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

3

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25 **ABSTRACT**

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

41

42 **KEYWORDS**

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

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50 INTRODUCTION

51 In 1956, the occurrence of the Minamata Bay accident in Japan initiated intensive research and
52 monitoring of mercury (Hg) concentrations in fish used for human consumption. The accident was
53 caused by releases of the neurotoxic Methyl-Hg (MeHg), which was biomagnified in aquatic food webs
54 and has since proved to have harmful effects on aquatic organisms¹ and their consumers², including
55 humans^{3,4}. Although the toxic effects of Hg have been known for more than half a century⁵, our ability
56 to predict impacts of changed Hg emissions on exposure, accumulation, and biomagnification of Hg in
57 food webs remains limited because of the complex biogeochemical cycling of Hg. Thousands of
58 freshwater lakes worldwide have fish Hg concentrations exceeding limits advised for human
59 consumption (0.3 – 1.0 ppm Hg wet weight (w.w.))⁶. Freshwater fish are considered being critical
60 receptors of long-range transboundary air pollution of Hg⁷. The *Minamata Convention on Mercury*
61 (hereafter *Minamata Convention*) aims to protect human health and the environment from adverse
62 effects of Hg at a global scale⁸. The agreement requires the parties to evaluate its effectiveness, based
63 on information and reporting, including adequate methodologies to detect trends of Hg
64 concentrations in biota⁸.

65 In Fennoscandia, environmental monitoring of Hg was initiated in the mid-1960s, following
66 the awareness of use of Hg in paper and pulp mill factory processes (from the 1960s to the 1980s)^{9,10}.
67 Initially, monitoring was focused on lakes close to known point sources of Hg, but during the 1980s it
68 was revealed that lakes in remote and pristine areas were exposed to increased loads of
69 predominantly atmospherically deposited Hg^{11,12}. High levels of Hg in monitored fish initiated new
70 environmental legislations, including changes in the forest industry processes, and local emissions and
71 releases were generally reduced⁹. Still, several Northern areas show significant increases in fish Hg
72 concentrations the last decades, including Sweden¹³, Finland¹⁴, and Ontario (Canada)¹⁵, although this
73 rising trend is not found in all regions and for all fish species. In fact, a study of lakes in Sweden¹⁶ shows
74 declining Hg concentrations in fish between 2005 and 2015, something which fits with the observed
75 declining trend of Hg deposition since at least the 1990s throughout Europe¹⁷. However, most studies,

76 including the examples mentioned here, usually provide no or very limited information on local
77 pollution history (i.e. whether Hg catchment input is of local and/or long-range origin)¹⁸. Another
78 limitation in most of the studies available in current literature is that temporal fish Hg trends are
79 analysed within country or state borders rather than per bio- or ecoregions which are potentially more
80 meaningful regarding controls of biochemical Hg cycling such as climate and deposition^{17,19}.

81 In many boreal, subarctic, and Arctic lakes in Fennoscandia, long-range atmospheric transport
82 of Hg is the main source of Hg contamination¹¹ and has led to long-term accumulation of Hg in
83 catchments²⁰, similar to remote areas in North America²¹. Deposited Hg reaches surface waters either
84 gradually through enrichment of soils²¹ and subsequent leaching (transported by organic matter, OM)
85 to surface waters^{3, 22}, or as direct deposition to the lakes. The gradual release contrasts with point
86 source releases of Hg to the environment, and leaching of Hg from catchment soils is controlled by a
87 range of environmental drivers, characteristics, and processes which in their turn potentially affect
88 food web exposure to Hg and subsequent bioaccumulation (summarised by Driscoll et al., 2013³). In
89 order to document the effectiveness of global Hg emission reduction measures, established under the
90 Minamata Convention⁸ and the Convention on Long-Range Transboundary Air Pollution (CLRTAP)²³,
91 and to distinguish their effects from earlier legislation, it is useful to attribute key sources of Hg
92 pollution (i.e. long-range versus local) in different water bodies.

93 We examined a 50-year database of >50 000 measurements of Hg in freshwater fish across
94 wide climate, geography, and deposition gradients in Fennoscandia (Norway, Sweden, Finland, and
95 the Murmansk area in Russia). We evaluated temporal trends and spatial patterns of Hg
96 concentrations for fish species with different foraging and thermal guilds, and assessed temporal
97 trends related to predominant sources of Hg for the lakes, i.e. local point industrial sources (*point*
98 *source lakes*) and long-range atmospherically transported Hg (*LRTAP lakes*, referring to CLRTAP¹⁸).
99 Hypothesizing that fish Hg trends in LRTAP lakes, directly or indirectly, are sensitive to environmental
100 drivers, including climate (temperature)²⁴⁻²⁶, lake browning, and atmospheric deposition (especially
101 Hg and sulphur, S²⁷⁻²⁹), we also tested for temporal trends of Hg in LRTAP lakes in southern (boreal)

102 and northern (subarctic) ecoregions. The results are placed in a context of demands for suitable
103 monitoring programmes to evaluate policies aimed to reduce global Hg pollution.

