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Review

Anthropogenic contributions to mercury levels in present-day Arctic animals—A review

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ABSTRACT

Background: Because of concern about the recently increasing levels of biological Hg in some areas of the Arctic, we examined the literature concerning the long-term changes of Hg in humans and selected Arctic marine mammals and birds of prey since pre-industrial times (i.e. before 1800 A.D.), to determine the anthropogenic contribution to present-day Hg concentrations and the historical timing of any changes.
Methods: Mercury data from published articles were extracted on historical and pre-industrial concentrations as percentages of the recent maximum, as well as the man-made contribution was calculated and depicted in a uniform manner to provide an overview of the development over time.
Results and discussion: Trends of [Hg] in hard tissues such as teeth, hair and feathers consistently showed that there had been an order-of-magnitude increase of [Hg] in Arctic marine foodweb-based animals that began in the mid- to late-19th Century and accelerated in the 20th Century. The median man-made contribution to present-day Hg concentrations was 92.4% ranging from 74.2 to 94.4%. Confidence in our data was increased by accompanying data in some studies on stable isotopes ($\delta^{13}C$, $\delta^{15}N$), which allowed us to normalize where necessary for changes in animal trophic position and feeding location over time, and by careful attention to the possibility of sample chemical diagenesis (Hg contamination or loss) which can alter the Hg content of ancient hard tissues.
Conclusions: Wildlife hard tissue matrices provide consistent information with respect to the steep onset of Hg exposure of Arctic wildlife beginning in the latter half of the 19th Century. Today the man-made contribution was found to be above 92%. Stable isotope analyses provide important information to normalize for possible changes in diet over time, and are highly relevant to include when interpreting temporal trends, baseline concentrations as well as man-made anthropogenic contribution of Hg.

Contents

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1. Introduction

Mercury (Hg) in the Arctic is one of the highest-profile metal contamination issues of recent decades. Despite the absence of any major Hg point sources in the region, an estimated 200–300 t of Hg is transported each year from various human (anthropogenic) and natural sources into the Arctic by atmospheric processes, ocean currents, coastal erosion and rivers (AMAP, 2003; Skov et al., 2004; Outridge et al., 2008). A series of processes unique to or accelerated in the Arctic then influences Hg dynamics and biological uptake. For example, gaseous elemental Hg is photo-catalytically depleted from the atmosphere in the Arctic each spring (Schroeder et al., 1998; Lindberg et al., 2002), followed by photo-reduction and revolatilization processes in snowpacks which greatly reduce the net amount of Hg deposition (Lahoutifard et al., 2005; Kirk et al., 2006; Constant et al., 2007). There is evidence that rapid climate warming has altered Hg dynamics in Arctic lakes (Outridge et al., 2007), and its accumulation by marine biota (Gaden et al., 2009). These findings underscore the global nature, multi-disciplinarity and complexity of Arctic Hg pollution science.

Although Hg is a naturally occurring element and, as such, was always present in the environment, global human activity has led to a several-fold increase of Hg emissions into the atmosphere and into oceans and rivers (Nriagu and Pacyna, 1988; Pacyna et al., 2006). In some areas of the Arctic, Hg concentrations in marine foodwebs have significantly increased in recent decades (Braune et al., 2005), causing levels in some marine mammals, birds, and fish to reach the point where adverse biological effects might be expected (AMAP, 2003). Mercury also biomagnifies in Arctic ecosystems, and concentrations in higher trophic-level marine species (seabirds, seals, and whales) are relatively high compared to elsewhere in the world (Dietz et al., 1998), resulting in high human exposure via consumption of these animals. In certain Arctic and sub-Arctic locations (e.g., the Faroe Islands), humans may have a daily dietary intake of Hg that well exceeds the threshold for negative effects on children's neurological development (AMAP, 2003; Choi and Grandjean, 2008). Recently the economic impacts of Hg polluted food sources in the Arctic were compiled to document to policy makers and the public that it would be cost effective to ban the trade in Hg and limit its emissions (Hylander and Goodsite 2006).

The question remains to what extent anthropogenic Hg emissions around the world have actually altered Hg concentrations in Arctic biota, and therefore the exposure of humans who continue to use this wildlife as food. To answer this question, the pre-industrial (i.e. pre-1800 A.D. natural baseline) levels of Arctic biological Hg first need to be established. These data can then be compared to current concentrations to determine the anthropogenic contribution in modern animals. Samples of animal tissues from the pre-industrial era are not available. However, calcified and keratinaceous hard tissues such as teeth, hair and feathers have the potential to help resolve this question because they tend to survive well in dry, cold climates like polar regions (Outridge, 2005) and are well represented in various museum collections. 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] (Born et al., 1991; Eide and Wesenberg, 1993; Eide et al., 1995; Outridge et al., 2000; Bearhop et al., 2000a,b). Thus, analysis of historical and modern samples of hard tissues can be informative about the long-term changes of Hg intake and body burdens in wildlife (Outridge, 2005).

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 at 1300 A.D. from Northwest Greenland (Dietz et al., 2006a). The tooth Hg content of scores of Canadian beluga (*Delphinapterus leucas*), walrus (*Odobenus rosmarus 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, 2005, in press), and the long-term changes assessed in the context of animal age structure and dietary information inferred from tooth stable carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotope data. Finally, Hg time trends in primary feathers of West Greenland gyrfalcons (*Falco rusticulus*), peregrine falcons (*Falco peregrinus*), and white-tailed sea eagles (*Haliaeetus albicilla*) covering the period 1850–2004 were documented (Dietz et al., 2006b). In this review, we combine the published time series to calculate the anthropogenic contribution to present-day Hg concentrations in Arctic biota, and to determine the historical timing of any changes. We also discuss the caveats of our interpretations, especially the effects of possible changes in feeding behaviour (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.

2. Material and methods

2.1. Important features of hard tissues from a mercury monitoring perspective

Because this review is concerned with animal hard tissues as indicators of biotic Hg trends, a short discussion of the attributes of different bioarchives is appropriate, focussing on the three types of hard tissues—hair, teeth, and feathers—and the animal species for which long-term data are available. Outridge (2005) treated this subject in greater detail.

