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Improved environmental status: 50 years of declining fish mercury levels in boreal and subarctic Fennoscandia

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ABSTRACT

Temporally (1965-2015) and spatially (55°-70°N) extensive records of mercury (Hg) in freshwater fish showed consistent declines in boreal and subarctic Fennoscandia. The database contains 54560 fish entries (n: pike>perch>>brown trout>roach≈Arctic charr) from 3132 lakes across Sweden, Finland, Norway, and Russian Murmansk area. 74% of the lakes did not meet the 0.5 ppm limit to protect human health. However, after 2000 only 25% of the lakes exceeded this level, indicating improved environmental status. In lakes where local pollution sources were identified, pike and perch Hg concentrations were significantly higher between 1965 and 1990 compared to values after 1995, likely an effect of implemented reduction measures. In lakes where Hg originated from long-range transboundary air pollution (LRTAP), consistent Hg declines (3-7‰ per year) were found for perch and pike in both boreal and subarctic Fennoscandia, suggesting common environmental controls. Hg in perch and pike in LRTAP lakes showed minimal declines with latitude, suggesting that drivers affected by temperature, such as growth dilution, counteracted Hg loading and foodweb exposure. We recommend that future fish Hg monitoring sampling design should include repeated sampling and collection of supporting information (pollution history, water chemistry, fish age, stable isotopes) to enable evaluation of emission reduction policies.

KEYWORDS

Atmospheric pollution; climate; Convention on Long-Range Transboundary Air Pollution; freshwater; Minamata Convention; point source pollution

INTRODUCTION

In 1956, the occurrence of the Minamata Bay accident in Japan initiated intensive research and monitoring of mercury (Hg) concentrations in fish used for human consumption. The accident was caused by releases of the neurotoxic Methyl-Hg (MeHg), which was biomagnified in aquatic food webs and has since proved to have harmful effects on aquatic organisms[1] and their consumers[2], including humans[3, 4]. Although the toxic effects of Hg have been known for more than half a century[5], our ability to predict impacts of changed Hg emissions on exposure, accumulation, and biomagnification of Hg in food webs remains limited because of the complex biogeochemical cycling of Hg. Thousands of freshwater lakes worldwide have fish Hg concentrations exceeding limits advised for human consumption (0.3 – 1.0 ppm Hg wet weight (w.w.))[6]. Freshwater fish are considered being critical receptors of long-range transboundary air pollution of Hg[7]. The *Minamata Convention on Mercury* (hereafter *Minamata Convention*) aims to protect human health and the environment from adverse effects of Hg at a global scale[8]. The agreement requires the parties to evaluate its effectiveness, based on information and reporting, including adequate methodologies to detect trends of Hg concentrations in biota[8].

In Fennoscandia, environmental monitoring of Hg was initiated in the mid-1960s, following the awareness of use of Hg in paper and pulp mill factory processes (from the 1960s to the 1980s)[9, 10]. Initially, monitoring was focused on lakes close to known point sources of Hg, but during the 1980s it was revealed that lakes in remote and pristine areas were exposed to increased loads of predominantly atmospherically deposited Hg[11, 12]. High levels of Hg in monitored fish initiated new environmental legislations, including changes in the forest industry processes, and local emissions and releases were generally reduced[9]. Still, several Northern areas show significant increases in fish Hg concentrations the last decades, including Sweden[13], Finland[14], and Ontario (Canada)[15], although this rising trend is not found in all regions and for all fish species. In fact, a study of lakes in Sweden[16] shows declining Hg concentrations in fish between 2005 and 2015, something which fits with the observed declining trend of Hg deposition since at least the 1990s throughout Europe[17]. However, most studies, including the examples mentioned here, usually provide no or very limited information on local pollution history (i.e. whether Hg catchment input is of local and/or long-range origin)[18]. Another limitation in most of the studies available in current literature is that temporal fish Hg trends are analysed within country or state borders rather than per bio- or ecoregions which are potentially more meaningful regarding controls of biochemical Hg cycling such as climate and deposition[17, 19].

In many boreal, subarctic, and Arctic lakes in Fennoscandia, long-range atmospheric transport of Hg is the main source of Hg contamination[11] and has led to long-term accumulation of Hg in catchments[20], similar to remote areas in North America[21]. Deposited Hg reaches surface waters either gradually through enrichment of soils[21] and subsequent leaching (transported by organic matter, OM) to surface waters[3, 22], or as direct deposition to the lakes. The gradual release contrasts with point source releases of Hg to the environment, and leaching of Hg from catchment soils is controlled by a range of environmental drivers, characteristics, and processes which in their turn potentially affect food web exposure to Hg and subsequent bioaccumulation (summarised by Driscoll et al., 2013[3]). In order to document the effectiveness of global Hg emission reduction measures, established under the Minamata Convention[8] and the Convention on Long-Range Transboundary Air Pollution (CLRTAP)[23], and to distinguish their effects from earlier legislation, it is useful to attribute key sources of Hg pollution (i.e. long-range versus local) in different water bodies.

