NORTHERN GANNETS *MORUS BASSANUS* AS INDICATORS OF MERCURY LEVELS IN THE MARINE ENVIRONMENT

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CANDIDATE'S DECLARATION

I declare that the work recorded in this thesis is entirely my own, unless otherwise stated, and it is of my own composition. No part of this work has been submitted by me for any other degree.

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March 1993

SUMMARY

1. The use of seabirds as monitors of heavy metals in marine environments was reviewed.

2. Intra-population variables related to sampling of seabird feathers for mercury analysis were examined using data for Northern Gannets *Morus bassanus* and Shags *Phalacrocorax aristotelis*.

3. Feather of nestlings (including down) appeared to be the most useful, given that intra-population variation is less than for feathers of adults

4. The number and type of feathers analysed were less important for nestlings than for adults.

5. If adult feathers are to be mercury-analysed, pooled samples of small body-feathers are best.

6. Moulted body-feathers of adults are potentially useful (if pooled before analysis).

7. Geographical patterns of mercury concentrations were examined for feathers of adult and nestling Gannets and Shags, and for Gannet eggs.

8. Mercury concentrations in feathers (including down) of nestling Gannets showed ca. three- to four-fold variation in mean levels between North Atlantic colonies. Concentrations were highest in feathers from Little Skellig (SW Ireland), followed by Grassholm (south Wales).

9. Feathers of adult Gannets showed much less clear-cut patterns than feathers of nestlings, but mercury concentrations were again highest in birds from Grassholm (and, possibly, Alderney in the Channel Islands).

10. Gannet eggs from British and Irish colonies showed similar patterns of mercury concentrations to feathers of nestling Gannets.

11. Feathers of nestling Shags showed similar patterns of mercury concentrations to nestling Gannets.

12. Patterns of mercury concentrations shown by Gannets and Shags may reflect a combination of localised anthropogenic inputs of mercury to coastal waters, and current-mediated inputs.

13. The possible role of variation in Gannet diet in influencing apparent patterns of mercury concentration is discussed, and remains a possible contributing factor.

14. The potential role of biological samples for detecting historical changes in global metal pollution is discussed, with particular reference to cadmium, mercury and lead. Mercury is the metal most amenable to such analysis.

15. Historical changes in mercury concentrations in north and northeast Atlantic seabirds were assessed using feather samples from museum specimens and live-sampled birds. Four out of five species, including Gannet, showed significant increases in plumage mercury concentrations since the mid 19th century; two Northern Fulmar *Fulmarus glacialis* populations showed significant decreases in mercury concentrations over the same period.

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Chapter 1

GENERAL INTRODUCTION AND METHODS

1.1 Mercury and seabirds

Mercury is widely recognised as a highly toxic metal, and, in its organic mono-methyl form, may be the most environmentally hazardous of the non-radioactive elements (Leatherland 1990). Large scale mortality of seed-eating birds has been attributed to the use of mercury seed-dressings in agriculture (e.g. Borg et al. 1969), and industrial inputs have led to highly elevated mercury levels in freshwater and enclosed marine waters (e.g. Fimreite 1974). Mortality incidents involving man have also occurred, notably at Minamata Bay in Japan (Kurland et al. 1960), and widespread efforts have been made to monitor mercury levels in human foodstuffs, especially marine fish which can accumulate high levels of mercury (e.g. Gardner 1978). Assessments of mercury levels have been made in many biota, and, in the case of birds, have been particularly aimed at detecting deleterious effects on reproduction.

The present study aimed to investigate the potential use of seabirds, and in particular the Northern Gannet *Morus bassanus*, as monitors of mercury levels in the marine ecosystem of the North Atlantic and adjacent seas. Feathers are known to accumulate high concentrations of mercury, and a particular aim of this study was to develop this avenue of monitoring in more detail. The general use of seabirds as biomonitors of heavy metals would also be reviewed.

1.2 Format of the thesis

Three of the chapers in this thesis have already been published, and are included here as published, except for repagination. The first of these Chapters (Chapter 2) reviews the factors which need to be taken account of when attempting to use seabird tissues in assessing levels of heavy metals in marine environments. Chapters 5 and 6 were published in collaboration with Dr R. W. Furness and Dr D. R. Thompson, and deal with the use of seabirds and other organisms as sources of data on historical changes in environmental levels of heavy metals, in particular mercury.

The remaining chapters (3 and 4) focus on the use of seabird feathers to assess or monitor mercury levels or bioavailability in marine environments. Chapter 3 examines the intra-population variables which may complicate (or, in some cases, simplify) attempts to use seabird feathers in this way. Chapter 4 deals with a particular case study of mercury in feathers of Northern Gannets, based on analyses of feather tissues from a range of North Atlantic colonies, and draws comparisons with supporting data for Gannet eggs and for feathers of Shags *Phalacrocorax aristotelis*.

1.3 Sampling procedures

All feather samples were collected at or near Gannet or Shag colonies during the breeding season (April-September), in most cases from living birds. Feathers (variously, one or two large scapulars, or pooled samples of small body-feathers or nestling down, usually ca. 0.2-0.5 g total) were sampled by careful plucking or by snipping feathers as close as possible to their bases with scissors. Feathers were also collected from Gannets found dead at some colonies, usually through falls or

other trauma. Moulted Gannet feathers were collected from some colonies, and, under appropriate licences, fresh and addled Gannet eggs.

Samples were collected by PMW (sometimes with help from other fieldworkers) from Gannet or Shag colonies at the Bass Rock, Hermaness, St Kilda, Ailsa Craig, Grassholm, Great Saltee and Little Skellig. Additional samples were collected from these or other colonies (including Norwegian and Canadian colonies) by a range of workers, listed in Acknowledgments. Sampling sites are mapped in Figures 4.1 and 4.4 (Chapter 4).

Feathers were stored in mercury-free polythene bags until analysis, at room temperature if the samples were completely dry. Any damp feathers were stored in a deep freeze until preparation for analysis.

1.4 Pre-treatment and mercury analysis of samples

1.4.1 Feathers

Before analysis, any gross surface contamination in feathers was removed by rinsing feathers in chloroform, acetone and water. Samples from each bird were placed in individual glass test-tubes, to which sufficient chloroform was added to cover the feathers. The tubes were then placed in an ultrasonic bath and washed for ten minutes. This procedure was then repeated, following which the tubes were placed in an incubator at 50^0 C for one hour, to evaporate off excess chloroform. Samples were then ultrasonically washed in acetone for ten minutes, followed by three successive five-minute rinses in water. Samples were left to dry in an incubator at 50^0 C for 24 hours, allowing feathers to reach a relatively constant weight. Tubes containing the feathers were then individually sealed with clingfilm

(to reduce absorption of water from the air), and allowed to cool before feathers were weighed to 0.01 g on a Mettler balance.

All glassware used for analysis or storage of samples was thoroughly rinsed in cold and hot water after each set of mercury analyses was completed, and then soaked (usually overnight) in Decon commercial solution. After further rinsing, glassware was dried at high temperatures in an incubator.

The mercury-analysis method used was cold-vapour, ultra-violet absorption spectrophotometry, using the methods of Furness et al. (1986) and Muirhead (1986).

In general, thirty individual feather samples were analysed in any two-day session. Weighed samples, usually of 0.08-0.13 g of dry feather, were placed in Kjeldahl flasks, to which 4 ml of sulphuric acid and 1 ml of nitric acid were added, to digest the sample. Flasks were placed in racks, and partly immersed in a water bath at 50-55^0 C, to speed digestion. After digestion was complete (clarified acid solution), 15 ml of potassium permanganate were added in 3 ml amounts, and samples were placed, still in tubes on racks, in a freezer overnight, to reduce 'frothing.' The following day, hydrogen peroxide was added to dissolve any precipitate, and water was added to bring the sample up to 25 ml. A 1 ml subsample was then added to a Dreschel flask containing 25 ml of acid permanganate (2% potassium permanganate and 50% sulphuric acid in equal volumes). Mercury was driven off by addition of 10 ml of stannous chloride solution (stannous chloride in 50% hydrochloric acid in a 1g/5ml ratio), and passed through a Data Acquisition Ltd. DA 1500.DP6 Mercury Vapour Detector. Calibrations were made using 50 ng or 100 ng standard mercuric nitrate solutions.

In some cases, replicate readings were taken for each sample, at different times during the procedure (i.e. thirty samples each analysed once, then analysis repeated). Throughout the period of analysis, blank readings were taken at regular intervals and subtracted from the Detector readings. Readings were converted to ng of mercury, and the figure for each sample was divided by the dry weight of sample to give mercury concentration (ug/g).

Precision of analyses was high, based on replicate analyses (which were usually within ±5% of their midpoint). Accuracy was assessed by analysis of standard horse kidney Reference Material H-8 from the International Atomic Energy Agency. The average of 22 subsamples analysed was 0.88 ug/g dry weight (± s.d. 0.09), which fell well within 95% confidence limits for accepted determinations.

1.4.2 Eggs

Egg contents (albumen and yolk) were homogenised by compression in a 50-100 ml plastic syringe, through which the tissue was repeatedly forced. This reduced the contents to a uniform consistency. About 5 ml of each homogenised sample were pipetted into a clean glass vial of known weight, and the egg contents were dried to a constant weight (incubator at 50^0 C for one week). The dried contents of each vial were, when almost dry, ground into a gritty powder using a glass rod, before drying continued. When dry, samples were sealed to prevent absorption of water before analysis.

Weighed samples of ca. 0.10 g were acid-digested, and their mercury concentrations determined, as for feathers (see above).

Chapter 2

THE USE OF SEABIRDS AS MONITORS OF HEAVY METALS IN THE MARINE ENVIRONMENT

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This chapter has been published in Furness, R.W. & Rainbow, P.S. (1990). Heavy Metals in the Marine Environment, CRC Press, Boca Raton, Florida, pages 183-204. I am the sole author of this chapter. The chapter is presented in the format required by the publisher.

I. INTRODUCTION

In attempts to quantify geographical variations in heavy metal levels in marine environments and assess the impact of anthropogenic inputs of metals, analysis of levels in biological indicator organisms offers several advantages over seawater or sediment analysis.[1] The choice of indicator depends on many variables, such as ability to regulate tissue concentrations or tendency to accumulate high concentrations of specific metals; macroinvertebrates are generally favored (see also Chapter 6).[1,2]

Although seabirds may average out ("integrate") very localized or short-term variations in contaminant levels of marine food chains, and in some cases (notably organochlorines and mercury) accumulate higher tissue concentrations than in food items, they may offer no overall advantage over other marine biological indicators for metal analysis.[3] However, seabirds have a high conservation profile, and mercury has been implicated as a cause of mortality or of reduced breeding success in terrestrial, freshwater, and coastal populations of birds.[4-8] Mercury concentrations in eggs and body tissues of seabirds have thus been analyzed on a large scale since the mid 1960s, particularly in the northern hemisphere,[9-19] and levels of cadmium, lead, copper, zinc, and other metals are increasingly being measured in seabirds.[20-31]

Most studies have not been specifically designed to quantify environmental variations in metal levels but are primarily concerned with possible adverse effects of elevated metal levels on seabirds. However, a few studies have specifically aimed to use seabirds as indicators of geographical variation,[32-37] while seabird feathers have been used for assessment of historical variations in mercury inputs to the sea.[38,39] The establishment of baseline concentrations of metals for seabird tissues is difficult, given geological, dietary, taxonomic, and other likely influences on 'natural' levels, but there is a growing database of metal levels in populations remote from sources of localized pollution.[28,30,31,40-42]

The present review aims primarily to draw attention to factors which need to be taken into account in designing programs to monitor geographical or temporal variations in marine levels of heavy metals using seabird tissues, or in interpreting existing work.

II. METABOLIC REGULATION OF METALS

Of the metals most often analyzed in avian tissues, copper and zinc are biologically essential, with important roles in, for example, enzyme function.[43] Seabird data support the idea that tissue concentrations of these metals are regulated (see also Chapter 9).[10,20,30] Copper concentrations are relatively constant between species and populations, with arithmetic means ranging from 2.3 to 18.5 (mainly 4 to 8) μg/g wet weight in liver and 1.0 to 7.8 (3.5 to 6.5) μg/g in kidney.[28,32,40,42] Similarly, zinc concentrations vary only 19 to 110 (mainly 30 to 50) μg/g in liver, 18 to 78 (25 to 50) μg/g in kidney.[24,32,42]

Further evidence for regulation of copper and zinc is provided by relatively low intrapopulation variation (Table 1).[20] Tissue concentrations of environmental contaminants are often not normally distributed in population samples,[44,45] with arithmetic means disproportionately influenced by a few individuals exhibiting high concentrations. However, copper and zinc tend to be normally distributed in seabirds,[20,46] as evident from comparison of means with the meager data available on median concentrations (Table 1).

For mercury and lead, coefficients of variation (CV) and skewness tend to be high (Table 1),[46] implying a lack of regulation, and concentrations vary widely between species and populations (see also Chapter 9).[42] Mean concentrations of mercury in adult seabirds range from 0.06 to 268 μg/g wet weight in liver (Figure 1),[31,41] at least 0.06 to 5.4 μg/g in kidney,[41,47] and 0.05 to 11.1 μg/g in egg contents.[19,48] More limited data for lead indicate mean concentrations <0.01 to 5.3 μg/g in liver and <0.01 to 2.1 μg/g in kidney.[41,49]

TABLE 1
Coefficients of Variation (100 SD/Mean) and "Skewness Ratios" (Arithmetic Mean/Median) for Metal Concentrations in Seabird and Wader Tissues (Averages of Individual Species Means)

Coefficient of Variation

Species	Location	Tissue	Cd	Cu	Hg	Pb	Zn
14 Seabird species[31]	Gough Island	Liver	38	27	57	—	21
		Kidney	30	26	—	—	18
2 Wader species[45]	Netherlands	Liver	202	21	—	105	16
		Kidney	114	27	—	267	10
Brown pelican[14,54]	South Carolina	Egg	53	17	37	78	14

Skewness ratio

14 Seabird species	Gough Island	Liver	0.99	1.05	1.19	—	1.02
		Kidney	1.04	1.02	—	—	1.00
2 Wader species	Netherlands	Liver	1.73	1.08	—	1.88	1.02
		Kidney	3.18	1.03	—	1.23	1.00
Brown pelican	South Carolina	Egg	1.31	0.96	1.02	1.42	1.01

Cadmium concentrations also vary widely between populations, with means <0.1 to 32 μg/g wet weight in liver and 1.5 to 138 μg/g in kidney (Figure 1).[31,49,50] However, intraspecific variation and skewness tend to be less than for mercury and lead in seabirds, though not, apparently, in waders (Table 1) or seaducks,[51,52] despite lower concentrations in the latter groups. It may be that, as an evolutionary consequence of long-term exposure to proposed high 'natural' levels of cadmium in oceanic food chains,[21,53] seabirds have some capacity to regulate their tissue levels of cadmium.

Available data for other metals imply metabolic regulation of the essential metals manganese and iron (e.g., coefficients of variation 12 to 27%, skewness ratios of 1.00 to 1.04 in waders),[45,49] but not of arsenic, chromium, and nickel (e.g., CVs 69 to 157%, skewness 1.2 to 1.79 in brown pelican *Pelecanus occidentalis* eggs).[14]

Metabolic regulation decreases the potential for detecting variations in environmental levels of essential metals using seabirds as sampling agents.[28] However, regulatory ability may not always be sufficient to cope with acute local pollution, allowing some potential for monitoring,[50] at least qualitatively. Such a 'breakdown' of regulation might be evident as an increase in skewness and intrapopulation variation of metal concentrations.

At the other extreme, marked geographical trends may be seen for tissue concentrations of some toxic metals despite apparent evidence for regulation. For example, mercury can show low intrapopulation variability and skewness in seabird eggs (Table 1) and nestling feathers.[35,37] In such cases, it may be that the tissue or age-class is a particularly good indicator of localized or short-term exposure to metals (see Sections IV and VI), and thus less likely to reflect temporal or spatial heterogeneities in exposure within a population.

III. CHOICE OF SPECIES: TAXONOMIC AND ECOLOGICAL FACTORS

An ideal seabird monitor of heavy metal pollution should be resident within a restricted radius of its breeding colony throughout the year. Its diet should be well known and should show no systematic variation between sampling locations; species feeding on few prey species are particularly suitable. However, the possibility that seasonal movements of prey species

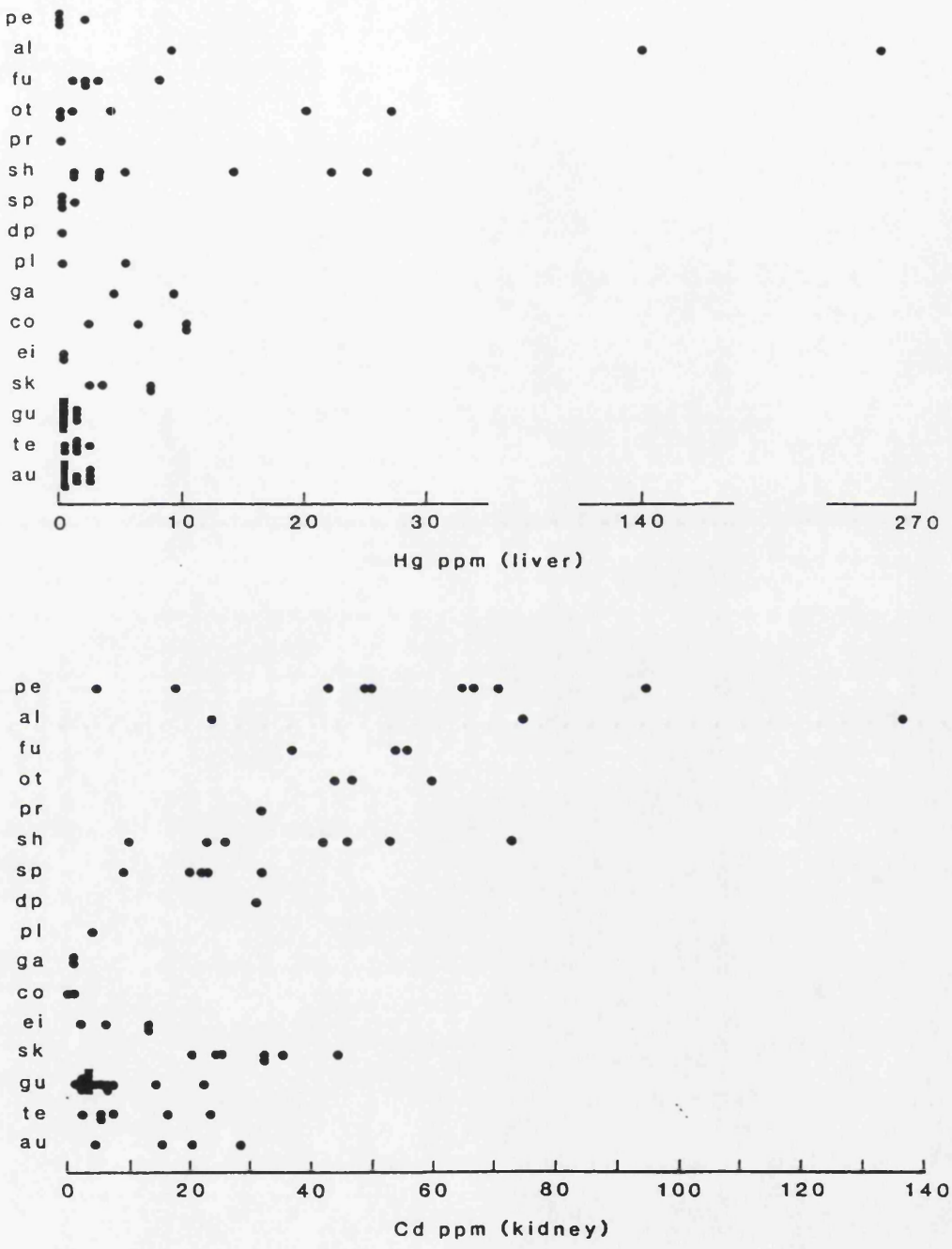

FIGURE 1. Variation of liver mercury and kidney cadmium concentrations between seabird taxa (ppm = parts per million or μg/g wet weight). Each point represents an arithmetic population mean based on 2+ individuals (small square = 2, large square = 4 means). Pe = penguins (Spheniscidae), al = albatrosses (Diomedeidae), fu = fulmars *Fulmarus*, ot = other tubenoses *(Pagodroma, Pterodroma)*, pr = broad-billed prion *Pachyptila vittata*, sh = shearwaters *(Puffinus, Calonectris)*, sp = storm petrels (Hydrobatidae), dp = common diving petrel *Pelecanoides urinatrix*, pl = brown pelican, ga = northern gannet *Sula bassana*, co = cormorants *Phalacrocorax*, ei = common eider *Somateria mollissima*, sk = skuas *Catharacta*, gu = gulls *(Larus, Rissa)*, terns *Sterna* and auks (Alcidae). Data from References 11, 14, 15, 17, 19—21, 23—33, 40, 41, 46, 47, 49, 51, 57, 59—64, 95.

could affect metal levels should be noted.[3] Furthermore, reliability of metal analyses, and ability to discriminate environmental variation, will be improved if the seabird species, or their prey, tend to accumulate relatively high tissue concentrations of the specific metal. The seabird species should also be relatively common and accessible,[3] and sufficiently widespread to provide good coverage of the proposed region being monitored for geographical variation.

Most seabirds meet only some of the above criteria. For many seabirds, however, diet, seasonal movements, and molt are reasonably well known,[38] so some allowance can be made for influences of these variables. Since different species tend to feed at varying distances from land, it is also possible in theory to select species which will reflect metal levels in either inshore or offshore waters.[3] Sampling at the breeding colony narrows down the sampling radius, particularly in the case of eggs or nestling tissues, which more closely reflect metal uptake from local foraging than do tissues of adults.[13,15,18,37] For example, eggs of common murres *Uria aalge*, which feed on relatively few species of small fish, [55] seem to be particularly good indicators of coastal variations in mercury levels.[13]

Variations in heavy metal concentrations between seabird species potentially reflect many factors, including feeding and migratory habits, body size, lifespan, molt strategy, and taxonomic influences on physiology. Inferences about geographical variations in environmental metal levels based on tissue analyses of different species are therefore open to question, though interspecific comparisons may shed light on relative metal loads in different food chains. Apparent tendencies for particular taxonomic or ecological groups to accumulate high levels of particular metals may also recommend suitable indicator species.

Figure 1 summarizes most of the published work on mercury and cadmium concentrations in liver and kidney tissues, respectively, of adult seabirds. For mercury, much of the variation reflects the well-known tendency for concentrations to be highest in species feeding on fish (or on other seabirds).[11,12,15,25,35,47] However, markedly high concentrations have been found in some procellariform species, notably albatrosses and gadfly petrels, from Gough Island,[31] where no association with diet was evident. In the latter case, a possible influence of molt-related or other constraints on mercury elimination was suggested.[31] Marked interspecific variation is also evident for cadmium, with a tendency for concentrations to be highest in species feeding on pelagic invertebrates, and in skuas feeding on pelagic seabirds.[3,21,25,30,31,41,56] For both cadmium and mercury, pelagic seabirds might seem ideal candidates for assessment of possible global oceanic pollution. However, difficulties posed by such factors as age accumulation (Section VI) and, in the case of cadmium, the apparent lack of a suitable tissue for historical analysis,[39] would need to be overcome.

Interspecific variation in zinc concentrations is comparatively slight, and in general follows the trends shown by cadmium (Section VII). For copper, the only notable feature is the occurrence of concentrations of 100 to 270 μg/g wet weight or more in liver tissue of common eiders from northern Europe,[3,30,51,57] 20 to 30 times higher than in "true" seabirds. Copper concentrations also tend to be high in other duck species,[52,58] though nowhere near as high as in eiders. It would appear either that eiders are physiologically unable to regulate their liver concentrations of copper, or that their dietary uptake of copper exceeds their regulatory capacity. Among wholly marine or coastal birds, eiders may thus be the best potential indicator of coastal pollution by copper.

The highest concentrations of lead occur in inshore or estuarine seabirds, notably laughing gulls *Larus atricilla*,[22,33] though this may partly reflect a bias towards sites known or suspected to be lead polluted. Interspecific differences for other metals, including silver, arsenic, chromium, iron, manganese, and nickel, are unclear in view of the paucity of data, and possible variations in analytical reliability.[14,20,23,32,41,57,59]

IV. VARIATION OF METAL CONCENTRATIONS BETWEEN TISSUES

Table 2 summarizes published data on relative concentrations of mercury, cadmium, lead, copper, and zinc in the most frequently analyzed tissues of adult seabirds. Such tissue/tissue ratios, although highly variable between studies, indicate the tissues which tend to accumulate high concentrations of particular metals and which may thus be 'good' indicators of metal uptake from the environment. The latter assumption has not been tested critically in seabirds, but various studies have looked for parallel variation of metal levels in different seabird tissues.[23-25,30,46,59,65,66] However, many multiple-tissue studies have made little attempt to investigate intertissue correlations, and no quantitative information is available on the relationship between concentrations in particular seabird tissues and the total metal burden in those tissues or in the whole body.

Tissues tending to have low concentrations could still reflect environmental variations, but analytical difficulties may be encountered. Also, such low concentrations might imply a saturation of metal-accumulatory ability. Conversely, some tissues may be more prone to the influence, and potential bias, of age accumulation of metals (Section VI), though it is sometimes possible to choose a specific tissue to reflect either long-term or recent exposure.[52,67-69]

Note that in Table 2, the water-content figures used for any necessary wet-weight/dry-weight conversions are very approximate, and may vary between studies,[19,25,51,59] for analytical, physiological, or taxonomic reasons. The figure for 10% water content of feathers may be a good average for full-grown feathers, but will vary with collecting conditions and will often be higher in chicks: the use of surface-cleaned,[70] dried samples is recommended. A 10% water content in bone is rather notional, and will depend on the degree of adhering soft tissue. Intertissue comparisons of metals on a wet-weight basis are rather meaningless, however, except in terms of ease of analysis. Wet-weight comparisons between eggs of different species, e.g., in attempts at toxicological assessment, are also questionable, as the water content tends to be highest in species hatching altricial (naked, poorly developed) chicks.[71] For example, fresh eggs of northern gannets and common eiders averaged 83.7 and 66.4% water, respectively, in Canadian samples,[19] representing a two-fold difference in relative dry-tissue content.

Compared to inorganic mercury, which accumulates mainly in the kidney in experimentally dosed animals, methylmercury distributes more evenly between the soft tissues.[67,72,73] Tissue concentrations in seabirds fit this pattern, with a liver/kidney ratio close to unity, although liver concentrations are usually highest. Liver and kidney concentrations are often significantly correlated,[25,59,66] and this correlation is also evident between populations (Table 2 data). However, kidney tissue alone might provide an underestimate of exposure, as the liver/kidney ratio tends to be higher at high liver and kidney concentrations of mercury. Liver and pectoral muscle concentrations are also significantly, but more variably, correlated between populations, again with some evidence of an exponential increase in liver concentration. The apparent position of liver as the best single soft tissue to indicate mercury exposure is further supported by the high degree of average correlation with other tissue concentrations in nestling common terns *Sterna hirundo*.[66]

Mercury shows a particularly strong tendency to be incorporated in feathers, becoming firmly bound to keratin during feather growth. Once growth is completed, feathers are physiologically isolated from other tissues, and their mercury content is remarkably resistant to further change under the influences of heat, ultraviolet light, and other forms of weathering,[74] with little evidence for external contamination under field conditions.[75] Additional advantages of feathers as mercury monitors are the tendency for concentrations to be high relative to other tissues and the ease with which live birds can be sampled.

TABLE 2
Between-Tissue Ratios of Metal Concentrations in Seabird Adults

			Ratio							
			l/k[a]	l/pm	l/br	l/bo	l/f	k/l	bo/l	f/l
Hg	md, n[b]	d[c]	1.12, 18	4.9, 19	6.4, 11	21, 6	0.35, 25	0.88, 18	0.05, 6	2.8, 25
	range		0.69—4.8	1.0—18.5	1.3—21.8	5.4—36	0.15—95	0.20—1.4	0.03—0.2	0.01—6.6
	md, n	w[c]	1.35, 18	5.9, 19	9.0, 11	7.0, 6	0.12, 25	0.73, 18	0.14, 6	8.5, 25
	range		0.82—5.8	1.1—22.2	1.8—30.4	1.8—12.0	0.05—32	0.17—1.2	0.08—0.6	0.03—20
Cd	md, n	d	0.17, 62	13.0, 26	6.1, 14	19.1, 14	>25.1, 7	5.9, 62	0.06, 14	<0.04, 7
	range		0.03—0.3	1.9—108	0.9—>276	0.97—577	2.7—304	1.9—35.2	0.002—1	0.003—0.4
	md, n	w	0.20, 62	15.6, 26	8.5, 14	6.4, 14	>8.4, 7	4.9, 62	0.17, 14	<0.11, 7
	range		0.03—0.4	2.3—130	1.2—>385	0.32—193	0.89—101	1.6—29.3	0.01—3.1	0.01—1.1
Pb	md, n	d	0.89, 12	1.44, 9	0.92, 11	0.97, 8	1.79, 4	1.12, 12	1.31, 8	0.57, 4
	range		0.01—2.1	0.87—3.3	0.04—3.3	0.37—3.1	<0.11—3	0.47—69	0.33—2.7	0.33—>9.3
	md, n	w	1.06, 12	1.73, 9	1.29, 11	0.32, 8	0.60, 4	0.93, 12	3.9, 8	1.70, 4
	range		0.02—2.5	1.04—4.0	0.53—3.4	0.12—1.0	<0.04—1	0.39—58	0.98—>7	1.0—>28
Cu	md, n	d	1.04, 36	1.39, 17	2.6, 3	2.4, 5	1.66, 3	0.95, 36	0.43, 5	0.60, 3
	range		0.55—2.5	0.39—17	2.41—3.8	1.65—22	1.09—5.9	0.40—1.8	0.04—0.6	0.17—0.9
	md, n	w	1.25, 36	1.67, 17	3.7, 3	0.78, 5	0.55, 3	0.79, 36	1.28, 5	1.81, 3
	range		0.66—3.0	0.47—20	3.36—5.3	0.55—7.5	0.36—2.0	0.33—1.5	0.13—1.8	0.51—2.7
Zn	md, n	d	0.81, 38	2.4, 19	2.1, 12	0.90, 10	1.54, 7	1.26, 38	1.12, 10	0.67, 7
	range		0.49—1.3	1.13—6.3	1.10—6.3	0.45—1.5	0.33—3.7	0.75—2.0	0.64—2.2	0.26—3.0
	md, n	w	0.98, 38	2.9, 19	2.9, 12	0.30, 10	0.49, 7	1.05, 38	3.3, 10	2.0, 7
	range		0.59—1.6	1.36—7.6	1.53—8.8	0.15—0.5	0.11—1.2	0.62—1.7	1.94—6.6	0.80—9.0

Note: Data from References 20, 22—29, 31—33, 41, 46, 47, 49, 51, 59, 65.

[a] Tissues = liver (l), kidney (k), pectoral muscle (pm), brain (br), bone (bo), feather (f).

[b] md = median, sample size, and range of population mean ratios.

[c] w = wet, d = dry tissue; conversions assume water content of kidney and muscle 75%, liver 70%, brain 78.5%, feather 10%, bone ?10%.

Although feathers have been shown experimentally to incorporate mercury in a dose-dependent fashion,[67] the relationship of feather to other tissue concentrations in seabirds is poorly documented. In part, this reflects the paucity of multitissue studies, but it also reflects the great variation of mercury concentrations among feathers of individual birds. Feathers replaced early in a molt cycle incorporate the highest concentrations of mercury, newly mobilized after accumulation in soft tissues between molts.[47,70] This is most strikingly shown by the tendency for mercury concentrations to decrease linearly along such feather sequences as the primaries, corresponding to the order in which feathers have been dropped and renewed.[47,70,76] Thus comparisons between studies are greatly complicated unless feathers from a similar position in the molt sequence are analyzed. However, suggestions that the mercury exposure of individual migratory birds on their wintering and breeding grounds can thus be compared, on the basis of where a particular feather was grown,[76,77] are based on the erroneous assumption that feather concentrations largely reflect recent dietary intake.

For environmental monitoring using feathers of adults, the effect of molt-influences on mercury levels of individual feathers can be minimized by pooling small body feathers from a defined plumage area.[70] If nestlings are used, little variation due to growth sequence can be expected; however, part-grown feathers tend to have disproportionately high concentrations of mercury.[37]

Dietary methylmercury is dose-dependently transferred to the contents of the egg,[67] and there is abundant evidence from seabirds of a relationship to environmental exposure.[13,15,16,29,78] Mercury accumulates particularly in the egg-white proteins,[67] which derive from serum proteins,[71] and egg concentrations thus apparently more closely reflect mercury from recent dietary uptake than from accumulated tissue stores.[18,65,69,79] (There is also evidence that the ovalbumin fraction of egg white has a specific affinity for dietary mercury, while the globulin fraction tends to accumulate low levels of "nondietary" mercury.[80]) Eggs may thus provide a particularly good indicator of mercury exposure in the vicinity of a breeding colony in the immediate prelaying season.[18,69]

Cadmium concentrations are invariably highest in seabird kidney, the critical organ for chronic exposure.[25,67] Nevertheless, liver may be a better indicator of recent exposure,[52,68] and may thus be an appropriate organ for short-term, localized monitoring. Liver/kidney ratios >1 indicate acute exposure to high cadmium doses,[67] which could occur in cases of severe coastal pollution. A possible advantage of liver over kidney is that elevated levels of cadmium can, by inducing tubular dysfunction, lead to an actual lowering of kidney concentrations,[67] though this has not been demonstrated in seabirds. Kidney and liver concentrations of cadmium tend to be significantly correlated both between and within populations.[24,30,46,59,62] No clear correlations with other tissues are evident from population means, but significant liver/kidney/pancreas and kidney/duodenum correlations have been noted in Atlantic puffins *Fratercula arctica*.[24]

Information on cadmium concentrations in seabird feathers is conflicting, with population means ranging from 0.02 to 27μg/g and little evidence of a relationship to other tissue levels.[24,32,41,56,65,81] The highest concentrations have been noted in pelagic seabirds (notably blue-gray ternlets *Procelsterna cerulea*),[56] but some pelagic populations have shown very low concentrations.[24,65] Nevertheless, the potential use of feathers in assessing historical variation in exposure to cadmium, and the possible influences of external contamination and molt sequence, merit further study.