2.1.1. Hair

Hair (or fur) is a keratin-based tissue, metabolically inert once formed, which has been widely studied as an indicator of blood Hg levels and dietary exposure in humans (FAO/WHO, 2003). Human hair appears to precisely reflect blood Hg concentrations, with an average ratio of blood:hair Hg concentrations of about 1:250 across a range of low- and high-exposure populations (FAO/WHO, 2003). Controlled dosing studies with rodents showed that of the four 'indicators' of inhaled inorganic Hg that were studied, i.e. hair, blood, molar teeth and incisor teeth, hair displayed the highest correlation coefficient with exposure (Eide and Wesenberg, 1993). Hair Hg was also significantly correlated with concentrations in animal soft tissues including kidney, liver, muscle, cerebrum and cerebellum in both laboratory and free-living populations of different species (Eide and Wesenberg, 1993; Evans et al., 1998; Mierle et al., 2000; Fortin et al., 2001; Ikemoto et al., 2004; Cardona-Marek et al., 2009). Lack of correlations between soft tissue and hair Hg have also been observed in some fur-bearing species, for example, for organic and total Hg in the liver and kidney of river otter (*Lutra Canadensis*; Halbrook et al.,

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1994; Fortin et al., 2001), total Hg in the muscle, kidney and liver of northern fur seals (*Callorhinus ursinus*), and the liver and kidney of Caspian seals (*Pusa caspica*, Ikemoto et al., 2004). A possible explanation for these different outcomes may be inter-specific differences in metabolic Hg demethylation and excretion via hair, which confounds the hair:tissue Hg relationship.

For many Arctic species, winter fur is formed during autumn and hence hair sampling represents an integrated measure of exposure mediated through the blood concentration at this time of the year. Born et al. (1991) explained how polar bear winter fur is grown from September to October, and how part of this fur is molted at the end of May. The remaining fur, named summer fur (June–August), does not differ in [Hg] from the winter fur (September–May) (Born et al., 1991), probably because hair is inert once formed. Seal species show a similar pattern with distinct molting periods which is August for harbour seals (*Phoca vitulina*) and July for harp seals (*Pagophilus groenlandicus*) and hooded seals (*Cystophora cristata*) (Reeves and Ling, 1981; Ronald and Healey, 1981). This means that seal hair represents a very uniform matrix, which will not show seasonal variation. In other species, hair grows at an approximately constant and continual rate (~1 cm per month in humans), allowing the short-term Hg exposure to be reconstructed by sequential analysis of hair segments (e.g. Legrand et al., 2007).

2.1.2. Teeth

Like hair, the teeth of mammals are living tissues in which minerals and metals are deposited directly from the bloodstream into actively growing increments. Three chemically and functionally distinct types of tooth material are found in mammals: enamel, dentine and cementum, although in some species such as some toothed whales only dentine and cementum are present (Hillson, 1986). Enamel is believed to generally cease growth around the time of sexual maturity. In the teeth of seals, bears and humans, the growth of dentine and cementum ceases or slows down once the pulpa cavity has been filled, whereas the teeth of toothed whales including beluga continue to grow and thus contain material that has been laid down continuously over a lifetime. Dental wear, however, may remove early lifetime layers (Hillson, 1986).

Only a few studies have examined relationships between Hg concentrations in teeth and in soft tissues or exposure regimes. Eide and Wesenberg (1993) found that Hg levels in incisor and molar teeth of laboratory rats were significantly correlated with kidney Hg following inhalation exposure, with the correlation coefficients between tissues and teeth higher than they were for hair or blood (although hair was more highly correlated with exposure level). Eide et al. (1995) further demonstrated that rat molars and incisors were indicators of inorganic and methyl Hg dietary intake. A study with free-living beluga showed that Hg concentrations in tooth cementum were highly correlated with those in liver, muscle, kidney and muktuk, i.e. the skin with attached fat (Outridge et al., 2000).

2.1.3. Feathers

The use of feathers to monitor the Hg body burden of birds has attracted a good deal of scientific investigation. The growth of primary feathers shows a similar seasonal pattern as for hair in Arctic mammals. Feathers from the various primaries and different species are laid down at specific times of the year. In the study of Dietz et al. (2006b), the distal end of the fifth primary (P5) feather was sampled from white-tailed eagles (*H. albicilla*), gyrfalcons (*F. rusticulus*) and peregrine falcons (*F. peregrinus*), as these are believed to be molted and generated in Greenland during summer. However, for other primaries the seasonality will be different, and other species may show different seasonal and annual patterns in their feather shedding and regeneration (Cramp, 1979). Virtually all feather Hg is in the methyl form (Thompson and Furness, 1989; Thompson et al., 1990), and is deposited in relatively high concentrations (μg/g DW range) which are correlated with blood Hg levels at the time of feather growth (Bearhop et al., 2000a,b). Once deposited into feathers, Hg is physiologically isolated from other tissues and is firmly bound to disulphide linkages of keratin. However, caution is warranted. Many birds, unlike most mammals, are capable of mobilizing most (70—>90%) of their methyl Hg body burden, and excreting it in feathers during each seasonal molt cycle (Honda et al., 1986, Braune and Gaskin, 1987). Consequently, the Hg kinetics of blood and tissues in birds are more complex than in mammals, and are probably influenced by high variability in excretion rates during molting (Bearhop et al., 2000a,b). Feathers formed at different times during this cycle, or on different parts of a bird's body, can exhibit markedly different Hg contents (Appelquist et al., 1984; Furness et al., 1986; Braune and Gaskin, 1987; Becker et al., 1994).

Another complication is that birds, like mammals, are capable of demethylating ingested methyl Hg to varying degrees, resulting in the storage and accumulation of apparently inert inorganic Hg in liver (Wolfe et al., 1998). In seabird species with a high proportion of inorganic Hg in liver, liver [Hg] may not be correlated with feather Hg concentrations because they are affected by two decoupled mechanisms—demethylation, and the excretion of methyl Hg (Thompson et al., 1990). Thus, while they reflect blood Hg levels, changing feather [Hg] during the lifetime of individuals, or differences between individuals of the same species, may be possibly more strongly influenced by physiological state than by dietary intake or organ Hg concentrations. Studies of Hg in modern and pre-industrial bird feathers should therefore attempt to control for parameters such as time of sampling in relation to the stage of the molting cycle, type and sequence of the feathers, and sex (Outridge, 2005).

2.2. Calculation of the anthropogenic component of Hg in modern animals

Like other long-term archives such as lake sediments and peat bogs, the fundamental approach to estimation of the anthropogenic Hg contribution in modern biota is to first establish the average natural background concentration in a species at a location using samples from the pre-industrial period. The modern [Hg] increase, if any, in the species at that location is taken to be the anthropogenic contribution. An important qualification is that the original Hg content of the historical material has been well preserved, and that all other factors which might affect Hg concentrations in animals (such as a population's demographics or feeding behaviour) have remained constant or can be corrected for, to ensure that the pre-industrial and modern data are directly comparable. These issues are discussed later in this review.

