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Temporal trends of mercury in Arctic biota: 10 more years of progress in Arctic monitoring



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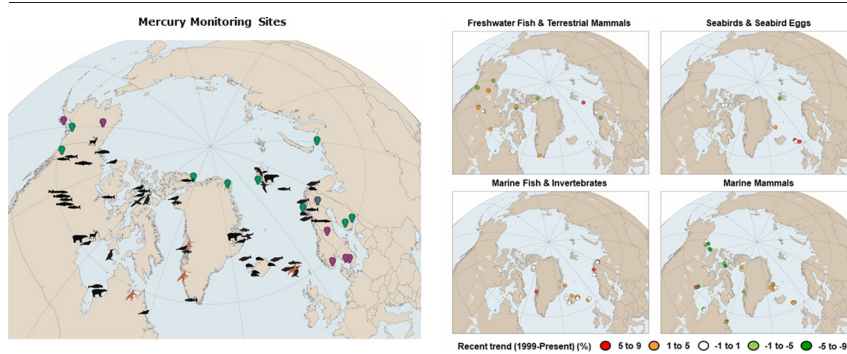
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HIGHLIGHTS

- Temporal trends of mercury levels were assessed in 110 time series for Arctic biota.
- 70% of the time series could detect a 5% annual increase with statistical power $\geq 80\%$.
- All biota groups exhibited significant trends, but few geographic patterns were observed.
- Mercury trends in seabirds appeared to increase in Arctic areas of the NE Atlantic.
- Mercury trends in marine mammals tended to be increasing at easterly longitudes.

GRAPHICAL ABSTRACT



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ABSTRACT

Temporal trend analysis of (total) mercury (THg) concentrations in Arctic biota were assessed as part of the 2021 Arctic Monitoring and Assessment Programme (AMAP) Mercury Assessment. A mixed model including an evaluation of non-linear trends was applied to 110 time series of THg concentrations from Arctic and Subarctic biota. Temporal trends were calculated for full time series (6–46 years) and evaluated with a particular focus on recent trends over the last 20 years. Three policy-relevant questions were addressed: (1) What time series for THg concentrations in Arctic biota are currently available? (2) Are THg concentrations changing over time in biota from the Arctic? (3) Are there spatial patterns in THg trends in biota from the Arctic? Few geographical patterns of recent trends in THg concentrations were observed; however, those in marine mammals tended to be increasing at more easterly longitudes, and those of seabirds tended to be increasing in the Northeast Atlantic; these should be interpreted with caution as geographic coverage remains variable. Trends of THg in freshwater fish were equally increasing and decreasing or non-significant while those in marine fish and mussels were non-significant or increasing. The statistical power to detect trends was greatly improved compared to the 2011 AMAP Mercury Assessment; 70% of the time series could detect a 5% annual change at the 5% significance level with power $\geq 80\%$, while in 2011 only 19% met these criteria. Extending existing time series, and availability of new, powerful time series contributed to these improvements, highlighting the need for annual monitoring, particularly given the spatial and temporal information needed to support initiatives such as the Minamata Convention on Mercury. Collecting the same species/tissues across different locations is recommended. Extended time series from Alaska and new data from Russia are also needed to better establish circumpolar patterns of temporal trends.

1. Introduction

The Arctic Monitoring Assessment Programme (AMAP) produces regular assessments of mercury (Hg) in the Arctic that include reviewing available information on temporal trends in Hg levels in environmental media. Consideration of temporal trends is an important element in the evaluation of the effectiveness of measures to reduce Hg emissions, such as the global Minamata Convention on Mercury. Understanding the underlying reasons for the direction and strength of temporal trends is equally important and requires knowledge of global, regional and local factors of influence. The work reported here is based on this component of the most recent (2021) AMAP Assessment of Mercury in the Arctic (AMAP, 2021), which updates an earlier review of biotic time series of mercury performed in the 2011 AMAP Mercury Assessment (AMAP, 2011). That earlier work made use of long-term datasets (i.e., those comparing modern with historical or pre-industrial Hg concentrations) to estimate the relative importance of natural and anthropogenic Hg inputs in modern biota and the environment. It also included analysis of short-term datasets (i.e., those covering the past one to three decades) which reflect more recent changes in Hg concentrations.

In the 2011 AMAP Mercury Assessment and related publication (AMAP, 2011; Rig  t et al., 2011), only 19% of the time series assessed had power metrics considered adequate to describe temporal trends with high confidence. Of the 86 trends of total mercury (THg) in the 2011 assessment, 45 were non-significant, 21 were changing in complex, non-linear patterns, 13 were increasing linearly, and only four trends were decreasing linearly (AMAP, 2011; Rig  t et al., 2011). The majority of increasing trends were found in liver of marine mammals (5, from Canada and Greenland) and freshwater fish muscle or liver (5, from Canada and the Faroe Islands) (AMAP, 2011; Rig  t et al., 2011). More recent publications have updated some of these time series of THg or added new ones, reporting temporal trends of THg in caribou (*Rangifer tarandus*; Canada) (Gamberg et al., 2020), Arctic fox (*Vulpes lagopus*; Norway) (Hallanger et al., 2019), beluga (*Delphinapterus leucas*; Canada) (Loseto et al., 2015), ringed seal (*Pusa hispida*; Canada, Greenland) (Houde et al., 2020; Rig  t et al., 2012), polar bears (*Ursus maritimus*; Canada, Norway) (Morris et al., 2022; Lippold et al., 2020), freshwater fish in Subarctic (Evans et al., 2013) and High Arctic (Hudelson et al., 2019) lakes, marine fish (Canada) (Evans et al., 2015), and seabird eggs or tissues (Canada, Greenland, Faroe Islands, Norway) (Fort et al., 2016; Foster et al., 2019; Morris et al., 2022; Tartu et al., 2022). Despite almost a decade of additional data, some recent publications still reported many non-significant temporal trends (e.g., Gamberg et al., 2020; Houde et al., 2020), as observed in the 2011 assessment (AMAP, 2011).

The lack of significance of some long-term time series of THg may be due to the myriad of complex interactions affecting Hg cycling and fate in the environment and ultimately levels in biota (see Dastoor et al., 2022; McKinney et al., 2022; Ch  telat et al., 2022; Jonsson et al., 2022; McKinney et al., 2022). Including climate-related and/or ecological factors in the analyses has identified factors of influence and in some cases strengthened or changed temporal relationships in multivariate models (e.g., Foster et al., 2019; Lippold et al., 2020; Morris et al., 2022; Rig  t et al., 2012). The effects of climate and ecological changes on Hg levels in biota are explored in further detail in McKinney et al. (2022).

Since the 2011 assessment, continued international and national efforts have generated new time series for THg for a range of species and locations in the circumpolar Arctic, and several existing biota time series have been substantially extended. Indigenous Peoples, who rely on fish and wildlife for subsistence have greatly contributed to the long-term monitoring of Hg around the Arctic (Houde et al., 2022). The increasing length (and associated statistical power) of many of the biota time series facilitated a more consistent comparison of temporal trends observed at different locations and in different species over the most recent 20-year period. In the case of the longer time series, some of which extend back to the late 1960s, the changes over the past 20 years can also be compared with those over the entire period of monitoring. These comparisons provide insight not only into trends that may be related to changing Hg emissions but also into trends that may be influenced by other changes that the Arctic is undergoing, including those directly and indirectly associated with climate warming. Wildlife and fish are exposed to Hg primarily through their diet, so environmental changes that affect Arctic food web structure and composition are particularly relevant when assessing trends in contaminants in biota over time. These factors are explored in other articles of this special issue, including changes in the deposition and concentrations of Hg in air over time (MacSween et al., 2022), emissions, transport and fate of Hg, and the impacts of climate change on abiotic and biotic systems (Ch  telat et al., 2022; Dastoor et al., 2022; Jonsson et al., 2022; McKinney et al., 2022). The temporal trends described in this article are also evaluated in relation to risk thresholds for wildlife health effects in Dietz et al. (2022), Chastel et al. (2022) and Barst et al. (2022).

The present study and trend assessment addressed three policy-relevant scientific questions: (1) What time series for THg concentrations in Arctic biota are currently available? (2) Are concentrations of Hg changing over time in Arctic biota? (3) Are there spatial patterns in temporal trends of Hg in Arctic biota? It focuses, in particular, on trends over the past two decades, evaluating these trends using a consistent statistical approach with

the objective of producing results that can be compared in a meta-analysis and considered in more detail in other articles in this issue. Given the importance of many of the species investigated here for the subsistence of Northern and Arctic Peoples, understanding how Hg levels are changing in these species is of importance to human health and food security (AMAP, 2011, AMAP, 2015; Basu et al., 2022; Houde et al., 2022). Establishing significant temporal trends in a range of species is also essential for future effectiveness evaluations of the UNEP Minamata Convention on Mercury (Evers et al., 2016), which will assess the impacts of control measures recommended through the convention on reductions of Hg in the environment, biota and vulnerable human populations including Northern and Arctic peoples (Evers et al., 2016; UNEP, 2018; UNEP, 2020).

2. Materials and methods

2.1. Available time series

Time series were available from all Arctic countries except for Russia and the USA; Alaskan seabird time series datasets were reported in the 2011 AMAP Mercury Assessment (AMAP, 2011), but had not been updated with data for recent years. The time series available for the current assessment are shown in Fig. 1 and are summarized in SI Table A1.

Biota time series of THg concentrations were assessed from data extracted from databases maintained at the AMAP marine Thematic Data Centre at ICES (www.ices.dk; mainly Icelandic and Norwegian fish and

shellfish monitoring) and additional data reported from national monitoring and research programs in Canada (www.science.gc.ca/NCP), the Kingdom of Denmark (Greenland and Faroe Islands), Norway and Sweden. Time series covered the period 1968 to 2018 inclusive and were assessed based on THg as it is broadly and relatively easily monitored.

