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Review

How closely do mercury trends in fish and other aquatic wildlife track those in the atmosphere? – Implications for evaluating the effectiveness of the Minamata Convention

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ABSTRACT

The Minamata Convention to reduce anthropogenic mercury (Hg) emissions entered into force in 2017, and attention is now on how to best monitor its effectiveness at reducing Hg exposure to humans. To address this, a key question is how closely Hg concentrations in the human food chain, especially in fish and other aquatic wildlife, will track the changes in atmospheric Hg that are expected to occur following anthropogenic emission reductions. We investigated this question by evaluating several regional groups of case studies where Hg concentrations in aquatic biota have been monitored continuously or intermittently for several decades. Our analysis shows that in most cases Hg time-trends in biota did not agree with concurrent Hg trends in atmospheric deposition or concentrations, and the divergence between the two trends has become more apparent over the past two decades. An over-arching general explanation for these results is that the impact of changing atmospheric inputs on biotic Hg is masked by two factors: 1) The aquatic environment also contains a large inventory of legacy emitted Hg that remains available for bio-uptake leading to a substantial lag in their response time to a change in external inputs; and 2) Biotic Hg trends reflect the dominant effects of changes in multi-causal, local and regional processes (e.g., aquatic or terrestrial biogeochemical processes, feeding ecology, climate) that control the speciation, bioavailability, and bio-uptake of both present-day and legacy emitted Hg. Globally climate change has become the most prevalent contributor to the divergence. A wide range of biotic Hg outcomes can thus be expected as anthropogenic atmospheric Hg emissions decline, depending on how these processes operate on specific regions and specific organisms. Therefore, evaluating the effectiveness of the Minamata Convention will require biomonitoring of multiple species that represent different trophic and ecological niches in multiple regions of the world.

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1. Introduction

The entry into force in August 2017 of the Minamata Convention on Mercury was an important step in international efforts to control mercury (Hg) releases into the environment and to protect human health from its harmful effects (www.mercuryconvention.org). Subsequently, in accordance with Article 22 of the Convention, the Second Conference of the Parties (COP2) in November 2018 turned its attention in part to the question of how best to evaluate the effectiveness of the Convention in meeting its goals (www.mercuryconvention.org/Meetings/Intersessional%20work/tabid/6325). Monitoring of biotic Hg concentrations and trends should logically be an important part of

this evaluation, given the central role of aquatic food chains (fish and other aquatic wildlife) in defining human Hg exposure (Basu et al., 2018; Eagles-Smith et al., 2018; Gustin et al., 2016). However, a key question remains as to which extent measurements of Hg in aquatic biota will track the changes in atmospheric Hg that are expected to occur following future anthropogenic emission reductions.

The atmospheric and aquatic chemistry of Hg is one of the most complex of all trace metals. The Hg that is emitted from anthropogenic (and natural) sources to air is almost exclusively inorganic as elemental (Hg^0) and divalent compounds (Hg^{II}) in gaseous and particulate forms (Obrist et al., 2018). Redox reactions between Hg^0 and Hg^{II} in the atmosphere are primarily photochemically driven (Ariya et al., 2015; Saiz-Lopez et al., 2018), with the bulk of the atmospheric Hg deposited onto the Earth's surface (oceans, land, and freshwaters) being in the form of inorganic Hg^{II} . The aquatic environment also receives Hg input, primarily as inorganic Hg^{II} , from rivers,

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catchment runoff, and submarine groundwater discharge (Fitzgerald and Lamborg, 2014; Outridge et al., 2018). Once in the aquatic environment, the net transformation of inorganic Hg^{II} through methylation to methylmercury (MeHg) is the key step in the Hg cycle leading to wildlife and human exposure. Methylmercury is one of the most bioaccumulative and toxic Hg species and is the only form that is biomagnified through food chains (Eagles-Smith et al., 2018; Fitzgerald and Lamborg, 2014).

Therefore, the relationship between anthropogenic Hg emissions to the atmosphere and Hg trends in aquatic biota is influenced by many environmental and ecological factors (e.g., temperature, light intensity, pH, redox condition, organic carbon and nutrient concentrations, and food web structure and dynamics) that control the rates of Hg deposition from the atmosphere, MeHg production (methylation) and degradation (demethylation) in the aquatic environment, and the uptake of Hg, especially MeHg, by biota (Eagles-Smith et al., 2018; Hsu-Kim et al., 2018). The complexity of these processes, along with the large inventories of legacy anthropogenic and natural Hg stored long-term in terrestrial and aquatic systems, suggests that biotic Hg may be only tenuously connected to atmospheric Hg which has a much shorter life-time (0.5 to 2 years) than the decadal or century-scale life-times of Hg in soils and oceans (Horowitz et al., 2017; Saiz-Lopez et al., 2018). Furthermore, even if atmospheric and biotic Hg do follow similar trends, there could be a significant time lag between their response. The response also likely differs in ecosystems of different sizes and complexity: changes in biotic Hg in marine ecosystems could lag significantly behind those in smaller, freshwater ecosystems (Sunderland and Mason, 2007; Sunderland and Selin, 2013).

One aspect of the United Nations Environment Programme (UNEP)'s Global Mercury Assessment (AMAP/UNEP, 2019) was a review of relationships between the trends of atmospheric Hg and Hg in fish and other aquatic wildlife, and the reasons for the match or mis-match between those trends. These relationships are important because of their relevance to predicting the efficacy of international regulatory actions, as prescribed in the Minamata Convention, in ultimately reducing Hg exposure in humans and wildlife. This review examines major case studies on biotic Hg trends in globally distinct regions, and compares them with concurrent atmospheric Hg trends

in the same region, summarized in four regional groups of case studies. The studies are from North America, Europe, China, and the Arctic, where Hg concentrations in certain aquatic biota have been monitored continuously or intermittently for several decades. No such long-term time-series data for biotic Hg are available for other regions of the world, especially for the Southern Hemisphere. Our findings are then discussed in the context of monitoring the effectiveness of the Minamata Convention.

2. Trends in atmospheric Hg concentration and wet deposition

Before presenting the biotic Hg trend case studies, we first review the general trends in Hg concentrations in air and in wet deposition, with which the aquatic biotic Hg trends will be compared.

2.1. North America and Europe

North America and Europe are considered together here because their overall atmospheric Hg concentrations and deposition fluxes have trended similarly over the past few decades (Zhang et al., 2016). Measured Hg concentrations in the air (as gaseous elemental mercury (GEM), or total gaseous mercury (TGM)) declined by 10–40% between 1990 and 2010 throughout most of North America and Europe (Cole et al., 2014; Slemr et al., 2011; Weiss-Penzias et al., 2016), a pattern that has been matched by trends in Hg concentrations and fluxes in wet deposition (Brigham et al., 2014; Prestbo and Gay, 2009; Weiss-Penzias et al., 2016; Zhang and Jaeglé, 2013), as well as by lake sediment core records (e.g., Drevnick et al., 2016). This general declining trend agrees very well with the declining Hg point-source emissions in North America and Europe over the same period (AMAP, 2010; AMAP/UNEP, 2013), and has been reasonably well reproduced by a recent modelling study with revised global Hg emissions inventories (Fig. 1) (Zhang et al., 2016). This model study accounted for the declining emissions from commercial Hg-containing products since 1990, corrected for shifts in the speciation of airborne Hg emissions related to air pollution control technology, and reduced the putative importance of atmospheric Hg emissions from artisanal and small-scale gold mining (ASGM).

