends in egg Hg also varied between Ailsa Craig and Bass Rock with a long-term decline at Ailsa Craig but a slight increase, though not statistically significant, at Bass Rock. A decline in Hg levels in eggs, as seen at Ailsa Craig, may have been expected given there has been a decrease in the emission levels of Hg in Western Europe. However this has not occurred in gannet eggs at Bass Rock (this study) nor at some other colonies off the British coast (Newton et al., 1990). It is notable that mercury concentrations in the eggs of thick-billed

murres (*Uria lomvia*) and Northen fulmars (*Fulmarus glacialis*) have also increased significantly between 1975 and 1998 (Braune et al., 2002; Lindberg et al., 2002).

Various factors may account for the spatio-temporal differences in egg PCB and Hg concentrations between Ailsa Craig and Bass Rock. Gannets forage within 150 km of their colonies in the breeding season (Tasker et al., 1985; Hamer et al., 2001) and so pollutants in gannet eggs from Ailsa Craig and Bass Rock would be expected to reflect local environmental contamination (Thyen et al., 2000; Braune et al., 2001) of the eastern Atlantic/Irish Sea and the North Sea, respectively. Our results indicate that contaminants in gannet prey that are then assimilated by females and mobilised into their eggs differ between these two areas. Contamination can occur via riverine inputs, direct discharges and atmospheric deposition, the first two being largely coastal processes. Sediments from sites closer to urban/industrial storm runoff discharges have been found to have heavier PCBs than those far away from the discharge site (Becker et al., 2001) and differences between colonies in absolute concentrations and temporal trends may reflect differences between areas in contaminant inputs from coastal sources. However, redistribution from sinks, such as contaminated sediments, is also potentially a major source of higher chlorinated PCB and of re-suspended inorganic and methylated Hg.

Although gannets feed mainly on herring, mackerel and to a lesser extent sandeels (Hamer et al., 2001), they also exploit locally abundant fish (Martin, 1989). Differences in exposure pathways and trophic level between fish species are likely to affect their contaminant uptake and the subsequent assimilation of contaminants by predators. Thus, inter-colony variation in feeding preferences could account for differences in contaminant levels and trends between eggs from Ailsa Craig and Bass Rock. Furthermore, a study of Bass Rock gannets in 2002-2003 found that discarded demersal fish were an important

component of the diet (Kakela et al., 2007). If gannets feed substantially on discard from commercial trawlers (Hamer et al., 2001), their exposure to contaminants via diet may not necessarily reflect local contamination. Furthermore, demersal fish tend to have higher concentrations of Hg than pelagic fish (Arcos et al., 2002) suggesting that gannets feeding in demersal fish would have a higher intake of Hg than those feeding on pelagic fish. Short and long term spatio-temporal variation in egg contaminant concentrations could in part signal temporal variation and the importance of discard in the diet.

4.4. Potential toxicity of PCBs and Hg in gannet eggs

No Observable Effect Concentrations (NOECs) for toxic equivalents (TEQs) of between 1.5 and 200 pg TEQs g^{-1} wet wt. and Lowest Observed Effect Concentrations (LOECs) of between 10 and 2200 pg g^{-1} wet wt. have been established in the eggs of various experimental and wild birds (AMAP, 1998). The LD_{50} for embryo mortality in white leghorn chickens, one of the more sensitive species, has been shown to range between 115 and 147 pg g^{-1} TEQ wet wt (AMAP, 1998). Thus, there is considerable overlap between NOEC, LOEC and LD_{50} TEQ values which, in part, reflects species variation in sensitivity. The (geometric mean) TEQ concentration in the gannet eggs in the current study in both colonies were within the range of the NOEC values or were at the lower end of the established LOECs. Therefore based on the TEQ concentrations, it appears likely that the levels of PCBs in gannet eggs are not toxicologically significant, and should not affect reproduction.

Hg concentrations ranging between 1.5 and 18 µg g^{-1} dry wt. in bird eggs have been associated with adverse effects, including decrease in egg weight, embryo malformations,

lower hatchability, decrease chick growth, and reduced survival of the young (Thompson, 1996; Burger and Gochfeld, 1997). In our study, the levels of Hg measured in all gannet eggs from Ailsa Craig were within the range of concern (1.5 to 5.1 µg g^{-1} dry wt.), as well as in some eggs from Bass Rock (0.8 to 2.7 µg g^{-1} dry wt.). Therefore, it is possible that some gannet embryos from both colonies may experience adverse effects due to Hg. However, we have no specific evidence of Hg-induced toxicity from either colony, and, furthermore, there is marked species variation in sensitivity to Hg. Seabirds naturally accumulate relatively high Hg tissue concentrations because the physical-chemical aquatic conditions can facilitate the conversion of inorganic Hg into bioavailable methyl-mercury which accumulates in fish and their predators (Monteiro and Furness, 2001; Wiener et al., 2003), and may be relatively insensitive compared with other species.

5. Conclusions

Our long term monitoring has demonstrated clear spatio-temporal differences in PCB congener and Hg contamination in gannet eggs between the Ailsa Craig and Bass Rock colonies. Our results suggest that contamination levels may differ between the eastern Atlantic and the North Sea regions off the British coast. However, some or all of the variation between colonies may also be explained by variation in prey selection. Parallel analysis of sediments, water and fish, together with food web studies using isotopic labelled compounds (^{13}C and ^{15}N), are needed to elucidate the underlying factors controlling inter-colony variation in PCB and Hg bioaccumulation by gannets. The lack of any decline in some contaminants, for example some of the heavier PCB congeners and Hg in gannets at Bass Rock, highlights a need for further monitoring to determine future risk.