Where appropriate and available, the time series were sub-divided by age, sex, or size (see Section 3.2.5). The 124 biota time series were filtered to ensure they met the criteria of having at least 6 years of data, with at least one of those years being in the period 2013–2018. A total of 110 biota time series were appropriate for trend assessment, ranging from 6 to 46 years of data with a mean of ~16 years (SI Table A1). The bulk of these time series focus on marine mammals (53 time series); followed by freshwater fish (19), seabirds (12), marine invertebrates (mussels; 12), marine fish (11), and terrestrial mammals (3). In addition to the analysis for trends, the time series data were examined with respect to their adequacy to detect trends with a given statistical power (Section 2.3). Some time series included in the AMAP Mercury Assessment (AMAP, 2021) were not included in the detailed results here, as separate publications are in preparation but are not yet available.

2.2. Analysis of temporal trends

Consistency in sampling and analyses are important when performing large-scale assessments such as the present investigation. Data used in the current investigation are primarily from published sources (references in

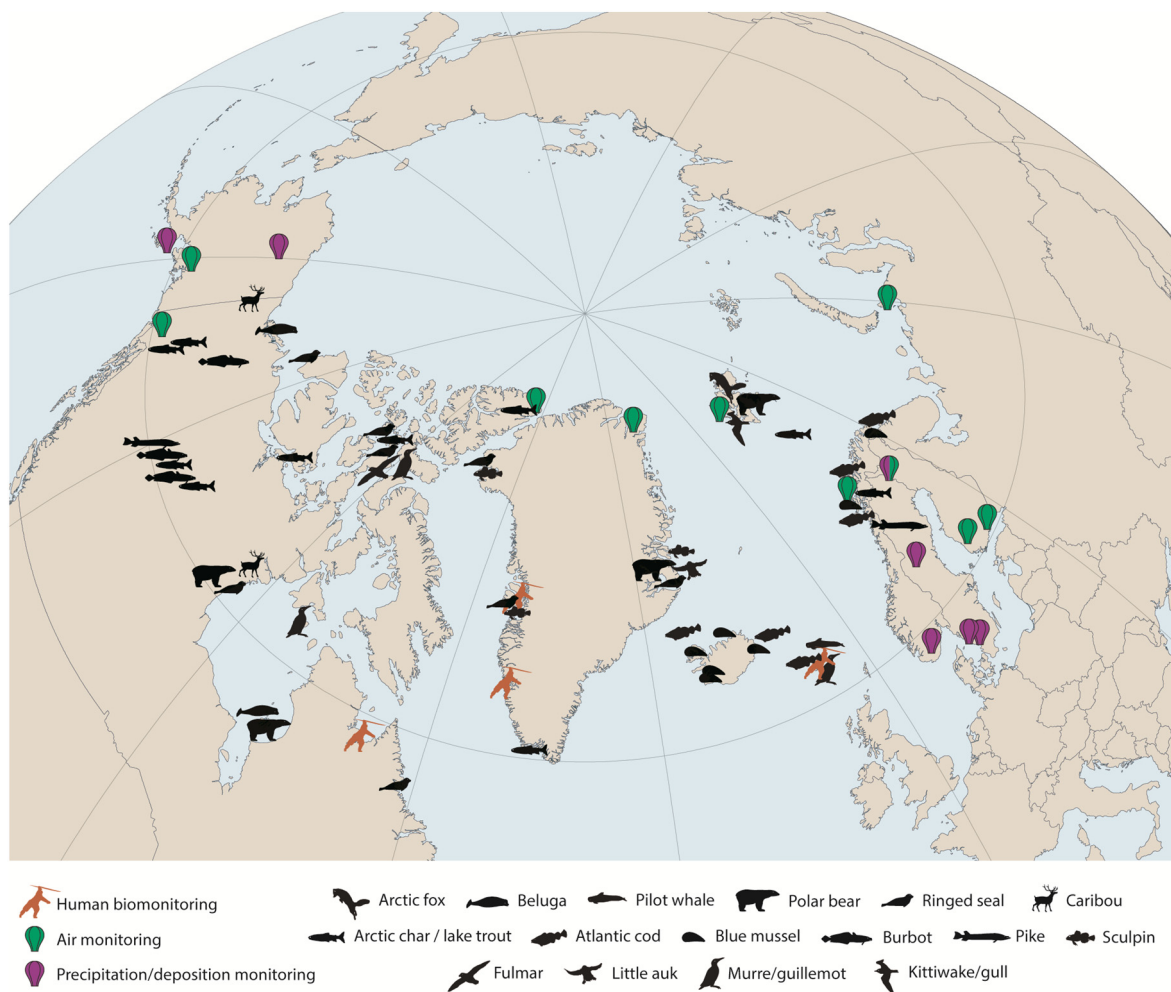


Fig. 1. Map of sampling locations for available time series of total mercury (THg) in biota included in the 2021 AMAP Mercury Assessment and the present study (see SI Table A1 for details on biota). Human biomonitoring, air and precipitation/deposition sites are described in other articles in this special issue. Figure reproduced with permission from AMAP (2021), with modification.

SI Table A1), or are extensions of these previously published time series, that were subject to stringent quality assurance-quality control. As part of upcoming effectiveness evaluations of the Minamata Convention, a monitoring guidance document has also been developed that will ensure that Hg monitoring data will be comparable and of good quality. This guidance document will be made public once complete, but it is not yet available (see www.mercuryconvention.org for more information).

Temporal trends can be calculated using a wide range of statistical methods, however if the goal is to compare trends across spatiotemporal scales, using a consistent statistical method is also crucial as it eliminates bias introduced by the variation in methods. Temporal trends of Hg in biota were analyzed using mixed models based on individual log_e-transformed THg concentrations (μg/kg). The analyses used a dedicated statistical application (AMAP, 2020) coded in the R statistical environment (R Core Team, 2020). This application has evolved from a method originally developed by Fryer and Nicholson (1999) and has also been used to analyse contaminant time series in biota, sediment and water under the OSPAR (<https://www.ospar.org>) and HELCOM (<https://helcom.fi/baltic-sea-trends/data-maps/>) Programmes. The current approach was similar to that used in previous AMAP assessments, which employed the PIA statistical application (Bignert et al., 2004; Rigét et al., 2011). However, it modelled individual concentrations rather than annual medians and used smooth terms rather than 3-years running means to evaluate the degree of non-linearity. A full description of the methods can be found in AMAP (2020).

Two key components of the analysis were the selection of a parsimonious model that adequately described the pattern of change over time, and the construction of a summary metric that allowed patterns of change to be compared across time series. To assess the pattern of change, a model with a (log_e-)linear trend:

$$\log_e \text{ THg concentration} \sim \alpha + \beta \text{ year} \quad (1)$$

was fitted to the data, where α is the intercept and β is the slope of the fitted line. This model was then compared with a model with a non-linear trend:

$$\log_e \text{ THg concentration} \sim s(\text{year}) \quad (2)$$

in which log_e THg concentrations varied smoothly (and non-linearly) over time. Smoothers (s) were fitted on up to 4 degrees of freedom depending on the length of the time series (as described in AMAP (2020)). The selected model was that with the lowest Akaike Information Criterion corrected for small sample size (Burnham and Anderson, 2002). The significance of the fitted trend (either linear or non-linear) was assessed by a likelihood ratio test.

The metric used to summarise the pattern of change in each time series was the annual percent (%) change in concentration in the most recent 20-year period (1999–2018) which, for convenience, is referred to as the ‘recent trend’. The metric was chosen because it focussed on a period covered by all the time series (only 29 of the 110 time series started after 1999) and because it allowed meaningful comparisons between time series with linear and non-linear trends. When the fitted trend was linear, the recent trend was calculated as:

$$\text{recent trend (\%)} = 100 \left(\exp \left(\hat{\beta} \right) - 1 \right) \quad (3)$$

where $\hat{\beta}$ was the estimate of β (slope of the log_e-linear trend) in Eq. (1). The exponentiation in Eq. (3) back-transforms the annual absolute change in log concentration, measured by β , to the annual percentage change in concentration. When the fitted trend was non-linear, the recent trend was calculated by taking the change in concentration between the start and end of the 20-year period and converting it to the equivalent annual percentage change had the trend been linear. Specifically:

$$\text{recent trend (\%)} = 100 \left(\exp \left(\frac{\hat{s}(2018) - \hat{s}(1999)}{2018 - 1999} \right) - 1 \right) \quad (4)$$

where $\hat{s}(2018)$ and the $\hat{s}(1999)$ were the fitted log concentrations in 2018 and 1999 respectively (see Eq. (2)). Where time series began after 1999 or ended before 2018, the start and end years were adjusted accordingly.

As well as assessing the significance of the linear or non-linear trend over the whole time series (using the likelihood ratio tests mentioned above), it was also possible to assess the significance of the recent trend. For linear trends, the significance levels were identical, since they were both based on $\hat{\beta}$. For non-linear trends, the significance levels differed depending on the pattern of change. For example, it would be possible to have a significant non-linear pattern of change over the whole time series, but a non-significant recent trend if, for example, concentrations declined in the early part of the time series (before 1999) and then stabilised.

2.3. Power analysis for biota

The previous 2011 AMAP Mercury Assessment (AMAP, 2011; Rigét et al., 2011) included statistical power analyses that assessed whether the time series available at the time could detect statistically significant trends ($p < 0.05$) with power greater than or equal to (\geq) 80%.

The previous assessment concluded that additional years of data added through continued monitoring would improve the ability of the AMAP programme to detect trends at greater statistical significance and power. Such improvements, along with the inclusion of new time series, were expected to yield an increased number of interpretable trends in future assessments. Therefore, a power analysis was repeated in the 2021 AMAP Mercury Assessment to evaluate how well the AMAP monitoring program for THg in biota can now detect significant trends, and how much it has improved since 2011.

