Chapter 5

Are Mercury Levels in Arctic Biota Increasing or Decreasing, and Why?

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5.1. Introduction

An analysis of the temporal trends for mercury in Arctic biota is important for scientists and managers concerned with making sound science-based policy with respect to changes in Hg in the Arctic environment. Long-term datasets (i.e., comparing modern with historical or pre-industrial Hg concentrations) can be used to estimate the relative importance of natural and anthropogenic Hg inputs in modern biota and the environment, while short-term datasets (i.e., covering the past one to three decades) illustrate how Hg concentrations have changed in recent times and suggest likely trends in the near-term future.

In the previous AMAP assessment of Hg in the Arctic environment (AMAP, 2005), evidence of increasing Hg concentrations from pre-industrial or historical to more recent times was presented for Arctic marine biota. Those trends were shown to continue in recent (short-term) datasets, primarily in higher-order marine biota from Canada and West Greenland. In contrast, time series for lower-order marine biota for Iceland and the European Arctic showed a general pattern of recently stable or declining Hg concentrations. The reasons for this spatial difference in trend patterns were not clear. Data for higher-order marine biota from Europe and Iceland were lacking as were temporal trend datasets for biota from Russia and Finland. One of the recommendations resulting from the previous AMAP assessment was that temporal trend monitoring of Hg concentrations should be continued in order to obtain longer and more statistically reliable trend analyses.

This chapter presents an update of the information available for both long-term and short-term trends, including a statistical meta-analysis of recent temporal trends in Hg concentrations in Arctic biota. The final section presents a discussion, using case studies, of the possible mechanisms driving the temporal trends observed.

5.2. How much higher are current mercury levels in Arctic biota than in the pre-industrial period, and thus what is the anthropogenic contribution to mercury in modern biota?

Information in this section is based on Dietz et al. (2009a).

5.2.1. Introduction

Although Hg is a naturally occurring element and, as such, has always been present in the environment, global human activity has led to a several-fold increase in Hg emissions to the atmosphere and inputs to oceans and rivers (Nriagu and Pacyna, 1988; Pacyna et al., 2006). In some areas of the Arctic, Hg concentrations in marine food webs have significantly increased in recent decades (Braune et al., 2005; see also Section 5.3), causing levels in some marine mammals, birds, and fish to reach the point where adverse biological effects might be expected (AMAP, 2003; see also Chapter 6). The question remains as to what extent global anthropogenic Hg emissions since the pre-industrial period have increased Hg concentrations in Arctic biota, and thus the exposure of humans who continue to use this wildlife as food. Calcified and keratinaceous hard tissues such as teeth, hair and feathers have the potential to help answer this question, because they tend to preserve well in dry, cold climates like polar regions (Outridge, 2005a) and are well represented at Arctic archeological sites and in museum collections. Also, work on laboratory and wild animal populations indicates that Hg concentrations in mammalian hair and teeth and bird feathers are correlated with the intake of organic and inorganic Hg and with organ Hg levels (Born et al., 1991; Eide and Wesenberg, 1993; Eide et al., 1995; Outridge et al., 2000; Bearhop et al., 2000a). Thus, analysis of historical and modern samples of hard tissues can be informative about the long-term changes in Hg intake and body burdens in wildlife.

The literature on this subject is not extensive. Several early studies reported data on biotic Hg concentrations spanning several centuries: in human teeth from Norway (Eide et al., 1993; Tvinnereim et al., 2000), human and seal hair from Greenland (Hansen et al., 1989), and human hair from Arctic Canada (Wheatley and Wheatley, 1988). These studies compared two time periods (pre-industrial and modern), and were based on relatively few samples; data on age and possible dietary differences between time periods were not available. More recently, Hg concentrations were examined in Greenland polar bear (*Ursus maritimus*) hair collected at intervals during the period 1892 to 2001 as well as in two pre-industrial samples dated 1300 AD from northwestern Greenland (Dietz et al., 2006a). The tooth Hg content of scores of Canadian beluga (*Delphinapterus leucas*), walrus (*Odobenus rosmarus*) and ringed seals (*Phoca hispida*) from pre-industrial, historical (19th and early to mid-20th century) and modern populations were examined by Outridge et al. (2002, 2005a, 2009), and the long-term changes were assessed within the context of animal age structure and dietary information inferred from tooth stable carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotope data. Time trends in Hg concentrations in primary feathers of West Greenland gyrfalcons (*Falco rusticulus*), peregrine falcons (*F. peregrinus*), and white-tailed sea eagles (*Haliaeetus albicilla*) for the period 1850 to 2004 have also been reported (Dietz et al., 2006b). This section combines the various published time series to calculate the average anthropogenic contribution to present-day Hg concentrations in Arctic biota, and to determine the historical timing of any changes. Note that the modern values used are the maximum annual mean values, which generally fell within the period 1973 to 2003, for specific species and locations. The caveats for these interpretations are also discussed, especially the effects of possible changes in feeding behavior (diet and location) as extrapolated from $\delta^{13}C$ and $\delta^{15}N$ measurements, and potential contamination or loss of Hg in archaeological and museum collections of ancient biological tissues.

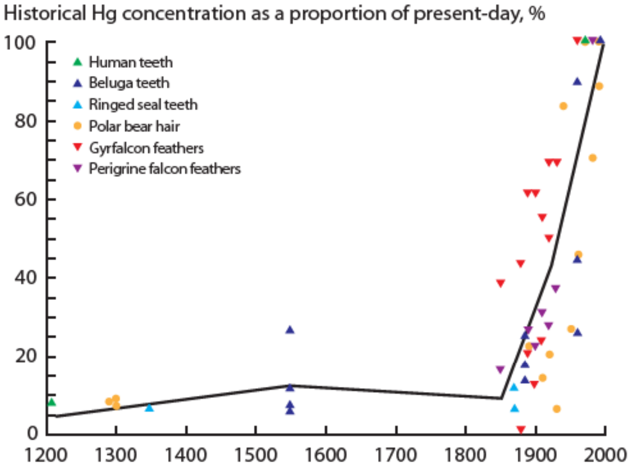


Figure 5.1. Historical trends in Hg concentration in hard tissues of various Arctic biota, expressed as a percentage of modern maximum annual average concentrations. Most points plotted represent mean values of multiple samples; for beluga and seal teeth, these are presented for different age classes (see Dietz et al., 2009a). Original data sources: beluga teeth (Outridge et al., 2002, 2005a, 2009); ringed seal teeth (Outridge et al., 2009); human teeth (Eide et al., 1993; Tvinnereim et al., 2000); polar bear hair (Wheatley and Wheatley, 1988; Dietz et al., 2006a); gyrfalcon and peregrine falcon feathers (Dietz et al., 2006b). Regions covered by these datasets include parts of the Canadian Arctic, Greenland Arctic, and northern Norway.

5.2.2. The anthropogenic mercury contribution in modern Arctic biota

Similar to other long-term archives such as lake sediments and peat bogs, estimating the anthropogenic Hg contribution to modern biota requires a comparison of modern Hg concentrations (Hg concentrations denoted as [Hg]) in hard tissues against those from the pre-industrial period (taken to be pre-1800 AD) in a given animal population from a defined region. The modern [Hg] increase, if any, in the population is taken to be the anthropogenic contribution, which was calculated in two ways by Dietz et al. (2009a). First, however, the published Hg data need to be represented in a consistent and comparable manner across tissues and species, because absolute concentrations in the hard tissues of various species varied by three to four orders of magnitude both in the pre- and post-industrial period (Dietz et al., 2009a). Therefore, the maximum present-day annual median concentration in each dataset was initially set to 100%, and the median historical and pre-industrial concentrations are given as percentages of the recent maximum, using the following formula:

Historic proportion of present-day [Hg] (%) = (median historic [Hg] / median recent maximum [Hg]) × 100

The resulting data distribution, which extends back over eight centuries, is shown in Figure 5.1.

The conversion of absolute concentrations to percentages of the modern maxima indicates a similar overall pattern across species and regions. Pre-industrial period hard tissues contained a median of 7.6% (range: 5.6% to 25.8%) of the maximum annual average Hg levels recorded for the same species in the same areas during recent decades. This finding means that, on average, 92.4% (range: 74.2% to 94.4%) of the present-day Hg in Arctic wildlife is likely to be of anthropogenic origin. This conclusion assumes that the long-term increase in Hg concentrations in Arctic biota occurred solely as a result of increased inputs of Hg from pollution sources, and that other potentially important factors such as changes in the cryosphere, biogeochemical processes or ecological factors (see Section 5.4) did not significantly affect the long-term increasing trend of biotic Hg levels. The possible effect of many of these other processes is difficult to assess within this context, first because of a lack of long-term data concerning how these processes have changed since pre-industrial times in the specific areas in which the long-term biotic studies were conducted, and second because of uncertainty as to how exactly and to what degree these types of change affect Hg levels in Arctic biota (see Chapter 4).

The possible effect of dietary changes on long-term trends in Hg concentration were assessed in beluga and ringed seal in the Canadian Arctic using stable carbon (C) and nitrogen (N) isotope data by Outridge et al. (2002, 2005a, 2009), and discussed in greater detail by Dietz et al. (2009a) (see also Section 5.2.5). Briefly, the Beaufort beluga trends (Outridge et al., 2002, 2009) were not accompanied by significant changes in C and N isotope ratios; which indicates that dietary changes do not explain the order of magnitude Hg increases in that population since pre-industrial times. The trophic position of Amundsen Gulf ringed seals, inferred from stable N isotopes, did not change over time, but feeding location inferred from stable C isotopes may have changed. $\delta^{13}C$ values in modern seals were significantly lower than in those from the 14th and 19th centuries, which could indicate a recent shift to more pelagic, offshore feeding (Outridge et al., 2009). A shift to more pelagic feeding over the centuries may have exposed the modern population to prey items with a lower average Hg content than inshore, benthic prey (Stern and Macdonald, 2005). If this was the case, then this shift would have caused the anthropogenic contribution to modern seal Hg to have been under-estimated rather than over-estimated. Section 5.2.5 includes an example in which a slight change in trophic position ($\delta^{15}N$) resulted in an over-estimate of the anthropogenic component of a central Arctic beluga population. However, there is no *a priori* reason to believe that adjustments for long-term dietary changes across the entire long-term biotic dataset, covering several species in different Arctic regions, would consistently bias the Hg trend results in one particular direction (higher or lower) compared to analyses of the unadjusted trends.

The only Arctic species studied so far that did not show a significant modern increase was walrus near Igloolik, Canada (Outridge et al., 2002). It is also the only species in this compilation which usually feeds at a very low trophic level (on clams and other bivalves). Low trophic-level species were predicted to be relatively immune to increases in environmental Hg levels compared to higher trophic-level species, because biomagnification would have little effect on Hg concentrations in these species or their prey in response to elevated environmental levels (Bernhard and Andreae, 1984). The results reported here support this prediction and so the walrus data were not included in the above calculations.

Although there are no long-term studies close to developed areas of the world, recent findings from Antarctica indicate that both the Arctic and the Antarctic have experienced significant increases in biotic Hg levels since the Industrial Revolution, with the increases in Antarctic biota markedly less than those in the Arctic. Hair of southern elephant seal (*Mirounga leonine*) retrieved from an Antarctic lake sediment core displayed considerable variation in [Hg] in the pre-industrial period (Sun et al., 2006). However, the minimum values (~ 1 µg/g) were on average about 60% of those in the uppermost sediment layer (1.7 µg/g), which would suggest an anthropogenic contribution of about 40% in modern elephant seals.

The long-term increases found by Hansen et al. (1989) in seal and human hair between 15th century Qilakitsoq Inuit mummies and 1970 were less pronounced (pre-industrial levels 23.1% of present [4.3-fold increase] and 31.6% [3.2-fold increase], respectively) than the above studies. Likewise, Wheatley and Wheatley (1988) reported that modern Hg levels in human hair from the Canadian Arctic were only several times higher than in pre-industrial samples. Both studies were excluded from the calculations above, but the effect was minimal; including these datasets would have only increased the median pre-industrial baseline value to 12.6% from 7.6% (with a resulting anthropogenic contribution of 87.4%). They were excluded because of the possibility of confounding factors which may have decreased the calculated anthropogenic component in the samples. A lower intake of high trophic-level marine foods by present-day Inuit compared to their forebears could explain the lower than expected modern increase in hair Hg (see Kinghorn et al., 2006). Also, modern hunting pressures may have significantly reduced the average age of harvested seals (and other human food items) compared to historic times when hunting pressure was presumably lower (R. Dietz, National Environmental Research Institute, Denmark, unpubl. data). This would have tended to reduce the Hg increases in harvested seal hair and the modern human Hg intake from those seals (as well as Hg levels in modern human hair). To what extent diagenetic contamination could have increased the historic hair Hg concentrations is difficult to assess, although the fact that the Qilakitsoq mummies and their clothing were lying on dry rock and not in soil (Hansen et al., 1989) may make it unlikely. Alternatively, the ancient light and heating technique of burning blubber oil lamps in small enclosed spaces may have created a substantial external Hg contamination of the hair of the Inuit and their seal-skin clothing which resulted in higher than normal, and inaccurate, hair Hg concentrations.

5.2.3. Timing of mercury increases over the past 150 years

Dietz et al. (2009a) used two separate calculations of the long-term change in the anthropogenic Hg component in Arctic biota. While both formulas agreed on the average input in modern biota (i.e., 92.4%), there were subtle differences in the rate of change between 1850 and recent decades. However, these differences are minor and do not detract from the overall picture. As is clear from Figure 5.2, both measures indicated that the 20th century showed a steep increase in biotic Hg compared to relatively constant values over the previous four to six centuries. As no Hg data were available from the period between the 16th century and about 1850, it is not possible to precisely identify the onset of the steep industrial era increase. However, based on the shape of the curves, it seems plausible that Hg started to increase somewhere between 1850 and 1900, with a clear acceleration in the rate of increase after 1900. The overall pattern is clearly seen in several individual studies. When assessing the temporal trends for two adjacent and complementary datasets – beluga in the Beaufort Sea and ringed seal in the Amundsen Gulf – Outridge et al. (2009) concluded that Hg levels in marine biota in this region had

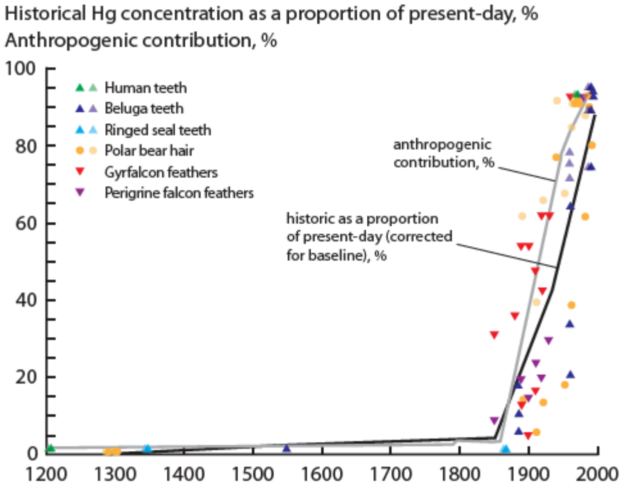


Figure 5.2. Change in the anthropogenic contribution to Hg levels in Arctic biota since the pre-industrial period, calculated from the data presented in Figure 5.1. The figure shows the historic proportion of the present level corrected for the baseline percentage (black line and darker symbols) and the percentage contribution of anthropogenically-derived Hg (grey line and lighter symbols). See Dietz et al. (2009a) for details.

not increased from pre-industrial times up to at least the late 19th century, with the most substantial increases occurring between then and 1960/61. Beluga teeth collected around Somerset Island during 1894 to 1998 showed Hg increases of 4.1- to 7.7-fold overall, but with no change between the late 19th century and the 1920s to 1940s, indicating that most or all of the increase has taken place after the early 20th century (Outridge et al., 2005a). Similarly, Hg trends in polar bear hair from eastern and northwestern Greenland displayed minimal increases until as late as 1950 (Dietz et al., 2006a). However, Hg trends in Greenlandic birds of prey since the 1850s (Dietz et al., 2006b) generally increased earlier and more rapidly than the previously discussed marine mammal datasets. As the bird species exploited a mixed terrestrial and aquatic food chain, the dichotomy between species with different food-chain bases suggests the possibility of systematic differences between marine- and land-based food webs in terms of the immediacy and effectiveness of atmospheric Hg deposition as a rapid forcer of biological Hg trends (Outridge et al., 2009).

The average rate of Hg increase over the past 150 years was usually within the range of 1% to 4% per year, based on linear regressions of tissue Hg concentration versus time. Polar bear hair showed a significant average 2.1% per year increase in Hg concentration from 1920 to 1991 in northwestern Greenland, and a 3.1% per year increase in the period 1892 to 1973 in northeastern Greenland (Dietz et al., 2006a). Mercury in primary feathers of western Greenland gyrfalcons, peregrine falcons, and white-tailed sea eagles from the period 1880 to 1935 showed average increases in the range 1.1% to 4.5% per year, whereas for 1880 to 1960 the average increase was slower, at 0.4% to 0.9% per year (Dietz et al., 2006b).

5.2.4. Preservation of the original mercury concentration in pre-industrial samples

Pre-industrial samples were invariably retrieved from archaeological sites, and both pre-industrial and historical era specimens were often stored for many decades in museums or other facilities prior to their recent analysis, sometimes open to the ambient air or in poorly-sealed bags and boxes. Archaeological material, which may have been in contact with soil, sediment or moisture for extended periods, can potentially experience physical, chemical or biochemical changes ('diagenesis'; Jackson, 1997) that may increase or decrease the original biogenic Hg concentration. Two additional preservation issues are: exposure to inorganic Hg contamination in museums which employed elemental Hg sublimate as a fungicide, and possible Hg volatilization from specimens over long periods at typical Arctic soil temperatures. The consequences of diagenesis and inadequate preservation can be bi-directional: either over-estimation of the modern increase and thus the anthropogenic contribution (i.e., when Hg was lost from the historical material) or under-estimation (when Hg contamination occurred). Outridge (2005a) and Dietz et al. (2009a) addressed these issues in detail. This section briefly reviews the evidence for the stability of Hg in the bioarchives included in this review.

Empirical evidence for the stability of Hg in hard tissues over any time span is very limited. However, Dietz et al. (2009a) concluded that over-estimation of the anthropogenic component in modern biota (because of losses of Hg from historical material) was extremely unlikely. Keratin-based samples such as hair and feathers exhibit surprising resistance to Hg loss. After up to eight months exposure to various harsh treatments including continuous ultraviolet (UV) light, heating to 100 °C, *in situ* exposure at various northern sites, and freezing, bird feathers lost less than 10% of their Hg content (Appelquist et al., 1984). Tanning of skins significantly alters the inorganic Hg and methylmercury (MeHg) content of hair (Newman et al., 2005), but none of the datasets included here used tanned skins.