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References

Alcock, R.E., Boumphrey, R., Malcolm, H.M., Osborn, D., Jones, K.C., 2002. Temporal and spatial trends of PCB congeners in UK Gannet eggs. Ambio 31, 202-206.

AMAP, 1998. AMAP assessment report: arctic pollution issues. Arctic monitoring and assessment programme (AMAP), Oslo, Norway.

Arcos, J.M., Ruiz, X., Bearhop, S., Furness, R.W., 2002. Mercury levels in seabirds and their fish prey at the Ebro Delta (NW Mediterranean): the role of trawler discards as a source of contamination. Marine Ecology-Progress Series 232, 281-290.

Becker, P.H., Cifuentes, J.M., 2004. Contaminants in Bird Eggs in the Wadden Sea. Recent Spatial and Temporal Trends, Seabirds at Risk? Effects of Environmental Chemicals on Reproductive Success and Mass Growth of Seabirds Breeding at the Wadden Sea in the Mid 1990s. Wadden Sea Ecosystem, No 18.

Becker, P.H., Cifuentes, J.M., Behrends, B., Schmieder, K.R., 2001. Contaminants in Bird Eggs in the Wadden Sea. Spatial and temporal trends 1991-2000. Wadden Sea Ecosystem, No 11, p. 68.

Becker, P.H., Thyen, S., Mickstein, S., Sommer, U., Schmieder, K.R., 1998. Monitoring pollutants in coastal bird eggs in the Wadden Sea. Wadden Sea ecosystem No 8 report, final report of the pilot study 1996-1997. Germany, pp. 59-98.

Bishop, C.A., Koster, M.D., Check, A.A., Hussell, D.J.T., Jock, K., 1995. Chlorinated hydrocarbons and mercury in the sediments, red-winged blackbirds (*Agelaius Phoeniceus*) and tree swallows (*Tachycineta bicolor*) from wetlands in the Great lakes- St Lawrence river basin. Environmental Toxicology and Chemisty 14, 491-501.

Borlakoglu, J., Wilkins, J., Walker, C., Dils, R., 1990a. Polychlorinated biphenyls (PCBs) in fish-eating sea birds. III. Molecular features and metabolic interpretations of PCB isomers and congeners in adipose tissues. Comparative Biochemistry and Physiology 97, 173-177.

Borlakoglu, J.T., Wilkins, J.P.G., Walker, C.H., Dils, R.R., 1990b. Polychlorinated biphenyls (PCBs) in fish-eating seabirds - II. Molecular features of PCB isomers and

congeners in adiposer tissue of male and female puffin (*Fratercula arctica*), guillemots (*Uria aalge*), shags (*Phalacrocorax aristotelis*) and cormorants (*Phalacrocorax carbo*) of British and Irish coastal waters. Comparative Biochemistry and Physiology 97C, 161-171.

Braune, B., Norstrom, R., 1989. Dynamics of organochlorine compounds in herring-gulls - .3. Tissue distribution and bioaccumulation in Lake-Ontario Gulls. Environmental Toxicology and Chemistry 8, 957-968.

Braune, B.M., Donaldson, G.M., Hobson, K.A., 2001. Contaminant residues in seabird eggs from the Canadian Arctic. Part I. Temporal trends 1975-1998. Environmental Pollution 114, 39-54.

Braune, B.M., Donaldson, G.M., Hobson, K.A., 2002. Contaminant residues in seabird eggs from the Canadian Arctic. II. Spatial trends and evidence from stable isotopes for intercolony differences. Environmental Pollution 117, 133-145.

Braune, B.M., Outridge, P.M., Fisk, A.T., Muir, D.C.G., Helm, P.A., Hobbs, K., Hoekstra, P.F., Kuzyk, Z.A., Kwan, M., Letcher, R.J., Lockhart, W.L., Norstrom, R.J., Stern, G.A., Stirling, I., 2005. Persistent organic pollutants and mercury in marine biota of the Canadian Arctic: An overview of spatial and temporal trends. Science of the Total Environment 351, 4-56.

Burger, J., 1994. Heavy metals in avian eggshells: another excretion method. Journal of Toxicology and Environmental Health 41, 207-220.

Burger, J., Gochfeld, M., 1997. Risk, mercury levels, and birds: relating adverse laboratory effects to field biomonitoring. Environmental Research 75, 160-172.

Carpi, A., 1997. Mercury from combustion sources: a review of the chemical species emitted from their transport in the atmosphere. Water Air and Soil Pollution 98, 241 - 254.

Cifuentes, J.M., Becker, P.H., Sommer, U., Pacheco, P., Schlatter, R., 2003. Seabird eggs as bioindicators of chemical contamination in Chile. Environmental Pollution 126, 123-137.

Connell, D.W., Fung, C.N., Minh, T.B., Tanabe, S., Lam, P.K.S., Wong, B.S.F., Lam, M.H.W., Wong, L.C., Wu, R.S.S., Richardson, B.J., 2003. Risk of breeding success of fish-eating Ardeids due to persistent organic contaminants in Hong Long: evidence from organochlorine compounds in eggs. Water Research 37, 459-467.

