Contemporary patterns and historical rates of increase of mercury contamination in different marine food chains

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Summary: We present measured trends in mercury contamination over the past 100 years in epipelagic, mesopelagic and deep sea marine food webs, showing 3-10 fold increases in mercury concentrations. The most rapid increases, and highest contemporary levels, occur in mesopelagic food chains in southern ICES areas. Congruent patterns are evident in fish and in seabirds, but the latter permit ready analysis of long term trends because of the availability of museum study-skin collections. Laboratory and field studies fully validate the use of seabird feathers as a reliable monitor of mercury contamination in their food. Although local mercury contamination 'hot-spots' can be identified within the North Sea, most of the North Sea food chains can be considered 'mercury-depleted' relative to those in North Atlantic environments. This pattern supports the view that point source and riverborne mercury is of minor importance relative to inputs from atmospheric deposition. The latter suggests that jet stream transport of mercury pollution from North America is the primary cause of mercury contamination in European waters. Methylation of inorganic mercury deposited from the atmosphere is thought to occur especially in low oxygen environments such as the deep sea. This is important because vertebrates assimilate a very high proportion of ingested organic mercury but only a small fraction of ingested inorganic mercury. High accumulation of mercury in mesopelagic and deep sea fish and in predators on mesopelagic animals suggests that methylation is a key process determining mercury accumulation in marine animals.

Introduction

Among the toxic trace metals, mercury is of particular concern as a hazard to human health, as levels in foods can exceed recommended exposure limits and neurotoxic effects can be seen in humans and in wild vertebrates with elevated exposures (Clarkson 1994, Thompson 1996). Mercury can occur, and be transported around the globe, in a variety of chemical forms, as elemental mercury, inorganic mercury, monomethylmercury and dimethylmercury (Mason and Fitzgerald 1990, 1991, 1993, Mason et al. 1992, 1995). Methylmercury is many times more toxic to vertebrates than are elemental or inorganic mercury (Thompson 1996), and accumulation of methylmercury tends to be much more efficient than accumulation of elemental or inorganic mercury (Southworth et al. 1995, Atwell et al. 1998, Downs et al. 1998). The biogeochemical cycle of mercury involves interchange between these chemical forms in atmospheric, aquatic and terrestrial compartments, but accumulation of mercury by animals tends to involve methylmercury owing to its high assimilation, and the fact that, unlike inorganic mercury, methylmercury is lipid-soluble (Kannan et al. 1998). Anthropogenic emissions of mercury are thought to represent about two-thirds of the total annual input of some 7000 t mercury (predominantly elemental mercury) to the atmosphere. Exposure of people to mercury occurs especially through human consumption of fish, as a consequence of monomethylmercury (MMHg) biomagnification through aquatic food chains, and particularly its accumulation in fish. It is estimated that mercury emissions to the atmosphere have increased due to increased coal and gas combustion, metal mining and smelting, industrial emissions and waste incineration to the extent that mercury concentrations in oceans are likely to be about three times as high as they were 100 years ago (Mason et al. 1994). However, mercury inputs to marine ecosystems will vary between the more industrialised northern hemisphere and the less developed southern hemisphere, and in relation to patterns of atmospheric transport from major industrial areas towards more pristine environments (Fitzgerald and Mason 1998). Patterns of contamination can be expected to vary on a more local scale in relation to discharges from point sources of mercury and in relation to discharges from contaminated rivers into shallow shelf seas (Ebinghaus et al. 1994).

Despite the prediction of significant increase in mercury contamination of oceans, historical data sets demonstrating such increases are very few (Slemr and Langer 1992, Swain et al. 1992, Fitzgerald 1995, Petersen et al. 1995). Techniques to make accurate measurements of mercury species and concentrations in seawater have only been available for a short time, and analyses of mercury in ice cores has been constrained by sampling and analytical limitations. Lake sediments and peat cores suggest an increase of mercury contamination of terrestrial and freshwater sites consistent with the predicted three-fold increase of atmospheric mercury, but from marine ecosystems the only detailed historical analyses to date have been of very small numbers of preserved fish, or from seabirds (Thompson et al. 1992a, 1993a,b, 1998a, Monteiro and Furness 1997).