First, however, the published Hg data need to be represented in a consistent and comparable way across tissues and species. Published studies in general report absolute Hg concentrations which are not easy to compare and sometimes do not provide a clear picture of increases over time. As Table 1 and Fig. 1 show, absolute concentrations in the hard tissues of various species varied over 3–4 orders of magnitude both in the pre- and post-industrial period. Also, it is difficult to describe in a uniform way the onset and magnitude of the anthropogenic increase in such different matrices. Therefore, we initially set the maximum present-day annual median concentration in each dataset to 100%, and represented the median historical and pre-industrial concentrations as percentages of the recent maximum, using the formula:

$$\textit{Historic proportion of present}\,(\%) = (\textit{Median historic}\,[Hg] / \textit{Median recent maximum}\,[Hg]) \times 100 \quad (1)$$

The resulting data distribution, which extends backwards eight centuries, is shown in Fig. 2.

As seen from this figure the asymptotic line prior to ca. 1850 represents the percent baseline contribution. If this baseline percentage

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Table 1
Selected time series elucidating increases above baseline and historic concentrations as well as an estimate of % man-made Hg above baseline.

Matrix	Species (age group)	Region	Year (on graph)	Year (estimate or range)	Hg (μg/g dw)	*n*	Historic proportion of present	Historic proportion of present - Baseline percentage (Eqs. (2a), (2b))	% Man made contribution (Eq. (3))	Reference
Hair	Ringed seal	Central West Greenland	1470	1500	0.600	9	***23.1***	***0.0***	***0.0***	Hansen et al., 1989
			1975	1975	2.600	10	***100.0***	***76.9***	***76.9***	
	Human	Central West Greenland	1470	1500	3.100	6	***31.6***	***0.0***	***0.0***	Hansen et al., 1989
			1975	1975	9.800	22	***100.0***	***68.4***	***68.4***	
	Polar bear	Northeast Canada	1290	300 B.C.-1500 A.D.	0.700	6	**7.9**	**0.0**	**0.0**	Wheatley and Wheatley, 1988
			1975	1971-1977	8.900	10	**100.0**	**92.1**	**92.1**	
		Northwest Greenland	1300	1300	0.520	2	**6.8**	**0.0**	**0.0**	Dietz et al., 2006a
			1920	1915-1924	1.530	2	**20.1**	**13.3**	**66.0**	
			1940	1935-1944	6.370	2	**83.7**	**76.9**	**91.8**	
			1960	1955-1964	3.460	1	**45.5**	**38.7**	**85.0**	
			1990	1985-1994	7.610	62	**100.0**	**93.2**	**93.2**	
		Northwest Greenland	1300	1300	0.520	2	**8.6**	**0.0**	**0.0**	Dietz et al., 2006a
		Central East Greenland	1890	1885-1994	1.360	2	**22.5**	**13.9**	**61.8**	Dietz et al., 2006a
			1910	1905-1914	0.858	3	**14.2**	**5.6**	**39.4**	
			1930	1925-1934	0.391	1	**6.5**	**0.0**	**-33.0**	
			1950	1945-1954	1.610	2	**26.7**	**18.1**	**67.7**	
			1970	1965-1974	6.040	58	**100.0**	**91.4**	**91.4**	
			1980	1975-1984	4.250	20	**70.4**	**61.8**	**87.8**	
			1990	1985-1994	5.360	147	**88.7**	**80.1**	**90.3**	
			2000	1995-2002	4.900	97	**81.1**	**72.5**	**89.4**	
Teeth	Ringed seal (5 year)	Holman	1350	14th century	0.0005	29	**11.4**	**0.0**	**0.0**	Outridge et al., submitted
			1870	late 19th century	0.0005	5	**11.4**	**0.0**	**0.0**	
			2002	2001 and 2003	0.004	38	**100.0**	**88.6**	**88.6**	
	(25 years)		1350	14th century	0.0005	29	**5.8**	**0.0**	**0.0**	
			1870	late 19th century	0.0005	5	**5.8**	**0.0**	**0.0**	
			2002	2001 and 2003	0.009	38	**100.0**	**94.2**	**94.2**	
	Walrus	Igloolik	1350	1200-1500	0.001		115.8	0.0	0.0	Outridge et al., 2002
			1992	1988 and 1996	0.001		100.0	0.0	0.0	
	Beluga whale	Mackenzie Delta	1550	1450-1650	0.004	28	**6.5**	**0.0**	**0.0**	Outridge et al., 2002
			1993	1993	0.055	25	**100.0**	**93.5**	**93.5**	
	(20 year)[a]	Mackenzie Delta	1550	1550	0.005		**25.8**	**0.0**	**0.0**	Outridge et al., submitted
			1960	1960	0.017		**89.5**	**63.7**	**71.2**	
			1993	1993	0.019		**100.0**	**74.2**	**74.2**	
	(40 year)[a]		1550	1550	0.010		**11.0**	**0.0**	**0.0**	
			1960	1960	0.039		**43.9**	**33.0**	**75.0**	
			1993	1993	0.089		**100.0**	**89.0**	**89.0**	

	(60 year)[a]		1550	1550	0.017		**5.6**	**0.0**	**0.0**	
			1960	1960	0.075		**25.5**	**19.8**	**77.9**	
			1993	1993	0.292		**100.0**	**94.4**	**94.4**	
	(20 year)[b]	Somerset Island	1886	1886	0.007		**24.6**	**17.0**		Outridge et al., 2005
			1995	1995	0.027		**100.0**	**92.4**		
	(40 year)[b]		1886	1886	0.013		**17.1**	**9.5**		
			1995	1995	0.078		**100.0**	**92.4**		
	(60 year)[b]		1886	1886	0.023		**12.9**	**5.3**		
			1995	1995	0.178		**100.0**	**92.4**		
	Human	Northern Norway	1210	Around 1200	0.24	57	**7.4**	**0.0**	**0.0**	Eide et al., 1993
			1972	1971–1972	3.23	124	**100.0**	**92.6**	**92.6**	Tvinnereim et al., 2000
Feather	Gyrfalcon (Juvenile)	Central West Greenland	1850	1845–1854	0.380		**38.0**	**30.4**		Dietz et al., 2006b
			1880	1875–1884	0.430	13	**43.0**	**35.4**		
			1890	1885–1894	0.610	40	**61.0**	**53.4**		
			1900	1895–1904	0.610	73	**61.0**	**53.4**		
			1910	1905–1914	0.550	35	**55.0**	**47.4**		
			1920	1915–1924	0.690	49	**69.0**	**61.4**		
			1930	1925–1934	0.690	20	**69.0**	**61.4**		
			1960	1955–1964	1.000		**100.0**	**92.4**		
	(Immature)		1880	1875–1884	0.570	1	**0.6**	**0.0**		Dietz et al., 2006b
			1890	1885–1894	4.380	2	**20.1**	**12.5**		
			1900	1895–1904	2.650	4	**12.2**	**4.6**		
			1910	1905–1914	5.160	4	**23.7**	**16.1**		
			1920	1915–1924	10.840	1	**49.7**	**42.1**		
			1930	1925–1934	15.040	1	**69.0**	**61.4**		
	Peregrine falcon (Juvenile)		1850	1845–1854	1.030	2	**15.8**	**8.2**		Dietz et al., 2006b
			1890	1885–1894	1.710	3	**26.3**	**18.7**		
			1900	1895–1904	1.430	15	**22.0**	**14.4**		
			1910	1905–1914	1.990	14	**30.6**	**23.0**		
			1920	1915–1924	1.770	20	**27.2**	**19.6**		
			1930	1925–1934	2.390	4	**36.8**	**29.2**		
			1980	1975–1984	6.500	2	**100.0**	**92.4**		
Bold	Median historic %					10	**7.6**	**0.0**	**0.0**	
Bold	Median present contribution					10	**100.0**	**92.4**	**92.4**	
All	Median historic %					12	***8.2***	***0.0***	***0.0***	
All	Median present contribution					12	***100.0***	***91.8***	***91.8***	