However, a closer analysis of the observational data in Fig. 1 suggests that this systematic declining trend may have become less clear

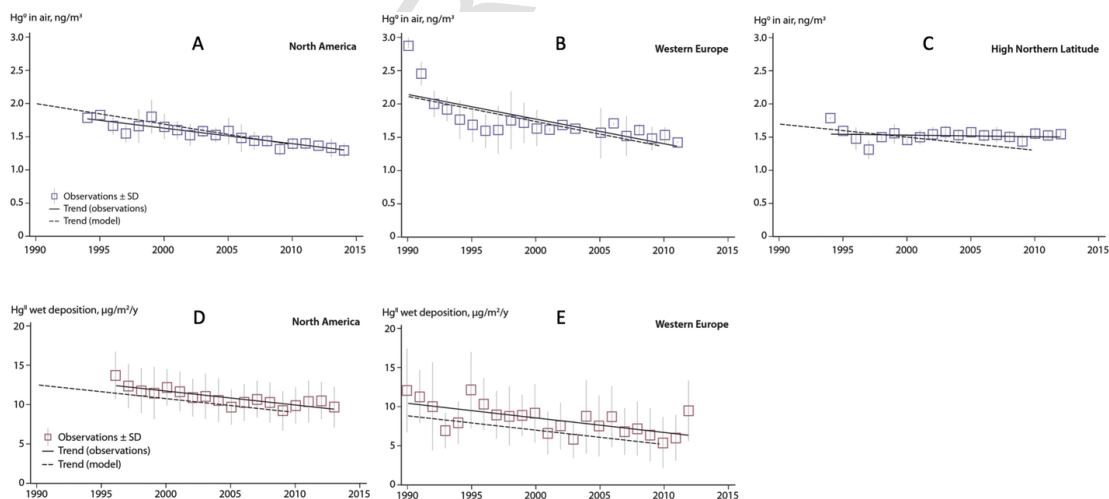


Fig. 1. Observed and modelled trends for the period of 1990 to 2013 in atmospheric gaseous elemental mercury (Hg^0) concentrations (A–C) and divalent Hg^{II} wet deposition fluxes (D and E) in different regions of the Northern Hemisphere (Zhang et al., 2016). Observations for individual years are shown as squares, with the vertical bars showing \pm one standard deviation. The solid line represents a linear regression, whereas the dashed line is the trend line from the GEOS-CHEM simulation. The data are averaged regionally across for North America (A and D), Western Europe (B and E), and high northern latitude regions (C).

and uniform, and even reversed, over the most recent decade (i.e., since ~2008). This is demonstrated by a recent study in North America (Weiss-Penzias et al., 2016) that showed gaseous elemental Hg concentrations in the air had stabilized since 2008 with a flat slope over time. While Hg concentrations and fluxes in wet deposition continued to decline in some regions (e.g., Risch and Kenski, 2018), an increasing trend has also been observed throughout North America (Weiss-Penzias et al., 2016). Such a divergency in the recent air Hg trend likely reflects varying local, regional and global influences, and is not reproduced by the model of Zhang et al. (2016) (see Fig. 1).

2.2. China

China is the largest national emitter of anthropogenic Hg to the atmosphere (Fu et al., 2015). In contrast to the global trend, anthropogenic Hg emissions in China increased rapidly from 1978 to around 2007 at an average rate of ~5.5% per year, except for the period 1998–2000 when emissions decreased due to the Asian financial crisis that led to a reduction in fuel consumption (Wu et al., 2016). Mercury emissions in China are reported to have plateaued around 2007 to 2010, and have shown a declining trend in the past few years (Wu et al., 2016). Available but limited data on atmospheric Hg concentrations over the past decade in China generally agree with this emissions trend. Direct measurements of GEM at Guiyang, an urban site in southwest China, showed that annual mean GEM concentrations increased at a rate of ~2.5% per year between 2002 and 2010 (Fu and Feng, 2015). GEM concentrations also increased at Mt. Changbai, a remote alpine site in northeastern China, at about the same rate from 2009 to 2013, but appear to have stabilized since (Fu et al., 2015; Fu et al., 2016). Mercury passive sampling and plant biomonitoring on the Tibetan Plateau suggest that atmospheric Hg concentrations were stable during 2006 to 2009 and decreased during 2010 to 2015 (Tong et al., 2016) that agrees with the Hg records in sediment and glacier ice cores from the Tibetan Plateau (Kang et al., 2016).

2.3. The Arctic

Long-term air Hg monitoring data are available at several Arctic sites (above 60°N), such as Alert (Nunavut, Canada), Villum Research Station at Station Nord (Greenland), and Zeppelin Station (Svalbard, Norway). The results show that atmospheric Hg concentrations have been generally declining, but at a markedly slower rate than elsewhere (Cole et al., 2013, 2014). The observed and modelled trends also disagreed more than in other regions, with observed GEM concentrations decreasing by ~0.2% per year since 1994, compared to a modelled decrease of 1.3% per year (Fig. 1). Deposition of atmospheric mercury can occur in the form of oxidized mercury to the Arctic coastal and marine environment during the spring-time atmospheric mercury depletion events (Schroeder et al., 1998), and in the form of gaseous elemental mercury via tundra uptake in the interior Arctic (Obriest et al., 2017). Unfortunately, there are no decade-long observational data of Hg deposition flux available for the Arctic or subarctic regions; existing depositional data are limited to one to two years of measurements only (Sanei et al., 2010).

3. Trends in Hg in aquatic biota and possible causes

3.1. Case study 1: fish and birds in lakes and coastal waters of North America

A large number of studies have reported inconsistent, diverging, or mixed temporal trends of Hg in aquatic biota throughout North America. The Great Lakes are an especially interesting study area because of the wide diversity of species monitored within the same general area, and the length of some of the biotic time trend datasets. Also, there is Hg isotopic evidence from two predatory Great Lakes fish (lake trout *Salvelinus namaycush* and burbot *Lota lota*) that established that the atmosphere, rather than sediments or watershed soils, was the ultimate source of most of the Hg accumulated into fish tissues in these large ecosystems (Lepak et al., 2015). This is consistent with earlier mass balance studies (Mason and Sullivan, 1997). However, intervening ecological, geochemical or climatic processes may be acting to drive biological time trends along divergent paths. Blukacz-Richards et al. (2017) evaluated the temporal trends since the 1970s of Hg concentrations in eggs of a piscivorous bird (herring gull *Larus argentatus*), in muscles of two piscivorous fish (lake trout and walleye *Sander vitreus*) and of a planktivorous fish (rainbow smelt *Osmerus mordax*) in the Great Lakes. The results presented a mixed temporal pattern (Fig. 2), with declining biotic Hg trends in all species in the first few decades (up to about 1995–2000) that matched the declining atmospheric Hg trend in North America (see Fig. 1). After about 2005, however, trend reversals were detected in most species at some locations. Zhou et al. (2017) also detected a breakpoint at ~2010 in fish Hg trends in the Great Lakes, with a significant decreasing trend before 2010 and no trend or an increasing trend since then.

Similar trend results were reported in fish populations across western Canada and the USA. When examining Hg data on 96,000 fish muscle samples from 206 species in over 4200 lakes, Eagles-Smith et al. (2016) found a significant, rapid decline in length-adjusted tissue Hg concentrations during the 1970s, with no subsequent significant trend up to 2012. In all of these studies, the authors attributed the early decline in biotic Hg to regional and local declines in atmospheric Hg concentrations and deposition related to regional anthropogenic emissions controls. They suggested that the subsequent trend reversal or lack of a significant trend, was caused by a variety of factors such as increasing local emissions, food web changes, eutrophication, or climate change.