104

105 **MATERIALS AND METHODS**

106 **Selection of data**

107 Records of total Hg measurements in freshwater fish muscle tissue from Sweden, Finland, Norway,
108 and the Murmansk Oblast (i.e. a federal subject) in Russia were collated from literature and existing
109 databases. Records that did not meet a set of criteria, including availability of Hg content, fish weight
110 and fish length, and a minimum of five records for a single fish species per lake, were excluded (11904
111 of initially 66464 individual records, see **Figure S1** in *Supporting Information*). Relations between Hg
112 concentrations and fish size, length and/or weight, and length-weight relationships were tested for
113 further quality assurance³⁰. Following these relations, residual outliers (i.e. entries outside 75%
114 quartile plus 1.5*interquartile range, n = 70) were excluded. The database was limited to fish species
115 that are typically distributed in all the four countries, resulting in records of Northern pike (*Esox lucius*,
116 42.4 %), perch (*Perca fluviatilis*, 34.1 %), Arctic charr (*Salvelinus alpinus*, 1.2 %), brown trout (*Salmo*
117 *trutta*, 3.1 %), and roach (*Rutilus rutilus*, 1.3 %). Finally, the database consisted of 54560 entries from
118 3132 lakes (**Figure S1**), collected between 1965 and 2015, spanning a south-north gradient from
119 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°
120 E on the Kola Peninsula (Murmansk, Russia, **Figure 1**).

121 The fish species differ in their thermal and foraging guilds^{31,32}. Arctic charr, brown trout, perch
122 and roach are generalist species that may forage across both pelagic and littoral habitats. The cold-
123 water adapted Arctic charr and brown trout are present in oligotrophic lakes; the cool-water species
124 perch is often the dominating species in mesotrophic lakes, and the warm-water species roach are
125 abundant in eutrophic lakes³³. Arctic charr, brown trout and perch undergo ontogenetic dietary shifts
126 from invertebrates to fish prey, but roach feed exclusively on invertebrate prey^{32, 34}. Pike is a cool-
127 water obligate piscivore that historically has been a key species, together with perch, for Hg

128 monitoring due to its wide distribution range, location at the top of food webs (i.e. combining both
129 littoral and pelagic energy sources due to its capacity to feed on all available prey fish species in
130 lakes³⁴), and its importance for recreational fishing.

131 Pike and perch were the most abundant species in the database, both spatially and temporally,
132 and they were selected for detailed analyses in this work. In the database, pike size (i.e. weight) centre
133 around 1 kg (mean \pm one standard deviation: 998 ± 579 g; median: 905 g), historically a target size for
134 many Fennoscandian Hg studies¹⁶. Because perch undergo an ontogenetic dietary shift from
135 invertebrates to fish³⁴, it is important to consider different size groups in the data analysis. In our
136 dataset, there was a significant decrease in perch size between those collected before year 2000 (140
137 ± 176 g) compared to those collected in year 2000 and later (61 ± 79 g, **Figure S2**). This shift in size for
138 collected fish is likely related to either sampling gear (i.e. a change in gill nets from traditional large
139 mesh gill nets to Nordic nets including small mesh sizes (<12 mm)), or sampling strategy (i.e. increased
140 focus on small, remote lakes with slow-growing perch). We have therefore chosen a selection of perch
141 sizes, including weights of 65-95 g (14-25 cm), to assess the potential trends in our data set. The size
142 selection of 65-95 g is based on *i*) the prevalence of these sizes throughout the whole database time-
143 period 1965-2015 (**Figure S2**); and *ii*) that the fish of these sizes have likely undergone an ontogenetic
144 shift to become piscivory³².

145

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

147 Lakes were classified per dominant Hg pollution source based on expert judgement: 1. *Lakes with no*
148 *local Hg pollution sources*, implying that atmospheric deposition of Hg is the dominating pollution
149 source (hereafter *LRTAP lakes*); 2. *Lakes with known local industry point source(s)* (hereafter *point*
150 *source lakes*); and 3. *Unknown*. We did not classify per timing of contamination. In the current work,
151 $n = 167$ lakes ($n = 13938$ specimens) were classified as being *point source lakes*, while $n = 474$ lakes (n
152 $= 14072$ specimens) were classified as being *LRTAP lakes* (**Figure S1** and **Figure 1**).