Bold red figures indicate values used to estimate average present increase factor above baseline. Bold blue figures indicate baseline percentage of present and % man-made contribution. Bold black italics figures are only used in the lower two lines estimating historic and present contribution based on all values except for the normal caption walrus figures where the present concentration is lower than historic concentration.

[a]Three out of 10 available age-group data selected.

[b]Age have been doubled due to recent information on age determination where one growth layer group represents one year.

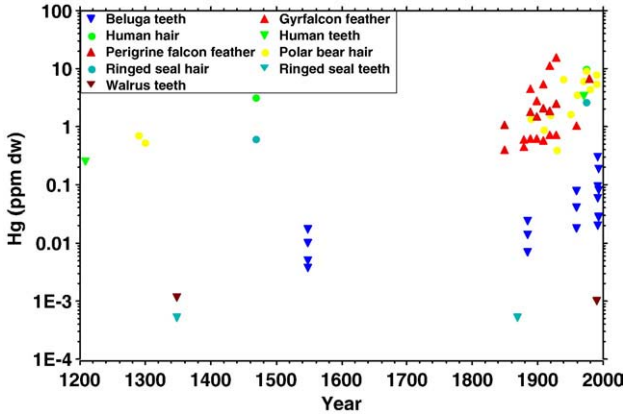

Fig. 1. Mercury concentrations in various present-day and historic hard tissues, plotted on a log scale against year. For sources and date details see Table 1.

is subtracted from anthropogenic sources to biota at any point in time we are able to calculate: (i) Historic proportion of present corrected for the baseline percentage (Eqs. (2a) and (2b), see below), which is a new measure developed for this review, where the baseline percentage is kept constant over time. Compared to the more generally used term (ii) Percent man-made contribution (Eq. (3), see below) the values were the same for the baseline and for the present man-made contribution. The historic proportion of present corrected for the baseline percentage was calculated in two ways: when pre-industrial baseline data were available for the species in the same region,

$$\begin{aligned} &\textit{Baseline} - \textit{Corrected Historic Proportion} \\ &= (\textit{Median historic}\,[Hg] \,/\, \textit{Median recent maximum}\,[Hg] \times 100) \\ &\quad -(\textit{Median baseline}\,[Hg] \,/\, \textit{Median recent maximum}\,[Hg] \times 100); \end{aligned} \tag{2a}$$

and where no specific pre-industrial baseline data were available,

$$\begin{aligned} &\textit{Baseline} - \textit{Corrected Historic Proportion} \\ &= (\textit{Median historic}\,[Hg] \,/\, \textit{Median recent maximum}\,[Hg] \times 100) \\ &\quad -\textit{Median of all baseline data} \end{aligned} \tag{2b}$$

[i.e. 7.6%; see Table 1].

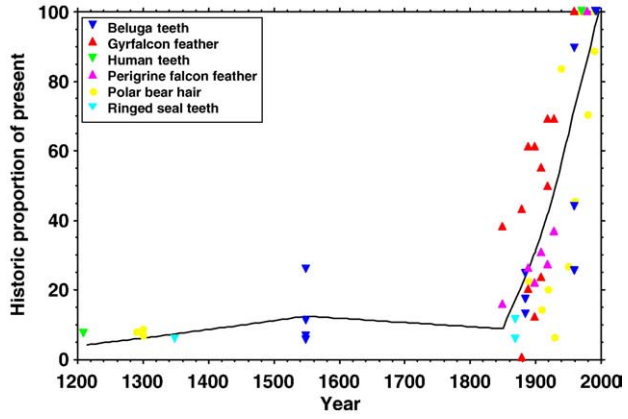

Fig. 2. Historical trends of Hg concentrations in various animal hard tissues, expressed as percentage of present-day maximum annual average concentrations. For sources and data details see Table 1.

The percent man-made contribution was calculated only for datasets which included pre-industrial baseline data, using the standard formula:

$$\begin{aligned} &\textit{Man-made Hg}\,(\%) \\ &= (\textit{Median modern}\,[Hg] - \textit{Median baseline}\,[Hg]) \,/\, \textit{Median modern}\,[Hg] \\ &\quad \times 100 \end{aligned} \tag{3}$$

The results are listed in Table 1 for all studies and depicted in Fig. 3.

3. Results and discussion

3.1. Mercury increases from pre-industrial baseline up to late 20th century

The conversion of absolute concentrations to percentages of the modern maxima (using Eq. (1)) showed that there was a similar overall pattern across species and regions. Pre-industrial era hard tissues contained a median of 7.6% (range: 5.6–25.8%) of the maximum Hg levels recorded for the same species at the same locations during 1975 to 2003 (Table 1 and Fig. 2). Both sets of formulas (Eqs. (2a), (2b) and (3)) agreed on this result. This finding means that 92.4% (range: 74.2–94.4%) of the present-day Hg in Arctic wildlife is of man-made origin (Table 1, Fig. 3). The datasets contributing to this result included polar bear hair, and teeth from humans, beluga and ringed seals, from the Canadian, Greenland and Norwegian regions of the Arctic (Wheatley and Wheatley, 1988; Eide et al., 1993; Tvinnereim et al., 2000; Outridge et al., 2002, 2005; Dietz et al., 2006a,b; Outridge et al., in press).

Several but not all studies included pre-industrial samples (before 1800 A.D.) which allowed for calculations of baseline% of present as well as man-made contribution (Wheatley and Wheatley, 1988; Eide et al., 1993; Tvinnereim et al., 2000; Outridge et al., 2002; Dietz et al., 2006a; Outridge et al., in press). Some of the studies on polar bear hair and birds of prey feathers covered several decades within the recent 150 years (Dietz et al., 2006a,b). From the beluga and seal studies, using teeth as the monitoring matrix and covering a broad range of ages, there is a clear age-related variance in the anthropogenic contribution, with older animals exhibiting a larger man-made percentage than younger animals (Outridge et al., 2002, 2005, in press). This was predicted by Bernhard and Andreae (1984) because the multiplicative effects of lifetime bioaccumulation on tissue Hg in response to environmental Hg increases is greater in individuals with longer lifetimes.