Evers, D.C., Taylor, K.M., Major, A., Taylor, R.J., Poppenga, R.H., Scheuhammer, A.M., 2003. Common loon eggs as indicators of methylmercury availability in North America. Ecotoxicology 12, 69-81.

Fowler, D., McDonald, A.G., Crossley, A., Nemitz, E., Leaver, D., Cape, J.N., Smith, R.I., Anderson, D., Rowland, P., Ainsworth, G., Lawlor, A.J., Guyatt, H., Harmens, H., 2006. UK Heavy Metal Monitoring Network. Centre for Ecology and Hydrology, p. 120.

Fox, G.A., Weseloh, D.V., Kubiak, T.J., Erdman, T.C., 1991. Reprodutive outcomes in colonial fish-eating birds - A biomarker for developmental toxicants in Great-Lakes food

chains.1. Historical and ecotoxicological perspectives. Journal of Great Lakes Research 17, 153-157.

Furness, R.W., 1993. Birds as monitors of pollutants, in: Furness, R.W., Greenwood, J.J.D. (Eds.), Birds as monitors of environmental change. Chapman & Hall, London, pp. 86-143.

Furness, R.W., Monaghan, P., 1987. Seabird ecology. Blacky & Son., Glasgow, London.

Giesy, J.P., Ludwig, J.P., Tillitt, D.E., 1994. Deformities in birds of the Great Lakes region: assessing casualty. Environmental Science and Technology 28, 128-135.

Gochfeld, M., Burger, J., 1998. Temporal trends in metal levels in eggs of endangered Roseate tern (*Sterna dougallii*) in New York. Environmental Research Section A 77, 36-42.

Grassman, K.A., Seanlon, P.F., Fox, G.A., 1998. Reproductive and physiological effects of environmental contaminants in fish-eating birds of the Great Lakes: a review of historical trends. Environmental Monitoring Assessment 53, 117-145.

Guruge, K., Tanabe, S., 1997. Congener specific accumulation and toxic assessment of polychlorinated biphenyls in common cormorants, *Phalacrocorax carbo*, from Lake Biwa, Japan Environmental Pollution 96, 425-433.

Hamer, K.C., Phillips, R.A., Hill, J.K., Wanless, S., Wood, A.G., 2001. Contrasting foraging strategies of gannet *Morus bassanus* at two North Atlantic colonies: foraging trip duration and foraging area fidelity. Marine Ecology Progress Series 224, 283-290.

Herzke, D., Kallenborn , R., Nygård, T., 2002. Organochlorines in egg samples from Norwegian birds of prey: Congener-, isomer- and enantiomer specific considerations. The Science of the Total Environment 291, 59-71.

Kakela, A., Furness, R.W., Kelly, A., Strandberg, U., Waldron, S., Kakela, R., 2007. Fatty acid signatures and stable isotopes as dietary indicators in North Sea seabirds. Marine Ecology-Progress Series 342, 291-301.

Kambamanolidimou, A., Kamarianos, A., KIilikidis, S., 1991. Transfer of methylmercuty to hens eggs after oral-admnistration Bulletin of Environmental Contamination and Toxicology 46, 128-133.

Lam, J.C.W., Tanabe, S., Wong, B.S.F., Lam, P.K.S., 2004. Trace element residues in eggs of little egret (*Epretta garzetta*) and Black-crowned Night Heron (*Nycticorax nycticorax*) from Hong Kong, China. Marine Pollution Bulletin 48, 390-396.

Lead, W.A., Steinnes, E., Bacon, J.R., Jones, K.C., 1997. Polychlorinated biphenyls in UK and Norwegian soils: Spatial and temporal trends. Science of the Total Environment 193, 229-236.

Lewis, S.A., Furness, R.W., 1991. Mercury accumulation and excretion in laboratory reared Black-headed Gull *Larus ridibundus* chicks. Archives of Environmental Contamination and Toxicology 21, 316-320.

Lindberg, S., Brooks, S., Lin, C., Scott, K., Landis, M., Steven, R., Goodsite, M., Rihter, A., 2002. A dynamic oxidation of gaseous mercury in the Artic troposphere at polar sunrise. Environmental Science and Technology 36, 1245-1256.

Martin, A.R., 1989. The diet of Atlantic Puffin *Fratercula arctica* and Northen Gannet *Sula bassana* chicks at a Shetland colony during a period of changing prey availabilty. Bird Study 36.

Mitchell, P.I., Newton, S.F., Norman, R., Dunn, T.E., 2004. Seabird Populations of Britain and Ireland. T. & A. D. Poyser Publishers, London.

Monteiro, L.R., Furness, R.W., 2001. Kinetics, dose-response, and excretion of methylmercury in free-living adult Cory's shearwaters. Environmental Science & Technology 35, 739-746.

Moriarty, F., 1990. Ecotoxicology - the study of pollutants in ecosystems. Academic Press, London.

Navarro, M., Lopez, H., Sanches, M., Lopez, M., 1993. The effect of industrial-pollution on mercury levels in water , soil, and sludge in the coastal area of Motril, Southeast Spain. Archives of Environmental Contamination and Toxicology 24, 11-15.