153

154 **Classification of lakes – boreal and subarctic ecoregions**

155 We divided the LRTAP lakes into subarctic (> 65° N) and boreal (< 65° N) (**Figure 1**), a simplified
156 classification following De Wit et al. (2016)³⁵. The regions contrast each other with respect to
157 atmospheric pollution (e.g. total Hg and S, primarily as oxidised S or SO₄)³⁶, temperature, and aqueous
158 OM concentrations. Deposition of Hg and S is lower in the subarctic region compared to the boreal,
159 and the subarctic lakes are colder and less coloured, i.e. lower OM concentrations. Deposition of SO₄
160 has been shown to promote methylation^{27, 37} and lately reduced acid deposition (primarily of SO₄) has
161 been shown to promote increased browning of surface waters²⁹. Temperature determines fish growth
162 with subsequent effects on Hg concentrations in muscle via dilution and condensation cycles^{33, 38}, but
163 temperature also controls terrestrial productivity and thus regional variation in aqueous OM³⁹. OM is
164 a transport vector for Hg^{22, 40}, but can also reduce photo-demethylation⁴¹ and bioaccumulation⁴².

165

166 **Data treatment**

167 Covariation between Hg concentration and fish size (length and weight^{43, 44}) and age⁴⁵ requires a
168 standardization to allow for investigation of spatial and temporal trends of Hg concentrations. We
169 used the individual fish weight and Hg concentration in combination with fish species information and
170 sampling year to find the modelled (i.e. expected) Hg concentration for fish at a standard weight.
171 Different linear regression models were applied to describe the log[Hg] concentrations
172 (Supplementary information, **Table S1**), where potential explanatory variables included fish weight,
173 fish species, sampling year, and the interaction terms year x species and weight x species, to evaluate
174 changes in fish Hg concentrations with weight and species over time.

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

180
$$\log \text{ALM} = \alpha + \beta * \text{year} + \gamma * Z + \delta * \text{year} * Z + \varepsilon \quad (1)$$

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

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

193

194 **RESULTS AND DISCUSSION**

195 **Fennoscandic fish Hg concentrations (observed data)**

196 Consumption of fish is considered the main Hg exposure route to humans and wildlife^{46, 47} and
197 measures taken under the CLRTAP²³, the Minamata Convention⁸, and the EU Water Framework
198 Directive (WFD) are therefore targeted to improve the quality of aquatic ecosystems with respect to
199 Hg. Fish Hg concentrations in lakes across Fennoscandia generally have concentrations that exceed
200 maximum limits set to protect human health (0.3-0.5 ppm w.w., **Table S2**)^{6, 48}. In the Fennoscandian
201 fish database, pike had the highest mean ALM concentration (0.67 ppm), with Arctic charr (0.37 ppm),
202 brown trout (0.22 ppm), perch (0.29 ppm), and roach (0.37 ppm) having lower concentrations. Pike is
203 a fish representing high trophic levels in Fennoscandic freshwater food webs and an obligatory
204 piscivore feeding on all types of prey fish, hence elevated Hg levels are expected^{13, 16, 49}. The levels

205 from the current work are similar to the median Hg concentrations observed in pike data from Munthe
206 et al. (2007)⁵⁰ and Åkerblom et al. (2014)¹⁶: 0.69 (1965-2004) and 0.68 ppm (1966-2012), respectively.

207 The large majority of fish caught in Fennoscandia over the last six decades shows observed Hg
208 concentrations above the WFD Environmental Quality Standard (EQS) of 0.02 ppm⁵¹. Of the 54560 fish
209 samples included in the entire database, 99.8% had concentrations above 0.02 ppm, and good
210 chemical condition is not met for any water body. Hg is a priority substance under the WFD, where
211 protection from biomagnification in the food chain (i.e. top predators including fish and wildlife) is a
212 main aim (i.e. “secondary poisoning”). For Hg, the EQS is based on a 365 days *No Observed Effect*
213 *Concentration* (NOEC) for MeHg, and a (relatively low) assessment factor of 10 is applied due to the
214 large number of NOECs available for MeHg⁵¹. Although the WFD EQS *secondary poisoning* for Hg in
215 biota has relevance for assessing the risks of ecosystem Hg exposure in Fennoscandia, it does not
216 differentiate between lakes with higher and lower Hg risks. A different threshold for Hg in
217 Fennoscandian fish is the limit to protect *human health* of 0.5 ppm⁵¹, where 74% of the water bodies
218 in our database would *not* meet this criterion. However, if only samples collected after year 2000 are
219 considered, the relative number of lakes with an individual fish Hg concentration above 0.5 ppm is
220 25%, testifying to improved environmental status in Fennoscandia.