Two alternative tests for the effect of diagenetic alteration of Hg in hard tissues were proposed by Outridge (2005a): evaluation of hard tissue Hg–age relationships, and of C:N concentration ratios. The first test is based on the premise that Hg concentrations in modern animal hard tissues are almost universally correlated with age. If diagenesis was absent in a historical group of samples, then a significant tissue Hg–age correlation should also be found in the group. However, if diagenetic alteration of tissue Hg levels had occurred, then the measured Hg values in the historical group should not be correlated with animal age. This test assumes that sample diagenesis or Hg volatilization from samples is independent of the original Hg concentration. This assumption is reasonable because elemental Hg(0), which is the only Hg form exhibiting marked volatilization at normal environmental temperatures, does not occur in significant quantities in biological tissues because of its rapid biological reactivity. Methylmercury, a non-volatile Hg species, dominates in hair and feathers (Bearhop et al., 2000b; FAO/WHO, 2003), and may also in teeth (Eide et al., 1994). When this test was applied (i.e., in the beluga tooth studies by Outridge et al. 2002, 2005a, 2009), significant tooth Hg–age regressions were found in pre-industrial or historical groups, suggesting that the baseline beluga Hg data were reliable. Mercury levels in the pre-industrial and 19th century ringed seal teeth studied by Outridge et al. (2009) were below detection, thus invalidating this test. However, the second test involving elemental C:N concentration ratios can be applied in cases like this. This test is based on the preferential metabolism by bacteria of carbon over nitrogen during diagenetic alteration of ancient calcified material, which results in a decrease of C:N values (DeNiro, 1985). In historical Somerset Island beluga teeth (Outridge et al., 2005a), no evidence of diagenetic alteration was found; C:N ratios were virtually identical between modern (3.1 ± 0.1) and historical (3.3 ± 0.1) samples. Similarly good preservation of ringed seal teeth was reported by Outridge et al. (2009), with 14th and 19th century samples having C:N values (3.2 ± 0.06 and 3.2 ± 0.02, respectively) similar to modern teeth (3.2 ± 0.06; 1-way ANOVA $p > 0.10$).

5.2.5. Using stable carbon and nitrogen isotopes in long-term trend studies

Another factor which could influence the interpretation of long-term biotic Hg trends is the possibility that feeding behavior (dietary trophic level and location) of the animal population changed significantly during the study period. One means of assessing this possibility, and of correcting [Hg] for any changes, involves the concurrent measurement of different stable isotopes along with Hg concentrations in animal hard tissues.

Food web stable isotope measurements, especially using stable isotope assays of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$), can provide a time-integrated assessment of the source of feeding and relative trophic level of animals, respectively. Since source of feeding determines the exposure of organisms to Hg, and trophic level influences the degree of bioaccumulation or biomagnification of Hg in an organism's tissues, stable isotope measurements can assist in the interpretation of trend data to a degree previously unavailable, by ruling out the possibility of, or correcting for, concomitant changes in population feeding behavior (e.g., Braune et al., 2001, 2002; Outridge et al., 2002, 2005a, 2009). This is especially true for organisms which can markedly alter their trophic level or migratory or feeding origin. An in-depth discussion of the application of stable isotope systems to contaminant trend interpretation is provided by Dietz et al. (2009a). As discussed in Section 5.2.2, most of the beluga and seal long-term datasets were either not accompanied by dietary changes, or evidenced dietary changes which would, if anything, result in an under-estimation of the anthropogenic input.

As an example of how a relatively subtle shift in trophic position over the long term can result in over-estimation of the anthropogenic component, the paper by Outridge et al. (2005a) which reported historical changes in Hg levels in teeth of beluga at Somerset Island, Northwest Territories, Canada was revisited. That study determined that tooth Hg levels increased with age for both late 19th century and mid-1990s animals but that this relationship was much more pronounced in the modern animals ($r^2 = 0.63$ *vs* 0.17). Tooth $\delta^{15}N$ values of modern animals were slightly higher than those of historical samples (18.5 ± 0.4‰ *vs* 17.8 ± 1.3‰). This corresponds to a mean trophic level increase of 0.18 (using the trophic enrichment value of 3.8‰ for Arctic marine food webs, see Campbell et al., 2005). Although it is not clearly understood how changes in the mean trophic level of beluga alters its average Hg content, Loseto (2007) estimated a total Hg (THg) biomagnification factor (BMF) of 12.8 to 16.0 for beluga feeding primarily on a fish diet. Thus, a trophic increase over time of 0.18 should correspond to a correction (subtraction) of the Hg increase from the 1890s to 1990s of about 2.2 to 2.9 (i.e., 0.18 times the BMF of 12.8 to 16.0). Using an average correction factor of 2.5, corrected [Hg] increases in Somerset beluga teeth from the 1890s to the 1990s ranged from a factor of 1.6 in younger animals to 5.2 in older animals, which are smaller than indicated by the isotopically-uncorrected data (Table 5.1). Corrections like these rely on assumptions including no temporal variation in food web stable isotopic values.

Changes in food web $\delta^{13}C$ values can occur due to changes in nutrients available to primary producers or other factors influencing plant growth rates (e.g., Laws et al., 1995). Another factor of interest regarding $\delta^{13}C$ is the isotopic change in atmospheric carbon dioxide (CO_2) that has occurred due to anthropogenic burning of fossil fuels. Normalizing terrestrial or freshwater food webs for this effect is relatively straightforward due to the well-mixed atmosphere and fast equilibrium with lakes (e.g., Bada et al., 1990). However, it has proven to be much more difficult to predict the consequences of this effect in the world's oceans (but see Hilton et al., 2006). This phenomenon, known as the 'Seuss effect' (Quay et al., 1992), is complicated because of the differential effects of deep ocean upwelling and mixing processes that may not be in equilibrium with atmospheric CO_2. Certainly at high latitudes, upwelling and lack of mixing may depress the Seuss effect in marine biota (Schell, 2000; but see Cullen et al., 2001). Recently, based on a long-term isotopic record of teeth from northern fur seals (*Callorhinus ursinus*), Newsome et al. (2007) provided more convincing evidence that the Seuss effect has been responsible for decreases in $\delta^{13}C$ of animal tissues over that period. However, unlike the model used to correct for trophic level changes using $\delta^{15}N$ measurements, quantitative corrections to Hg trends using $\delta^{13}C$ values are not yet possible because it is not known how Hg levels vary across a benthic-to-pelagic gradient of beluga prey. Better use of $\delta^{13}C$ values in future studies of Hg trends will be possible when the isotopic and Hg benthic-pelagic effect is better described for areas of interest (e.g., Stern and Macdonald, 2005; Loseto, 2007; Loseto et al., 2008a).

5.3. In which areas and species have mercury levels recently increased or decreased?

In the previous AMAP Hg assessment (AMAP, 2005), compelling evidence was presented for increasing trends in Hg concentration in recent decades, primarily for marine birds and mammals from the Canadian and West Greenland Arctic. In contrast, Hg concentrations in the European Arctic in lower-order marine biota and terrestrial mammals were either stable or declining. Furthermore, based on the analysis of 42 datasets with 4 to 29 years of data for Hg in Arctic biota, Bignert et al. (2004) concluded that only 10% of the 42 time series were 'adequate' with respect to the number of years of data required to meet reasonable standards of statistical power. For this assessment, a review of Hg trends reported in the recent literature was undertaken followed by an updated meta-analysis of time-series datasets for Hg concentrations in Arctic biota.

Table 5.1. Changes in geometric mean mercury concentrations in teeth of beluga at Somerset Island, Northwest Territories, Canada between the late 19th century and the 1990s, with and without normalization using $\delta^{15}N$ values. The analysis is based on data provided by Outridge et al. (2005a), except that the ages have been doubled because of a change in beluga ageing technique (Stewart et al., 2006). The assumed increase in beluga mercury concentration and trophic level was a factor of 12.8 to 16 based on Loseto (2007).

Sample	[Hg] 19th century, ng/g dw	[Hg] 1990s, ng/g dw	Uncorrected factor increase	Corrected factor increase
10y old	6.6	26.8	4.1	1.2 – 1.9
20y old	13.4	78.4	5.9	3.0 – 3.7
30y old	23.0	178	7.7	4.8 – 5.5

5.3.1. Selection of tissues and chemical forms of mercury for monitoring

The chemical form of Hg and the tissues in which it is measured varies among monitoring programs. The most bioavailable and toxic form of Hg is MeHg (Wiener et al., 2003). In seabirds, concentrations of MeHg appear to be less variable across tissues than THg with a rank order of MeHg concentrations in various tissues as follows: liver > kidney > muscle (Kim et al., 1996). The rank order for concentrations of THg across tissues seems to be similar for marine mammals: liver > kidney > muscle > *muktuk* (Lockhart et al., 2005b) or in the case of polar bear: kidney > liver > muscle > adipose (Dietz et al., 1995, 2000a). In birds, nearly 100% of the Hg transferred by breeding female birds to their eggs is in the form of MeHg (Wiener et al., 2003), making eggs a valuable, non-intrusive monitoring matrix for Hg. However, interpretation of Hg concentrations measured in biota is a complex issue requiring an understanding of sources and chemical forms of Hg in the environment, food web biomagnification, and animal physiology. For a detailed discussion of these factors, see Chapter 3.

It has been shown that the percentage of THg present as MeHg in liver decreases as concentrations of THg increase in some higher trophic-level species (Dietz et al., 1990). This has also been documented for seabird liver (Kim et al., 1996). This may be due, in part, to the capability of some seabirds, as well as marine mammals, to demethylate MeHg into tiemanite in the liver (Dietz et al., 1990; Wagemann et al., 1998; Chapter 6). Given this potential for changing concentrations of MeHg in some tissues, it is perhaps best to monitor concentrations of THg rather than MeHg for tracking environmental trends.

5.3.2. Mercury trends in the recent literature

A summary of recently published temporal trends in THg in Arctic biota is presented in Table 5.2. The literature contains many datasets which include only two or three years of data. This table includes only those datasets with at least three years of data. The majority of such datasets were from Canada and Greenland with only one dataset from Alaska, three from Norway, and four from the Faroe Islands.

Table 5.2 includes datasets ranging from 3 to 14 years of data and covering variable time periods between 1973 and 2007. Of the four published time series for terrestrial biota, two showed a decreasing Hg trend and two showed no change. All except one of the recently published time series for freshwater fish were from Canada. Of the 18 datasets reported for freshwater fish, none were significant; one showed a non-significant increasing trend, three showed a decreasing tendency, and 14 showed no trends at all. However, of 167 short time series for

Table 5.2. Summary of temporal trends for total mercury in Arctic biota from the recent literature.

Area	Species[a]	Tissue	Period (n)[b]	Trend	Trend significance according to author[c]	
Terrestrial						
Faroe Islands	Mountain hare	liver	1997-2006 (5)	→	Ns	Hoydal and Dam, 2009
Central West Greenland	Caribou	liver	1995-1999 (4)	→	Ns	Rigét et al., 2004
Yukon, Canada	Caribou	kidney	1994-2003 (10)	↓	*	Gamberg et al., 2005
Yukon, Canada	Moose	kidney	1994-2003 (10)	↓	*	Gamberg et al., 2005
Freshwater						
Lake Á Mýrunum, Faroe Islands	Arctic char (landlocked)	muscle	2002-2007 (3)	→	Ns	Hoydal and Dam, 2009
Lake Hazen, Canada	Arctic char (landlocked)	muscle	1990-2006 (7)	→	Ns	Gantner et al., 2009
Amituk Lake, Canada	Arctic char (landlocked)	muscle	1989-2003 (5)	→	Ns	Muir et al., 2005
Char Lake, Canada	Arctic char (landlocked)	muscle	1993-2003 (5)	→	Ns	Muir et al., 2005
Resolute Lake, Canada	Arctic char (landlocked)	muscle	1993-2003 (7)	→	Ns	Muir et al., 2005
Lac Ste. Therese, Canada	Lake trout	muscle	1980-2002 (4)	→	Ns	Evans et al., 2005a
Lac Ste. Therese, Canada	Walleye	muscle	1975-2002 (5)	→	Ns	Evans et al., 2005a
Lac Ste. Therese, Canada	Pike	muscle	1980-2002 (4)	→	Ns	Evans et al., 2005a
Lac Ste. Therese, Canada	Whitefish	muscle	1992-2002 (3)	→	Ns	Evans et al., 2005a
Mackenzie River, Canada	Burbot (males)	muscle	1985-2001 (6)	↑	Ns	Evans et al., 2005a
Mackenzie River, Canada	Burbot (females)	muscle	1985-2001 (6)	→	Ns	Evans et al., 2005a
Great Slave Lake, Canada	Pike	muscle	1976-2002 (12)	→	Ns	Evans et al., 2005a
Great Slave Lake, Canada	Lake trout	muscle	1979-2002 (9)	→	Ns	Evans et al., 2005a
Great Slave Lake, Canada	Burbot	muscle	1975-2002 (10)	→	Ns	Evans et al., 2005a
Slave River, Canada	Burbot	muscle	1991-2002 (8)	→	Ns	Evans et al., 2005a
Lake Laberge, Canada	Lake trout	muscle	1993-2002 (6)	↓	Ns	Evans et al., 2005a
Kusawa Lake, Canada	Lake trout	muscle	1993-2002 (4)	↓	Ns	Evans et al., 2005a
Quiet Lake, Canada	Lake trout	muscle	1992-2002 (4)	↓	Ns	Evans et al., 2005a

Marine						
Northwest Greenland	Shorthorn sculpin (large)	liver	1987-2004 (4)	→	Ns	Rigét et al., 2007
Central West Greenland	Shorthorn sculpin (small)	liver	1994-2002 (5)	→	Ns	Rigét et al., 2007
Central West Greenland	Shorthorn sculpin (large)	liver	1994-2004 (7)	→	Ns	Rigét et al., 2007
Northern Norway	Herring gull	egg	1983-2003 (3)	→	Ns	Helgason et al., 2008
Northern Norway	Atlantic puffin	egg	1983-2003 (3)	→	Ns	Helgason et al., 2008
Northern Norway	Black-legged kittiwake	egg	1983-2003 (3)	→	Ns	Helgason et al., 2008
Koltur, Faroe Islands	Black guillemot	egg	1999-2006 (6)	→	Ns	Hoydal and Dam, 2009
Skúvoy, Faroe Islands	Black guillemot	egg	1999-2006 (6)	↑	Ns	Hoydal and Dam, 2009
Seymour Island, Canada	Ivory gull	egg	1976-2004 (3)	↑	Ns	Braune et al., 2006
Lancaster Sound, Canada	Thick-billed murre	egg	1975-2003 (8)	↑	*	Braune, 2007
Lancaster Sound, Canada	Northern fulmar	egg	1975-2003 (7)	↑	*	Braune, 2007
Lancaster Sound, Canada	Black-legged kittiwake	egg	1975-2003 (6)	→	Ns	Braune, 2007
Northwest Greenland	Walrus	liver	1977-2003 (7)	→	Ns	Rigét et al., 2007
Northwest Greenland	Ringed seal	liver	1984-2004 (5)	↑	Ns	Rigét et al., 2007
Central West Greenland	Ringed seal	liver	1994-2004 (7)	→	Ns	Rigét et al., 2007
Central West Greenland	Ringed seal	liver	1999-2004 (6)	↑	*	Rigét et al., 2007
Hudson Strait, Canada	Ringed seal	liver	1989-2002 (4)	↑	Ns	Braune et al., 2005
Ungava Bay, Canada	Ringed seal	liver	1989-2002 (3)	↓	Ns	Braune et al., 2005
Qausuittuq, Canada	Ringed seal	liver	1976-2000 (3)	↓	Ns	Braune et al., 2005
Ulukhaktok, Canada	Ringed seal	muscle	1973-2007 (10)	→	Ns	Gaden et al., 2009
Faroe Islands	Pilot whale	muscle	2001-2007 (6)	→	Ns	Hoydal and Dam, 2009
Mackenzie Delta, Canada	Beluga	liver	1981-2002 (8)	↑	Ns	Lockhart et al., 2005b
Pangnirtung, Canada	Beluga	liver	1984-2002 (5)	→	Ns	Lockhart et al., 2005b
Coral Harbour, Canada	Beluga	liver	1993-2000 (3)	→	Ns	Lockhart et al., 2005b
Lake Harbour, Canada	Beluga	liver	1994-2001 (3)	→	Ns	Lockhart et al., 2005b
Central East Greenland	Polar bear	liver	1983-2000 (9)	→	Ns	Rigét et al., 2004
Central East Greenland	Polar bear	hair	1973-2001 (14)	↓	*	Dietz et al., 2006a
Canada	Polar bear	liver	1982-2002 (3)	↑	Ns	Rush et al., 2008
Alaska	Polar bear	liver	1993-2002 (8)	↓	Ns	Kannan et al., 2007

[a] Mountain hare (*Lepus timidus*); caribou (*Rangifer tarandus*); moose (*Alces alces*); Arctic char (*Salvelinus alpinus*); lake trout (*Salvelinus namaycush*); walleye (*Stizostedion vitreum*); pike (*Esox lucius*); whitefish (*Coregonus clupeaformis*); burbot (*Lota lota*); shorthorn sculpin (*Myoxocephalus scorpius*); herring gull (*Larus argentatus*); Atlantic puffin (*Fratercula arctica*); black-legged kittiwake (*Rissa tridactyla*); black guillemot (*Cepphus grylle*); ivory gull (*Pagophila eburnea*); thick-billed murre (*Uria lomvia*); northern fulmar (*Fulmarus glacialis*); walrus (*Odobenus rosmarus*); ringed seal (*Phoca hispida*); pilot whale (*Globicephala melas*); beluga (*Delphinapterus leucas*); polar bear (*Ursus maritimus*); [b] 'n' represents number of sample years during time period; [c] statistically significant (*) and non-significant (Ns) time trends.

fish from 45 Canadian lakes and rivers covering variable periods between 1971 and 2002, and which had been measured for Hg on more than one occasion, 34 comparisons (20%) suggested that Hg levels had increased while 22 (13%) indicated decreases when two years were compared (Lockhart et al., 2005a). The majority showed no statistically significant changes with time. The highest number of increasing trends in Hg concentration was seen in marine biota. Of the 29 datasets reported in the literature for the marine environment, nine (31%) suggested increasing trends (only three of those were significant), and only four (14%) showed decreasing concentrations (one was significant). The other 16 (55%) datasets showed no trend.

The following section presents the results of an updated meta-analysis of available time series datasets. Some of the datasets included in the meta-analysis are based on the datasets presented in Table 5.2 which have been updated with more recent data. In some cases, data subsets have been created for analysis based on size, age, or sex. Inclusion / exclusion of co-variables may also differ between the published Hg trends and the meta-analysis presented here. The objective was to analyze Hg time-series datasets in a consistent and comparable manner.

5.3.3. Meta-analysis of recent temporal trends of mercury in Arctic biota

The present 'meta-analysis' of recent temporal trends in Hg concentration in Arctic biota is a part of the AMAP Hg assessment process. Previous temporal trend workshops were held in Copenhagen in 2001 and Stockholm in 2006 (see AMAP, 2007).

5.3.3.1. Available data

At the AMAP Mercury Expert Group meeting held in Quebec City in December 2008, it was decided that only time series with at least six years of data, and including years both before and after 2000, be included in the meta-analysis of temporal trends. The inclusion of pre-2000 data was intended to ensure that the time series were representative of a time period sufficiently long to detect any change. A total of 83 time series fulfilling these requirements were available as follows: Canada (30), Faroe Islands (7), Denmark (Greenland) (15), Iceland (15), Norway (9), Sweden (6), and the United States (Alaska) (1). The datasets included time series of Hg levels in various tissues and organs of a range of species (including shellfish, freshwater and marine fish, marine mammals, terrestrial mammals, and seabirds). In a few cases, Hg was analyzed in different tissues or organs of the same animals and these were treated as separate time series. No time series were available from Russia or Finland, and so the geographical coverage was essentially semi-circumpolar. The time series covered periods ranging from 7 to 38 years (average time span 17.3 years), with data available for an average of 11.0 years. However, within this overall pattern, datasets were generally characterized either by series based on annual sampling, or by non-contiguous series of samples representing different time periods with gaps of several years between them.