Experimental studies showing little transfer of cadmium to eggs are confirmed by seabird data, with concentrations usually <0.7 (mainly 0.003 to <0.05) μg/g wet weight.[67,14,15,20,27,29,33,35,41,49,62] Such an apparent barrier to cadmium transfer might break down if other tissues became overloaded,[25] possibly accounting for the anomalously high concentrations reported in one study of sooty tern *Sterna fuscata* eggs from Hawaii (75.0 μg/g).[26] There is limited evidence that cadmium concentrations in egg shells may be relatively

high, with means of 1.39 and 1.75 μg/g recorded for two laughing gull populations on the Texas coast (vs. <0.028 μg/g in egg contents).[33]

Although recorded lead concentrations vary rather little between seabird tissues, the avian kidney tends to accumulate higher levels than other soft tissues and may thus be a good indicator of recent exposure to lead.[67] However, it has been suggested that liver may be a better short-term monitoring tissue,[52] and that increased liver/kidney ratios may indicate recent acute exposure.[82] Current or recent exposure can also be monitored by lead concentrations in blood, most readily assessed by indirect techniques based on inhibition of certain enzymes by lead.[67] In birds, measurement of aminolevulinic acid dehydratase activity is particularly reliable,[67] but has not been applied to seabirds.

Lead accumulates most strongly in bone, which thus provides probably the best measure of lifetime or chronic exposure.[67,83] However, for assessment of localized or recent uptake of lead, particularly in migratory species, the use of soft tissues is preferable.[22] Although lead concentrations can be high in seabird feathers, interpretation is difficult as there is evidence from waders of heavy contamination by secretory products.[75] However, this still allows some potential for indirect monitoring of lead exposure over known periods. Little lead is transferred to eggs, which have invariably low concentrations in seabirds (usually <0.4 μg/g), often below detection limits.[20,22,26,29,33,49,54,81]

Copper concentrations tend to be highest in seabird kidney and, especially, liver. Significant liver/kidney correlations have been noted in individual studies,[30,59] and are also evident between species and populations. Common eiders are exceptional in showing a marked (8-fold to 17-fold) excess of liver over kidney concentration.[30,57] No liver/pectoral muscle correlation is evident between populations, but the liver/muscle ratio increases with increasing liver concentration. Concentrations in seabird eggs are comparatively low (means 0.15 to 1.8 μg/g).[15,20,34,41,81]

Zinc concentrations are relatively high in kidney, bone, liver, and feathers. There is a significant liver/kidney correlation between seabird populations, and at Gough Island kidney/liver ratios of zinc tended to be highest in species having high kidney concentrations of cadmium.[31] Zinc concentrations are lower in pectoral than in femoral and gastrocnemius muscle, probably reflecting differences in the content of zinc metalloenzymes.[41,46] Pancreatic concentrations are also high,[24,59] and correlate with feather levels in brown pelicans.[59] In waders, the zinc concentration of feathers is apparently 'fixed' in the shaft and highly variable in the vane, but how this variation relates to zinc exposure is unclear.[75] Mean concentrations of zinc in seabird eggs range from 1.5 to 22 μg/g.[15,20,29,34,41]

A significant feather/liver correlation of arsenic concentrations has been noted for estuarine gulls in New Zealand,[23] but concentrations may only be meaningful in feathers of known age, in view of external, probably secretory, contamination noted in waders.[75] Feather concentrations of iron, manganese, and nickel are poorly documented, though nickel levels may be comparatively high.[41,84] Concentrations of chromium in bone tissue of Antarctic and Pacific procellariform species exceeded liver and pectoral muscle concentrations in the ratio 5/2/1 (dry-weight terms),[20] and concentrations may also be high in the kidney and salt glands.[14,32] Liver tends to have the highest concentration of iron,[32,41,57] while manganese concentrations tend to be highest in liver, salt-gland, pancreas, and bone.[32,41,49] Silver, cobalt, and nickel dry-weight concentrations in bone/liver/pectoral muscle followed approximate ratios of 60/1/2, 12/1/1, and 3/2/1, respectively, in several procellariforms,[20] while common eiders can accumulate disproportionately high levels of silver in liver.[57] In seabird egg contents, mean concentrations of 0.13 to 0.30 μg/g have been noted for arsenic,[14,34] 0.01 to 0.55 for chromium,[20] 5.8 to 6.0 for iron,[41] 0.04 to 1.2 for manganese,[41,49] and 0.02 μg/g for nickel.[14]

V. PHYSIOLOGICAL AND SEASONAL INFLUENCES

Insufficient attention is often paid to possible physiological or seasonal influences on the metal content of seabird tissues.[85] Seasonal changes in tissue concentrations of essential metals, notably zinc, have been documented in house sparrows *Passer domesticus*,[86] starlings *Sturnus vulgaris*,[87] and waders.[3] Some such changes are associated with molt and can be accompanied by elevated concentrations of nonessential metals (e.g., cadmium and mercury). Seasonal variation in fat or protein content of tissues, or in enzyme activity, may also be important.[24,87] For example, an increase in metalloprotein content may increase the number of binding sites available to essential metals (e.g., zinc) and their toxic competitors (e.g., cadmium).[24,88] The most pessimistic view is that metal concentrations in tissues "depend as much on physiology as they do on exposure."[85]

In Atlantic puffins at a colony in southeast Scotland,[85] liver mercury concentrations declined significantly from a mean of 1.73 μg/g dry weight in March (prelaying) to 0.59 μg/g in July (chick rearing), kidney cadmium from 15.5 to 4.48 μg/g. No change was seen for zinc, probably reflecting only partial coverage of the annual cycle (excluding molt, when peak concentrations would be expected).[85] A proposed peak of cadmium during early to mid molt may have contributed to differences in cadmium concentrations between three pelagic seabird species at St. Kilda, sampled in June but molting at different seasons.[24]

Fairy prions *Pachyptila turtur* in Tasmania showed no variation in cadmium or zinc concentrations between breeding and nonbreeding seasons,[36] although tissue concentrations of copper and lead differed significantly. A significant difference in zinc and iron concentrations in greater crested terns *Sterna bergii* between two New South Wales colonies was considered an artifact of sampling date, reflecting seasonal declines.[32]

Seasonal variation in mercury concentrations, largely reflecting mobilization and incorporation into plumage,[70] is well known, but not always taken into account when 'soft-tissue' levels are presented. Body burdens of mercury (excluding plumage) in Bonaparte's gulls *Larus philadelphia* declined by 93% during autumn molt in southeast Canada.[89] Average body burdens in Japanese black-eared kites *Milvus migrans* declined by somewhere in the range 44 to 53% during molt, and concentrations in liver by up to 64%, in kidney by up to 59%, and in other tissues by 28 to 78%.[90] Soft-tissue comparisons between populations of adult seabirds are thus ideally based on samples collected immediately before molt, when mercury concentrations are highest and least variable.

Mercury incorporated into feathers is stably bound, and concentrations in individual feathers thus remain constant.[74,89] However, given that mercury concentrations tend to be highest in feathers renewed early in a molt sequence,[47,70] the average mercury concentration of the plumage can, in theory, alter. If individual feathers tend to be replaced at the same stage of each molt period, average concentrations will be reduced after such high-mercury feathers are dropped but before they are regrown. As molt proceeds, this effect may be reversed as high-mercury feathers are replaced and low-mercury feathers dropped. While the occurrence and potential magnitude of such effects remain to be confirmed, it is likely that feather mercury concentrations will be most consistent in samples taken outside the molt period.

No significant correlation between mercury concentration in eggs and season date was found for common terns in Ontario, but mercury in breast muscle of five adults increased significantly with date during the breeding season.[69] Since egg proteins are largely derived from blood serum,[71] this discrepancy was taken to imply an equilibrium between mercury in eggs (but not muscle), blood, and diet.

Egg laying had little influence on metal concentrations in female Adélie penguins *Pygoscelis adeliae* in Antarctica.[41] Efficiency of transfer to the egg was greatest for mercury, but the total egg content was negligible relative to adult burdens. Females had higher zinc

concentrations in bone than males; zinc concentrations are also highest in female house sparrows, and peak during egg laying, apparently related to alkaline phosphatase activity associated with calcium mobilization.[86] Male Adélie penguins took the first, about 20-d incubation shift, and muscle concentrations of iron appeared to decline during this starvation period, while liver concentrations increased, implying redistribution of iron. This probably contributed to the higher iron concentrations in livers of males (987 μg/g wet weight) than of females (478 μg/g).[41]

Although evidence from seabird studies is lacking, sex differences in tissue (especially bone) concentrations of lead can be expected. Reproductively active female mallard *Anas platyrhynchos* and Barbary doves *Streptopelia 'risoria'* accumulate lead more rapidly than males, resulting in order-of-magnitude differences in bone concentrations of experimentally dosed birds.[67] Increased synthesis of calcium-binding protein during eggshell formation apparently increases intestinal absorption of both calcium and lead.[67,91]

Possible influences of starvation should be taken into account in metal analyses of seabirds found dead. Even if a metal "does not appear to be freely mobilized in starved birds" (mercury in liver),[11] changes in tissue weight may alter metal concentrations without altering tissue burdens. Thus in northern gannets found dead around British coasts, a twofold difference in liver concentrations of mercury and zinc between east and west coasts was largely offset by a twofold difference in liver weights, reflecting the emaciated condition of corpses in the west coast sample.[10,15] Stress associated with slow death from injury or starvation could also influence metal concentrations. For example, lead, which follows calcium during bone formation,[67] might be released with calcium during stress, as known for mammals.[83]

VI. AGE-RELATED VARIATION

Variations in the age composition of a sample of breeding or other 'adult' seabirds could potentially contribute to apparent differences in tissue concentrations of metals between species, populations, or years. If a species can be aged at all using morphological characters, this is usually possible only in the first few, mainly nonbreeding, years of life. Sampling of known-age adults, therefore, usually requires a population of birds which have been banded as chicks.

The metal levels of few such populations have been studied, greatly limiting information on possible accumulation of metals over adult life. In 13 3- to 12-year-old breeding great skuas *Catharacta skua* from Shetland, cadmium concentrations showed statistically significant positive correlations with age in kidney, liver, and pectoral muscle.[25,62] This was considered to reflect the long biological half-life of cadmium and its binding to a metallothionein-like protein and subsequent retention in the kidney.[25] Mercury concentrations in primary feathers, kidney, and liver also increased with age in great skuas,[62] though nonsignificantly.[25] In fact, both metals showed great variation between birds of the same age. Moreover, a further analysis of cadmium and mercury in liver, kidney, and muscle of Shetland great skuas found only weak age-accumulatory tendencies, only that for cadmium in muscle (n = 27 adults) being significant.[92,93]

No age accumulation of cadmium, mercury, or zinc was detected in 2- to 11-year-old herring gulls *Larus argentatus* from southeast Scotland.[25,46] For cadmium, this was thought possibly to reflect a lower dietary exposure than in great skuas.[46] Recaptures of black guillemots *Cepphus grylle* in the Baltic Sea likewise indicated no systematic variation of (plumage) mercury concentrations with age.[38] It may be that elimination of mercury into feathers during molt,[70] almost entirely as methylmercury,[39] is usually efficient enough to prevent age accumulation of this particularly toxic form. However, there is also evidence for demethylation of methylmercury by seabirds and subsequent storage of inorganic mercury

in the liver.[94] This suggests that age accumulation of mercury is likely to occur in species having particularly high liver concentrations of inorganic mercury, e.g., certain albatrosses *Diomedea* with slow molt patterns.[31,94]

Evidence for accumulation of metals in the first few months or years posthatching is much more readily available. While of little direct relevance to adult seabirds, such studies help elucidate the relative merits of different tissues or age classes as metal-monitors.

Several studies have compared cadmium and lead concentrations in tissues of adult and nestling (or newly fledged) laughing gulls and terns (Figure 2). For cadmium, age accumulation is most marked in kidney, with adult:chick ratios varying from 13 in a laughing gull study to 387 in royal tern.[27,49] Ratios range from 1.9 to 148 in liver and only 1 to 7.3 in other soft tissues, with conflicting evidence for accumulation in bone. Liver and kidney concentrations of cadmium in first-year common eiders from Denmark were about half those in older birds.[51] Age accumulation of lead is most evident in bone, with adult:chick ratios of 1.8 to 152 (Figure 2).[33,49] For both cadmium and lead, discrepancies between studies presumably reflect differences in temporal availability of metals, in physiological responses or, perhaps, in analytical accuracy (especially at low tissue concentrations).

Surprisingly little has been published on mercury levels in chicks. An adult:prefledgling ratio of 13.3 was recorded for liver tissue of great cormorants *Phalacrocorax carbo* in the Netherlands,[95] and adult:first year ratios of 6.9, 6.4, and 10 for pectoral muscle of Baltic great cormorants, common murres, and black guillemots, respectively.[77] Limited data for brown pelicans in the eastern U.S. suggest an adult:nestling ratio of 1.6 to 9.2.[14] Plumage mercury concentrations of northern gannets average about three to five times higher in adults than in well-grown chicks.[37]

Very few data are available for other metals,[14,32] though significant adult:prefledgling differences were recorded in greater crested terns from New South Wales for manganese (ratio 1.3) and iron (2.2) in liver.[32]

To summarize, while metal concentrations may differ considerably between chicks and adults, evidence for gradual accumulation of metals in adults is very limited. It seems likely that most metals reach a dynamic equilibrium in adult seabirds, while others, such as cadmium, and perhaps inorganic mercury, may increase in some tissues of some species throughout adult life. Chicks of known age may thus provide the most reliable indication of between-colony variations in metal exposure, assuming that tissue concentrations fall within the range of analytical reliability. A further advantage, that chicks will reflect dietary uptake within a comparatively restricted radius of a breeding colony, should be noted.

VII. CORRELATIONS BETWEEN METALS, AND EVIDENCE FOR DETOXIFICATION MECHANISMS

Correlations between concentrations of different metals in seabird tissues are of interest for several reasons. Such correlations may indicate antagonistic, 'detoxifying' interactions between certain essential and nonessential metals. Clues may also be provided to the mechanisms by which metals are distributed between tissues and accumulated. In addition, if several metals tend to coaccumulate for physiological or toxicological reasons, only one of those metals may be amenable to monitoring of environmental variation.

There is particularly strong evidence in seabirds for a relationship between tissue concentrations of zinc and cadmium. Significant positive correlations have been noted for kidney and liver tissue (usually both) between individual Cory's shearwaters *Calonectris diomedea*,[29] great skuas, and herring gulls.[25,46] Similar relationships occur both within and between species from Spitsbergen, Antarctica, and Gough Island.[30,31] The zinc/cadmium relationship is particularly strong in kidney, and is summarized in Figure 3 over a range of seabird populations.

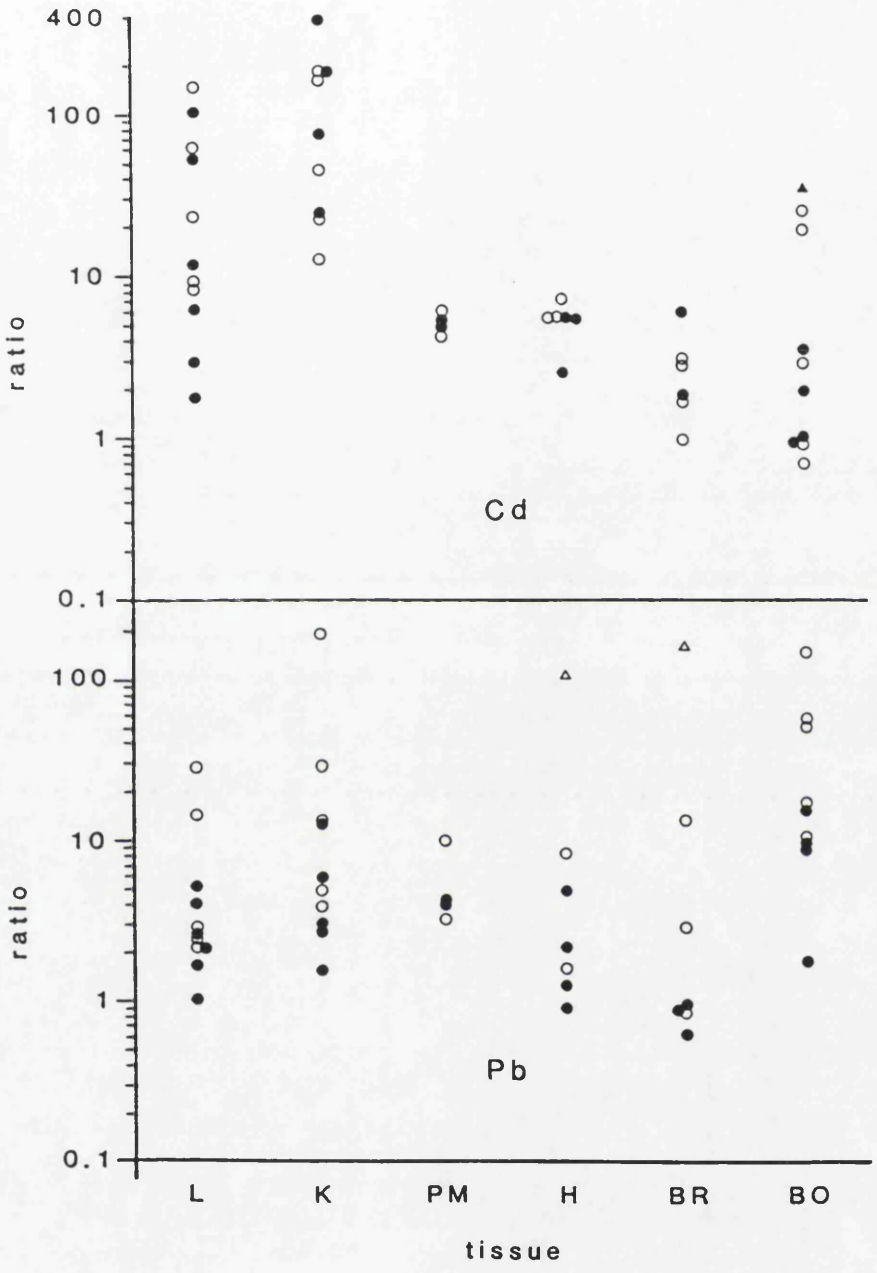

FIGURE 2. Adult:chick ratios of cadmium and lead concentrations (wet weight) in seabird tissues: laughing gulls,[22,33,49] greater crested,[32] royal *Sterna maxima* and sandwich terns *S. sandvicensis*.[27] Filled circles = well-feathered or newly fledged chicks, open circles = downy chicks; triangles = minimum ratio. Tissues: liver (L), kidney (K), pectoral muscle (PM), heart (H), brain (BR), and bone (BO). Note logarithmic scale of y-axes.

Most of the cadmium and zinc in seabird kidney and liver appears to be bound to a low molecular weight, metallothionein-like protein.[25,88] Such proteins can be experimentally induced in mammals and birds by dosing with heavy metals, including cadmium and zinc.[67] Increased synthesis of cadmium-induced metallothionein at the same time increases the number of binding sites available to zinc, thereby accounting for parallel accumulation of

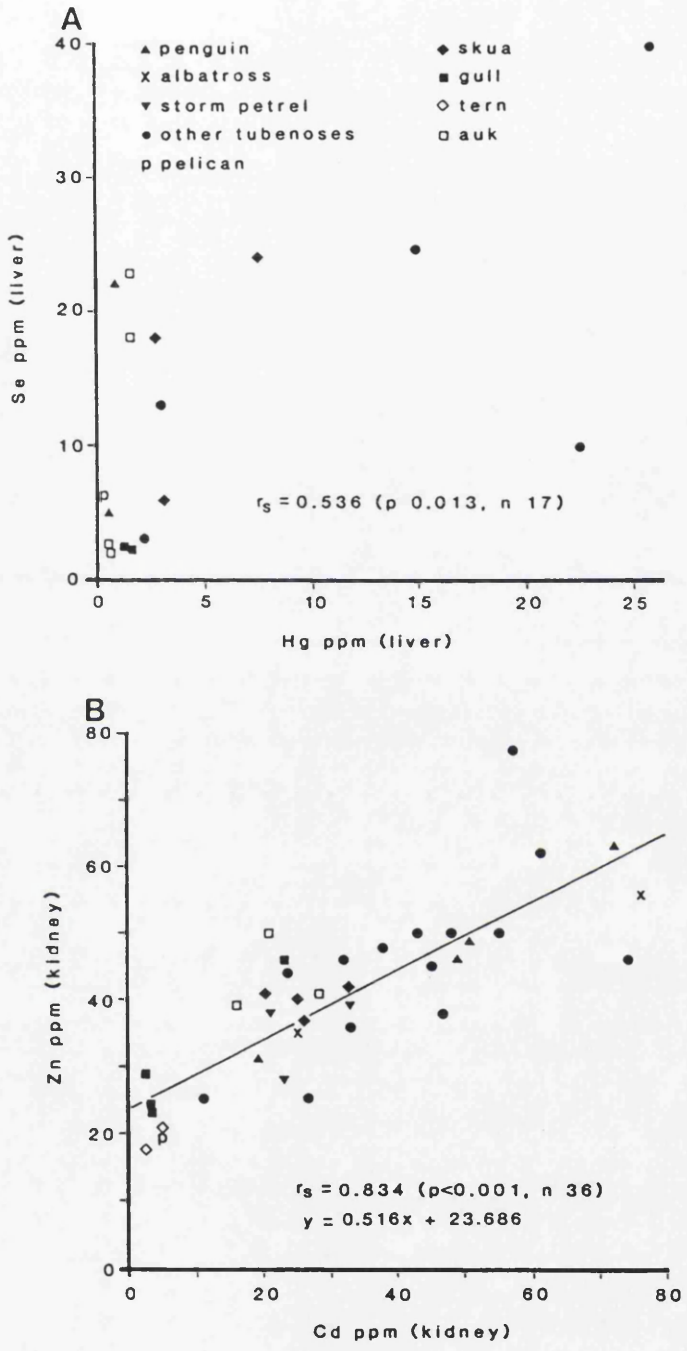

FIGURE 3. Selenium/mercury and zinc/cadmium relationship in liver and kidney tissue, respectively, of seabird adults (ppm = parts per million or μg/g wet weight). r_s = Spearman rank correlation coefficient, n = number of species or population means (based on 4+ individuals) plotted. Other significant ($p < 0.05$) Spearman rank correlations were obtained for: Zn/Cd (liver) all species (n = 52), tubenoses (25); Zn/Cd (kidney) penguins (4), tubenoses (19), skuas/gulls/terns (10), skuas/gulls/terns/auks (12); Cu/Cd (K) skuas to terns (9), skuas to auks (11); Cu/Zn (L) skuas (3), gulls (3), skuas to terns (8), skuas to auks (10); Cu/Zn (K) skuas to terns (6); Zn/Hg (L) all species (41); Se/Hg (K) all species (8); Se/Cd (L) all species (16), skuas (3), skuas to terns (12); Hg/Cd (L) all species (43), skuas/gulls (14), skuas to terns (16), skuas to auks (18). Data from References 20, 23—26, 28—32, 36, 40, 41, 46, 57, 59, 108.

the two metals. There is considerable evidence that, by binding cadmium, metallothionein helps prevent its toxic interactions with other proteins.[67] Nevertheless, kidney damage associated with high concentrations of cadmium has been noted in pelagic seabirds,[96] consistent with the known nephrotoxicity of metallothionein-bound cadmium.[67]

The primary role of metallothionein may in fact be as a zinc store,[97] in which case cadmium binding could represent a toxic interference with zinc metabolism.[88,96] It is also possible that seasonal variation in zinc requirements, e.g., for molt, could result in increased retention of dietary cadmium at particular seasons.[85,88] Zinc itself has a protective influence against various toxic effects of cadmium,[68,98] though the relative importance of zinc-induced metallothionein synthesis and outcompetition of cadmium for other binding sites is unclear.

Cadmium ingestion in experimental mallard can result in increased kidney concentrations of both zinc and copper, possibly related to metallothionein synthesis.[99] Significant correlations of zinc/copper in liver and cadmium/copper in kidney have been reported in brown pelican.[59] However, in Gough Island seabirds and over a range of populations (Figure 3), there is only limited evidence of such a relationship.[31,92]

Although inorganic mercury is known to bind to metallothionein in mammalian kidney,[67] mercury was not associated with the metallothionein-like protein in northern fulmar *Fulmarus glacialis* liver and great skua kidney.[25,88] This was suspected to reflect a preponderance in seabird tissues of methylmercury, which may not be able to bind to metallothionein.[46,85]

Methylmercury does in fact often comprise the bulk of mercury in seabirds, e.g., up to 97% in liver.[24,30,47] Being lipid soluble, unlike inorganic mercury, methylmercury can readily cross the 'blood/brain' and other membrane barriers, and is thus particularly toxic to the central nervous system.[67] Demethylation to the less toxic inorganic form has been shown to occur in experimental mammals,[100] and has also been inferred in marine mammals.[42] In South Polar skuas *Catharacta maccormicki* and a range of Gough Island seabirds, the percentage of mercury present as methylmercury is negatively correlated with the total mercury concentration in liver tissue.[30,94] This may indicate that seabirds are capable of demethylating organic mercury,[94] as has been suggested for freshwater and predatory species,[73,101] though experimental evidence in birds is apparently lacking. However, while the tissue concentration of methylmercury is the critical factor from a toxicological viewpoint, variations in dietary exposure to mercury are likely to be most clearly reflected by tissue levels of total mercury.

More prevalent may be a detoxification mechanism involving mercury and selenium, a nonmetallic element. Selenium has a protective effect against both inorganic mercury and methylmercury in experimental animals, including birds,[67,102] and approximate 1:1 molar (or atomic) ratios and increments have been noted between mercury and selenium in marine mammals.[103] A direct linkage between mercury and selenium, involving sulfur, has been postulated, the resulting complex being stable and nontoxic.[67,104,105] Nevertheless, this interaction may increase retention and accumulation of mercury,[105] especially in liver and kidney. There is also some evidence for involvement of selenium in demethylation of methylmercury.[67,105]

The mercury/selenium relationship is not as clearcut in seabirds as in marine mammals,[65,104] but this is not necessarily evidence that no protective effect is operating. Certainly there is rarely a direct 1:1 relationship, but the mean selenium:mercury molar ratio is always >1 (6.0 to 69 in kidney and 1.1 to 70 in liver over a range of species).[25,26,29,30,59] Perhaps reflecting this 'surplus' of selenium, correlations between the two elements in liver and kidney of different seabird populations are not very clearcut (Figure 3). Nonetheless, significant positive correlations have also been noted within populations.[25,29,30] A positive correlation has also been noted in eggs of little terns *Sterna albifrons*,[106] but no relationship is apparent between species, though again there is usually a (1.2- to 26-fold) molar excess of selenium over mercury.[14,26,29,34,35] However, the toxicological significance of selenium

in eggs is unclear, given that dietary selenium increases the transfer of dietary methylmercury to eggs and that the two bind preferentially to egg white and yolk, respectively.[80]

Significant selenium/cadmium correlations have been noted in kidney or liver of several seabirds,[25,30] possibly related to a protective role of selenium against cadmium.[107] Significant correlations also occur in kidney, but not liver, over a range of populations (Figure 3). A strong correlation between molar liver concentrations of combined cadmium plus mercury and selenium plus zinc in species from Spitsbergen and the Antarctic was interpreted as suggesting integration of detoxifying interactions between these elements.[30]

Although evidence from seabirds is lacking, coaccumulation of zinc and lead in bone, unrelated to dietary levels of zinc, can apparently occur in feral rock doves *Columba livia*.[98] It is unclear whether this reflects the known antagonistic interaction between these metals or an influence on both metals of active bone metabolism during eggshell formation.[86,98] An antagonistic interaction also exists between lead and calcium, since intestinal absorption of lead is greatest from calcium-deficient diets.[67]

Various other metal/metal correlations, of doubtful toxicological or physiological significance, have been reported in seabirds.[14,50,59] There is an overall, though highly variable, positive correlation between cadmium and mercury in liver of different populations (Figure 3), but this largely reflects the tendency for recorded concentrations of these metals to be higher in procellariforms and skuas than in gulls, terns, and auks.

VIII. DOSE-RESPONSE RELATIONSHIPS

The use of seabirds as metal-monitors depends on the assumption that environmental variations will be reflected by levels of heavy metals in tissues. This assumption has not, however, received as much scrutiny in seabirds as in the case of other proposed marine biological indicators.[1,2] At the very least, tissue concentrations should reflect dietary uptake of the particular metal being monitored. The evidence in seabirds tends to be rather anecdotal, being based on inadequately quantified associations with dietary levels of metals or with known sources of metal pollution.[11,15,18,25,27,31,33,35,36,38,41,49,50]

Experimental studies of non-seabird species confirm that increased dietary doses of mercury, cadmium, and lead are reflected in increased tissue concentrations (particularly in specific 'target' organs).[67] In some studies, tissue levels have increased in direct proportion to dietary levels over a particular dose range,[109] but dose responses are often nonlinear, with a lower ratio of tissue to dietary concentration at higher doses. For cadmium, there is some evidence of an increased dose response at higher doses, but there are also studies suggesting up to 75% decreases in response.[67] Similarly, for lead, decreases in dose response of up to 60% have been noted.[110] Mercury may tend to show a more linear tissue/dose relationship,[109] although there is also evidence for a decrease in dose response of up to 20% in eggs.[111] For analogy with natural rates of metal uptake, however, a critical appraisal of dose responses over lower, chronic dose ranges is needed.

Even assuming that a coherent picture can be deduced from existing experimental studies, applying such dose-response relationships to free-living seabirds is far from straightforward. In particular, the exposure of individual seabirds to heavy metals over periods of years or even decades, with associated seasonal variations in uptake and elimination rates, may invalidate comparisons with experimental dosing over weeks or months. Seabird nestlings would be more amenable to direct comparison, but experimental dosing would be far more informative. The most practicable approach, however, might be a quantitative assessment of tissue concentrations in relation to dietary uptake for nestlings at several colonies varying in exposure to heavy metals. Given such information, attempts at using seabirds to quantify environmental variations in heavy metals, at least in terms of biological availability, would be based on firmer ground.

IX. CURRENT EVIDENCE FOR GEOGRAPHICAL VARIATION IN EXPOSURE OF SEABIRDS TO HEAVY METALS

Particularly striking patterns of geographical variation are evident from studies of mercury in single or closely related species in European and North Atlantic waters. Common murres provide a particularly good example. Eggs collected from 16 colonies around Britain and Ireland in the early 1970s showed a 20-fold variation in mean mercury concentrations, with lowest concentrations (0.8 to 1.9 μg/g dry weight or about 0.25 to 0.6 μg/g wet weight) from northwest Scotland around to northeast England but markedly high concentrations (about 1.9 to 4.9 μg/g wet weight) at five Irish Sea colonies.[13] Anthropogenic inputs of mercury to the Irish Sea, in industrial and other waste, are high and are probably exacerbated by low rates of water exchange; mercury levels in seawater and in fish are also high.[112] Eggs from other parts of the murre's range are lower in mercury, with means of 0.12 μg/g at a Canadian colony,[19] about 0.20 to 0.25 in the Faroes, [13,113] <0.07 to 0.13 in northern Norway, [12,18] and slightly higher levels of 0.25 to 0.54 μg/g in the Baltic Sea.[77,113]

Elevated mercury levels in the enclosed Baltic were also reflected in a study of murre feather concentrations: north Baltic samples averaged 3.3 μg/g, south Baltic 2.6, Faroes 1.2, and Greenland (thick-billed murres *Uria lomvia*) 1.1 μg/g.[38] Black guillemots showed a similar pattern, which in both species was also evident, though less marked, even in feathers from pre-1900 museum specimens.[38] In addition to historical trends revealed by auk feather samples,[38,39] there is limited evidence of a decline in Baltic mercury levels within the 1970s, possibly reflecting reduced mercury discharges from Swedish and Finnish chlorine-alkali and paper/pulp industries.[38,114,115]

Other seabirds showing clear geographical variations in mercury levels include northern gannets (mean egg concentrations about 0.6 to 1.7 μg/g wet weight at British and Irish colonies, again peaking in the Irish Sea) and herring gulls (a fairly even trend of increasing egg concentrations from 0.09 μg/g in northern Norway to 1.28 in the south).[12,15,16] Herring gull and common tern eggs from seven regions of the German North Sea coast averaged about 0.2 to 0.5 and about 0.3 to 0.7 μg/g respectively, except for pronounced peaks of 1.55 and 4.58 μg/g at the Elbe estuary.[48]

Further south, mercury concentrations in eggs and adult tissues of Cory's shearwaters from three Mediterranean colonies were consistently higher than at an Atlantic colony (Selvagen),[29] e.g., egg/liver concentrations about 1.3 to 1.9/14.9 to 25.9 vs. about 0.5/3.8 μg/g wet weight. High levels of mercury in the Mediterranean, reflecting both anthropogenic and geological influences,[29] are also reflected in eggs of Audouin's gulls *Larus audouinii* (means 0.76 to 1.32 μg/g).[116,117]

Assessment of variations in seabird mercury levels on a broader, global scale, and of the relative roles of human and natural inputs of mercury,[20,31,65] is difficult, given differences in species and food chains between studies and the current lack of data on historical levels in pelagic seabirds. However, it has been suggested that Antarctic species (Antarctic fulmar *Fulmarus glacialoides* and skuas *Catharacta*) may be exposed to higher mercury levels than comparable species from Spitsbergen (northern fulmar and glaucous gull *Larus hyperboreus*).[61] Comparison between pelagic and inshore populations of seabirds further suggests that mercury in oceanic food chains may be largely natural in origin, despite high tissue concentrations in some pelagic species and evidence for coastal mercury pollution.