Turning to studies of the spatial pattern of mercury contamination, we find a much wider range of possibilities; mercury concentrations can be compared between regions in seawater, in suspended particulate organic matter, in sediments, plankton, fish, mammals or birds. Concentrations of mercury in seawater, in POM or in sediments may not provide much indication of the degree of contamination of local food chains, since that will depend not only on the amount of mercury and chemical form, but also on its availability for uptake into organisms. Thus with regard to considerations of mercury contamination of marine biota, especially fish of interest for human consumption, it would seem best to investigate mercury levels in these fish. However, mercury levels in top predators such as seabirds may be useful indicators of geographical patterns in contamination, as seabirds are relatively easy to sample, and they can integrate mercury contamination over relevant spatial scales related to their foraging distribution. Furthermore, fish show age-related, and growth-rate related accumulation of mercury, and so local contamination must be corrected for variations in fish age and/or growth rates, and mercury can be taken up across the gills as well as from food (Canli 1995) although food seems to be the main source (Hall et al. 1997). This complication does not arise with seabirds because mercury concentrations in adult seabirds are not affected by bird age, and the mercury in birds is derived entirely from food and almost entirely as monomethylmercury (see below).

Seabirds as biomonitors of mercury in marine food webs

Seabirds tend to feed in the upper trophic levels of marine food webs. Unlike fish (Joiris et al. 1995b, 1997a) and marine mammals (Hansen et al. 1990, Thompson 1990, Andre et al. 1991), seabirds do not show life-long age-related accumulation of mercury. Although mercury levels in young seabirds tend to be lower than in adults, after they have reached one year old, there is no evidence for any further age-related increase in methylmercury concentrations in any seabird tissues (Furness et al. 1990, Thompson et al. 1991). This results from the fact that feather keratins represent the major route for excretion of methylmercury by seabirds (Furness et al. 1986, Braune and Gaskin 1987a,b, Burger 1993, Becker et al. 1993a,b, 1994, Bearhop et al. 2000a,b,c). Seabirds incorporate methylmercury in growing feathers, where it is tightly bound in a highly stable form linked to sulphur amino acid sulphur-sulphur double bonds (Appelquist et al. 1984). Once feathers are fully formed, they are dead tissue, and methylmercury in the feather remains there. Most seabirds renew feathers once per year, in the autumn moult. It is only during the active growth of feathers that mercury can be incorporated. As a result, methylmercury tends to accumulate in seabird soft tissues during intermoult periods, and is then excreted into growing feathers such that the entire annual intake of mercury is lost with each annual moult cycle. Studies with captive and with wild seabirds have demonstrated that mercury concentrations in feathers reflect mercury concentrations in the blood during the period of feather growth (Thompson et al. 1990, Bearhop et al. 2000b,c), and that this in turn reflects dietary uptake of methylmercury (Lewis and Furness 1991, Stewart et al. 1997, Monteiro et al. 1998). Thus mercury concentrations in birds are dependent on the dietary dose and can be used to estimate dietary exposure to methylmercury (Furness 1993, Monteiro et al. 1998). Thus mercury concentrations in seabird feathers can be used as measures of mercury contamination of the food chain within which the particular seabird feeds. Monteiro et al. (1998) showed that the ‘bioconcentration factor’ of mercury concentration in food (dry mass) to mercury concentration in body feathers of the seabird was fairly consistent across a range of seabird species. Although dietary intake and mercury concentrations in seabirds varied by more than an order of magnitude among species, the bioconcentration factor from food to plumage was between 125 and 146 in 5 of 6 species studied. Whether this ratio is a general one for all birds and ecosystems has yet to be examined. It may vary as a function of the proportion of plumage moulted each year as this can differ among species; the one outlier in the study by Monteiro et al. (1998) is the only seabird in that sample that apparently moults rather less than 100% of plumage each year. If the bioconcentration factor is similar across species then this would permit comparisons of plumage mercury concentrations between different seabird species in different regions rather than requiring comparisons to be made within species between regions.