The power to detect a trend in a time series depends on the magnitude and pattern of the trend, the number of years of data and the number of samples collected each year, whether the series is sequential or contains gaps, the magnitude of the variance components in the data, the test used and the significance level of the test. The current investigation considered the power to detect a log-linear change in concentration using a two-sided F-test at the 5% significance level ($\alpha = 0.05$). In particular, the following metrics were calculated for each time series:

- Power to detect a 5% annual increase given the current configuration of years.
- Lowest annual increase detectable with power $\geq 80\%$ given the current configuration of years.
- Power to detect a 5% annual increase given 10 sequential years of monitoring.
- Lowest annual increase detectable with power $\geq 80\%$ given 10 sequential years of monitoring.

Full details of the power calculations can be found in AMAP (2020).

In the current assessment, 77 of the 110 time series (70%) had (statistical) power $\geq 80\%$ to detect a 5% annual increase with their current configuration of years, meeting or exceeding the specified criteria for detection of trends (Fig. 2). This represents a major improvement over the previous assessment (AMAP, 2011), where only 16 of the 84 time series (19%) had power $\geq 80\%$ (recalculated from Rigét et al., 2011). The improvement in power was also evident in the lowest annual increase detectable with $\geq 80\%$ power, which had a median value of 3.3% in the current assessment compared to a value of 11% in the 2011 assessment (recalculated from Rigét et al., 2011) (Fig. 2).

Much of the improvement in power is likely due to the additional years of monitoring, which have extended existing time series from a mean length of 11 years in the previous assessment (AMAP, 2011) to 16 years in the current assessment. The addition of new time series with high power (seabirds, mammals) has also contributed to this improvement (SI Table A1).

2.3.1. Implications of the power of trend detection for AMAP monitoring

The selection of species and tissue for monitoring temporal trends in THg concentrations in biota involves considerations and objectives that

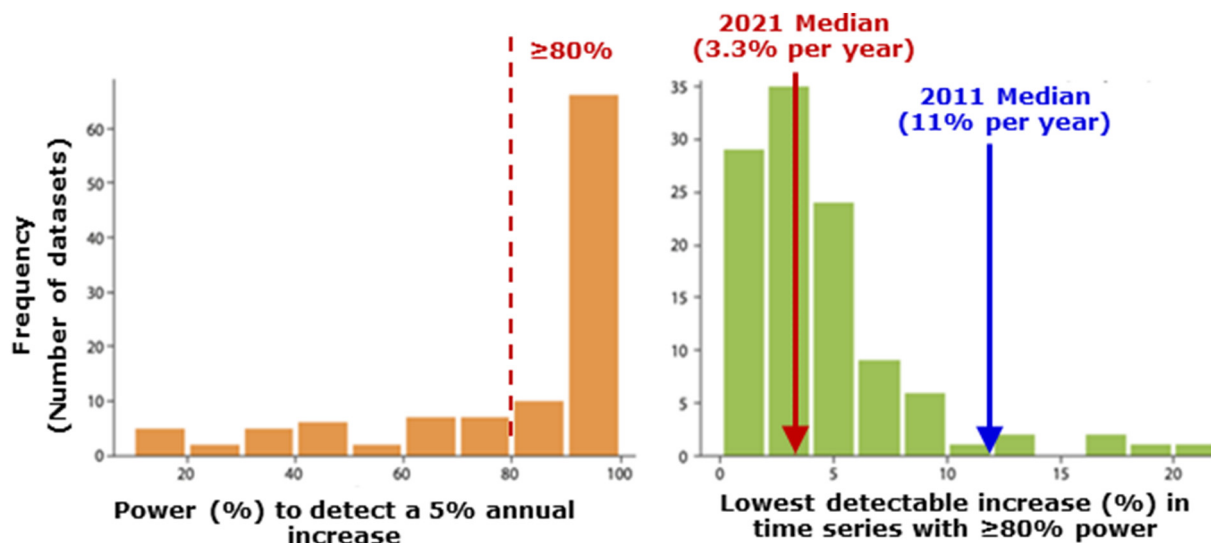


Fig. 2. Histogram of the power to detect a 5% annual increase ($\geq 80\%$ power indicated by dashed line, left panel) and the lowest annual increase detectable with $\geq 80\%$ power (right panel) given the current configuration of years in the available THg time series. Figure reproduced with permission from AMAP (2021), with modification.

are often decided at the national or institutional level. These include trend detection for purposes such as the assessment of health of threatened species, ecosystem state, effectiveness of contaminant regulation and interactions between climate change and pollutant exposure. Practical considerations include funding limitations, species availability, the collection of samples under challenging Arctic conditions, and the maintenance of monitoring efforts over a long period (decades). Table 1 presents the power of the available time series for each animal group/tissue assuming 10 years of sequential annual monitoring were available. This calculation allows power to be compared between groups/tissues while ignoring the effect of different numbers of years and gaps in time series when there was no sampling. In the current assessment, time series based on seabird eggs had the greatest power but are also the least represented group. Seabirds are used in trend monitoring studies across the Arctic, including retrospective trend studies using archived samples. Species selection is an important consideration as some species are migratory, while others are resident and overwinter in the Arctic and different species occupy different trophic positions. Monitoring using bird eggs generally assumes that Hg concentrations represent the uptake in the adult birds in the local (breeding) area and not areas outside the breeding season (e.g., wintering areas). However, contaminant levels are likely affected by contamination in wintering areas in (partly) capital breeders such as common eider (*Somateria molissima*) (Sénéchal et al., 2011).

Invertebrates (mussels) provided powerful time series for THg trend evaluation (Table 1). However, their spatial coverage in the current assessment was limited to Norway and Iceland and, despite the high power, few significant trends were detected. Mussels, a sessile filter-feeding organism,

Table 1

Mean statistical power (standard deviation in parentheses) and number of time series by animal group/tissue to detect a 5% annual increase in total mercury given 10 sequential years of monitoring at the 5% significance level. Only group/tissue combinations with 5 or more time series are shown. See SI Table A2 for individual results. Table reproduced with permission from AMAP (2021).

Animal group	Tissue	Power (%)	Number of time series
Seabirds	Egg homogenate	68 (12)	5
Invertebrates (mussels)	Soft body tissue	52 (31)	12
Freshwater fish	Muscle	49 (24)	18
Marine mammals	Muscle	48 (23)	15
Marine mammals	Hair	31 (9)	6
Marine fish	Muscle	29 (16)	8
Marine mammals	Liver	19 (13)	30

provide valuable spatio-temporal information on contaminant distributions and trends. Establishing circumpolar bivalve monitoring could be a cost-effective approach to fill geographical gaps in THg monitoring but, given that several long and powerful time series did not produce significant, interpretable temporal trends, mussels may not be ideal for this purpose. Both seabird eggs and mussels are relatively easy to sample in a standardized manner (e.g., with respect to time of sampling), though care is needed if sampling from small populations or endangered species. Powerful time series were also detected for marine mammal (muscle) and freshwater fish, the latter notably more powerful than the time series for marine fish (Table 1). Trends in these animal groups are particularly important to monitor as they are important traditional foods for Northern and Arctic Indigenous Peoples, and some high trophic level marine mammals have levels that may be a concern for toxicological effects in the wildlife (Dietz et al., 2022).

The biota time series presented here will provide globally relevant insights regarding the effectiveness of measures taken to reduce and eliminate anthropogenic Hg emissions through UNEP's Minamata Convention on Mercury. As general indicators of ecosystem state, time series would ideally represent a range of ecosystem components and animal groups, taking into account potential confounding factors. Some of these factors include local sources of pollution for example, or direct and indirect influences of climate change on processes that affect exposure and uptake by biota, including changes in the structure of ecosystems, migration patterns, etc. These are addressed in more detail in Jonsson et al. (2022) and McKinney et al. (2022).

3. Results and discussion

3.1. Are concentrations of mercury changing in Arctic biota over time?

The 2011 AMAP Mercury Assessment (AMAP, 2011) evaluated around 80 biota time series using a consistent statistical methodology. Newly available data have extended many of these time series and added new time series, though other time series were discontinued or lacked new data and were not included in the current assessment. The increasing length and power of many of the time series facilitated comparison of trends over a synchronous time-period, with emphasis placed on changes observed over the past 20 years. Based on the updated data and statistical analyses, the question of whether concentrations of THg in the Arctic are changing over time has been re-evaluated and compared to the conclusions made in 2011 (AMAP, 2011), and will be discussed in the following subsections.