The tendency for cadmium levels to be relatively high in pelagic species also suggests a natural origin.[21,24,26,31,53,56,96] This is supported by data for individual species, e.g., kidney concentrations in Atlantic puffins from St. Kilda (about 29 μg/g wet weight) about 12 times higher than from the more coastal Isle of May population.[85] Similarly, for Cory's shearwaters, kidney cadmium concentrations were higher in Atlantic (Selvagen) adults (about 53.5 μg/g) than at Mediterranean colonies (about 10.7 to 46.9 μg/g).[29] However, cadmium variations between broad oceanic regions are unclear.

Anthropogenic inputs of cadmium should be more marked in inshore waters, but little information is available for coastal seabirds. A comparison of laughing gull tissue concentrations between Galveston Bay and Matagorda Bay, Texas found no consistent variation, though prefledgling kidney concentrations were significantly higher in the more polluted Galveston Bay.[33] Fairy prions apparently reflected dumping of industrial waste off the coast of Tasmania, with significantly higher tissue concentrations of cadmium at a nearby colony than at colonies more remote from the dumping site.[36] However, cadmium concentrations in greater crested terns in New South Wales and in estuarine gulls in New Zealand showed no relationship to degree of industrialization.[23,32]

There is some evidence of geographical variation in lead levels of coastal seabirds, reflecting local pollution. Tissue concentrations were significantly higher in laughing gulls from Galveston Bay than from Matagorda Bay, Texas.[33] The mean liver concentration of 5.3 μg/g wet weight in Galveston adults is apparently the highest yet reported for living seabirds, but declined (to 1.1 μg/g) between 1977 and 1980, as did other tissue levels.[33] In Britain, many birds, including gulls, were poisoned by trimethyllead in the Mersey Estuary during 1977 to 1981, the gull casualties having liver concentrations of about 1 to 8 μg/g.[82] However, despite the apparently high contribution of anthropogenically derived atmospheric lead to surface oceanic waters,[2,20] little or no information is available for pelagic seabirds.

Copper and zinc concentrations vary little between seabird populations, as may be expected from their metabolic regulation (Section II). Geographical differences can be evident for zinc,[85] but may largely reflect variation in exposure to cadmium (Section VII).[31] However, copper and zinc concentrations in nestling common terns from Rhode Island, U.S., apparently reflected industrial pollution, and showed the same localized patterns of variation as metals in food items.[50]

Variations in tissue concentrations of arsenic, chromium, iron, manganese, and nickel in some cases appear to reflect coastal pollution,[20,23,32,50,59] but interpretation is difficult at present, as natural levels and the suitability of particular tissues as metal monitors are particularly unclear.

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Chapter 3

SAMPLING SEABIRD FEATHERS FOR MERCURY ANALYSIS: INTRA-POPULATION VARIABLES IN NORTHERN GANNETS AND SHAGS

3.1 Introduction

The usefulness of feathers, in relation to other body-tissues, for monitoring exposure of birds to mercury has been examined in a range of studies (see also Chapters 2 and 4). Once a feather is completely grown, it becomes physiologically isolated from the rest of the body (Voitkevitch 1966). The keratin which forms the bulk of the feather is not easily biodegradable, and, during feather-growth, mercury probably becomes firmly associated with the disulphide bonds of the keratin (Appelquist et al. 1984). Compared to other tissues, mercury concentrations in feathers tend to be high (even if comparisons are on the basis of dry weight), and the affinity of mercury for keratin is evidently higher than for other toxic metals (Chapter 2, section IV).

As shown by Appelquist et al. (1984), the mercury content of fully-grown feathers shows little or no change when feathers are subjected to various rigorous treatments, including exposure to extremes of temperature and to UV light. In addition, there is no indication that leaching of mercury from the feather occurs under normal environmental conditions (including exposure to salt-water), or that the mercury content of feathers is increased by secretions from the uropygial (preen) gland (Goede & de Bruin 1984).

The bulk of data on mercury levels in seabirds (and other groups) is based on soft-tissues (mainly liver, kidney and pectoral muscle) and on eggs. Various studies have examined the relationship between mercury concentrations in different tissues, and significant positive correlations between plumage and soft-tissue concentrations have often been demonstrated (e.g. Furness & Hutton 1979; Ohlendorf et al. 1985; Thompson *et al.* 1991). Similar results have been found for other birds, including waterfowl and waders (e.g. Bacher &

Norman 1984; Vermeer & Armstrong 1972; Turner et al. 1978; Goede 1985). More frequently, positive correlations are apparent but can not be demonstrated statistically (e.g. Gochfeld 1980, and individual tissues in some of above studies). Small sample sizes are sometimes to blame, but perhaps more important are soft-tissue variations reflecting seasonal or physiological influences.

Some older studies have attempted to estimate soft-tissue mercury concentrations based on a supposed 7: 3: 1 ratio between feather, liver and pectoral muscle concentrations. Thompson et al. (1990) showed that such ratios can vary greatly, depending on such factors as moult and the chemical form of mercury involved.

Mercury concentrations in soft tissues have been found to decline during feather-growth in the moult period, and to increase again in the periods between moults, associated with incorporation of mercury in the growing feathers. A good demonstration of this was given by Honda et al. (1986) for Black-eared Kites *Milvus migrans*, in which about half of the 'non-plumage' mercury burden was lost between the beginning and end of moult. Similarly, for Bonaparte's Gulls *Larus philadelphia*, Braune & Gaskin (1987) estimated that feather-growth accounted for 60-70% of mercury-losses over the period of autumn moult. Thus the relationship between mercury concentrations in feathers and other tissues may vary seasonally.

All, or virtually all, of the mercury incorporated in feathers appears to be in the organic form (methyl mercury) (Thompson & Furness 1989a). This introduces a further complication, in that some species have been shown to accumulate high concentrations of inorganic mercury, mainly in the liver (apparently following de-methylation of methyl mercury which has not been 'eliminated' in feathers) (Thompson & Furness 1989b). Thus the relationship between feather and liver concentrations of mercury will not always be clearcut.

Seasonal (including moult-related) and other factors obviously need to be taken into account when assessing the 'plumage mercury concentration' of a given sample or population of birds. Other relevant intra-population variables would include the type of feather analysed and the age of birds, among others. Without allowing for such variables, there is a danger that inaccurate conclusions may be drawn from data on plumage-content of mercury.

This chapter examines some of the intra-population or sampling variables which may be relevant, based on feather-samples from Gannets *Morus bassanus* and Shags *Phalacrocorax aristotelis*. Relevant findings from other studies are also discussed. Geographical or year-to-year variation in mercury concentrations of feathers are not dealt with here (see Chapters 4 and 6), nor is the possible role of dietary variation (Chapters 4).

A note on Gannet moult and feather-growth

Adult Gannets undergo a complete annual moult, beginning around May and ending as late as December (body-feathers) or February (primary feathers of wing) for at least some individuals (Cramp & Simmons 1977, Nelson 1978, Walsh unpublished). Moult of the primaries in Gannets is said to be 'serially descendant', i.e. there are several (usually three) active moult 'centres' along the ten primaries of each wing, with moult preceding outward within several distinct series of feathers. This contrasts with moult in most (smaller) species of bird, where primary feathers are moulted and replaced from innermost to outermost feather along the entire primary sequence, i.e. beginning at primary 1 and ending at primary 10. Moult of secondary feathers on the Gannet wing is serially ascendant, i.e. from outermost to innermost feather within several (possibly many) different sequences. Moult of

tail-feathers is irregular or perhaps serially descendant. For all these series of large feathers, the positions at which moult begins can differ between individual Gannets or even between the left and right wing (or side of tail) of an individual. The sequence of moult within, or between, feather tracts (including the large scapulars) on the body has apparently not been established for Gannets.

Gannets chicks are almost naked when they hatch, but growth of white down feathers is apparent after three days and proceeds most rapidly on the dorsal surface (Kirkham & Montevecchi 1982). By the time the chick is three weeks old, down covers all feather tracts. This first generation of short down is replaced by a second generation of fluffy, generally longer down, which is obvious by the end of the fourth week (Nelson 1978). The detailed timing of down growth has not been assessed, but there appears to be some growth up to the fifth or sixth week. Growth of dark scapulars, wing and tail feathers begins about this time (with near-simultaneous growth of individual feathers, i.e. no obvious 'moult' sequences). These juvenile feathers, including body feathers which appear over the following weeks, gradually push out and replace the second-generation down. The last trace of down is lost by the time the chick is 11-12 weeks old, about a week before fledging from the nest (Nelson 1978). Limited growth of the primaries continues immediately after fledging. Subsequent moult patterns for juvenile and older immature Gannets are complex (Cramp & Simmons 1977, Nelson 1978), but not relevant to the feather samples analysed in this study.

3.2 Methods

3.2.1 Collection of samples

Feathers were sampled from adult and nestling Gannets at a wide range of North Atlantic colonies, and from adult and nestling Shags at several British and Irish colonies, mainly during the breeding seasons of 1986-88. In most cases, samples of scapulars, small body-feathers or nestling down were taken from live birds, usually from one or two plumage areas only. Some additional samples were taken from Gannets found dead at or near colonies, in most cases birds thought to have died as a result of accidents (particularly falls from the cliff-top). Moulted body-feathers were also collected at some colonies.

In addition to samples collected by PMW, fieldworkers at a range of other colonies provided feathers. Fuller details of these, and of sampling procedures in general, are given in Chapters 1 and 4.

3.2.2 Laboratory methods

Methods are outlined more fully in Chapter 1, but are summarised briefly here.

Most feather samples were stored at room temperature (or, in some cases, refrigerated at ca. 5^{o}C), in the polythene bags originally used. Samples of down from chicks (which generally held more moisture) and any samples which were damp when collected were stored in a deep freeze.

Before analysis, feathers were rinsed several times in chloroform, acetone and distilled water to remove surface contamination. Feathers were then dried at 50°C for 24 hours, sealed in individual test-tubes and allowed to cool before weighing. This reduced variation in sample weight (although, for most feather samples, variation would usually be less than ±5% in any case).

Analysis of the mercury content of feathers was by cold-vapour ultra-violet absorption spectrophotometry following standard acid digestion. Accuracy was assessed by analysing horse kidney Reference Material H-8 from the International Atomic Energy Agency.

3.3 Results

3.3.1 Age of bird

Differences in the mercury content of adult and nestling feathers can be expected on the basis of such factors as different exposure times to environmental mercury and different patterns of feather-growth. Some additional comparisons between adult and chick samples are made under later sub-headings.

For a given colony-year, pooled mantle-feathers of adult Gannets and Shags always had mercury concentrations at least twice as high as in similar samples from well-grown chicks (Table 3.1).

For a given type of feather sample, however, variability within a population (as indicated by the Coefficient of Variation [CV = 100 x Standard Deviation/Mean] of mercury concentration) was generally much lower for well-grown chicks than for adult Gannets and Shags (Table 3.2). For example, the median CV for mercury in mantle-feathers was 20.7% for six population samples from Gannet chicks, compared to 31.9% for nine samples from adult Gannets.

The lower variability of mercury concentrations in chicks can be explained partly on the basis that feather-growth is less complex than in adults. Juvenile feathers in a given feather tract (e.g. scapulars) grow more or less simultaneously (see Introduction), and are thus likely to show less variation in mercury concentrations than feathers of adults (cf. sections 3.3.2 and 3.3.3). This applies whether or not feathers are pooled to reduce variation (section 3.3.3).

Table 3.1 Ratio of mean mercury concentration in feathers of adult birds to concentration in feathers of well-grown chicks, for colonies of Gannets and Shags

Species	Colony	Year	Adult : large chick ratio of Hg concentration in mantle feathers*
Gannet	Grassholm	1987	3.8
	St Kilda	1986	2.4
	Bass Rock	1986	4.8-6.6[2]
		1987	4.0-4.0[2]
		1988	4.2-4.6[3]
Shag	Great Saltee	1988	5.5
	Bass Rock	1988	6.3

*Ratio based on arithmetic means of samples of 10+ individuals (ranges indicate [2] or [3] samples from adults in a season).

Table 3.2 Coefficients of variation of mercury concentrations in feathers of (i) Gannets and (ii) Shags during breeding season (samples of 10+ birds). For some colony-years, two samples were available.

Colony	Year	chick down	chick mantle	chick scapular	adult mantle	adult scapular
(i) Gannet						
Bass Rock	1986	-	21.2	31.2	31.2	48.7/55.7
	1987	-	20.2	-	27.3/32.7	-
	1988	20.3/25.4	29.0	-	28.7/51.8	
Noss	1987	-	-	14.1	-	-
Hermaness	1988	-	-	-	33.0	-
St Kilda	1986	-	-	20.5	47.9	-
Ailsa Craig	1987	-	-	25.3	-	-
Grassholm	1986	-	19.8	33.4	45.2	70.0
	1987	19.0	-	-	-	-
Great Saltee	1986	22.0	-	-	-	-
	1987	18.9	-	-	-	-
Little Skellig	1987	14.5	12.2	-	-	-
Hovsflesa	1986	-	-	-	-	35.0
Skarvlakken	1986	-	-	-	-	53.4
Bonaventure	1986	-	-	18.4	-	35.5
no. of samples		**6**	**6**	**6**	**9**	**6**
median		**19.7**	**20.7**	**22.9**	**31.9**	**51.0**
(ii) Shag						
Bass Rock	1988	-	21.8	-	46.9	-
Isle of May	1988	-	-	-	-	17.8
Great Saltee	1986	-	-	32.4	-	-
	1988	-	25.1	-	45.6	-
Blaskets	1988	-	-	33.1	-	-
no. of samples		**0**	**2**	**2**	**2**	**1**
median		**-**	**23.4**	**32.7**	**46.2**	**(17.8)**

Samples of feathers (other than down) from Gannet chicks were taken from well-grown nestlings (9-13 weeks old: Nelson 1978) or newly-fledged birds. Apart from down, few feather-samples were taken from younger chicks, and possible influences of chick-growth or age on mercury concentration in juvenile plumage were thus not examined.

Coefficients of variation were also low for mercury concentrations in mantle-down of Gannet chicks (Table 3.2), despite the fact that some samples included chicks ranging in age from two to seven weeks old (cf. Table 3.6), with a median CV of 19.7% for six samples.

Downy chicks sampled varied greatly in size, and mercury concentrations were examined in relation to three measures of body-size for several samples. The measures used were: total weight of chick; standard wing-length (maximum chord of outer wing, excluding down at tip); and length of the 10th (outermost) primary feather (which appears when the chick is in its fifth week) (Nelson 1978). Weight provides only a crude measure of chick age, as it can vary greatly between individual chicks (and may include the undigested parts of a recent meal). In addition, both weight and wing-length (as defined here) do not increase linearly with age, but begin to level off after six or seven weeks. Growth of the tenth primary is more linear with age (Poulin 1968).

Chick weight and wing-length were recorded for four samples of downy Gannet chicks, and were compared with mercury concentration in mantle down by linear regression (Figures 3.1-3.4). In all cases, there was a very poor relationship with mercury concentration, and inspection of the data indicated that non-linear regressions would likewise fail to demonstrate any relationships.

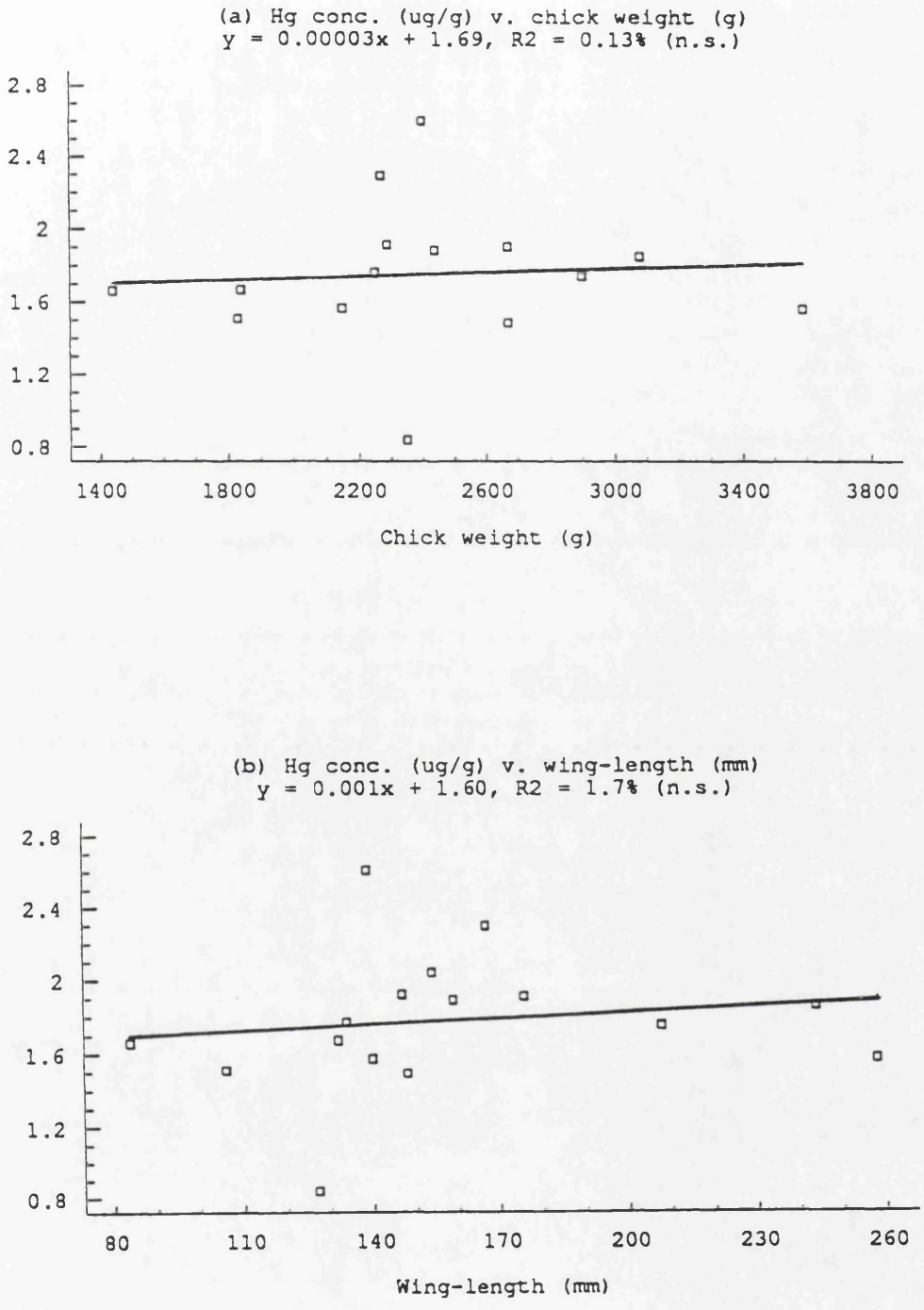
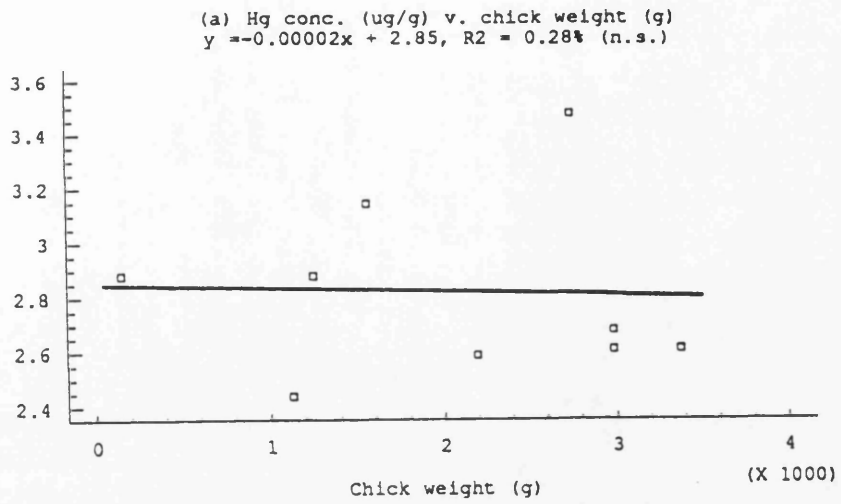
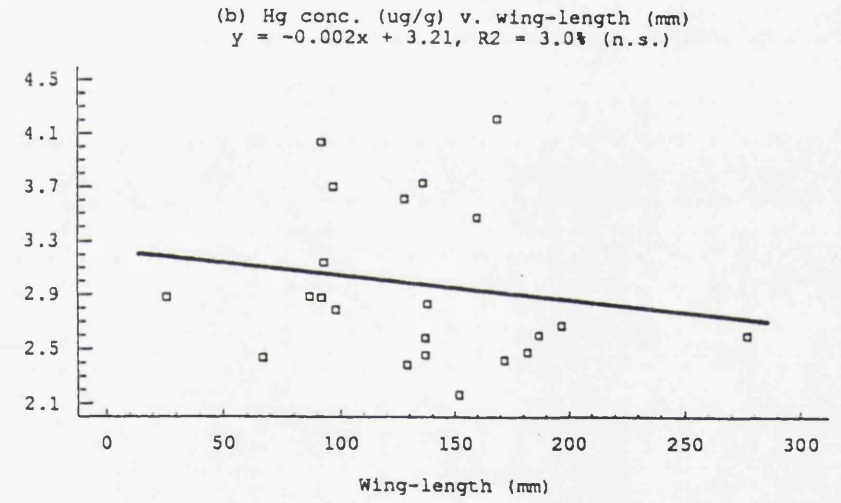


Figure 3.1 Mercury concentration (ug/g dry weight) in mantle down of Gannet chicks, Great Saltee, 25 June 1986, in relation to (a) weight of chick and (b) wing-length (maximum chord of outer wing, excluding down). Each point represents a sample from one bird; fitted line is a simple regression of mercury concentration against chick weight or wing-length (neither regression is significant).





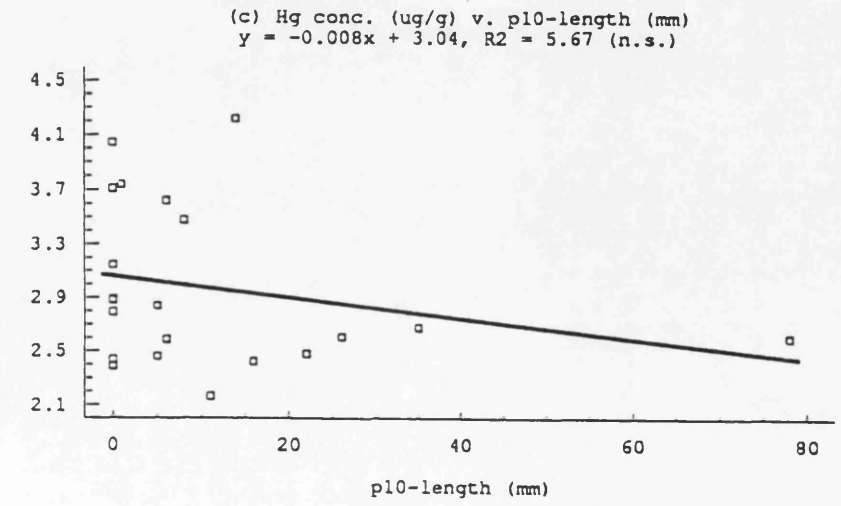


Figure 3.2 Mercury concentration (ug/g dry weight) in mantle down of Gannet chicks, Grassholm, 1 July 1987, in relation to (a) weight of chick, (b) wing-length (maximum chord of outer wing, excluding down), and (c) length of 10th (outermost) primary-feather of wing. Each point represents a sample from one bird; fitted line is a simple regression of mercury concentration against chick weight, wing-length or p10-length (all regressions non-significant).

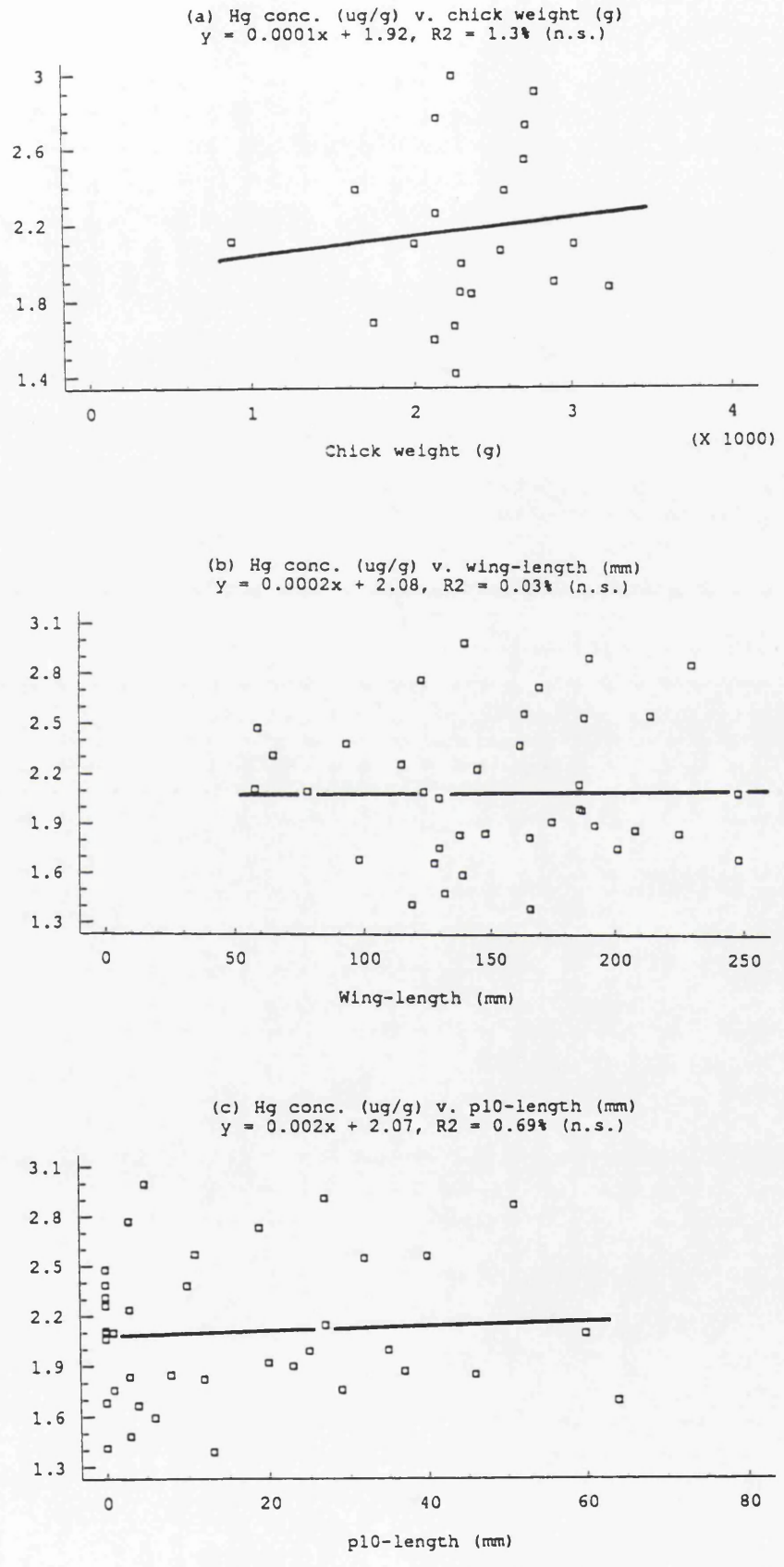


Figure 3.3 Mercury concentration (ug/g dry weight) in mantle down of Gannet chicks, Bass Rock,14 July 1988, in relation to (a) weight of chick, (b) wing-length (maximum chord of outer wing, excluding down), and (c) length of 10th (outermost) primary-feather of wing. Each point represents a sample from one bird; fitted line is a simple regression of mercury concentration against chick weight, wing-length or p10-length (all regressions non-significant).

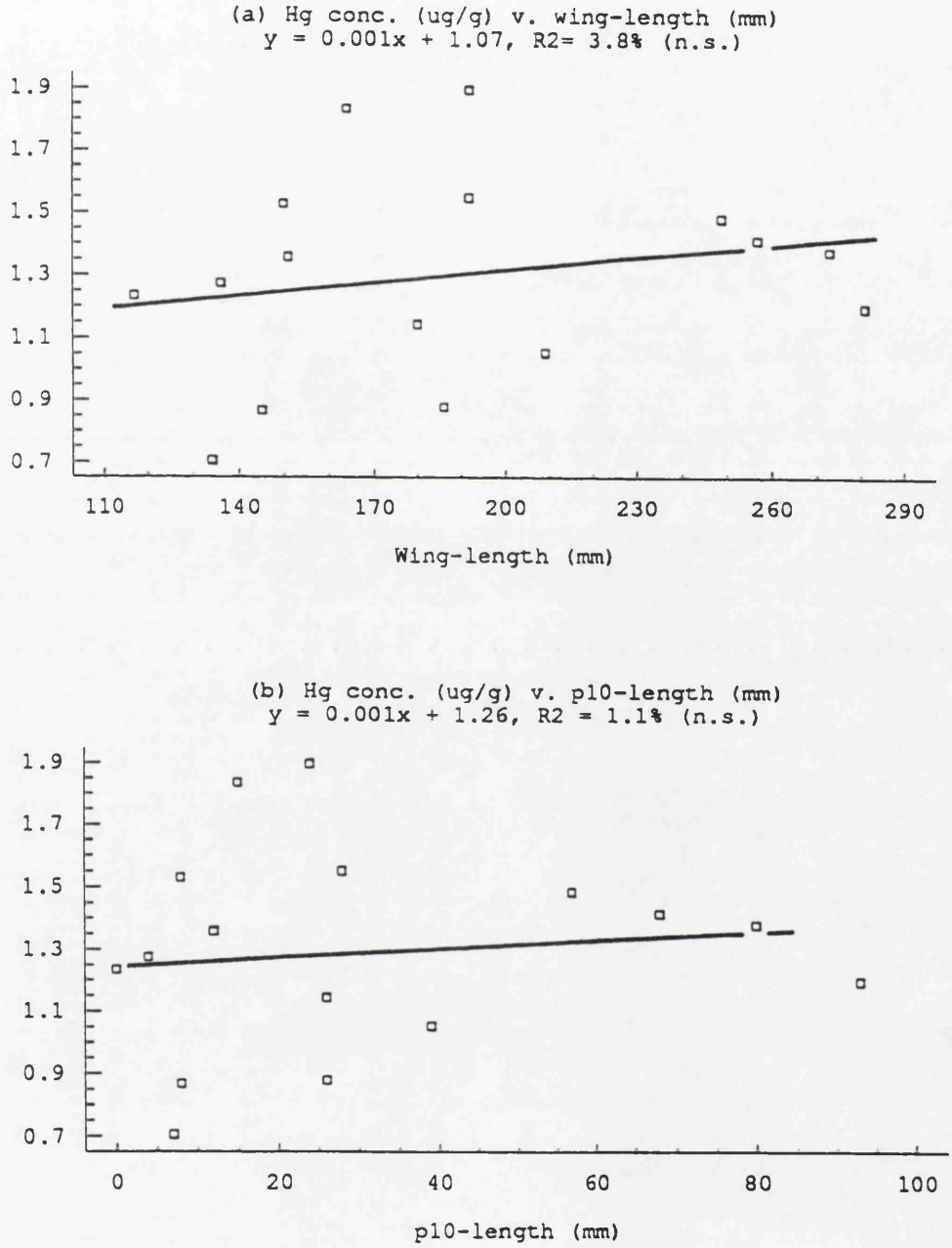


Figure 3.4 Mercury concentration (ug/g dry weight) in mantle down of Gannet chicks, Bass Rock, 20 August 1988, in relation to (a) wing-length (maximum chord of outer wing, excluding down) and (b) length of 10th (outermost) primary-feather of wing. Each point represents a sample from one bird; fitted line is a simple regression of mercury concentration against chick weight or wing-length or p10-length (neither regression is significant).

For three of these samples (Figures 3.2-3.4), the length of primary 10 was also known, but, again, no relationships were evident. In the Grassholm sample (Figure 3.2), there was a suggestion that mercury concentrations in down were highest for the younger chicks, i.e. those with the shortest 10th primary ('p10'). The median mercury concentrations for chicks with p10 = 0 mm, and those with p10 = 0-10 mm or p10 = 0-20 mm, were all 2.88 ug/g, compared to 2.61 ug/g for those with p10 >0 mm, > 10 mm and >20 mm, but in no case was this difference significant (Mann-Whitney tests). Similar tests for the other samples likewise showed no significant variation between these broad age-/size-categories.

For both Gannet and Shag, too few adults or post-juvenile immatures of known age were sampled to allow assessment of possible changes in mercury concentrations in the plumage after the fledglings have left the colony.

3.3.2 Moult and feather growth

Mercury concentrations along feather-sequences of wing- and tail-feathers are shown for an adult Gannet found dead at the Ailsa Craig colony in Figures 3.5-3.8. In each case, the highest mercury concentrations were found in recently-grown feathers, either still growing or showing traces of the feather-sheath. None of the feather sequences shows a single, linear sequence of increasing or decreasing mercury concentration, and the pattern of mercury concentrations shown by the secondary feathers looks particularly haphazard.

In fact, the patterns of mercury concentrations in all these feather-sequences are consistent with the complex moult patterns seen in Gannets (see Introduction, this chapter), whereby loss and replacement of feathers may proceed near-simultaneously along several short sequences of feathers, within a longer series.