5.3.3.2. Statistical application

The statistical application used was PIA developed by Anders Bignert – a custom application based on Fortran code and developed to run on Windows systems. A ‘PIA distribution package’ (a zip file containing the program, documentation and examples) is available on the public area of the AMAP website (www.amap.no).

The PIA application applies a trend assessment on ‘index values’ (by default, the annual geometric mean concentration) for each data year and employs a running-mean smoother (default, 3 years) to test for non-linear trend components (as described by Bignert, 2001 and Bignert et al., 2004). The application can also analyze trends in concentrations adjusted for a single selected covariate. In addition to the statistical results, the application output includes a trend plot and a simplified ‘trend interpretation’. A directive file specifies options for the analysis to be conducted, including options to vary the power and alpha values used in the statistical tests, and to apply an analysis based on median or geometric mean index values. The categorization of trends by the software is based on a robust regression-based analysis to detect trends, described in detail by Nicholson et al. (1998), as well as an ordinary log-linear regression. The two methods differ slightly when testing for a log-linear trend component and, in a few cases, these results conflicted (one showed a p-value just below and the other just above 5%). In these few cases, the ultimate trend evaluation was based on closer examination of the detailed results, such as how well the smoother described the trend compared to a log-linear regression (comparing the standard deviation of the residuals from the two trend lines), and visual inspection of the PIA trend plot to check for dubious points.

5.3.3.3. Trend analyses

At the AMAP Mercury Expert Group meeting held in Quebec City in December 2008, it was decided that the trend analyses should be run using annual geometric means and the 3-year running-mean smoother. However, in two cases, annual median values were applied instead because of the occurrence of several data points below the analytical detection limit. Medians are less influenced than means by the assumptions necessary to deal with below detection values.

It was the intention of this exercise to perform the temporal trend analyses in as consistent a manner as the available data allowed. However, the data providers are the scientists most familiar with their data, and their advice for treatment of covariates and the basis of concentrations (wet weight or dry weight) has been followed. During the AMAP Workshop on Statistical Analysis of Temporal Trends of Mercury in Arctic Biota held in November 2006, valuable experience was gained in making adjustments for covariates in trend analyses of Arctic Hg time series. Results from the workshop (AMAP, 2007) demonstrated that age and length or weight were powerful covariates, and the data runs also indicated the need to transform Hg data (e.g., using log transformations) prior to conducting trend analysis. Stable isotope values (δ^{13}C, δ^{15}N) can also be used to adjust Hg concentrations for species known to undergo shifts in diet (see Chapter 3). However, stable isotope values were not included as a covariate in the statistical analyses; rather they were used prior to PIA analysis to group some populations into sub-groups according to feeding behavior.

5.3.3.4. Data adjustment for covariables

The procedures of data adjustment for covariates used in this assessment are described here. Time-series of Hg in blue mussels (Iceland and Norway) involved the collection of animals in specific length / weight classes; additionally, the limited size ranges available made it difficult to establish relationships between the covariates and Hg concentrations. For these reasons, no adjustment for biological covariates in these samples was performed. Furthermore, the trend analyses were carried out using dry weight concentrations to address potential problems related to variable water content.

It is well known that Hg concentrations in fish muscle (freshwater and marine) are related to the size of the fish (AMAP, 1998). Therefore, most of the fish trend analyses were based on length-adjusted data using log-transformed length as a covariate (one case used log-transformed weight, and one square-root transformed age). In a few cases, fish were divided into two length intervals. Stable isotope data were available for a landlocked Arctic char (*Salvelinus alpinus*) population, which was separated into two well-defined morphs with different feeding behavior according to the δ^{15}N value. Trend analyses were carried out separately for the two morphs.

For seabird eggs, no adjustment was applied. The Canadian Hg time series for seabird eggs were based on dry weight concentrations while the Faroe Islands time series were based on wet weight concentrations.

Terrestrial mammal Hg data were available from Sweden and Canada. The Swedish monitoring program is based on

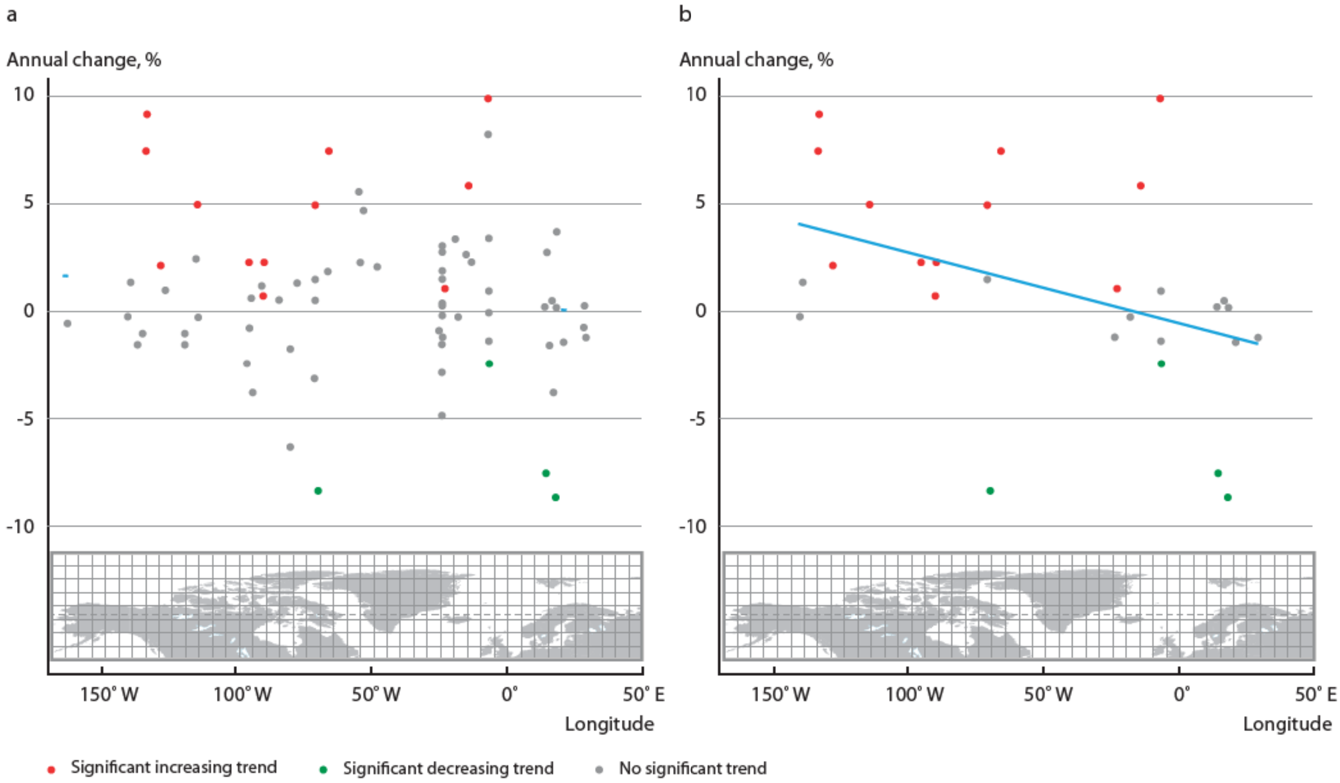


Figure 5.3. Annual change estimated (a) for all 83 analyzed time series versus longitude, and (b) for only those time series (n = 28) showing linear statistical significance or which meet the data adequacy requirements. The blue line indicated a significant linear regression ($R^2 = 0.2$, $p = 0.02$).

reindeer of specific age and gender, and thus no adjustment for covariates was done. The Canadian caribou kidney time series were separated by sex, and the square root of age was applied as a covariate according to the guidelines of the data providers.

It is well known that Hg in marine mammals is positively correlated to animal age (AMAP, 1998). In some cases, data were separated into age groups (e.g., juveniles and adults) and by sex, and no further adjustment for covariates was done. In other datasets, square-root transformed ages were applied as a covariate. In the case of Canadian beluga, data were separated into two time series above and below the overall mean age, and in addition, square-root transformed age was applied as a covariate. This procedure was chosen because the age range of beluga was very large (0 to 84 years old), and the assumption that there was a linear relationship between square-root transformed age and log-transformed Hg concentration over the total age span created unreliable results. In the case of polar bears from Alaska, only male adults and sub-adults were included (i.e., cubs were excluded) and the data were analyzed with square-root transformed age as a covariate. In one case, no covariate adjustment was applied due to missing data.

5.3.3.5. Adequacy of time series

In the Stockholm Workshop Report (AMAP, 2007), a concept termed 'Adequacy' was used which is closely related to, yet distinct from, the 'statistical power' of datasets. 'Adequacy' (expressed as a percentage or a proportion) is defined as the number of actual monitoring years in a time series divided by the number of years of sampling required to detect a 5% annual change in Hg concentration, with a significance level of $p < 0.05$ and 80% statistical power (see Bignert et al., 2004). Adequacy should not be confused with the concept of power, which is only one component of adequacy, albeit a very important one. Statistical power is defined as 1.0 minus the probability of a false positive result (β or Type II Error) (Sokal and Rohlf, 1981); it is affected by factors such as the number of samples taken each year, variability in Hg concentrations within and between years of sampling, and the desired detectable rate of change in Hg levels. The desired power level can also be pre-set (in these analyses, a conventional value of 80% was used). When this is done, other parameters of practical interest can be calculated, such as the number of years of data required to statistically detect a 5% annual change in Hg concentration (i.e., used in this measure of adequacy). Time series which have fewer than the minimum number of years required to detect a 5% annual rate of change are classed as 'inadequate', while those with years of data equal to or more than the minimum required are 'adequate'.

5.3.3.6. Annual change

The annual change in average Hg concentration ranged from -8.6% to 10% across all time series, with a median value of 0.5%. The annual change showed no apparent trend with longitude (linear regression, $p = 0.20$) (Figure 5.3a) or latitude (linear regression, $p = 0.65$) (Figure 5.4a). Considering only those datasets with significant linear trends, or which met the data adequacy requirements, the median annual rate of change was 2.4%. The annual rate of change decreased significantly from west to east ($p = 0.02$) (Figure 5.3b) but showed no trend with latitude ($p = 0.54$) (Figure 5.4b).

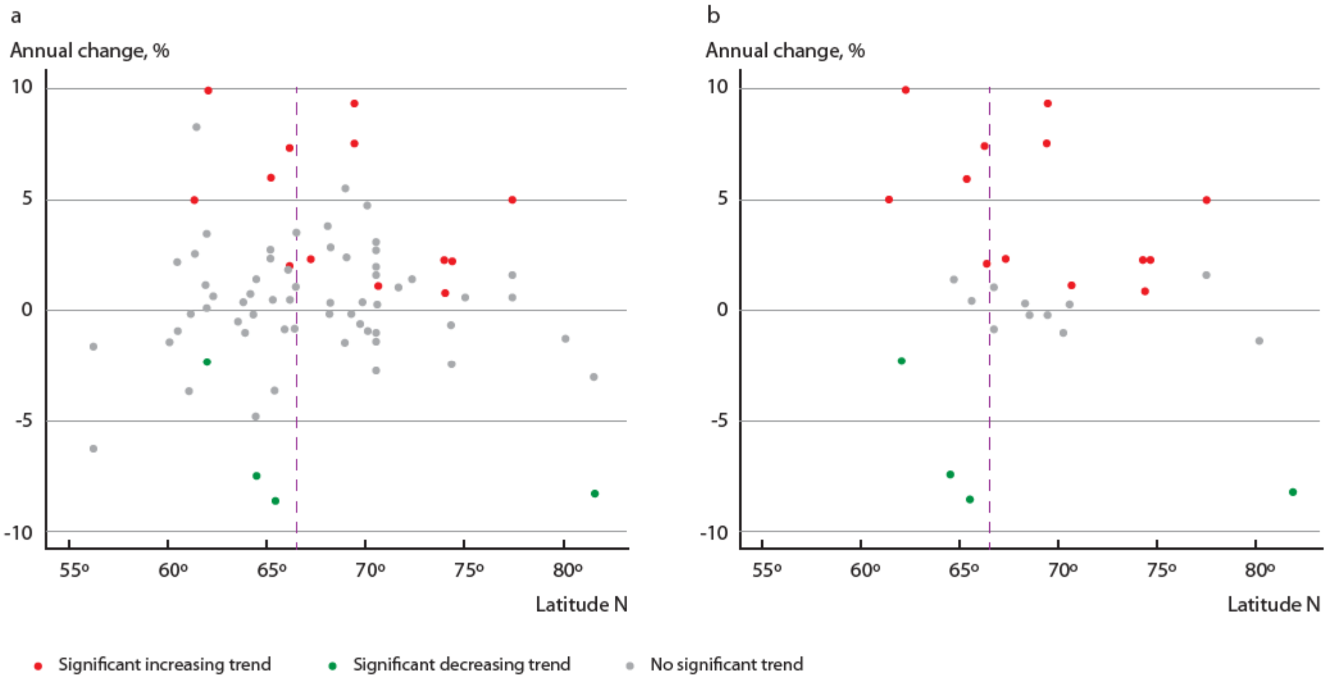


Figure 5.4. Annual change estimated (a) for all 83 analyzed time series versus latitude, and (b) for only those time trends (n = 28) which show linear statistical significance or which meet the adequacy requirements. The Arctic Circle is indicated by the dashed line.

One of the outputs of the temporal trend analysis is an estimation of the statistically detectable annual trend, assuming a sampling period of ten years, a significance level of 0.05 and a statistical power of 80%. The minimum detectable annual change ranged from 1.9% to 100% with a median value of 11% (Figure 5.5). Based on the datasets available for the previous AMAP Hg assessment (the 'Phase II' Assessment), and using the same criteria as above, Bignert et al. (2004) determined that the minimum detectable annual change in datasets available at that time ranged from 2.2% to 57% with a median value of 12%.

The utility of different taxa or biotic media for monitoring Hg can also be evaluated using the magnitude of the residual standard deviation from the trend; i.e., linear or non-linear. The

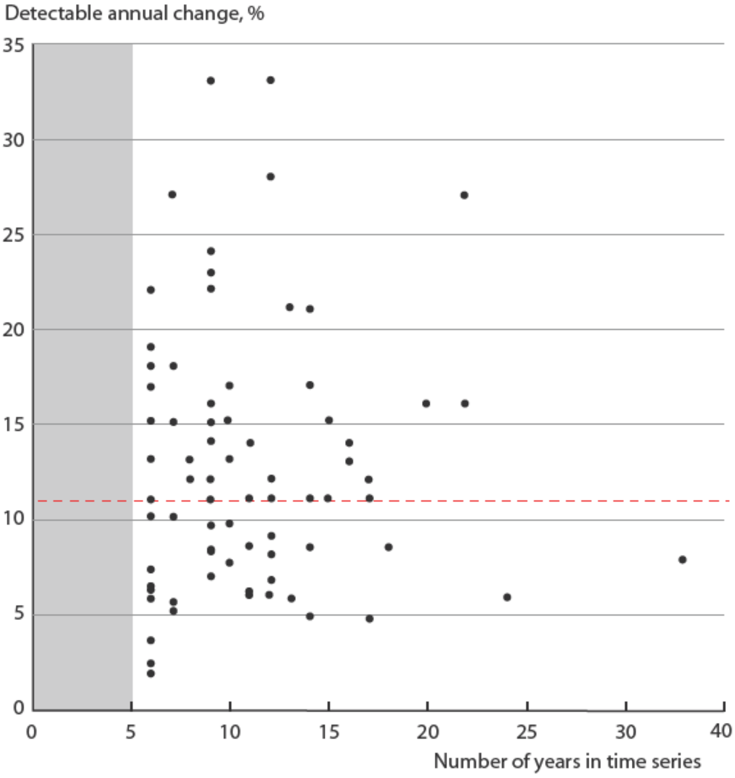


Figure 5.5. Statistically detectable annual change in Hg concentrations (assuming a sampling period of ten years, α = 0.05, power = 80%) plotted against number of years in available time series. Time series with less than six years of data were not included. Horizontal dashed line indicates the median detectable change.

lower the residual standard deviation, the more powerful the dataset. Seabird eggs had the lowest residual standard deviations (median = 0.17), while terrestrial and marine mammals had the highest (median = 0.41 and 0.40, respectively). The other groups fell within this range.

5.3.3.7. Evaluation of temporal trends

A summary of results from analysis of all 83 datasets included in this assessment is found in Table 5.3 and a summary of the statistical outputs resulting from analysis of the 45 time series with adequate data or which were statistically significant is presented in Table 5.4. It is outside the scope of this assessment to evaluate the individual time series; these are considered in far greater detail in related scientific papers and publications. The aim of this meta-analysis is to deduce possible general geographical patterns in Hg trends within the Arctic, or any similarities or dissimilarities between freshwater, terrestrial and marine ecosystems. The results have been categorized into four groups: significant increasing or decreasing trends (log-linear

Table 5.3. Results of trend analysis of mercury time series for all datasets (n = 83) analyzed using the PIA program.

Ecosystem/ Country/Species[a]	Tissue	Location	Range of years	No. years	Temporal trend	Annual change, %	Adequacy of data
Terrestrial Animals							
Sweden							
Reindeer	muscle	Abiskojaura	1983-2005	22	Significant non-linear trend	3.8	0.81
Reindeer	liver	Ammarnäs	1996-2005	10	Significant decreasing trend	-8.6	0.50
Reindeer	liver	Abiskojaura	1983-2005	22	No trend	0.2	1.05
Reindeer	muscle	Ammarnäs	1996-2005	10	Significant non-linear trend	-3.8	0.48
Canada							
Caribou (female)	kidney	Yukon	1994-2007	14	No trend	-0.3	1.00
Caribou (male)	kidney	Yukon	1994-2007	12	No trend	1.3	1.00
Freshwater Fish							
Sweden							
Pike	muscle	Storvindeln	1968-2006	33	Significant non-linear trend	0.4	2.54
Arctic char, landlocked	muscle	Abiskojaura	1981-2006	24	Significant non-linear trend	-0.3	2.18
Faroe Islands							
Arctic char, landlocked	muscle	Á Mýrunum	2000-2007	6	Significant increasing trend	10.0	0.50
Greenland							
Arctic char, landlocked	muscle	Isortoq	1994-2008	6	Significant non-linear trend	2.1	0.55
Canada							
Arctic char, landlocked	muscle	Char Lake	2000-2008	6	Significant increasing trend	2.3	0.86
Arctic char, landlocked (small morph)	muscle	Lake Hazen	1990-2008	9	Significant decreasing trend	-4.4	0.82
Arctic char, landlocked (large morph)	muscle	Lake Hazen	1990-2008	9	No trend	1.3	0.50
Arctic char, landlocked	muscle	Amituk Lake	1989-2008	9	Significant non-linear trend	0.5	0.47
Arctic char, landlocked	muscle	Resolute Lake	1997-2008	11	Significant non-linear trend	-0.7	0.69
Burbot	muscle	West Basin	1996-2007	9	Significant increasing trend	5.1	0.75
Burbot	liver	Fort Good Hope	1985-2008	12	Significant increasing trend	2.2	0.86
Burbot	muscle	Fort Good Hope	1985-2008	13	Significant increasing trend	2.4	1.18
Lake trout	muscle	Kusawa	1993-2008	9	No trend	-1.6	0.45
Lake trout	muscle	West Basin- Hay River	1999-2007	7	No trend	2.5	0.64
Lake trout	muscle	East Arm Great Slave	1995-2007	9	No trend	-0.3	0.64
Lake trout	muscle	Laberge	1993-2008	11	No trend	-1.0	0.79
Marine Invertebrates							
Norway							
Blue mussels	soft tissue	Brashavn	1998-2007	9	No trend	0.2	0.64
Blue mussels	soft tissue	Fensneset Grytoya	1994-2007	6	No trend	-1.6	0.67
Blue mussels	soft tissue	Skallneset	1994-2007	14	No trend	-1.0	1.40
Blue mussels	soft tissue	Elenheimsundet	1994-2007	6	Significant non-linear trend	0.3	1.00
Iceland							
Blue mussels	soft tissue	Mjoifjordur Hofs	1995-2007	12	Significant increasing trend	6.1	0.75
Blue mussels	soft tissue	Mjoifjordur Dalatangi	1997-2007	9	No trend	2.4	0.56
Blue mussels	soft tissue	Ulfsa Skutulsfjordur	1997-2007	9	No trend	0.4	0.60
Blue mussels	soft tissue	Dvergasteinn Alftafjordur	1996-2006	10	No trend	-0.9	0.77
Blue mussels	soft tissue	Hvassahraun	1992-2007	15	No trend	0.4	0.79
Blue mussels	soft tissue	Eyri Hvalfjordur	1992-2007	16	No trend	0.7	0.84
Blue mussels	soft tissue	Grimsey	1992-2007	14	No trend	3.5	0.88
Blue mussels	soft tissue	Hvalstod Hvalfjordur	1992-2007	16	No trend	-0.3	0.89
Blue mussels	soft tissue	Mjoifjordur head	1996-2007	11	No trend	2.6	0.92
Blue mussels	soft tissue	Straumur Straumsvik	1992-2007	15	Significant non-linear trend	-1.0	0.75
Blue mussels	soft tissue	Hvitanes Hvalfjordur	1992-2007	16	Significant non-linear trend	0.8	0.89

Table 5.3 (continued).