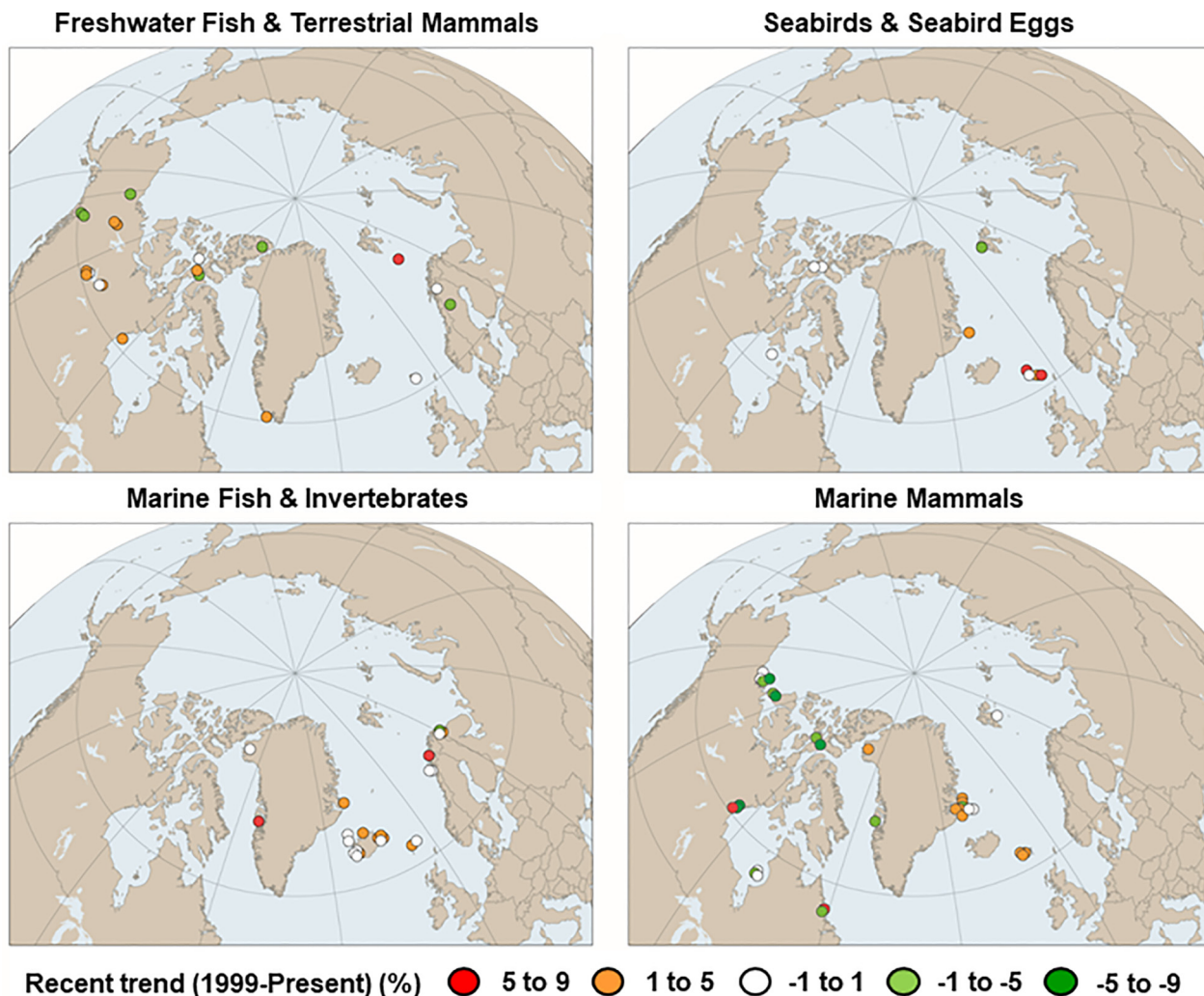


Fig. 3. Geographical patterns in recent (last 20 years) trends of THg concentrations in tissues of biota, where trends were significant and/or time series were of a statistical power $\geq 80\%$. The figures show freshwater fish and terrestrial mammals (top left); seabirds (top right); marine invertebrates (mussels) and marine fish (bottom left) and marine mammals (bottom right). Overlapping symbols have been dispersed to make them visible, resulting in clusters of points around some monitoring locations where trends are available for multiple time series (see SI Table A2). Figure reproduced with permission from AMAP (2021), with modification.

3.2. Recent trends of mercury in biota

Results of the statistical analysis of the 110 time series for the current assessment are summarized in SI Table A2, with significant and/or high power ($\geq 80\%$) trends also shown in Fig. 3. The desired criteria of having a statistical power of $\geq 80\%$ to detect a 5% annual increase in THg concentrations was met by 77 time series. Of these, 37 showed a significant trend

($p < 0.05$) of which 18 were non-linear and 19 were linear. Focussing on the past 20 years, 44 time series had a positive recent trend of which 17 were significant, and 32 had a negative recent trend of which 14 were significant (one time series had a recent trend of $\sim 0\%$). Table 2 summarises the results for the 77 time series with adequate power ($\geq 80\%$) by animal group. All animal groups included time series with both increasing and decreasing recent trends, with median recent trends ranging from 0.0% to 1.1% per year.

Table 2

Summary of trend results by animal group for the time series with statistical power $\geq 80\%$: the number of time series, the number of nonlinear and linear significant trends, the number of decreasing and increasing recent trends (with significant recent trends in parentheses) and the minimum, median and maximum recent trend (% change per year). Table reproduced with permission from AMAP (2021).

Species group	# of time series	# of Significant trends (full time series)		Recent trend (% change per year)				
		Nonlinear	Linear	Decreasing	Increasing	Minimum	Median	Maximum
Invertebrates (blue mussels)	12	3	1	6 (1)	6 (2)	-2.4	~ 0.0	2.2
Marine fish ^a	8	3	1	1 (0)	6 (2)	-0.3	1.1	4.0
Freshwater fish	18	5	8	7 (6)	11 (6)	-3.3	0.8	5.3
Seabirds	9	1	5	4 (1)	5 (4)	-3.1	1.0	6.1
Marine mammals	28	6	4	13 (6)	15 (3)	-8.6	0.2	2.8
Terrestrial mammals	2	0	0	1 (0)	1 (0)	-1.0	0.8	2.6
All species	77	18	19	32 (14)	44 (17)	-8.6	0.6	6.1

^a One marine fish time series had a recent trend of $\sim 0\%$.

The most rapid decline (-8.6% per year) was in small size group beluga whales (*Delphinapterus leucas*) in the Eastern Beaufort Sea, and the fastest increase (6.1% per year) was in Faroese black guillemot (*Cepphus grylle*).

3.2.1. Invertebrates

Blue mussel (*Mytilus edulis*) time series ($n = 12$) were available from several locations in Iceland and northern Norway. These data are reported annually to the AMAP marine thematic data centre at the International Council for the Exploration of the Sea (ICES) and were extracted from the ICES databases (ICES, 2020). One statistically significant decreasing recent trend of THg was observed in Skallneset, in northern Norway (-2.4% per year), and two significantly increasing recent trends were detected at two stations in Mjóifjörður fjord in eastern Iceland (1.5% and 2.0% per year; Fig. 3; SI Table A2). These results confirmed that THg continued to increase in blue mussels from eastern Icelandic stations, where the only significant trend in mussels was observed in the 2011 AMAP Mercury Assessment (AMAP, 2011; Rigét et al., 2011), although the previous trend was observed at a different station in Mjóifjörður. Two of the recent trends were part of more complex non-linear trends over the whole time series (SI Table A2, SI Fig. A1). At Skallneset, concentrations were stable and then decreased (from about 2007) and at one Mjóifjörður station, concentrations first decreased before increasing in the latter part of the time series (from about 2005). A third non-linear trend was detected at Husvågen, Norway, where concentrations decreased and then increased, with concentrations at the end of the time series similar to those at the start.

No recent temporal investigations of THg in blue mussels from these northern regions are available in the literature for comparison. Despite increasing trends of THg in some Icelandic mussels, the concentrations in Arctic bivalves remain low and are not likely of toxicological concern to the organisms (Barst et al., 2022). As sessile, low trophic level filter-feeders, bivalves (like other invertebrates) accumulate the toxic form of Hg, methylmercury (MeHg) (Braune et al., 2015) and, since they are monitored throughout Europe, are convenient for spatial comparisons of environmental pollution. However, even with relatively long time series (15–22 years) of adequate power, THg concentrations in mussels produced few significant trends. The lack of significance, conflicting results, and narrow geographic distribution of the Arctic time series of Hg in mussels mean that no general trend of THg has been observed for blue mussels.

3.2.2. Fish

3.2.2.1. Marine fish. Eleven time series in three species of marine fish, short-horn sculpin (*Myoxocephalus scorpius*; $n = 3$), Atlantic cod (*Gadus morhua*; $n = 7$) and sea-run Arctic char (*Salvelinus alpinus*; $n = 1$; char were primarily sampled in late summer during migration from the sea) (Evans et al., 2015) provided information on THg trends across the circumpolar Arctic, with locations in Canada, Greenland, the Faroe Islands, Iceland, and Norway (SI Table A1).

There were four significant recent trends in THg in marine fish, all of which were increasing (Fig. 3; SI Table A2). They were in the liver of sculpin from central East and central West Greenland (2.1% and 6.1% per year, respectively), and in the muscle of Atlantic cod from the northwest Faroe Islands (of “undefined” sizes) and northern Norway (3.9% and 7.4% per year, respectively). However, the significant Norwegian cod trend should be interpreted with caution as the time series spanned less than 10 years with only seven time-points, and the power was $<80\%$ (Fig. 3; SI Table A2). Three time series showed significant non-linear trends over the full range of years, all in the muscle of Atlantic cod; two from the Faroe Islands and one from northern Norway (SI Fig. A1; note the recent trend based on the non-linear pattern was only significant in one of the Faroese time series). In all three, concentrations decreased until about 2000, increased until about 2010 and then either plateaued or decreased once more (although more time-points are required to confirm the most recent decrease). The overall reduction in THg concentrations between the start and end of the time series was about 70% in the two Faroese time series (1979–2016 or 2017) and 50% in the Norwegian time series

(1994–2017). No other time series showed a significant trend. The range of time series for marine fish in the 2011 AMAP Mercury Assessment was limited (i.e., from the early 1990s to the early- or mid-2000s) with only two significant linear trends, both of which were decreasing levels in Atlantic cod from the Faroe Islands and Iceland (AMAP, 2011; Rigét et al., 2011).