We examined a 50-year database of >50 000 measurements of Hg in freshwater fish across wide climate, geography, and deposition gradients in Fennoscandia (Norway, Sweden, Finland, and the Murmansk area in Russia). We evaluated temporal trends and spatial patterns of Hg concentrations for fish species with different foraging and thermal guilds, and assessed temporal trends related to predominant sources of Hg for the lakes, i.e. local point industrial sources (*point source lakes*) and long-range atmospherically transported Hg (*LRTAP lakes*, referring to CLRTAP[18]). Hypothesizing that fish Hg trends in LRTAP lakes, directly or indirectly, are sensitive to environmental drivers, including climate (temperature)[24-26], lake browning, and atmospheric deposition (especially Hg and sulphur, S[27-29]), we also tested for temporal trends of Hg in LRTAP lakes in southern (boreal) and northern (subarctic) ecoregions. The results are placed in a context of demands for suitable monitoring programmes to evaluate policies aimed to reduce global Hg pollution.

MATERIALS AND METHODS

Selection of data

Records of total Hg measurements in freshwater fish muscle tissue from Sweden, Finland, Norway, and the Murmansk Oblast (i.e. a federal subject) in Russia were collated from literature and existing databases. Records that did not meet a set of criteria, including availability of Hg content, fish weight and fish length, and a minimum of five records for a single fish species per lake, were excluded (11904 of initially 66464 individual records, see ***Figure S1*** in *Supporting Information*). Relations between Hg concentrations and fish size, length and/or weight, and length-weight relationships were tested for further quality assurance[30]. Following these relations, residual outliers (i.e. entries outside 75% quartile plus 1.5*interquartile range, n = 70) were excluded. The database was limited to fish species that are typically distributed in all the four countries, resulting in records of Northern pike (*Esox lucius*, 42.4 %), perch (*Perca fluviatilis*, 34.1 %), Arctic charr (*Salvelinus alpinus*, 1.2 %), brown trout (*Salmo trutta*, 3.1 %), and roach (*Rutilus rutilus*, 1.3 %). Finally, the database consisted of 54560 entries from 3132 lakes (***Figure S1***), collected between 1965 and 2015, spanning a south-north gradient from 55.50° N in Sweden to 70.03° N in Norway, and a west-east gradient from 6.00° E in Norway to 37.37° E on the Kola Peninsula (Murmansk, Russia, ***Figure 1***).

The fish species differ in their thermal and foraging guilds[31, 32]. Arctic charr, brown trout, perch and roach are generalist species that may forage across both pelagic and littoral habitats. The cold-water adapted Arctic charr and brown trout are present in oligotrophic lakes; the cool-water species perch is often the dominating species in mesotrophic lakes, and the warm-water species roach are abundant in eutrophic lakes[33]. Arctic charr, brown trout and perch undergo ontogenetic dietary shifts from invertebrates to fish prey, but roach feed exclusively on invertebrate prey[32, 34]. Pike is a cool-water obligate piscivore that historically has been a key species, together with perch, for Hg monitoring due to its wide distribution range, location at the top of food webs (i.e. combining both littoral and pelagic energy sources due to its capacity to feed on all available prey fish species in lakes[34]), and its importance for recreational fishing.

Pike and perch were the most abundant species in the database, both spatially and temporally, and they were selected for detailed analyses in this work. In the database, pike size (i.e. weight) centre around 1 kg (mean ± one standard deviation: 998 ± 579 g; median: 905 g), historically a target size for many Fennoscandian Hg studies[16]. Because perch undergo an ontogenetic dietary shift from invertebrates to fish[34], it is important to consider different size groups in the data analysis. In our dataset, there was a significant decrease in perch size between those collected before year 2000 (140 ± 176 g) compared to those collected in year 2000 and later (61 ± 79 g, ***Figure S2***). This shift in size for collected fish is likely related to either sampling gear (i.e. a change in gill nets from traditional large mesh gill nets to Nordic nets including small mesh sizes (<12 mm)), or sampling strategy (i.e. increased focus on small, remote lakes with slow-growing perch). We have therefore chosen a selection of perch sizes, including weights of 65-95 g (14-25 cm), to assess the potential trends in our data set. The size selection of 65-95 g is based on *i*) the prevalence of these sizes throughout the whole database time-period 1965-2015 (***Figure S2***); and *ii*) that the fish of these sizes have likely undergone an ontogenetic shift to become piscivory[32].

Classification of lakes – point pollution sources versus long-range atmospheric deposition

Lakes were classified per dominant Hg pollution source based on expert judgement: 1. *Lakes with no local Hg pollution sources,* implying that atmospheric deposition of Hg is the dominating pollution source (hereafter *LRTAP lakes*); 2. *Lakes with known local industry point source(s)* (hereafter *point source lakes*); and 3. *Unknown*. We did not classify per timing of contamination. In the current work, n = 167 lakes (n = 13938 specimens) were classified as being *point source lakes*, while n = 474 lakes (n = 14072 specimens) were classified as being *LRTAP lakes* (***Figure S1*** and ***Figure 1***).