Contrasting biotic Hg trends have also been reported for other lakes in the same region. When examining temporal fish Hg trends in hundreds of small lakes in Ontario, Canada, Gandhi et al. (2014) found a general decline in length-adjusted fish muscle Hg concentrations from the 1970s to 1990s for northern pike (*Esox lucius*), walleye and lake trout, followed by relatively small increases in some lakes starting about 1995–2000. Both the early declining trends and recent increasing trends were more pronounced in northern lakes and in northern pike and walleye compared to lake trout. In contrast, when examining changes in fish muscle Hg from 873 Ontario lakes, Tang et al. (2013) reported no significant decreases over the past several decades in any of the seven species (walleye, northern pike, lake trout, burbot, smallmouth bass *Micropterus dolomieu*, whitefish *Coregonus clupeaformis* and white sucker *Catostomus commersonii*). Instead, mean fish Hg concentrations were found to be slightly higher in the period 2005–2010 than in 1974–1981, and were significantly so in northern pike. Such divergent results were also reported in fish Hg in four small lakes within a national park in northern Minnesota

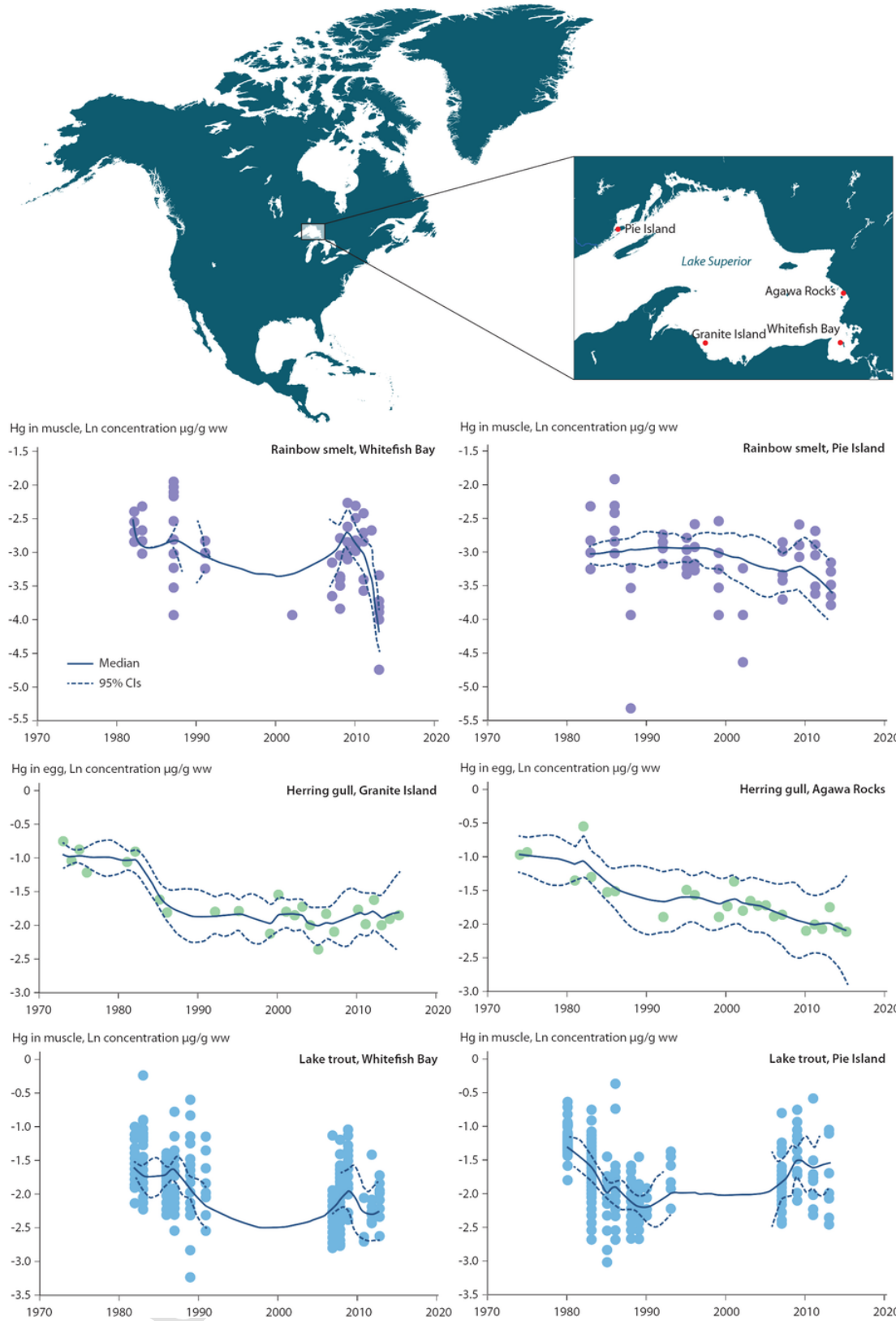


Fig. 2. Mercury concentrations (wet weight; natural logarithm-transformed) in muscle of rainbow smelt (planktivorous) and lake trout (piscivorous), and in eggs of herring gull (piscivorous) from the Great Lakes region, North America from the 1970s to 2010s (Blukacz-Richards et al., 2017). The solid and dashed lines correspond to the median and the 95% confidence intervals (CIs) of the predicted Hg concentrations.

(Brigham et al., 2014) and throughout the State of Massachusetts (Hutcheson et al., 2014) in the USA. Whereas the majority of these lakes showed a decreasing trend in fish Hg between ~2000 and ~2010, a smaller set of lakes showed an increasing or no trend. The diverging trends in fish Hg among lakes from the same region clearly demonstrate the complexity of ecosystem responses to changes in atmospheric Hg concentration and deposition, and suggest that other factors besides Hg inputs, likely related to human and climate induced variability, were more important drivers.

Similarly, a declining trend was found in different populations of the marine species, bluefish (*Pomatomus saltatrix*), along the northeast coast of the USA. Substantial reductions in muscle Hg, of ~30–40%, were reported when comparing fish caught in the early 1970s with those caught between 1994 and 2011; the New York regional data suggested no change in bluefish Hg concentrations after ~1994 (Fig. 3) (Cross et al., 2015).

Most of these studies did not include carbon and nitrogen isotope data, making it impossible to investigate whether changes in feeding behavior (e.g., prey trophic level and feeding location) influenced the Hg trends. The value of including trophic dynamic information in the interpretation of Hg temporal trends was demonstrated by Burgess et al. (2013) in a study of Hg in herring gull eggs on the eastern Canadian seaboard. Between 1972 and 2008, two sites displayed a trend of significantly declining egg Hg that was consistent with the declining atmospheric Hg deposition occurring at that time (see Fig. 1). However, when trophic level changes over time were factored into the analysis using $\delta^{15}\text{N}$ isotope data, it was found that the Hg declines were due to feeding behavior shifts. $\delta^{15}\text{N}$ is a widely-used indicator of the trophic level of species' prey selection, and was highly correlated with egg Hg in the birds. These results suggest that Hg in the coastal ecosystem in that region may have remained relatively constant over the past few decades despite the reduction in airborne Hg fluxes.

3.2. Case study 2: freshwater fish in Fennoscandia

Braaten et al. (2017) and Åkerblom et al. (2014) assessed the spatial and temporal trends of Hg in various species of freshwater fish (e.g., northern pike, Eurasian perch *Perca fluviatilis*, brown trout *Salmo trutta*, Arctic char *Salvelinus alpinus*, roach *Rutilus rutilus*) during the past 50 years (1965–2015) based on 54,560 samples from 2775 lakes across Fennoscandia (Sweden, Finland, Norway, and the Kola Peninsula in Russia). Some of the lakes were impacted by historical, local industrial emissions of Hg directly to surface water,

whereas others were impacted primarily by atmospheric Hg deposited onto catchment soils as well as water surfaces.