Newton, I., Dale, L., Little, B., 1999. Trends in organochlorine and mercurial compounds in the eggs of British merlins *Falco columbarius.* Bird Study 46, 356-362.

Newton, I., Haas, M.B., Freestone, P., 1990. Trends in organochlorine and mercury levels in gannet eggs. Environmental Pollution 63, 1-12.

Norstrom, R., 1988. Bioaccumulation of polychlorinated biphenyls in Canadian wildlife., in: Crine, J. (Ed.), Hazards, decontaminations and replacement of PCB. Plenum New York, NY, pp. 85-100.

Rice, C.P., O'Keefe, P., Kubiak, T., 2003. Sources, pathways, and effects of PCBs, dioxins, and Dibenzofurans, in: Hoffman, D.J., Rattner, B.A., Burton Jr., G.A., Cairns Jr., J. (Eds.), Handbook of ecotoxicology. Lewis Publishers, London, New York, pp. 501-573.

Sell, J.L., Guenter, W., Sifri, M., 1974. Distribution of mercury among components of eggs following admnistration of methylmercuricchloride to chickens. Journal of Agricultural and Food Chemistry 22, 248-251.

Shore, R. F., Malcolm, H. M, Wienburg, C.L, Turk, A., Walker, L. A, , Horne, J., 2005. Wildlife and pollution: 2000/01: Annual report, JNCC Report, No. 351.

Simpson, V.R., 2007. Health status of otters in southern and south west England 1996-2003, Using science to create a better place. Environment Agency, Bristol, p. 91.

Stickel, L.F., Wiemeyer, S.N., Blus, L.J., 1973. Pesticide-Residues in Eggs of Wild Birds - Adjustment for Loss of Moisture and Lipid. Bulletin of Environmental Contamination and Toxicology 9, 193-196.

Tasker, M.L., Jones, P.H., Blake, B.F., Dixon, T.J., 1985. The marine distribution of the gannet *Sula bassana* in the North Sea. Bird Study 32, 82-90.

Thompson, D.R., 1996. Mercury in birds and terrestrial mammals, in: Beyer, W.N., Heinz, G.H., Redmon-Norwood, A.W. (Eds.), Environmental contaminants in wildlife: interpreting tissue concentrations. CRC Lewis Publishers, Boca Raton, pp. 341-356.

Thyen, S., Becker, P.H., Behmann, H., 2000. Organochlorine and mercury contamination of Little Terns (*Sterna albifrons*) breeding at the western Baltic Sea, 1978-1996. Environmental Pollution 108, 225-238.

Van den Berg, M., Birnbaum, L., Bosveld, A.T.C., Brunstrom, B., Cook, P., Feeley, M., Giesy, J.P., Hanberg, A., Hasegawa, R., Kennedy, S.W., Kubiak, T., Larsen, J.C., van Leeuwen, F.X.R., Liem, A.K.D., Nolt, C., Peterson, R.E., Poellinger, L., Safe, S., Schrenk, D., Tillitt, D., Tysklind, M., Younes, M., Waern, F., Zacharewski, T., 1998. Toxic equivalency factors (TEFs) for PCBs, PCDDs, PCDFs for humans and wildlife. Environmental Health Perspectives 106, 775-792.

Walker, C., 2001. Organic pollutants: an ecotoxicological perspective. Taylor & Francis, NY.

Walker, C., Livingstone, D.R., 1992. Persistent pollutants in marine ecosystems. Pergamon Press, Oxford.

Walker, C.H., 1994. The ecotoxicology of persistent pollutants in marine fish-eating birds., in: Walker, C., Livingstone, D.R. (Eds.), Persistent pollutants in marine ecosystems. Pergamon Press, Oxford, pp. 211-232.

Walker, L.A., Simpson, V.R., Rockett, L., Wienburg, C.L., Shore, R.F., 2007. Heavy metal contamination in bats in Britain. Environmental Pollution 148, 483-490.

Wiener, J.G., Krabbenhoft, D.P., Heinz, G.H., Scheuhammer, A.M., 2003. Ecotoxicology of mercury in: Hoffman, D.J., Rattner, B.A., Burton, G.A., Jr, Cairns, J., Jr (Eds.), The handbook of Ecotoxicology. Lewis Publishers, New York, pp. 409-463.

Figure Legends

Figure 1. Location of Ailsa Craig (West Scotland) and Bass Rock (East Scotland) colonies.

Figure 2. Individual congeners as a percentge of the congener sum in gannet eggs from Bass Rock and Ailsa Craig for 2002.

Figure 3. Time trends of dominant PCB congeners in gannet eggs from Bass Rock and Ailsa Craig.

Figure 4. Scores (A) and loadings (B) of the two first principal components from a principal component analysis of PCB congeners.

Figure 5. Temporal trends in Hg concentration (μg g^{-1} wet weight) in gannet eggs from Bass Rock (A) and Ailsa Craig (B).

Table Legends

Table 1. PCB congener concentrations (geometric mean and range, ng g^{-1} wet weight) from 1990 to 2004 in Gannet eggs collected from Bass Rock and Ailsa Craig. % detected – percent detected above limit of detection; n - is the number of eggs analysed per year. ND - below detection limit; Not Deter – Not determined.