The THg time series for sea-run char from Cambridge Bay (Queen Maud Gulf, Canada) did not show a significant trend in the current assessment and was not included in the 2011 AMAP Mercury Assessment for comparison. These char were generally caught annually by the domestic fishery in late summer on their return from the sea, though some may have been sampled later after their return to freshwater though they were confirmed to be fish that had recently returned from the sea. In previously published models, with fork length or body condition factor included as covariates, concentrations of THg in Cambridge Bay char were reported to be decreasing significantly between 2004 and 2013, although when assessed from 1977 an increasing trend was observed (Evans et al., 2015). The decreasing concentrations in the most recent time frames were attributed to warmer springs resulting in greater primary production, faster fish growth rates, improved condition factor and mercury biodilution (Evans et al., 2015). Trend analyses also were conducted from other sea-run char locations in Nunavut and Nunatsiavut in northern Canada and while some positive and negative trends were detected (Evans et al., 2015), the data base (years of sampling) was too small to have confidence in the results and these time series were not extended for re-analysis in the present study. Several potentially influential environmental and climate factors on the levels of Hg in the environment and biota are discussed in Jonsson et al. (2022) and McKinney et al. (2022).

All of the recent 20-year trends in marine fish were non-significant or significantly increasing. Comparisons between species are not discussed as they are complicated by differences in tissue analyzed and ecological niche, as they include benthic (sculpin), benthopelagic (Atlantic cod, char) and anadromous (char) fish. Circumpolar or national comparisons of trends among fish can be confounded by proximity to source regions (i.e., sources of THg emissions and/or deposition), differences in the bio-availability of THg at the base of the food web (particularly MeHg), species-specific differences in bioaccumulation, as well as the type, composition, and length of the organism's food web (Kirk et al., 2012). Reports of climate-related effects and their potential influence on THg levels are rarer for Arctic marine than for freshwater fish (see McKinney et al., 2022). Changes in productivity and related rates of methylation in marine systems as well as alterations to the structure of food webs have been identified as drivers of differences in THg across marine locations (Wang et al., 2018; Jonsson et al., 2022; Chételat et al., 2022; McKinney et al., 2022).

Broader collections of sea-run char from other circumpolar locations could be informative as the fish are widely distributed and are routinely fished for subsistence. Sculpin are also circumpolar and do not migrate long distances (Harley et al., 2015) and so could be monitored elsewhere for useful comparisons with Greenlandic populations. The concentrations of THg were increasing relatively rapidly in sculpin and Atlantic cod from West Greenland and Norway (respectively) from around 1999 to present as discussed above, and should continue to be monitored with high priority, as should other locations, to generate longer and more comparable trends. However, given the relatively low concentrations of THg and the low toxicological risk calculated for Arctic marine fish (Barst et al., 2022), these may be of relatively low priority for expanded monitoring.

3.2.2.2. Freshwater fish. Freshwater populations of landlocked Arctic char ($n = 9$) were available for comparison throughout northern Canada (Ellesmere and Cornwallis Islands, Nunavut) (Hudelson et al., 2019), Southwest Greenland, the Faroe Islands, Norway (Bjørnøya) and Sweden (SI Table A1). There were significant decreasing recent trends in THg levels in char from the two most northern Canadian lakes (Amituk and Hazen, both -3.3% per year), and in Lake Abiskojaure in northern Sweden (-0.90% per year) (Fig. 4). Conversely, there was a significant increasing recent trend of 5.3% per year in char from Lake Ellasjøen on Bjørnøya. The trend in Amituk Lake was non-linear and suggested that, although

THg concentrations were significantly lower at the end of the time series compared to the start (2001), concentrations have increased since 2011 (Fig. 4). An increase in THg concentrations since 2013 was also suggested by the non-linear trend in char from Resolute Lake in Canada, although concentrations at the end of the time series were similar to those at the start (1993) (SI Fig. A1). In both time series, more data are required to validate the possible increases in the last few years. There were no significant trends in Greenlandic or Faroese char.

Time series available for muscle of lake trout (*Salvelinus namaycush*) ($n = 4$) from northwest Canada showed significant, decreasing recent trends in THg levels in of -1.9% and -2.5% per year in two lakes in the Yukon (Laberge and Kusawa respectively) (SI Table A2). Conversely, there was a significant increasing recent trend in lake trout in the west basin of Great Slave Lake (Northwest Territories, Canada), but no significant trend in lake trout from the east arm of Great Slave Lake (AMAP, 2021). Time series were also available for concentrations of THg in muscle and/or liver of burbot (*Lota lota*) from the west basin and east arm of Great Slave Lake (Evans et al., 2013) and the Mackenzie River (Fort Good Hope, Canada) (Carrie et al., 2010) ($n = 4$) and all showed significantly increasing recent trends (Fig. 3; see AMAP, 2021 for detailed results). Northern pike (*Esox lucius*) were not broadly monitored ($n = 2$): there was one significant decreasing recent trend in Lake Storvindeln, Sweden (-2.1% per year) and no significant trend in pike from the west basin of Great Slave Lake (Fig. 3; AMAP, 2021). The full THg time series for trout from Lake Laberge and pike from Sweden showed non-linear trends in which concentrations first increased, peaking in the mid-2000s or early 1990s respectively, and then decreased as indicated by the recent trends (SI Fig. A1; SI Table A2).

Most of the time series for the freshwater fish added ~ 10 years to those analyzed in the previous AMAP mercury assessment (AMAP, 2011; Rigét et al., 2011), greatly improving the statistical power of the trends (see Section 2.3) and providing new insights. This is the first report of decreasing THg trends in lake trout from the Yukon (Gamberg et al., 2020) as well as the most recent assessment available for trends in Faroese and Swedish char, updating some existing time series (AMAP, 2011). The increasing concentrations in Canadian burbot (AMAP, 2021) and the decreasing trends in Lake Hazen char (Fig. 3) were consistent with those described in the last Hg assessment (AMAP, 2011). However, the previously described increasing trends in char from Lake á Mýrunum (Faroe Islands) and Char Lake (Canada) (AMAP, 2011) were no longer significant. These analyses previously spanned only seven to eight years, which is a short trend to interpret with confidence, and thus more credence can be given to the trends from the current assessment.

Temporal trends were recently described in some of the same populations of Cornwallis Island (Canada) landlocked char and some results were comparable to those here, although length-normalization resulted in some different trends. For example, Hudelson et al. (2019) observed significantly decreasing concentrations in both Amituk and Resolute lakes, which was not the case here though a non-linear pattern was identified in Resolute Lake. Technical reports have also described decreasing trends of THg in Lake Hazen (Canada) and in some of the time series from lakes on Cornwallis Island (Muir et al., 2021), that are consistent with the results described here. The trends described in the recent assessment (AMAP, 2021) were consistent with increasing temporal trends previously described through multivariate models (including length as a covariate) for trout and burbot from Great Slave Lake (up to 2012; Evans et al., 2013), and THg was also increasing in more complex models in burbot from Fort Good Hope (Canada), the most recent publication available for these data (Carrie et al., 2010).

Regional and habit-specific differences are key drivers of variation in THg trends in northern lake fish (Chételat et al., 2015). In Canadian High Arctic lakes with minimal inputs of terrestrial organic carbon such as those on Cornwallis and Ellesmere Islands food web energetics are driven by benthic algal production (Chételat et al., 2010). Methylmercury bioaccumulation and the resulting concentrations in aquatic invertebrates (the primary food source of the char) have been shown to be inversely related to dissolved organic carbon (DOC) in lakes across a latitudinal gradient,

with higher bioaccumulation observed in lakes with low DOC due to higher MeHg bioavailability (see McKinney et al., 2022; Chételat et al., 2018). In-lake water chemistry, including particulate organic carbon (POC) and/or DOC levels, were also shown to be more influential on levels of Hg in Arctic char than broader watershed variables in High Arctic lakes (Hudelson et al., 2019; McKinney et al., 2022). Increases in algal-derived organic matter linked to higher temperatures due to climate-related warming have also been hypothesized to drive the increases in burbot at Fort Good Hope (Carrie et al., 2010).

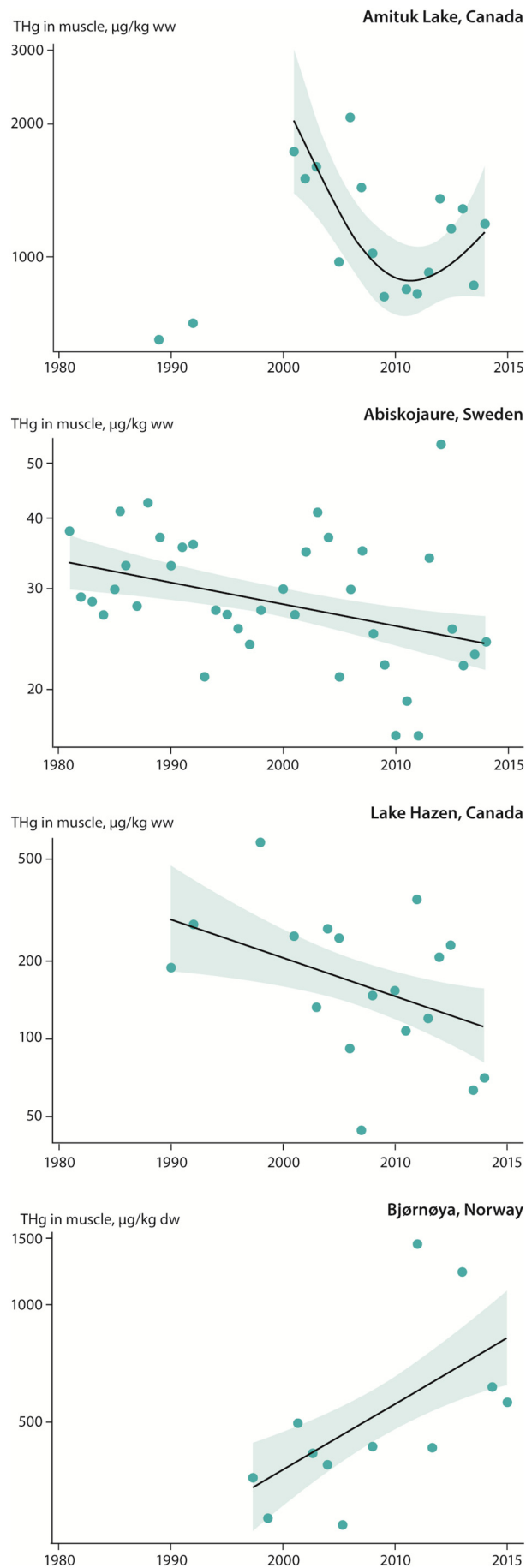
Climate variables that significantly relate to concentrations of Hg in freshwater fish have been identified for several time series (McKinney et al., 2022). These include, for char, the duration of ice cover, precipitation levels and the phase of the North Atlantic Oscillation Index (NAO) (Hudelson et al., 2019). Also, air temperatures, and the Pacific North American Pattern (PNA) in fish from Great Slave Lake (Evans et al., 2013); and air temperatures for burbot from Fort Good Hope (Carrie et al., 2010) as well as for Arctic char from Southwest Greenland (Rigét et al., 2010). Further inclusion of relevant climate/weather, biological and ecological factors as covariates in time series analysis may affect the trends and may improve significance of the time series and help explain variation in THg over time and between locations, as well as give important insight into drivers of the observed trends (McKinney et al., 2022).