Female seabirds can excrete methylmercury into developing eggs as well as into growing feathers (Lewis and Furness 1993, Burger 1994). This may be expected to lead to lower levels of mercury in breeding females than in males, and such a difference has been detected in some studies (Lewis et al. 1993). However, the amount of mercury lost into eggs is very small compared to the amount lost into feathers, and so the difference in plumage mercury or soft-tissue mercury concentrations between male and female seabirds is trivial (Furness 1993, Lewis et al. 1993). However, this permits the use of eggs to monitor mercury , as mercury concentrations in eggs relate to mercury levels in food consumed in the few days immediately prior to egg laying, when the birds are known to be feeding in a defined area around their breeding site (Furness 1993, Barrett et al. 1996).

One particular advantage of seabird feathers as a biomonitor of mercury contamination is that large collections of seabird study skins exist in museums around the world, many dating back to 100-150 years ago and with information on collection date and locality. This permits analysis of historical change in mercury contamination by comparing mercury levels in particular feathers from seabird study skins collected over an extended time period from a particular locality or region (Thompson et al. 1992a,b, 1993a,b, 1998a,b, Furness et al. 1995, Monteiro and Furness 1995, 1997).

Several potential pitfalls in using seabirds as biomonitors of spatial and temporal patterns in mercury contamination can be identified, and these have been given detailed attention (Walsh 1990, Burger 1993, Furness 1993, Monteiro and Furness 1995, Furness and Camphuysen 1997, ICES 1999). Concentrations of mercury in different feathers can indicate seasonal variations in mercury uptake if the pattern of moult is known. In particular, primary feathers are generally renewed in precise sequence and timing and in some species some may be grown in breeding areas and others in the wintering area. This has been used to study seasonal patterns of mercury exposure in several studies, such as those by Hario and Uuksulainen (1993) and Bearhop et al. (2000a). Broadly speaking, body feather mercury tends to reflect exposure of seabirds to mercury in food eaten during the breeding season (Monteiro and Furness 1995, 1997, Bearhop et al. 2000a). Seabirds that are omnivorous and feed on a wide range of prey in different habitats are likely to provide a less reliable measure of mercury in a particular food chain than seabirds feeding on a narrowly-defined prey category. This makes such omnivores as many of the Larus gulls less suitable since mercury levels vary considerably between food chains. In particular, mercury levels are much lower in terrestrial habitats than in marine ones, and so seabirds that feed in both terrestrial and marine habitats are likely to be less suitable as biomonitors. Long-term changes or spatial differences in feeding habits within species can cause unexpected trends. An example is the northern fulmar, where diet of fulmars around Britain as indicated by stable isotope signatures appears to have changed over decades (Thompson et al. 1995). However, such long-term changes have not been detected in other British seabirds.