As expected, lakes that were affected by local direct pollution sources had higher mean observed fish Hg concentrations than lakes that were predominantly affected by long-range atmospherically deposited Hg. When the fish Hg concentrations were normalized to a standard 1-kg pike, the Hg concentrations in “1-kg pike equivalent fish” showed a consistent and significant decreasing trend for the entire database (Fig. 4). This declining trend matches very well with the general declining atmospheric Hg trend over northern Europe (see Fig. 1). Of particular interest is the finding that the declining fish Hg trend was much stronger for the entire database than for lakes only impacted by atmospheric Hg deposition. This could suggest that reduced Hg emissions, especially a reduction in local emission sources, lead to lower Hg in fish. However, Braaten et al. (2017) noted that the temporal trends varied with different standardization methods, and cautioned that a better understanding of possible confounding environmental processes (such as the impact of temperature and dissolved organic matter) is needed prior to concluding that the two declining trends are causally linked.

Also of note is that some of the lakes in Sweden were treated with lime in the 1970s and early 1980s to mitigate the impact of acidification. Although liming reduced fish Hg concentrations (Meili, 1995), fish Hg concentrations in these limed lakes remained consistently higher than in lakes that were not limed decades after the liming (Åkerblom et al., 2014). The temporal trends in both limed and non-limed lakes were, however, similar (Åkerblom et al., 2014).

3.3. Case study 3: fish in reservoirs in North America and Europe versus China

Some of the longest time series of aquatic Hg data exist for man-made reservoirs due to concerns about the effects of impoundment on Hg methylation rates and thus on fish Hg concentrations. Although these reservoirs are not natural habitats for aquatic life, they contain abundant fish and invertebrate communities, and support important recreational fisheries in some areas (e.g., in North America and Europe) and large aquaculture operations in others (e.g., in Asia). Extensive studies in the past decades have revealed very different fish Hg trends in reservoirs in North America and Europe from those in China, neither of which followed the atmospheric Hg trend.

Studies in North America and Europe have shown that following impoundment, the large influx of flooded vegetation and organic matter in submerged soil stimulates microbial methylation of Hg, result-

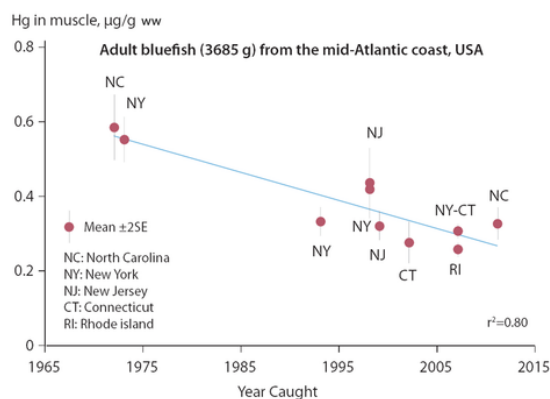


Fig. 3. Mercury concentrations (wet weight) in muscle of piscivorous bluefish (average adult of 3685 g) along the mid-Atlantic coast of the USA from 1972 to 2011 (Cross et al., 2015). The vertical bars show \pm two times standard errors (SE), and the solid lines represents a linear regression.

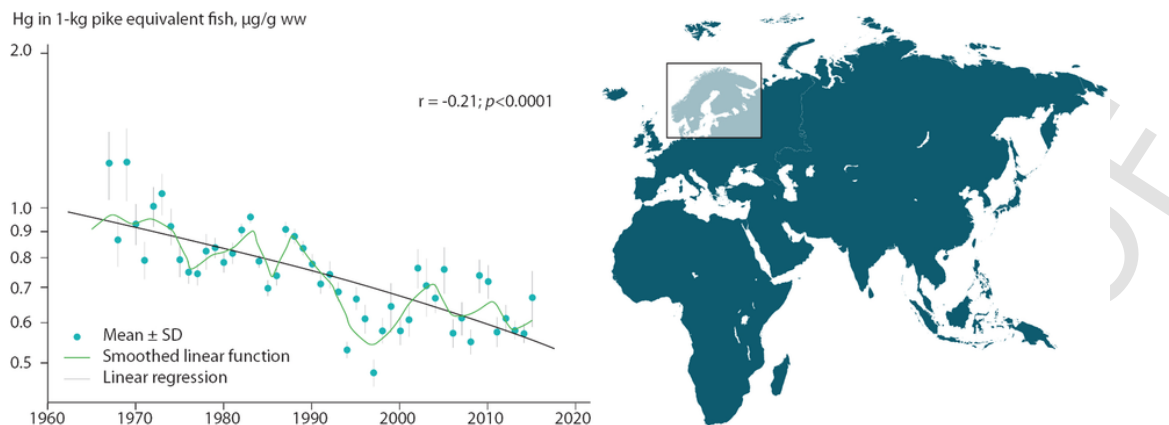


Fig. 4. Annual mean mercury concentrations (wet weight) in muscle of five main freshwater fish species (Arctic char, brown trout, perch, pike and roach; normalized to a “standard 1-kg pike equivalent fish”) in 2775 lakes across Fennoscandia from 1965 to 2015 (Braaten et al., 2017). The vertical bars show \pm one standard deviation (SD). The green line represents a smoothed linear function, while the grey line represents a linear regression. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

ing in a sharp increase in fish Hg due to biomagnification of MeHg (Bodaly et al., 2007; Hall et al., 2005; Lucotte et al., 1999; St. Louis et al., 2004). Mercury methylation rates and fish Hg concentrations typically decrease as the reservoir ages and the organic matter further decomposes (Bodaly et al., 2007). This was demonstrated in a recent analysis of the temporal trends of Hg in a range of fish species from 883 reservoirs across western North America (Willacker et al., 2016). Temporal patterns (normalized for confounding variables such as species and body length) were clearly related to the time elapsed since reservoir impoundment, with maximum fish Hg concentrations being reached on average three years after the impoundment (Fig. 5). Fish Hg concentrations thereafter declined relatively rapidly for 4-to-12 years, followed by a monotonic slow decline over many decades. Because the reservoirs were built at different dates over the past 150 years, it may be concluded that the fish Hg trends are not directly related to changing atmospheric Hg deposition. Instead, water storage management is shown to be a key factor influencing this temporal pattern. Fish in reservoirs that experienced maximum drawdown dur-

ing summer months (May–July) exhibited significantly higher concentrations (up to 11-fold) than fish in reservoirs in which drawdown occurred during other times of the year, likely due to oxidation of reduced sulfide in the sediment while exposed as well as higher microbial activities upon inundation, both of which stimulate methylation of Hg.

Indeed, for reservoirs in China, however, fish Hg concentrations may be even less connected with atmospheric inputs (Feng et al., 2018; Hsu-Kim et al., 2018). Extensive studies over the past decade on reservoirs in the Wujiang Basin in southwest China, most of which were actively used for aquaculture, have shown that Hg concentrations in the various fish species studied were remarkably low in the reservoirs along the Wujiang River regardless of the age of the reservoir (Feng et al., 2018). While biodilution and simple (short) food web structures clearly contribute to the generally low fish Hg concentrations, comparisons of fish Hg concentrations in reservoirs of different age in the same basin reveal three distinct stages of evolution (primary, intermediate, and advanced stages) due to changes in