Figure 1

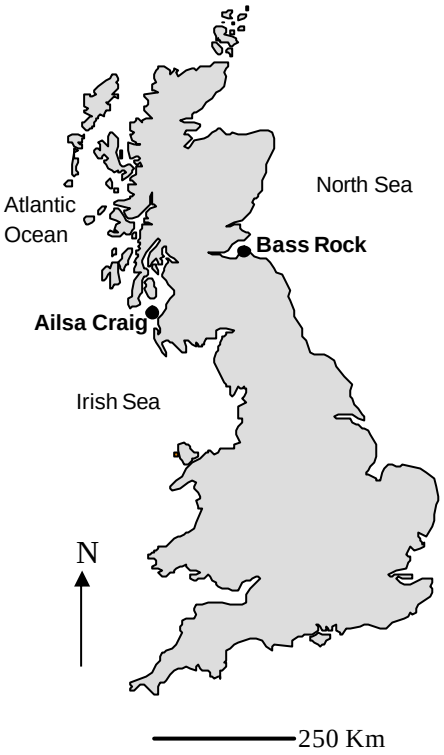
North Sea
Atlantic
Ocean
Bass Rock
Ailsa Craig
Irish Sea
N
250 Km

Figure 2

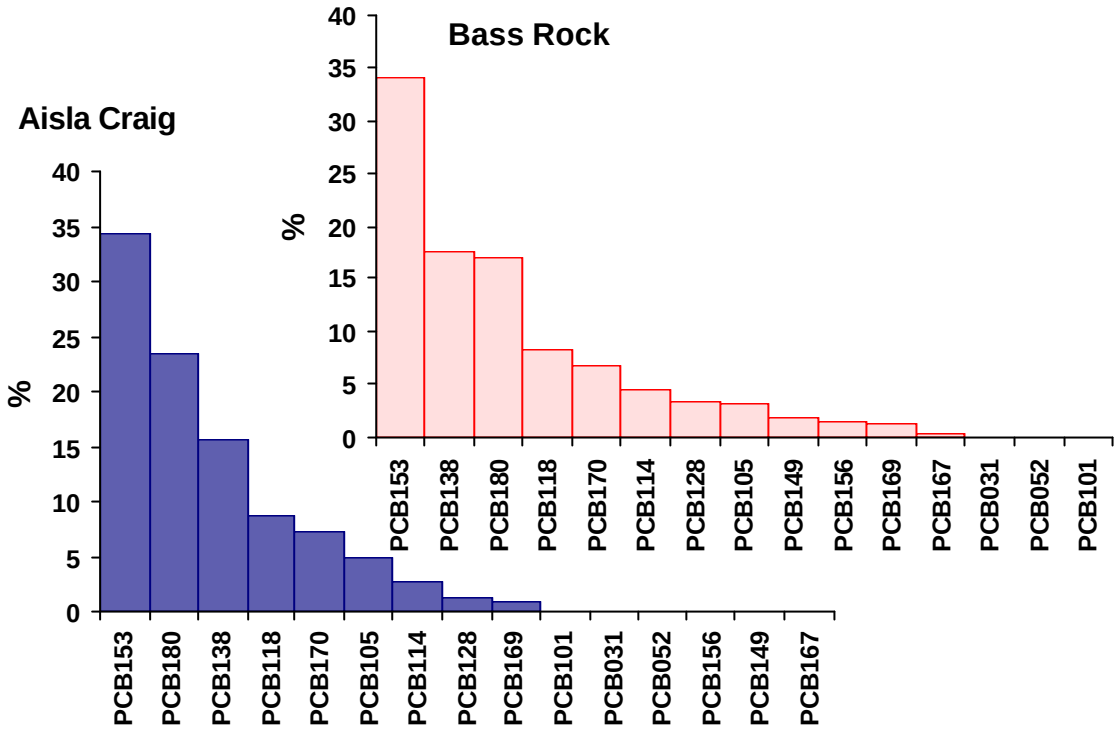
Aisla Craig
%
40
35
30
25
20
15
10
5
0
PCB153
PCB180
PCB138
PCB118
PCB170
PCB105
PCB114
PCB128
PCB169
PCB101
PCB031
PCB052
PCB156
PCB149
PCB167
Bass Rock
%
40
35
30
25
20
15
10
5
0
PCB153
PCB138
PCB180
PCB118
PCB170
PCB114
PCB128
PCB105
PCB149
PCB156
PCB169
PCB167
PCB031
PCB052
PCB101