Classification of lakes – boreal and subarctic ecoregions

We divided the LRTAP lakes into subarctic (> 65° N) and boreal (< 65° N) (***Figure 1***), a simplified classification following De Wit et al. (2016)[35]. The regions contrast each other with respect to atmospheric pollution (e.g. total Hg and S, primarily as oxidised S or SO_4)[36], temperature, and aqueous OM concentrations. Deposition of Hg and S is lower in the subarctic region compared to the boreal, and the subarctic lakes are colder and less coloured, i.e. lower OM concentrations. Deposition of SO_4 has been shown to promote methylation[27, 37] and lately reduced acid deposition (primarily of SO_4) has been shown to promote increased browning of surface waters[29]. Temperature determines fish growth with subsequent effects on Hg concentrations in muscle via dilution and condensation cycles[33, 38], but temperature also controls terrestrial productivity and thus regional variation in aqueous OM[39]. OM is a transport vector for Hg[22, 40], but can also reduce photo-demethylation[41] and bioaccumulation[42].

Data treatment

Covariation between Hg concentration and fish size (length and weight[43, 44]) and age[45] requires a standardization to allow for investigation of spatial and temporal trends of Hg concentrations. We used the individual fish weight and Hg concentration in combination with fish species information and sampling year to find the modelled (i.e. expected) Hg concentration for fish at a standard weight. Different linear regression models were applied to describe the log[Hg] concentrations (Supplementary information, ***Table S1***), where potential explanatory variables included fish weight, fish species, sampling year, and the interaction terms year x species and weight x species, to evaluate changes in fish Hg concentrations with weight and species over time.

The standardised fish Hg data were used to calculate *annual lake-specific medians* (ALMs) for each fish species (***Table S2***), which were used in further statistical analysis. Long-term temporal trends in fish Hg concentrations were investigated through linear regression models of the ALMs, by fish species, pollution history, and ecoregions. Differences in regression coefficients were tested using multiple linear regression models (MLR, ***Equation 1***).

$$\log \text{ALM} = \alpha + \beta^*\text{year} + \gamma^*Z + \delta^*\text{year}^*Z + \varepsilon \qquad \textbf{(1)}$$

where α represent the intercept, β the partial regression coefficient for time, γ the indicator variable of groups representing either fish species (perch and pike) or lakes subject to Hg pollution from different sources (LRTAP and point source lakes), δ the interaction between time and indicator variable, and ε the random error. Including δ for different groups (Z) enabled the comparison of regression coefficients between ecoregions and fish species by a t-test to test the difference of the temporal trend slopes.

Latitudinal gradients in ALM fish Hg concentrations were tested separately for pike and perch using the Pearson product-moment correlation coefficient. A probability for each correlation coefficient was used to estimate the significance for each gradient. To test for differences between grouped data, *Analysis of Variance* (ANOVA) models were applied, where the groups (Z, fish species and Hg pollution sources) were included as fixed variable and lakes as a random variable. A significance level of $p = 0.05$ was used.

RESULTS AND DISCUSSION

Fennoscandic fish Hg concentrations (observed data)

Consumption of fish is considered the main Hg exposure route to humans and wildlife[46, 47] and measures taken under the CLRTAP[23], the Minamata Convention[8], and the EU Water Framework Directive (WFD) are therefore targeted to improve the quality of aquatic ecosystems with respect to Hg. Fish Hg concentrations in lakes across Fennoscandia generally have concentrations that exceed maximum limits set to protect human health (0.3-0.5 ppm w.w., ***Table S2***)[6, 48]. In the Fennoscandian fish database, pike had the highest mean ALM concentration (0.67 ppm), with Arctic charr (0.37 ppm), brown trout (0.22 ppm), perch (0.29 ppm), and roach (0.37 ppm) having lower concentrations. Pike is a fish representing high trophic levels in Fennoscandic freshwater food webs and an obligatory piscivore feeding on all types of prey fish, hence elevated Hg levels are expected[13, 16, 49]. The levels from the current work are similar to the median Hg concentrations observed in pike data from Munthe et al. (2007)[50] and Åkerblom et al. (2014)[16]: 0.69 (1965-2004) and 0.68 ppm (1966-2012), respectively.

The large majority of fish caught in Fennoscandia over the last six decades shows observed Hg concentrations above the WFD Environmental Quality Standard (EQS) of 0.02 ppm[51]. Of the 54560 fish samples included in the entire database, 99.8% had concentrations above 0.02 ppm, and good chemical condition is not met for any water body. Hg is a priority substance under the WFD, where protection from biomagnification in the food chain (i.e. top predators including fish and wildlife) is a main aim (i.e. "secondary poisoning"). For Hg, the EQS is based on a 365 days *No Observed Effect Concentration* (NOEC) for MeHg, and a (relatively low) assessment factor of 10 is applied due to the large number of NOECs available for MeHg[51]. Although the WFD EQS *secondary poisoning* for Hg in biota has relevance for assessing the risks of ecosystem Hg exposure in Fennoscandia, it does not differentiate between lakes with higher and lower Hg risks. A different threshold for Hg in Fennoscandian fish is the limit to protect *human health* of 0.5 ppm[51], where 74% of the water bodies in our database would *not* meet this criterion. However, if only samples collected after year 2000 are considered, the relative number of lakes with an individual fish Hg concentration above 0.5 ppm is 25%, testifying to improved environmental status in Fennoscandia.