Historical trends in mercury contamination in different marine food chains

Two typical examples of the historical increase in mercury contamination in marine food chains of the northern hemisphere are shown in Figure 1. In addition to showing a clear and approximately linear increase, these graphs also show the characteristically high individual variation in mercury burdens with the highest levels about 5 times the lowest within the population at any period of time. This high level of individual variation in mercury levels within populations is found to a similar, or even greater, extent in other animal groups, and creates a requirement for large sample sizes to be analysed to give reliable mean values. This consistent upward trend fits predictions from models that indicate that atmospheric deposition of mercury is the main input into marine environments (Mason et al. 1994). Not all regions show the same pattern. Even though mercury levels in southern hemisphere albatrosses and petrels tend to be very high (Muirhead and Furness 1988), southern hemisphere seabirds show only slight increases in mercury burden or no significant change (ignoring local point-source effects) (Thompson et al. 1993b), while seabirds in the southeastern North Sea show a more complex pattern that seems to relate to local riverborne inputs of mercury from European industry rather than from long distance atmospheric transport. Becker et al. (1993b) showed that eggs, and down and feathers of chicks, of gulls and terns from colonies along the German North Sea coast indicated very local patterns of mercury contamination. Concentrations of mercury were very high in birds breeding at the mouth of the River Elbe, but much lower at coastal colonies only some 40-50 km away from this well known source of mercury contamination. Thompson et al. (1993a) measured changes in mercury concentrations in adult herring gulls from southeastern North Sea colonies from 1880 to 1990. This pattern differs from the more common one of an approximately constant increase over the last 100 years (see above). In the case of German North Sea herring gulls, mercury contamination briefly rose to very high levels during and immediately after the Second World War, then followed an increasing trend from the 1950 to 1980, after which levels decreased (Figure 2). These patterns correlate very closely with the quantity of mercury used in German industries in the postwar decades (Thompson et al. 1993a) suggesting that the mercury in these gulls is largely derived from riverborne mercury contamination entering the southeastern North Sea, rather than from long-distance transport of atmospheric mercury contamination.

Tables 1 and 2 summarise some examples of rates of increase in mercury contamination of seabirds, presenting data from warm-temperate North Atlantic (Azores) and cool-temperate (British Isles). They show some clear features that are consistent in other similar data sets. Firstly, rates of increase of mercury contamination have been higher in seabirds feeding on mesopelagic prey than in seabirds feeding on epipelagic prey. Secondly, both the pre-1931 and contemporary concentrations of mercury in seabirds feeding on mesopelagic prey tend to be especially high compared to seabirds feeding on epipelagic or coastal prey, or on terrestrial or freshwater prey.

Spatial patterns in mercury contamination

Concentrations of mercury and rates of increase tend to be highest in southwest Europe (Monteiro et al. 1995) and decrease in a clinal pattern towards the northeast. Concentrations and rates of increase are high in seabirds breeding on the southwest of the British Isles, moderate in the northwest, low among seabirds breeding in the northern North Sea, and especially low in seabirds in north Norway (Thompson et al. 1992a, b, 1998a, Furness et al. 1995, Monteiro and Furness 1995, 1998, Monteiro et al. 1999). Another region showing particularly elevated concentrations of mercury in seabirds is northwest Iceland, where concentrations tend to be higher than in the same species in Britain or Norway

(Thompson et al. 1992b, R. Palsson unpubl.). It is particularly noteworthy that seabirds at North Sea colonies tend to have lower levels of mercury than conspecifics at Atlantic colonies (Thompson et al. 1992b, Walsh 1990). The exception to this is birds in colonies close to the mouth of major European rivers discharging into the southeast North Sea, where in very localised areas mercury levels can be greatly elevated (Becker et al. 1993a, Furness et al. 1995). Seabirds spending the winter in the southern North Sea can also accumulate higher burdens of mercury than conspecifics wintering in the northern North Sea (Joiris et al. 1997b). In contrast, seabirds at Mediterranean colonies tend to have higher mercury levels than conspecifics from Atlantic colonies (Renzoni et al. 1986).