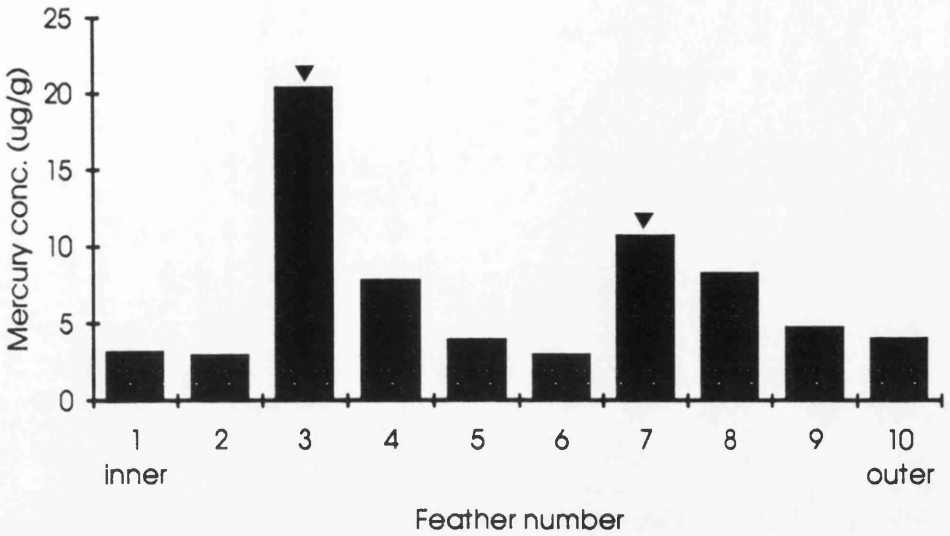


Figure 3.5 Pattern of mercury concentrations (ug/g dry weight) in primary feathers of an adult Gannet (right wing) found dead on Ailsa Craig, August 1985. Direction of moult is from inner to outer primaries (in more than one linear sequence). Recently-grown feathers (including partly-grown ones) are highlighted.

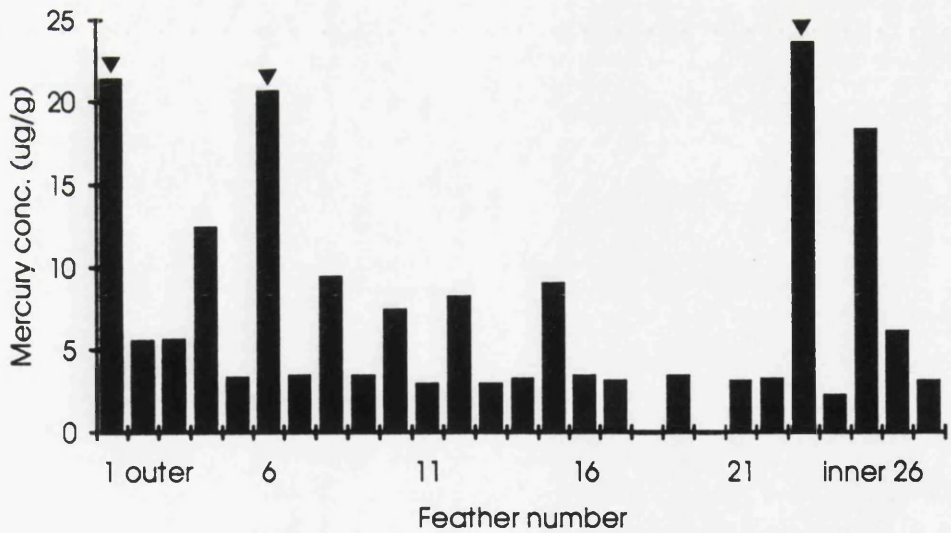


Figure 3.6 Pattern of mercury concentrations (ug/g dry weight) in secondary feathers of an adult Gannet (left wing) found dead on Ailsa Craig, August 1985. Direction of moult is from outer to inner feathers (in more than one linear sequence). Recently-grown feathers (including partly-grown ones) are highlighted; two feathers have been moulted but not replaced yet.

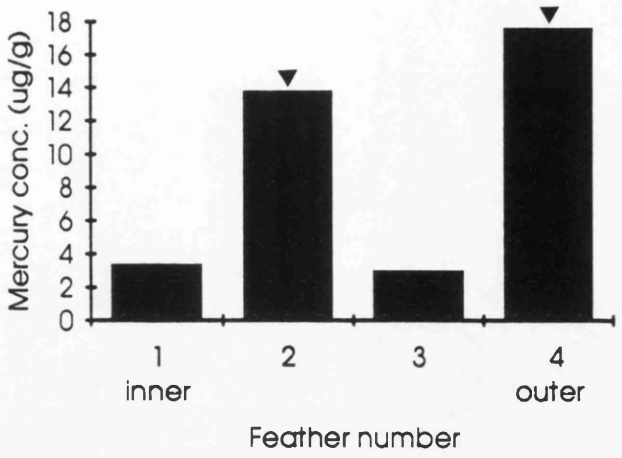


Figure 3.7 Pattern of mercury concentrations (ug/g dry weight) in alula feathers of an adult Gannet (right wing) found dead on Ailsa Craig, August 1985. Recently-grown (part-grown) feathers are highlighted.

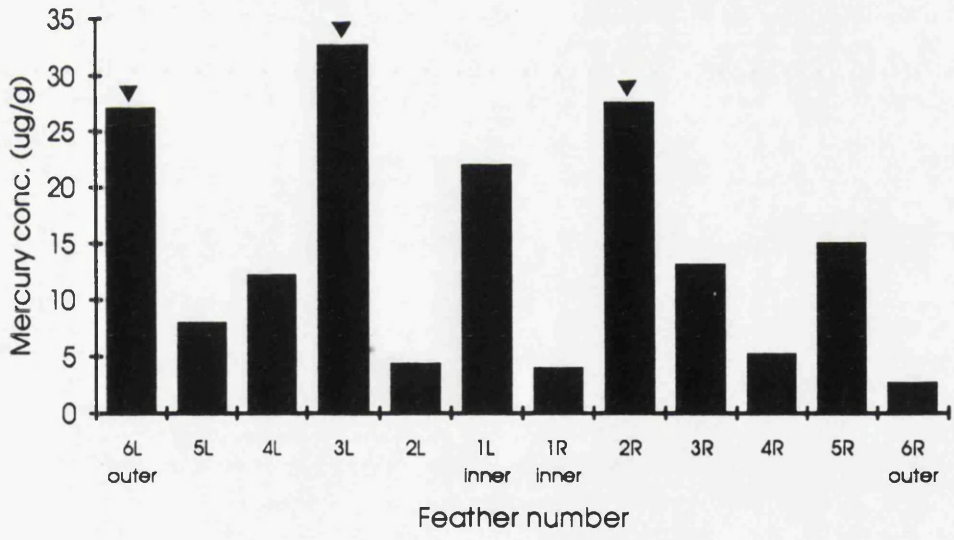


Figure 3.8 Pattern of mercury concentrations (ug/g dry weight) in tail-feathers of an adult Gannet found dead on Ailsa Craig, August 1985. Left half of tail = feathers 1L-6L, right half = 1R-6R. Direction of moult is either irregular, or possibly from inner to outer feathers (more than one moult-sequence each side). Recently-grown feathers (including partly-grown ones) are highlighted.

For adult Gannets sampled at a range of colonies in 1986, mercury concentrations in individual large scapular feathers were analysed. If two or more scapulars were available, one was chosen randomly. Each feather was assigned to one of the feather-wear categories in Table 3.3. These categories covered the range of variation seen in large feathers of Gannets, although extremely worn feathers (category 'F') were rarely seen among scapulars (which are more protected from abrasion than, e.g., tail -feathers). The degree of abrasion shown by a scapular was taken to reflect its relative age, i.e. how recently it had been moulted and regrown.

In Figure 3.9, mercury concentrations are plotted against feather-wear categories for five samples of scapulars from adult Gannets (including combined data for two Norwegian colonies). In each case, there is a negative relationship between mercury concentration and degree of wear (relative age) of feather, i.e. the most recently-grown feathers tended to have the highest concentrations. However, this relationship is significant for only two of the samples (Bass Rock [September] and Bonaventure Island).

For any given category, there is substantial variation in mercury concentrations. Part of this may reflect variation between individual adults, and errors in assigning feathers to a particular category. A more important influence is likely to be that feather-wear may not always accurately reflect the relative position of a feather within a moult sequence, i.e. feathers of a given category may have been replaced at different stages within the moult cycles of different birds. There may also be variation in the mercury-concentration / feather-wear relationship between different colony samples, reflecting differences in timing of breeding (and by implication moult) or in sampling date.

Table 3.3 Feather-wear categories defined for scapulars and other large feathers of adult Gannets. In most cases, a 10x binocular microscope was used to examine feathers for abrasion around the edges of the feather-webs (particularly at the tip of the feather). Where the precise wear-category was uncertain, intermediate categories were used e.g. CD (or, if closer to D than C, CDD). For scapular feathers sampled in July-September, feathers in category A certainly, and category B almost certainly, would have been grown since the start of May; feathers in category E and F would almost certainly have been grown the previous year. Feathers at intermediate stages of wear could include a combination of feathers from two different years.

Category	Description
A	Completely fresh, unfaded/undiscoloured (pure white in the case of scapulars, pure black in the case of primaries); includes part-grown feathers, or recently-grown feathers still in sheath (with separate note kept of these)
B	Fresh, apparently unabraded, but slight fading/discolouration
C	Slight abrasion at the edges, at least near near tip of feather, barely visible with unaided eye; often more obvious fading/discolouration.
D	Obvious abrasion and fading/discolouration
E	Very worn/abraded feather, with obvious fading/discolouration
F	Extremely worn, faded/discoloured feather (FF = part of feather-shaft broken at tip, especially on tail)

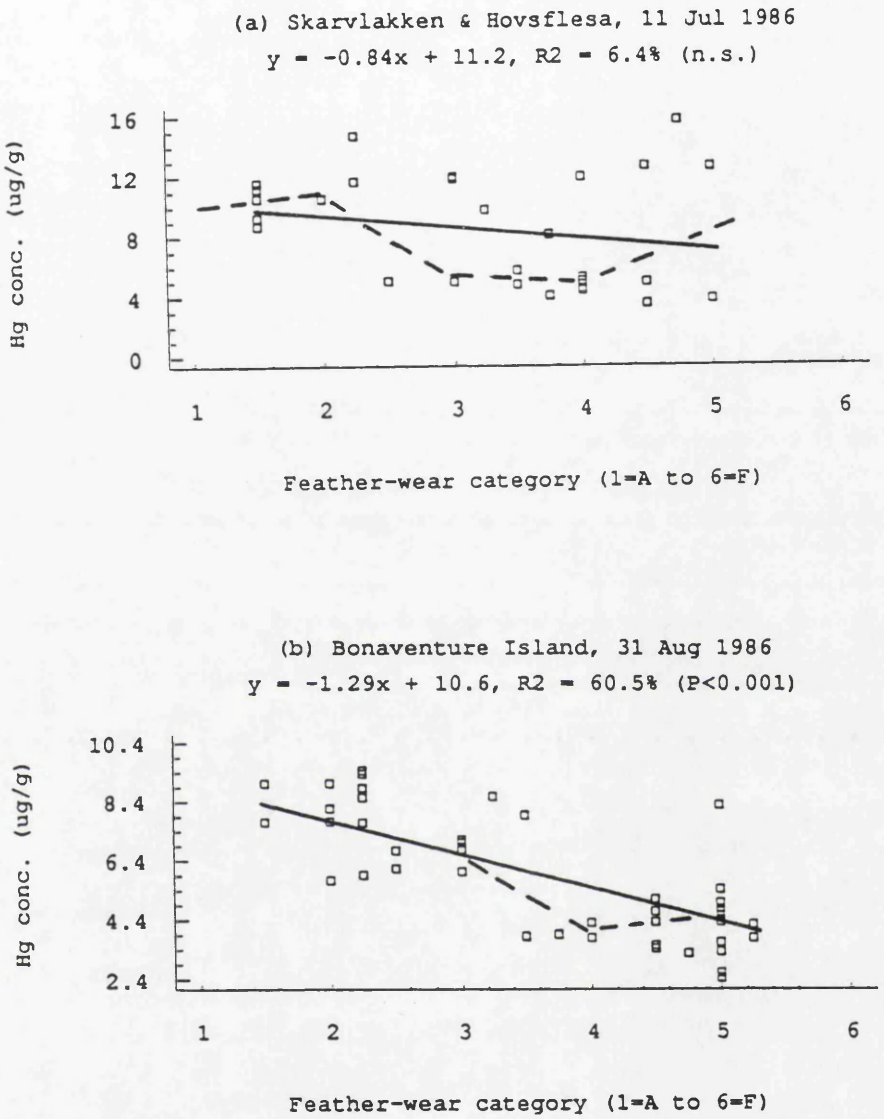
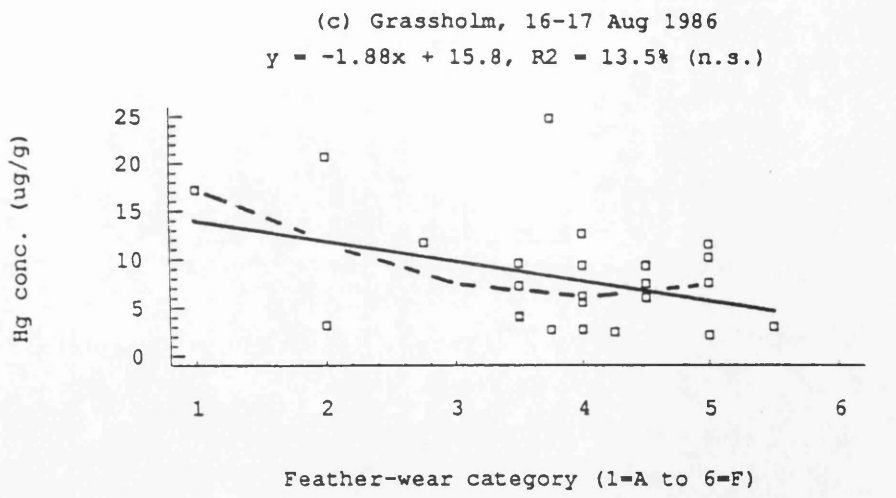
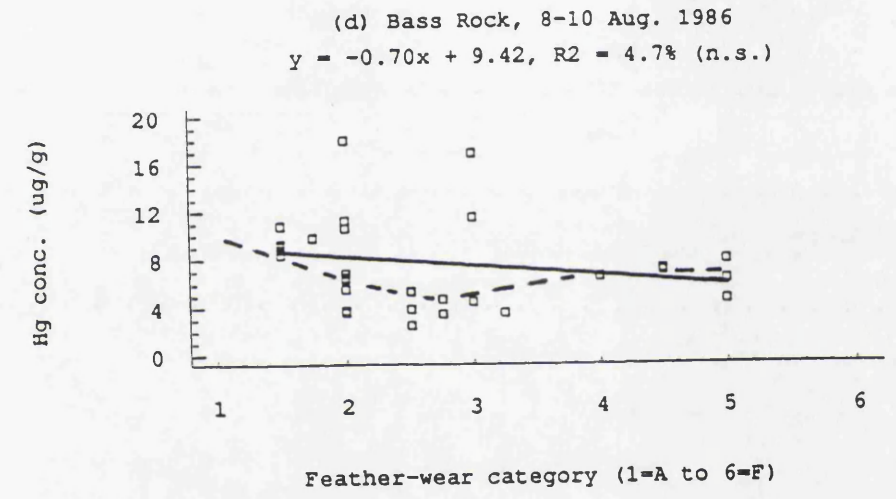


Figure 3.9 Mercury concentrations (ug/g dry weight) in single scapular feathers of individual adult Gannets in relation to feather-wear category (relative age of feathers). Categories range from A (perfectly unabraded, unfaded, recently grown feather) to F (extremely abraded), with intermediates (equivalent to numerical scale from 1 to 6). Each point represents a single feather from a different adult Gannet; highlighted points are part-grown feathers or recently-moulted feathers still in sheath. Continuous line is simple regression of Hg concentration on feather-wear category (negative relationships significant for Bonaventure and for September Bass rock samples only). Broken lines connect median mercury concentrations for broad wear-categories (BC-CD, CD-DE, etc., with overlap at category boundaries).





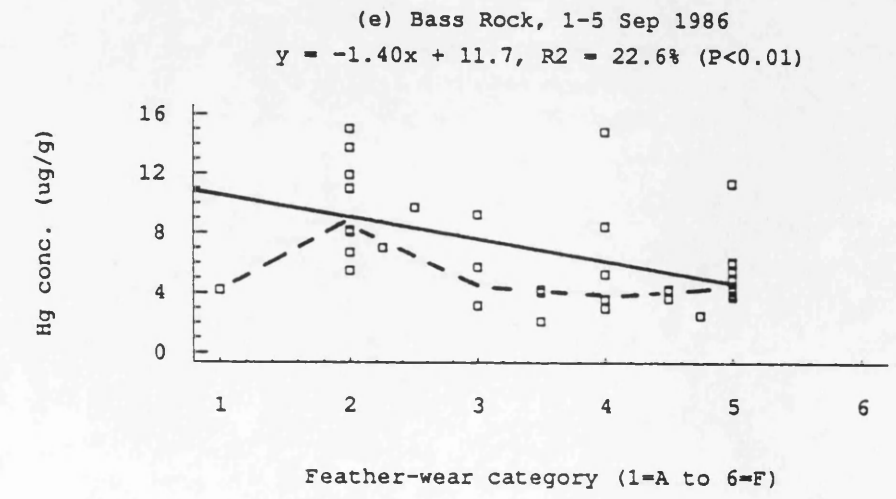


Figure 3.9 (continued)

If scapular feathers are assigned to broader categories (combined categories A-BC, CD, and DE-F), the highest mercury concentrations are seen in the 'least worn' category for four out of five samples (Table 3.4). Again, variation between wear-categories is significant only for the Bonaventure and September Bass Rock samples

There is no clear tendency for the most heavily worn scapulars to have mercury concentrations higher or lower than scapulars in other categories (Figure 3.9, Table 3.4). In fact, both 'low-mercury' and 'high-mercury' feathers are seen within each wear-category. This can be expected even if specific scapulars tend to be replaced at the same stage of each successive moult-cycle (athough this is not certain), despite the fact that late-moulted feathers will tend to have lower mercury concentrations. During the late breeding season (July-September), the most heavily worn feathers are likely to be those moulted at an intermediate stage of the previous moult-cycle, if the oldest feathers have already been moulted and replaced (i.e. at least categories A-B). However, some of the previous year's 'early-moulted' feathers may not yet have been replaced, and could account for some of the high mercury concentrations seen in heavily worn feathers. In addition, although adult Gannets appear to undertake a complete (though prolonged) annual moult (Cramp & Simmons 1977, Walsh unpublished), it may happen that moult of some feathers falls 'out of phase', e.g. if a late-moulted feather is not replaced again until two moult-cycles later.

3.3.3 Number and type of feathers sampled

Within a given population of adult seabirds, mercury concentrations in individual, large feathers are likely to vary more than concentrations in pooled samples of

Table 3.4 Mercury concentrations in single scapulars from individual adult Gannets in relation to feather-wear category (cf. Table 3). Data for scapulars in feather-wear categories A-BC (least worn), C-D, and DE-F (most worn), respectively, are combined separately (data for some intermediate/uncertain categories are excluded).

Colony (date)	Feather-wear categories	n	Hg concentration (ug/g dry wt) mean	s.d.	median	Kruskal-Wallis test-statistic
Skarvlakken / Hovsflesa (11 July 1986)	A-BC	9	10.39	2.62	10.59	
	C-D	13	7.17	3.17	5.33	2.74 (P = 0.25, ns)
	DE-F	6	8.91	5.41	8.82	
Bonaventure* (1 Sept 1986)	A-BC	14	7.88	1.29	7.97	
	C-D	11	6.06	1.76	6.80	23.14 (P<0.001***)
	DE-F	20	4.30	1.24	4.35	
Grassholm (16 Aug 1986)	A-BC	3	13.74	9.29	17.25	
	C-D	11	8.13	6.38	6.22	1.21 (P = 0.54, ns)
	DE-F	8	7.20	3.34	7.57	
Bass Rock (9 Aug 1986)	A-BC	19	7.90	3.64	8.41	
	C-D	5	8.60	5.56	6.55	0.34 (P = 0.84, ns)
	DE-F	5	6.62	1.26	7.05	
Bass Rock* (3 Sept 1986)	A-BC	10	9.13	3.62	8.11	
	C-D	11	5.82	3.74	4.27	11.39 (P<0.01 **)
	DE-F	17	4.86	1.91	4.29	

*Significant differences noted for the following two-sample comparisons (Mann-Whitney tests):
Bonaventure - A-BC v. C-D (P<0.05), C-D v. DE-F (P<0.05), A-BC v. DE-F (P<0.001);
Bass Rock (Sept 1986) - A-BC v. C-D (P<0.05), A-BC v. DE-F (P<0.01).

body-feathers. This can be predicted on the basis of moult influences on the mercury content of feathers (section 3.3.2).

To examine the value of pooling small feathers to reduce variation, Table 3.2 compares Coefficients of Variation (CV) for mercury concentrations in single scapular feathers and pooled mantle-feathers of Gannet and Shag chicks and adults. For chicks, CV values are all within the range 12-36%, compared to 18-70% for adults. However, CVs are generally lower for pooled mantle-feathers than for single scapulars of adult Gannets, and to a lesser extent of chicks. This can be seen for Grassholm and Bass Rock adults in August 1986, from which both types of feather-sample were analysed for each bird (Figure 3.10).

Variations in average mercury concentration between different plumage areas of adult Gannets are potentially high, and might be expected to reflect differences (if any) in the stage of the moult-cycle or in the rate at which different feather-tracts are renewed. Table 3.5 presents limited data for different plumage areas of two adult Gannets, suggesting that mercury concentrations show relatively low variation if pooled samples of small body-feathers are used. Variability is again high between individual large feathers (scapulars, wing and tail).

The relationship between mercury concentration in scapular and mantle feathers is examined in Figures 3.11-3.12 for two samples of adult Gannets (cf. Figure 3.10). In both cases there is a broad positive correlation, although this is significant only for the Bass Rock sample (Figure 3.11). Median mercury concentrations were higher for pooled mantle-feathers than for scapulars in both cases, although in neither case was the difference significant.

In a smaller sample, mercury concentrations in pooled mantle and pooled flank feathers showed a significant positive correlation (Figure 3.13), and median

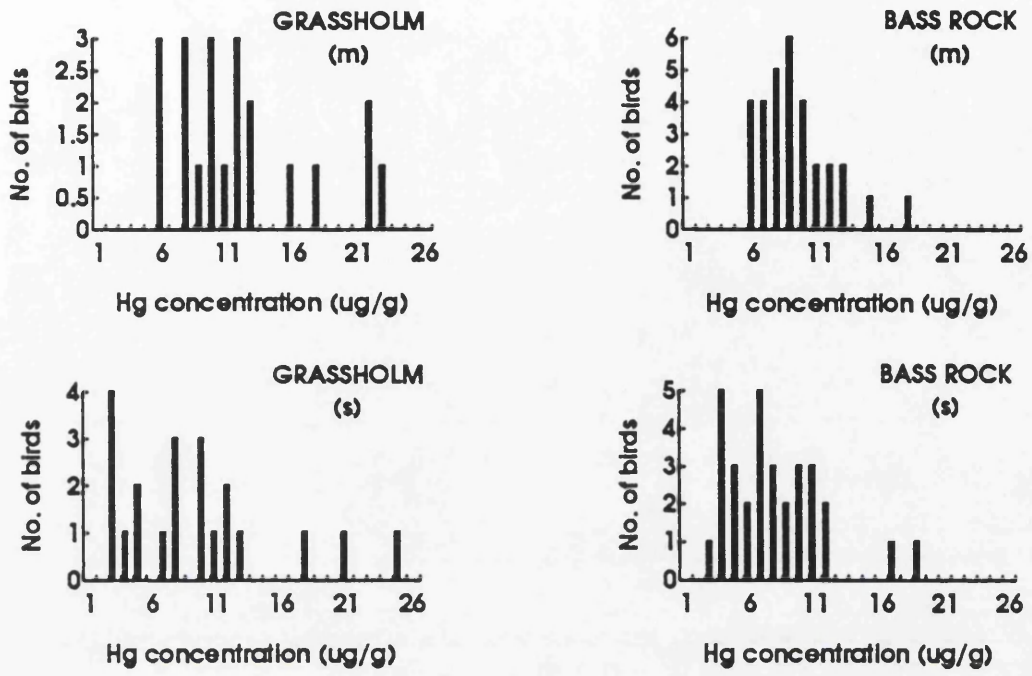


Figure 3.10 Comparison of mercury concentrations (ug/g dry weight) in pooled mantle (m) feathers v. single scapular (s) feathers of adult Gannets at two colonies. Both feather-types were sampled from the same 31 birds on the Bass Rock and 21 birds on Grassholm.

Table 3.5 Mercury concentrations (ug/g dry weight) in feathers from different plumage areas of two adult Gannets (found dead Ailsa Craig, August 1985, and Bass Rock, May 1988, respectively).

Plumage area	Ailsa Craig			Bass Rock
	weighted mean	median	range, n	single samples
Crown	-	2.89	2.73-3.06, 2	8.35
Nape	-	2.94	2.66-3.23, 2	6.35
Lower hind-neck	-	7.59	6.99-8.19, 2	8.61
Foreneck	-	-	-	8.65
Mantle	-	8.85	6.92-11.43, 3	10.96
Breast	-	9.16	8.30-9.88, 3	6.62
Rump/lower back	-	8.18	7.50-8.87, 2	7.56
Belly	-	7.82	6.54-9.10	8.09
Upper flank	-	-	-	8.18
Scapulars*	10.7	9.04	2.28-18.88, 8	-
Lesser & median upperwing-coverts	8.5	8.62	6.79-10.45, 2	-
Greater upperwing-coverts	3.8	3.46	2.39-5.46, 3	-
Alula*	8.7	8.5	2.9-17.5, 4	-
Secondaries: full-grown*	6.4	3.4	2.2-20.6. 23	-
Secondaries: part-grown*	22.2	21.9	21.3-23.6, 2	-
Primaries: full-grown*	5.4	4.0	2.9-10.6, 9	-
Primaries: part-grown*	20.3	20.3	20.3, 1	-
Tail: full-grown*	7.6	7.9	2.5-14.9, 9	-
Tail: part-grown*	28.5	27.0	27.0-32.6, 3	-

*Individual feathers analysed; other samples are pooled small feathers.

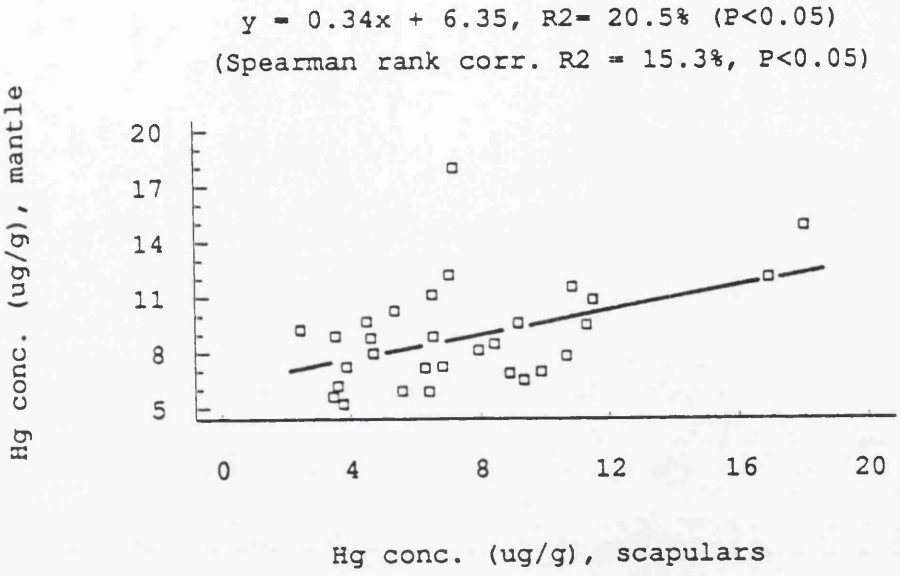


Figure 3.11 Relationship between mercury concentrations(ug/g dry weight) in pooled mantle feathers and in single scapular feathers of 31 individual adult Gannets, Bass Rock, 9 August 1986. Fitted line is simple regression of mantle against scapular data. Difference in median concentrations is not quite significant (8.48 ug/g for mantle v. 6.84 for scapulars: Wilcoxon signed rank test, Z = 1.95, P = 0.051, n.s.).

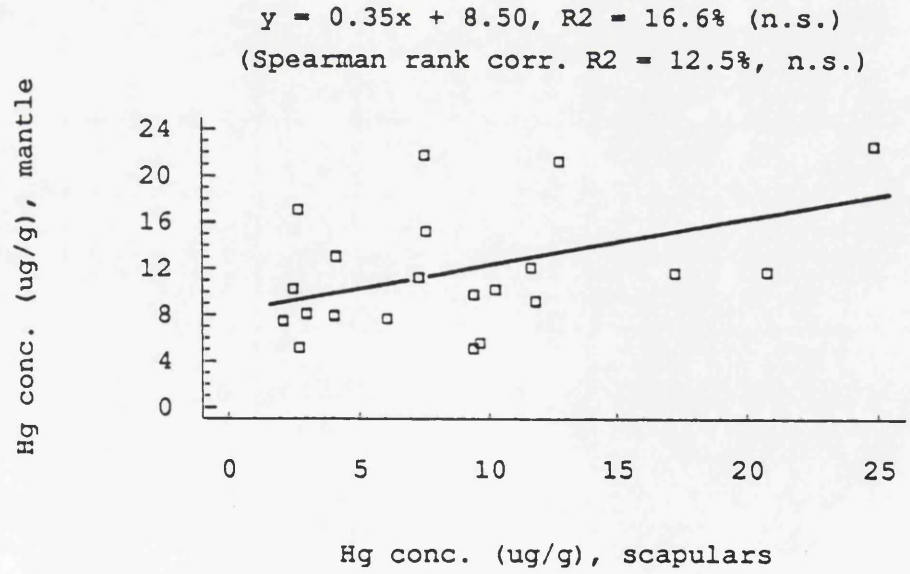


Figure 3.12 Relationship between mercury concentrations(ug/g dry weight) in pooled mantle feathers and in single scapular feathers of 21 individual adult Gannets, Grassholm, 17 August 1986. Fitted line is simple regression of mantle against scapular data. Median concentrations do not differ significantly (10.25 ug/g for mantle v. 8.93 for scapulars: Wilcoxon signed rank test, Z = 1.67, P = 0.095, n.s.).

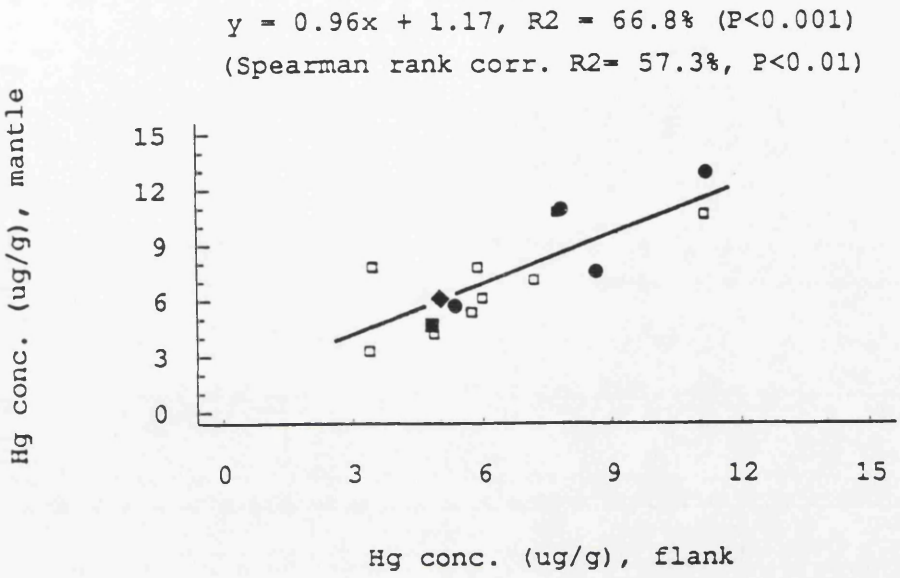


Figure 3.13 Relationship between mercury concentrations(ug/g dry weight) in pooled mantle feathers and in pooled upper-flank feathers of 15 individual adult Gannets, May-August 1985-88, mainly from Ailsa Craig, but some from Bass Rock (●), Sule Stack (■) and St Kilda (♦). Fitted line is simple regression of mantle against scapular data.

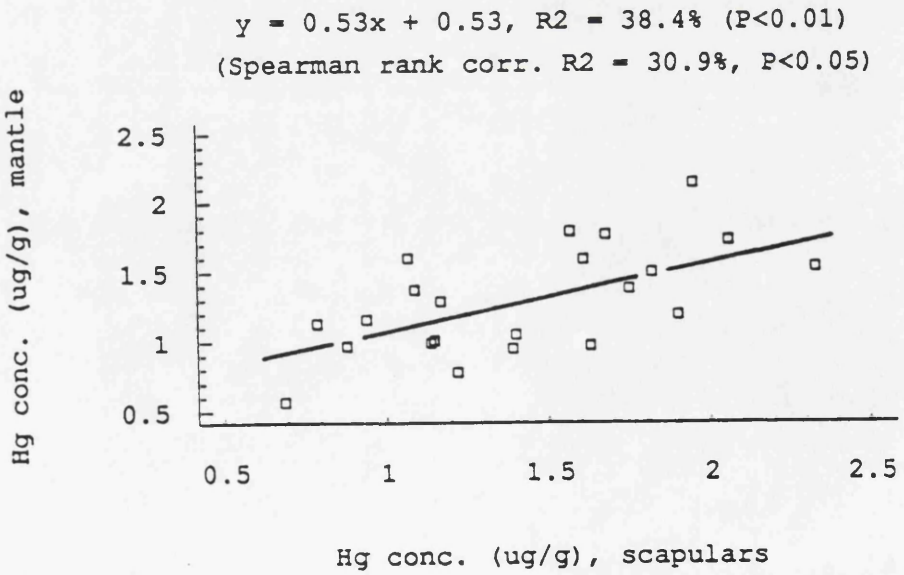


Figure 3.14 Relationship between mercury concentrations(ug/g dry weight) in pooled mantle feathers and in single scapular feathers of 22 well-grown Gannet chicks, Bass Rock, 4 September1986. Fitted line is simple regression of mantle against scapular data. Median concentrations do not differ significantly (1.24 ug/g for mantle v. 1.39 for scapulars: Wilcoxon signed rank test, Z = 1.57, P = 0.11, n.s.).

mercury concentrations did not differ significantly between the two feather-types (Wilcoxon signed rank test).