Fish Hg concentrations in relation to atmospheric Hg deposition and local sources

Abatement measures introduced to reduce emissions and releases from industry, including closure or removal of Hg releasing facilities, may have been very effective, but previous pollution has left legacy Hg in soils or lake sediments[9]. Thus, lakes with historical local Hg sources are likely to add an additional concentration signal compared to lakes only influenced by long-range atmospherically transported Hg. Lower fish Hg concentrations in LRTAP lakes (LRTAP lakes: 0.28 ± 0.16 ppm, n = 474 lakes, mean ± one standard deviation of ALMs, all five species) compared to point source lakes (0.46 ± 0.22 ppm, n = 167 lakes, ANOVA: F-ratio=116, p<0.0001, r^2=0.15) support this hypothesis. The same pattern is evident on individual fish species level for the two main fish species in the database (***Table S2***). Despite a large body of evidence suggesting that between-lake variation in fish Hg levels is controlled by catchment and foodweb characteristics (including fish species composition), in addition to climate[52], our database indicates that pollution sources matter, i.e. that atmospheric pollution has resulted in much lower Hg loading to lakes than point sources, and therefore lower Hg in fish. As an illustration, a small lake (0.5 km^2) catchment (5 km^2) without a local pollution source, with a yearly atmospheric (10 µg Hg m^{-2} y^{-1}) and catchment (2.5 µg Hg m^{-2} y^{-1}) input[20, 53] of Hg from long-range atmospheric pollution receives total annual inputs of 17.5 g Hg. To put this into perspective, examples on abatement measures in Fennoscandia include a chlor-alkali plant that released from three to five tons of Hg annually to Lake Vänern, Sweden, before new legislations were introduced in the 1970s and 1980s[10], and a sulphide ore smelter emitting 3.5 tons of Hg annually to air in Northern Sweden in the late 1960s[54].

For the point source lakes, the temporal trends in ALMs showed a significant long-term decreasing trend between 1965 and 2015 (perch: annual decrease (ad)=-8‰ $year^{-1}$, p<0.001, pike: ad=-4‰ $year^{-1}$, p<0.0001). However, since 1995, the temporal trends are not significant (perch: ad=-1‰ $year^{-1}$, p=0.73, pike: ad=-4‰ $year^{-1}$, p=0.36), indicating that most of the change in concentrations happened earlier (***Figure 2***). In Fennoscandia, chlor-alkali industry can be recorded back to at least the 1920s[55], and a peak in industry emissions and releases are assumed to have occurred during the 1950s and 60s, when 20 to 30 tons of Hg were discharged annually from point sources in Sweden[56]. Since the 1980s local emissions and releases in Fennoscandia were reduced significantly[57]. In Norway, the official governmental total emissions to the atmosphere and releases to soil and water have declined from 5.0 tons in 1985 to 2.5 tons in 1995 and 0.9 tons in 2005[58]. These declines fit well with the temporal fish Hg data from the point source lakes, where there is a significant difference between samples collected in 1990 or earlier and those collected in 1995 or later for both perch (65-95 g, 0.47 ± 0.12 ppm and 0.21 ± 0.03 ppm, ANOVA: F-ratio=352, p<0.0001, r^2=0.60) and pike (0.69 ± 0.10 ppm and 0.55 ± 0.14 ppm, ANOVA: F-ratio=188, p<0.0001, r^2=0.22) (***Figure 2***). The reasons for the decline in discharge and emissions in Scandinavia are, in addition to regional and national control legislation, improved technology, and reduction of polluting industrial production[56].

For the LRTAP lakes, the temporal decrease in ALMs were significantly larger for perch compared to pike (perch: ad=-7‰ year^{-1}, p<0.0001, pike: ad=-4‰ year^{-1}, p=0.0032) (***Table S3A***). This difference in trends between the fish species could indicate that perch and pike respond differently to changes in factors that relates to Hg biomagnification, potentially as a consequence of biological and ecological differences between species pike[59-61] and perch[62, 63]. To examine such differences between fish species, and to disentangle the cause for the different magnitude of decreases in fish Hg concentrations over time, data on age[45] and trophic level indicators (i.e. stable isotopes of nitrogen, N[64]) would be necessary[32, 33].

No other studies of temporal Hg trends exist, covering such a large geographical area with understanding of sources of Hg contamination. Our trends are only partially supporting findings from large North American fish databases. Similar to this study, Eagles-Smith et al. (2016)[65] show that fish Hg concentration trends are declining from 1969 to 1977 in a study from the Western US and Canada (n = 96310 specimens, n = 4262 locations), but show no trend from 1978-2012. In two studies from Ontario, Canada, Gandhi et al. (2014)[15] reveal declining or unchanging fish Hg concentrations between the 1970s and 2012 (n = 31743 specimens, n = 1167 locations), depending on the fish species considered, and Tang et al. (2013)[66] found no significant decline between the time periods 1974-1981 and 2005-2010 (n = 5215 specimens, n = 73 locations). For a more recent time period, Zhou et al. (2017)[67] demonstrate declining fish Hg concentrations between 2004 and 2015 for specimens of lake trout (*Salvelinus namaycush*) from the Laurentian Great Lakes (n specimens unknown, n = 8 locations).