Implied mechanisms of mercury transformation and bioavailability

The high increase in mercury contamination in mesopelagic food chains is presumably a consequence of long-distance atmospheric transport of mercury (Swain et al. 1992, Petersen et al. 1995, Fitzgerald and Mason 1998), deposition into the oceans (Mason et al. 1994, Sorensen et al. 1994), and sinking to depth (Joiris 1995a), where under low-oxygen conditions bacteria convert inorganic mercury to methylmercury (Cossa et al. 1994, Mason et al. 1995). This renders the mercury more suitable for uptake into the food chain and biomagnification through the food chain (Downs et al. 1998, Kannan et al. 1998, Monson and Brezonik 1999). Furthermore, close to the sea surface, photodegradation of methylmercury to inorganic mercury will tend to deplete surface waters of methylmercury relative to deeper water (Sellers et al. 1996), reducing the availability of mercury to organisms living close to the surface. Consistent with this interpretation, Monteiro et al. (1996) found that mercury concentration increased with depth inhabited by different small fish in the vicinity of the Azores. Similarly, some deep sea fish have rather higher mercury burdens than found in most shelf-sea fish (S. Fleming unpubl.).

How seabirds that feed primarily on mesopelagic fish and squids catch these prey is not known, although the importance of these foods in their diets is well documented. Madeiran storm petrels and Bulwer's petrels certainly cannot dive to great depths, and are thought to catch prey close to the sea surface mainly at night. Most likely the vertical migrations of mesopelagic animals make them available to these seabirds.

The higher accumulation of mercury in seabirds from southwestern regions of the North Atlantic suggests that atmospheric deposition may be particularly high in those regions, and lower further northeast. Possibly this might relate to transport by jet stream of mercury contamination across the Atlantic from North America. Whatever the source, the process of methylation of mercury appears to be a crucial step in enhancing uptake and accumulation into marine food chains. The continued accumulation of mercury by fish throughout their life may also tend to particularly elevate mercury concentrations in food chains containing slow growing long-lived fish, as is the case in many deep-sea environments.

Implications for assessment of toxic hazards of mercury

Seabirds appear to be particularly well adapted to coping with high concentrations of mercury (Thompson and Furness 1989a). Concentrations found in many healthy breeding seabirds exceed those that cause breeding failure and even fatalities in freshwater or terrestrial birds (Barr 1986, Scheuhammer 1987, 1988, Spalding et al. 1994), and there is no compelling evidence that any seabirds display toxic effects due to mercury (Thompson et al. 1991, Thompson 1996), though high levels in tern chicks at German North Sea rivermouth colonies may in combination with high levels of other contaminants, have reduced breeding success of these birds (Becker et al. 1993c). However, there have been no studies of breeding performance of Bulwer's petrels in relation to the particularly large increase in mercury contamination in that species. It seems likely that the naturally higher exposure of deep sea fish to methylmercury as a consequence of methylation in such environments and the long life-span of fish in such habitats (Berg et al. 1997), would mean that these species will show greater tolerance of mercury than seen in freshwater or shallow-sea fish. Most studies of toxicity of mercury to fish have been carried out in laboratory conditions using easily kept species such as carp (e.g. Canli 1995, 1996). However, if toxic effects are occurring, they would be most likely to be seen in local areas with particularly high contamination with mercury from point source discharges (as at mouths of major European rivers) or where mercury levels have increased most (apparently the southwest North Atlantic deep sea environments).

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Figure 1. Mercury concentrations (µg g^{-1} fresh weight of body feathers) in adults of two seabird species from the Azores archipelago sampled from study skins in museum collections (birds collected during the breeding season only) and from contemporary birds caught at colonies, showing the typical pattern seen in many European seabirds of a considerable and approximately linear increase in mercury contamination over the last 100 years. Data from Monteiro and Furness (1997).