221

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

223 Abatement measures introduced to reduce emissions and releases from industry, including closure or
224 removal of Hg releasing facilities, may have been very effective, but previous pollution has left legacy
225 Hg in soils or lake sediments⁹. Thus, lakes with historical local Hg sources are likely to add an additional
226 concentration signal compared to lakes only influenced by long-range atmospherically transported Hg.
227 Lower fish Hg concentrations in LRTAP lakes (LRTAP lakes: 0.28 ± 0.16 ppm, $n = 474$ lakes, mean \pm one
228 standard deviation of ALMs, all five species) compared to point source lakes (0.46 ± 0.22 ppm, $n = 167$
229 lakes, ANOVA: F-ratio=116, $p < 0.0001$, $r^2 = 0.15$) support this hypothesis. The same pattern is evident
230 on individual fish species level for the two main fish species in the database (**Table S2**). Despite a large

231 body of evidence suggesting that between-lake variation in fish Hg levels is controlled by catchment
232 and foodweb characteristics (including fish species composition), in addition to climate⁵², our database
233 indicates that pollution sources matter, i.e. that atmospheric pollution has resulted in much lower Hg
234 loading to lakes than point sources, and therefore lower Hg in fish. As an illustration, a small lake (0.5
235 km²) catchment (5 km²) without a local pollution source, with a yearly atmospheric (10 µg Hg m⁻² y⁻¹)
236 and catchment (2.5 µg Hg m⁻² y⁻¹) input^{20,53} of Hg from long-range atmospheric pollution receives total
237 annual inputs of 17.5 g Hg. To put this into perspective, examples on abatement measures in
238 Fennoscandia include a chlor-alkali plant that released from three to five tons of Hg annually to Lake
239 Vänern, Sweden, before new legislations were introduced in the 1970s and 1980s¹⁰, and a sulphide
240 ore smelter emitting 3.5 tons of Hg annually to air in Northern Sweden in the late 1960s⁵⁴.

241 For the point source lakes, the temporal trends in ALMs showed a significant long-term
242 decreasing trend between 1965 and 2015 (perch: annual decrease (ad)=-8‰ year⁻¹, $p<0.001$, pike:
243 ad=-4‰ year⁻¹, $p<0.0001$). However, since 1995, the temporal trends are not significant (perch: ad=-
244 1‰ year⁻¹, $p=0.73$, pike: ad=-4‰ year⁻¹, $p=0.36$), indicating that most of the change in concentrations
245 happened earlier (**Figure 2**). In Fennoscandia, chlor-alkali industry can be recorded back to at least the
246 1920s⁵⁵, and a peak in industry emissions and releases are assumed to have occurred during the 1950s
247 and 60s, when 20 to 30 tons of Hg were discharged annually from point sources in Sweden⁵⁶. Since
248 the 1980s local emissions and releases in Fennoscandia were reduced significantly⁵⁷. In Norway, the
249 official governmental total emissions to the atmosphere and releases to soil and water have declined
250 from 5.0 tons in 1985 to 2.5 tons in 1995 and 0.9 tons in 2005⁵⁸. These declines fit well with the
251 temporal fish Hg data from the point source lakes, where there is a significant difference between
252 samples collected in 1990 or earlier and those collected in 1995 or later for both perch (65-95 g, 0.47
253 ± 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
254 and 0.55 ± 0.14 ppm, ANOVA: F-ratio=188, $p<0.0001$, $r^2=0.22$) (**Figure 2**). The reasons for the decline
255 in discharge and emissions in Scandinavia are, in addition to regional and national control legislation,
256 improved technology, and reduction of polluting industrial production⁵⁶.

257 For the LRTAP lakes, the temporal decrease in ALMs were significantly larger for perch
258 compared to pike (perch: $ad=-7\% \text{ year}^{-1}$, $p<0.0001$, pike: $ad=-4\% \text{ year}^{-1}$, $p=0.0032$) (**Table S3A**). This
259 difference in trends between the fish species could indicate that perch and pike respond differently
260 to changes in factors that relates to Hg biomagnification, potentially as a consequence of biological
261 and ecological differences between species pike⁵⁹⁻⁶¹ and perch^{62, 63}. To examine such differences
262 between fish species, and to disentangle the cause for the different magnitude of decreases in fish Hg
263 concentrations over time, data on age⁴⁵ and trophic level indicators (i.e. stable isotopes of nitrogen,
264 N⁶⁴) would be necessary^{32, 33}.

265 No other studies of temporal Hg trends exist, covering such a large geographical area with
266 understanding of sources of Hg contamination. Our trends are only partially supporting findings from
267 large North American fish databases. Similar to this study, Eagles-Smith et al. (2016)⁶⁵ show that fish
268 Hg concentration trends are declining from 1969 to 1977 in a study from the Western US and Canada
269 (n = 96310 specimens, n = 4262 locations), but show no trend from 1978-2012. In two studies from
270 Ontario, Canada, Gandhi et al. (2014)¹⁵ reveal declining or unchanging fish Hg concentrations between
271 the 1970s and 2012 (n = 31743 specimens, n = 1167 locations), depending on the fish species
272 considered, and Tang et al. (2013)⁶⁶ found no significant decline between the time periods 1974-1981
273 and 2005-2010 (n = 5215 specimens, n = 73 locations). For a more recent time period, Zhou et al.
274 (2017)⁶⁷ demonstrate declining fish Hg concentrations between 2004 and 2015 for specimens of lake
275 trout (*Salvelinus namaycush*) from the Laurentian Great Lakes (n specimens unknown, n = 8 locations).