The Gandhi et al. (2014)[15] study was considering time trends for different predatory fish species (pike, lake trout, walleye, *Sander vitreus*) between 1970 and 2012. It was shown that while fish Hg concentrations from 1970 to 1990 were generally declining, concentrations in recent decades (time periods 1985-2005 and 1995-2012) were increasing, especially for pike and walleye. For comparison, our data shows that there is no significant trend for pike (ad=-4‰ year^{-1} in LRTAP lakes; ad=-4‰ year^{-1} in point source lakes) or perch (ad=-2‰ year^{-1} in LRTAP lakes; ad=-1‰ year^{-1} in point source lakes) in either LRTAP or point source lakes between 1995 and 2015. Gandhi et al. (2014)[15] also demonstrate overall (1970-2012) neutral or declining trends (depending on the fish species considered). A similar study as the one by Gandhi et al. (2014)[15] was done by Åkerblom et al. (2014)[16], documenting an overall long-term decline from 1965 to 2012 in Swedish pike (n = 44927).

Spatial patterns of fish Hg in boreal and subarctic Fennoscandia

For the LRTAP lakes (all species combined), fish Hg concentrations (mean ± one standard deviation of ALMs) showed a pattern where the boreal region (0.32±0.18 ppm) had significantly (p=0.017) higher concentrations than the subarctic region (0.29±0.16 ppm). As indicated in ***Table S2***, the inter-regional variation is not the same for all the fish species, and we observe that the difference between the regions is larger for pike (0.56±0.15 ppm versus 0.48±0.16 ppm, ANOVA p<0.001) than for perch (65-95 g, 0.23±0.07 ppm versus 0.21±0.05 ppm, p=0.027). The difference is surprisingly small between the ecoregions, as higher concentrations in the boreal region compared to the subarctic region was to be expected, given that elevated levels of Hg in fish often are associated with humic lakes[42, 68, 69]. In Fennoscandia there is a strong increasing west-to-east and north-to-south aqueous OM concentration gradient[70], likely to influence the fish Hg concentrations. OM can have both indirect and direct effects on Hg accumulation in aquatic food webs. Higher concentrations of OM, particularly higher molecular weight terrestrially derived OM, may reduce bioavailability of MeHg for uptake at the base of the food web[71]. However, in contrast, increased OM could also act as a substrate for increased in-situ MeHg production, with more labile algal-derived OM supporting higher methylation[72].

Relationships between observed fish Hg concentrations and aqueous OM often leaves a considerable amount of variation unexplained[12,42], and disguises other complex processes influenced by climate, catchment characteristics and biology/ecology[33]. An example is deposition of Hg, which in Fennoscandia follows a pattern of decreasing levels from south to north[17], suggesting that fish Hg concentrations in LRTAP lakes should be expected to decline with increasing latitude[73]. This hypothesis is only partly confirmed by our fish data, where concentration trends are decreasing with increasing latitude for pike (r=-0.27, p=0.0005), but where the perch data decline is not significant (r=-0.11,

p=0.078) (***Figure 3***). However, subarctic lakes typically have higher Hg biomagnification rates than lakes located further south[74], related to combined temperature effects on growth dilution and starvation[38, 75], trophic transfer efficiency and excretion rates[76]. Hence, the limited declines in fish Hg concentrations with increasing latitude observed for the LRTAP lakes, suggests that climate related effects potentially counteract Hg deposition and Hg effects from aqueous OM (i.e. increased foodweb exposure). In subarctic lakes, seasonality is much stronger than in boreal lakes located further south, likely strengthening growth dilution and starvation cycles in fish[38, 75]. In fish, the lower temperature of the subarctic region will directly reduce growth, metabolic activity, and excretion of Hg in these lakes[76].

Temporal fish Hg trends in Fennoscandia

Differences in temporal trends between ecoregions (i.e. boreal and subarctic) could potentially document to what extent fish Hg concentrations respond to changes in Hg biomagnification in LRTAP lakes. Given the strong relationships between cycling of Hg and aqueous OM[40], a naturally emerging hypothesis is that observed browning of many North American and northern European lakes[29] could influence fish Hg concentrations[42]. For both perch and pike, our data from LRTAP lakes demonstrate significantly declining trends of Hg in both boreal and subarctic regions (***Figure 4***, ***Table S4***). For perch, the annual decreases were -7‰ per year and -6‰ per year for the boreal and subarctic regions, while for pike the decreases were -3‰ per year and -5‰ per year. The inter-regional and inter-species differences in trends were not significant (***Table S3***). From a comparison of the long-term linear trend curve and the smoothed kernel curve it is obvious that the annual decrease in fish Hg levels does not represent the inter-annual and inter-decadal trends and changes in fish Hg levels (***Figure 4***). Studies investigating lake-specific increases in fish Hg during the period 1995-2005 suggests that temporal trends reflect processes in accumulation of Hg that is controlled by environmental drivers such as OM in lakes[13].