In well-grown Gannet chicks, there appears to be little variation between scapular feathers and pooled mantle feathers in mean mercury concentration for a given colony-year (Chapter 4). For chicks on the Bass Rock in 1986, mercury concentrations showed a significant positive correlation between the two plumage areas (Figure 3.14).

Mercury concentrations in different (randomly selected) scapulars of Gannet chicks were also positively correlated in small samples from three colonies, although in most cases non-significantly (Figures 3.15-3.17). Variation appeared to be high for some individual birds (although less so than would be expected for adults), but median mercury concentrations did not differ significantly between the paired samples.

3.3.4 Variation within individual feathers

This was not examined in detail for Gannets or Shags, as with these species it is rarely necessary to analyse small portions of individual feathers. (Even when sampling from valuable museum specimens, small body feathers can be used - see Chapter 6). However, for some Gannet analyses, mercury concentrations in the distal and proximal 'halves' (40%-60% by weight) of individual scapular feathers were analysed separately, providing a gross indication of variation within a feather.

For a combined sample of scapulars from 14 adult Gannets at the Bass Rock and Grassholm in August 1986, the relative concentration of mercury did not differ significantly between distal and proximal halves (Wilcoxon signed ranks tests).

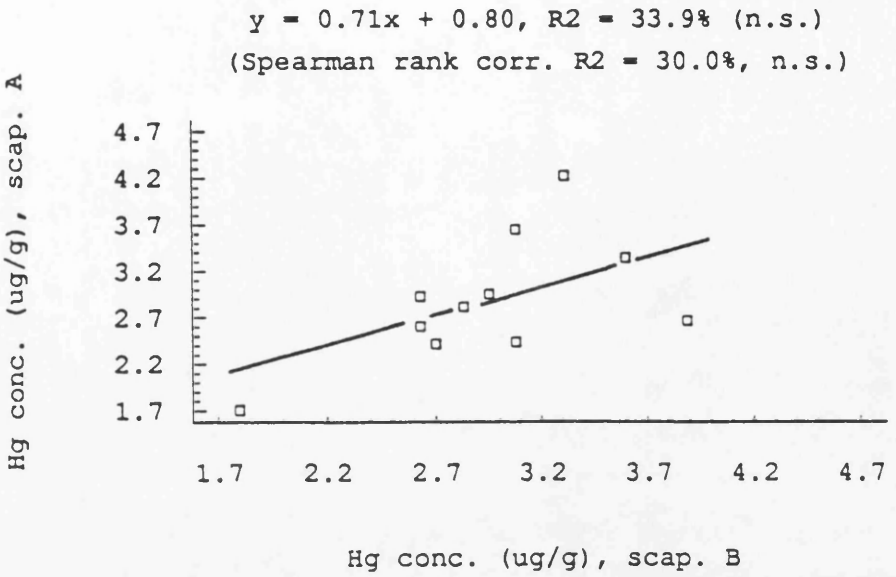


Figure 3.15 Relationship between mercury concentrations (ug/g dry weight) in different scapular feathers of 11 well-grown Gannet chicks, Grassholm, 17 August 1986 (each sample is a single feather). Fitted line is simple regression of 'scapular A' v. 'scapular B' data. Median concentrations do not differ significantly (2.84 ug/g for scapular A v. 2.82 ug/g for scapular B: Wilcoxon signed ranks test, Z = 0.58, P = 0.56, n.s.).

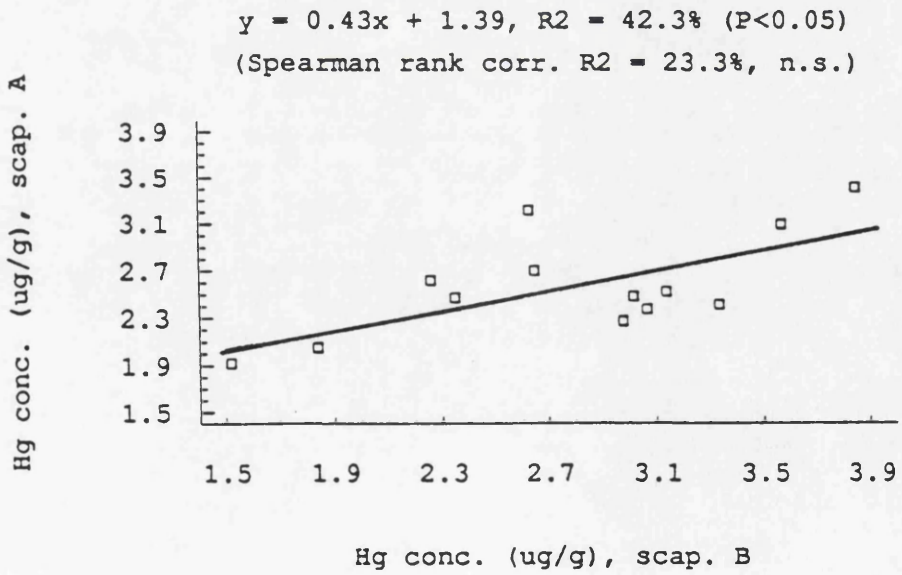


Figure 3.16 Relationship between mercury concentrations (ug/g dry weight) in different scapular feathers of 13 well-grown Gannet chicks, St Kilda, 5 August1986 (each sample is a single feather). Fitted line is simple regression of 'scapular A' v. 'scapular B' data. Median concentrations do not differ significantly (2.48 ug/g for scapular A v. 2.98 ug/g for scapular B: Wilcoxon signed ranks test, Z = 1.47, P = 0.14, n.s.).

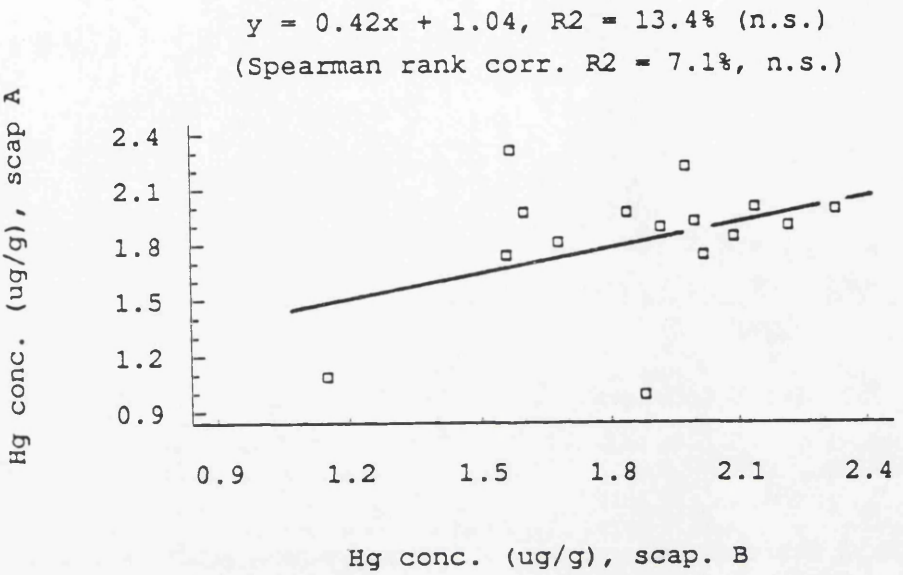


Figure 3.17 Relationship between mercury concentrations (ug/g dry weight) in different scapular feathers of 15 well-grown Gannet chicks, Bonaventure Island, 1 September 1986 (each sample is a single feather). Fitted line is simple regression of 'scapular A' v. 'scapular B' data. Median concentrations do not differ significantly (1.90 ug/g for scapular A v. 1.92 ug/g for scapular B: Wilcoxon signed ranks test, Z = 0.60, P = 0.55, n.s.).

Proximal and distal concentrations were significantly correlated with each other (Spearman rank correlation coefficient for nine Bass Rock birds = 0.92, $P<0.001$).

For 20 Gannet fledglings from Ailsa Craig (August 1986), mercury concentrations were significantly higher in the distal (median 2.44 ug/ g) than in the proximal halves (2.12 ug /g) of scapulars (Wilcoxon signed ranks test, $Z = 3.21$, $P<0.01$). As in the case of adult Gannets, distal and proximal concentrations were significantly correlated (Figure 3.18).

In two samples of scapular feathers from Great Saltee Shags (n = 9 and 20 individuals, respectively), mercury concentrations were significantly higher distally than proximally (median concentrations 1.12 v. 0.71 and 1.25 v. 1.14 ug/g-1, $Z =$ 2.61 [$p<0.01$] and 2.41 [$p<0.05$], respectively). For the sample of 20, distal and proximal concentrations were significantly correlated (Spearman $R^2 = 47.6\%$, $p<0.01$).

3.3.5 Seasonal variation

Mercury concentrations in pooled mantle feathers from breeding adult Gannets on the Bass Rock showed little evidence of any systematic variation over the breeding season, based on six samples from April-September (Figure 3.19). In part, this may reflect the inclusion of data from a three-year period 1986-88 (with the August 1986 sample having significantly higher mercury concentrations compared to 1987-88 samples).

Whether or not the 1986 sample is included, there is no significant trend in mercury concentration over the season. There was no significant variation between months in 1987-88, but there is a slight indication that average or median

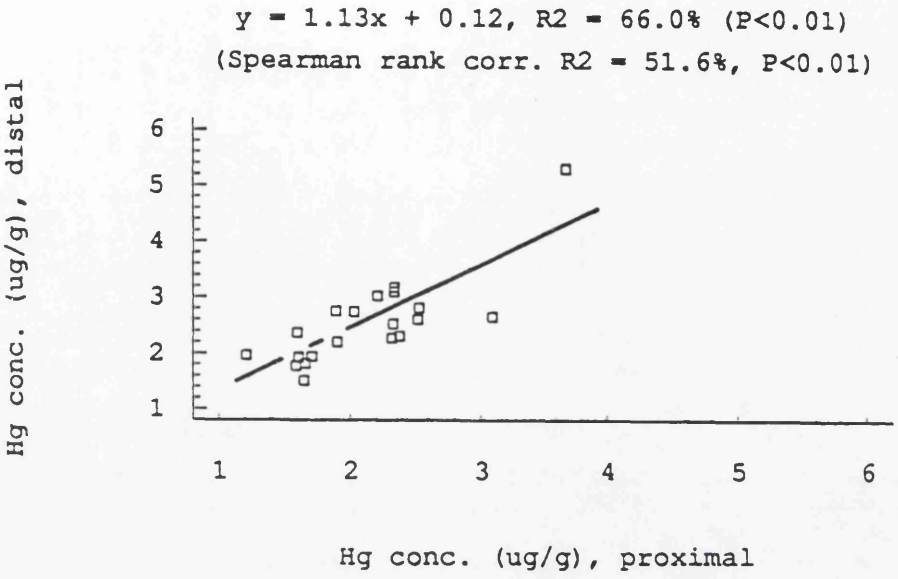
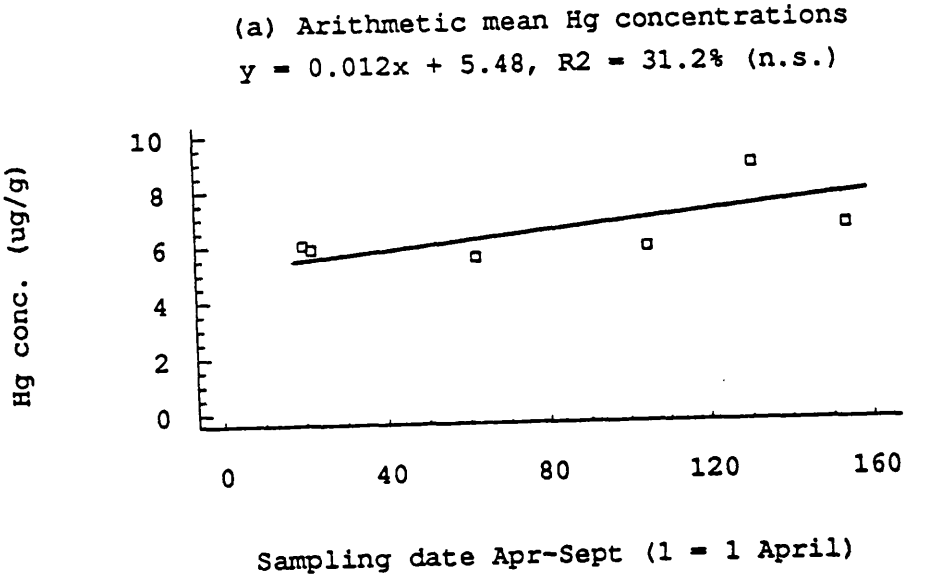


Figure 3.18 Relationship between mercury concentrations in distal and proximal halves of scapular feathers from Gannet fledglings, Ailsa Craig, August 1987. Fitted line is simple regression.



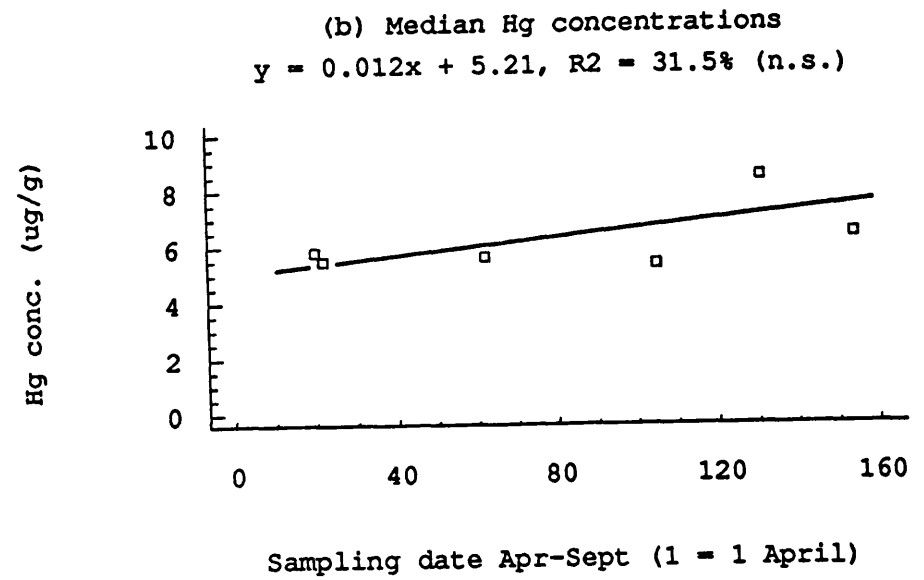


Figure 3.19 Mercury concentrations (ug/g dry weight) in mantle feathers of adult Gannets on the Bass Rock in relation to sampling date. Data points are (a) arithmetic mean and (b) median mercury concentrations, based on samples of 12-22 adults (August 1986; April and September 1987; April, June, and July 1988). Sample from each adult generally comprised 5-15 small feathers. Fitted lines are simple regression of mercury concentration against date (0 = 31 March, 160 = 7 September) - neither regression is significant. There was significant variation between the six 1986-88 samples (Kruskal-Wallis test-statistic = 31.23, $P<0.001$), but not between the five 1987-88 samples (K.-W. statistic = 4.17, $P = 0.38$).

mercury concentrations in mantle feathers of adult Gannets may have increased later in the season (perhaps with a dip around the middle of the breeding season).

With more comprehensive data from a single season, such a pattern might be confirmed to occur, if feathers renewed at the start of a moult cycle tend to have relatively high concentrations of mercury (section 3.3.2) and if individual body-feathers tend to be renewed at the same stage of each moult cycle. If both these assumptions held true, the average mercury content of the plumage would tend to decrease as 'high-mercury' feathers are moulted, then increase again as these are replaced by newly grown feathers. Moult of adult body-feathers had apparently not yet begun when April samples were collected from Bass Rock adults, but was very evident by early May in 1987 (Walsh, unpublished).

No information was collected on possible seasonal variations in the mercury levels in feathers of well-grown Gannet chicks. However, most samples were collected at a similar stage of the season (generally mid August to early September at British and Irish colonies, around the peak fledging period). Where well-grown chicks were present at colonies earlier in the season, they were generally too sparsely distributed to allow adequate sampling without excessive disturbance.

Downy Gannet chicks of various ages are available for sampling on a wider spread of dates, so any seasonal effects are potentially more important. On the Bass Rock in 1988, mercury concentrations in mantle down were significantly higher for samples collected during 13-15 July than for samples collected during 19-20 August (Table 3.6). Downy chicks sampled on the latter date were about a week older than those in the first sample (based on estimates of median age). However, the lack of any relationship between mercury concentration in down and the size of chicks within each sample (section 3.3.1) indicates that age differences alone cannot account for differences between the July and August samples.

Table 3.6 Mercury concentrations in mantle down from two samples of Gannets chicks on the Bass Rock, 1988. Mercury concentrations differed significantly between the July and August samples ($t_{52} = 6.74$, $P<0.001$; Mann-Whitney $Z = 5.05$, $P<0.001$). Biometric data are also presented for these chicks, but note that, within each sample, there was no significant relationship between mercury concentration and chick-size (Figures 3.3-3.4).

	14 July	20 August
Sample size (chicks)	38	16
Arithmetic mean mercury concentration (ug/g) ± s.d. (range)	2.10 ±0.43 (1.39-2.99)	1.30 ±0.33 (0.71-1.90)
Median mercury concentration (ug/g)	2.07	1.31
Median weight of chicks (g) (range)	2300 (890-3250)	- -
Median wing-length (mm) (range)	155 (58-248)	183 (117-281)
Median length of 10th primary (mm) (range)	9 (0-64)	25 (0-93)
Estimated median age of chicks (weeks)* (range)*	4-5 (2.5-7)	5-6 (3.5-8)

*Estimated ages based on plumage and biometrics (cf. Nelson 1978, Kirkham & Montevecchi 1982).

Great Saltee was the only other colony where downy chicks were sampled at different stages of the season, although in different years. In this case, the mercury concentration in mantle down was significantly lower in samples collected on 25 June 1986 (arithmetic mean ±s.d.1.77 ±0.39 ug/g, median 1.76 ug/g, n= 16) than on 8 August 1987 (mean 2.06 ±0.39, median 2.06 ug/g, n = 33) ($t_{47} = 2.47$, $P<0.05$; Mann-Whitney test, $Z = 2.31$, $P<0.05$). However, this may have reflected a difference between years.

3.3.6 Use of moulted feathers

Collections of moulted body-feathers at two Gannet colonies in 1986 allowed direct comparison with mercury concentrations in body-feathers plucked from adult Gannets at the same time (Table 3.7). Because the moulted samples were not assignable to individual birds, and were pooled for analysis, statistical comparison is not possible. However, it can be seen from Table 3.7 that mercury concentrations in the moulted samples were comparatively low, while showing a similar degree of inter-colony difference.

There are several possible explanations for the lower concentrations in the moulted feathers. Some of these feathers may have come from immature, non-breeding birds at each colony, but this is unlikely to have had a major influence on the results (not least because far fewer immature than adult birds were present). The moulted feathers will also have come from a range of plumage areas, and so may not be directly comparable with mantle feathers. However, variation in mercury concentration appears to be small between different types of body-feather (section 3.3.3).

Table 3.7 Mercury concentrations (ug/g dry weight) in body-feathers of adult Gannets at three colonies, August 1986: comparison between pooled samples of moulted feathers and mantle-feathers plucked from individual adults.

	*moulted body-feathers		------plucked mantle-feathers------			
	pooled mean	(sample size)	median	arithmetic mean	geometric mean	(sample size)
Bass Rock	4.84	(7)	8.48	8.94	8.58	(31)
Grassholm	6.72	(10)	10.25	11.61	10.58	(21)
% difference	+39		+21	+30	+23	

*5-9 small, moulted body-feathers analysed per sample, with mercury concentrations weighted by the number of feathers to provide a pooled mean figure. (Pooling by weight of feathers per sample gave almost identical figures.)

A more likely explanation is that moulted body-feathers collected in August 1986 had, on average, been recently moulted. Body moult had probably been in progress, on a large scale, since early May (as noted in 1987), so collections of moulted feathers in August were probably biased towards the middle of the moult period. Mercury concentrations in those feathers are likely to have been comparatively low (section 3.3.2), in this case much lower than the average mercury-concentration in 'live-plucked' feathers. The latter category included some feathers replaced after early moult, and likely to contain relatively high concentrations of mercury (section 3.3.2).

Individual, large moulted feathers of Gannets were not analysed in this study. However, analysis of large feathers (remiges, rectrices and scapulars) sampled from corpses or live birds (sections 3.3.2 and 3.3.3) suggests that variation in mercury concentrations would be high (see Discussion). This is likely to be true even if the relative position of a feather within, e.g., a sequence of wing- or tail-feathers is known (based on size and shape of the feather), because of the complexity of Gannet moult (Introduction, this chapter).

3.3.7 Sex

For some August-September samples of adult Gannets, the sex of individual birds was tentatively assessed on the basis of the buff coloration of the crown and nape. In most adult females, this coloration has become paler and patchier than in males by this stage of the season (Nelson 1978) No significant differences in the mercury concentration in mantle feathers was noted between apparent males and females (Table 3.8). There was a slight suggestion that mercury concentrations were lower in 'female' birds, but samples were small and the sex of individual birds was not known for certain.

Table 3.8 Mercury concentrations (ug/g) in samples of pooled mantle feathers of adult Gannets in relation to probable sex of each bird (see text); n = number of birds. No significant differences between sexes were detected (t-tests on arithmetic and geometric means, Mann-Whitney tests on medians).

Colony (date)	probable sex	n	mercury concentration (ug/g dry wt)				
			arithmetic $\bar{x}$	s.d.	range	median	geometric $\bar{x}$
Bass Rock	female	7	5.16	1.75	2.86-7.38	5.26	4.87
(September 1986)	male	6	7.70	2.80	3.78-10.99	7.96	7.22
Grassholm	female	6	12.36	5.08	8.09-21.73	11.83	11.62
(August 1986)	male	8	12.83	6.27	5.57-22.52	10.73	11.56

3.4 Discussion

Age

The comparatively few studies of mercury allowing direct comparison between soft-tissues of adult and nestling/fledgling seabirds from the same populations all show higher concentrations in adults (e.g. Koeman et al. 1973; Jensen et al. 1972; Blus et al. 1977). This is also the case where comparable feather-samples have been used, in studies of Herring Gulls *Larus argentatus* (Struger et al. 1987), Red-billed Gulls *L. novaehollandiae* (Furness et al. 1990) and Great Skuas *Catharacta skua* (Thompson et al. 1991). As with Gannets and Shags (section 3.3.1), feather concentrations in body-feathers of adult Herring Gulls and Great Skuas were at least twice as high as in chicks; the difference was slight for Red-billed Gulls . Various studies of other bird groups have also found mercury concentrations substantially higher in feathers of adults than of chicks or juveniles (i.e. fledged birds before they begin their first moult), with average adult:chick ratios varying 1.4-10 (e.g. Driver & Derksen 1980; Lindberg & Odsjo 1983; Goede 1985).

Nestling and adult birds of any species are clearly exposed differently to mercury in the environment, if only on a temporal scale, as adults can accumulate mercury over a longer period (at least, the period since their last moult). Both age-classes may reflect environmental mercury levels on the breeding grounds, but adults may also reflect mercury in other habitats or areas (especially wintering areas if different). Where exposure to mercury in the breeding area is much higher than elsewhere, adult:chick ratios might be expected to be lower than otherwise expected (as may have been the case for Ospreys *Pandion haliaetus* in parts of Sweden (Jensen et al. 1972).

Variation in exposure to mercury may also be higher among adults than among nestlings of a given population (e.g. reflecting variations in non-breeding areas), and this may account for the generally lower variability of mercury concentrations in seabird chicks. This is clearly seen in feathers from Shag and Gannet chicks, and has also been shown for Great Skuas (Thompson et al. 1991). In Red-billed Gulls (Furness et al. 1990), mercury concentrations were very variable in both adults and chicks (Coefficients of Variation 64% in adults, 57% in chicks), perhaps indicating that, even during the chick-feeding period, individual adults were specialising in their feeding areas or habits.

The generally higher mercury levels in feathers of adults may facilitate accurate analysis, as problems of 'sensitivity' caused by low tissue concentrations are obviated. The higher variability of samples from adults, however, will reduce the analyst's ability to demonstrate significant differences between samples.

Short-term, age-related variations in mercury concentrations could potentially increase the variability of results for chick plumage. This was examined for mantle-feathers of nestling Great Skuas aged on the basis of wing-length, but no relationship was found (Thompson et al. 1991).

The above studies relate to mercury levels in the 'definitive' plumage of chicks or fledged, pre-moult juveniles, as opposed to the downy feathers of younger nestlings. Very few studies have looked at mercury levels in chick down, and there is little information on possible age-influences. Lambertini (1982) compared mercury concentrations in down from Audouin's Gull *Larus audouinii* and Yellow-legged Gull *Larus michahellis* chicks on a Mediterranean island, using down cut from the upperparts 'immediately after hatching.' Mercury in those feathers was thus believed to derive wholly from that in the egg. Average concentrations were at least twice as high (15 ug/g for Audouin's Gull) as for Gannet chicks in the

present study. It may be that concentrations in down from newly-hatched chicks will be higher than in older chicks. There are no data to test this, however.

Gannet chicks are naked and much less well-developed than gull chicks at hatching. Adequate samples of down for analysis were not available until Gannet chicks were a week or more old, from which stage no relationship between age and mercury concentration was evident over a four- to seven-week period (section 3.3.1). Downy Gannet chicks of any age would thus appear suitable for analysis, although adequate samples are more readily obtained from older chicks.

Several studies have looked for age-related variations in mercury content of the plumage of 'adult' birds, as such variation could (a) increase the variability of mercury concentrations within a population sample and (b) lead to false conclusions regarding exposure of different populations to mercury (if age-structures differ between populations). Work on chicks (see above) suggests that there might be a gradual increase in plumage mercury-concentrations during at least the first few years post-fledging. This has not been demonstrated, but few studies have sampled adequate numbers of immatures/'young adults' to test for this. For adult-plumaged birds as a whole, no relationship with age was evident for Great Skuas (Thompson et al. 1991), Red-billed Gulls (Furness et al. 1990) or Herring Gulls (Hutton 1981).

For Gannets, this study indicates that the variability of mercury concentrations in down or definitive feathers of chicks is of a similar magnitude to variability in fresh Gannets eggs sampled from the same colonies. Further comparisons between results for chicks and eggs (a favoured tissue for pollutant analysis in seabirds) are made in Chapter 4.

Sex

As with age-related variation, there has been little work on possible sex influences on mercury concentrations in bird tissues. The only detailed study for a sample of seabirds has been that of Thompson et al. (1991), who found no significant difference between plumage mercury-concentrations of male and female Great Skuas (using small body-feathers). In other groups, Lindberg & Odsjo (1983) found no significant differences between male and female Peregrine Falcons *Falco peregrinus* (although this was based on individual primary and tail feathers). Even for soft tissues, there is little evidence for any sex-related variation (e.g. Bacher & Norman 1984 for several waterfowl species).

Moult and feather-growth

For a range of bird species, analyses of mercury concentrations have shown decreases of mercury along linear sequences of flight-feathers (especially primaries) and tail-feathers. Furness et al. (1986) reviewed the available studies, and provided data for primary sequences of nine species of seabird. In almost every case, mercury concentrations of primary feathers were highest in the innermost feathers and decreased gradually towards the outermost. Previous workers had often interpreted such patterns as indicating a decline in exposure to mercury over the period of moult, basing this on the assumption that dietary intake at the time of feather-growth was the critical factor (e.g. Johnels & Westermark 1969). However, Furness et al. concluded that the patterns seen in primary and other feather-sequences in fact reflected the order in which feathers were moulted. Under this model, mercury accumulates in soft tissues during the period between moults, is mobilised at the beginning of moult, and becomes incorporated in growing feathers (with highest concentrations in those grown first).

Braune (1987) found a similar relationship between mercury concentration and moult-position of primary feathers in several seabirds. For auks (Alcidae), which moult and re-grow all their primaries near-simultaneously, no such pattern was seen. Common Terns *Sterna hirundo* also differed from the general pattern, in that mercury concentrations declined sequentially from primary no. 1 (innermost) to primary 4, but increased again in primary 5 with a separate decline to primary 10 (outermost). This matched the particular pattern of moult in this species, where adults moult primaries 1-4 before autumn migration, then 'suspend' moult before resuming at primary 5 in their winter quarters (Cramp 1985). A build-up of mercury in soft-tissues during the period of moult-suspension could thus occur, to produce a higher concentration than 'expected' in primary 5.

An adult Osprey *Pandion haliaetus* studied by Jensen et al. (1972) also showed an 'unexpected' pattern of mercury concentrations along its primaries. In this case, concentrations declined from primary 3 to primary 6, with a higher concentration in primary 7 and a further decline to primary 10 (outermost). Moult in Ospreys is complex during the first five years of life, and there may be two overlapping moult sequences within the primaries (because of moult cycles overlapping between different years). This could account for the pattern of mercury concentrations seen. In older Ospreys, moult is suspended during autumn migration (Cramp & Simmons 1980), and this could also account for the pattern.

Data for an adult Gannet (section 3.3.2) are consistent with the influence of moult sequence on mercury concentrations in individual feathers. In this species, moult of flight- and tail-feathers is also complex, and there are usually about three active 'centres' of moult within a sequence of primaries (see Introduction).

When analysing individual flight- or tail-feathers, some knowledge of the timing and sequence of moult is thus required to place the mercury concentrations in context. For species with a relatively simple moult-sequence, comparisons of mercury concentrations between populations or individuals could be based on feathers from a known position within a moult-sequence. For a species such as Gannet, however, moult can begin at different positions within feather-sequences of different birds.

Number and type of feathers sampled

An obvious implication of the influence of moult on feather mercury-concentrations is that pooled samples of feathers are preferable to individual feathers (possibly of unknown position within a moult-sequence). Furness et al. (1986) demonstrated the lower variability of mercury concentration of pooled body-feathers compared to single flight- or tail-feathers for several species of seabird. This is confirmed here for adult Gannets (section 3.3.3).

If different plumage areas moult, on average, at different stages within a moult-cycle, the average mercury concentrations of these areas are also likely to vary. For example, Furness et al. (1986) explained differences between plumage areas of a Great Skua and a Kittiwake *Rissa tridactyla* on the basis of timing of moult in each feather-tract. Feathers from the head and neck of those species showed more variable mercury concentrations than other 'body-feathers', perhaps a consequence of a partial pre-breeding moult of head-feathers. (Pooling of feathers in samples will still tend to reduce variability, however.) Various other studies have indicated differences between plumage areas (e.g. Doi & Fukuyama 1983, Buhler & Norheim 1982), although the differences are usually difficult to establish conclusively from the available sample-sizes.

Moulted feathers

Moulted flight-feathers or tail-feathers have been used by various workers to assess the exposure of, in particular, raptors to mercury (e.g. Berg et al. 1966). Unless the position of these feathers within a moult-sequence is known, however, assessments of mercury exposure are difficult to make. At seabird colonies, it is usually possible to sample large numbers of moulted, small body-feathers, and pooled samples of these are potentially more useful, as variability is reduced. As noted in section 3.3.6, it may not be possible to compare mercury concentrations directly with unmoulted feathers sampled directly from birds. It may also be important to standardise the stage of the season (relative to onset of moult) at which moulted feathers are collected.

A potential problem at mixed seabird colonies is that small feathers may not be identifiable to species. It is easy to distinguish between different families of seabird on the basis of the barbule structure of the feather-bases, and this will allow separation, e.g., of Gannet from gull feathers (Brom 1986). Species-separation of more closely-related seabirds is usually not possible, however.

Seasonal variations

There is little evidence at present for seasonal variation in the average mercury content of the plumage of either adult or nestling seabirds. For adults of species which moult during the breeding season (e.g. Gannet), it might be expected that mercury levels in plumage would vary somewhat over the season (see section 3.3.5

and Chapter 2, section VI). Significant variation may be difficult to demonstrate, and perhaps so small as to be of little consequence compared to other variables. Variability of mercury concentrations should, in theory, be lowest in feather samples collected outside the moult period (e.g. in April for Gannets). For both adults and chicks, broad standardisation of collection dates between years or colonies would help to reduce possible effects.

Variation within individual feathers

Limited information is available on variation of mercury concentrations between different feather-parts, and most relates to large feathers sampled from raptors or waders. Goede & de Bruin (1984) found that mercury concentrations in the vane of specific primaries from two species of wader averaged 1.4 times higher than in the feather shaft; in each species, vane and shaft concentrations of mercury were significantly correlated. However, they recommended that, when using older feathers (especially moulted ones), the shaft should be analysed in preference to the vane, since the vane might be more subject to external contamination. Doi & Fukuyama (1983) also found mercury concentrations slightly higher in the barbs (vane) than the shaft of feathers of adult birds. In a sample of scapulars from adult Gannets, no significant difference was detected between basal and distal halves of the feathers (section 3.3.4).

Lindberg & Odsjo (1983) compared mercury concentrations in the tips and bases of secondary and tail feathers of two fully-feathered Peregrine Falcon chicks ; concentrations were ca. four times higher at the feather tip than at the base (although the weight of feather sampled was not stated). They interpreted this as transfer of mercury from the egg to early-grown parts of feathers, but this is a rather simplistic view (which omits mention of the preceding downy feathers). For

three samples of scapulars from Gannet and Shag chicks, less marked variation was seen, but concentrations were significantly higher in the distal halves.