For Arctic biota, such a high man-made contribution across multiple species feeding at high trophic levels, and on-going increasing Hg trends in Arctic Canada and Northwest Greenland biota (Braune et al., 2005; Dietz et al., 2006a), suggests that toxic effects studies in the highest exposed species and regions would be particularly relevant, as a toxicological impact from Hg may be more likely in these high trophic-level species. Although there is an absence of comparable long-term studies of Hg in biota in other regions of the world closer to urban-industrial areas, recent findings from Antarctica indicate that both polar regions have experienced significant increases of biotic Hg since the Industrial Revolution, with the increases in Antarctic biota less than those in the Arctic. Hairs of southern elephant seal (*Mirounga leonine*) retrieved from a lake sediment core displayed considerable [Hg] variation 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

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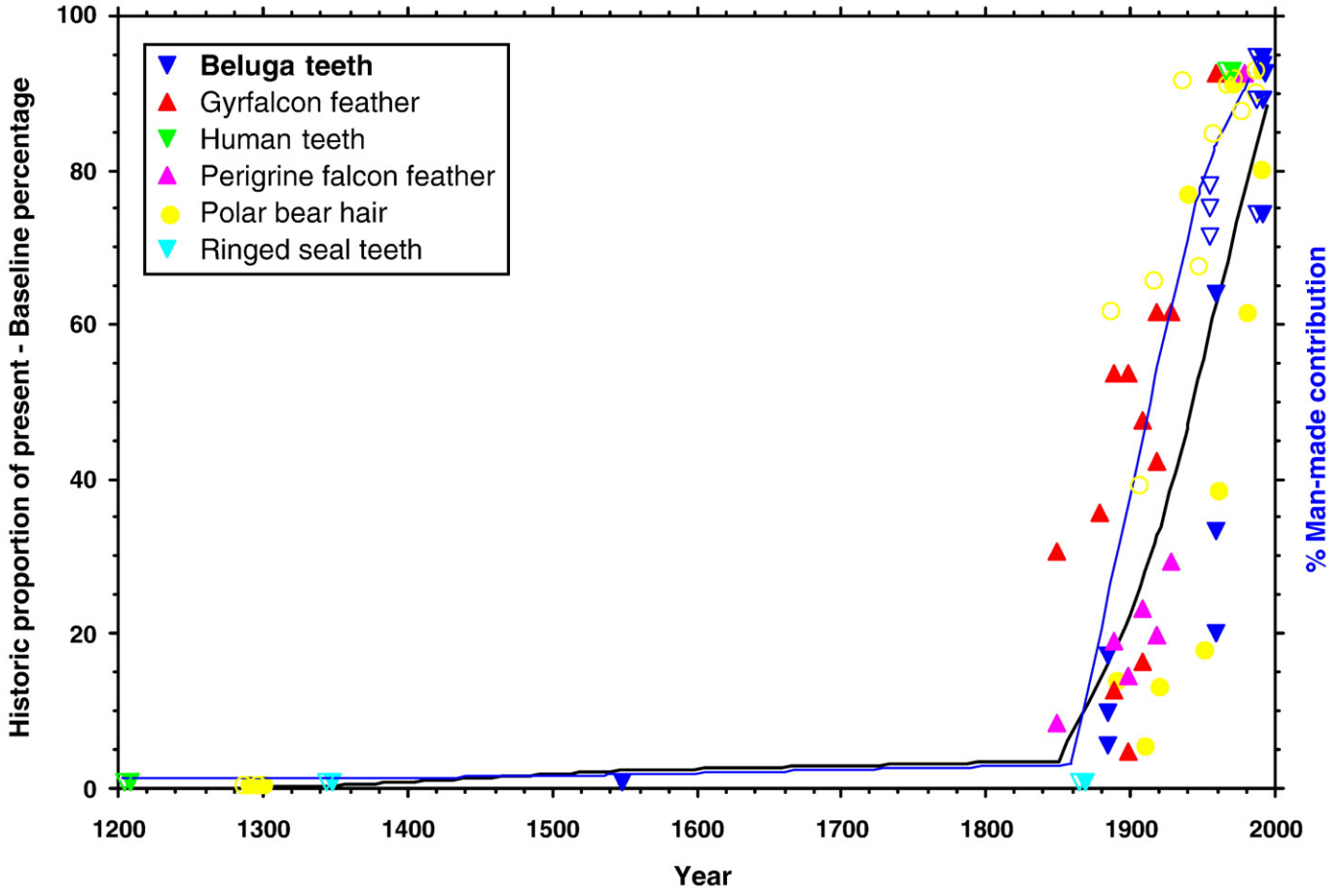

Fig. 3. The historic proportion of present corrected for the baseline percentage (black line and filled symbols; Eqs. (2a), (2b)) compared to the percent man-made Hg contribution (blue line and open symbols; Eq. (3)) in modern biota, in various biological hard tissues. For sources and date details see Table 1. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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% (Table 1). There are several possible explanations for these discrepant findings. 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 of hair Hg in these populations (see Kinghorn et al., 2006). Another possible reason is that modern hunting pressures may have significantly reduced the average age of harvested seals (and other human food items) compared to historic times when the hunting pressure was presumably lower (Dietz, unpublished data). This would have acted to reduce the Hg increases in harvested seal hair and the modern human Hg intake from those seals (and Hg in human hair), giving an inaccurate comparison between time periods. 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 enclosed small 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.

The only Arctic species not to show a significant modern increase was walrus (*O. rosmarus rosmarus*) near Igloolik, Canada (Outridge et al., 2002). This 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 environmental Hg increases compared to higher level species, because biomagnification would have little effect on Hg concentrations in these species or their prey in response to elevated environmental Hg levels (Bernhard and Andreae, 1984). Our results are in accord with this prediction and hence the walrus data were not included in the above calculations.

3.2. Timing of mercury increases over the last 150 years

As seen from Figs. 2 and 3 (and Table 1), the 20th Century showed a much steeper increase of Hg than the previous 4 to 6 centuries. As no Hg data are available from the period between the 16th Century and ca. 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. When assessing the temporal trends of two adjacent and complementary datasets—beluga in the Beaufort Sea and ringed seal in the Amundsen Gulf, Outridge et al. (in press) concluded that Hg levels in marine biota in this region were stable from pre-industrial times up to at least the late 19th Century, with substantial increases occurring between then and 1960/61. The large 20th Century anthropogenic effect on beluga Hg occurred mostly before 1960, when man-made Hg had already attained ~75% of total Hg.