A recent study of Scandinavian lakes suggests that the largest lake browning trends between 1990 and 2013 were found in regions with strong reductions in S deposition. Hence, the change in OC concentrations was largest furthest south in the boreal wet (+1.7 % per year) and dry (+1.5 % per year) regions, and lower in the subarctic (+0.8 % per year) region[35]. A larger input of OC to lakes could influence Hg cycling in several ways, including increased loading of aqueous Hg[22], decreased MeHg degradation[77] and production[78], and increased/decreased fish bioaccumulation factors[42], all potentially affecting fish Hg concentrations. In our study, we found no evidence of significantly increasing concentrations for either perch or pike in any of the ecoregions for the same time-period (1990-2013) studied by de Wit et al. (2016) (***Figure 4***). In sum, it is challenging to document and quantify the potential influence from climatic differences and changing OC concentrations on fish Hg trends, likely because of biological and ecological factors also playing an important role.

Recommendations for the use of fish Hg databases for international environmental agreements

To evaluate the effectiveness of the Minamata Convention, there is a need for identification of legacy Hg sources and for separating these sources from long-range atmospheric sources of Hg (***Figure 2***), per the scheme in this paper. An important aspect in combining monitoring efforts for documentation of convention effectiveness would be to define regional biological species for monitoring, to minimize the effects of species-specific physiological differences. Based on the present work, especially pike would be an ideal species for this work in Northern Europe and North America, because: it is widely distributed in both continents; it accumulates significant amounts of Hg due to its position at the top of food webs; it poses a potential risk for human health via frequent consumption; and it exists in numerous historical studies. We also recommend that for future monitoring of LRTAP of Hg, relevant lakes must be selected (i.e. a selection of equal number of lakes from different ecoregions) for annual measurements of fish Hg concentrations. This will reduce the errors caused by targeting lakes impacted and affected by multiple stressors, instead of more pristine lakes.

Although fish Hg trends are declining, concentrations are still high (i.e. exceeding maximum limits set to protect human health) and effective actions are needed to solve the Hg problem. To be able to potentially explain the main drivers behind the spatial patterns and temporal trends of fish Hg concentrations, and how these patterns and trends change under influence of different and emerging drivers (environmental/climate change, deposition change, etc.), a set of minimum target information should be developed. For each location this should include lake and catchment morphology, pollution deposition patterns, and local pollution history, and for each fish species: length, weight, and age. Samples (i.e. fish muscle) for determination of total Hg concentrations, should also be analysed for stable N and carbon (C) isotopes for a better understanding of trophic position and energy sources[33,38,64]. To conclude, we stress that a deeper understanding of Hg dynamics in relations to evaluating policies aimed to reduce global Hg pollution requires long-term monitoring of fish Hg concentrations in lakes unaffected by local pollution industry.

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SUPPORTING INFORMATION

Additional figures (***Figure S1*** and ***S2***) and tables (***Table S1-S4***) referenced in the main text includes a summary scheme for data selection and organising, xy-plot of perch fish size versus sampling year, methods for fish standardisation, a summary of fish Hg concentrations, a summary of temporal trend models, and additional acknowledgements.