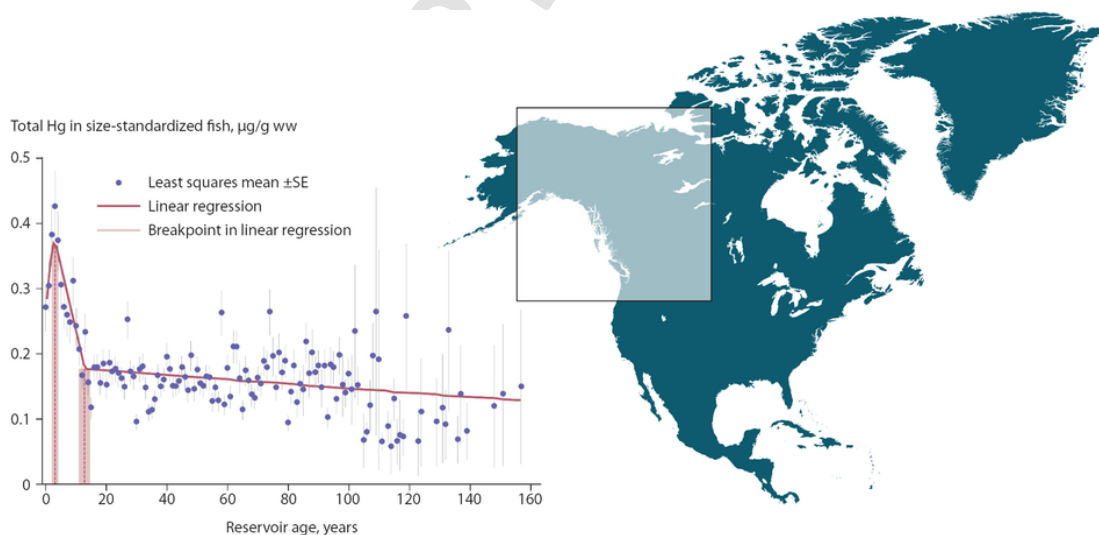


Fig. 5. Mercury concentrations in muscle of fish (wet weight, size-standardized) from reservoirs across western North America as a function of the reservoir ages (Willacker et al., 2016). The data show least squares mean \pm one standard error (SE); the least squares means account for the effects of ecoregion, waterbody, species, and sampling year. Vertical dashed lines and shaded regions indicate estimated breakpoints (\pm SE) from segmented linear regression (solid line) on fish Hg concentration when accounting for the effects of ecoregion, waterbody, species, and sampling year.

the source and concentration of organic matter in the submerged soil/sediment as the reservoir ages and cage aquaculture activities increase (Feng et al., 2018). The newly constructed reservoirs in the Wujiang Basin are not active sites of net Hg methylation due to the low organic carbon content in the submerged soils and/or low primary productivity (Meng et al., 2010; Yao et al., 2011). Consequently, during the primary stage the newly constructed reservoirs are not a net source of MeHg and instead represent a net sink (Zhao et al., 2017; Feng et al., 2018). As these reservoirs become more productive (mesotrophic to eutrophic) with time, the organic matter content in the sediment increases due to continuous increases in autochthonous productivity due to the cage aquaculture activities. This tends to promote in situ Hg methylation, and as such reservoirs at this intermediate stage have transitioned from a net MeHg sink to a net MeHg source. Over the long-term evolution of the reservoir, primary productivity continues to increase and the reservoir will eventually become eutrophic. Phytoplankton-derived organic matter, and fish food and feces, become significant sources of organic matter input to the surface sediments. The increased oxygen consumption during fresh organic matter degradation causes progressively more anoxic conditions at the sediment-water interface (Meng et al., 2010; Meng et al., 2016) that promotes the microbial Hg methylation processes during this advanced stage of evolution, when both the surface sediment and the hypolimnetic water were sites of net MeHg production (Feng et al., 2009; Meng et al., 2010; Meng et al., 2016). Thus, in contrast to fish in North American reservoirs, and despite the relatively high atmospheric Hg loading across much of China, fish Hg concentrations in Chinese impoundments reflect within-impoundment processes, especially organic matter loadings to sediments, water/soil quality, food web structure, and bidilution, rather than atmospheric inputs (Feng et al., 2018).

3.4. Case study 4: the Arctic

Rigét et al. (2011) summarized all available temporal Hg datasets on Arctic biota up to about 2009, and found that some species in some locations had shown significant increases over recent decades, whereas others with closely adjacent or overlapping distributions exhibited decreases or non-significant changes. Most of the increasing biotic Hg trends occurred in marine species in the North American and west Greenlandic sector of the Arctic, whereas most declining trends were observed in east Greenlandic and European Arctic biota. This regional dichotomy is clearly seen in the hair of polar bears (*Ursus maritimus*), and has been suggested to be due to increased emissions from Asia entering the western Arctic coincident with decreasing emissions from North America and Europe in the eastern Arctic (Dietz et al., 2006).

A few additional studies have since been published. Rigét et al. (2012) analyzed temporal trends of Hg in livers of ringed seals (*Pusa hispida*) collected from the early 1980s to 2010 from Greenland. Increasing concentrations of Hg were found in ringed seals in two out of three Greenlandic seal populations (Central East and Northwest Greenland), rising at a rate of 10.3% per year and 2% per year, respectively. In addition to age and trophic position, the study showed that the Atlantic Oscillation Index, a parameter related to climate variability, was positively associated with Hg concentrations in seals although the specific mechanism involved was not clear. Similarly, the long-term variability in beluga Hg from the Beaufort Sea region has been shown to be associated, with a lag time, with the Pacific Decadal Oscillation (Loseto et al., 2015).

By analyzing Hg in the teeth of polar bear from Svalbard in the Norwegian Arctic, Aubail et al. (2012) reported a decreasing trend in

Hg concentration over the period 1964–2003 (Fig. 6B). Since no temporal changes were found in tooth $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$, they concluded that the decrease of Hg was not due to changes in trophic dynamics; instead, it was more likely to be due to a lower environmental Hg exposure in the region. McKinney et al. (2017) also reported a significant declining trend in hair Hg of the southern Beaufort Sea polar bear population, at an average rate of -13% per year, between 2004 and 2011. This finding differs from the general west-east trend patterns in Arctic biota Hg noted above. However, only male Polar Bears in the southern Beaufort Sea area exhibited significant decreases; females from the same area showed no significant trend. Mercury concentrations in the bears' main prey (ringed seal) also showed no change up to 2007 (Gaden et al., 2009), which argues against changes in Hg inputs or the biogeochemical Hg cycle as contributing to the decline. Analyses of body condition and diet led to the conclusion that the bears' Hg trend was due to changing foraging patterns over time and not to alteration in environmental Hg concentrations (McKinney et al., 2017).

Braune et al. (2014) reported the temporal trend of Hg in thick-billed murre (*Uria lomvia*) eggs from Coats Island, northern Hudson Bay, and Prince Leopold Island in Lancaster Sound, Nunavut. Although there was no significant change in Hg concentrations in murre eggs from Coats Island from 1993 to 2013, $\delta^{15}\text{N}$ values for the eggs were found to be decreasingly significantly, suggesting a decline in trophic position for the bird due to a dietary switch from Arctic cod (*Boreogadus saida*) to capelin (*Mallotus villosus*). After adjusting egg Hg concentrations for the decline in trophic position, time trends in Hg concentration at Coats Island changed from non-significant to significantly increasing. In contrast, at Prince Leopold Island, after adjustment for trophic position the egg Hg time trends changed from non-significant to significantly decreasing over the same period. These results suggest that in addition to trophic change in diet, there may have been other geographic factors at play that influenced Hg concentrations at the base of the marine food web, such as differences in Hg deposition, or in Hg bioavailability related to climate change.

Subsequently, Braune et al. (2016) updated the Hg trends in High Arctic seabird eggs at Prince Leopold Island to 2014 for five species: thick-billed murre, northern fulmar (*Fulmarus glacialis*), black-legged kittiwake (*Rissa tridactyla*), black guillemot (*Cepphus grylle*), and glaucous gull (*Larus hyperboreus*). The first three species' eggs had been collected from the Island as early as 1975, while the guillemots and gulls were sampled from 1993 to 2013. Egg Hg trends were adjusted for possible shifts in trophic position of the birds using $\delta^{15}\text{N}$ data. Adjusted Hg concentrations in eggs of murre, fulmars and kittiwakes increased from 1975 to the 1990s, followed by a plateauing or slight decline from the 1990s to 2014 (Fig. 6A). However, the kittiwake trend was strongly influenced by the 1975 samples; when these were excluded, kittiwake eggs displayed a significant decreasing trend from 1976 to 2013. Trends in the eggs of murre, fulmars, kittiwakes, and guillemots had negative slopes between 1993 and 2013. The pattern in glaucous gull eggs was different: decreasing by 50% from 1993 to 2003 before increasing again.