276 The Gandhi et al. (2014)¹⁵ study was considering time trends for different predatory fish
277 species (pike, lake trout, walleye, *Sander vitreus*) between 1970 and 2012. It was shown that while
278 fish Hg concentrations from 1970 to 1990 were generally declining, concentrations in recent decades
279 (time periods 1985-2005 and 1995-2012) were increasing, especially for pike and walleye. For
280 comparison, our data shows that there is no significant trend for pike ($ad=-4\% \text{ year}^{-1}$ in LRTAP lakes;
281 $ad=-4\% \text{ year}^{-1}$ in point source lakes) or perch ($ad=-2\% \text{ year}^{-1}$ in LRTAP lakes; $ad=-1\% \text{ year}^{-1}$ in point
282 source lakes) in either LRTAP or point source lakes between 1995 and 2015. Gandhi et al. (2014)¹⁵ also

283 demonstrate overall (1970-2012) neutral or declining trends (depending on the fish species
284 considered). A similar study as the one by Gandhi et al. (2014)¹⁵ was done by Åkerblom et al. (2014)¹⁶,
285 documenting an overall long-term decline from 1965 to 2012 in Swedish pike (n = 44927).

286

287 **Spatial patterns of fish Hg in boreal and subarctic Fennoscandia**

288 For the LRTAP lakes (all species combined), fish Hg concentrations (mean \pm one standard deviation of
289 ALMs) showed a pattern where the boreal region (0.32 ± 0.18 ppm) had significantly ($p=0.017$) higher
290 concentrations than the subarctic region (0.29 ± 0.16 ppm). As indicated in **Table S2**, the inter-regional
291 variation is not the same for all the fish species, and we observe that the difference between the
292 regions is larger for pike (0.56 ± 0.15 ppm versus 0.48 ± 0.16 ppm, ANOVA $p < 0.001$) than for perch (65-
293 95 g, 0.23 ± 0.07 ppm versus 0.21 ± 0.05 ppm, $p=0.027$). The difference is surprisingly small between the
294 ecoregions, as higher concentrations in the boreal region compared to the subarctic region was to be
295 expected, given that elevated levels of Hg in fish often are associated with humic lakes^{42, 68, 69}. In
296 Fennoscandia there is a strong increasing west-to-east and north-to-south aqueous OM concentration
297 gradient⁷⁰, likely to influence the fish Hg concentrations. OM can have both indirect and direct effects
298 on Hg accumulation in aquatic food webs. Higher concentrations of OM, particularly higher molecular
299 weight terrestrially derived OM, may reduce bioavailability of MeHg for uptake at the base of the food
300 web⁷¹. However, in contrast, increased OM could also act as a substrate for increased in-situ MeHg
301 production, with more labile algal-derived OM supporting higher methylation⁷².

302 Relationships between observed fish Hg concentrations and aqueous OM often leaves a
303 considerable amount of variation unexplained^{12,42}, and disguises other complex processes influenced
304 by climate, catchment characteristics and biology/ecology³³. An example is deposition of Hg, which in
305 Fennoscandia follows a pattern of decreasing levels from south to north¹⁷, suggesting that fish Hg
306 concentrations in LRTAP lakes should be expected to decline with increasing latitude⁷³. This hypothesis
307 is only partly confirmed by our fish data, where concentration trends are decreasing with increasing
308 latitude for pike ($r=-0.27$, $p=0.0005$), but where the perch data decline is not significant ($r=-0.11$,

309 $p=0.078$) (**Figure 3**). However, subarctic lakes typically have higher Hg biomagnification rates than
310 lakes located further south⁷⁴, related to combined temperature effects on growth dilution and
311 starvation^{38, 75}, trophic transfer efficiency and excretion rates⁷⁶. Hence, the limited declines in fish Hg
312 concentrations with increasing latitude observed for the LRTAP lakes, suggests that climate related
313 effects potentially counteract Hg deposition and Hg effects from aqueous OM (i.e. increased foodweb
314 exposure). In subarctic lakes, seasonality is much stronger than in boreal lakes located further south,
315 likely strengthening growth dilution and starvation cycles in fish^{38, 75}. In fish, the lower temperature of
316 the subarctic region will directly reduce growth, metabolic activity, and excretion of Hg in these
317 lakes⁷⁶.