These variations within nestling feathers could, in part, reflect the relative contribution of vane and shaft to the distal and basal sample-weights (if mercury concentrations are lower in the shaft). It is possible, however, that a growth-related influence is operating (e.g., mercury concentration highest in first-grown parts of feather), which might be predicted from the moult-influences seen in adult birds (see above). This might be clarified by analysis of part-grown feathers at various stages of development.

To minimise such influences (especially in chicks), mercury-analysis of entire feathers (or as much of the feather as possible above the base) may be advisable. Part-grown feathers, less than ca. 50% of their 'final' weight should perhaps be excluded (except when included in pooled samples of feathers).

Conclusions

1. Feathers of nestlings (including down) appear to be them most useful feathers for assessment of geographical variation in seabird exposure to mercury (showing similar low variability to fresh eggs).

2. The number and type of feathers analysed are less important for chicks than for adults. If large feathers are sampled, take as much of the feather as possible, to reduce possible effects of variation of mercury-concentrations within feathers.

3. Further work on possible seasonal variations in mercury content of chick feathers would be useful.

4. If adult feathers are used, pooled samples of small body feathers are best. Age- and sex-influences appear to be slight or non-existent.

5. Adult feathers should, if possible, be sampled between moult periods, although evidence for seasonal variation in average mercury content of small body-feathers is poor.

6. Moulted adult feathers are potentially useful if small body-feathers are collected at similar stage of season at different colonies or in different years. (Larger feathers, from the wing or tail, may also be useful for species - not including Gannet - where a feather's position with a moult sequence can be judged on size and structure alone.)

Chapter 4

GEOGRAPHICAL VARIATION IN MERCURY LEVELS IN FEATHERS OF NORTHERN GANNETS

4.1 Introduction

4.2 Methods

4.3 Results

4.3.1 Mercury in feathers of adult Gannets

4.3.2 Mercury in feathers of Gannet chicks

4.3.3 Mercury in Gannet eggs

4.3.4 Mercury in feathers of adult and nestling Shags

4.3.5 Gannet diet, and mercury in Mackerel Scomber scombrus

4.4 Discussion

4.4.1 Comparison of geographical patterns of mercury concentration shown by different sample types

4.4.2 Other studies of geographical variation in mercury levels of North Atlantic seabirds

4.4.3 Factors contributing to geographical patterns seen, and evidence from other monitoring or baseline studies on mercury in North Atlantic waters

4.4.4 Influences of at-sea distribution, seasonal movements and foraging ranges of Gannets and Shags

4.1 Introduction

There have been many studies of mercury concentrations in feathers of wild birds since the 1960s (e.g. Berg et al. 1966, Goede 1985, Furness et al. 1986). However, comparatively few attempts have been made to assess geographical variation in environmental mercury levels using such samples (examples for land-birds include Hakkinen & Hasanen 1980, Lindberg & Odsjo 1983) . Work on geographical variation in mercury levels of seabirds has, in most cases, been based on eggs or on soft-tissues (e.g. Renzoni et al. 1986, Parslow & Jefferies1975). Only a few studies have assessed geographical variation in mercury levels of seabird feathers, including work on auks and several other species in parts of the North Atlantic (Appelquist et al. 1985, Thompson et al. 1992).

Mercury levels in soft tissues of Northern Gannets *Morus bassanus* found dead around British coasts by Dale et al. (1973) and Parslow et al. (1973) were higher than in many other seabirds examined at the same time, with the exception of some inshore species such as Cormorant *Phalacrocorax carbo* and Shag *P. aristotelis*. Analyses of Gannet eggs in the 1970s also revealed high concentrations of mercury (among other pollutants) for some colonies. These results, and the Gannet's fairly wide distribution in the North Atlantic, suggested it might be a suitable species for wide-scale assessment of geographical patterns in environmental levels of mercury. An attempt was thus made to sample feathers of adult and nestling Gannets at as many colonies as possible during 1986-88.

The influence of such variables as age of bird and type of feather sampled was examined in Chapter 3. Some samples have been used in assessment of historical changes in mercury levels (Chapter 6). The present chapter reports on the geographical variation found for specific types of feather-sample. For British and

Irish colonies, comparative data are also provided for Gannet eggs, which have been used more widely as a tissue for mercury-monitoring (Newton et al. 1989, 1990). Mercury concentrations in feathers of Shags are also examined, to assess whether geographical patterns are similar for this more inshore-feeding species. The role of variation in Gannet diet is also considered.

4.2 Methods

A summary of methods used for sampling feathers, and analysing mercury concentrations, is given in Chapter 3, with fuller details in Chapter 1. In addition, samples of fresh or addled (unhatched, failed) eggs were collected from five Gannet colonies around Britain and Ireland (see Chapter 1 for details of preparation and analysis). The locations of all sampled colonies are shown in Figures 4.1-4.2. Samples of lateral muscle from Mackerel *Scomber scombrus* caught commercially in inshore waters in the Firth of Clyde (near Ailsa Craig), off the Firth of Forth (near Bass Rock) and off Co. Wexford (near Great Saltee) in 1987 were also analysed.

All statistical comparisons made here are based on non-parametric tests (Mann-Whitney U-tests and Spearman's rank correlations), because of deviations from normality shown by mercury concentrations in some of the samples. Median mercury concentrations provide the basis for most comparisons made, but arithmetic means, standard deviations and geometric (logarithmically-derived) means are also presented to aid comparison with other studies.

Where feathers have been sampled from a given colony in more than one year, data are presented separately if more than ten birds were sampled each year. Where several samples were taken from a colony in a given year, data are combined in tables unless significant differences were noted. (Data were examined for seasonal variation in Chapter 3.)

Limited data were collected on Gannet diet at six colonies during 1986-88, by examining regurgitations from chicks or breeding adults, and will be considered briefly here.

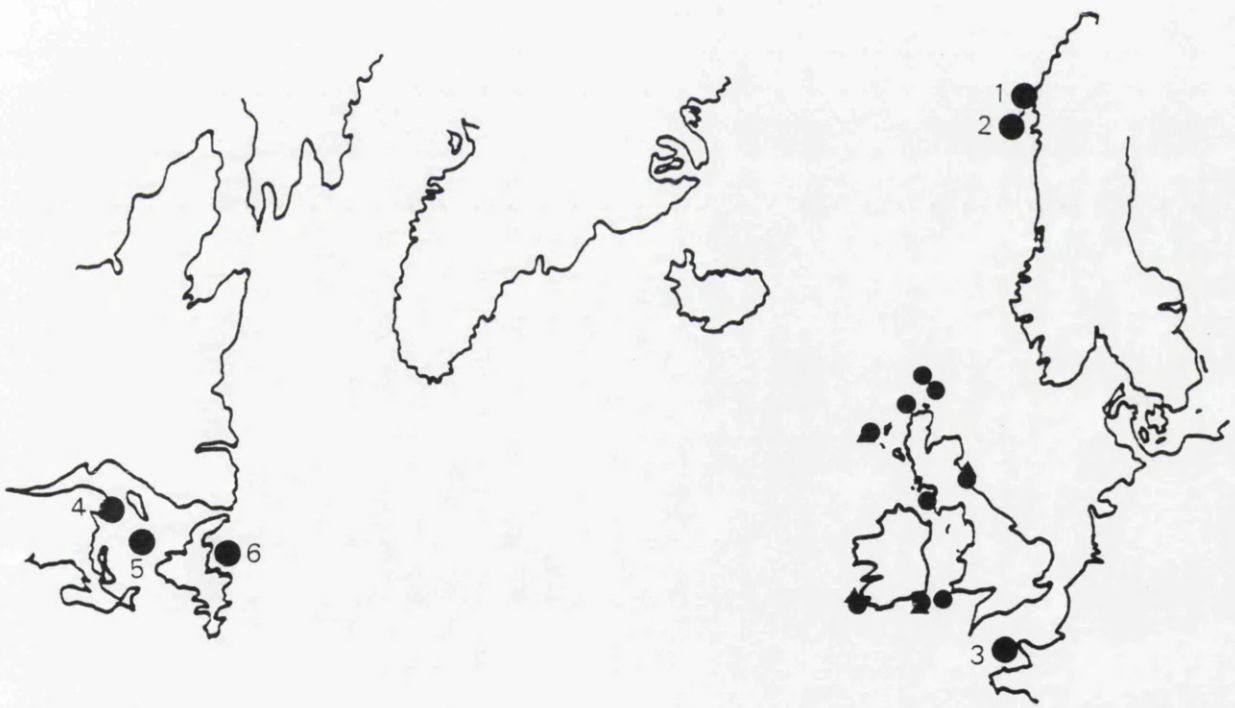


Figure 4.1 Gannet and Shag colonies from which feathers were sampled during 1986-88. See Figure 4.2 for British and Irish colonies. Norwegian colonies are: 1 = Skarvlakken; 2 = Hovsflesa. Channel Islands: 3 = Alderney (Gannets)/Jersey (Shags). Canadian colonies: 4 = Bonaventure Island; 5 = Bird Rocks; 6 = Funk Island.

Figure 4.2 British and Irish colonies from which feathers of Gannets (●), Shags (▲) or both species (■) were sampled during 1986-88.

4.3 Results

4.3.1 Mercury in feathers of adult Gannets

Individual scapular feathers were analysed for Gannets from two Norwegian, three British and one Canadian colony (Table 4.1). As discussed in Chapter 3, mercury concentrations in scapulars from any given colony tend to be very variable. It thus proved difficult to distinguish clear geographical patterns. Median concentrations of mercury ranged from 5.1 to 10.6 ug/g, but few inter-colony differences were significant. Concentrations were highest overall in samples from Hovsflesa (Norway) and Grassholm (south Wales), and lowest in samples from Hermaness (Shetland) and Bonaventure Island (Gulf of St Lawrence, Canada).

The range of variation in mercury concentrations was highest for scapular samples from Grassholm (2.1-24.9 ug/g). Most other samples included some feathers with high mercury concentrations (Figure 4.3). However, it was notable that feathers from Bonaventure Island all had concentrations < 10 ug/g.

Samples of pooled, small body-feathers were collected from five colonies around Britain and one in Canada (Table 4.2). These showed less variation than single scapulars in mercury concentrations for any given colony-year (cf. Chapter 3), but inter-colony differences are again not very clear-cut. Median mercury concentrations varied 4.6-10.2 ug/g between colony-years, highest for Grassholm. High concentration (medians 7.6-8.5 ug/g) were also recorded for Ailsa Craig (Firth of Clyde), Funk Island (eastern Newfoundland, Canada), and the Bass Rock (Firth of Forth) 1986 sample. Concentrations were significantly lower in 1987-88 samples from the latter colony.

Table 4.1 Mercury concentrations (ug/g dry weight) in single scapular feathers from adult Gannets at Norwegian, British, Irish and Canadian colonies in 1986. $\bar{X}a$ = arithmetic mean, $\bar{X}g$ = geometric mean.

Colony	date	n	mercury concentration (ug/g)					key to significance*
			$\bar{X}a$	s.d.	median	range	$\bar{X}g$	
Skarvlakken	Jul. 1986	13	**7.45**	3.98	**5.09**	3.52-15.79	**6.60**	A B - -
Hovsflesa	Jul. 1986	15	**9.56**	3.35	**10.59**	4.11-14.75	**8.90**	A - - -
Bass Rock	Au-Se.1986	71	**7.01**	3.68	**6.03**	2.15-18.02	**6.19**	- B C -
Hermaness	Jn. 1986-87	28	**5.63**	3.39	**5.09**	1.93-18.06	**4.91**	- B - D
Grassholm	Aug. 1986	24	**8.44**	5.90	**7.43**	2.13-24.88	**6.74**	A - C D
Bonaventure	Aug. 1986	47	**5.86**	2.08	**5.45**	2.46-9.49	**5.48**	- B - D

*Samples with the same letter do not have significantly different mercury concentrations (Mann-Whitney U-tests); for example, all colonies with 'A' in last column of table have mercury concentrations not significantly different from each other. Significance level is set as $p<0.01$ because of the number of comparisons made.

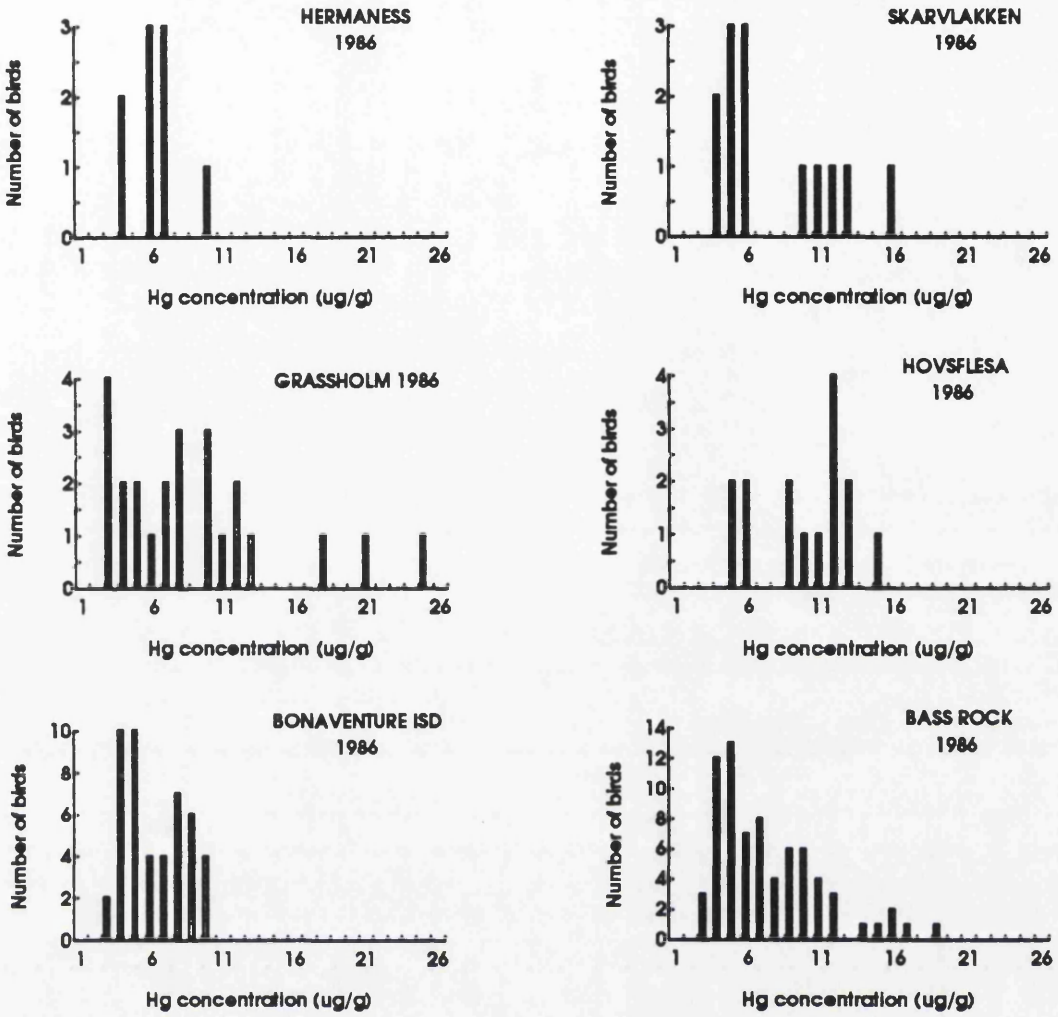


Figure 4.3 Mercury concentrations (ug/g dry weight) in scapular feathers of adult Gannets at North Atlantic colonies (single feather per bird).

Table 4.2 Mercury concentrations (ug/g dry weight) in pooled mantle feathers (usually 5-15 per bird) from adult Gannets at British, Irish and Canadian colonies in 1986-88. $\bar{X}a$ = arithmetic mean, $\bar{X}g$ = geometric mean. Data in *italics* were provided by R. W. Furness (unpublished).

Colony	date	n	mercury concentration (ug/g)					key to significance*
			$\bar{X}a$	s.d.	median	range	$\bar{X}g$	
Bass Rock	Aug. 1986	31	**8.95**	2.79	**8.48**	5.13-17.98	**8.58**	A- - - -
	Ap-Se. 1987	35	**6.36**	2.00	**5.87**	2.86-10.94	**6.07**	- B- - -
	Ap-Jul. 1988	61	**5.88**	1.89	**5.69**	2.55-13.13	**5.60**	- B C - -
Hermaness	May 1987	29	**4.80**	1.58	**4.72**	2.06-7.53	**4.52**	- - C - -
St Kilda	Jn-Jl.1986-87	18	**5.36**	2.86	**4.57**	2.90-12.30	**4.92**	- - C - -
Ailsa Craig	*Aug. 1985*	*12*	***10.48***	*10.95*	***7.92***	*3.0-43.8*	***7.88***	*A B C - -*
	Jn-Au.1986	11	**7.31**	3.00	**7.64**	3.76-13.39	**6.77**	A B C - -
Grassholm	Aug. 1986	21	**11.61**	5.25	**10.25**	5.11-22.52	**10.58**	A- - - -
Funk Isd**	Aug. 1987	14	**8.05**	3.05	**7.83**	3.40-16.63	**7.55**	A- - - -

*Samples with the same letter do not have significantly different mercury concentrations (Mann-Whitney U-tests); for example, all colonies with 'A' in last column of table have mercury concentrations not significantly different from each other. Significance level is set as $p<0.01$ because of the number of comparisons made.

**Funk Isd 1987: two mantle feathers per bird.

Mercury concentrations of up to 22.5 ug/g were recorded for mantle feathers from Grassholm (Figure 4.4) and one Ailsa Craig sample had a concentration of 43.8 ug/g. Since these were pooled samples of up to 15 feathers, the influence of any individual feathers with high mercury concentrations should have been reduced (see Chapter 3). It would thus seem that some individual Gannets from the Firth of Clyde and the Irish Sea had been exposed to particularly high environmental levels of mercury. Concentrations > 10 ug/g were also recorded for individual birds from some other colonies, notably Bass Rock in 1986.

It did not prove possible to sample feathers from individual adults at either of the colonies off Alderney, in the Channel Islands. However, moulted small body-feathers were collected in August 1986, and seven pooled samples of these mixed feathers were analysed. Weighting each sample by the number of feathers, the average concentration of mercury was 8.0 ug/g (range 6.1-9.4). Comparable figures for other colonies, also in August 1986, were 4.8 ug/g (4.1-5.6, n = 7) for the Bass Rock and 6.7 ug/g (6.1-7.3, n = 7) for Grassholm. Mantle-feathers sampled directly from Grassholm adults had the highest concentrations of any colonies. The figures from moulted feathers (see Chapter 3, Table 3.7) suggest that levels are even higher in Alderney birds.

4.3.2 Mercury in feathers of Gannet chicks

Juvenile feathers were sampled from well-grown Gannet nestlings, or recently fledged juveniles, from five British, two Irish and three Canadian colonies (Table 4.3, Figure 4.5). Median concentrations ranged 1.3-3.6 ug/g between colony-years, and inter-colony differences were much more clear-cut than in the case of feathers from adult Gannets.

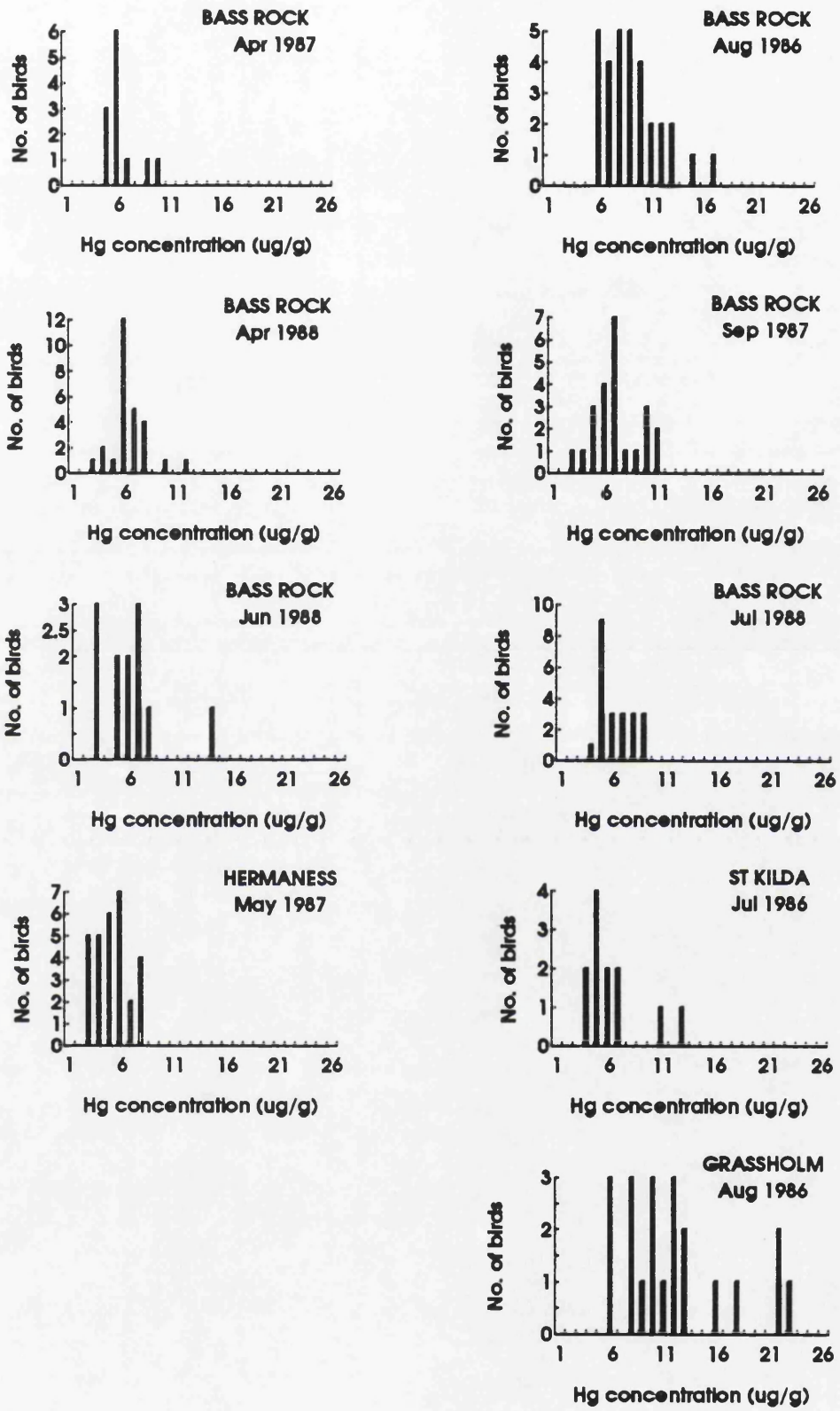


Figure 4.4 Mercury concentrations (ug/g dry weight) in mantle feathers of adult Gannets at North Atlantic colonies.

Table 4.3 Mercury concentrations (ug/g dry weight) in pooled mantle feathers or individual scapular feathers from well-grown Gannet chicks at British, Irish and Canadian colonies in 1985-88. $\bar{X}a$ = arithmetic mean, $\bar{X}g$ = geometric mean. Annual samples <10 are combined.

Colony	year	feather type*	n	mercury concentration (ug/g) $\bar{X}a$	s.d.	median	range	$\bar{X}g$	key to significance**
Bass Rock	1986	M	38	**1.36**	0.39	**1.33**	0.57-2.14	**1.30**	A - - - - - -
	1987	M	30	**1.47**	0.31	**1.48**	0.82-2.22	**1.44**	A - - - - - -
	1988	M	20	**1.33**	0.27	**1.36**	0.77-1.85	**1.29**	A - - - - - -
Noss	1986-87	S	37	**1.37**	0.19	**1.39**	1.01-1.75	**1.36**	A - - - - - -
St Kilda	1986	S	20	**2.46**	0.50	**2.59**	1.44-3.63	**2.49**	- B C - - - -
Ailsa Craig	1985-86	M	13	**2.49**	0.78	**2.21**	1.65-4.2	**2.38**	- B C - - F G
	1987	S	21	**2.35**	0.59	**2.32**	1.57-4.29	**2.27**	- B C - - F -
Grassholm	1986	M	62	**3.05**	0.60	**2.95**	1.48-4.47	**2.96**	- - - - E - -
Great Saltee	1987	M/S	30	**2.45**	0.52	**2.38**	1.72-4.46	**2.41**	- - C - - - -
Little Skellig	1987	M	33	**3.60**	0.44	**3.60**	2.67-4.36	**3.57**	- - - - - - -
Bird Rocks	1987-88	S/B	21	**1.81**	0.38	**1.69**	1.41-2.73	**1.77**	- - - D - - -
Bonaventure	1986	S	30	**1.90**	0.35	**1.90**	0.99-2.82	**1.86**	- - - D - F -
Funk Isd***	1987	PM	10	**3.12**	(0.30)	**3.11**	(2.72-3.67)	-	- - - - E - G

*Feather-type: S = single scapular per bird; M = 5-20 small mantle feathers per bird; B = two medium/large body-feathers per bird; PM = 2 small mantle feathers per bird, with 2-5 individual birds analysed in each pooled sample. Where both mantle and scapular feathers were analysed for a given colony-year, data for the largest sample are presented (no significant differences were noted between these feather-types: Chapter 4).

**Samples with the same letter do not have significantly different mercury concentrations (Mann-Whitney U-tests); for example, all colonies with 'A' in last column of table have mercury concentrations not significantly different from each other. Significance level is set as $p<0.01$ because of the number of comparisons made.

***Funk Island 1987: 10 samples analysed (31 chicks, 2 feathers per chick, 2-5 chicks per sample).

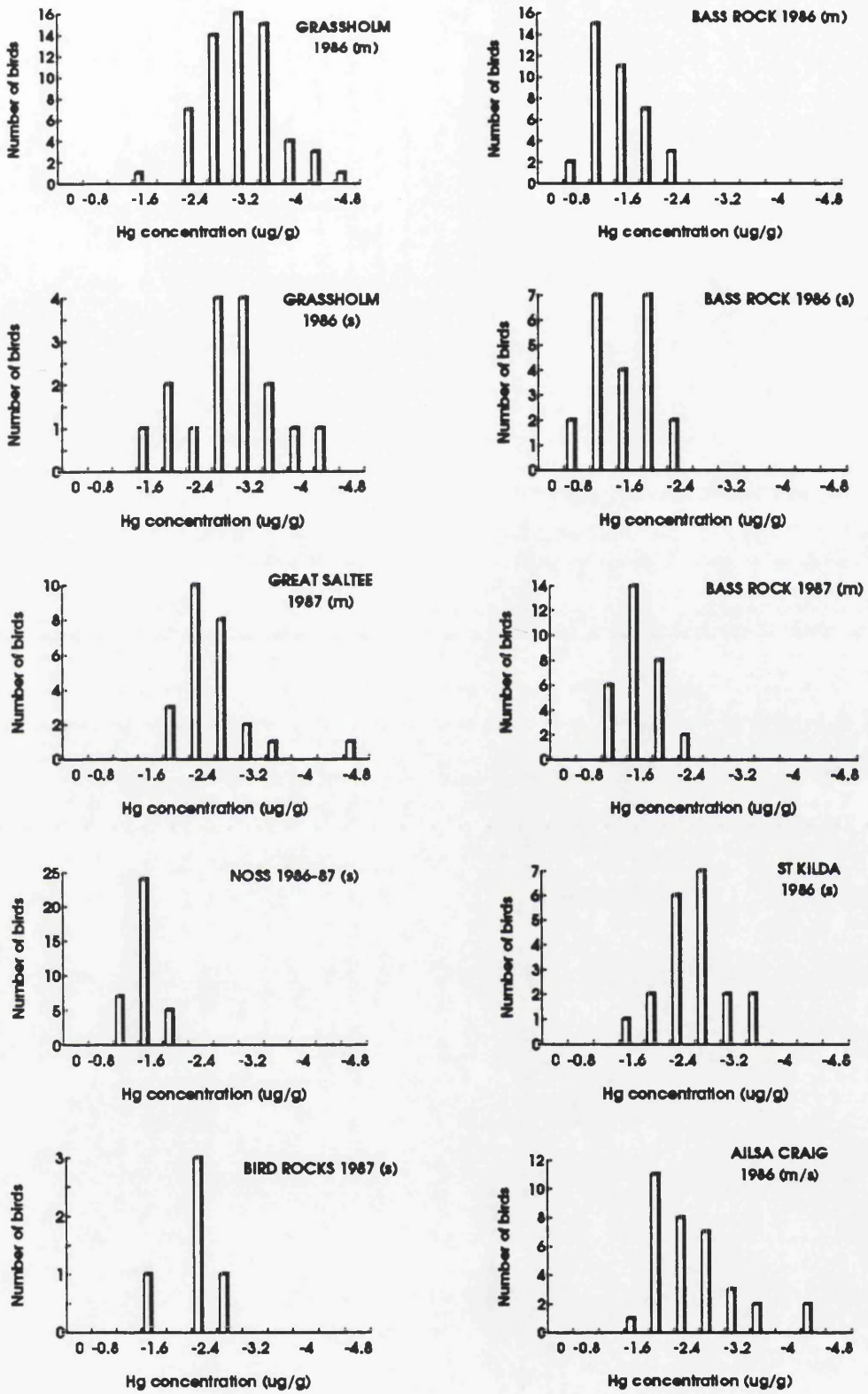


Figure 4.5 Mercury concentrations (ug/g dry weight) in feathers of well-grown Gannet chicks at North Atlantic colonies. Feather-types are indicated by: (m) = mantle feathers, (s) = scapular feather(s).

Concentrations were significantly lower (medians 1.3-1.5 ug/g) in feathers from the Bass Rock (Firth of Forth) and Noss (Shetland) than in any other samples. Chicks from Little Skellig (off SW Ireland) had significantly higher concentrations (median 3.6 ug/g) than those from all other colonies. Of the other colonies within Britain and Ireland, birds from Grassholm (southern Irish Sea) had the highest concentrations (2.9 ug/g). These colonies, on the whole, showed a broad trend of increasing mercury concentrations from the northeastern to the more western and southwestern colonies.

Of the Canadian colonies sampled, chicks from the two in the Gulf of St Lawrence (Bird Rocks and Bonaventure Island) had the lowest mercury concentrations (medians 1.7-1.9 ug/g). Figures for Funk Island, east of Newfoundland, were markedly higher (3.1 ug/g).

At four of the British and Irish colonies, adequate samples of downy feathers were also obtained, from less well-grown chicks (Table 4.4). Although age-related variation in the mercury concentration of down appeared to be lacking (Chapter 3), two samples from the Bass Rock in 1988 differed significantly from each other. If the median of these and other figures in Table 4.4. are compared, the same qualitative trend is seen as for definitive feathers: lowest concentrations for chicks from the Bass Rock, highest for Little Skellig, and next highest for Grassholm.

4.3.3 Mercury in Gannet eggs

Three British and two Irish colonies were sampled. Concentrations were lowest in eggs from the Bass Rock and Great Saltee (medians both 2.0 ug/g), and significantly higher in eggs from Grassholm and Ailsa Craig (2.9-3.1 ug/g) (Table 4.5).

Table 4.4 Mercury concentrations (ug/g dry weight) in down feathers of one- to eight-week old Gannet chicks at British and Irish colonies, 1986-88. $\bar{X}a$ = arithmetic mean, $\bar{X}g$ = geometric mean.

Colony	date	n	mercury concentration (ug/g)					key to significance*
			$\bar{X}a$	s.d.	median	range	$\bar{X}g$	
Bass Rock	July 1988	38	**2.11**	0.43	**2.08**	1.39-3.00	**2.07**	A - - -
	Aug. 1988	16	**1.30**	0.33	**1.32**	0.71-1.90	**1.26**	- B - -
Ailsa Craig	Aug. 1987	3	**2.43**	0.81	**2.87**	1.49-2.93	**2.32**	A B C D
Grassholm	July 1987	24	**2.95**	0.56	**2.82**	2.17-4.22	**2.90**	- - C D
Great Saltee	June 1986	16	**1.77**	0.39	**1.76**	0.84-2.61	**1.73**	A - - -
	Aug. 1987	33	**2.06**	0.39	**2.06**	1.38-2.82	**2.03**	A - - -
Little Skellig	July 1987	22	**3.04**	0.44	**2.93**	2.42-3.92	**3.02**	- - - D

*Samples with the same letter do not have significantly different mercury concentrations (Mann-Whitney U-tests); for example, all colonies with 'A' in last column of table have mercury concentrations not significantly different from each other. Significance level is set as $p<0.01$ because of the number of comparisons made.

Table 4.5 Mercury concentrations (ug/g dry weight) in fresh and addled eggs of Gannets from five British and Irish colonies in 1987. $\bar{X}a$ = arithmetic mean, $\bar{X}g$ = geometric mean.

Colony		n	mercury concentration (ug/g)					key to significance*
			$\bar{X}a$	s.d.	median	range	$\bar{X}g$	
Bass Rock	fresh	9	**1.94**	0.29	**1.97**	1.57-2.49	**1.92**	A - C
Ailsa Craig	fresh	10	**3.22**	0.94	**3.15**	2.03-5.23	**3.10**	- B -
	addled	14	**2.98**	1.11	**2.91**	1.27-5.01	**2.78**	- B C
	total	24	**3.08**	1.03	**3.08**	1.27-5.23	**2.91**	- B -
Grassholm	fresh	20	**3.19**	0.71	**3.16**	1.96-4.46	**3.12**	- B -
	addled	10	**3.27**	1.38	**2.84**	2.26-6.76	**3.09**	- B -
	total	30	**3.22**	0.96	**2.88**	1.96-6.76	**3.11**	- B -
Great Saltee	fresh	30	**2.11**	0.36	**1.97**	1.73-2.99	**2.08**	A - -
Little Skellig	addled	6	**2.76**	1.00	**2.65**	1.62-4.07	**2.61**	A B -

*Samples with the same letter do not have significantly different mercury concentrations (Mann-Whitney U-tests); for example, all colonies with 'A' in last column of table have mercury concentrations not significantly different from each other. Significance level is set as $p<0.01$ because of the number of comparisons made.