The two measures of anthropogenic contribution (baseline-corrected historic proportion of present, and percent man-made Hg), showed somewhat different results for the intermediate period between pre-

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industrial and modern times. This reflects the *chi*-squared distribution of percentage values, whereas the baseline-corrected proportion values are more closely related to relative changes in recent concentrations (see Table 2 for an idealized example contrasting the two approaches). Consequently, the percent man-made Hg formula (Eq. (3), blue line in Fig. 3) indicated a faster initial increase and then leveled off, whereas the opposite was the case for the historic proportion of present corrected for the baseline percentage (black line in Fig. 3, Table 1). In addition it was not possible to calculate the percent man-made Hg contribution for the species, regions and matrices with no pre-industrial baseline samples, as the baseline concentration was part of the formula. This means that more data points could be calculated, drawn and used for the Lowess smoother curve for the historic proportion of present corrected for the baseline percentage. It should be noted that the assumption in using the median baseline percentage value in this way is that the overall median approximates the baseline values for the species and locations missing such data.

Polar bear hair samples showed a significant average 2.1% per year increase in Hg concentrations for the period 1920 to 1991 in Northwest Greenland, and 3.1% per year increase in the period 1892 to 1973 in Northeast Greenland (Dietz et al., 2006a). Although not covering unbroken time series, feathers from West Greenland birds of prey have provided useful insight to the temporal increase of Hg over the last 150 years. Mercury in primaries of West Greenland gyrfalcons, peregrine falcons, and white-tailed sea eagles covering the period 1850–2004 increased in 7 out of 8 comparisons (3 species and 2–3 age groups), of which 4 were significant. The linear regressions on unbroken time series from the period 1880 to 1935 showed average increases in the range of 1.1–4.5% per year, and for the period 1880 to 1960 the average increase was 0.4–0.9% per year (Dietz et al., 2006b). Beluga teeth collected around Somerset Island during 1894–1998 showed [Hg] increases of 4.1- to 7.7-fold, with no change between the late 19th Century and the 1920s to 1940s, indicating that most or all of the increase had taken place after the early 20th century (Outridge et al., 2005). In East Greenland polar bear hair, the highest mean [Hg] was detected in samples from 1965 to 1974, but as no data were available from 1950 to 1965, the peak may have appeared earlier than 1965. A clear increase of Hg has likewise been found from 1835 to 1969 in primary feathers sampled from common guillemot (*Uria aalge*) and Brünnich's guillemot (*Uria lomvia*) from the Baltic and Kattegat areas, whereas levels were lower and the trend less pronounced in samples from the Faroe Islands and Greenland for the same period (Appelquist, 1985).

3.3. Preservation of the original Hg concentration in historical samples

A key factor in the correct interpretation of long-term trends of biotic Hg is ensuring that the Hg concentrations measured in historical samples are the same as when those samples were formed. The pre-industrial samples were invariably retrieved from archaeological sites, and both pre-industrial and historical samples were 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. In some cases, their storage history is unknown. Archaeological material, which may have been in contact with soil, sediment or moisture for extended periods of time, may experience physical, chemical or biochemical changes ("diagenesis"; from Jackson, 1997) that could possibly increase or decrease the original biogenic Hg concentration. Two additional preservation issues are: (a) exposure to inorganic Hg contamination in museums which employed elemental Hg sublimate as a fungicide; and (b) possible Hg volatilization from specimens over long periods of time at normal environmental temperatures. The consequences of diagenesis and inadequate preservation can be bi-directional: either over-estimation of the modern Hg increase and thus the anthropogenic contribution (i.e. when Hg was lost from the historical material) or under-estimation (when Hg contamination occurred). This section of the paper will briefly review the known mechanisms of diagenesis, and evidence for the stability of Hg concentrations in the bioarchives included in this review. Unfortunately, Hg changes during diagenesis have not been specifically studied and so discussion about this element must be extrapolated from studies on other elements.

Table 2
Idealized example showing how the two approaches to calculation of the anthropogenic contribution differ.

Year	Hg concentratiom	Historic % proportion of present	Baseline-corrected historic proportion (Eq. (2a))	% Man-made contribution (Eq. (3))
1500	0.5	5	0	0
1900	1	10	5	50
1950	5	50	45	90
2000	10	100	95	95

3.4. Diagenesis

Among calcified tissues, those with higher organic contents such as bone, dentine and cementum are generally more susceptible to diagenesis than enamel (Budd et al., 2000) because degradation of the organic component by bacterial action is typically the first stage of chemical alteration (Molleson, 1990). It follows that organic matrices such as feathers and hair are likely to be less resistant to degradation in many environments than calcified tissues in general. However, feathers and hair have been less well studied from this perspective than teeth and bones because of their central importance to archaeology. Archaeological studies of calcified tissues have shown that diagenetic processes are highly site- and matrix-specific, and their effect can vary at the micro-scale within archaeological sites because of varying amounts of water pooling around samples, the organic content of surrounding soils, presence or absence of surrounding protective tissues (e.g. teeth embedded within jaws), and degree of exposure to sunlight and freeze/thaw cycles.

The form of Hg in calcified tissues is relevant to this issue, but largely unknown. The fact that 95% of Hg in blood is MeHg (Sherlock et al., 1984), and that all of the nutrients, and major, minor and trace elements in bones and teeth are deposited from the bloodstream (Hillson, 1986), suggests that most Hg in calcified tissues occurs as methyl Hg bound with the proteinaceous fraction (Eide et al., 1994, 1995), as it does with hair and feather keratin (Bearhop et al., 2000a,b; FAO/WHO, 2003). If so, then conditions leading to rapid collagen degradation are of most concern from a Hg perspective. Collagen decomposition by microbes in calcified tissue is strongly favoured by warm temperatures, the presence of soil moisture and circum-neutral pH (Turkian and Bada, 1972; Lynch and Jefferies, 1982; Hillson, 1986; Trueman and Tuross, 2003). Preservation of collagen-containing samples like teeth should therefore generally be better in cold, dry climates such as the Arctic.

On the basis of extensive study of bones dating from the mid-Holocene to the late 18th Century, Smith et al. (2007) recognized three types of calcified tissue diagenesis: accelerated collagen hydrolysis, microbially-attacked bone, and catastrophic mineral dissolution, while Nielson-Marsh et al. (2007) described the environmental and soil conditions associated with each type. The first two types (characterized by loss of density, increased porosity, collagen decay, and microbial colonization) could substantially increase trace element concentrations in bone and teeth by increasing the internal surface area available for element adsorption, as is commonly found in obviously degraded samples from the Holocene (e.g. Trueman and Tuross, 2003). These types of diagenesis occur predominantly in wet, neutral to alkaline soils (Nielson-Marsh et al., 2007). The third diagenetic type, catastrophic dissolution, initially results in obvious damage to outer surfaces of specimens and ultimately in complete

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decomposition; this process typically occurs only in acidic, organic soils. Thus, leaching of Hg from calcified material should only occur in samples which exhibit apparent external signs of erosion, and which can therefore be easily identified and removed from a study.