Ecosystem/ Country/Species[a]	Tissue	Location	Range of years	No. years	Temporal trend	Annual change, %	Adequacy of data
Marine Fish							
Norway							
Plaice	muscle	Lille Mola/Husholmen	1993-2006	11	No trend	2.8	0.58
Plaice	muscle	Skogerøy	1997-2007	9	No trend	-1.1	0.75
Atlantic cod	muscle	Lille Mola/bjørnerøya	1993-2006	12	No trend	0.3	0.71
Atlantic cod	muscle	Varangerfjorden	1994-2007	14	Significant non-linear trend	-0.8	0.58
Faroe Islands							
Atlantic cod (≤73 cm)	muscle	Faroe Islands	1979-2007	20	Significant decreasing trend	-2.4	1.00
Iceland							
Atlantic cod	muscle	Southeast	1990-2003	7	Significant decreasing trend	-7.4	0.64
Atlantic cod	muscle	Southwest	1990-2001	6	Significant non-linear trend	-4.9	0.33
Atlantic cod	muscle	Northeast	1990-2007	17	No trend	1.0	1.00
Atlantic cod	muscle	Northwest	1990-2007	17	Significant non-linear trend	-0.9	1.00
Greenland							
Sculpin (<27 cm)	liver	Qeqertarsuaq	1994-2006	7	No trend	4.8	0.26
Sculpin (>27 cm)	liver	Qeqertarsuaq	1994-2008	9	No trend	5.5	0.38
Sculpin	liver	Ittoqqortoormiit	1995-2008	6	Significant non-linear trend	2.7	0.46
Marine Mammals							
Norway							
Polar bear (>2 yrs)	hair	Svalbard	1995-2008	12	No trend	-1.3	1.09
Faroe Islands							
Pilot whale (adult male)	muscle	Faroe Islands	1979-2006	13	No trend	1.0	0.54
Pilot whale (immature)	muscle	Faroe Islands	1979-2007	14	No trend	3.4	0.67
Pilot whale (adult female)	muscle	Faroe Islands	1979-2007	15	No trend	0.1	0.94
Greenland							
Polar bear (>2 yrs)	hair	Ittoqqortoormiit	1984-2008	17	Significant increasing trend	1.2	1.70
Polar bear (adult)	kidney	Ittoqqortoormiit	1984-2001	6	No trend	-1.4	0.25
Polar bear (juvenile)	kidney	Ittoqqortoormiit	1983-2001	9	No trend	-2.8	0.47
Polar bear (juvenile)	liver	Ittoqqortoormiit	1983-2006	10	No trend	1.8	0.67
Polar bear (>2 yrs)	hair	Avanersuaq	1987-2006	18	No trend	1.6	1.29
Polar bear (adult)	liver	Ittoqqortoormiit	1984-2006	8	Significant non-linear trend	1.8	0.44
Ringed seal (juvenile)	liver	Avanersuaq	1984-2008	7	Significant increasing trend	5.0	0.32
Ringed seal (juvenile)	liver	Ittoqqortoormiit	1986-2008	9	No trend	2.9	0.36
Ringed seal (adult)	liver	Ittoqqortoormiit	1986-2008	9	No trend	1.4	0.53
Ringed seal (juvenile)	liver	Qeqertarsuaq	1994-2008	9	Significant non-linear trend	2.3	0.53
Walrus (9-16 yrs)	liver	Avanersuaq	1977-2003	7	No trend	0.6	0.37
Canada							
Beluga (≤30 yrs)	liver	Hendrickson Island	1981-2006	12	Significant increasing trend	9.4	0.43
Beluga (>30 yrs)	liver	Hendrickson Island	1981-2006	12	Significant increasing trend	7.7	0.40
Beluga (≤20 yrs)	liver	Pangnirtung	1984-2002	10	Significant increasing trend	7.5	0.53
Beluga (>20 yrs)	liver	Pangnirtung	1984-2002	9	No trend	1.9	0.36
Beluga (≤27 yrs)	liver	Sanikiluaq	1994-2004	6	Significant non-linear trend	-1.7	0.29
Beluga (>27 yrs)	liver	Sanikiluaq	1994-2004	6	Significant non-linear trend	-6.3	0.32
Narwhal	liver	Pond Inlet	1978-2004	8	No trend	1.3	0.44
Ringed seal	liver	Arviat	1992-2007	6	No trend	-3.7	0.26
Ringed seal	liver	Sachs Harbour	1987-2007	6	No trend	1.0	0.38
Ringed seal	muscle	Holman	1973-2007	10	No trend	-1.1	0.56
Ringed seal	liver	Holman	1973-2007	12	No trend	-1.3	0.86
Ringed seal	liver	Resolute	1993-2007	6	Significant non-linear trend	-2.5	0.29
Alaska							
Polar bear (male)	liver	Chucki/Bering Sea	1987-2007	13	No trend	-0.6	0.22
Marine Birds							
Faroe Islands							
Black guillemot	egg	Koltur	1999-2006	6	No trend	-1.3	0.50
Black guillemot	egg	Skuvoy	1999-2006	6	Significant non-linear trend	8.3	0.38
Canada							
Black-legged kittiwake	egg	Prince Leopold Island	1975-2008	7	No trend	1.0	0.44
Northern fulmar	egg	Prince Leopold Island	1975-2008	11	Significant increasing trend	0.9	1.00
Thick-billed murre	egg	Prince Leopold Island	1975-2008	12	Significant increasing trend	2.4	1.00
Thick-billed murre	egg	Coats Island	1993-2008	7	Significant non-linear trend	0.5	0.64

[a] Caribou, reindeer (*Rangifer tarandus*); pike (*Esox lucius*); Arctic char (*Salvelinus alpinus*); burbot (*Lota lota*); lake trout (*Salvelinus namaycush*); blue mussels (*Mytilus edulis*); plaice (*Pleuronectes platessa*); Atlantic cod (*Gadus morhua*); sculpin (*Myoxocephalus scorpius*); ringed seal (*Phoca hispida*); walrus (*Odobenus rosmarus*); pilot whale (*Globicephala melas*); narwhal (*Monodon monocerus*); beluga (*Delphinapterus leucas*); polar bear (*Ursus maritimus*); black guillemot (*Cepphus grylle*); black-legged kittiwake (*Rissa tridactyla*); northern fulmar (*Fulmarus glacialis*); thick-billed murre (*Uria lomvia*).

Table 5.4. Summary of statistical results for all 83 mercury time series analyzed and for the 45 time series showing significantly increasing/decreasing/non-linear trends or with adequate datasets.

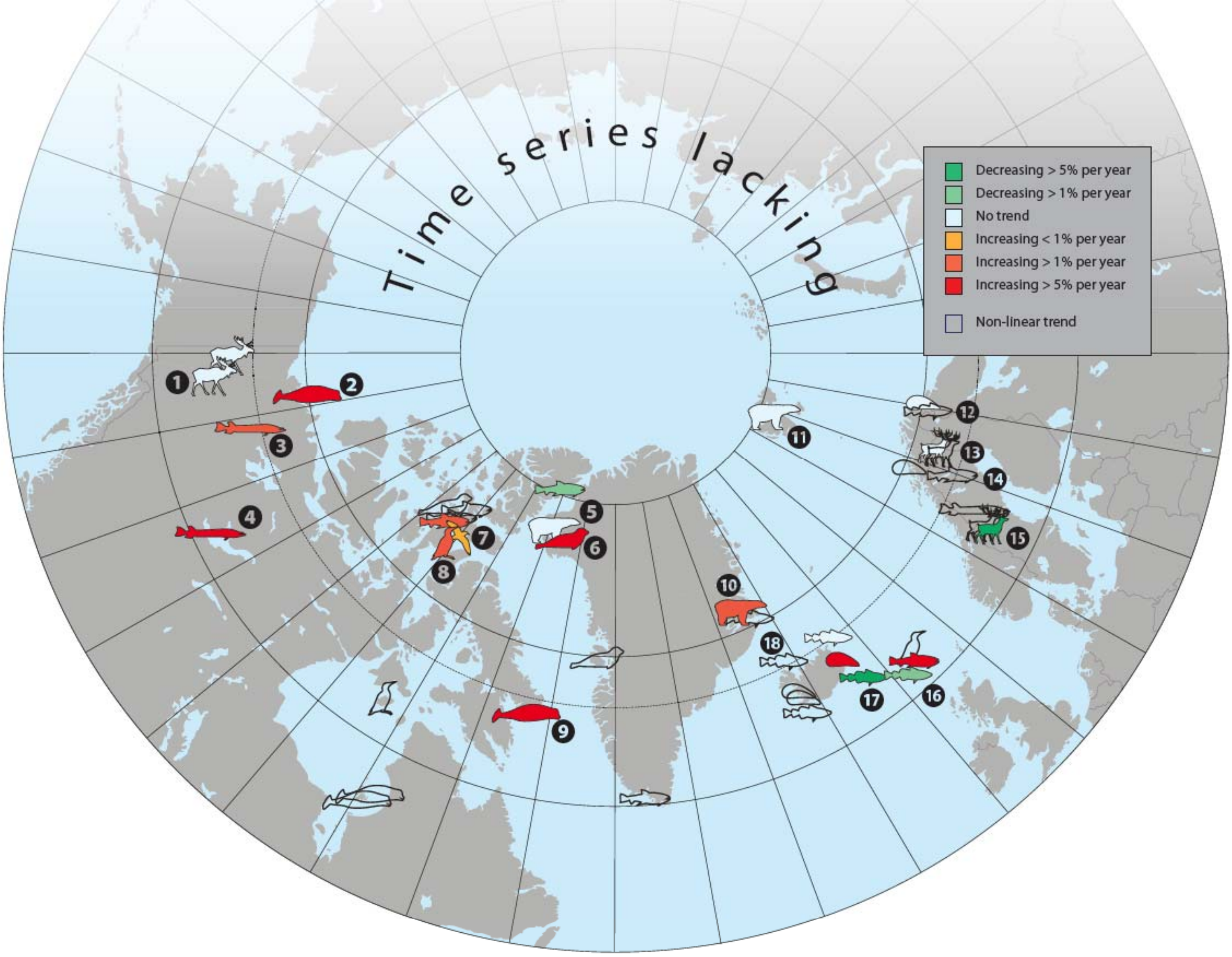
Significant increasing trend	Significant decreasing trend	No trend	Significant non-linear trend	Median annual percentage change	Median lowest detectable trend (%) in 10 years with α = 5% and a power of 80%	Median power (%) in 10 years to detect 5% with α = 5% and a power of 80%
All 83 time series						
13 (16%)	4 (5%)	45 (54%)	21 (25%)	0.6	11.0	24
45 times series with increasing/decreasing/non-linear trend or adequate datasets						
13 (29%)	4 (9%)	7 (16%)	21 (47%)	0.8	11.0	27

trend component is significant), significant non-linear trends (non-linear trend component is significant) and no trend.

Of the 83 time-series analyzed, 13 (16%) showed a significantly ($p < 0.05$) increasing linear trend and one of these also had a significant non-linear trend component within the overall trend. Two of the significant datasets were from the same animals but different tissues. Four time series (5%) showed a significantly decreasing linear trend and one of these also had a significant non-linear component. Twenty-one time series showed a significant non-linear trend. If the required α level of significance is weakened from 5% to 10%, then the number of time series showing an increasing trend would increase slightly, from 13 to 16, and the number showing a decreasing trend would increase from 4 to 5.

The average annual increases for the 13 significantly increasing datasets ranged between 0.9% and 10% (median 5.0%). Breaking the results down by country, nine significantly increasing time series were found for Canada (30% of the Canadian datasets), two for Greenland (13%), and one each for the Faroe Islands (14%) and Iceland (7%) (Figure 5.6, see next page spread). Broken down by taxonomic grouping, increasing trends were found in marine invertebrates (1; 8% of the significantly increasing datasets), seabirds (2; 15%), marine mammals (5; 38%) and freshwater fish (5; 38%). Only four (5%) of the time series showed a significantly decreasing trend and one of these also had a significant non-linear trend component. The annual decreases ranged between -2.4% and -8.6% (median -5.9%). Two of these were for Atlantic cod (*Gadus morhua*) from the Faroe Islands and Iceland, one was for Arctic char from Canada, and one was for reindeer (*Rangifer tarandus*) from Sweden. None of the Arctic marine mammal or seabird datasets showed a significantly declining trend. The remaining 66 Hg time series showed either no trend or a significant non-linear trend component. The overall median annual rate of change of these time series was 0.4%.

These overall results are, as expected, similar to those obtained during the Stockholm Workshop in 2006 (AMAP, 2007), particularly as the majority of the significant time series are identical except for a few additional years of data in some cases.


Time series lacking
🟩 Decreasing > 5% per year
🟩 Decreasing > 1% per year
☐ No trend
🟧 Increasing < 1% per year
🟥 Increasing > 1% per year
🟥 Increasing > 5% per year
☐ Non-linear trend
1
2
3
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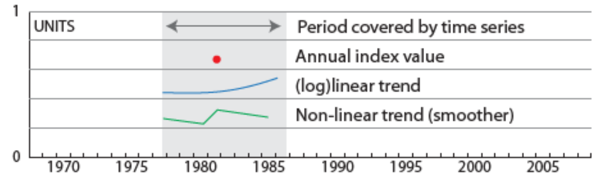
1
0
UNITS
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Annual index value
(log)linear trend
Non-linear trend (smoother)
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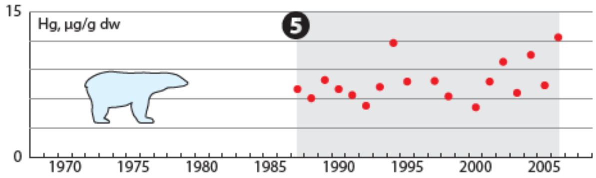
1975
1980
1985
1990
1995
2000
2005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Hg, µg/g dw
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1970
1975
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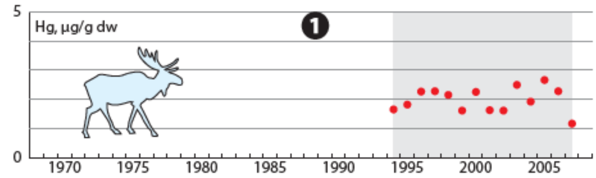
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Hg, µg/g dw
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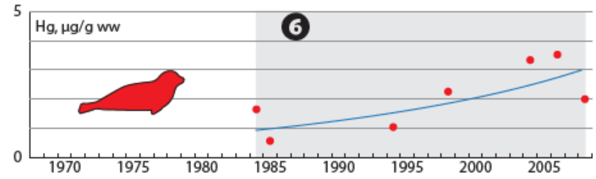
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Hg, µg/g ww
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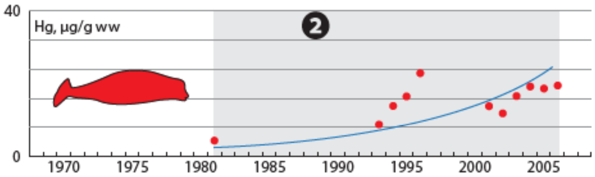
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1975
1980
1985
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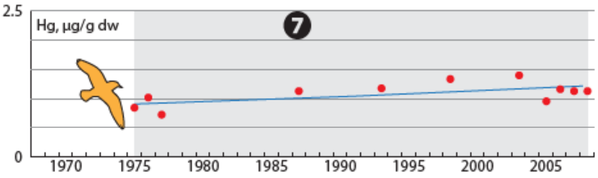
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Hg, µg/g dw
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1970
1975
1980
1985
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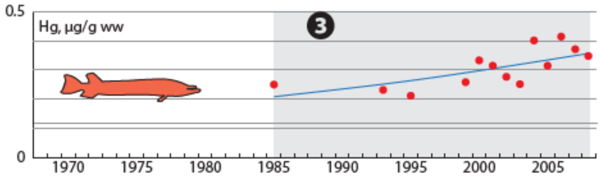
1995
2000
2005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3
1970
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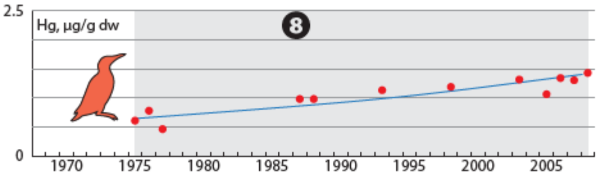
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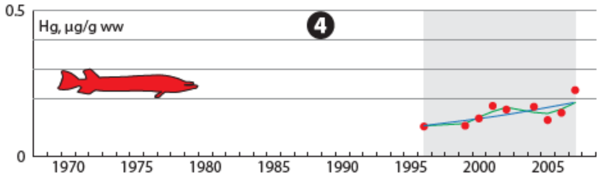
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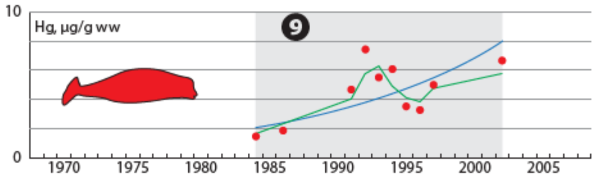
10
0
Hg, µg/g ww
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1970
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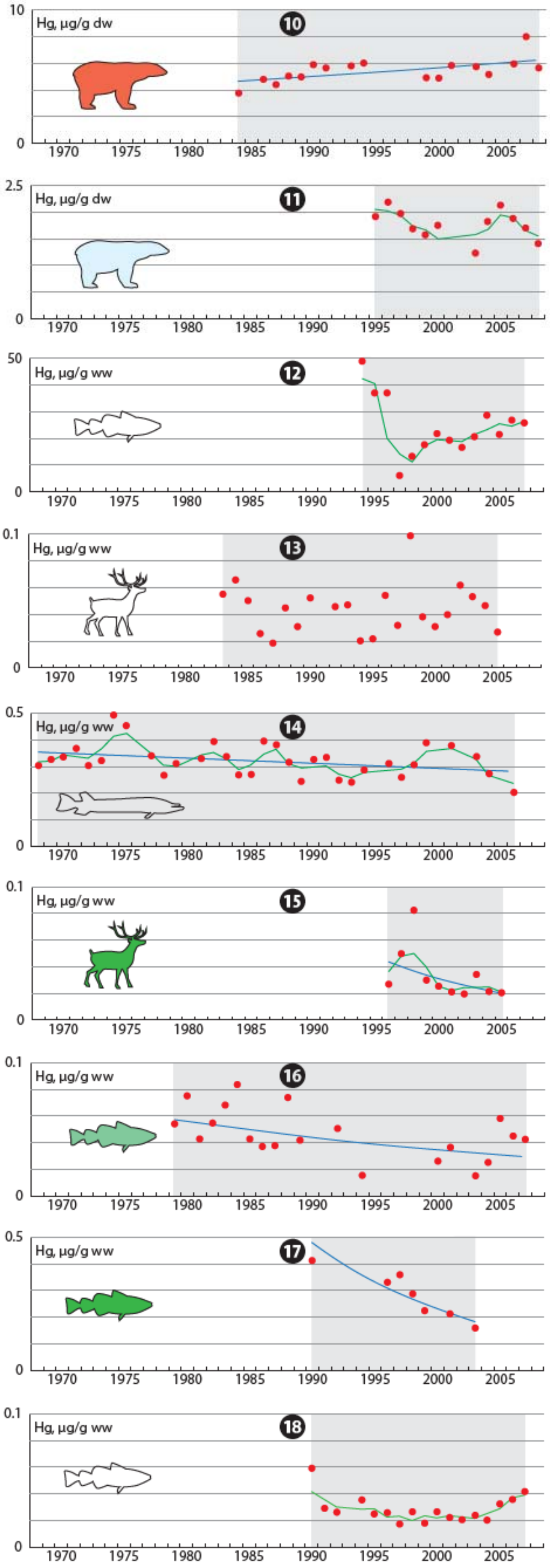

Figure 5.6. Summary of trends which are statistically significant or meet the data adequacy requirements, with details of trends for selected time series from Canada and West Greenland (1-9) and East Greenland and the European Arctic (10-18).