318

319 **Temporal fish Hg trends in Fennoscandia**

320 Differences in temporal trends between ecoregions (i.e. boreal and subarctic) could potentially
321 document to what extent fish Hg concentrations respond to changes in Hg biomagnification in LRTAP
322 lakes. Given the strong relationships between cycling of Hg and aqueous OM⁴⁰, a naturally emerging
323 hypothesis is that observed browning of many North American and northern European lakes²⁹ could
324 influence fish Hg concentrations⁴². For both perch and pike, our data from LRTAP lakes demonstrate
325 significantly declining trends of Hg in both boreal and subarctic regions (**Figure 4, Table S4**). For perch,
326 the annual decreases were -7‰ per year and -6‰ per year for the boreal and subarctic regions, while
327 for pike the decreases were -3‰ per year and -5‰ per year. The inter-regional and inter-species
328 differences in trends were not significant (**Table S3**). From a comparison of the long-term linear trend
329 curve and the smoothed kernel curve it is obvious that the annual decrease in fish Hg levels does not
330 represent the inter-annual and inter-decadal trends and changes in fish Hg levels (**Figure 4**). Studies
331 investigating lake-specific increases in fish Hg during the period 1995-2005 suggests that temporal
332 trends reflect processes in accumulation of Hg that is controlled by environmental drivers such as OM
333 in lakes¹³.

334 A recent study of Scandinavian lakes suggests that the largest lake browning trends between
335 1990 and 2013 were found in regions with strong reductions in S deposition. Hence, the change in OC
336 concentrations was largest furthest south in the boreal wet (+1.7 % per year) and dry (+1.5 % per year)
337 regions, and lower in the subarctic (+0.8 % per year) region³⁵. A larger input of OC to lakes could
338 influence Hg cycling in several ways, including increased loading of aqueous Hg²², decreased MeHg
339 degradation⁷⁷ and production⁷⁸, and increased/decreased fish bioaccumulation factors⁴², all
340 potentially affecting fish Hg concentrations. In our study, we found no evidence of significantly
341 increasing concentrations for either perch or pike in any of the ecoregions for the same time-period
342 (1990-2013) studied by de Wit et al. (2016) (**Figure 4**). In sum, it is challenging to document and
343 quantify the potential influence from climatic differences and changing OC concentrations on fish Hg
344 trends, likely because of biological and ecological factors also playing an important role.