Braune et al. (2016) concluded that the general increasing trends in egg Hg during the 1970s and 1980s were consistent with atmospheric Hg increases over the Arctic during that period. They noted that the migratory habits of the five bird species, which overwinter in different southern regions away from Lancaster Sound, complicated interpretation of the reasons for the temporal trends. Environmental Hg changes in their wintering areas could have been different to those in the Arctic. Interpretation is also complicated by significant differences in the findings from glacier archives of atmospheric Hg on the western and eastern edges of the North American Arctic.

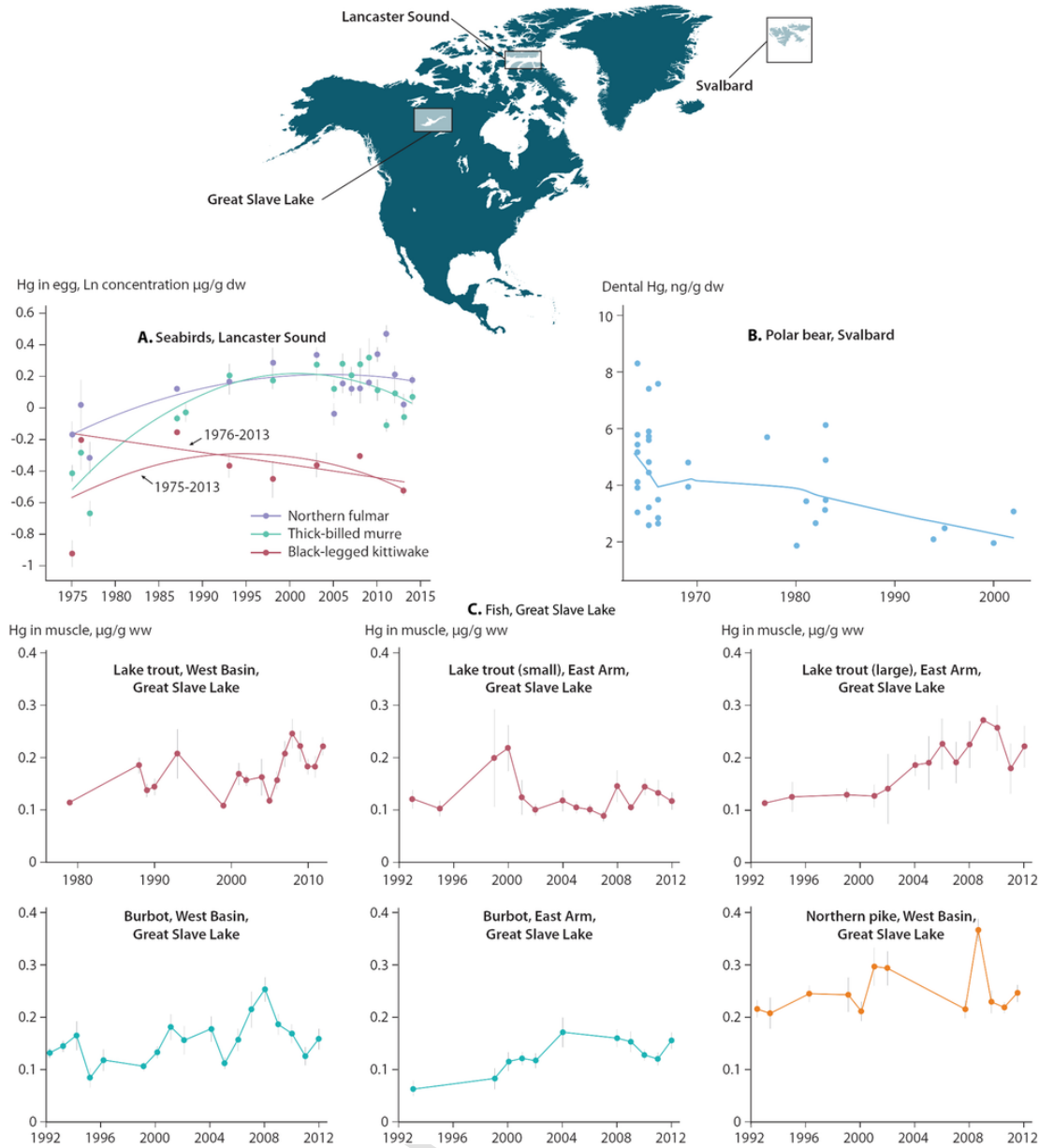


Fig. 6. Mercury trends in Arctic aquatic biota. A) Annual mean Hg concentrations (dry weight (dw); natural logarithm-transformed) adjusted for trophic position in eggs of thick-billed murre, northern fulmar, and black-legged kittiwake from Lancaster Sound, Nunavut, from the 1970s to the 2010s (Braune et al., 2016). Vertical lines show \pm one standard error, and the trend lines represent second-order polynomial regressions for each of the species. B) Mercury concentrations (dry weight) in teeth of polar bears from Svalbard, aged from three to 10 years, from the 1960s to the 2000s (Aubail et al., 2012). The line represents the fitted non-linear trend. C) Mercury concentrations (wet weight) in muscle of burbot and lake trout collected from the West basin and east Arm of Great Slave Lake, Northwest Territories, Canada, from the 1980s to 2010s (Evans et al., 2013). Vertical lines show \pm one standard error.

Greenland glacial snow/firn (Faïn et al., 2009) showed a monotonic decline in atmospheric GEM concentrations during the 1970s and 1980s, following peak concentrations in the 1950s to 1960s. Glacial snow and ice core reconstructions of atmospheric Hg deposition from Mt. Logan (Yukon) showed increases in deposition through the 1990s, which could be an indication of increasing trans-Pacific contamination from Asia (Beal et al., 2015). Overall, these data, especially the declining GEM trend on Greenland through the 1970s and 1980s, are inconsistent with the conclusions of Braune et al. (2016). However, the flat or slightly declining egg Hg data from about 1990 onwards is consistent with the recent modelling of atmospheric GEM in the Arctic (see Fig. 1). Zheng (2015), on the other hand, reported that

20th-century total Hg accumulation in a Greenland ice core was relatively constant until increasing during the 1970s to 2000s, a pattern similar to those in most of the bird species but not in agreement with the (Zhang et al., 2016) modelling. Thus, uncertainty about the actual trends in Arctic atmospheric Hg deposition is a limiting factor in assessing agreement between environmental and biological Hg trends in this region.

In Great Slave Lake in the western Canadian Arctic, temporal trends of Hg in lake trout, burbot, and northern pike were monitored irregularly between the late 1980s or early 1990s and 2012 (Fig. 6C) (Evans et al., 2013). Muscle Hg data were adjusted for fish length, but not for trophic shifts over time. Mercury concentrations generally

increased over time in lake trout and burbot, but not in northern pike, with considerable interannual variation. These increasing or flat patterns are inconsistent with atmospheric GEM concentrations and wet deposition fluxes that were declining at the time (see Fig. 1) and with the Mt. Logan atmospheric deposition record of Beal et al. (2015). Statistical analysis of climate factors suggested that varying annual mean air temperatures, and particularly cold season temperatures, were related to the fish Hg patterns although a precise mechanism linking temperature to fish Hg could not be elucidated (Evans et al., 2013).