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FIGURES

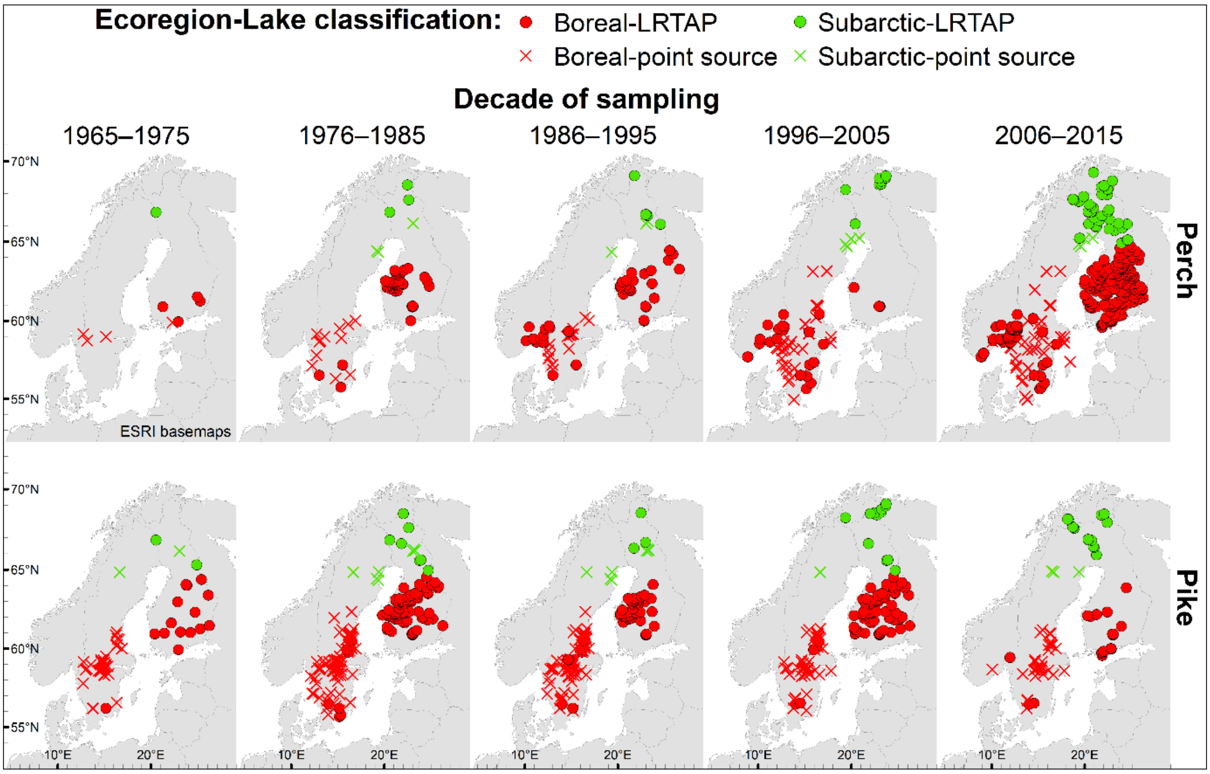

Figure 1 The geographical distribution of the LRTAP lakes (circles, n=474) and the point source lakes (crosses, n=167) and what decade they were sampled (from left to right: 1965-75; 1976-85; 1996-2005; 2006-15). Top and bottom panels show the lakes where perch and pike were represented, and the colours demonstrate the ecoregion they belong to: boreal (red) and subarctic (green).

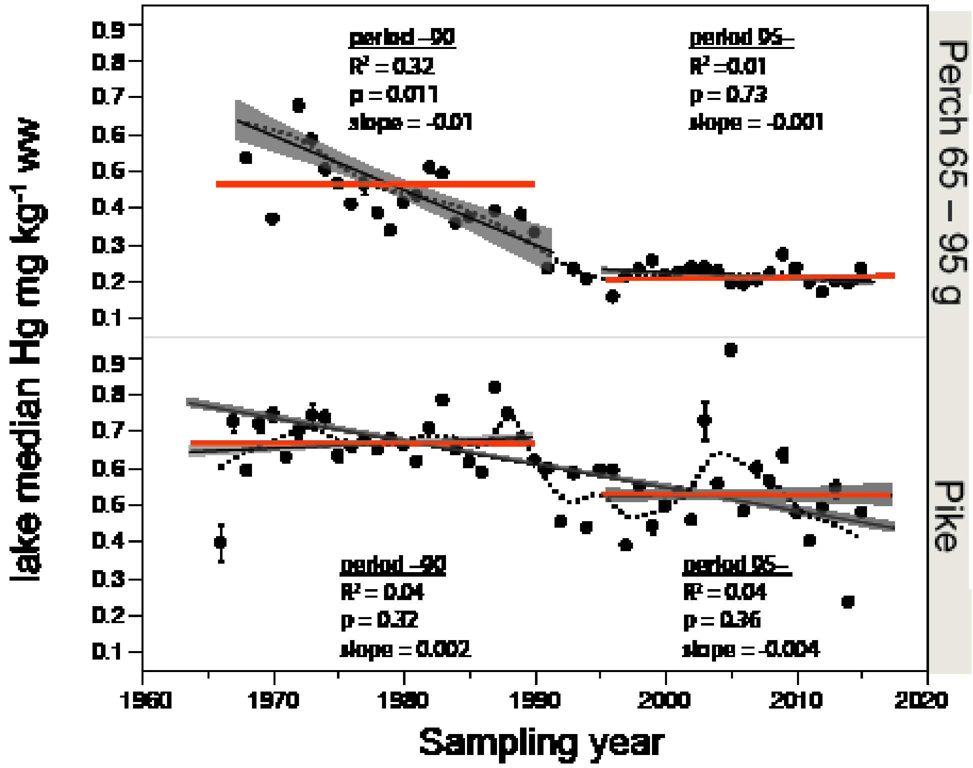

Figure 2 Temporal trends in annual lake medians (ALMs ± standard error) of Hg concentrations (wet weight, ww) of perch (65-95 g) and pike from point source lakes. The overall trends (1965-2015) are presented both as a linear regression (solid black line) and a smoothed kernel curve (dotted black line). For the separated periods 1965-90 and 1995-2015, both the linear regression (solid black line) and the mean concentration for the periods (solid orange line) are shown. 95 % confidence intervals around the linear regression lines are indicated in grey.