Volatilization of Hg from bioarchives under natural conditions has not been studied, however, heat treatments of feathers lasting months showed that Hg volatilization under natural conditions is likely not significant (Appelquist et al., 1984). Elemental Hg(0), which is the only Hg form exhibiting marked vapour pressure at normal environmental temperatures, does not occur in significant quantities in biological tissues because of its reactivity with biological ligands such as thiols; MeHg dominates in hair and feathers (Bearhop et al., 2000a,b; FAO/WHO, 2003), which may also be the case for teeth (Eide et al., 1994). The points above suggest that over-estimation of the anthropogenic contribution because of Hg leaching or volatilization from historical specimens is unlikely. An increase of historical Hg concentrations and consequent underestimation of the anthropogenic contribution is likely if diagenesis has occurred and is not recognized. Surface abrasion and acid-washing of samples, which was a common practice to all of the long-term tooth studies discussed here, effectively removes surface metal contamination but is not always effective at eliminating subsurface contamination of more porous samples such as bones (Shafer et al., 2008).

3.5. Storage preservation

Museums are generally the main storage locations of pre-industrial specimens collected from archaeological sites and of historical era material. What potential is there for significant Hg losses or contamination from specimens in artificial storage environments?

There are few empirical studies of this question, and none concerning teeth. However, organic samples such as hair and feathers exhibit surprising resistance to alteration under various harsh treatment regimes. For up to eight months, Appelquist et al. (1984) exposed bird feathers to different treatments, including continuous UV light, heating to 100 °C, *in situ* exposure at various northern sites, and freezing. A small (<10%) decrease in total Hg content occurred within the first four months of weathering, UV and heat treatments, but nothing thereafter. Hogstad et al. (2003) reported that washing of feathers with a detergent and a preservative commonly used in museums resulted in significant losses of total Hg from some types of feathers in some species but not in others, for reasons that were unclear. Tanning of skins significantly alters inorganic and methyl Hg content of hair (Newman et al., 2005), but none of the datasets included here used tanned skins. When using museum specimens, there is a possibility that they have been contaminated by Hg-containing preservation agents, such as sublimate ($HgCl_2$) which was used as a fungicide up until the late 19th to mid 20th century. Examples of sublimate contamination of polar bear hair from Canadian and Danish museums were reported by Eaton and Farant (1982), Sirois (2001) and Dietz et al. (2006a). In situations where inorganic Hg contamination is suspected, analysis of the methyl Hg content has avoided this problem (e.g. Thompson et al., 1990).

3.6. Evidence for good preservation of historical samples

Several tests for the occurrence of diagenetic contamination of calcified tissues have been proposed which rely on a sample-by-sample evaluation (Schoeninger et al., 1989; Budd et al., 2000; Trueman and Tuross, 2003). These tests also require additional analyses beyond those required for the Hg trend study (namely, Hg concentrations, ageing, and stable isotopes). Two alternative approaches, which do not require further analytical work, include an evaluation of sample Hg-age relationships, and of C:N concentration ratios. The first test, which provides an assessment of the status of the overall group of historical samples, is based on the premise that Hg concentrations in modern animal hard tissues are almost universally correlated with age (Outridge, 2005). Assuming that sample diagenesis or Hg volatilization is independent of the original Hg concentration, the measured Hg values in the historical group should not be correlated with animal age when significant, widespread contamination or loss of Hg has occurred. When this test was applied (i.e. the beluga tooth studies by Outridge et al., 2002, 2005, in press), significant tooth Hg–age regressions were found in pre-industrial or historical sample groups, suggesting that the baseline beluga Hg data were not significantly altered over time. Mercury levels in the pre-industrial and 19th Century ringed seal teeth studied by Outridge et al. (in press) were below detection, thus invalidating this test. However, the second test involving elemental C:N concentration ratios can be applied in cases like this, and samples can be individually evaluated. 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 the Somerset Island beluga study (Outridge et al., 2005), 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. (in press), 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 $P > 0.10$).

3.7. Possible changes in species feeding behaviour over time

Another factor which could influence the interpretation of biotic [Hg] trends is the possibility that feeding behaviour (dietary trophic level and location) of the animal population also shifted significantly. One means of assessing this possibility, and of correcting [Hg] for any changes, involves the concurrent measurement of stable isotope ratios along with Hg concentrations in animal hard tissues. The measurement of abundances of naturally occurring stable isotopes of several light elements common to foodwebs (e.g. C, N, O, H, S) has emerged as a valuable tool in the environmental sciences, particularly their integration with contaminants studies in biota (Macko and Ostrom, 1994; Kidd, 1998). This advance owes its origins to the fact that foodweb 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 source of feeding and relative trophic level. Since source of feeding determines the exposure of organisms to contaminants, and trophic level influences the degree of bioaccumulation or biomagnification of contaminants in an organism's tissues, stable isotope measurements can assist in the interpretation of contaminants data to a degree previously unobtainable by more conventional techniques.

In addition to the examination of contaminants in contemporary foodwebs, the stable isotope approach is suited to better interpreting contaminant exposure and levels in historic or archaeological samples and hence to inform studies concerned with contaminant trends through time (e.g. Braune et al., 2001, 2002; Outridge et al., 2002, 2005). This is especially true for systems in which the organism of interest can change trophic level or migratory or feeding origin. For example, if [Hg] increases with trophic level and an organism shows an increasing Hg trend over a few hundred years, does this reflect actual increase in exposure or simply a temporal shift to higher trophic-level prey as a result of fundamental changes in prey availability or foodweb structure? Similarly, if a species shifted from benthic to more pelagic feeding over that time period, how would that influence tissue Hg levels?

The basic premise of stable isotope tracing of foodwebs is that the relative abundance of heavier to lighter isotopes of various elements

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Table 3
Summary of derived biomagnification factors for Hg in aquatic foodwebs.

Ecosystem	Slope	Reference
Arctic marine	0.2	Atwell et al. (1998)
Arctic marine	0.2	Campbell et al. (2005)
Arctic marine	0.23 to 0.26	Loseto (2007)
Arctic freshwater	0.19	Power et al. (2002)
Boreal freshwater	0.17 to 0.48	Kidd et al. (1995)

These values are the slope of the relationship between ln [Hg] and tissue δ^{15}N values of foodweb components.

in prey (p) tissues are passed on to the consumer (c) with slight or moderate isotopic discrimination:

$$\delta Xc = \delta Xp + \Delta\delta Xcp \quad (4)$$

where X is the heavier isotope species (e.g. ^{13}C, ^{15}N) and $\Delta\delta Xcp$ is the isotopic discrimination factor between consumer and prey tissue. This relationship is tissue specific.