5.3.3.8. Evaluation of adequacy of time series to detect trends

At the Stockholm workshop in 2006, only five time series were found to be 'adequate'. Four of those are also classified as 'adequate' in this update, whereas the fifth (Finnish freshwater fish influenced by construction of hydroelectric reservoirs) is not included in this update. Here, a total of 16 (19%) of the 83 time series were classified as 'adequate' (Figure 5.7). These included seven datasets showing no trend, four showing a significant non-linear trend, four showing a significantly increasing trend, and one showing a significantly decreasing trend (see Table 5.3). It was predicted in 2006 that a larger number of time series would be classified as 'adequate' in this updated assessment because of the additional years of data. However, in some cases, adequacy may have declined or remained constant, largely attributed to increased between-year variability in the extended time series. Overall, however, the results from this assessment represent an increase from 10% to 19% of datasets deemed 'adequate' relative to the previous AMAP Hg assessment (Bignert et al., 2004).

If an additional five years of data were to be added to the time series, it is predicted that 44 of the 83 time series would meet the criteria for adequacy defined here (as indicated by the broken line in Figure 5.7). However, this assumes that other factors, such as between-year variability, remain the same.

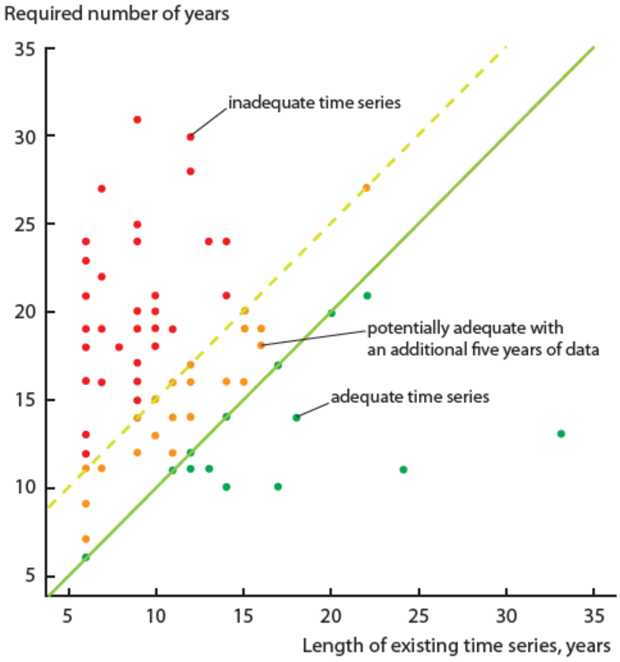

Figure 5.7. Statistical adequacy of the datasets analyzed. Points at or below the solid line have an adequacy of ≥ 1 and points above the solid line have an adequacy of < 1. The dashed line predicts the situation if the time series are extended with an additional five years of data (assuming unchanged between-year variation).

5.3.4. Comparison of meta-analysis results with mercury trends published in the recent literature

In both the published literature (Table 5.2) and this meta-analysis (Table 5.3), Hg concentrations in terrestrial biota showed either no trend (this includes significant non-linear trends) or a decreasing trend. Of the time series for freshwater fish, none of the published time series showed statistically significant trends (Table 5.2). However, of the 16 time series for freshwater fish included in the meta-analysis, five of the time series (31%) showed significant increases in Hg concentration (although two of those were for different tissues from the same fish), and one (6%) showed a significant decrease. The highest number of increasing trends in Hg concentration was seen in marine biota. Of the 29 datasets reported in the literature for the marine environment (Table 5.2), 31% suggested increasing trends while the other 69% showed either no trend or decreasing concentrations. Of the 61 time series for marine biota included in the meta-analysis, a similar percentage compared to those in the literature (13% *vs* 10%) showed significantly increasing trends.

Since there was a geographical bias in the time series represented, it is difficult to evaluate with great confidence the meaning of the overall spatial / circumpolar distribution of increasing or decreasing trends. However, the seabird egg datasets provide an opportunity to do so since there are data available from Canada, the Faroe Islands and Norway (Table 5.5). Three of the five Canadian egg datasets showed increasing Hg trends and one of two datasets from the Faroe Islands showed a significant non-linear trend which included a recently increasing trend component, whereas none of the three datasets from Norway nor the remaining datasets from Canada or the Faroe Islands showed any linear trend. Although few in number, these egg datasets support the general pattern of higher numbers of time series showing an increasing trend in Canada and Greenland compared with northern Europe (see Figures. 5.3 and 5.6).

Factors that may be driving the Hg trends observed, both spatially and across ecosystems, are discussed in Section 5.4.

5.4. Why are mercury levels in Arctic biota increasing or decreasing?

It becomes clear from Sections 5.2 and 5.3 that, while Hg concentrations in some Arctic populations have increased more than 10-fold on average compared to pre-industrial times, no consistent trend can be generalized across tissues and species of the entire circumpolar Arctic for the past 30 years or so. There was, however, a clear west-to-east gradient in the occurrence of recent increasing Hg trends, with larger numbers and a higher proportion of biotic datasets in the Canadian and West Greenland region of the Arctic showing significant increases than in the North Atlantic Arctic. Most of the increasing datasets were for marine-based species, especially marine mammals.

Since the potential biases of important biological covariates such as age and (in some cases) trophic change have been accounted for in the statistical analyses, the biotic Hg increases and decreases, or lack of change, presumably approximate real variations (or lack thereof) in Hg levels in the animals' environments and food webs. A clearer picture of the variability and consistencies in Hg trends in Arctic biota over time spans of recent centuries and decades is now emerging. This section discusses the possible drivers of these trends, grouped under the general categories of sources (anthropogenic and natural emissions) and environmental processes (mainly but not exclusively climate-driven).

5.4.1. Sources- vs. processes-driven mercury bioaccumulation in the Arctic

As detailed in Section 5.2, the significant increase in Hg concentrations in present-day Arctic animals compared to their pre-industrial counterparts is most likely to have been driven primarily by the increase in anthropogenic Hg influx to the Arctic ecosystems. This is supported by the significant increase in anthropogenic Hg depositional flux recorded in Arctic lake sediments and peat bogs (Bindler et al., 2001b; Shotyk et al., 2003, 2005a; Fitzgerald et al., 2005; Lindeberg et al., 2006, 2007; Outridge et al., 2007; Landers et al., 2008; Muir et al.,

Table 5.5. Mercury trends in seabird eggs.

Location/species	Published trend[a]	Meta-analysis trend[b]
Northern Norway		
Herring gull	No trend	n/a[c]
Atlantic puffin	No trend	n/a
Black-legged kittiwake	No trend	n/a
Faroe Islands (Koltur)		
Black guillemot	No trend	No trend
Faroe Island (Skúvoy)		
Black guillemot	Increasing trend since 2000	Significant non-linear trend
Canada (Prince Leopold Island)		
Black-legged kittiwake	No trend	No trend
Northern fulmar	Significant increasing trend	Significant increasing trend
Thick-billed murre	Significant increasing trend	Significant increasing trend
Canada (Coats Island)		
Thick-billed murre	No published trend	Significant non-linear trend
Canada (Seymour Island)		
Ivory gull	Non-significant increasing trend	n/a

[a] See Table 5.2.; [b] see Table 5.3.; [c] n/a not analyzed because dataset did not meet criteria for meta-analysis.

2009), by global Hg emission inventories (see Chapter 2), and by evidence of significant amounts of Hg contamination in the world's oceans (Sunderland and Mason, 2007) (see Chapter 3).

However, this anthropogenic source-driven scenario appears not to be the sole explanation for biotic trends in recent decades (Wang et al., 2010). Although there has been a regional shift in Hg emission to the atmosphere, the global size of the atmospheric pool of Hg has been either stable since the mid-1970s or may have decreased (Pacyna et al., 2003; Lindberg et al., 2007; Fain et al., 2009b; Streets et al., 2009). *In situ*, high resolution measurements at Alert, Canada, have shown no significant trend in the annual average concentration of gaseous elemental Hg (GEM) in High Arctic air since 1995 (Steffen et al., 2005). Prior to 1995, a reconstructed GEM history from Greenland Summit firn snow showed a decline in atmospheric Hg levels from the early 1970s to early 1990s (Faïn et al., 2009b). Similarly, analysis of particulate Hg in archived air filters collected from Resolute, Canada, has shown a significant declining trend for summer/autumn during the period 1974 to 2000 (Li et al., 2009). These constant or declining trends in Arctic atmospheric Hg do not explain the continued recent increasing trends of Hg in Arctic wildlife (see Section 5.3). As some of the case studies in Section 5.4.2 illustrate, the lack of a direct atmosphere-biota 'dose-response' relationship for Hg in the Arctic has precipitated several recent studies on alternative drivers for Hg bioaccumulation in the Arctic. These alternative drivers include additional Hg sources, as well as cryospheric, biogeochemical and ecological processes that control the movement of Hg from the abiotic environment to Arctic animals (see Chapter 3).

5.4.1.1. Mercury sources other than atmospheric deposition

Atmospheric deposition is often the dominant source of Hg to Arctic lakes. However, migratory seabirds have also been shown to contribute Hg via their guano locally in a few small, isolated Arctic ponds that are visited by a large number of seabirds (Blais et al., 2005, 2007). Transport of Hg to the Arctic marine environment also occurs via riverine discharge, thawing permafrost, coastal erosion, and oceanic circulation (Macdonald et al., 2005; Leitch et al., 2007; Outridge et al., 2008). Recent studies on the mass budget of Hg in the Arctic Ocean (Outridge et al., 2008) and Hudson Bay (Hare et al., 2008) suggested that the atmospheric influx of Hg to the Arctic Ocean may have been significantly overestimated and that the contribution of atmospherically deposited Hg needs to be considered within the context of the total Hg flux including terrestrial and marine sources.

5.4.1.2. Cryospheric processes that control the timing, speciation, and net flux of atmospheric mercury deposition

Although the annual average concentration of GEM has stayed essentially unchanged since at least 1995 (Steffen et al., 2005), the discovery of the tropospheric atmospheric Hg depletion events (AMDEs; see Chapter 2) during polar sunrise (Schroeder et al., 1998) raises the question of the significance of photochemically oxidized Hg during AMDEs in Hg uptake in Arctic ecosystems. This is, however, complicated by the fact that most of the surfaces of Arctic marine and coastal systems are frozen at the time of AMDEs. A time lag thus exists between when the Hg is deposited onto the frozen surface and when it is transported to the underlying aquatic or terrestrial ecosystems, during which post-depositional changes in concentration and speciation are known to occur. Therefore, the net contribution of atmospherically transported Hg to the Hg flux in the Arctic ecosystem is highly modified by cryospheric conditions, which is reflected in the large uncertainty in the estimate of atmospheric Hg flux to the Arctic Ocean (8 to 300 t/y; Outridge et al., 2008).

5.4.1.3. Biogeochemical processes that control the methylation of mercury

Although uptake of inorganic Hg is possible, it has been well established that Hg uptake and accumulation in Arctic aquatic animals occurs mainly in the form of MeHg (see Chapters 3 and 6). No mass budget is currently available for MeHg in the Arctic Ocean, but the majority of MeHg is believed to be formed by methylation of the existing pool of inorganic Hg in the aquatic environment. Mercury methylation can occur in deep sediments (Kraepiel et al., 2003; Hammerschmidt and Fitzgerald, 2005) as well as in the water column (Kirk et al., 2008; Cossa et al., 2009; Sunderland et al., 2009) of the ocean, and is controlled by biogeochemical processes that affect the influx of inorganic Hg and organic matter, and the presence and activity of Hg methylators. Any change in these processes will thus change the locality and pool of MeHg available for uptake by Arctic ecosystems.

5.4.1.4. Biological and ecological processes that control the transport of methylmercury in food webs

Because MeHg biomagnifies in food webs, any change in the foraging behavior (e.g., habitat use, diet preferences) of an Arctic animal, and in the structure and dynamics of Arctic food webs in general, would result in a change in the Hg concentration in the animal.

Therefore, to better understand the temporal and spatial variation in Hg concentrations in Arctic animals, changes both in Hg sources and post-depositional Hg processes need to be considered (Wang et al., 2010). This requires an understanding of changes in anthropogenic activities and the effects of climate change which has occurred in the Arctic over recent decades (Post et al., 2009).

5.4.2. Case studies

This section presents eight case studies as examples for discussing the relative importance of different drivers that may be responsible for the temporal variations in Hg concentration in Arctic animals. These case studies demonstrate that the interactions between the various factors are complex, with atmospheric Hg deposition, and environmental and ecological processes, in some cases coupled with climate change, playing a role, as does the biology and behavior of the species.

Case Study 1. Landlocked Arctic char from Lake Hazen

Muscle tissue of landlocked Arctic char (*Salvenlius alpinus*) from Lake Hazen was used to investigate temporal trends in Hg concentration using recent data in addition to data published by Gantner et al. (2009). Lake Hazen is the largest lake in the Canadian High Arctic and Arctic char from this lake have been relatively well studied compared to most other lakes in the Canadian Arctic Archipelago. Up to three different morphotypes of Arctic char are present in Lake Hazen (Reist et al., 1995; Guiguer et al., 2002). All the char are considered to be non-anadromous (resident) (Babaluk et al., 1997). Archived and newly collected char consisting of two morphotypes (large piscivores and smaller insectivores) were analyzed by Gantner et al. (2009). The morphotypes differed in Hg concentration, and the resulting variability was accounted for by adjusting the Hg concentrations using their nitrogen stable isotope ratio ($\delta^{15}N$) values. The adjustment using $\delta^{15}N$ was justified by the fact that the relationship of $\delta^{15}N$ with Hg was the same each year (parallel slopes of the $\delta^{15}N$–Hg concentrations). Adjusted Hg concentrations showed no significant change over the 16-year study period (1990 to 2006) (Gantner et al., 2009).

More recent measurements (2007 and 2008) were added to the original dataset from Gantner et al. (2009) and contributed to the meta-analyses described in Section 5.3.3. Results show a significant decreasing trend (-4.4%) for the small morphotype (Table 5.3) and no trend (1.3%) for the large morphotype. Combining both morphotypes into a single dataset and adjusting for $\delta^{15}N$, as done by Gantner et al. (2009) showed an overall non-significant trend (+17%) for Hg in Lake Hazen char using PIA. The lowest detectable change in the time series is 16% (with a power of 80% and one-sided test, $\alpha = 0.05$). The non-significant trend and relatively high variation among char result in statistical inadequacy for the combined dataset (50%) or the individual morphotypes (50% and 82%, Table 5.3). The unadjusted values and geometric mean concentrations in Lake Hazen char are shown in Figure 5.8.

One explanation for the different Hg trends in small and large morphotypes could be that small morphotypes may respond more immediately to changes in Hg (deposited in the lake) associated with sediments. Small char in Lake Hazen feed mainly on sediment-dwelling organisms (chironomids) and therefore feed closer to the 'source' of bioavailable Hg (i.e., the site of methylation). While large char that feed on juvenile char have greater Hg concentrations owing to greater biomagnification, their Hg concentrations also are more variable, making trend analysis more difficult. Unfortunately, the available sediment record (Muir et al., 2009) does not extend past 2004, making a parallel examination of the trends in sediment and small char impossible. Another explanation may be that the statistical approach for this assessment was different from the original trend (Gantner et al., 2009). While the two groups were separated using a $\delta^{15}N$ cut-off, unadjusted Hg values were used in the PIA analysis here. The overall decline in Hg at Lake Hazen of -5.4% appears to be greater than the modest declines estimated by PIA for landlocked char in Resolute Lake and in Lake Abiskojaure (Sweden) (Table 5.3). In two other lakes in the Canadian Arctic and also in Lakes Á Mýrunum (Faroe Islands) and Isortoq (Greenland), the Hg trend in char increased or showed a non-significant positive trend (Table 5.3).

The reason for the apparent decline in Hg concentration in small morphotype char in Lake Hazen is unknown. With a surface area of $537.5\ km^2$, Lake Hazen is the tenth largest Arctic lake and the world's largest lake north of 74° N. It is much larger than all the other landlocked char lakes with Hg time series, it is also the most remote from global anthropogenic sources and the only one that is glacier fed. Several indicators of the physical environment of Lake Hazen point to the stability of the system. Basic water chemistry data from multi-year monitoring indicated little change over time (Babaluk, 1999; Babaluk et al., 2009), and Hg fluxes in a dated sediment core from Lake Hazen collected in 2005 showed only an overall modest increase in recent horizons compared to the pre-industrial era (flux ratio = 1.13; compared with an average flux ratio of 2.2 for 16 other Arctic lakes) (Muir et al., 2009). The Hg trends in char and the recent trend in the anthropogenic Hg flux in sediment are compared in Figure 5.8. Nearby measurements of GEM at Alert, Nunavut, Canada show no significant decline to mid-decade (Temme et al., 2007). Thus three different lines of investigation of Hg trends from northern Ellesmere Island are not entirely in agreement although, given inter-annual variation, they suggest very little overall change.