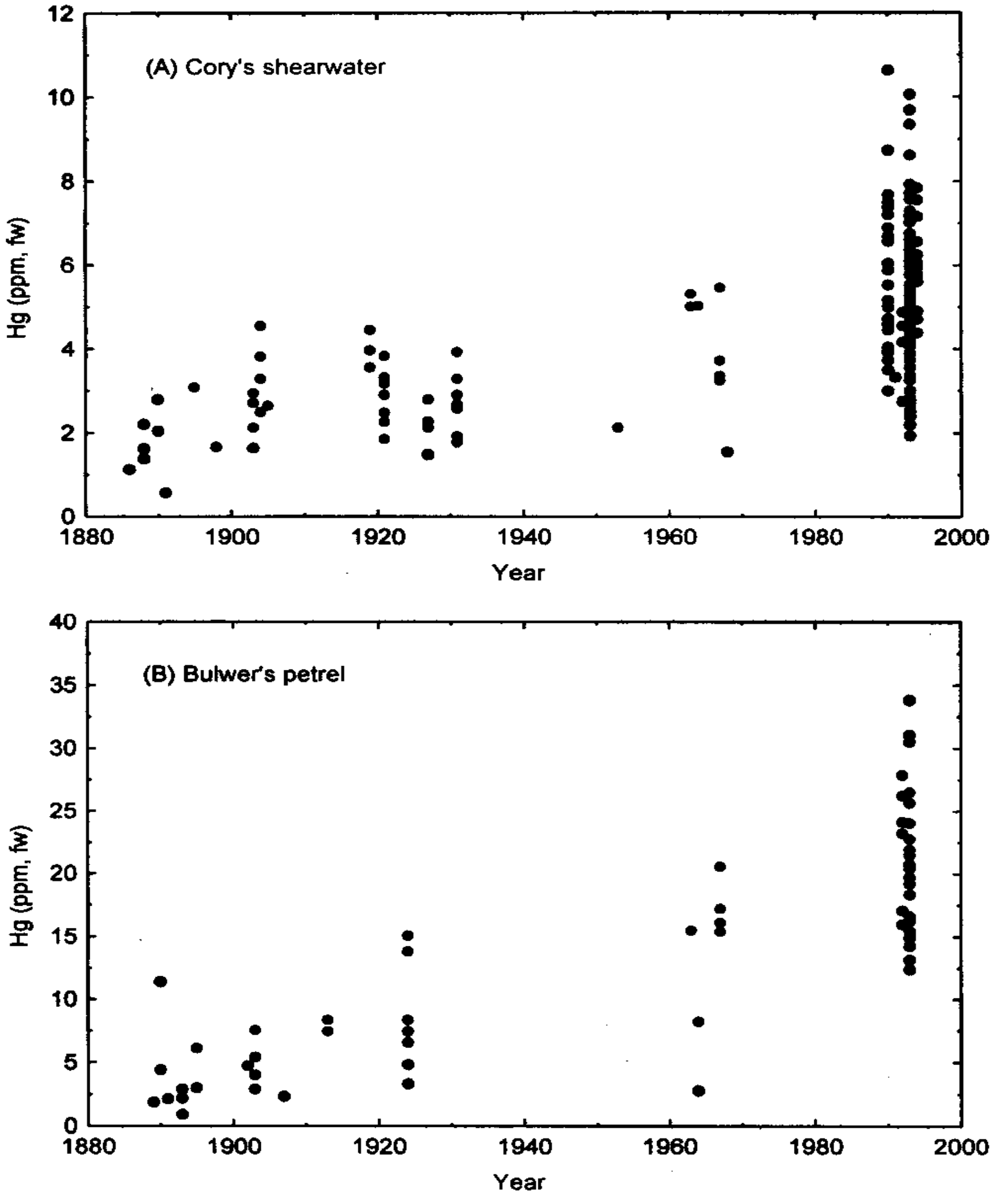

Figure 2. Mercury concentrations (µg g^{-1} fresh weight of body feathers) in adult herring gulls collected from German North Sea colonies during the breeding season, showing changes between decades in mercury contamination. Postwar trends correlate closely with quantities of mercury used by local industry, with clear evidence of declining contamination since the 1980. For comparison, levels in herring gulls from east Scotland during the late 1980s are shown as an open circle plus confidence limits, showing the higher levels in the southeastern North Sea gulls compared to those from east Scotland. Data from Furness et al. (1995).