Both fresh and addled eggs were collected from the latter two colonies, and in each case mercury concentrations were not significantly different in the addled eggs. Standard deviations were higher in addled relative to fresh eggs, however.

4.3.4 Mercury in feathers of adult and nestling Shags

Mantle feathers from adult Shags were sampled at two colonies in Britain* and Ireland (Table 4.6). Mercury concentrations were slightly, but not significantly, higher in birds from Great Saltee (SE Ireland) than those from the Bass Rock (Firth of Forth). Additional samples over the same period from the Isle of May (also Firth of Forth) and Foula (Shetland) had significantly lower concentrations (data from Thompson et al. 1992).

Nestling Shags were sampled at five colonies, with additional data for one colony provided by D. R. Thompson (unpublished). Chicks from Foula had the lowest mercury levels in feathers (median 0.34 ug/g), and figures were also low for Bass Rock birds (0.6 ug/g) (Table 4.7). In contrast, a median concentration of 2.3 ug/g was recorded from chicks on Jersey (Channel Islands).

4.3.5 Gannet diet, and mercury in Mackerel Scomber scombrus

Mackerel and sandeels (Ammodytidae) were the most frequent food items overall at six colonies in Britain and Ireland, occurring on average in 37% and 27%, respectively, of regurgitations (averaged between colonies; n = 420 regurgitations in total). Some colony samples were small, or referred to only one or two dates. However, there appears to be a broad pattern, in that sandeels were more

frequently recorded at the northern, Scottish colonies, than at colonies in Wales and southern Ireland (cf. Discussion and Figure 4.8). The pattern for Mackerel was roughly the opposite. Other frequent food items were 'whitefish' (Gadidae) (average 15% of regurgitations) and Herring *Clupea harengus* (16%), while a mixed group of at least 11 other species of fish occurred on average in 31% of samples.

Mercury concentrations in lateral muscle of Mackerel from the vicinity of three Gannet colonies were low, averaging 0.07 (±s.d. 0.02) ug/g wet weight for both Firth of Forth (n = 12) and Wexford (n = 18) samples, and 0.10 (±0.05) ug/g for the Firth of Clyde (n =25) sample (differences not significant: Mann-Whitney U-tests).

Table 4.6 Mercury concentrations (ug/g dry weight) in pooled mantle feathers (usually 5-15 per bird) from adult Shags at British and Irish colonies in 1986-88. $\bar{X}a$ = arithmetic mean, $\bar{X}g$ = geometric mean. Data in *italics* are from Thompson *et al.* (1992).

Colony	date	n	mercury concentration (ug/g)					key to significance*
			$\bar{X}a$	s.d.	median	range	$\bar{X}g$	
Bass Rock	Jul. 1988	27	**3.90**	1.83	**3.03**	2.07-8.34	**3.58**	A - -
*Isle of May***	*1987-91*	*33*	***2.3***	*0.6*	-	*1.5-4.4*	***2.2***	- - -
Foula	*1987-91*	*40*	***1.9***	*0.6*	-	*0.8-3.8*	***1.8***	- - -
Great Saltee	May 1988	34	**4.42**	2.02	**4.43**	1.35-8.63	**3.95**	A - -

*Samples with the same letter do not have significantly different mercury concentrations (*t-tests* or Mann-Whitney U-tests); for example, all colonies with 'A' in last column of table have mercury concentrations not significantly different from each other. Significance level is set as $p<0.01$ because of the number of comparisons made.

**Isle of May 1988: sample of single scapulars from 16 adults also analysed: mean mercury concentration 3.90 ug/g ± s.d. 0.69 (range 2.12-5.12 ug/g).

Table 4.7 Mercury concentrations (ug/g dry weight) in single scapular feathers (1-2 per bird) from well-grown Shag chicks in 1986-88. $\bar{X}a$ = arithmetic mean, $\bar{X}g$ = geometric mean. Data in *italics* were provided by D. R. Thompson (unpublished).

Colony	year	n	mercury concentration (ug/g)					key to significance*
			$\bar{X}a$	s.d.	median	range	$\bar{X}g$	
Bass Rock	1988	21	**0.61**	0.14	**0.61**	0.35-0.84	**0.60**	A - -
Foula	*1987*	*20*	***0.33***	*0.08*	***0.34***	*0.22-0.47*	***0.32***	- - -
St Kilda	1986	8	**0.91**	0.31	**0.81**	0.61-1.35	**0.81**	A B C
Great Saltee	1986	14	**0.88**	0.16	**0.90**	0.58-1.18	**0.87**	- B -
	1988	21	**1.25**	0.39	**1.14**	0.70-2.27	**1.20**	- - C
Blasket Islands	1988	13	**1.36**	0.45	**1.42**	0.67-1.99	**1.28**	- - C
Jersey**	1987	7	**2.30**	(0.62)	**2.30**	(1.04-2.84)	**2.20**	- - -

*Samples with the same letter do not have significantly different mercury concentrations (Mann-Whitney U-tests). Significance level is set as $p<0.01$ because of the number of comparisons made.
**Jersey: 1 small mantle feather per bird, 1-5 birds per sample (n = 7 samples, comprising 25 individual birds).

4.4 Discussion

4.4.1 Comparison of geographical patterns of mercury concentration shown by different sample-types

Mercury concentrations in feathers of Gannet chicks, and in Gannet eggs, showed the most coherent geographical patterns. The broad pattern seen for these samples at British and Irish colonies was for mercury concentrations to increase from northeast to west and southwest (Figure 4.6). Within this pattern, however, are disproportionately high mercury concentrations in samples from Ailsa Craig and Grassholm, in the more enclosed waters of the Firth of Clyde and the southern Irish Sea, respectively.

Elsewhere in the Gannet's North Atlantic breeding range, chick feathers were sampled only at three of the Canadian colonies. Mercury concentrations here were within the range of British and Irish figures, but again a geographical pattern was evident. Mercury levels were lowest in feathers from colonies in the enclosed Gulf of Lawrence (Bird Rocks and Bonaventure Island), but markedly higher in feathers from Funk Island, in eastern Newfoundland.

At four British and Irish colonies, both down and juvenile feathers were sampled from chicks (Figure 4.6). In each case, the geographical ranking of median mercury concentrations was the same: Little Skellig > Grassholm > Great Saltee > Bass Rock (rank correlation coefficient = 1.0, $P<0.001$).

For the three (British) regions where both scapulars and pooled mantle-feathers were sampled from adult Gannets, the ranking of median concentrations was the same: Grassholm > Bass Rock > Shetland (Noss or Hermaness) (Figure 4.6). Both

chick feathers and adult mantle feathers were sampled from five colonies but the ranking from high to low median concentrations were not identical (although Grassholm figures were the highest in each case).

Body-feathers of adult and nestling shags sampled from the same three colonies in Scotland and SE Ireland showed the same ranking of median mercury concentrations: Great Saltee > Bass Rock > Foula (Figure 4.7). For Shag and Gannet chicks, comparative data were available for five colonies or regions. In this case, the precise ranking of sample areas was not identical, but median mercury concentrations can be broadly ranked: Kerry > Wexford/St Kilda > Forth/Shetland.

4.4.2 Other studies of geographical variation in mercury levels of North Atlantic seabirds

Mercury concentrations in Gannet eggs have been assessed on a regular basis at several British colonies since 1971 (Newton et al. 1989, 1990); a summary of these results is given in Table 4.8. The Bass Rock and Ailsa Craig colonies have been sampled most frequently, and mercury concentrations were higher in the Ailsa Craig eggs in all nine years when both colonies were sampled. This is consistent with results from the present study, for both eggs and feathers of Gannets. Mercury concentrations were highest in eggs from the Scar Rocks colony, in the north Irish Sea (especially in 1972-73: geometric means 9.6-10.5 ug Hg/g, dry weight). Feather samples were not collected from this colony.

Gannet eggs have also been sampled at one of the Norwegian colonies (Skarvlakken). Mean mercury concentrations in four years 1972-83 ranged ca. 2.2-4.9 ug/g (dry weight) (Fimreite et al. 1982, Barrett et al. 1985), near the mid-range of British results. (Conversion of 'wet-weight' to dry-weight results is based

on the average water content of Gannet eggs [83.7%] noted by Noble & Elliott [1986]).

Eggs of Common Guillemots *Uria aalge* eggs around Britain in the early 1970s showed even more marked regional variations (Parslow & Jefferies 1975). Mercury concentrations were lowest (0.8-1.9 ug/g) at colonies from St Kilda around to NE England (lowest at Foula, Shetland). Mean concentrations were highest (10.1-15.7 ug/g) at four colonies in the northeastern Irish Sea, from Anglesey to the Mull of Galloway (near the Scar Rocks gannetry). Concentrations were lower for the Firth of Clyde (3.6 ug/g at Ailsa Craig) and for colonies south of Anglesey (2.1-5.7 ug/g, highest off south Devon and the Bristol Channel).

The low mercury concentration in Guillemot eggs from St Kilda contrasts with the rather high mercury concentrations of Gannet feathers there. Data for liver and kidney tissue of Puffins *Fratercula arctica* (Osborn 1984) showed a similar pattern to Gannet feather results, however, with concentrations two-three times higher in adults from St Kilda than in birds from the Isle of May (Firth of Forth).

Recent data are available on mercury levels in feathers of a range of adult seabirds at Scottish colonies (St Kilda, Foula and Firth of Forth colonies) (Thompson et al. 1992). Levels were generally lowest in birds from Foula, but no consistent difference between St Kildan and Firth of Forth populations was seen (Kildan concentrations > Forth for Fulmar *Fulmarus glacialis* and Puffin; < Forth for Razorbill *Alca torda*). This study also included colonies in NW Iceland and in NW and NE Norway, and up to five-fold variation of mean concentrations was seen overall for individual species. Where data were available, mercury concentrations were usually highest in Icelandic samples, and lowest in NE Norwegian samples (Thompson et al. 1992).

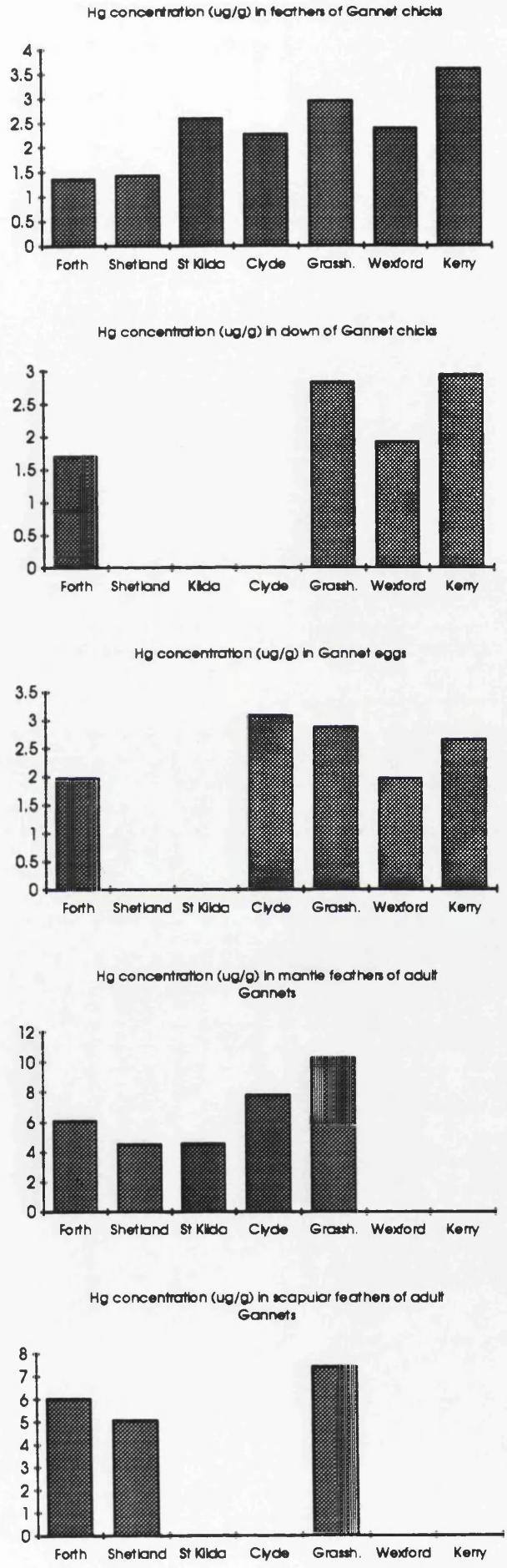


Figure 4.6 Regional variation of median mercury concentrations (ug/g dry weight) in Gannet feathers and eggs from British and Irish colonies.

Earlier work on feathers of auks (Alcidae) had compared mercury levels in feathers of birds from the Baltic Sea, the Kattegat, the Faroe Islands and Greenland (Appelquist et al. 1985). Mercury concentrations were highest in Common Guillemots and Black Guillemots *Cepphus grylle* from the northern Baltic, and markedly lower for Faroese and Greenland auks. Dyck and Kraul (1984) also found mercury levels (in Common Guillemot eggs) lower for Faroese than Baltic samples.

In the western Atlantic, Pearce et al. (1979) summarised mercury levels in seabird eggs sampled from eastern Canadian colonies during 1970-76. Geographical coverage was widest for Double-crested Cormorants *Phalacrocorax auritus* and Puffins. Average mercury concentrations in cormorant egg-contents were mainly in the range 0.21-0.36 ug/g (wet weight) (equivalent to ca. 1.3-2.2 ug/g dry weight: conversion factor from Noble & Elliott 1986). A figure of 0.50 ug/g wet weight was recorded for one colony, but no clear geographical pattern was obvious and colonies in SE Newfoundland were not sampled. For Puffins, mercury concentrations were highest at a colony in SE Newfoundland (0.23-0.26 ug/g wet weight or 0.8-0.9 ug/g dry weight), and lowest at colonies in the Gulf of St Lawrence (0.15-0.16 ug/g/ wet weight). There is thus only limited agreement with the pattern seen for feathers from Gannet chicks, where mercury concentrations were highest for Funk Island in eastern Newfoundland and lowest at two colonies in the Gulf.

Other North Atlantic studies are less relevant to the Gannet data presented here, but include some striking results for colonies on the German North Sea coast. Mercury concentrations in eggs of several seabirds (notably Common Tern *Sterna hirundo*) have shown marked, and consistent, regional variations, with highest concentrations (averages up to 2.2 ug/g wet weight or ca. 9.2 ug/g dry weight for Common Tern) in the Elbe Estuary (Becker 1989).

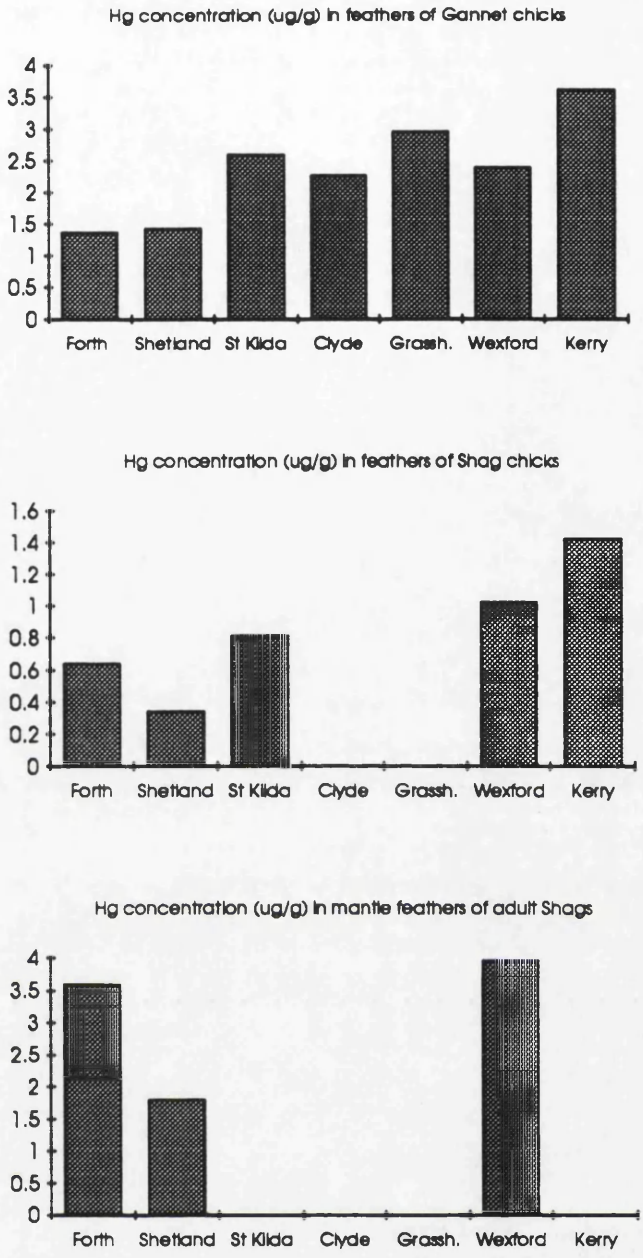


Figure 4.7 Regional variation of median mercury concentrations in feathers of Gannets chicks, and comparison with median mercury concentrations in feathers of Shag chicks and adults, for British and Irish colonies.

Table 4.8 Comparative data on geometric mean mercury concentrations (ug/g dry weight) in Gannet eggs analysed by the Institute of Terrestrial Ecology during 1971-87 (Newton et al. 1989), and 1987 data from this study. All data are for fresh eggs except for Little Skellig 1987 (addled eggs, this study).

Colony	Years sampled	mean mercury concentration (ug/g dry wt) range of means	1987	1987 this study	Net increase or decrease
Bass Rock	11 (1973-1987)	**0.80-2.74**	**2.05**	**1.92**	**Increase
Hermaness	4 (1980-83)	**1.16-2.49**	-	-	***Increase
St Kilda	3 (1979-87)	**0.44-2.31**	**2.09**	-	***Increase
Ailsa Craig	11 (1971-87)	**2.04-5.07**	**2.97**	**3.10**	***Decrease
Scar Rocks	4 (1972-83)	**4.01-10.47**	-	-	***Decrease
Grassholm	2 (1980/1984)	**1.45-2.92**	-	**3.12**	***Increase
Great Saltee	-	-	-	**2.08**	-
Little Skellig	1 (1973)	**3.10**	-	**(2.61)**	-

** or *** = significant (P<0.01 or P<0.001) net increases or decreases in mercury concentrations over the relevant periods, determined by regression of log of individual mercury determinations against time or (where only two years' data available) by t-test (Newton et al. 1989), based on ITE data only.

4.4.3 Factors contributing to geographical patterns seen, and evidence from other monitoring or baseline studies on mercury in North Atlantic waters

The geographical patterns shown by mercury levels in Gannet and Shag tissues should reflect, at least qualitatively or in general terms, dietary uptake of mercury. Laboratory studies on birds have shown that mercury is passed into developing eggs or growing feathers in a dose-dependent fashion (e.g., Heinz 1984, Scott et al. 1975, Scheuhammer 1987). Not all studies have noted a linear increase of feather or egg mercury concentrations with dosage. For example, March et al. (1983) found that mercury input into growing feathers of juvenile domestic fowl *Gallus domesticus* approximately doubled for every three-fold increase in dietary uptake. Further complications might also arise because of variation in type of food, magnitude of mercury dosage, inter-specific differences and other factors. Quantitative estimation of mercury uptake of wild birds, based solely on their tissue concentrations, is thus difficult. Nevertheless, it is clear from experimental work that, for a given species, major differences in mercury uptake over comparable periods should be reflected, at least in gross terms, by feather or egg samples.

Dietary differences between bird species are known to contribute to differences in their tissue mercury levels (see Chapter 2). Known dietary differences between different populations of a given species have also been used to account for differences in mercury levels between different populations of a given species. Newton et al. (1989), for example, noted that mean mercury concentrations in eggs of British Peregrine Falcons *Falco peregrinus* were ca. six times higher for coastal than for inland populations. Tissue concentrations of mercury were lower in prey species available to inland Peregrines, compared to high levels in seabirds and waders (Charadrii) preyed on by coastal Peregrines. Furness et al. (1989) and Newton & Galbraith (1991) noted similar results for Golden Eagles *Aquila*

chrysaetos in Scotland, where much higher mercury levels in feathers and eggs from coastal sites reflected the importance of seabird prey.

An ideal 'indicator' species for marine pollutants would, among other attributes, show little geographical variation in diet. In contrast, Gannets are notably catholic in their diet, and can feed on a wide range of pelagic shoaling fish, ranging in size from young sandeels to large Mackerel (Nelson 1978, Cramp & Simmons 1977, Wanless 1984, Montevecchi & Porter 1980, and section 3.3.5). Additional fish species, including mid-water or bottom-dwelling species, may be consumed as discards from trawlers (Furness et al. 1988). Although data are not always clear-cut, differences in mean mercury concentration are likely to occur between different fish species (e.g. Portmann 1972, Clark & Topping 1989). In particular, small fish such as sandeels will tend to have lower mercury concentrations (a trend also seen generally for size-classes within a given fish species: Thompson 1990).

When the average mercury concentration in Gannet eggs from a Norwegian colony more than halved between 1978 and 1979, Fimreite et al. (1982) suggested that a dietary change was more likely to be responsible than any major, short-term change in environmental levels of mercury. For British Gannets, Newton et al. (1989) similarly suggested that dietary changes could have accounted for some year-to-year changes in organochlorine levels of eggs. At Ailsa Craig, in particular, organochlorine levels (and mercury levels to a lesser extent) in eggs declined markedly in the early 1980s. This appeared to coincide with a change in Gannet diet, from predominantly Mackerel to sandeels (Wanless 1984).

Such dietary influences could also, in theory, account for some geographical variations in seabird mercury levels. Figure 4.8 compares the relative frequency of two major food items, Mackerel and sandeels, in Gannet regurgitations at British and Irish colonies with data on mercury concentrations in Gannet chicks. There is

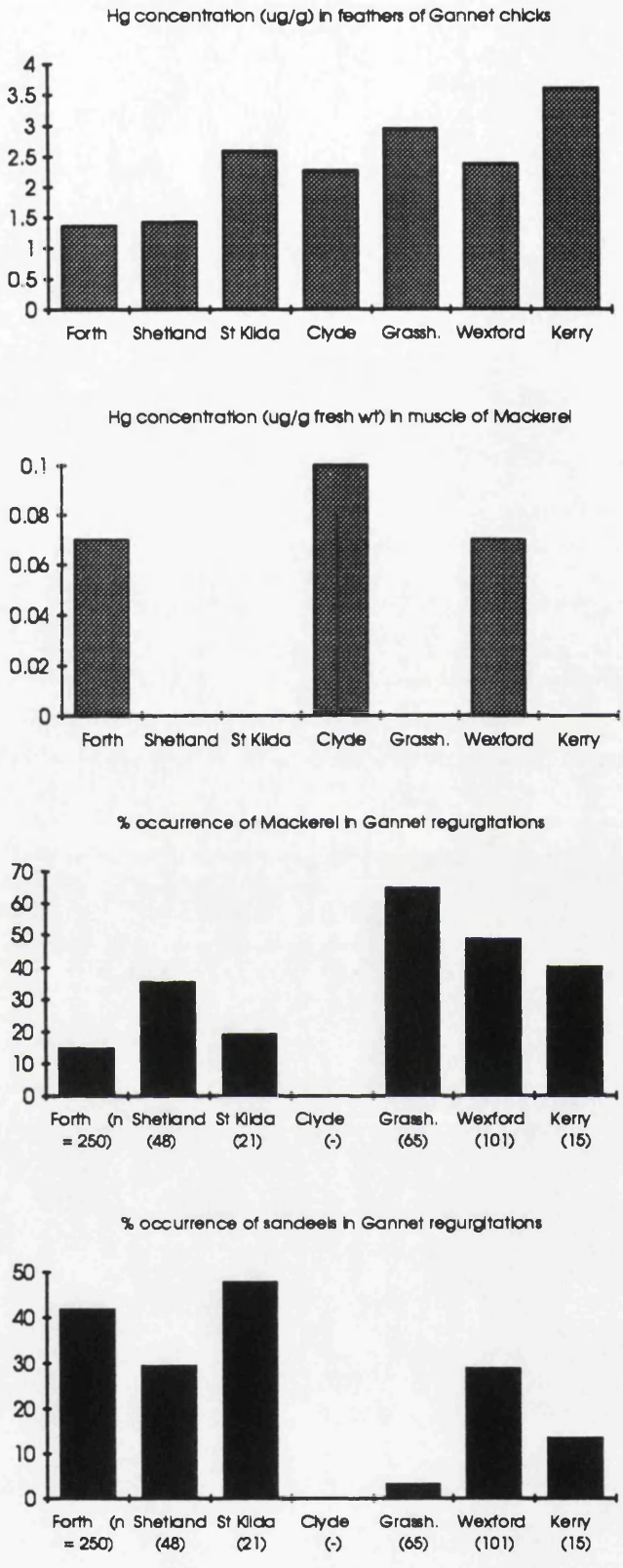


Figure 4.8 Regional variation of median mercury concentrations in feathers of Gannet chicks from British and Irish colonies, and comparison with mercury concentrations in Mackerel muscle and the percentage occurrence of Mackerel and sandeels in Gannet regurgitations.

a broad correlation, in that mercury levels in chick feathers were lowest for colonies where many sandeels, or relatively few Mackerel, were regurgitated. Without more comprehensive data, however, this relationship can only be speculative. Other food items, including trawler discards, showed no obvious geographical patterns, or relationships to mercury data.

The limited data from this study on mercury concentrations in Mackerel tissue are also plotted in Figure 4.8, but show no clear pattern. In each of the samples, mercury concentrations broadly increased with size of fish, and differences in size-composition may have clouded any regional differences. Given the low concentrations, however, and the variability of the mercury levels for fish of a given size, it was unlikely that any real pattern could be demonstrated.

In contrast to Gannets, Shags feed mainly on relatively few species of small fish. Sandeels predominate in virtually all studies during the breeding and, in at least one study, pre-breeding periods (Cramp & Simmons 1977, Harris & Wanless 1991). Despite this, Shag colonies sampled during the present study showed similar geographical patterns of mercury concentrations in feathers. This tends to support the idea that mercury levels seen in Gannet feathers were reflecting real geographical variation in bioavailability of mercury.

The geographical variation seen for Gannet mercury levels around Britain and Ireland also has some parallels in data for mercury in other biota, and in data for mercury concentrations in seawater.

Mercury concentrations in surface waters around the U.K. are, on the whole, highest in the Irish Sea (Gardner & Riley 1973, Baker 1977). Concentrations there are particularly high in the north-eastern section, in a broad strip northwards from the coast between Anglesey and the Mersey Estuary. This corresponds to known

high inputs of mercury from industrial and other sources around Liverpool Bay. In the early 1970s, mercury concentrations in commercially-caught species of fish in these waters averaged ca. 0.5 ug/g (wet weight) (Portmann 1972, Gardner 1978). This was more than twice as high as the average for other parts of the Irish Sea, and marginally exceeded statutory safety limits for human consumption (Gardner & Riley 1973, Anon 1990). Efforts have since been made to reduce mercury inputs here. Apparently associated with this, mercury concentrations have since fallen to <0.3 ug/g in fish there (Dickson 1987), and concentrations of mercury in Mersey Estuary sediments halved during 1974-83 (Anon 1990). Nevertheless, Law et al. (1991), in a review of metals in marine mammals during 1988-89, recorded particularly high levels of mercury in three Grey Seals *Halichoerus grypus* in this region.

Baker's (1977) results for total mercury and 'reactive' mercury in UK waters suggested that concentrations were lowest around Shetland and northern Scotland, and higher further south in the North Sea and off west and southwest coasts of Britain. Particularly high concentrations were recorded in the English Channel, the Bristol Channel and, especially, Liverpool Bay. High sediment loads caused by severe weather may have contributed, however (Baker 1977), so it is not clear whether a real SW-NE trend existed.

The above studies, and work on mercury levels in mussels *Mytilus edulis* (Dickson & Boelens 1988), do indicate, however, that the western channel of the Irish Sea is the least contaminated part. High mercury concentrations in feathers and eggs of Gannets from Grassholm would appear most likely to reflect the influence of Bristol Channel waters.

In the North Sea, mercury concentrations are higher in coastal waters than further offshore, and tissue levels of mercury and fish tend to be highest in the southern /

south-eastern North Sea (Anon 1987, Lee 1978). High levels of mercury have been recorded in mussels from the Firth of Forth (Alzieu 1987). However, evidence from fish, mussels, sediments and water indicates that the Firth of Forth is far less contaminated with mercury and other metals than the inner Forth estuary (Essink 1985, Balls & Topping 1987).

Levels of mercury in fish recorded by Lee (1978) were particularly low around Shetland. Portmann (1972) noted that mercury concentrations in fish >25 miles offshore in the North Sea were half those recorded in similar samples from the Irish Sea; figures for fish in coastal waters were similar to those for the Irish Sea margins, and included particularly high concentrations in the Humber / Wash / Thames region.

The relative contributions of 'natural' mercury inputs, localised anthropogenic inputs and inputs from wider global contamination, even to relatively enclosed waters such as the North Sea, Irish Sea or Firth of Clyde, with heavily industrialised and urbanised margins, is not always clear. For the Irish Sea, the most important sources of mercury input have been ranked as: 1. rivers (including natural plus waste-discharges), 2. dumping activity, 3. Atlantic inflow, 4. industrial discharges, 5. atmospheric and 6. sewage discharge (Anon 1990). Industrial discharge and dumping of dredge spoil were the main inputs to Liverpool Bay. For the North Sea, Barbour (1987) estimated annual input of mercury as 75 tonnes, of which 70 came from land and 5 from the atmosphere. Globally, however, man's input of mercury to the environment was estimated as <1% of total inputs (e.g., through natural weathering, and from hydrothermal vents on ocean floors).

Different views have also been expressed regarding global anthropogenic inputs of mercury. Gardner (1975) found higher mercury concentrations in seawater of the

northern hemisphere than of the southern hemisphere. She proposed that this reflected pluvial deposition from the northern jet streams carrying atmospheric industrial inputs from North America, Europe and Japan. If this effect occurs, it would, for example, contribute to mercury input in the Atlantic inflow, either in currents or by direct pluvial deposition, to the Irish Sea and British southwest approaches. Elevated mercury levels in seawater would also tend to reach other western waters of Britain and Ireland. Currents west of Scotland (Lee & Ramster 1979) flow through the Pentland Firth, around northern Scotland, before turning down into the North Sea. Gradual loss of mercury from the water column, through sequestration by plankton or loss to suspended particular matter (cf. Chester et al. 1973, Gardner 1975), could thus contribute to a SW-NE 'cline' of mercury concentrations (if, indeed, one exists).

4.4.4 Influences of at-sea distribution, seasonal movements and foraging movements of Gannets and Shags

The precise sea areas being 'sampled' when mercury levels in seabird tissues are assessed are not always clear, because of such factors as seasonal movements (migratory or dispersive) or uncertainty about foraging ranges from a colony. Some tissues (and age-classes) of seabirds may be more likely to reflect mercury uptake from a given sea area. Thus, we might expect that some soft tissues of a species spending a substantial part of the year away from its breeding grounds might not necessarily reflect mercury levels in the vicinity of the colony in a straightforward way. Likewise, since mercury concentrations in feathers appear to reflect mobilisation of soft-tissue mercury during moult, feathers of adult birds might provide just as imprecise a measure of relative 'contamination.'

Overlap between seabirds from different breeding areas occurs particularly outwith the breeding season, and it may not be easy to quantify the degree of overlap based on available ring recoveries. When species are 'tied' to colonies (March/April to September/October for Gannets), there will tend to be less overlap between different populations.

Available evidence for laboratory and field studies indicates that mercury levels in eggs reflect dietary uptake from a relatively limited period before egg-laying. For example, Becker (1989) noted that 'microgeographic' patterns of mercury levels in eggs of several species at a range of colonies on the German North Sea coast were consistent from year to year. This was despite differences in feeding ecology and migratory habits between species. Parslow & Jefferies (1975) also noted that, for many seabirds, pollutant residues in eggs tended to fall within relatively narrow limits for one locality [at least within a given year], particularly for more sedentary species having a narrow food range.

Growing feathers of seabird chicks will incorporate mercury derived from a relatively restricted radius of a colony, over a limited period of time, so can be predicted to provide less variable results than adults. This proved to be the case for Gannets and Shags (Chapter 3).

Gannet foraging ranges have been estimated for various colonies. Tasker et al. (1985) found that most foraging around Noss (Shetland) was within 120 km of the colony (and the bulk with a 40-km radius), with a maximum of ca. 150 km. Webb et al. (1990) noted that St Kilda some adults were apparently foraging 100 km or more from the colony. For the Bass Rock, Nelson (1966) suggested a normal feeding radius of up to 160 km during the breeding season. In the western Atlantic, Newfoundland Gannets as a whole were suggested to forage usually

within 120 km of colonies, while Kirkham et al. (1985) noted that Gannet densities around Funk Island were particularly high within a 60 km radius.

At both west-coast and North Sea gannetries around Britain, at-sea densities were particularly high around colonies early in the breeding season (Tasker et al. 1985, Webb et al. 1990). Later in the season, from July onwards, adult Gannets were somewhat more widely dispersed, perhaps in response to seasonal changes in food availability (Tasker et al. 1985). Perhaps a similar situation occurs off Funk Island (Newfoundland), where few birds occur as far north as the Labrador coast before July (Kirkham et al. 1985).

In the pre-breeding period, the bulk of the adult population arrives at the colony in March or April, with some birds as early as January or February, depending on the colony. For example, the main return was in mid March at the Bass Rock, but about a month later at Noss (Tasker et al. 1985). Distribution of Gannets at seas during this period was also markedly concentrated around colonies during surveys off the west and east coasts of Britain by Tasker et al. (1985) and Webb et al. (1990).