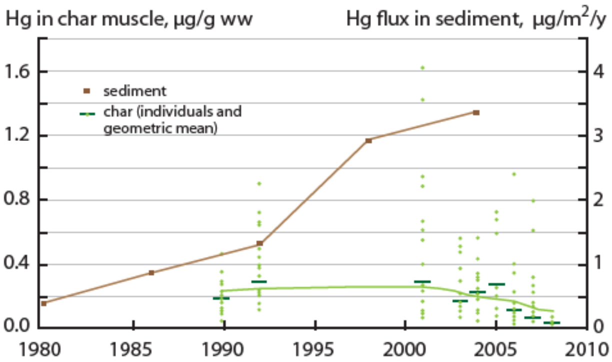

Figure 5.8. Trend in Hg in landlocked Arctic char and a dated sediment core from Lake Hazen, Canada, over the period 1980 to 2008. For char, the plot shows the individual data, annual geometric means, and the 3-point running mean. For sediments, the plot shows the anthropogenic flux, estimated from the average recent (1998 to 2004) flux minus the pre-industrial (1830 to 1840) flux. Symbols represent mid-points of 0.5 cm thick sediment horizons. Data from Gantner et al. (2009) and Muir et al. (2009).

Case Study 2. Canadian Arctic seabirds

Eggs of thick-billed murres (*Uria lomvia*), northern fulmars (*Fulmarus glacialis*) and black-legged kittiwakes (*Rissa tridactyla*) from Prince Leopold Island in Lancaster Sound in the Canadian High Arctic have been monitored for contaminants since 1975 (Braune, 2007). Significant increases in concentrations of THg in eggs of northern fulmars and thick-billed murres were documented between 1975 and 2008, whereas the increase of THg in eggs of blacklegged kittiwakes was not significant over the study period (Figure 5.9, Table 5.3). Mercury concentrations in eggs of ivory gulls (*Pagophila eburnea*) from Seymour Island in the Canadian High Arctic also increased between 1976 and 2004 although the trend was not statistically significant (Braune et al., 2006).

The ivory gull is a resident Arctic species whereas the other three species are migratory, dispersing to various areas throughout the North Atlantic for the winter. Interpretation of

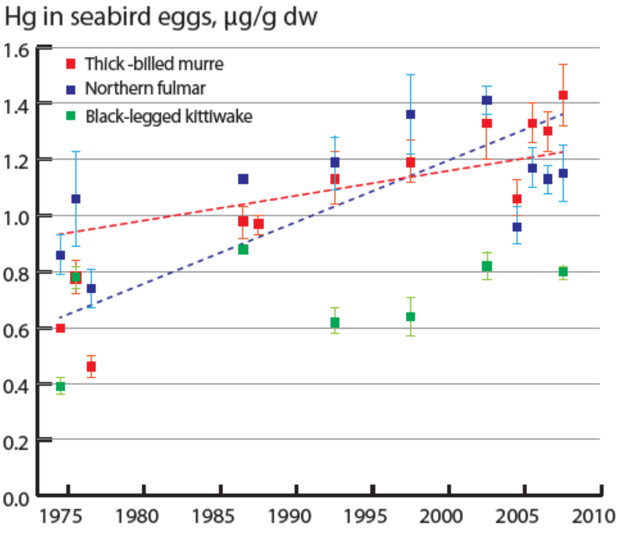

Figure 5.9. Annual mean concentrations (± standard error) of THg in eggs of thick-billed murres, northern fulmars and black-legged kittiwakes from Prince Leopold Island (Canada) for 1975 to 2008. Linear regression lines are shown for statistically significant relationships (*p*<0.05).

Hg levels and trends must include consideration of the year-round distribution of these birds since the components of each egg will contain materials gained from food items obtained at the breeding colony as well as those obtained over the winter and on migration. The kittiwakes probably overwinter on the Grand Banks off Newfoundland and possibly further south along the eastern seaboard of North America (Baird, 1994), whereas band return data suggest that the thick-billed murres from Prince Leopold Island overwinter in open waters off southwestern Greenland (Donaldson et al., 1997). The northern fulmars from the Canadian Arctic appear to overwinter anywhere from the Labrador Sea to the North East Atlantic (Hatch and Nettleship, 1998; Mallory, 2005). In general, then, the overwintering range of the kittiwakes breeding at Prince Leopold Island extends further south than that of the murres and fulmars (Figure 5.10).

The lack of a significant trend in THg concentrations in eggs of kittiwakes is similar to time series data showing no change or decreasing Hg concentrations in seabirds at lower latitudes such as herring gulls from the Great Lakes (Koster et al., 1996), common terns from the New Jersey coast (Burger and Gochfeld, 2004) and the Wadden Sea (Becker et al., 2001), and little terns from the Baltic Sea (Thyen et al., 2000). This is in agreement with the declining Hg emissions in North America (Lindberg et al., 2007), suggesting that, by overwintering at lower latitudes, kittiwakes may be exposed to less Hg over the year and are less impacted by the changing Hg bioavailability in the Arctic. In contrast, since the ivory gulls, murres and fulmars spend extended periods of time at higher latitudes, the trend of increasing Hg concentrations in their eggs may reflect different atmospheric sources and/or deposition processes in higher latitude regions when compared to lower latitude regions. Although there has not been a net change in the size of the atmospheric pool of Hg in the Northern Hemisphere since the mid-1970s (Lindberg et al., 2007), the proportionate emissions attributed to the various source regions have changed, with Hg emissions from Asia increasing, and those from Europe and North America decreasing (Lindberg et al., 2007). Further studies are needed to investigate changes in the bioavailability of Hg in the Arctic and to what extent changes in regional emission patterns are affecting the Hg concentrations in High Arctic seabirds.

Case Study 3. Non-predatory and predatory fish from Arctic reservoirs

A specific source for MeHg in freshwater biota is the construction of man-made reservoirs and river diversion systems. The process of decomposition of flooded organic material (in soil and in vegetation) appears to promote Hg methylation, but the physical parameters of reservoirs may also be important. Rates of Hg methylation are far greater in anaerobic-aerobic transition zones with enhanced activity of sulfate-reducing bacteria which are the main methylators of Hg (see Section 3.3). In Arctic and sub-Arctic regions, this has been shown in Canada (e.g., Bodaly et al., 1984) and Finland (Lodenius et al., 1983; Verta et al., 1985). Increases in MeHg were found in all compartments of the reservoirs: water, zooplankton, benthic invertebrates, and fish; downstream effects were reported (e.g., Bodaly et al., 1997; Lucotte et al.,

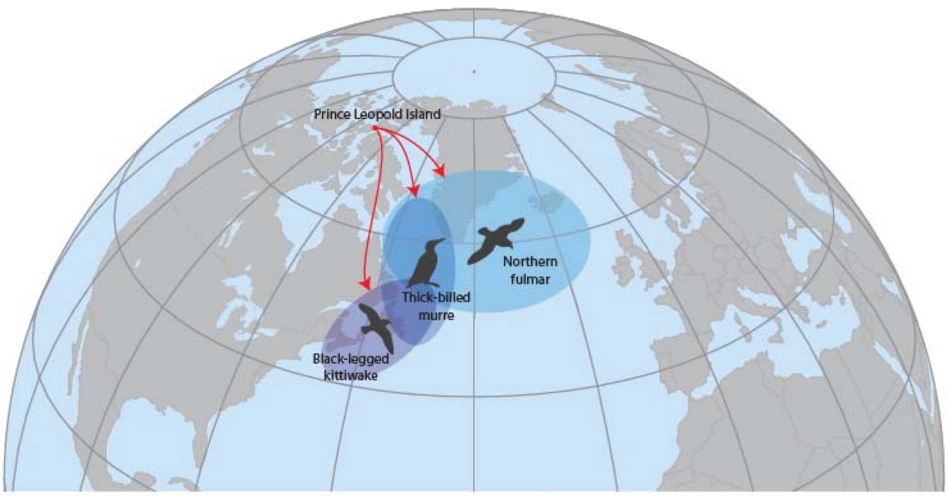

Figure 5.10. Assumed overwintering areas based on best available information for thick-billed murres, northern fulmars and black-legged kittiwakes breeding on Prince Leopold Island, Canada.

1999; Schetagne et al., 2000; St. Louis et al., 2004). In most cases, the MeHg concentration in water and in fish eventually decreases, with a typical recovery time of more than 10 years for water and more than 20 years for fish.

The post impoundment time trends for MeHg concentrations in non-piscivorous whitefish (*Coregonus clupeaformis*) and in piscivorous fish, walleye (*Sander vitreus*) and northern pike (*Esox lucius*), from some hydroelectric reservoirs in northern Manitoba and the Finnish Arctic are presented in Figure 5.11. These reservoirs were constructed in the late 1960s to mid-1970s. The muscle Hg concentration in all the fish increased shortly after the impoundment, with higher Hg concentrations in the predatory fish than the non-predatory fish. The Hg concentration in the fish peaked in the first ten years after the impoundment and then started to decline. By the 2000s (20 to 30 years after the impoundment) the Hg concentration in fish had recovered to background levels. Atmospheric Hg deposition plays a negligible role in the fish MeHg concentration in these reservoirs.

Case Study 4. Burbot and lake trout from the Mackenzie River basin, Canada

Long-term monitoring of THg in burbot (*Lota lota*) has been ongoing at the Rampart Rapids of the Mackenzie River near Fort Good Hope since 1985 and in Great Slave Lake since 1998

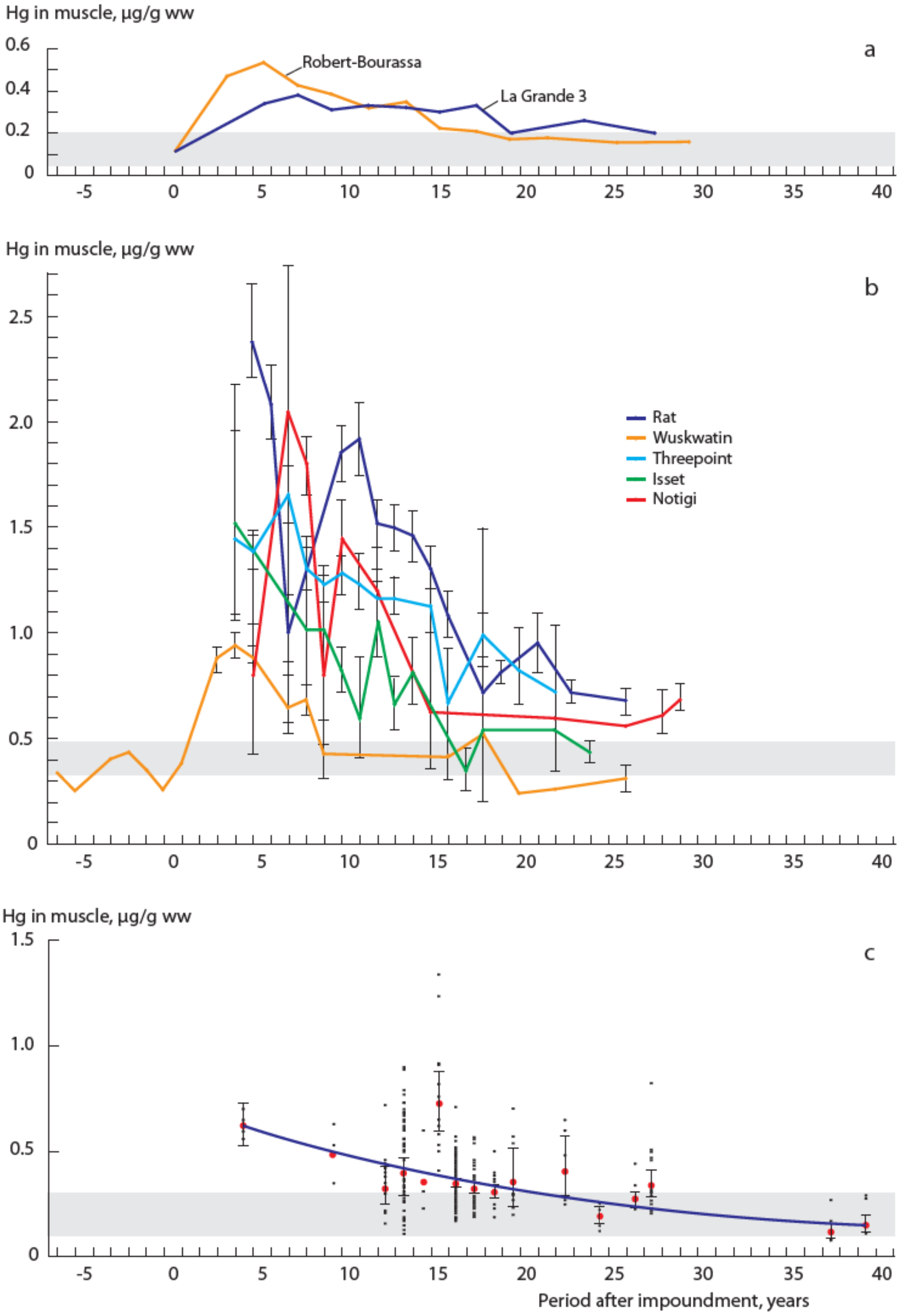

Figure 5.11. Mean Hg concentrations (± 95% confidence levels) in (a) muscle of lake whitefish from hydroelectric reservoirs in northern Québec, Canada, (b) muscle of walleye from hydroelectric reservoirs in northern Manitoba, Canada, and (c) muscle of northern pike from the Lokka hydroelectric reservoir in northern Finland. Adapted from R. Schetagne (Hydro Québec, Montréal, Canada, pers. comm., 21/12/2009), Bodaly et al., 2007; Porvari, 1998; E. Huttula (Kemijoki Group, Rovaniemi, Finland, pers. comm., 4/11/2009). Note that the time of impoundment is denoted as year 0. The grey bands indicate the probable concentration range in the region before impoundment.

(Evans et al., 2005a; Lockhart et al., 2005a; see Figure 5.12). Mercury levels have also been periodically monitored in lake trout (*Salvelinus namaycush*) in lakes along the Mackenzie River (Evans et al., 2005a; Lockhart et al., 2005a).

Burbot is a top predator freshwater fish and an important staple for northern Canadian communities. Carrie et al. (2010) recently examined the temporal trend for THg in muscle of burbot from the Fort Good Hope area and the causes for such a trend. The fish were collected in winter (December to January) by the local residents and analyzed for age (*via* otoliths), sex, morphometry, contaminants (THg and PCBs), and stable isotopes (δ^{13}C, δ^{15}N). Overall, Carrie et al. (2010) reported data for 13 time points over the 24-year period from 1985 to 2008. Over this period, muscle Hg concentrations increased around 2-fold (Figure 5.13) and cannot be explained by differing physiological characteristics or changes in feeding behavior. There is no significant change in δ^{15}N and δ^{13}C data over the period 1995 to 2005, implying that burbot have not been feeding differently; nor are there significant differences for δ^{15}N between years of lower Hg concentration (1999: 0.24 ± 0.06 µg/g; mean ± SD) and higher Hg concentration (2004: 0.40 ± 0.09 µg/g; mean ± SD). Additionally, no correlation was noted between muscle Hg concentration and δ^{15}N or δ^{13}C. There is also no correlation between muscle Hg concentration and fish size (weight or length), with the exception of a weak, although statistically significant, correlation with age. This last finding is contrary to observations for other fish species (e.g., Evans et al., 2005b; Gantner et al., 2009) and may be attributed to a relatively short half-life of Hg in burbot muscle and liver (Carrie et al., 2010). The overall increasing trend in burbot Hg concentration and its associated large degree of temporal variability (Figure 5.13) cannot be explained by the atmospheric Hg trend either, as atmospheric Hg concentrations have been stable or falling in the Arctic over the past 25 years (Chapter 2).

Using a dated sediment core from Hare Indian Lake, a tributary lake near the Mackenzie River sampling site (Figure 5.12), Carrie et al. (2010) showed striking temporal similarities between Hg concentration in burbot muscle and in Hare Indian Lake sediment core slices when plotted against year of collection and median year of deposition, respectively (Figure 5.13). Furthermore, there is an almost perfect point-by-point co-variation between the Hg concentration (normalized against the conservative element titanium (Ti) to correct for potential geogenic inputs of Hg) in the sediment core and labile, algal-derived organic matter (measured as S1 and S2 carbon by Rock-Eval pyrolysis; Sanei and Goodarzi, 2006) over the past 150 years, similar to what has been reported in two High Arctic lakes (Outridge et al., 2007). While tagging data on Mackenzie River burbot are scarce, the results of a study conducted in Aklavik (450 km away in the Mackenzie Delta) found that burbot for the most part stayed within 15 km of their tagging point (Stein et al, 1973). Also, Hare Indian Lake is characteristic of burbot feeding habitat (McPhail and Lindsey, 1970; Scott and Crossman, 1973). Based on these results, Carrie et al. (2010) suggested that the significant increase in Hg concentration in burbot muscle since the 1980s may be strongly influenced by increased concentrations of algal-derived organic matter in the water column. This is possible as algal blooms are efficient scavengers, concentrators, and possibly producers, of MeHg from and to the water column and hence sources

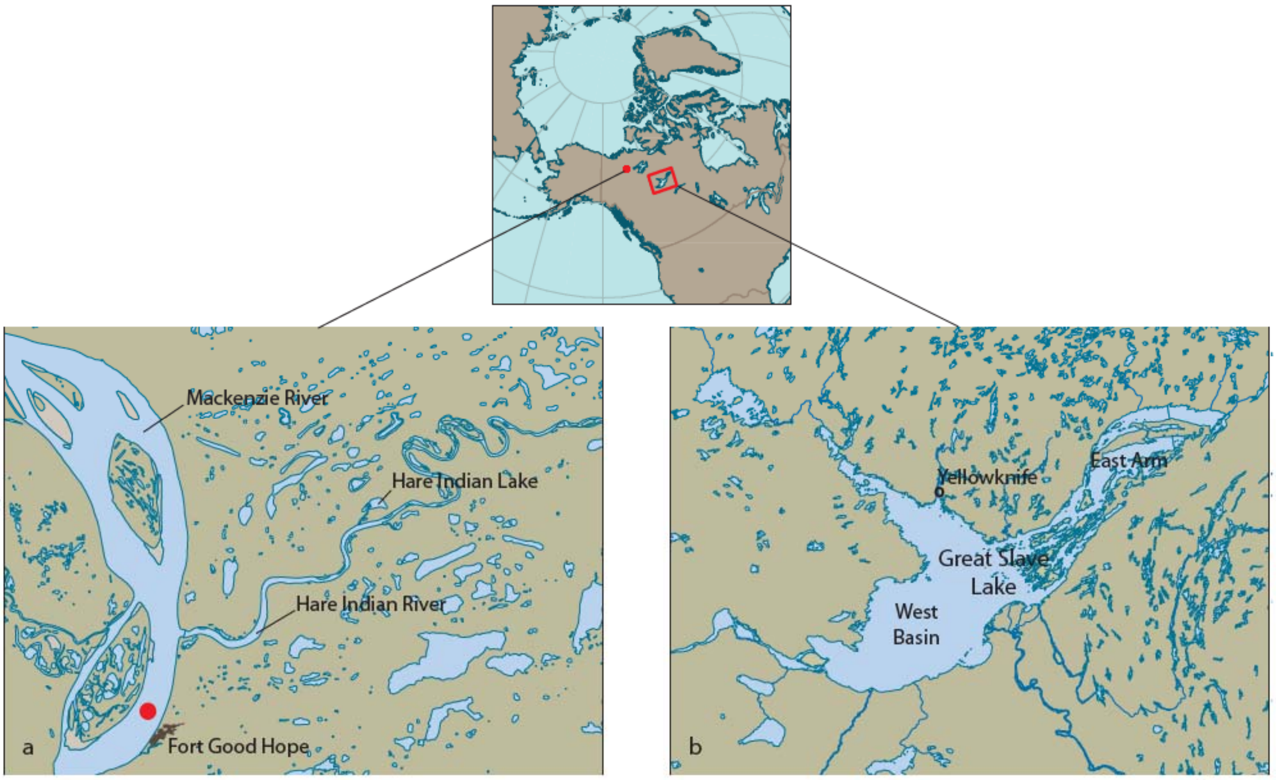

Figure 5.12. (a) Fort Good Hope region, showing the fish collection site (red dot) and the lake sediment core site (Hare Indian Lake), and (b) Great Slave Lake showing the West Basin and East Arm. Modified from Carrie et al. (2010).