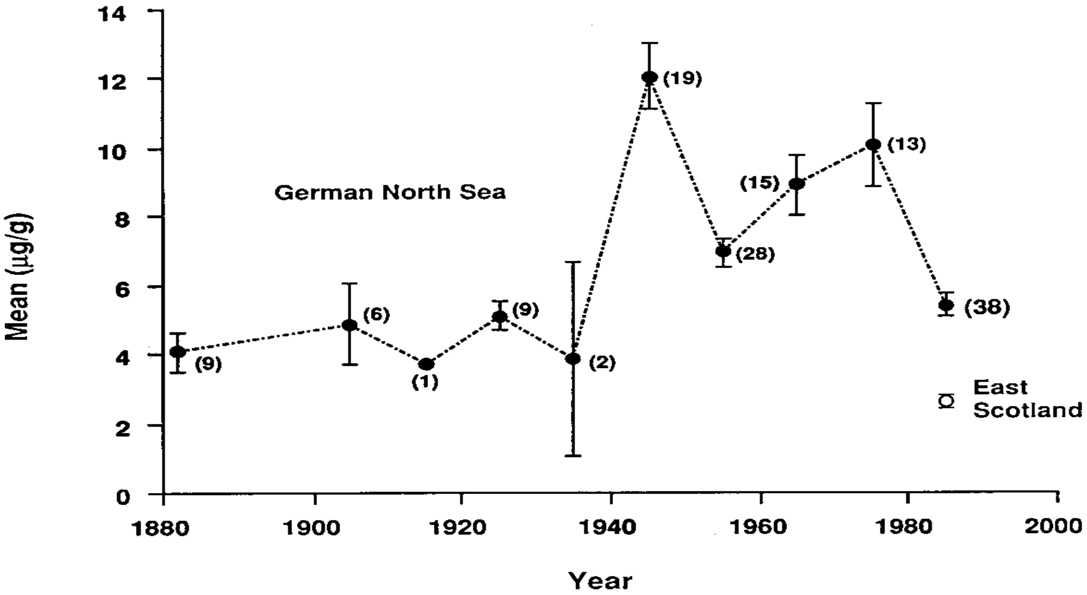

Table 1. Long-term trends in mercury concentrations (µg g^{-1}) in seabird feathers from warm-temperate North Atlantic (Azores) seabirds (from Thompson et al. 1998). Values presented are mean, standard error, sample size.

Species	Pre-1931	Post-1979	Increase (%)	Increase rate (% per year)	Main prey type
Madeiran storm petrel	3.0, 0.1, 16	14.9, 0.5, 47	394	4.1	Mesopelagic
Bulwer's petrel	6.0, 0.7, 30	21.6, 0.7, 55	260	2.9	Mesopelagic
Cory's shearwater	2.7, 0.1, 48	5.4, 0.1, 219	100	1.4	Epipelagic
Common tern	1.1, 0.1, 15	2.0, 0.1, 22	82	1.3	Epipelagic
Little shearwater	1.7, 0.2, 15	2.8, 0.2, 34	65	0.7	Epipelagic

Table 2. Long-term trends in mercury concentrations (µg g^{-1}) in seabird feathers from cool-temperate North Atlantic (British Isles) seabirds (from Thompson et al. 1998). Values presented are mean, standard error, sample size.

Species and subregion	Pre-1931	Post-1979	Increase (%)	Increase rate (% per year)
Manx shearwater; NW Scotland	1.7, 0.2, 14	4.7, 0.2, 78	176	2.3
Manx shearwater; SW Brit. Isles	1.5, 0.2, 17	2.9, 0.2, 22	93	1.0
Atlantic puffin; West Britain	1.8, 0.1, 49	4.4, 0.2, 61	144	1.5
Great skua; all NE Atlantic sites	4.2, 0.6, 13	7.0, 0.3, 212	67	0.4
Northern gannet; East Scotland	5.9, 0.6, 19	7.7, 0.5, 38	31	0.3