Elements such as C and S generally show little to no isotopic discrimination for whole organisms between trophic levels and so are reasonably faithful recorders of source inputs. Trophic discrimination for ^{15}N is more pronounced, the result of substantial relative loss of ^{14}N due to the amination and deamination of proteins and mechanisms of voiding nitrogenous wastes. Thus, δ^{15}N measurements are typically used as indicators of trophic level and δ^{13}C and δ^{34}S measurements as indicators of source or location of feeding.

Even without the direct integration of Hg or other contaminants data, the isotopic characterization of organisms within foodwebs relative to trophic position and source of primary production can directly inform the interpretation of Hg levels in biota (e.g. Bearhop et al., 2000a,b; Nisbet et al., 2002; Morrissey et al., 2004). This is because information on basic feeding ecology is lacking for many species of interest. Many Arctic marine birds and mammals are able to exploit a wide range of invertebrate and vertebrate prey, and the stable isotope approach has revealed an often greater reliance on lower trophic-level prey than previously suspected (Hobson, 1993; Hobson et al., 1994). The same is true of the relative use of benthic vs. pelagic foodwebs by many of these organisms, behaviour that can be traced effectively using δ^{13}C measurements (Hobson, 1993; Hobson et al., 1994).

3.8. Trend analyses and stable isotopes

Ideally, we would like to correct data for [Hg] trends for any changes in trophic level through time. So, for a given organism, if we arbitrarily consider the factor by which observed [Hg] changes between two time periods (t and t') as being the temporal change factor (TCF), then we can correct this factor for changes in trophical level (TL) as derived by δ^{15}N analysis as:

$$TCF_{\text{real}} = TCF_{\text{obs}} - (\Delta TL) \times (\partial[Hg] / \partial TL) \quad (5)$$

where ∂[Hg]/∂TL is the change in [Hg] with change in trophic level. For Arctic marine systems (e.g. Hobson et al., 2002), we can also approximate ΔTL as just $(\delta^{15}N_{t'} - \delta^{15}N_t)/3.8$. So,

$$TCF_{\text{real}} = TCF_{\text{obs}} - (\Delta\delta^{15}N / 3.8) \times (\partial[Hg] / \partial TL) \quad (6)$$

This permits all [Hg] observations to be normalized in any time trend analysis but requires that the rate of change in [Hg] with trophic level be derived for any given system of interest. Several researchers have estimated the slope of the relationship between ln [Hg] and δ^{15}N in aquatic systems (summarized in Table 3) that allows approximation of ∂[Hg]/∂TL in general. However, data on how specific species bioaccumulate and biomagnify Hg is preferred for specific case histories.

We revisited the paper by Outridge et al. (2005) which reported historic changes in Hg levels in teeth of beluga whales at Somerset Island, NWT, Canada. That study determined that tooth Hg levels increased with age for both late 19th Century and mid 1990s animals but this effect was much more pronounced in the modern animals ($r^2 = 0.63$ vs. 0.17). Tooth δ^{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 TL increase of 0.18 (using the trophic enrichment value of 3.8‰ for Arctic marine foodwebs). Values for ∂[Hg]/∂TL are not well established for beluga, but Loseto (2007) estimated a total Hg biomagnification factor (BMF) of 12.8 to 16.0 for beluga feeding primarily on a fish diet. So, a trophic increase of 0.18 should correspond to a correction (subtraction) of the Hg TCF 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, [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 uncorrected data (Table 4).

The important assumption in using animal δ^{15}N values to correct Hg trend analyses is that baseline foodweb δ^{15}N values did not change during the period of interest or that other ecological or physiological processes independent of trophic level *per se* did not occur (e.g. Hobson et al., 2002; York et al., 2008). Foodweb baseline δ^{15}N stability is a reasonably robust assumption for marine foodwebs in steady state that have not undergone major upwelling or water temperature changes but few long-term datasets exist.

Temporal variation in foodweb δ^{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; O'Reilly et al., 2003). Another factor of interest in historical foodweb reconstructions using δ^{13}C is the isotopic change in atmospheric CO_2 that has occurred due to anthropogenic burning of fossil fuels. Fossil fuels are isotopically light and their burning has resulted in a decrease in atmospheric CO_2 δ^{13}C of about 1‰ (i.e. from about −7 to −8‰). Normalizing terrestrial or freshwater foodwebs 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, 2001, but see Cullen et al., 2001). Recently, based on a long-term isotopic record of teeth from northern fur seals (*C. ursinus*), Newsome et al. (2007) provided more convincing evidence that the Seuss effect has been responsible for decreases in δ^{13}C of animal tissues over that

Table 4
Changes in geometric mean Hg concentrations in teeth of Somerset Island beluga between the late 19th Century and the 1990s with and without normalization using δ^{15}N values and Eq. (5).

Sample	[Hg] 19th Century	[Hg] 1990s	Uncorrected factor increase	Corrected factor increase
20 years old	6.6	26.8	4.1	1.2–1.9
40 years old	13.4	78.4	5.9	3.0–3.7
60 years old	23	178	7.7	4.8–5.5

Analysis based on data provided in Outridge et al. (2005), except that the ages of beluga have been doubled from those presented in that reference because of a change in beluga ageing technique (Stewart et al., 2006).Teeth Hg concentrations are given as ng/g dw. The assumed increase in beluga Hg concentration and trophic level was a factor of 12.8 to 16 based on Loseto (2007).

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period. A related phenomenon is the fact that due to increases in atmospheric $[CO_2]$ the $[CO_2]_{aq}$ of oceans is increasing. This increase can change overall fractionation between dissolved bicarbonate ions and plant cells during diffusion (Michener and Schell, 1994). The net result can be a relatively small but measurable change in marine biota $\delta^{13}C$ that needs to be considered in contaminant trend analyses using this stable isotope.

Unfortunately, unlike the model used to correct for TL changes using $\delta^{15}N$ measurements, quantitative corrections to Hg trends using $\delta^{13}C$ values are not yet possible because we do not know 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., 2008).

4. Conclusions and recommendations

We have shown that wildlife hard tissue matrices provide consistent information with respect to the steep onset of Hg exposure of Arctic wildlife. This onset was determined to begin around 1850. In addition the man-made contribution to the present time Hg concentration was determined to be 92.4% ranging from 74.2 to 94.4%. Stable isotopes provide important information to normalize for possible changes in diet over time, and is highly relevant to include when interpreting temporal trends, baseline concentrations as well as man-made anthropogenic contribution of Hg. Finally, contamination can take place in hard tissue matrices either from natural sources or from storage additives such as sublimate. Ways to avoid such contamination include proper cleaning of the samples before analysis and in addition organic Hg analysis can reveal whether items have been exposed to storage conserving inorganic Hg contamination.

Similar Hg reviews should be carried out in other regions of the world as well as for other abiotic matrices and compartments of the Arctic ecosystem.

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