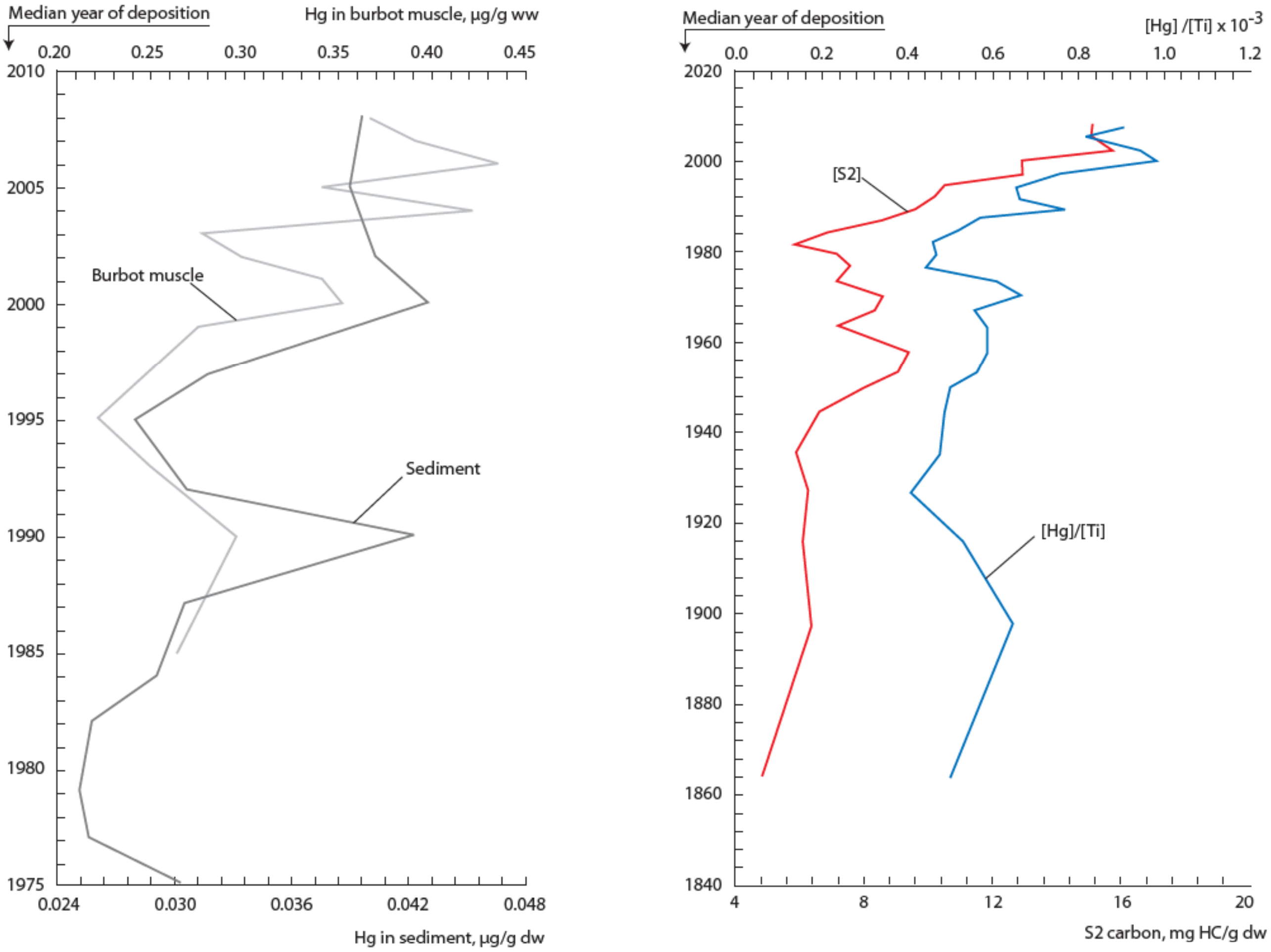

Figure 5.13. Chronological variations (a) in total mercury concentration in Hare Indian Lake sediment core and in Mackenzie River burbot and (b) by point comparison between [THg]/[Ti] and S2 carbon concentrations in the Hare Indian lake sediment core. Source: Carrie et al. (2010).

of bioaccumulative Hg to larger biota (Balogh et al., 2002; Pickhardt et al., 2002; Luengen and Flegal, 2009).

The significant increase in algal-derived organic matter as indicated from the sediment core is also consistent with the climatic record of the Mackenzie Basin. The mean annual air temperature for the nearby Norman Wells, for instance, has seen a statistically significant increase of about 1.9 °C since the early 1970s. This region has also seen a significant decrease in snow cover over the same period. Reduction in ice-season length, and consequent improvement in the light regime for algal growth, is thus a plausible explanation for the coincidence among rising air temperature, algal productivity, and increasing Hg in Hare Indian Lake sediment in Mackenzie River burbot.

A generally increasing trend, although not always statistically significant, has also been observed in burbot and lake trout in a broader range of lakes in the Mackenzie River Basin with different physical features and watersheds (Evans et al., 2005b). The greatest increases in Hg concentration in lake trout are observed in warmer, smaller, and shallower lakes with large watersheds. Small lakes with large watersheds have a proportionately larger littoral zone relative to the pelagic zone, a closer connection to watershed inputs of Hg and dissolved organic carbon, warm to a greater extent in summer, and thus potentially experience a higher rate of increase in algal productivity.

Furthermore, different time trends in different fish species in a given lake from this region suggest that the biology of the fish is also important (Evans et al., 2005b). This is most evident for Great Slave Lake which has two distinct sub-basins (see Figure 5.12): the West Basin is strongly influenced by the Slave River, while the East Arm is deeper and less productive. In the West Basin, burbot have shown a pronounced trend in Hg concentration, while trends are less evident for lake trout (Table 5.3). This is possibly because the lake trout is a coldwater stenotherm and thus more associated with deeper waters, whereas burbot is a nearshore fish that is exposed more to the littoral zone. Mercury concentrations are lower in both species in the East Arm where watershed influences are weaker and the waters deeper.

Case Study 5. Ringed seals from Ulukhaktok

Total Hg concentration in muscle tissue from ringed seals (*Phoca hispida*) sampled at Ulukhaktok (Holman), NWT, in the western Canadian Arctic was recently reported for ten years from the period 1973 to 2007 (Gaden et al., 2009). All samples were taken during the subsistence harvests primarily during June and prior to break-up of the sea ice. In total, 214 samples of muscle tissue from adult seals ranging in age from 7 to 36 yrs were analyzed for THg. Stable isotope ratios of nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) were also analyzed for 117 of the muscle tissue samples. These biomarkers, together with biological data (age and sex) and climate data (length of the ice-free season in the Eastern Amundsen Gulf), were examined for trends.

No significant difference was found in Hg concentration between the sexes (p = 0.31), therefore male and female seal samples were pooled for subsequent analysis. No temporal trends in THg were observed for the study period from 1973 to 2007 (p = 0.24), but a significant association of the muscle Hg concentration was found with the length of the ice-free season in the previous year to seal harvest. The THg concentrations displayed a 2nd-order polynomial trend in which concentrations increased from about 140 ice-free days onward and about 110 days and under (Figure 5.14), suggesting that the muscle Hg concentration in seals may have been influenced by the previous year's environmental conditions which affect the types and/or amounts of prey eaten by the ringed seals. This reasoning is based upon (i) the turnover of stable isotope ratios and Hg in muscle tissue, (ii) the seal population's seasonal distribution and diet and (iii) biological and ecological responses to shifts in climate parameters.

By knowing the turnover times of elements in certain tissues, the timeframe of prey consumption and subsequent deposition into tissues can be identified. In this study, the turnover of THg in seal muscle was found to be about twice as long (seven months) as the typical replacement time of tissue nitrogen and carbon (three months) (Gyrd-Hansen, 1981; Baumann et al., 1994). Since the seals were harvested during June–July, the turnover times of the δ^{15}N and δ^{13}C signatures correspond to the seal diet in March (late winter) and November (late autumn to early winter) for the THg concentrations. This mismatch in turnover times may explain why THg was strongly associated with the previous ice-free season but that δ^{15}N and δ^{13}C were not. It is likely that the length of the ice-free season in the previous year may have affected the seal diet in the consecutive autumn, reflecting the measured THg concentrations, in comparison to late winter (associated with the δ^{15}N and δ^{13}C signatures).

The next step, then, was to consider the diet of the study population during late autumn to early winter. Ringed seals are opportunistic foragers and have a varied diet that changes seasonally. Work by Smith (1987) showed that adults in the study population remain near Ulukhaktok in the Prince Albert Sound from late autumn to ice break-up. Throughout the ice-covered period, ringed seals primarily consume Arctic cod (*Boreogadus saida*) (Smith, 1987). Therefore, variation in the length of the ice-free season may have influenced the overall exposure of seals to Hg by means of changes in the cod

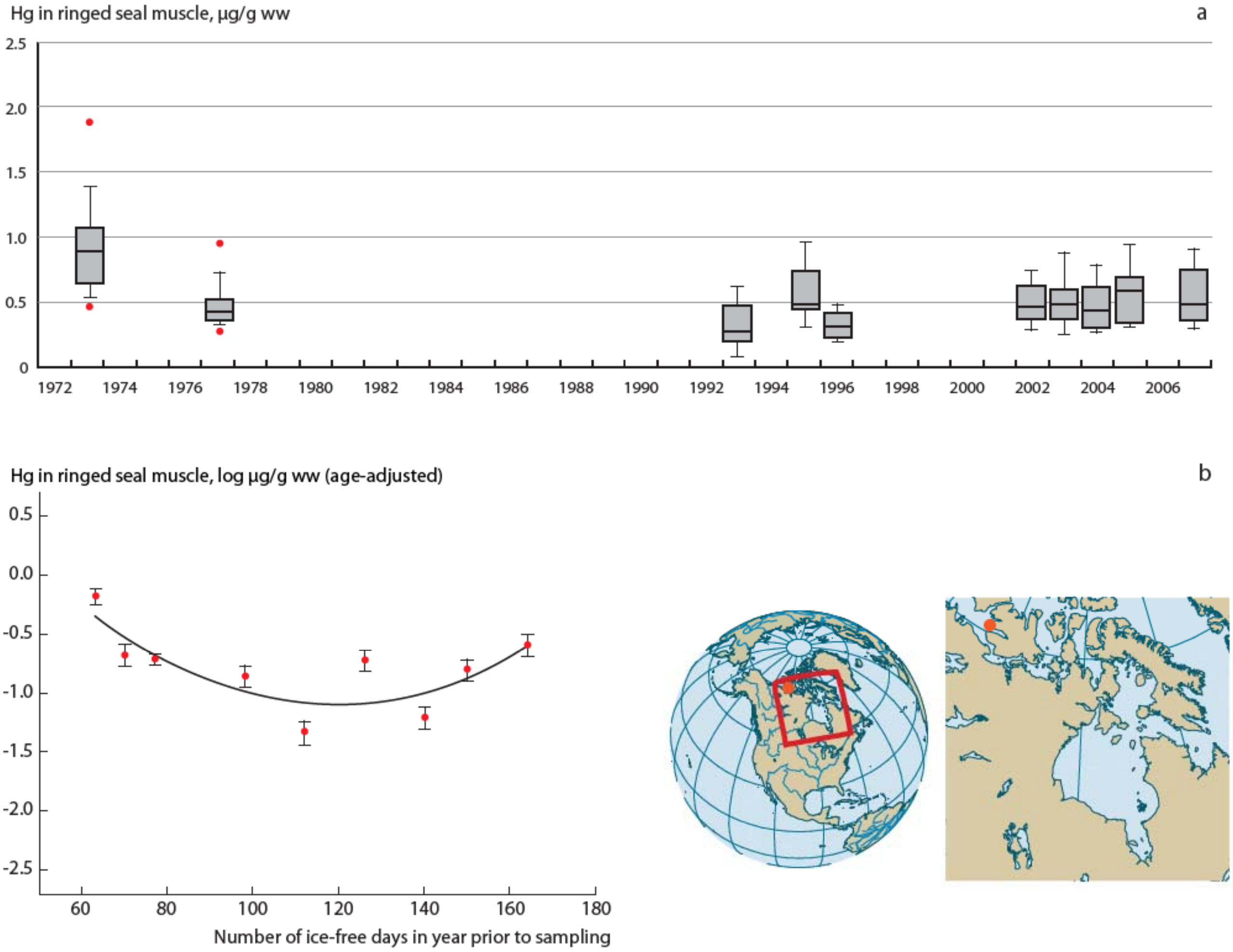

Figure 5.14. (a) Box and whisker plot of total Hg concentrations in muscle of adult (7+ yrs) ringed seals (males and females) sampled during the period 1973 to 2007, Ulukhaktok (Holman), NWT, Canada. Lines in boxes represent mean concentrations; whiskers represent 5th and 95th percentiles. (b) Log-transformed, age-adjusted mean total Hg concentration in muscle of adult (7+ yrs) ringed seals (males and females) plotted against ice-free days in the year prior to sampling. Source: Gaden et al. (2009).

population structure or abundance available to seals in the late autumn to early winter.

Gaden et al. (2009) postulated that during short ice-free seasons there may have been shifts in the cohort composition of the Arctic cod population, whereas long ice-free seasons could have resulted in higher numbers of the prey species. Young Arctic cod are particularly vulnerable to the harsh conditions imposed by the ice-covered period (Michaud et al., 1996). In years with long winters and shorter ice-free seasons, a majority of older, more contaminated cod may have out-lived the younger cohorts, exposing predators like the seals to a higher Hg intake (Lockhart and Evans, 2000). Alternatively, longer ice-free seasons are likely to have favored the survival and abundance of the cod via the longer growing season, enhanced ecological productivity and overall increased food availability in the pelagic food web. With the higher abundance of Arctic cod, ringed seals could have fulfilled a larger proportion of their diet with cod and subsequently accumulated higher Hg loads.

Findings suggest that shifts in climate over the study period indirectly influenced Hg uptake in ringed seals in the western Canadian Arctic. The length of the ice-free season in the Arctic is predicted to increase in the future (Walsh, 2008), and therefore it is plausible that Hg concentrations may rise accordingly.

Case Study 6. Reindeer and Arctic char in northern Scandinavia

Atmospheric deposition is the dominant source of Hg to northern Scandinavia. Early Hg deposition measurements were made at irregular intervals in Jergul and Överbygd in 1986 and 1987 (Iverfeldt, 1991). Between 1991 and 1996, Hg bulk deposition measurements were performed close to Kiruna on a monthly basis (Berg et al., 2002). Deposition measurements have been ongoing at the Pallas-Matorova station since 1996 (Wängberg et al., 2007). When comparing the bulk atmospheric deposition of Hg at these four locations, there seems to have been a 30% to 50% decrease within the past 20 years in this region, depending on whether the Överbygd data are included or not (Figure 5.15a). This reduction in Hg deposition is supported to some extent by a moss survey in Sweden. A monitoring program to measure atmospheric heavy metal deposition using mosses was established in 1980 (Harmens et al, 2008). Data collected at five-year intervals between 1985 and 2005 show Hg concentrations in moss from this region to range from < 0.05 to 0.1 μg/g dw for the period 1985 to 1995, but to decrease to < 0.05 μg/g dw after 2000 (www3.ivl.se/miljo/projekt/mossa/hg.asp).

Mercury levels in reindeer (*Rangifer tarandus*) and Arctic char (*Salvenlius alpinus*) from the Abisko National Park, Sweden, have been monitored since the early 1980s. The area is located around 450 to 700 m above sea level and is not affected by local pollution. Reindeer are strict herbivores feeding predominantly on lichen in winter, while the summer diet also includes low growing sedges and willows. Due to this short terrestrial food web, reindeer have been suggested as a useful species for monitoring change in Hg deposition as a result of the direct contaminant pathway air → plant → reindeer (Poissant et al., 2008; Lokken et al., 2009). As part of the Swedish contaminant monitoring program, muscle tissue

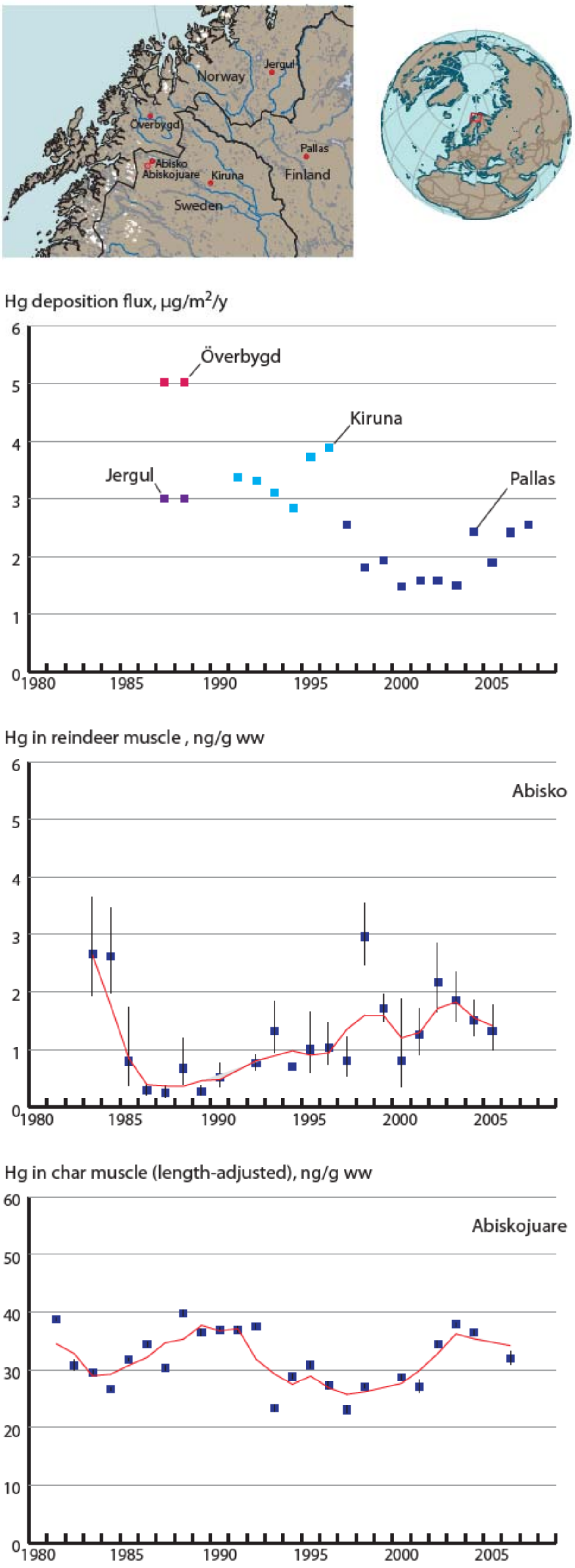

Figure 5.15. (a) Atmospheric Hg deposition at various sites in northern Scandinavia between 1986 and 2007. (b) Total Hg concentrations in muscle of reindeer from Abisko, northern Sweden (geometric mean ± 95% confidence limits). (c) Total Hg concentrations in muscle of Arctic char from Lake Abiskojaure, northern Sweden (length-adjusted geometric means). The red line shows a three-point running mean smoother ($p < 0.05$). Data from Wängberg et al. (2007), Odsjö et al. (2007) and Bignert (2002).

from the front tibia of male reindeer (3 years old) is sampled before the rutting period in September each year. The uptake of Hg in freshwater fish is probably more complex, due to several factors affecting uptake, such as the humic content of the water, acidity and temperature, although recent data suggest a rapid response in fish Hg content to changes in air deposition (Harris et al., 2007). Arctic char of equal size (ca. 30 cm, 220 g, 4 years old) are sampled in August/September each year from Lake Abiskojaure, which is an oligotrophic lake with no vegetation.