The concentration levels and related toxicological risk of THg to the fish varied considerably between freshwater fish populations; however, several have high proportions of individuals in the moderate to high-risk categories (Barst et al., 2022). Further monitoring is required to determine the change in risk to the fish populations over time.

3.2.3. Seabirds

Total Hg concentrations in circumpolar seabird populations produced 12 time series in five species for comparison across four distinct geographical regions: little auk (*Alle alle*; $n = 2$) in Greenland (Fort et al., 2016), black guillemot (*Cepphus grylle*; $n = 6$) in the Faroe Islands, northern fulmar (*Fulmarus glacialis*; $n = 1$) and thick-billed murre (*Uria lomvia*; $n = 2$) in Canada (Foster et al., 2019; Morris et al., 2022), and black-legged kittiwake (*Rissa tridactyla*; $n = 1$) in Norway (Tartu et al., 2022) (SI Table A1). Significant increasing recent trends in THg were greatest in the Faroe Islands in the liver of black guillemot collected at Sveipur (6.1% per year) and in their eggs collected at Koltur and Skúvoy (4.6% and 5.9% per year, respectively) (Fig. 3; SI Table A2). A more modest significant increasing recent trend was found in the eggs of northern fulmar at Prince Leopold Island in the Canadian High Arctic (1.0% per year). There was a non-linear trend in thick-billed murre eggs at Prince Leopold Island, which showed concentrations increasing from 1975 to around 2000 and then remaining stable (SI Fig. A1). Finally, there was a significant decreasing recent trend in red blood cells of Norwegian black-legged kittiwakes in Svalbard (-3.1% per year) (Fig. 3; SI Table A2). All other trends in seabirds were non-significant.

Trends in THg in Arctic seabirds are consistent with those published in the last AMAP mercury assessment (AMAP, 2011; Rigét et al., 2011). Additional years of data have improved the power of the trend analysis in most of the time series, though two black guillemot datasets (for feathers and liver) from the western Faroe Islands remained well below the desired power threshold of 80% (SI Table A2; Section 2.3). Though the trend for the black-legged kittiwakes at Svalbard was decreasing from 2000 to 2016 (SI Table A2), a very recent publication has described a U-shaped function for the same time series with an additional 3 years of data, with the concentrations increasing from 2010 through 2019 after decreasing from 2000 to 2010 (Tartu et al., 2022). The plateaued or slightly decreasing THg concentrations in northern fulmar and murre eggs after 1999 in the Canadian data series appear to reflect similar atmospheric trends (Braune et al., 2016). It could also be argued that changes in THg concentrations may reflect changes in diet composition and foraging habitats as prey fish exposure to THg varies with depth in the water column (and between species; reviewed in Braune et al., 2016). Changing sea-ice and oceanographical conditions might force seabirds to relocate their foraging grounds or



adjust consumed prey items (Grémillet et al., 2015; Vihtakari et al., 2018). However, changes in prey composition may not be reflected by a change in trophic position alone and could drive various THg or MeHg exposure scenarios in the marine environment.

The THg monitoring dataset for seabird eggs from Prince Leopold Island was analyzed in the context of environmental and climate change indices in a separate study (Foster et al., 2019). For both fulmars and murres, THg was related to NAO and temperature and (for murres) snowfall, while the fulmar model also included sea-ice; all variables had time lags of two to seven years. Morris et al. (2022) also described similar relationships in thick-billed murres from Coats Island (Canada), though consistency with published trend directions varied with length of the time series. The climate/weather factors of influence on THg trends in seabirds are relatively consistent with those identified for mammals (McKinney et al., 2022). Covariation of sea-ice cover and THg levels was also found for Greenland little auks and affected the body condition of adult birds as well as the growth rates of chicks, providing important indicators of health. However, the THg concentrations were, and largely remain, below known toxicological thresholds of concern (Amélineau et al., 2019; McKinney et al., 2022; Chastel et al., 2022).

3.2.4. Terrestrial mammals

Only three THg time series were available for terrestrial mammals. THg concentrations measured in kidneys in the Porcupine and Qamanirjuaq caribou herds from northern Canada (Gamberg et al., 2020) and in the liver of Arctic foxes from Svalbard (Hallanger et al., 2019) did not show any significant temporal trends (Fig. 3; SI Table A2). The caribou time series had adequate power indicating that a statistically significant trend would likely be detected if present, while that of foxes was below the desired 80% (40%) (SI Table A2; Section 2.3). Temporal trends in THg were also not detected in separated male and female and/or adult and immature caribou time series (data not shown). In the previous AMAP Mercury Assessment, shorter time series from the Porcupine herd (northwest Canada/Alaska; 1994–2007) also failed to produce significant trends, though one decreasing linear and one non-linear trend were detected in the muscle and liver of reindeer from Sweden (early 1980s/mid-1990s–2005) (AMAP, 2011). No additional data were available for the Swedish reindeer to extend that time series here.

Recent reports on the Porcupine and Qamanirjuaq caribou (that included the data analyzed here) also concluded that age-normalized THg trends were not significant (Gamberg et al., 2020; Morris et al., 2022). Dietary differences may be less influential on Hg variation in different caribou/reindeer herds across Canada and Greenland than environmental factors, which affect the movement and deposition of Hg to lichens, a key route of uptake in terrestrial herbivores (Gamberg et al., 2015). In addition to age, sex and season (Gamberg et al., 2020), recent studies have found relationships between concentrations of THg in Hudson Bay caribou and some climate-related factors such as sea-ice freeze-up, which can affect deposition to lichens (Morris et al., 2022; McKinney et al., 2022).

Hallanger et al. (2019) reported concentrations of THg in Svalbard foxes in the period from 1997 to 2014 increasing at ~3.5% annually, approximately the same value reported here, however the present trend is not significant and had low power (SI Table A2). In Hallanger et al. (2019), the relatively poor fit to the data improved when carbon source ($\delta^{13}\text{C}$), availability of reindeer carcasses and sea-ice extent were included in the model, resulting in a more rapid increase in THg (7.2% annually). Arctic foxes in Svalbard are opportunistic predators and scavengers with a highly variable diet, they feed extensively on marine resources (seabirds and their eggs, seal pups and carcasses) in addition to terrestrial resources (reindeer

Fig. 4. Selected temporal trends and 95% confidence bands of THg ($\mu\text{g}/\text{kg}$) in muscle of Arctic char from Canada (wet weight), Sweden (wet weight) and Norway (dry weight) (see SI Table A2). Data points are annual medians of \log_e concentrations, with the y-axis back-transformed for visualization. Figure reproduced with permission from AMAP (2021).

carcasses, ptarmigans, geese) which have different patterns and levels of Hg (Frafjord, 1993; Eide et al., 2005; Ehrich et al., 2015).

3.2.5. Marine mammals

Marine mammals provided temporal monitoring coverage across a broader geographic range in the present study than in the 2011 AMAP Mercury Assessment (AMAP, 2011; Rig  t et al., 2011), which affected the spatial interpretation of the trends (Section 3.3). Ringed seals provided 22 time series from adult and juvenile (<5-years-old) seals from multiple locations in Canada ($n = 16$; muscle and liver) (Houde et al., 2020) and in West and East Greenland ($n = 6$; liver) (SI Table A1). The time series in cetaceans included Canadian beluga whales ($n = 10$; liver, muscle and epidermis) (Loseto et al., 2015) and Faroese long-finned pilot whales (*Globicephala melas*) ($n = 5$; liver and muscle). Beluga were subdivided into small/young (<380 cm) and large/adult animals (≥ 380 cm), while pilot whales were subdivided into adult (≥ 495 cm), juvenile (<495 cm) male, and undefined (mixed) categories. There were 16 polar bear time series subdivided into adult males, adult females and juveniles from Greenland ($n = 8$; hair and liver), Svalbard ($n = 2$; hair) (Lippold et al., 2020) and Canada ($n = 6$; liver) (Morris et al., 2022) (SI Table A1). Bears were classified as juveniles if less than 5 years old in Norway and Canada and less than 5 years (female) or 6 years (male) in Greenland.

None of the time series for Greenlandic seals had significant trends of THg over time (SI Table A2). Seven time series in Canadian seals had significant recent trends: in the eastern Beaufort Sea (western Arctic), Lancaster Sound (High Arctic), Labrador Sea (eastern Arctic), and in western Hudson Bay. The only significant recent trend of THg concentrations in liver of ringed seals was increasing at 8.9% per year in adult animals from the Labrador Sea, but caution should be taken due to the low statistical power of the trend (34%). The six significant recent trends in muscle were more robust than liver, and were all decreasing between -2.4% and -8.0% per year (Fig. 3; SI Table A2). Two of the significant, recent downwards trends were non-linear; muscle concentrations in adult seals from western Hudson Bay decreased in the period from 2003 to 2012 and then stabilised, whereas those in adult seals from the Eastern Beaufort Sea were initially stable and then decreased from about 2009 (SI Fig. A2).