Figure 3

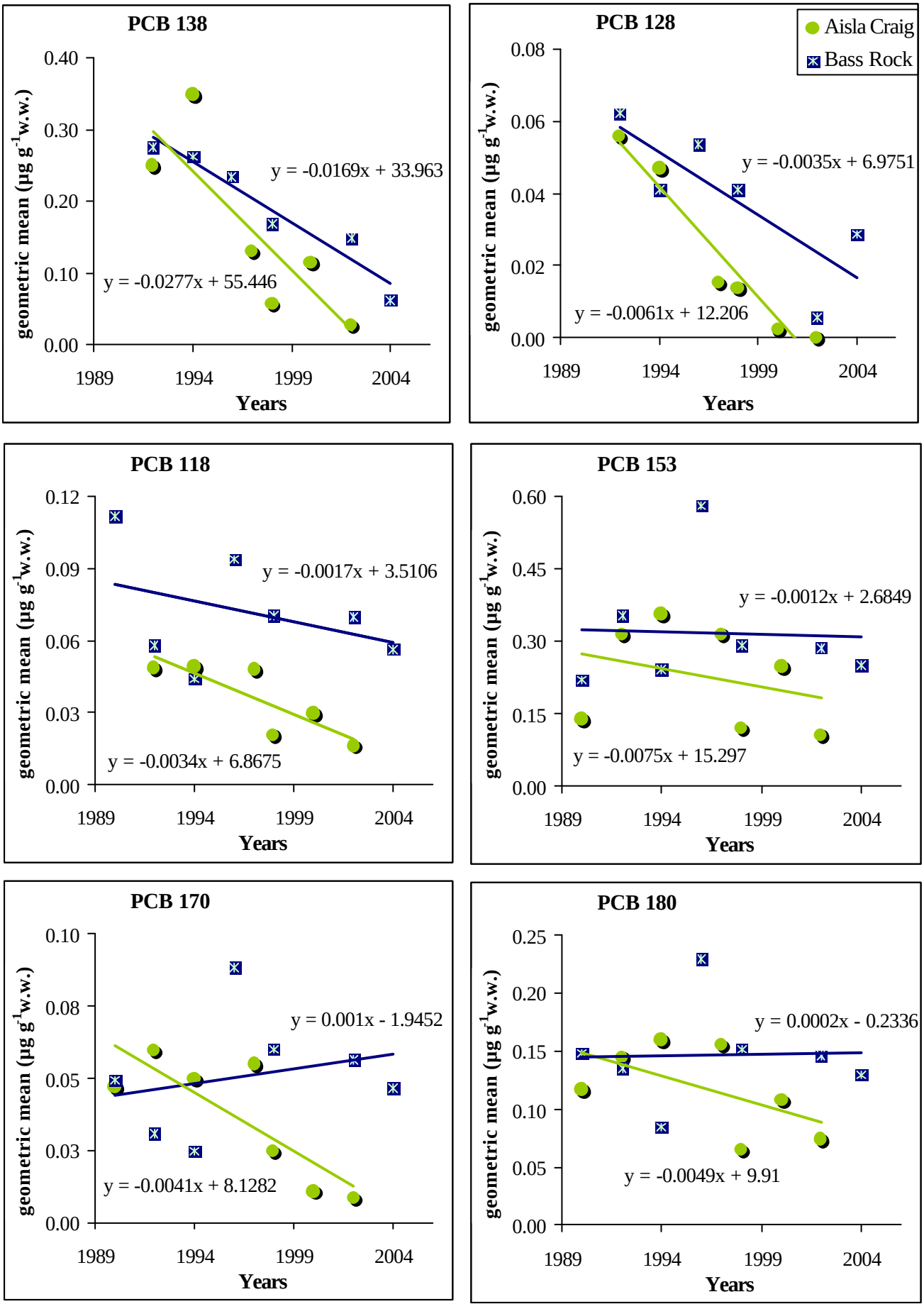
PCB 138
geometric mean (µg g-1w.w.)
0.40
0.30
0.20
0.10
0.00
y = -0.0169x + 33.963
y = -0.0277x + 55.446
1989
1994
1999
2004
Years
PCB 128
Aisla Craig
Bass Rock
0.08
0.06
0.04
0.02
0.00
y = -0.0035x + 6.9751
y = -0.0061x + 12.206
PCB 118
0.12
0.09
0.06
0.03
y = -0.0017x + 3.5106
y = -0.0034x + 6.8675
PCB 153
0.60
0.45
0.30
0.15
y = -0.0012x + 2.6849
y = -0.0075x + 15.297
PCB 170
0.10
0.08
0.05
0.03
y = 0.001x - 1.9452
y = -0.0041x + 8.1282
PCB 180
0.25
0.20
0.15
0.10
0.05
y = 0.0002x - 0.2336
y = -0.0049x + 9.91