345

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

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

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

371

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378

379 **SUPPORTING INFORMATION**

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

384

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FIGURES

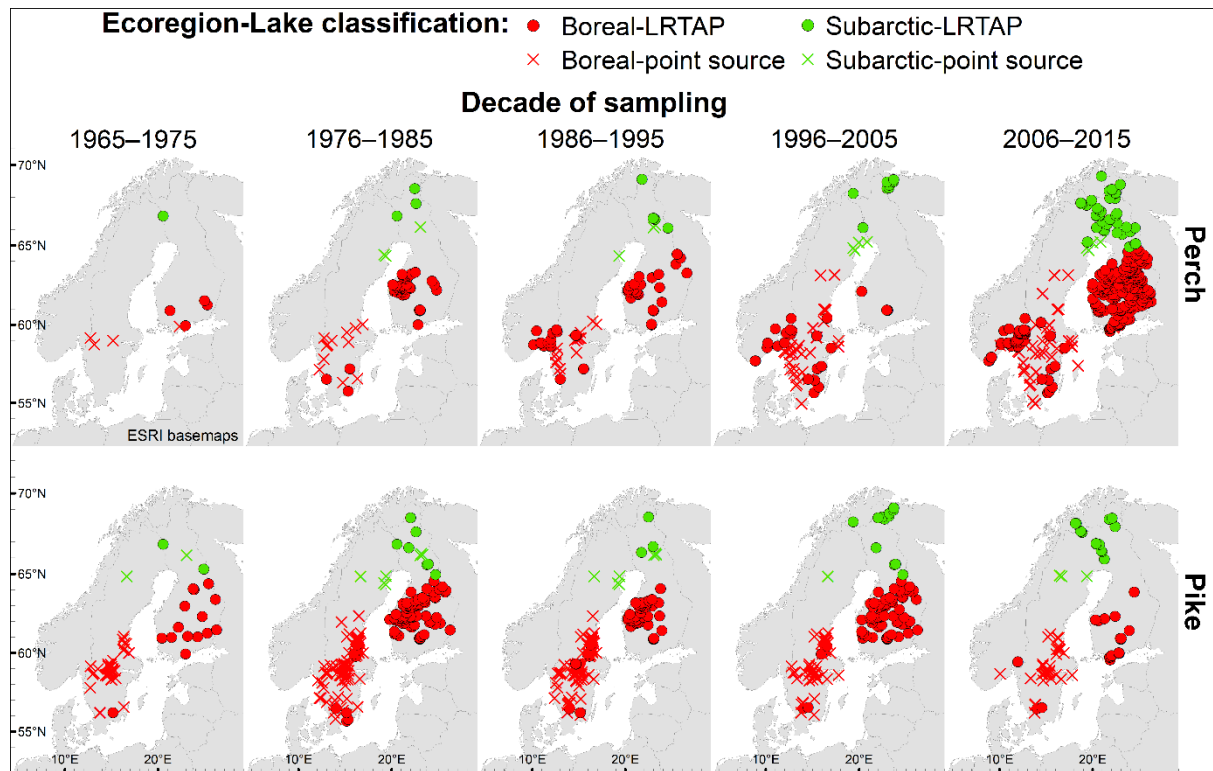


Figure 1 The geographical distribution of the LRTAP lakes (circles, n=474) and the point source lakes (crosses, n=167) and what decade they were sampled (from left to right: 1965-75; 1976-85; 1996-2005; 2006-15). Top and bottom panels show the lakes where perch and pike were represented, and the colours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

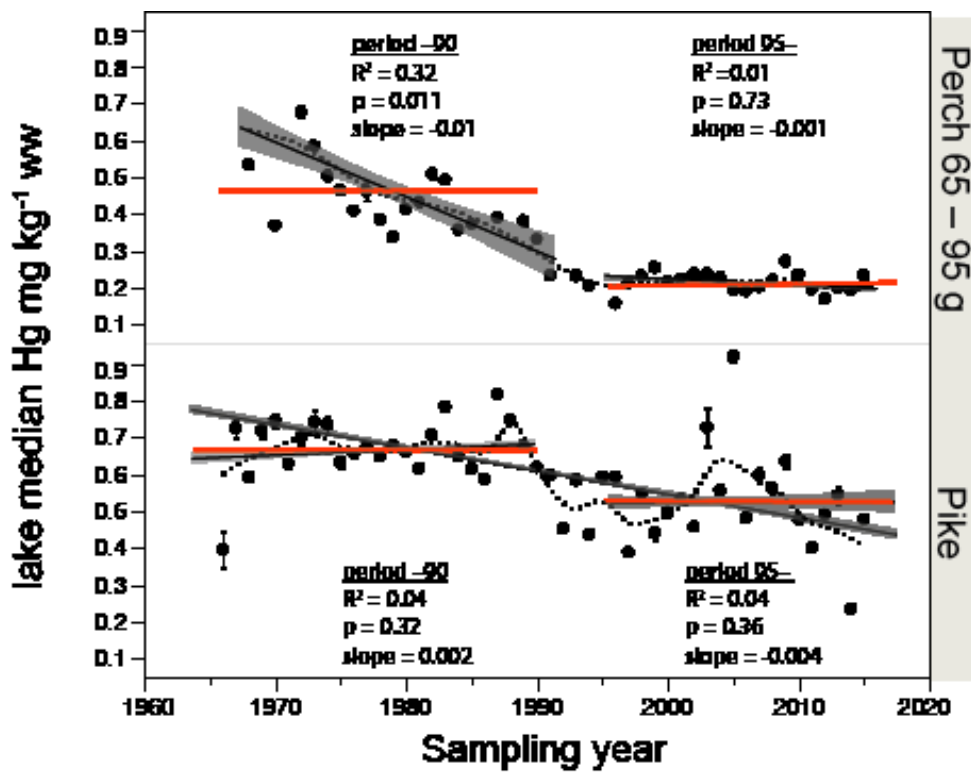


Figure 2 Temporal trends in annual lake medians (ALMs \pm standard error) of Hg concentrations (wet weight, ww) of perch (65-95 g) and pike from point source lakes. The overall trends (1965-2015) are presented both as a linear regression (solid black line) and a smoothed kernel curve (dotted black line). 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.