4. Causes of the divergence between aquatic biota and atmospheric Hg trends

There are marked differences between century-scale and recent decadal datasets in terms of how well atmospheric and biotic Hg trends track each other. The century-scale biotic Hg trends since about 1850 (from the Arctic; Dietz et al., 2009) generally matched the increasing atmospheric Hg deposition trends recorded in remote glacial ice cores and other natural archives (Beal et al., 2015; Kang et al., 2016; Zheng, 2015). Starting in the mid- to late-19th century, and shortly after major anthropogenic uses and global-scale emissions of Hg became more common (Outridge et al., 2018), Hg concentrations in the atmosphere and in aquatic biota increased steadily up to maxima typically attained at about the 1970s–1980s (Dietz et al., 2009; Rigét et al., 2011). The longer-term biotic datasets from the Arctic thus clearly indicate the effects of anthropogenic contamination of aquatic systems over the century or more since about 1850.

However, as anthropogenic emissions to air began to stabilize after the 1980s, it became increasingly apparent that a divergence between the biotic and atmospheric Hg trends was developing in some areas, and in some co-occurring species. In other areas and species, however, the atmospheric and biotic trends continued synchronously. The divergent patterns between the atmospheric and biotic Hg trends have become more apparent over the past two decades, as atmospheric and biological monitoring became more widespread and frequent.

Fundamentally, the divergence between the biotic and atmospheric mercury trends may be generally attributed to the large inventories of legacy Hg in soil and ocean reservoirs, and the exceptional sensitivity of Hg biogeochemical cycling to changes in physical (e.g., temperature, light, hydrology), geochemical (e.g., pH, redox status, complexing ligands), biological (e.g., feeding behavior of an organism) and ecological (e.g., primary productivity, microbial processes, food web structure and dynamics) conditions (Table 1; see also Hsu-Kim et al., 2018). Many of the effects have been recently exacerbated by climate change. Some of the major processes that trigger changes in these conditions, and thus cause the decoupling between biotic and environmental Hg, include changes in the terrestrial environment and landscape, changes in the aquatic ecosystem, and climate change.

4.1. Changes in the terrestrial environment and landscape

Globally the terrestrial environment represents the largest inventory of Hg (~950kt), with ~150kt stored in surface organic soils (Outridge et al., 2018). Terrestrial biota are much less prone to Hg bioaccumulation as the conversion of inorganic Hg to MeHg is not favored in the terrestrial environment (Fitzgerald and Lamborg, 2014). Also, on a global scale, anthropogenic inputs have altered the Hg inventory in the soils to a lesser degree than in the oceans due to the naturally large mass of Hg that is present in terrestrial systems (Outridge et al., 2018). However, major changes in landscape and

Table 1
Properties of mercury (Hg) and their implications for its biogeochemistry.
(Modified from Wang and Zhang, 2013).

Property	Implications
Redox between Hg ⁰ and Hg ^{II}	Sensitive to changes in redox and pH conditions; sensitive to photochemical and microbial processes
High vapor pressure of Hg ⁰	Sensitive to changes in temperature; long-range atmospheric transport; a global problem needing global solutions
Hg ²⁺ ions being one of the softest Lewis acids	Strong affinity to ligands (e.g., reduced sulfides, halogens); sensitive to changes in organic carbon and redox conditions
Methylation is primarily microbial, with methylmercury being the most bioavailable and toxic	Sensitive to changes in temperature, organic carbon, nutrients, redox and microbial processes; direct source control of methylmercury difficult
Methylmercury biomagnifies in the food chain	Sensitive to changes in temperature, food web structure and dynamics

land-use in the watershed, such as urbanization, agricultural activities, flooding, damming, and deforestation, may affect the net release of soil Hg to downstream aquatic systems (inland and coastal). More importantly, they also change the organic carbon flux from watersheds and redox conditions that directly influence Hg methylation processes in adjacent aquatic systems. The importance of such changes is perhaps best demonstrated by reservoir construction in the watershed. As shown in Case Study 3, fish Hg concentrations in reservoirs are primarily controlled by the influx and dynamics of terrestrially-derived organic carbon and Hg, and bear almost no relationship with trends in atmospheric Hg concentrations or deposition, even decades after reservoir impoundment.

4.2. Changes in the aquatic ecosystem

As MeHg biomagnifies in the aquatic food web (i.e., MeHg concentration increases from prey to predator), any changes in ecosystem structure, function, and dynamics may result in major changes in Hg concentrations within the ecosystem. Processes such as acidification/liming (Case Study 2) and eutrophication (see Case Study 3) affect MeHg production by altering Hg speciation and bioavailability, as well as Hg food-chain transfer and biotic Hg concentrations by altering species composition, biomass and growth rates (e.g., Clayden et al., 2013; Jardine et al., 2013). Aquaculture, overfishing, and invasion of non-native species can change not only the nutrient status of an aquatic ecosystem, but also the structure, function, and dynamics of food webs, and thus could result in major changes in biota Hg.

4.3. Climate change

On the global scale, climate change may be the most prevalent contributor to the divergence between biotic and environmental Hg trends. Changes in water temperature alone could affect mercury bioaccumulation in various ways. For instance, temperature increases could lead to changes in bioenergetics of primary consumers and fish that leads to higher ingestion but has less effect on elimination processes, resulting in an increase in biota Hg concentrations. The impact of climate change on biotic Hg is perhaps most profoundly felt in the Arctic, where rapid climate warming has resulted in dramatic changes in many biogeochemical and ecological processes that drive Hg cycling (Chen et al., 2018; Stern et al., 2012; Wang et al., 2010). For instance, the rapid decline in the aerial coverage and thickness of Arctic sea ice and the replacement of multi-year sea ice by first-year sea ice have been shown to influence Hg distribution

and transport across the ocean–sea ice–atmosphere interface, as ice prevents elemental Hg evasion and leads to a buildup of Hg⁰ under the ice (Dimento et al., 2018; Andersson et al., 2008), and can alter Hg methylation and demethylation rates, promote changes in primary productivity, and shift food web structures (bottom-up processes) (Beattie et al., 2014; Chaulk et al., 2011; Heimbürger et al., 2015; Wang et al., 2017). Arctic tundra plants have been shown to actively take up GEM from the atmosphere, leading to unexpectedly high Hg accumulation in tundra soils (Obriest et al., 2017). A warming climate thus will not only affect the tundra accumulation of atmospheric Hg, but also the amount of Hg that can be delivered from the watershed to the Arctic freshwater and marine ecosystems via snow melt and riverine discharge. The very large mass of mainly natural Hg in northern permafrost deposits, which is projected to be steadily released with further climate warming (Schuster et al., 2018), may profoundly affect biotic Hg concentrations around the Northern Hemisphere especially as large amounts of organic carbon that may stimulate Hg methylation rates will be simultaneously released.

Changes in animal social behavior associated with changing sea-ice regimes can also affect dietary exposure to Hg (top-down processes) (Stern et al., 2012). As shown in Case Study 4, the trophic position of thick-billed murre from Coats Island in northern Hudson Bay has declined, presumably due to feeding increasingly on capelin instead of Arctic cod (Braune et al., 2014). However, the population's egg Hg concentrations did not change significantly between 1993 and 2013; to explain this stable trend, the availability of MeHg in the environment and efficiency of Hg food web transfer must have increased. It has also been suggested that climate warming may cause a shift in energy flow from benthic to pelagic food webs as aquatic productivity increases in high Arctic lakes. Since zooplankton species such as *Daphnia* contain higher MeHg than benthic organisms, this shift could increase Hg transfer in the food web (Chetelat and Amyot, 2008). The impact of climate change on biotic Hg has also been observed in lower latitude regions (e.g., Pinkney et al., 2014).