Temporal trends show no significant change in THg concentration in the muscle tissue of reindeer (Odsjö et al., 2007) or char (Bignert, 2002) from Abisko over the past 25 years (Figure 5.15b and c). For reindeer, the same is true for THg liver concentrations, which generally follow the same pattern as for muscle concentrations (data not shown). Although not significant for the time period as a whole, the apparent increase in reindeer muscle Hg levels since around 1990 (Figure 5.15b) contradicts the calculated decrease in atmospheric Hg deposition between the late 1980s and 2000s in the same region. It should be noted, however, that the concentrations in reindeer muscle are very low, with a geometric mean of 1.00 ng/g ww (0.73 to 1.37 ng/g ww; 95% confidence limits), and often close to the detection limit. Liver concentrations are about 40 times higher but this tissue showed no trend (Table 5.3). Similarly for char, there is no notable relationship between Hg deposition levels and time trends in fish (Figure 5.15c).

It can be assumed that the Hg deposition in Abisko mimics reasonably well the measured deposition at the various stations in northern Scandinavia. The apparent decrease in Hg deposition and moss concentration in the area is not reflected in the Hg levels in terrestrial herbivore reindeer or in Arctic char over the same time period. This indicates that factors other than atmospheric Hg deposition are more important drivers for Hg concentration in biota in this region. These factors may be process-based and dependent on environmental characteristics, such as changes in food availability, food selection, or food web structure. Climate change may alter food availability and food selection for reindeer, thus affecting dietary exposure to Hg more than changes in deposition. Consequently, even moderate changes in deposition fluxes may not be reflected in these species over short periods.

Case Study 7. Beluga from the western Canadian Arctic

Mercury concentrations in various tissues of beluga from the western Canadian Arctic have been monitored since 1981, making this the longest temporal dataset for Hg concentrations in Arctic beluga (Lockhart et al., 2005b; Stern and Lockhart, 2009). Since 1981, Hg concentrations in the beluga population at Hendrickson Island have increased significantly (Table 5.3). Although there is a good relationship between beluga age and liver Hg concentration (Figure 5.16), there is no simple relationship between muscle Hg concentration and beluga age. However, larger individuals tend to have higher Hg concentrations in their muscle and this reflects habitat use and diet (Loseto et al., 2008a) (Figure 5.16). This temporal trend and size-dependence cannot be explained by the atmospheric Hg trend, and so there have been extensive studies in recent years on Hg sources other than atmospheric deposition (Leitch et al., 2007), as well as on the habitat and feeding behavior of beluga (Loseto et al., 2006, 2008b, 2009).

Evaluating the relative importance of Hg sources to food webs provided a bottom-up approach to examining drivers of Hg temporal trends in top predators. In addition to atmospheric deposition, Hg is also transported to the Beaufort Sea from riverine discharge, thawing permafrost, coastal erosion, and oceanic circulation (Leitch et al., 2007). The Mackenzie River was found to be a particularly important Hg source, transporting about 2 tonnes of THg per year to the Mackenzie Delta, which is similar to or higher than the atmospheric Hg depositional flux to the Beaufort Sea (Leitch et al., 2007). The Mackenzie River is the largest river emptying into the Beaufort Sea, providing many of the nutrients and much of the particulate organic matter that fuel nearshore and potentially offshore pelagic food webs (Carmack and Macdonald, 2002). The impact of the large Hg flux associated with the Mackenzie River outflow may not be limited to the nearshore food web; the Hg could be further deposited and re-distributed in the biologically active

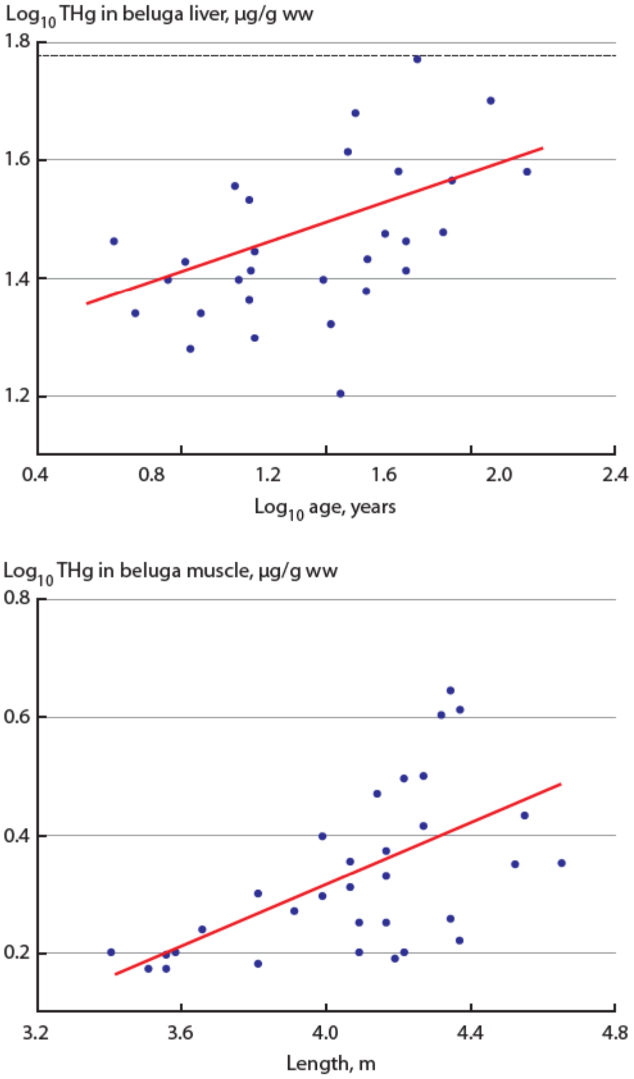

Figure 5.16. The relationship between THg concentrations in beluga liver and age (in years, estimated based on 1 growth layer group) is best described by an exponential relationship (R2 = 0.24; *p* = 0.006) demonstrating accumulation over time, whereas for THg in beluga muscle it is best described by an exponential relationship with length (R2 = 0.35; *p* <0.0001) where the biomagnification process dominates. The dashed line indicates the observed effects level in marine mammals of 60 µg/g ww (see Section 6.3.2). Modified from Loseto et al. (2008a).

shelf break area where Hg may be resuspended and/or made bioavailable to the offshore food web (Loseto et al., 2008b).

In contrast to evaluating sources to food webs that biomagnify to levels measured in top predators, a top-down approach was considered to evaluate food web processes (guild and trophic transfer of Hg) and predator behavior (habitat use, diet preferences) to understand Hg concentrations in the Beaufort Sea beluga population. First, beluga behavior was examined to gain insight into diet preferences and dietary exposure to Hg. Combining beluga movement patterns (with use of satellite telemetry) and features of the physical environment (sea ice, bathymetry), the Beaufort Sea beluga population was found to sexually segregate during their summer habitat use (Loseto et al., 2006). This provided a basis for considering food webs and dietary Hg sources associated with the different habitats to which individuals of the population were potentially exposed (see Chapter 3, Figure 3.6). Food webs were then characterized according to the three habitats the Beaufort Sea beluga population appeared to feed in (Loseto et al., 2008b). Hypothetical feeding groups and food webs provided a framework to begin merging the predator behavior, food web dynamics and dietary sources of Hg. Results demonstrated the role and importance both of Hg sources to food webs and the processes within food webs that may differ from benthic to pelagic systems (i.e., bioaccumulation and biodilution) and finally the importance of food web length toward describing sources to predators (i.e., beluga) (Loseto et al., 2008b).

A detailed fatty acid biomarker approach was also used to define diet (Loseto et al., 2009). Data revealed Arctic cod to be the dominant prey item for the Beaufort Sea beluga population. However, size-related dietary differences were evident, whereby larger-sized beluga preferred offshore Arctic cod, and smaller-sized beluga appeared to feed on prey in nearshore habitats that included nearshore Arctic cod. If these size trends can be explained by energetics then the large males need to maintain body mass by adjusting foraging behavior to feed either on energy-rich prey or to feed more often (Boyd et al., 1997), suggesting a greater abundance or availability of Arctic cod in the deep offshore Arctic Ocean.

Therefore, evaluating beluga Hg levels required that both a top-down and bottom-up perspective be considered. That is, the top-down factors influencing beluga habitat use, such as predation, resource selection, and nursing also defined feeding behavior and associated dietary Hg exposure among size and sex classes in the population. From the bottom-up perspective, different food webs, for example the nearshore estuarine food web, had lower Hg levels relative to the offshore pelagic food web that biomagnified to top predators (see Chapter 3). When integrating the sources of Hg, the food web processes and structure, and predator feeding ecology, they revealed the size-dependence of Hg concentration in beluga muscle. A concerted effort to merge the two approaches is needed to further clarify the dominance of source or process in driving Hg trends in the Beaufort Sea beluga and other higher trophic level species (Chapter 7).

Case Study 8. Greenland polar bears

Recent time series of Hg concentrations in polar bear hair from northwestern Greenland, East Greenland and Svalbard have been reported by Dietz et al. (2006a) and are updated in Figure 5.17. There appears to be an overall upward trend in the annual median Hg concentration in polar bear hair from Greenland and no overall trend for Svalbard. Further analysis was carried out to evaluate whether the medians as well as the residuals from the trend line calculated from the annual medians could

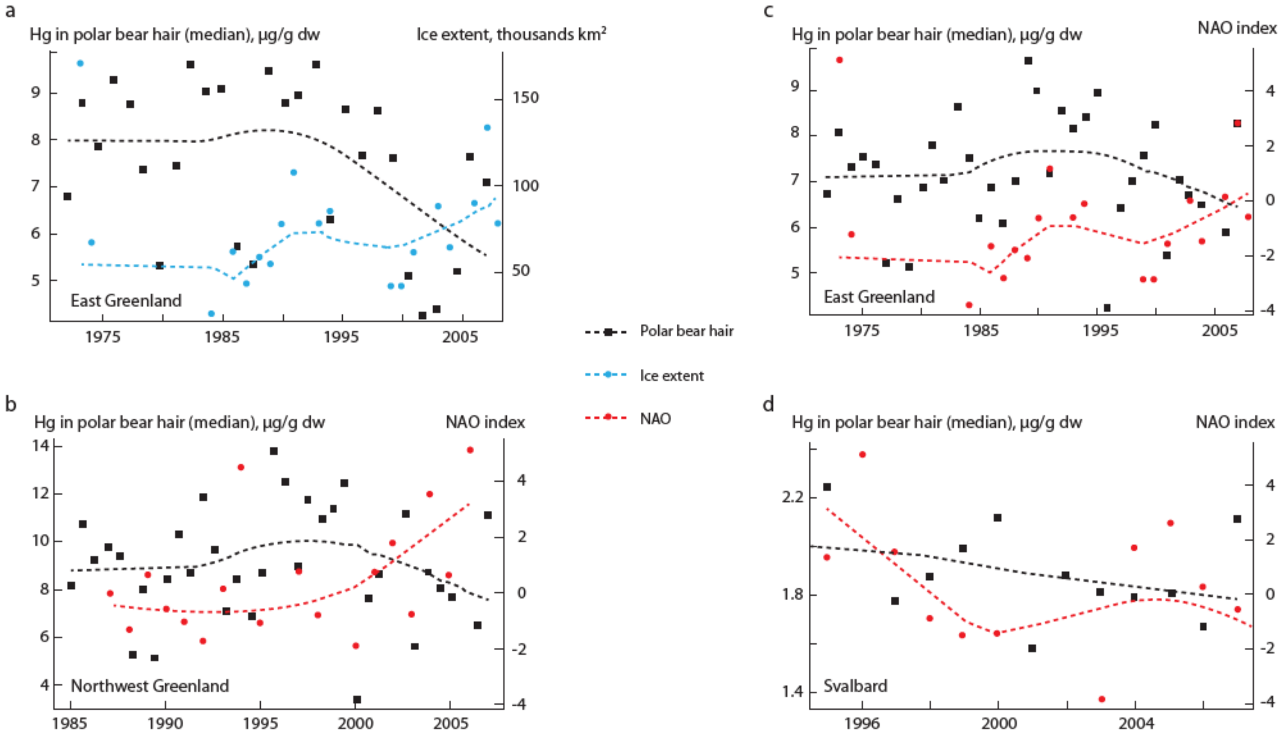

Figure 5.17. Temporal trends and Loess smoother estimates of (a) median Hg concentrations in polar bear hair and ice extent in East Greenland; and polar bear hair and the North Atlantic Oscillation Index for (b) northwestern Greenland, (c) East Greenland and (c) Svalbard. Source: Dietz et al. (2008a,b, 2009b).

Table 5.6. Correlation coefficients and significance levels (in brackets) between the Loess smoother estimate (see Figure 5.17) based on median mercury concentrations in polar bear hair and ice extent and the North Atlantic Oscillation Index for northwestern Greenland, East Greenland and Svalbard. Source: Dietz et al. (2006a, 2008a,b, 2009b).

Hg concentration vs	Ice extent	North Atlantic Oscillation Index
Region	Loess smoother estimate based on median values	Loess smoother estimate based on median values
Northwestern Greenland		-0.97 (0.001)
East Greenland	-0.59 (0.02)	-0.48 (0.04)
Svalbard		0.67 (0.02)

be linked to climate variables such as polar sea-ice coverage and the North Atlantic Oscillation (NAO) Index (Dietz et al., 2008a,b, 2009b). Using median Hg values, significant correlations were found with ice extent (average for September) in East Greenland and for the NAO Index in all three regions (Table 5.6, Figure 5.17). For the residuals deviating from the median, a significant negative correlation was detected between the NAO Index and Hg concentration in East Greenland polar bear hair, whereas a weak positive correlation was detected for bears from the Svalbard area. Significant negative correlations were also found between Hg concentration in East Greenland polar bear hair and the sea-ice extent in the same year. Overall the decreasing sea-ice extent and the decline in the NAO Index in East Greenland were correlated with increases in Hg concentration in polar bears, whereas the opposite was observed at Svalbard. Ongoing calculations on the effect on stable isotopes will be used to resolve whether changes in the food chain can explain part of the change in Hg concentration. It remains unresolved why such differences appear in these two regions and to what extent these are linked to changes in transport routes or the food chain. However, East Greenland polar bears have easier access to the harp and hooded seals breeding areas as the ice edge gets closer to the coast (R. Dietz, National Environmental Research Institute, Denmark, and E.W. Born, Greenland Institute of Natural Resources, unpublished data). On the other hand, reduced sea ice at Svalbard may result in longer fasting periods on land resulting in lower Hg exposure. Clearly, the relationships found indicate that climate parameters should be taken into consideration when predicting trends in biotic Hg concentration.

5.5. Conclusions and recommendations

Conclusions (in numbered bullets) are organized under section headings, followed by knowledge gaps / recommendations (in italics) when appropriate.

How much higher are current mercury levels in Arctic biota than in the pre-industrial period, and thus what is the anthropogenic contribution to mercury in modern biota?

1. Long-term trend studies using archeological and museum collections suggest that on average over 90% (median 92%, range 74% to 94%) of the present-day Hg in upper trophic level marine biota (beluga, ringed seal, polar bear, birds of prey) is of anthropogenic origin.

2. This roughly ten-fold increase in Hg concentration in upper trophic level marine biota over the past few centuries is much greater than the increase in ambient environmental Hg levels (~25% in global upper oceans, see Chapter 2), probably because of a non-linear relationship between increases in environmental Hg levels and biotic Hg levels due to biomagnification.

3. The late 19th to 20th century showed a steep increase in biotic Hg levels compared to relatively constant values over the previous four to six centuries. The average rate of Hg increase over the past 150 years was usually within the range of 1% to 4% per year.

In terms of long-term trends, available information is limited to fewer than ten wildlife species in the Canadian and Greenlandic regions of the Arctic; coverage of low trophic level species and invertebrates is particularly limited. Historical time series from the Alaskan and Russian Arctic would be a valuable addition to knowledge of anthropogenic impacts on modern Hg levels.

In which areas and species have mercury levels recently increased or decreased?

4. Of the 83 recent time series (i.e., covering the past few decades) which met the specified requirement of at least six years of data, 13 (16%) showed a statistically significant increasing trend (including two from different tissues in the same animals), four (5%) showed a statistically significant downward trend, 21 (25%) showed a statistically significant non-linear trend (non-linear trend component is significant), with the rest (54%) showing no statistically significant trend.

5. Most of the time series showing significantly increasing trends involved marine species, followed by freshwater fish species. No significant increases in Hg were found for terrestrial animals in recent decades.

6. A greater number of significantly increasing trends occurred in Canada and Greenland compared with northern Europe, suggesting that Hg concentrations are increasing to a greater extent in the western Arctic (west of 40° W) compared with the eastern Arctic (east of 40° W).

7. The statistical ‘adequacy’ of recent time series to detect pre-determined annual changes in Hg concentration has increased from 10% to 19% of total datasets over the past five years, owing to continued monitoring and the resulting growth in the statistical power of available time series.

For recent biotic time series, there is a regional imbalance in data availability. Few or no tissue monitoring data which met *the specified data requirements were available from the Alaskan, Russian or Finnish areas of the Arctic. Continued support for ongoing time series, and initiation of biotic Hg monitoring in regions presently lacking coverage, will further add to the accuracy of the picture of recent trends in biotic Hg concentrations.*

Why are mercury levels in Arctic biota increasing or decreasing?

8. The significant increase in Hg concentration in present-day Arctic animals compared to their pre-industrial counterparts is probably explained primarily by the increase in anthropogenic Hg in Arctic ecosystems.

9. The temporal variations in Hg concentration in Arctic animals over recent decades, however, cannot be solely explained by changes in anthropogenic atmospheric Hg emissions. Recent biotic Hg trends that are increasing in the Canadian and Greenlandic Arctic do not match regional atmospheric Hg records which show stable or declining trends over recent decades.

10. The extent to which regional shifts in Hg emissions from Europe and North America to Asia are currently affecting Hg concentrations in Arctic biota remains unknown. Environmental and ecological processes appear to play an increasingly important role in determining Hg trends in Arctic animals, particularly those at higher trophic levels.

Uncertainties concerning the net deposition rate of Hg from AMDEs and other wet and dry atmospheric deposition processes in the Arctic (see Chapter 3) limit the ability to evaluate competing theories about the important drivers of recent trends in biotic Hg levels. Little is known about how changes in the Arctic cryosphere (snow, lake and sea ice, brine, permafrost) are affecting Hg bioaccumulation in Arctic ecosystems. Also lacking is systematic information on habitat and feeding behavior for many Arctic species, which can affect Hg concentrations in biota through alterations in MeHg assimilation and biomagnification.