The previously observed increasing trend in juvenile ringed seals at Avanersuaq in Northwest Greenland (1984–2008; AMAP, 2011) was no longer significant in the current study ($p = 0.06$; also previously reported at $p = 0.08$ in Rig  t et al., 2012). Rig  t et al. (2012) also reported that time series of THg in ringed seal populations up to 2010 were significantly decreasing in Qeqertarsuaq in Central West Greenland and increasing at Ittoqqortoormiit in Central East Greenland. Published trends of THg in Canadian ringed seals were generally consistent, but not an exact match with the results described in the current study, as they found no significant trends in liver (Houde et al., 2020), while one significantly increasing trend in liver was described in the present study (adult seals from the Labrador Sea). In both the present study and Houde et al. (2020), THg was generally decreasing in muscle of ringed seals, including in those from the Labrador Sea and Hudson Bay (SI Table A2).

Three significant recent trends for polar bear hair or liver were observed and all were increasing (SI Table A2; Figs. 3, 5). THg concentrations in hair samples from adult male bears from East Greenland (Ittoqqortoormiit) increased at 2.3% per year, while the rate of increase in hair from juvenile bears from East Greenland was slower (1.7% per year). In western Hudson Bay, THg in liver of adult male polar bears increased at 5.8% per year. However, the power of most Canadian polar bear time series was below the desired level of 80% (SI Table A2). The two trends in the hair of the Norwegian bears were not significant; the power for the adult female time series exceeded 80%, however that in the juvenile bears was relatively low (42%) indicating that the time series is unlikely to detect a significant trend even if one was present. THg concentrations in the liver of juveniles from East Greenland showed a significant non-linear trend (SI Fig. A2; SI Table A2) with concentrations in 2018 that were significantly higher than in 1983. Concentrations were increasing from 1983 until the mid-1990s, decreasing through to about 2010 and then increasing through to 2018

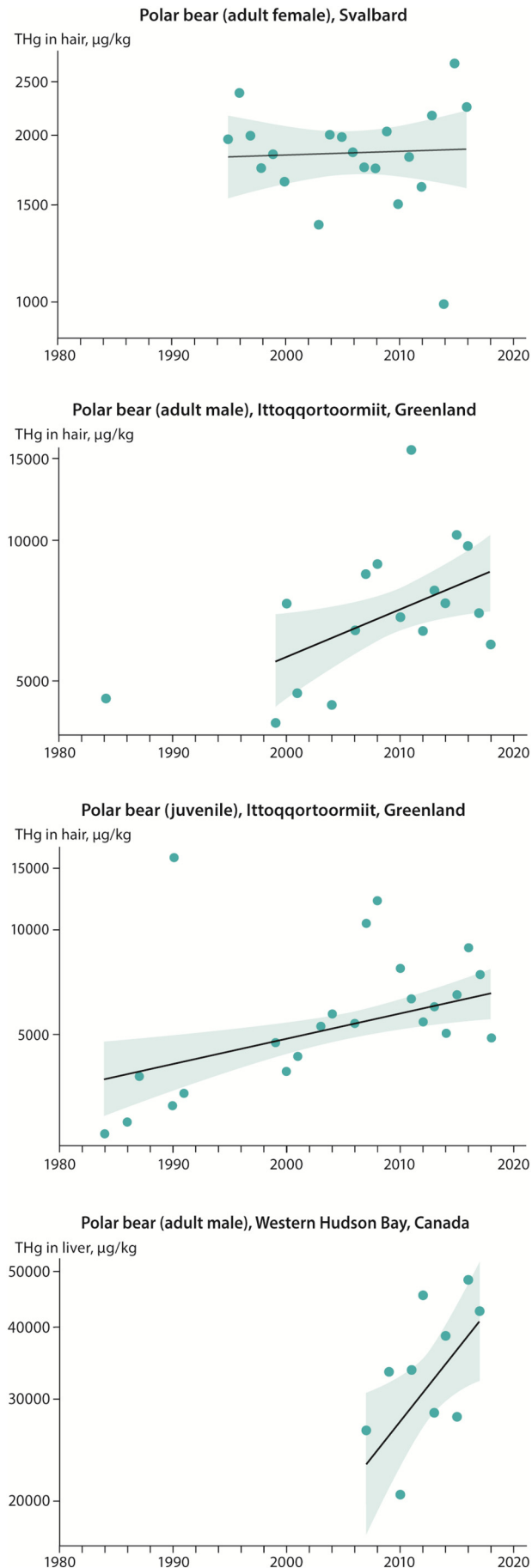
(SI Fig. A2), a possible cyclical trend; however, more data as well as linkages to explanatory variables are required to evaluate this possibility.

The results in the present study indicate that the trends of THg have continued to increase in the hair of polar bears from Ittoqqortoormiit, East Greenland as observed in the 2011 AMAP Mercury Assessment (AMAP, 2011; Rig  t et al., 2011). A recent report on Hudson Bay polar bears (liver) found non-significant temporal trends of Hg in shorter (2007/2008–2015/2016) time series from the Western and Southern Hudson Bay subpopulations (Morris et al., 2022). Dietary changes affect THg/Hg levels in polar bears significantly. Ecological changes in food web composition and corresponding increases in the dietary proportions of lower trophic level prey were found to decrease THg levels in the hair of Beaufort Sea polar bears (McKinney et al., 2017). This was further illustrated in Lippold et al. (2020) where the temporal increase of THg observed in polar bears from Svalbard was slightly faster when the trend was adjusted for the variation in carbon source (marine vs. terrestrial). Differences in the statistical approaches and years modelled, including truncation due to gaps in the time series, likely explain the relatively subtle differences with the published results for polar bears. Future assessments could include dietary factors such as stable isotopes of carbon in temporal models in order to compensate for dietary/food web differences affecting the THg levels in bears and other predators.

Three recent trends were significant in beluga, indicating that THg concentrations were decreasing in the epidermis of both large and small whales and in the liver of large whales from the eastern Beaufort Sea (from -2.5% to -8.6% per year) since 1999 (Fig. 3). The trend in the full liver time series was non-linear and showed that concentrations had increased from 1981 to about 1995 and then decreased, with concentrations at the end of the time series still significantly higher than those in 1981 (SI Table A2, SI Fig. A2). The trend in the epidermis of small whales was also non-linear and suggested local peaks in concentration around 1998 and 2013 with concentrations at the end of the time series significantly lower than those in 1999 but not at the start of the time series in 1993 (SI Fig. A2). By contrast with the beluga, the only significant recent trend in long-finned pilot whales was an increasing trend in the muscle of juvenile male whales (1.7% per year) (SI Table A2). The overall trend was non-linear with a period of slow decline from 1997 to ~ 2005 followed by a faster increase in concentration from ~ 2005 to 2018 (SI Fig. A2).

In the present study, the non-linear trend of THg in the liver of eastern Beaufort Sea beluga increased over the period from 1981 (liver) or 1993 (epidermis) to approximately 2002 (SI Fig. A2), consistent with the trend described in AMAP (2011) and Loseto et al. (2015). The previous AMAP Mercury Assessment also reported an increasing trend in beluga liver from Pangnirtung, Nunavut (Cumberland Sound population); however, this time series has not been extended past 2010 and was not re-analyzed for the current assessment. A previous publication for Beaufort Sea beluga divided whales into old and young categories for analysis of liver THg and subdivided muscle by size of the animals (Loseto et al., 2015). However, because age and size variables were significantly correlated in both populations of beluga here, size was used as a proxy for age in the current assessment for the sake of consistency in the analyses. The increasing trends previously reported for muscle and liver of eastern Beaufort Sea beluga from 1981 to ~ 2002 were less consistent from ~ 2002 to 2012, decreasing in older (liver) and large (muscle) whale categories but flattening out in young and small whales (Loseto et al., 2015). These previous findings are partially consistent with the decreasing, recent trends observed in liver and epidermis in eastern Beaufort Sea beluga in the current assessment (SI Table A2, SI Fig. A2).

Relationships between concentrations of THg in high trophic level Arctic wildlife and atmospheric oscillation indices, sea-ice conditions, temperatures and precipitation have been described for several marine mammals in Canada and Greenland (Rig  t et al., 2012; Loseto et al., 2015; Houde et al., 2020; Morris et al., 2022; McKinney et al., 2022). These studies highlight the potential for these factors to influence THg concentrations over time or independently. However, the role of climate/weather drivers on trends of THg requires further investigation, as these studies have been species and location-specific which limits our ability to make general



conclusions. In general, these results do suggest that climate and dietary changes are highly influential factors in several mammalian wildlife models (McKinney et al., 2022).

Except for beluga, all of the marine mammal species investigated had at least some time series with significantly increasing trends in THg in liver over the last 20 years. Based on the toxicological assessment undertaken in Dietz et al. (2022), this could indicate ongoing and potentially increasing risk to some individual animals in these populations.

3.3. Are there spatial patterns in mercury trends in biota from the Arctic?

Mercury temporal trends at individual sites and/or in a certain species/tissue reflect a combination of both regional and local environmental contamination as well as a broad range of factors and processes that can influence Hg levels in a particular type of sample. Temporal trend monitoring protocols are often designed to minimise the influence of some of these processes and factors (e.g., by sampling biota during specific seasons or of a certain age class). However, individual temporal trend studies require careful interpretation. In the 2011 AMAP Mercury Assessment (AMAP, 2011) a meta-analysis was conducted, combining trend results from multiple studies to see if consistent patterns of trends could be discerned at regional scales. Examining spatial patterns in Hg trends for a more integrated picture of trends may provide insight into the factors and processes driving regional trends that are potentially obscured in single time series. Spatial patterns in Hg trends can be considered using different approaches. In the current assessment, this included observations based on a qualitative (visual) interpretation of the trend patterns in biota supplemented by a quantitative analysis of the magnitude and direction of the trend versus latitude and longitude.