Figure 4

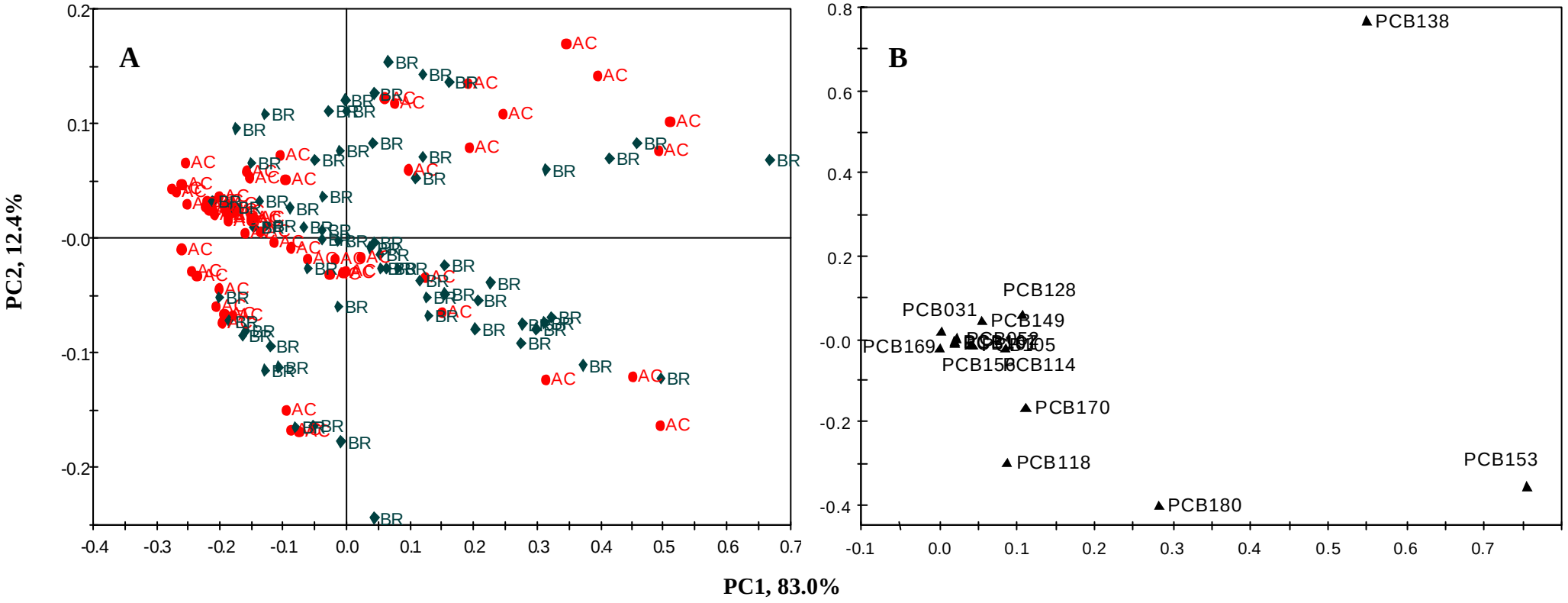
A
B
AC
BR
PCB138
PCB128
PCB031
PCB149
PCB169
PCB114
PCB170
PCB118
PCB153
PCB180
PC2, 12.4%
PC1, 83.0%

Figure 5

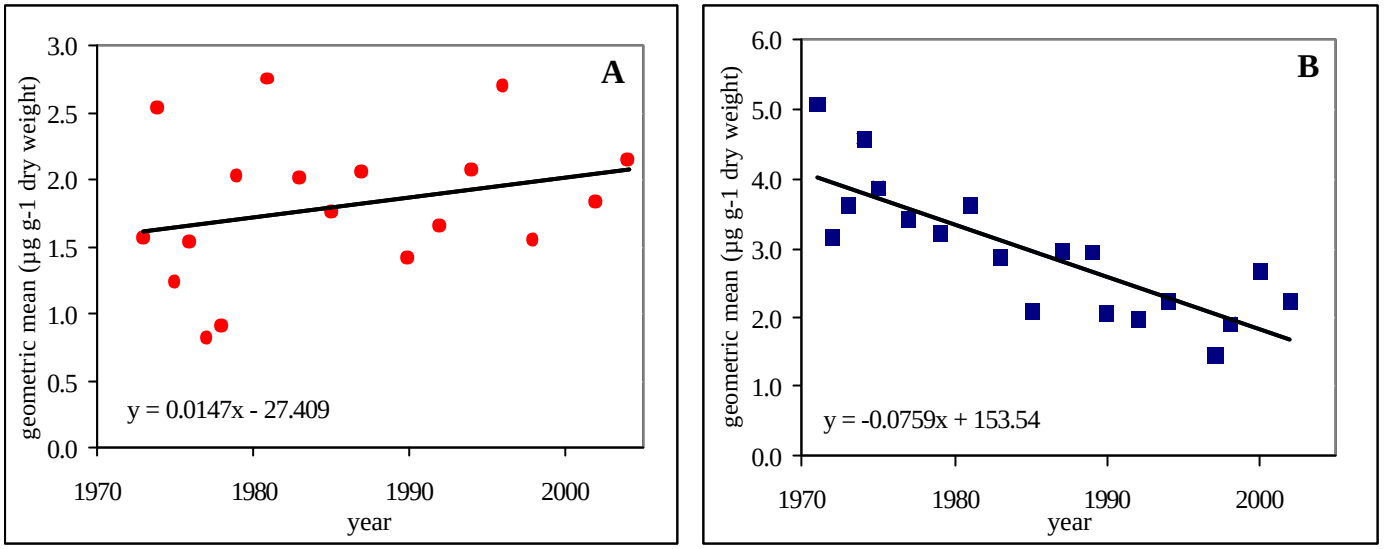
A
geometric mean (µg g-1 dry weight)
3.0
2.5
2.0
1.5
1.0
0.5
0.0
y = 0.0147x - 27.409
1970
1980
1990
2000
year
B
geometric mean (µg g-1 dry weight)
6.0
5.0
4.0
3.0
2.0
1.0
0.0
y = -0.0759x + 153.54
1970
1980
1990
2000
year

Table 1.