In winter, some adult Gannets remain in northern waters, but the majority of British birds move further south, mainly to the west coast of France and the Iberian peninsula (Thomson 1974). (Southward movements are more marked for younger Gannets, however.) At this season, the main concentrations of Gannets in the North Sea appear to be in the south (Tasker et al. 1986.) Norwegian Gannets appear to show the same dispersal pattern as British birds, with very few ever seen at breeding latitudes before February (Barrett 1988). Overlap of wintering ranges may account in part for the general lack of clear differences in plumage mercury concentrations for Gannets at different colonies. For example, the high levels recorded for some Bass Rock birds might reflect wintering, e.g., in the southern

North Sea (where mercury levels in fish are higher than further north: section 4.4.3).

Most Canadian Gannets winter from New England south to the Gulf of Mexico (Nelson 1978, Kirkham et al. 1985).

Compared to Gannets, foraging ranges of Shags are probably generally small. Around the Isle of May (off SE Scotland), Wanless et al. (1991) radio-tracked adults, and found that average foraging distance was 7.0 km from the island. Most birds, in fact, foraged either within 2 km of the island, or in two discrete areas 5-13 km away.

Galbraith et al. (1986) assessed regional variations in the dispersal patterns of Shags in northern Europe. For most populations, median winter movements shown by ring-recoveries were <100 km, and wintering ranges of different breeding populations were relatively discrete. Winter ranges were plotted for each population, based on November-March recoveries, excluding the most distant 10%. For birds breeding in the Firth of Forth and off NE England, the winter range stretched from Orkney south to the English Channel, while Shetland birds mainly occupied waters from Shetland south to Lewis (Western Isles) and NE Scotland (Galbraith et al. 1986). Birds from the southern Irish Sea winter as far south as NW France and the western English Channel, as well as around SW Ireland and the SW Approaches. Mercury levels in feathers of adult Shags from these populations could thus be influenced by dietary uptake from quite substantial inshore areas (though to a lesser extent than adult Gannets).

Chapter 5

EVIDENCE FROM BIOLOGICAL SAMPLES FOR HISTORICAL CHANGES IN GLOBAL METAL POLLUTION

TABLE OF CONTENTS

This chapter has been published in Furness, R.W. & Rainbow, P.S. (1990). Heavy Metals in the Marine Environment, CRC Press, Boca Raton, Florida, pages 219-225. I am a co-author of this chapter together with R.W. Furness and D.R. Thompson. The chapter is presented in the format required by the publisher.

I. INTRODUCTION

It has been argued that global, as distinct from local, pollution by heavy metals is most likely to arise with lead, mercury, and cadmium.[1-4] Relative to natural emissions, anthropogenic inputs of these elements are high[3,4] and atmospheric transport may be followed by deposition far from industrial sites. Attempts to quantify changes in heavy metal contamination of remote areas using ice cores or snow samples have been hindered by the extremely low concentrations of these elements in ice and snow and consequent technical difficulties of avoiding contamination of samples and of measuring such low levels with accuracy (as discussed by Wolff in this volume).

Since plants and animals may accumulate these metals to concentrations five to nine orders of magnitude higher than found in snow and ice samples, it is easy to measure levels in biological samples with little likelihood of sample contamination due to handling by the analyst. Where samples of biological material collected at dated times in the past can be obtained, it should be possible to compare metal levels in contemporary samples with levels found in the past. Such studies should not be considered as an alternative to analysis of ice cores, but as a valuable source of independent evidence to support data from ice and snow samples. However, a number of difficulties arise.[5] In particular, there are rather few suitable biological samples available for analysis. It is essential that samples should not have been treated (for example by use of preservatives) in such a way that metal levels will have been altered. The collection site for samples must be known so that geographical variations may be taken into account if these are pronounced, and the date of collection must not be open to doubt. The tissues must be suitable for the measurement of the metal of interest; for example, bird feathers or mammalian hair may provide excellent data on mercury levels, but cadmium is generally not transferred into feathers and hair in detectable amounts[6-8] and in studies where levels of cadmium and lead in feathers or hair have been measured, the levels found show little or no correlation with cadmium and lead levels in other organs.[8-10]

Since the nonessential heavy metals cadmium, lead, and mercury usually show high coefficients of variation in populations,[10-12] any attempt to compare population levels between past and present day samples requires a large sample size of both contemporary and historical tissues before any statistically meaningful comparison may be made. Where metal levels may vary, perhaps by an order of magnitude between individuals within any one population, attempts to test whether or not metal levels have changed as a result of human activities should take particular care not to draw conclusions from small numbers of tissue samples. Other factors, such as changes in the dietary composition or migration patterns of animals may also alter their metal levels and so care must be taken to avoid erroneous conclusions due to a lack of consideration of such possible alternative explanations.

In this chapter, we firstly review papers that have used historical biological material to assess time trends in metal levels in the environment, either to detect instances of local pollution or to seek evidence for global trends, and then we assess the likely value of this approach in current and future studies.

II. REVIEW OF PUBLISHED WORK

Rather few studies have been made in which metal levels in historical biological samples have been analyzed, and all have considered only mercury, since the tissue distribution of cadmium and lead makes those elements more difficult to measure in historical samples.

Three papers[13-15] describe mercury levels in museum specimens of fish collected 60 to 100 years ago compared with levels in present-day specimens. Miller et al.[13] analyzed total mercury levels in seven tuna (five skipjack *Katsuwonus pelamis*, one albacore *Thunnus alalunga*, and one bluefin *Kishinoella tonggol*) collected between 1878 and 1909 from a

wide range of Atlantic and Pacific sites, two tuna collected in 1970 to 1971 from the Pacific, three samples of tinned albacore tuna, a (physically deformed) swordfish *Xiphias gladius* head preserved in isopropanol since 1946 and six fresh swordfish from California. They found an order of magnitude difference in mercury levels between the six 1970 to 1971 swordfish, with the 1946 sample falling within this range. Similarly, the museum and contemporary tuna showed variations between 0.13 and 0.64 μg/g wet weight, with no significant difference between groups. Barber et al.[14] measured total mercury levels in recent and 90-year-old benthopelagic fish. The museum samples consisted of one *Antimora rostrata* collected in 1883 and one *Aldrovandia macrochir* collected in 1886. Both fish had mercury levels similar to those of nine and five contemporary individuals of those species, and these authors also concluded that the high levels in these fish species "are natural and may not be related to anthropogenic inputs of mercury to the oceans." Based on such meager sample sizes such a conclusion appears rash. Even if a difference did exist, given the high degree of individual variation in metal levels it would be virtually impossible to demonstrate a statistically significant difference between museum and contemporary fish with such a small sample size. Indeed, Rowland[15] cautioned that the results did not show the absence of oceanic mercury pollution but suggested that any anthropogenic increase in this deep ocean habitat was unlikely to be of a factor of as much as five. Nevertheless, these papers seem to have been taken by the scientific community to indicate that no global increase in mercury levels in marine ecosystems has occurred, despite the fact that the small sample sizes analyzed would prevent an increase from being readily detectable.

Analysis of polar bear *Ursus maritimus* hair for mercury has shown that mean levels within populations vary from 2.5 μg/g for 41 animals on the southern shore of Hudson's Bay to 18.5 μg/g for 5 animals at Amundsen Gulf[16] with no overlap in individual values sampled at these 2 localities. Intermediate values were obtained at other sampling sites in between and further northeast. The authors presented data for mercury levels in hair of 18 animals collected 1910 to 1927 but of these, 8 were not adults. Since young animals have lower mercury levels than adults,[10] these 8 cannot be considered further. Eight of the adults were collected from the vicinity of Amundsen Gulf and so were comparable with the contemporary sample of five from that area. However, separate measurements of total organic and inorganic mercury levels in the hair of the historical samples showed clear evidence of contamination during preservation or storage with inorganic mercury, such that one young bear had hair levels of 53.3 μg/g of inorganic mercury but only 0.3 μg/g of organic mercury. Although the authors washed the museum hair samples thoroughly, in our experience, if stored samples become at all damp, much inorganic mercury contamination will become bound to hair too strongly to be removed by any washing procedure. Normally polar bear hair contains very little inorganic mercury, the organic fraction representing over 80% of the total[16] as found in human hair. Unfortunately, Eaton and Farant did not analyze inorganic and organic mercury levels separately in the contemporary samples and so a direct comparison between organic mercury levels in the museum and recent samples is not possible. If we assume that 85% of the total mercury in the contemporary samples was organic as the authors suggest is normal, then the comparison (Table 1) indicates a statistically significant difference, with the modern samples having four times as much mercury as the museum samples, and yet Eaton and Farant concluded "observed high mercury levels in the Canadian Arctic reflect geological sources and are not connected with anthropogenic release." Geographical variation in mercury levels in polar bears around the Canadian Arctic seems to be at least as great as this historical increase, and may be attributed to differences in diets since different seal species have quite different mercury levels.[10,16,17]

Hansen, (see Chapter 13) describes a study of levels of metals in hair samples from 15th century mummified Eskimos compared with today's Eskimos. That study shows evidence of increases in mercury and lead levels that were attributed to increased global

TABLE 1
Mercury Levels (μg/g) in Hair Samples from Adult Polar Bears Collected in the Amundsen Gulf Area of the Canadian Arctic

Period	Sample size	Mean	S.D.	Range
1910—27	8	4.6	1.8	1.8—7.5
1977—80	5	18.5	14.5	9.3—44.3

Note: It is assumed that organic mercury levels represented 85% of total mercury for museum samples.

pollution, while some changes in selenium, iron, and bromine levels appeared to be related to a reduction in the relative importance of marine foods in the diet, a trend which would also have tended to reduce, rather than increase, mercury and lead exposure.[18]

Mercury and other heavy metals are taken up by bog plants to a particularly high degree and are strongly retained by the humic substances of peat and so peat cores may be used to examine historical changes in metal fluxes.[19] Madsen showed that the deposition rate of mercury on two peatbogs in Denmark increased in parallel with the known increase in consumption of mercury in European Community countries between 1740 and 1980, and suggested that some brief periodically elevated deposition rates could be attributed to major volcanic events occurring in Iceland.

Several papers have considered the extent to which changes in mercury levels in the environment may be assessed by analysis of time series of feather samples from bird populations.[20-25] Use of alkyl-mercury seed dressings in Scandinavia from the 1940s to the 1960s led to clearly elevated mercury levels in feathers of terrestrial and freshwater birds compared to levels in pre-1940 museum specimens.[20] This study did not distinguish between organic and inorganic mercury levels in feathers and so encountered problems due to contamination of some museum specimens with inorganic mercury. The authors arbitrarily discounted anomalously high values of total mercury from museum samples as due to contamination, though this procedure requires a detailed knowledge of the results to be expected in order that anomalous data can be omitted.

Study of total mercury levels in feathers of black guillemots *Cepphus grylle*, common murres *Uria aalge*, and thick-billed murres *U. lomvia* from the Baltic Sea, Kattegat, and Atlantic[21] showed that mercury levels had increased in the species in the Baltic Sea, from levels of around 1 to 2 μg/g in common guillemot primaries from before 1900 to 2 to 8 μg/g around 1960 to 1974 and from 2 to 5 μg/g in black guillemot primaries before 1900 to 9 to 25 μg/g around 1960 to 1974.[21] This work used neutron activation analysis to measure total mercury levels, and yet the authors made no mention of problems with post-mortem contamination during museum preparation or storage, which we have found to be a problem in some historical collections (see below). Elevated mercury levels in Baltic seabirds can be attributed to use of mercury seed dressings in Scandinavia and other local sources of pollution, and there is some evidence to suggest that levels decreased slightly after mercury seed dressing became less common.[21] Examination of changes in mercury levels in the birds around the Faeroes and Greenland suggest that an increase in mercury levels may have occurred in those areas too, but to a much smaller extent than in the Baltic; from around 1 μg/g in black guillemots from Greenland before 1900 to around 2 μ/g in 1973, and from 0 to 1 μg/g in primaries of common murres from the Faeroes before 1920 to 0.5 to 1.5 μg/g in 1973.[21]

After the outbreak of Minamata disease in the 1960s in Japan, caused by mercury discharge into the Shiranai Sea at Minamata Bay,[22] an analysis was made of mercury levels

in feathers of around 100 birds from this area collected between 1955 and 1980 and these values were compared with analyses of feathers from similar birds from *uncontaminated* sites in China and Korea. The authors concluded that feathers provided a reliable measure of geographical differences in mercury levels in the ecosystem, and that analysis of time series from museum collections should provide evidence for trends in mercury levels over periods of decades.[22]

III. FUTURE PROSPECTS AND PROBLEMS

A. CADMIUM

Since cadmium tends to accumulate to high concentrations in kidney tissue and is generally not transferred to the more stable tissues,[9] such as feathers and hair, the usefulness of preserved specimens in determining any historical changes in cadmium levels is likely to be limited.

B. MERCURY

As a result of being both chemically and physically stable, together with their relative abundance in preserved collections, feathers and fur should make suitable study tissues for the assessment of historical changes in mercury levels on a global scale. By contrast, soft, internal organs are prone to physical decay and chemical change and as such are likely to be less reliable for this purpose.

The elucidation of historical trends in mercury levels in the environment by use of time series of feather samples from appropriate bird populations[20-25] has been undertaken largely without consideration of the form of mercury incorporated into the feather keratin molecule, or of the consequences of mercury contamination via preservation processes. Additional, inorganic mercury used in the treatment of preserved specimens will result in erroneous total mercury values.

An initial investigation into the relationship between organic and inorganic feather mercury[26] has revealed that organic mercury represents about 100% of the total mercury level. This result is based on a comparison of total and organic feather mercury levels in a sample of 95 seabirds of various species. The variation around this figure is relatively large due to the destructive nature of the analytical technique, making direct comparisons within a given sample difficult. However, by performing an initial fractionation of the feather mercury to remove the intrinsic organic mercury only, a more meaningful assessment of the historical mercury concentration can be made. This technique should effectively overcome any contamination problems associated with study skin preservation processes, and the subjective discarding of high mercury values. Studies of the effects of storage, washing and exposure of hair samples or feathers to UV-B and other factors indicate that mercury is firmly bound in feathers and levels do not change with a variety of harsh treatments.[27,28]

Recent work on the dynamics of mercury in birds and of the relationships between molt patterns and mercury levels in feathers[29-32] gives a good basis for studies of time series of mercury levels in seabird feathers, and progress in this area can be expected.

C. LEAD

Lead has been shown to accumulate to relatively high concentrations in bone and teeth of marine vertebrates.[34-36] Other similarly hard and relatively stable tissues, such as horns and antlers, are likely to show a similar pattern. By taking into account both the possibility of lead contamination subsequent to sample collection, and the distribution of lead between these and other internal tissues, historical changes in global lead levels in a range of appropriate biological samples could be investigated. Although lead levels in hair have been shown to be quantifiable,[37] the relationships between lead concentrations in hair (and feathers)

and internal tissues are poorly documented.[9] Without a detailed knowledge of such distribution patterns and relationships, a meaningful interpretation of any hair/feather lead data will be difficult. Given the high industrial usage of lead in recent decades, it would be valuable to develop the possibility of using selected hard tissues of preserved biological samples to determine changes in lead concentrations over time.

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Chapter 6

HISTORICAL CHANGES IN MERCURY CONCENTRATIONS IN THE MARINE ECOSYSTEM OF THE NORTH AND NORTH-EAST ATLANTIC OCEAN AS INDICATED BY SEABIRD FEATHERS

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Historical changes in mercury concentrations in the marine ecosystem of the north and north-east Atlantic ocean as indicated by seabird feathers

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Summary

1. Body feather samples from preserved study skins held in museum collections and from contemporary specimens of a range of species of adult seabirds from the north and north-east Atlantic were analysed for mercury in order to assess historical trends in contamination of the marine environment with this toxic heavy metal.
2. Problems with the contamination of samples through the application of inorganic mercury to many study skins as a preservative were overcome by methyl mercury extraction (Thompson & Furness 1989a,b).
3. Fulmars *Fulmarus glacialis* (L.) from both St. Kilda and Shetland/Orkney exhibited significant declines in mercury concentrations associated with pronounced dietary change over the study period.
4. Other species analysed showed a general increase in mercury burdens, particularly towards the south and west of Britain.
5. Increased pluvial deposition of atmospheric mercury over specific oceanic areas, associated with jet-streams and the pollution of the northern hemisphere by mercury, is suggested as being a likely cause of the observed trends.
6. The use of time-series of feather samples, coupled with the extraction method for methyl mercury used in this study, offer great potential for elucidating changes in the mercury contamination of other environments.

Key-words: historical changes, mercury, seabird feathers.

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Introduction

Examples of local, relatively well-defined pollution due to anthropogenic emissions of mercury to the environment have been well documented. Mercury, of both industrial and agricultural origin, has been shown to be the cause of major toxicological problems, and in some cases human fatalities, in Japan (Kurland, Faro & Seidler 1960), Iraq (Bakir *et al.* 1973) and Sweden (Borg *et al.* 1969; Johnels & Westermark 1969). Despite the fact that anthropogenic mercury has been a major constituent of the overall total mercury released to the environment (Lantzy & Mackenzie 1979; Nriagu 1989), demonstration of a general and widespread increase in mercury concentrations in the ecosphere over the past 150 years has proven to be difficult.

Attempts have been made to measure mercury concentrations in polar ice-core samples (Appelquist *et al.* 1978). Such investigations have been hampered by the analytical difficulties associated with the measurement of the extremely low mercury concentrations (*c.* $1-10\,pg\,g^{-1}$) in ice samples. Furthermore, the low mercury concentrations encountered have led to problems with sample contamination, and reliable mercury data have been difficult to obtain (Wolff 1990).

Complementary to the use of ice-core samples have been the various studies incorporating mercury determinations in historical and contemporary biological samples. Biota accumulate mercury to concentrations five to nine orders of magnitude higher than those found in ice/snow samples, so analytical difficulties in measuring the mercury concentrations are much reduced.

A wide range of biological indicator species has been used, ranging from peat core samples (Madsen 1981), pelagic marine fish (Barber, Vijayakumar & Cross 1972; Miller *et al.* 1972), hair samples from polar bears *Ursus maritimus* Phipps or from humans (Eaton & Farant 1982; Hansen 1990), skin samples from harbour porpoises *Phocoena phocoena* (L.)

(Andersen & Rebsdorff 1976), to time-series of feather samples from numerous bird species (Berg *et al.* 1966; Doi, Ohno & Harada 1984; Appelquist, Drabaek & Asbirk 1985).

However, the findings of several of the above studies were based on the analysis of only a few individuals (Barber *et al.* 1972; Miller *et al.* 1972; Andersen & Rebsdorff 1976), whilst others encountered problems due to sample contamination by inorganic mercury (Berg *et al.* 1966; Eaton & Farant 1982). Such contamination has led to the subjective discarding of elevated results (Berg *et al.* 1966), an approach prone to error as it is difficult, if not impossible, to distinguish between high results due to contamination and those which truly reflect naturally high mercury concentrations.

Although the contamination of samples prepared for museums has been problematical, time-series of feather samples offer several advantages as a means to elucidate historical trends in mercury burdens of environments. Preserved collections of study skins are relatively large and well-documented, allowing accurately dated feather samples to be obtained.

Recent work by Thompson & Furness (1989a) indicated that the mercury present in seabird feathers is predominantly organic (methyl) mercury. It was concluded that by employing the extraction technique used in that study, problems of inorganic mercury contamination were overcome and reliable and meaningful results could be obtained (Thompson & Furness 1989a). Hence, feather samples from a range of British seabirds captured prior to 1930 at documented breeding sites have been obtained from collections of museum study skins and their methyl mercury concentrations determined. Comparisons were made with mercury concentrations in feather samples from contemporary specimens from the same geographical locations, in order to assess any change in mercury burdens of seabirds from the north and north-east Atlantic over a period of increased anthropogenic mercury production.

Methods

SAMPLE COLLECTION AND PREPARATION

Contemporary (post-1980) feather samples were obtained from apparently healthy adult birds during the breeding season. In most cases, four to ten body feathers were taken (Furness, Muirhead & Woodburn 1986) and placed in mercury-free polythene bags prior to analysis. Species and sites sampled were as follows: northern fulmar *Fulmarus glacialis* (L.) from St. Kilda and Foula, Shetland; Manx shearwater *Puffinus puffinus* (Brünnich) from Skomer, south-west Wales (body feathers or small wing coverts from freshly dead adults) and Rhum, Inner Hebrides; north Atlantic gannet *Sula bassana* (L.) from the Bass Rock; great skua *Catharacta skua* Brünnich from Foula and southern Iceland; and Atlantic puffin *Fratercula arctica* (L.) from St. Kilda, Foula and Great Saltee, southern Ireland.

Historical feather samples from the same species were obtained from preserved study skins held in British museum collections. In all cases, body feathers were taken and placed in mercury-free polythene bags prior to washing and analysis.

Although the organic mercury extraction technique discriminates against inorganic mercury (Thompson & Furness 1989a), contamination will alter the weight of the feathers; thus all museum feather samples were subjected to a chloroform/acetone washing regime (Muirhead 1986) in order to remove any gross surface contamination.

ANALYSIS OF MERCURY CONCENTRATIONS

Historical feather samples were subjected to an initial fractionation in order to extract only the organic mercury present. The method used is based on that of Uthe, Solomon & Grift (1972) and is outlined more fully in Thompson & Furness (1989a,b), where it was demonstrated that the analysis of organic mercury and total mercury in uncontaminated seabird feathers gave identical results. Mercury concentrations were determined using a cold vapour technique, incorporating a Data Acquisition Ltd. DA 1500-DP6 mercury vapour detector, preceded by standard acid digestion (Furness *et al.* 1986). The methyl mercury extraction technique was calibrated using standards of methyl mercuric chloride, matrix effects being negligible (Thompson & Furness 1989a). The accuracy of mercury determinations was assessed using International Atomic Energy Agency horse kidney Reference Material H-8 (Thompson & Furness 1989a). All mercury concentrations are expressed on a fresh weight basis; although washed feathers were dried in an oven at 50 °C for 24 h, they were then allowed to equilibrate to ambient laboratory temperature (*c.* 22 °C) for several hours before analysis as accurate and consistent 'dry' weights were difficult to obtain due to rapid reabsorption of water (Thompson 1989). Statistical comparisons between mercury concentrations in pre-1930 and post-1980 groups were made using Mann-Whitney *U*-tests.

Results

There was no significant difference between median mercury concentrations of historical Icelandic and historical northern isle feather samples of great skuas (Mann-Whitney *U*-test, $P = 0{\cdot}78$), nor between median mercury concentrations in contemporary feather samples of great skuas from these two areas (Mann-Whitney *U*-test, $P = 0{\cdot}84$). In the light of these results and in view of the small number of preserved historical specimens of this species, all

Table 1. Changes in mercury concentrations ($\mu g\ g^{-1}$ fresh weight) of body feather samples obtained from pre-1930 museum skins and post-1980 north and north-east Atlantic seabirds

Species	Location	Pre-1930	Post-1980	Change ($\mu g\ g^{-1}$)	Change (%)	Mann-Whitney *U*-test
Northern fulmar	Shetland/ Orkney	4·00 1·27–10·74 (25)	1·42 0·85– 3·90 (32)	−2·58	−65	$P < 0.0001$
Northern fulmar	St. Kilda	4·45 1·85– 6·83 (11)	2·90 1·17–11·53 (85)	−1·55	−35	$P < 0.001$
Manx shearwater	North-west Britain/ Ireland	1·61 0·24– 3·16 (14)	4·42 1·40–10·48 (78)	+2·81	+175	$P < 0.0001$
Manx shearwater	South-west Britain/ Ireland	1·32 0·08– 3·22 (17)	3·33 1·02– 5·05 (22)	+2·01	+152	$P < 0.001$
Atlantic puffin	South-west Britain/ Ireland	1·83 0·18– 3·68 (49)	4·03 1·92–11·40 (61)	+2·20	+120	$P < 0.0001$
North Atlantic gannet	Bass Rock	6·03 1·77–10·62 (19)	7·21 2·55–17·98 (38)	+1·18	+20	$P < 0.05$
Great skua	Iceland/ Shetland/ Orkney	3·67 2·41–10·62 (13)*	5·62 1·15–32·42 (212)†	+1·95	+53	$P < 0.05$

Values are medians; ranges are also shown and sample sizes are provided in parentheses.
* 7 birds from Shetland/Orkney plus 6 birds from Iceland.
† 197 birds from Shetland/Orkney plus 15 birds from Iceland.

data have been combined to compare historical and contemporary mercury concentrations.

Mercury concentrations in birds sampled pre-1930 and post-1980, together with the change and percentage change in mercury concentrations between these two periods, are presented in Table 1. In all species except the fulmar, an increase in mercury concentration was noted, this increase being most pronounced in Atlantic puffins (Table 1; Fig. 1) and Manx shearwaters (Table 1; Fig. 2). Fulmars from both St. Kilda and Shetland/Orkney exhibited significant decreases in mercury concentrations between the two periods (Table 1).

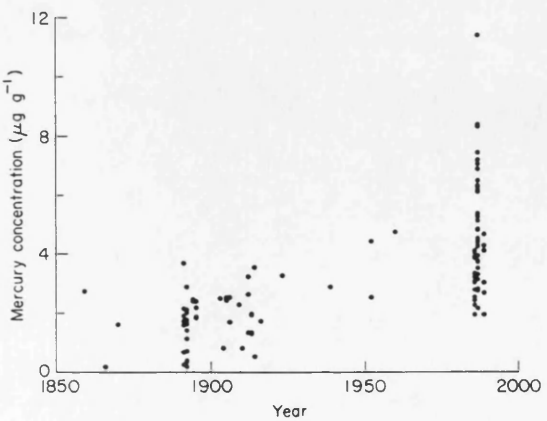

Fig. 1. Temporal variation in mercury concentrations of body feathers of Atlantic puffins sampled from the south-west and west of Britain and Ireland.

Discussion

Several assumptions must be met if the mercury concentrations of feathers are to reflect changes in environmental contamination with mercury.

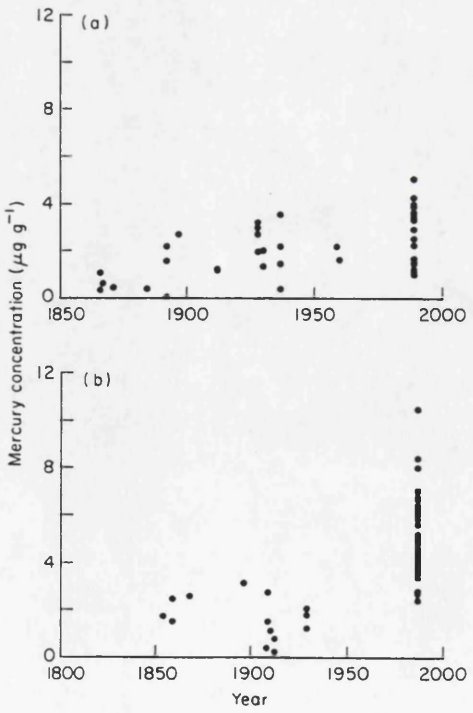

Fig. 2. Temporal variation in mercury concentrations of body feathers or small wing covert feathers of Manx shearwaters sampled from (a) the south-west of Britain and Ireland, and (b) the north-west of Britain and Ireland.

1. Mercury incorporated into the feather during feather growth must be relatively stable. Crewther *et al.* (1965) noted that mercury is bound strongly to disulphide linkages in keratin, whilst Appelquist, Asbirk & Drabaek (1984) found that mercury concentrations of feathers were unaffected by various rigorous treatments. Preserved soft internal tissues have been used to investigate temporal changes in mercury concentrations (e.g. Barber *et al.* 1972; Miller *et al.* 1972), although the fact that such tissues are prone to wastage and decay prior to preservation and could be altered chemically by the preservative used, renders these tissues less reliable (Furness, Thompson & Walsh 1990). Contamination of study skins with inorganic mercury and other preservatives can be overcome by extracting organic mercury from the feather sample, because the mercury in feathers is effectively all organic mercury (Thompson & Furness 1989a).

2. The mercury concentrations in feathers must reflect body burdens accumulated during the period prior to moult. Studies by Honda, Nasu & Tatsukawa (1986) and by Braune & Gaskin (1987a) have clearly demonstrated that mercury accumulated in internal tissues is deposited into growing feathers at the onset of moult, the mercury concentrations in soft tissues decreasing as moult proceeds and the 'body pool' of mercury diminishes. Indeed, the mercury concentrations in feathers have been shown to correlate well with their relative position in a given moult sequence, the first-moulted feathers having higher mercury concentrations (Furness *et al.* 1986; Honda *et al.* 1986; Braune & Gaskin 1987a). Thus, it is essential to sample a consistent feather area from all birds, and Furness *et al.* (1986) showed that in seabirds, body feathers provide the most consistent and reliable indicator of the total plumage burden of mercury. Many studies have demonstrated a significant correlation between the mercury concentrations of internal tissues and those in defined feathers (Fimreite 1974; Furness & Hutton 1979; Hutton 1981; Ohlendorf *et al.* 1985; Thompson, Hamer & Furness 1991), showing that feathers can be used to indicate mercury uptake by birds over intermoult periods.

3. The dietary composition of a particular seabird species must have remained constant over the sampling period. As prey from different marine taxa exhibit different mercury concentrations (Bryan 1984), a shift in diet might result in a change in exposure to mercury. Fulmar diets have changed during the last 100 years but it is probable that the diets of Atlantic puffins, Manx shearwaters, north Atlantic gannets and great skuas have remained much the same over this period. There is, unfortunately, rather little information on diets in previous decades.

4. Seasonal movements of seabirds and their consequent exposure to mercury in different localities should be considered. There are few data available on the concentrations of mercury to which seabirds are likely to be exposed outwith the breeding season, although a marked alteration in the wintering area of a given species over the sampling period would be likely to introduce an alteration in mercury uptake. However, there is no evidence to suggest that any of the species studied here have undergone such a shift of wintering area.

From the results, it can be seen that only one species, the fulmar, exhibited significant decreases in mercury burdens between the birds sampled pre-1930 and those sampled post-1980. Marked dietary change during this period in fulmars may, in part, explain this trend. Fulmars at both localities were considered to feed extensively on whale offal up until about 1919, when whaling ceased in British waters (Fisher 1952). During the breeding season, contemporary fulmars were found to feed predominantly upon pelagic zooplankton at St. Kilda whilst sandeels *Ammodytes marinus* Raitt formed the bulk of the diet in Shetland during the sampling years (Furness & Todd 1984). There is evidence that the availability of sandeels increased considerably in the 1960s and 1970s (Sherman *et al.* 1981). Mercury concentrations in whale offal are considerably higher than those in marine zooplankton, while concentrations in sandeels are very low (Braune & Gaskin 1987b; Thompson 1990; Thompson *et al.* 1991). It seems unlikely, therefore, that this trend represents a decline in the mercury burden of the environment, but rather a change in diet with a corresponding decline in mercury concentrations.

A general increase in mercury contamination is indicated by the present data for Manx shearwaters, Atlantic puffins, north Atlantic gannets and great skuas. However, the increase varies among different populations. Changes were most pronounced in birds sampled to the west of Britain and in Ireland (Manx shearwater and Atlantic puffin; Table 1), but were modest towards the north of Britain and into the North Sea (great skua and north Atlantic gannet; Table 1). Dietary influences may play a part in this respect. Atlantic puffins and Manx shearwaters both feed almost exclusively upon small shoaling fish while breeding and are unlikely to have changed in diet (Harris 1984; Brooke 1990). By contrast, the more catholic diets of gannets and great skuas (Nelson 1978; Furness 1979) may render these species less suitable as monitors of long-term trends in contamination. The more pronounced increases in mercury contamination measured in Atlantic puffins and Manx shearwaters (Table 1; Figs 1 & 2) may reflect an enhanced monitoring value through a well-defined, relatively narrow diet. However, an additional factor with respect to such geographical variation may be increased pluvial deposition of mercury to the oceans

over specific areas, associated with anthropogenic emissions of mercury.

Atmospheric mercury is derived from both anthropogenic and natural continental sources, together with air–sea exchange processes (Fitzgerald 1986; Kim & Fitzgerald 1986). Fitzgerald (1986) noted that scavenging by rainfall is a major process by which atmospheric mercury is removed to the oceans. Indeed, Gill & Fitzgerald (1987) estimated the pluvial flux of mercury to the oceas to be nearly an order of magnitude greater than the fluvial flux.

Gardner (1975) suggested that relatively elevated mercury concentrations in sea-water samples from the sub-tropical north Atlantic (including the south-west approaches to Britain and the south-west of Britain and Ireland) were the result of increased mercury deposition in rainfall below jet streams carrying industrial mercury, particularly from North America, Japan and Europe. It can be envisaged that the above processes would lead to an enhancement of the mercury concentrations in water approaching the south and west of Britain, resulting in more pronounced increases within marine biota from such regions. The implied reduction in this effect (Table 1) as the prevailing oceanic currents progress northwards to southern Iceland and round the north of Scotland, thence turning south into the North Sea, may reflect the assimilation of mercury into the marine food chain or the sequestration of mercury by particulate matter. Several studies have noted depressed mercury concentrations in sea water associated with biological activity (Chester *et al.* 1973; Gardner 1975; Gill & Fitzgerald 1987). Furthermore, this pattern would not only lead to a more pronounced historical increase in mercury concentrations towards the south and west of Britain, but would also lead to higher mercury concentrations at any given time in biota from the south-west of Britain, as observed in the present data and in north Atlantic gannets by Walsh (1988).

The historical increase in mercury concentrations measured in common guillemots *Uria aalge* (Pontoppidan) and black guillemots *Cepphus grylle* (L.) from the Baltic and Kattegat (Appelquist *et al.* 1985) resulted, it was argued, from local anthropogenic emissions of mercury into rivers, and the subsequent accumulation of the element through food chains by coastal birds. The atmospheric transport of mercury to areas far from the initial source of production, suggested as being a likely explanation for the patterns of increase in avian mercury concentrations observed in most species in this study, contrasts with any localised effect and, rather, appears to indicate a more widespread increase in mercury contamination of the marine envionment.

The results presented here indicate that the large numbers of bird study skins held by museums around the world provide an excellent opportunity for the assessment of historical changes in mercury contamination of ecosystems, providing that possible confounding effects such as changes in diet are taken into consideration.

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