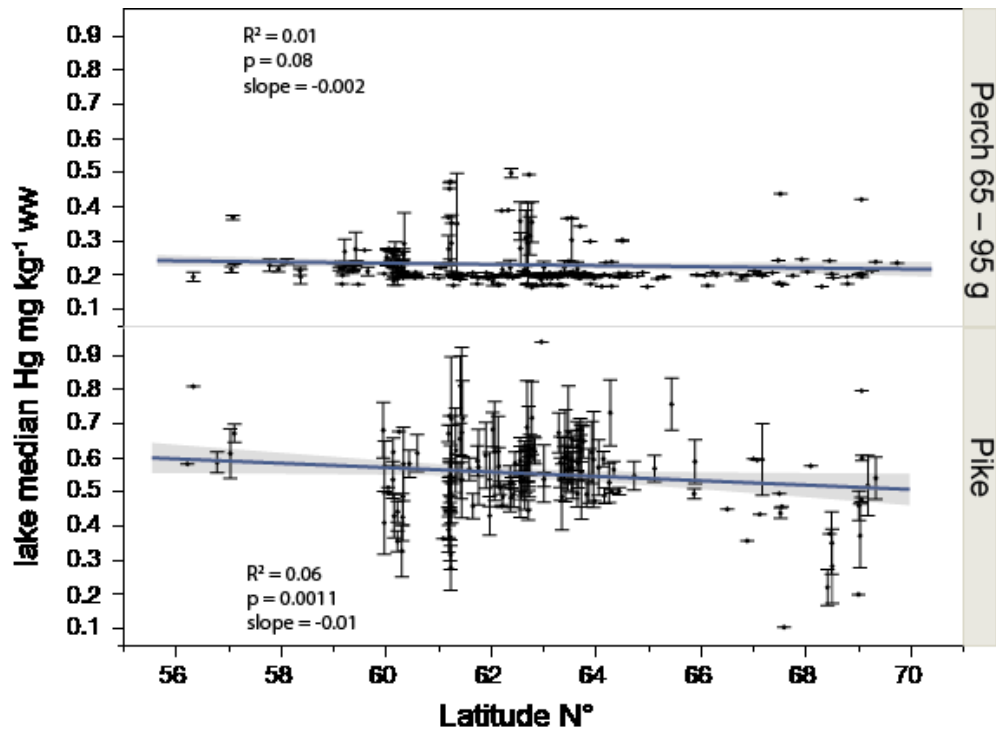


Figure 3 Latitudinal gradient in Hg concentrations (wet weight, ww) of perch (65 – 95 g, top panel) and pike (bottom panel) across Fennoscandian lakes subject to Hg loads from primarily long-range transported atmospheric pollution (LRTAP lakes). Each circle represents the mean annual lake median (ALM) 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.

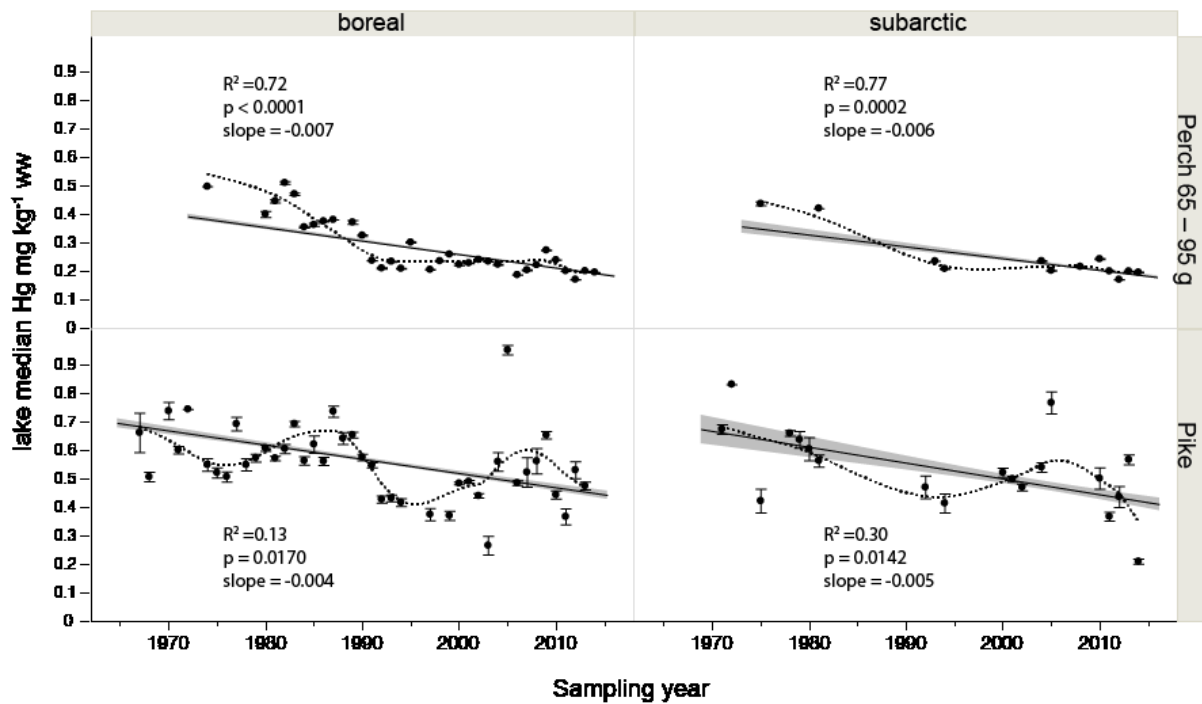


Figure 4 Temporal trends in lake Hg medians (wet weight, ww) of perch (65-95 g, top panels) and pike (bottom panels) between boreal (left panels) and subarctic regions (right panels) in Fennoscandia in lakes being subject to Hg loads from primarily long-range transported atmospheric pollution (LRTAP 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.