		Ailsa Craig							Bass Rock						
PCB	**% detected**	**1990** (n=10)	**1992** (n=9)	**1994** (n=9)	**1997** (n=10)	**1998** (n=9)	**2000** (n=10)	**2002** (n=10)	**1990** (n=10)	**1992** (n=10)	**1994** (n=10)	**1996** (n=10)	**1998** (n=9)	**2002** (n=10)	**2004** (n=10)
31	15.5	Not deter	0.3 ND-19.1	0.4 ND-7.1	ND ND-ND	ND ND-ND	ND ND-ND	ND ND-ND	Not deter	0.2 ND-40.0	0.5 ND-11.2	ND ND-ND	0.2 ND-6.1	ND ND-ND	ND ND-ND
52	15.4	ND ND-ND	1.0 ND-84.2	0.1 ND-8.2	ND ND-ND	0.1 ND-6.1	ND ND-ND	ND ND-ND	ND ND-ND	1.0 ND-61.3	ND ND-ND	2.9 ND-66.2	0.2 ND-11.0	ND ND-ND	0.1 ND-4.9
101	41.2	ND ND-ND	2.1 ND-33.2	1.9 ND-7.7	0.4 ND-91.2	0.2 ND-3.2	0.5 ND-139.1	ND ND-ND	ND ND-ND	0.3 ND-47.1	2.0 ND-22.1	0.9 ND-148.1	2.2 ND-22.2	ND ND-ND	19.2 11.8-32.8
105	82.8	Not deter	Not deter	Not deter	Not deter	6.7 5.6-9.0	0.6 ND-873.4	3.7 ND-41.0	Not deter	Not deter	Not deter	Not deter	21.6 6.3-46.4	16.4 ND-44.1	18.1 12.8-30.2
114	55.2	Not deter	Not deter	Not deter	Not deter	18.1 15.4-27.1	ND ND-ND	1.0 ND-24.1	Not deter	Not deter	Not deter	Not deter	30.8 ND-296.2	36.5 16.3-74.4	ND ND-ND
118	98.5	97.0 54.7-165.2	48.8 20.4-175.1	49.1 11.1-79.3	48.0 16.8-118.2	20.8 15.9-27.2	29.5 ND-204.8	16.4 ND-44.4	111.7 83.8-161.1	58.3 25.1-129.3	44.2 25.2-86.5	93.6 60.8-209.0	70.2 18.1-175.3	69.7 32.2-139.7	56.2 40.2-104.0
128	86.2	Not deter	55.7 23.0-133.6	46.8 8.7-86.5	15.1 ND-75.5	13.6 11.3-20.1	2.3 ND-70.2	0.2 ND-21.2	Not deter	62.2 33.4-126.2	40.8 22.8-86.2	53.6 36.1-130.4	40.8 12.3-95.1	5.6 ND-60.2	28.6 20.4-57.5
138	98.3	Not deter	248.8 101.8-714.4	346.9 56.5-905.2	128.3 53.6-301.2	56.1 43.5-74.2	114.0 34.1-399.1	26.9 ND-98.8	Not deter	275.0 109.2-565.1	261.5 126.2-585.6	233.6 159.2-567.4	168.5 50.6-455.2	147.5 70.1-277.4	62.6 ND-244.2
149	64.2	Not deter	17.3 ND-95.4	22.4 5.5-43.1	0.7 ND-49.1	0.1 ND-2.0	Not deter	ND ND-ND	Not deter	26.8 8.0-55.3	28.4 13.4-59.2	4.9 ND-91.3	2.0 ND-64.1	1.3 ND-56.1	6.1 ND-43.2
153	100.0	138.8 79.8-244.2	312.7 134.0-873.2	356.2 64.6-1022.2	314.6 140.2-714.5	119.6 101.8-157.2	246.5 76.6-905.1	105.3 51.6-203.3	220.7 142.1-106.5	352.8 146.3-768.6	241.8 113.9-561.4	581.2 407.0-1478.1	291.0 42.1-1059.4	285.1 133.0-542.4	251.2 161.7-461.9
156	60.3	Not deter	Not deter	Not deter	Not deter	9.3 7.8-12.3	0.2 ND-68.2	ND ND-ND	Not deter	Not deter	Not deter	Not deter	23.3 7.2-56.3	1.2 ND-34.2	20.1 13.3-33.7
167	53.4	Not deter	Not deter	Not deter	Not deter	2.7 2.1-3.0	0.1 ND-36.3	ND ND-ND	Not deter	Not deter	Not deter	Not deter	9.0 2.2-24.4	0.2 ND-20.3	9.7 7.4-13.6
169	28.3	Not deter	ND ND-ND	0.3 ND-3.2	ND ND-ND	4.1 2.0-6.4	0.2 ND-70.6	0.2 ND-17.2	Not deter	ND ND-ND	Not deter	ND ND-ND	1.5 ND-22.1	1.2 ND-33.1	ND ND-ND
170	96.3	48.7 21.6-102.3	59.7 26.1-206.5	49.9 10.7-116.3	55.0 23.8-134.2	24.6 20.3-33.6	10.8 ND-161.4	8.5 ND-48.1	49.2 15.5-106.2	30.6 ND-114.3	24.6 3.2-59.3	88.5 53.8-232.3	59.8 21.7-129.2	56.1 26.6-95.2	46.4 27.1-78.8
180	100.0	116.6 57.2-262.5	144.5 64.5-480.2	159.6 29.7-499.6	155.3 66.6-346.4	64.8 50.1-85.8	107.8 31.3-524.6	74.1 34.1-128.0	148.0 84.2-273.5	135.2 50.3-257.8	85.1 25.0-167.7	229.5 139.4-617.1	151.6 50.3-341.5	145.7 65.1-239.3	129.6 79.5-217.6