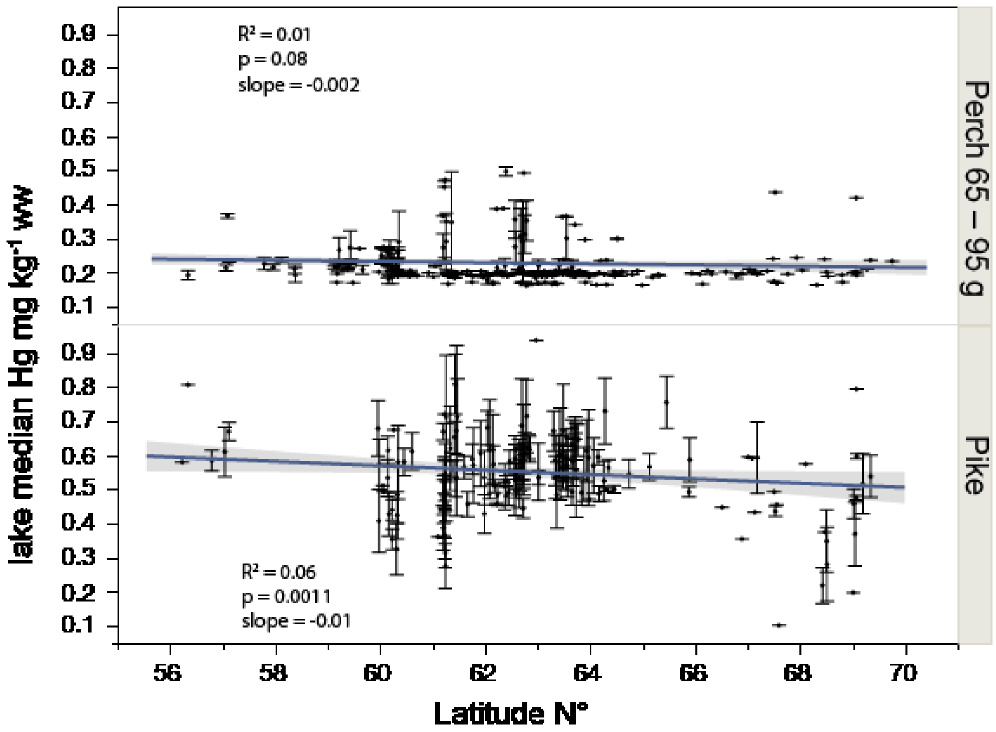

Figure 3 Latitudinal gradient in Hg concentrations (wet weight, ww) of perch (65 – 95 g, top panel) and pike (bottom panel) across Fennoscandian lakes subject to Hg loads from primarily long-range transported atmospheric pollution (LRTAP lakes). Each circle represents the mean annual lake median (ALM) for the period that each lake was sampled and error bars (standard error) represent the temporal variation for each lake. The regression lines are indicated with 95% confidence interval for a model using latitude as explanatory variable.

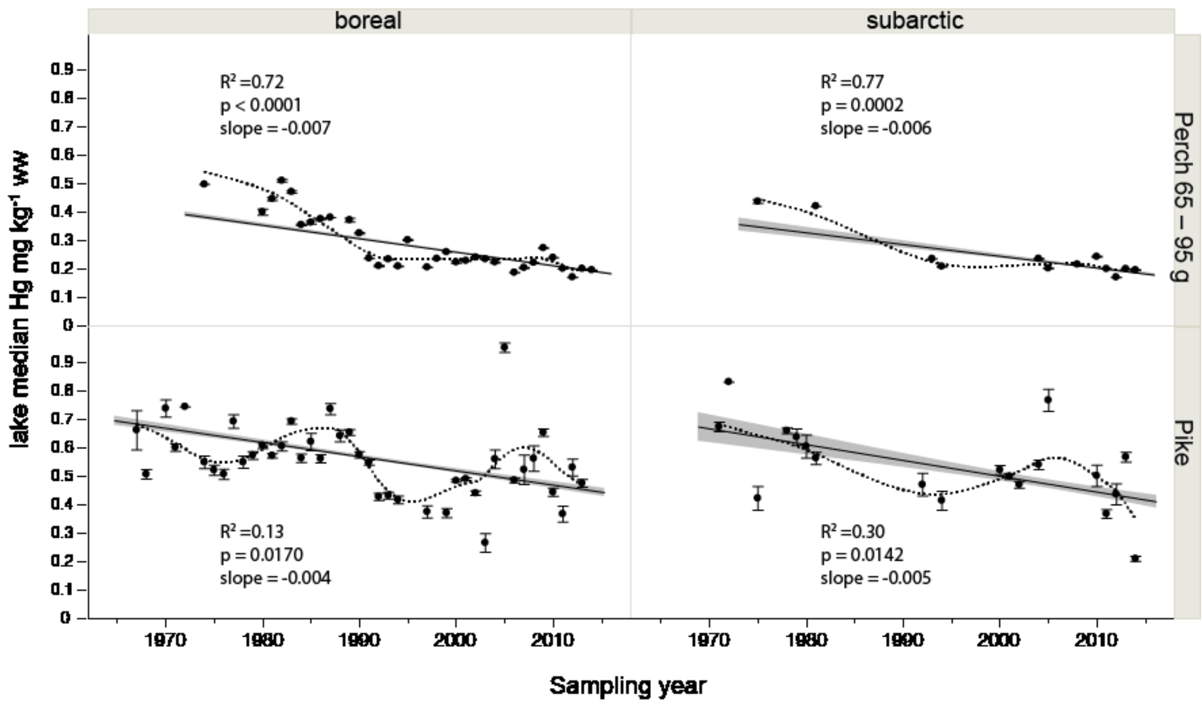

Figure 4 Temporal trends in lake Hg medians (wet weight, ww) of perch (65-95 g, top panels) and pike (bottom panels) between boreal (left panels) and subarctic regions (right panels) in Fennoscandia in lakes being subject to Hg loads from primarily long-range transported atmospheric pollution (LRTAP lakes). Trends are presented both as a linear regression (solid line) and a smoothed kernel curve (dotted line). 95 % confidence intervals around the linear regression lines are indicated in grey. Data is presented as annual mean and standard error for lake medians of fish Hg concentrations.