5. Evolution of the relationship between atmospheric and biotic Hg concentrations

Wang et al. (2010) and Wang and Zhang (2013) proposed that the divergence between biotic and environmental Hg trends is an indication that an aquatic ecosystem has entered a new 'paradigm' in which the key controls on Hg bioaccumulation have switched from being 'emissions-driven' to 'processes-driven'. This switch occurs because the biotic Hg concentrations in an aquatic ecosystem are influenced not only by Hg influx (natural or anthropogenic) to the system, but also by the internal processes in the ecosystem that control the recycling, speciation, bioavailability, methylation and biological uptake of Hg. As the accumulated mass of 'legacy' Hg in a water body becomes large relative to the loading rate of newly emitted Hg, the internal biogeochemical processes that control its permanent removal (e.g., burial), re-emission, or uptake into the biota, increasingly become the determining steps in bioavailability and bioaccumulation.

The changing relationship over time between atmospheric Hg concentrations (or deposition) and biotic Hg is shown in Fig. 7. Prior to anthropogenic influences (the exact timeline is subject to debate; Outridge et al., 2018), when Hg emissions were at their natural level, the flux of Hg to the aquatic system was generally low, and so were its biotic concentrations (Phase I – 'Natural background'). At around the mid-19th century, however, as rapid industrialization resulted in a sharp increase in anthropogenic Hg emissions, aquatic biota Hg concentrations responded rapidly due to increasing Hg deposition, exposure and uptake of Hg from a small but growing environmental Hg

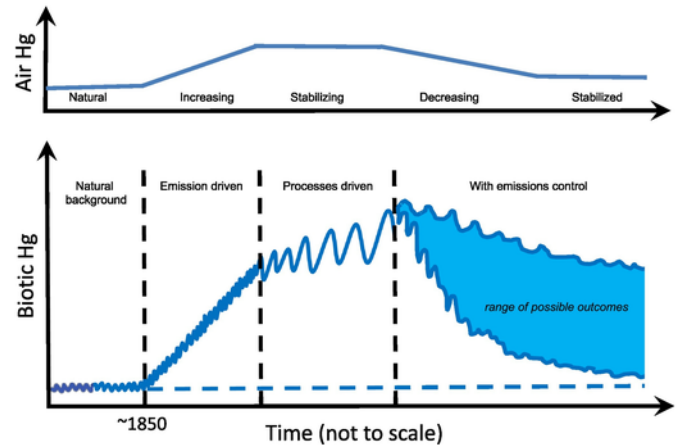


Fig. 7. A schematic representation of evolution in the Hg concentrations in the air (top) and aquatic biota (bottom), showing changes over time in the principal drivers of Hg bioaccumulation. The figure is updated from Wang et al. (2010). See the text for details.

inventory (Phase II – 'Emissions-driven'). This phase is clearly seen in long-term retrospective studies of Hg concentrations in Arctic biota (Dietz et al., 2009). Once an aquatic ecosystem has accumulated sufficient Hg, additional increases in Hg influx become secondary to the amount that has been accumulated in the system after decades to centuries of loading (i.e., legacy Hg). Bioaccumulation then draws predominantly on this legacy Hg, which is operated on by the internal biogeochemical processes (Phase III – 'Internal Processes-driven'). Throughout these three phases, biogeochemical processes (shown as sine-wave 'noise' in Fig. 7) determine the transport and transformation of Hg from the abiotic part of the ecosystem to biota. But it is in Phase III that these processes emerge to create a variability that is large enough to obscure the effect of the current external Hg inputs, and hence produce the divergence between biotic and atmospheric Hg trends (Wang et al., 2010).

In the period following the Minamata Convention, a new phase, Phase IV, can be envisioned (see Fig. 7) which is based on observations of biotic Hg trends in the recent past, as discussed above, and model predictions of future seawater Hg concentration trends. As anthropogenic Hg emissions decrease, atmospheric Hg concentrations will decrease and eventually stabilize at a new steady state. However, revolatilization of part of the large quantity of legacy anthropogenic Hg presently contained in the world's oceans and soils, and its recycling between oceans, soils and the atmosphere, means that atmospheric and seawater Hg concentrations are likely to decrease much more slowly than future changes in Hg emissions (Sunderland and Mason, 2007; Sunderland and Selin, 2013). While biotic Hg concentrations are also projected to decrease over the long term, the current phase of 'processes-driven' bioaccumulation dictates that it will take much longer to establish a new steady-state in biotic Hg.

The biotic Hg concentrations at the new steady-state are also likely to remain above the Holocene background levels for an extended period after atmospheric Hg levels have returned to natural levels, because of the continued presence of legacy Hg in aquatic and terrestrial systems. In the shorter term, in fact, aquatic biotic Hg concentrations in many instances, especially in some of the larger, deeper marine ecosystems, are likely to continue to increase despite recent emission controls (Sunderland and Selin, 2013). Biota in smaller waterbodies, such as lakes and coastal marine systems with restricted water mass turnover, are more likely to respond relatively rapidly to emissions controls because of the smaller mass of legacy Hg contained there relative to the decreased anthropogenic Hg loadings.

Examples of this long and ‘uneven’ recovery in biotic Hg can be found following the impoundment of a river, or after ‘de-acidification’ of a lake. As shown in Case Study 3, fish Hg in reservoirs decreased a few years after the impoundment, but remained above the pre-impoundment concentrations even after more than a century (see Fig. 5). In the 1970s, lime was applied to many Swedish lakes that were acidified due to atmospheric acid deposition, to help restore the lake ecosystem. Following the liming, fish Hg in those lakes declined by 10–20% by the 1990s (Meili, 1995) and continued to decline to the present day (Åkerblom et al., 2014). Yet, more than 30 years after the liming, fish Hg concentrations in these lakes remained considerably higher (twice as high on average) than those in lakes that were not impacted by acidification (and not subject to liming) (Åkerblom et al., 2014).

6. Implications for evaluating the effectiveness of the Minamata Convention

Reports of biotic Hg trends not following the atmospheric Hg trends in recent decades should not be regarded as discouraging news when considering the efficacy of regulations to reduce atmospheric and other releases of Hg. The fact that effective Hg emission control is expected to be followed by long delays, in some cases, before an ensuing reduction is seen in food web Hg concentrations makes it all the more pressing to control and reduce Hg emissions as early as possible (Chen et al., 2018; Wang et al., 2010).

Nonetheless, policy-makers should be prepared for the fact that the reductions in biotic Hg concentrations and human Hg exposure via aquatic food-chains are likely to be uneven and predictable only in the most general way, both in terms of their timing and in the degree of reduction. Different species within the same region, or the same species in different regions, can be expected to show quite different trajectories of tissue Hg over time, because of the unpredictable consequences of coincidental environmental or ecological process changes. Clearly, based on the recent literature reviewed above, no single species can be expected to reliably reflect the impact of reductions in anthropogenic emissions. Therefore, biomonitoring efforts designed to evaluate the effectiveness of the Minamata Convention in reducing biotic Hg concentrations should include multiple species occupying different ecological and geographical niches. In particular, piscivorous fish and small prey fish (e.g., young of the year perch) that are widely distributed across ecosystems and continents should be monitored, as they could serve as a better proxy of change (Mason et al., 2005; Wiener et al., 2007). Continuation of existing biological Hg monitoring programs should focus on supporting the longest and most statistically powerful datasets, because these will reflect any future changes in concentrations with greater confidence than short-term or high-variance datasets (Bignert et al., 2004). The collection of essential ancillary biological and ecological data (such as body length or weight, and trophic position using stable nitrogen isotope ratios), which will help to interpret observed changes in tissue Hg, should also be mandatory.

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