If taken all together, the geographical pattern of THg trends ($p < 0.05$ and/or power $\geq 80\%$) presents a complicated picture of increasing and decreasing trends, which differs from the pattern presented in the 2011 AMAP Mercury Assessment (AMAP, 2011) (Fig. 3). In the previous assessment, a greater number of increasing trends were observed in the North American Arctic and West Greenland, and a greater number of decreasing trends were observed in the European Arctic. In 2011, the European regions were represented primarily by lower trophic level species (fish and shellfish), whereas the North American Arctic included a larger number of time series for higher trophic level species such as marine mammals and seabirds (AMAP, 2011; Rig  t et al., 2011), which accumulate more Hg and produced a number of significant, powerful time series (Fig. 3; SI Table A2). In addition, the 2011 assessment reflected trends analyzed for the entire period of the available time series, which resulted in comparisons of temporally inconsistent trends (AMAP, 2011). The focus of the current assessment on trends over the most recent 20-year period provided a more consistent basis for comparison, as many of the time series considered included relatively complete monitoring sequences for the period since 1999. The inclusion of a greater number of marine mammal and seabird time series from European sites resulted in more uniform species and trend distribution throughout the different regions (i.e., North America, Greenland and Europe) (Fig. 3).

For the marine fish and blue mussel time series, increasing trends were observed in time series from the Faroe Islands, Iceland, and Greenland, noting that mussel series are only available from coastal sites in Iceland and northern Norway (Fig. 3, bottom right). Freshwater fish exhibited both increasing and decreasing trends, in some cases in the same lake systems (Fig. 3, top left). In general, increasing trends appeared to be associated with sites at lower latitudes (as well as on Bj  rn  ya) and decreases were often seen in lakes at higher latitudes. Only three terrestrial mammal time series were available, and only two with adequate power (Fig. 3, top left),

Fig. 5. Selected temporal trends and 95% confidence bands of THg concentrations in hair and liver ($\mu\text{g}/\text{kg}$) of polar bears. Bears are from Svalbard (hair of adult females, dry weight), Ittoqqortoormiit, Greenland (hair of adult males and juveniles, dry weight), and Western Hudson Bay, Canada (liver of adult males, wet weight) (see SI Table A2). Data points are annual medians of \log_e concentrations, with the y-axis back-transformed for visualization. Figure reproduced with permission from AMAP (2021).

both for caribou from sites in Canada – one with an increasing trend and the other a decreasing trend (neither significant). For seabirds (Fig. 3, top right), with some exceptions, decreasing recent trends were seen in the Canadian Arctic and Svalbard, and increasing trends in the Faroe Islands, and East Greenland; no new data were available for seabird time series from Alaska reported in the 2011 assessment. Like seabirds, the majority of the powerful and significant trends in marine mammals from Canada were decreasing with some exceptions (Fig. 3, bottom right); increasing trends were observed in Faroe Islands pilot whales as well as for marine mammals from East and West Greenland.

These patterns were further explored through statistical analyses of the relationship between observed trends, latitude and longitude. Few trends with latitude were evident when plotting all trend data together or when the data were separated by ecological group (i.e., blue mussels, freshwater fish, marine fish, marine mammals, terrestrial mammals, and seabirds). Only seabirds exhibited a significant linear correlation with latitude (Fig. 3, top right) with more rapidly decreasing trends observed at higher latitudes ($r^2 = 0.37$, $p = 0.035$). However, the time series available were primarily from lower latitudes, with few high latitude trends available, so this relationship should be interpreted with caution.

Significant relationships between trends of THg with longitude were non-linear, and unlike latitude, seabird trends had no significant longitudinal relationship. The most distinct relationship with longitude was observed in marine mammals (Fig. 6); THg was decreasing most rapidly in marine mammals at the most western longitudes in the Canadian Arctic (-125°) but switched from slowly decreasing to slowly increasing from west to east through Greenland before plateauing and remaining relatively consistent through the Northern European locations. As with plotting the relationship between THg trends and latitude, the data were weighted more heavily to lower longitudes, which affects the ability to infer that the observed relationships are consistent across the circumpolar Arctic.

The updated longitudinal pattern differed from that in the previous AMAP Mercury Assessment where trends of THg in marine mammals were found to be increasing in Canada and West Greenland while generally decreasing in Northern Europe (AMAP, 2011). In contrast, the current trends tended to be increasing at higher longitudes for several species groups (i.e., towards Europe, Fig. 3); including strong trends in marine mammals (Figs. 3, 6). The increase in the number of years available for analysis increased the number of resulting trends that could be interpreted in the current assessment, including several time series that showed decreasing trends at western longitudes in Canada that did not meet the criteria for inclusion in the analysis in 2011 (AMAP, 2011). These additions likely affected longitudinal relationships; however, many factors contributed to these differences.

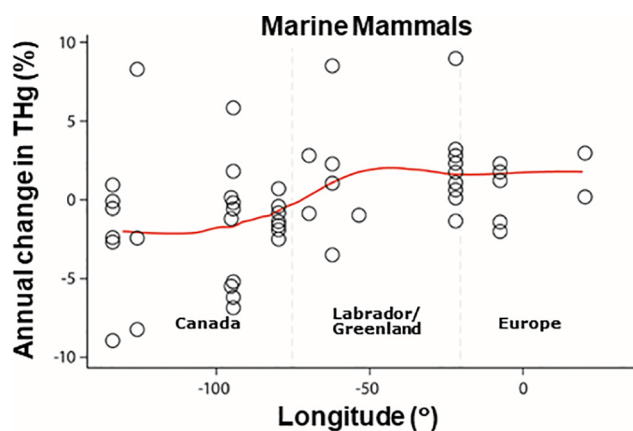


Fig. 6. Annual change of total mercury (THg, %) over time in marine mammals as a function of longitude ($^\circ$). Data points represent the recent trends (1999–present) for an individual time series. The red line represents the non-linear relationship that best describes the pattern of trends in marine mammals (trend values are available in SI Table A2). Figure reproduced with permission from AMAP (2021).

4. Conclusions and recommendations

The temporal trends presented in this article provide improved and relatively comprehensive Arctic coverage compared to the 2011 AMAP Mercury Assessment and related publication (AMAP, 2011; Rig et et al., 2011). Continued monitoring since 2011 has resulted in considerable improvements in the number and statistical power of available biota time series. This has improved our ability to generate, interpret and compare temporal trends and reinforces that these monitoring efforts should be continued annually where possible. The broadest geographic monitoring coverage and most powerful trends were observed for marine mammals, seabirds and freshwater fish, though there remains substantial regional variation in coverage within all species groups. The current marine mammal monitoring programs should continue and be expanded where possible. A broader geographic range of samples from toothed whales and ringed seals in particular, which are important subsistence animals and high trophic level predators, would make for useful spatiotemporal comparisons relevant to both ecosystem health and human exposure. Caribou/reindeer and other ungulates also have broad geographic distributions, are key subsistence animals, and have different exposure pathways than marine mammals; their monitoring should be expanded beyond the two species and three sites described here. Besides terrestrial mammals, geographic monitoring coverage was most limited for marine fish and invertebrates (e.g., blue mussels). Monitoring of common species should be maintained, and new time series added as needed, particularly for species of importance to the subsistence of Northern and Arctic Peoples and when the Hg levels and trends are unknown.

Concentrations of THg changed (increased or decreased) significantly in 37 of the 77 full time series that had sufficient statistical power ($\geq 80\%$), with 31 significant recent trends (1999–present) in these 77 time series. There were 14 decreasing and 17 increasing significant recent trends in biota, making generalizations about the direction of trends overall or within species groups difficult to draw. This mixture of increasing and decreasing temporal trends highlights the need for continued monitoring of Hg in Arctic environments and biota. Though the majority of the trends were log-linear, several time series exhibited complex, non-linear trends that may be related to changing environmental processes that require better understanding themselves. For marine mammals, more increasing trends were observed at higher (East Greenland/European) longitudes, while seabirds were the only species to vary with latitude, with increasing trends observed at lower latitudes within the North. These spatial comparisons have improved greatly since 2011, but should still be interpreted with caution as the distribution of time series remains unbalanced geographically. Differences in local processes and sources could play a substantial role in the geographic differences observed, and as discussed in other articles of this special issue, changes in a broad range biological, ecological and environmental parameters affect the fate and delivery of Hg to ecosystems and its uptake and bioaccumulation in biota (Dastoor et al., 2022; Jonsson et al., 2022; Ch etelat et al., 2022; McKinney et al., 2022).

Meeting the information needs of Northern peoples who consume Arctic biota remains a high priority for Northern research and monitoring. Monitoring of the selected species and tissues are relevant for assessing both ecosystem health and human exposure through consumption, so additional time series for tissues that are most often consumed in relatively large amounts (e.g., muscle tissue) should be considered. Priority should also be given to extending temporal trend monitoring to parts of the North currently lacking time series, including the Russian Arctic, and could include retrospective analyses to fill gaps while new monitoring systems are established. Trend analysis methods should be further developed to take account of additional factors (biological, ecological, environmental) and in particular those associated with climate change, in order to better understand the drivers of Hg contamination in ecosystems and food webs. Assessing temporal trends of Hg is a critical method of inquiry that can reveal insights into relationships among key environmental and intervention drivers and should remain a priority for Northern research and monitoring through AMAP and its member nations.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary information

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2022.155803>.

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