



Pike Mercury Concentration in Small Boreal Headwater Lakes During Four Decades of Regional and Local Changes

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Abstract We measured the total mercury (THg) concentration in the muscle of northern pike (*Esox lucius* L.) from 12 boreal headwater lakes once per decade during the 1980s to 2010s. Relations of pike THg to regional environmental changes, such as reduction of acid deposition and increase in water colour (i.e. browning), were examined as well as the effects of local lake-specific changes caused by introduced beavers (*Castor canadensis*) and intensive fishing of pike. No consistent decrease in pike THg took place during 1983–2013 despite the reductions in mercury emissions from European sources since the 1990s and the associated decreased mercury

deposition in Finland. In most of the lakes, individual pike exceeded the recommended limit for human consumption (1 mg kg⁻¹, wet weight). The highest THg levels were recorded in 2002 following intensive browning of the lakes in the 1990s in connection with decreased sulphate deposition. The effects of acid deposition were only seen as the highest pike THg in the two most acidic lakes in the early 1980s. After taking length effects into account, pike THg had a negative relationship with growth (i.e. growth dilution), but a positive relationship with water colour, iron, nutrients and maximum lake depth. Females grew faster and had lower THg than males. Pike THg was elevated in beaver-impacted lakes. Experimental removal of pike from three study lakes during 2005–2013 resulted in a decrease of pike THg. These findings emphasize the significance of local abiotic and

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biotic drivers when interpreting the regional long-term trends in pike THg.

Keywords THg · Pike growth · Browning of lakes · Beaver · Intensive fishing

1 Introduction

Mercury (Hg) is a toxic metal occurring naturally in the environment. Since the beginning of the industrial revolution, mercury has been widely released into the environment due to human activities like mining, industrial point sources, agriculture and atmospheric deposition from burning of fossil fuels, especially coal (e.g. Streets et al., 2017). Mercury occurs in various forms in nature: elemental mercury (Hg⁰), inorganic mercury salts (HgI and HgII) and organic mercury compounds, especially toxic methylmercury (MeHg). The symptoms of acute mercury poisoning are connected to disturbances in the central nervous system. Because of the risk to human health, upper limits for the mercury content of edible fish have been set widely (EU, 2006). More recently, the Minamata Convention on Mercury entered into force in August 2017 (Wang et al., 2019). This treaty obligates the signed countries to control Hg emissions and the release of Hg to ambient air, water bodies and soil. It addresses several aspects necessary to reduce Hg use globally and to protect human health and the environment from the adverse effects of mercury.

Fish accounts for most of the mercury intake by humans from daily food, and over 95 % of the Hg contained in adult predatory fish muscle is MeHg (Bloom, 1992; but see Lescord et al., 2018) that bioaccumulates in organisms and biomagnifies in aquatic food web (e.g. Lavoie et al., 2013). High fish mercury concentrations, exceeding the recommended limits for human consumption (1 mg kg⁻¹), were measured in Nordic lakes during the 1960s and 1970s (Häsänen and Sjöblom, 1968; Lindeström, 2001). Since then, a general decreasing trend, although fluctuating, has been reported (Åkerblom et al., 2014; Braaten et al., 2019). This is due to the restrictions of use of Hg in industrial processes (Lodénus, 1991; Lindeström, 2001) and due to the decrease of mercury emissions from European sources since the early 1990s (Travnikov et al., 2012) followed by decreased

mercury deposition (Wängberg et al., 2010; Ruoho-Airola et al., 2014).

In headwater lakes, where mercury mainly originates from atmospheric deposition to the catchment and directly to the lake surface, high and even increasing fish Hg levels were reported in recent studies (Wyn et al., 2010; Braaten et al., 2014; Rask et al., 2021). This is connected to long-term environmental changes, such as recovery of lakes from acidification after reduced sulphate deposition (Keller et al., 2007; Rask et al., 2021), subsequent browning of lake waters due to increased load of dissolved organic carbon (DOC) compounds (Vuorenmaa et al., 2006; Monteith et al., 2007; Arvola et al., 2010; Blanchet et al., 2022) and different effects of climate change (Gandhi et al., 2014; Ahonen et al., 2018; Eagles-Smith et al., 2018; de Wit et al., 2021). As a consequence of these changes, legacy mercury load from catchments to lakes has increased due to complexation reactions between DOC and Hg compounds. This has often resulted in increased mercury concentration of fish (Hongve et al., 2012; Braaten et al., 2018; Moslemi-Aqdam et al., 2023).

Besides the large-scale atmospheric deposition and climatic effects, DOC and Hg dynamics can be affected locally via beaver activities and by forestry (e.g. Blanchet et al., 2022). The effects of beavers are typically strongest during the first 3 years of flooding (Vehkaoja et al., 2015; Nummi et al., 2018). Increasing DOC flux from riparian areas brings bound mercury and subsequently lowers growth rate and elevate mercury levels in food webs especially in top predatory fish, such as northern pike *Esox lucius* L. (Moslemi-Aqdam et al., 2021, Moslemi-Aqdam et al., 2022). Forest harvesting as an important form of land use in countries of boreal region can increase the DOC and mercury load to lakes and bioaccumulation of mercury to fish (Rask et al., 1994; Garcia & Carignan, 2000; Porvari et al., 2003; Wu et al., 2018; Ahonen et al., 2018; Eklöf et al., 2018).

Pike is one of the most common fish species in the boreal lakes of the Nordic countries (Tammi et al., 2003). As a top predator of food chains in boreal lakes, pike often show the highest fish mercury concentrations (Häsänen and Sjöblom, 1968; Göthberg, 1983; Heiskary and Helwig, 1986; Verta, 1990; Porvari, 1998; Lindeström, 2001; Kozak et al., 2021). Available prey resources will directly affect the growth rate of predatory fish, which is generally

inversely correlated with the total mercury (THg) content, and is often referred to as mercury growth dilution, as described for many different species (Keva et al., 2017; Rask et al., 2021; Moslemi-Aqdam et al., 2022). Fish growth is dependent on both environmental and biotic factors, where intensive fishing of the fish community including predatory fish has often led to reduction of the mercury content of top predator fish such as pike (Göthberg, 1983; Verta, 1990; Sharma et al., 2008).

Considering the Hg dynamics of the entire food web of a lake ecosystem, the bioaccumulation of MeHg in organisms at the base of the food chain is an important controller of Hg concentrations in taxa at higher trophic levels (Yu et al., 2011; Painter et al., 2015; Kahilainen et al., 2017; Kozak et al., 2021). Habitat- and species-specific differences in Hg bio-availability are affecting the fish Hg levels, and the dominance of pelagic energy sources is often related to a higher mercury content in fish (Karimi et al., 2016; Kahilainen et al., 2016; Arcagni et al., 2018; Moslemi-Aqdam et al., 2022). Microbial communities in stratified lakes with anoxic hypolimnion are also important (Verta et al., 2010; Rask et al., 2010; Karhunen et al., 2013) because sulphate-reducing bacteria under anoxic conditions are mainly responsible for the methylation of inorganic mercury to MeHg in freshwater ecosystems (Ullrich et al., 2001; Eckley et al., 2005; Bravo et al., 2017).

We measured pike THg content in 12 small headwater lakes in the Evo forest area, southern Finland, during 1983–2013. During these four decades, sulphate and Hg deposition decreased (Vuorenmaa, 2004; Wängberg et al., 2010; Ruoho-Airola et al., 2014), and browning of lakes took place, especially in the 1990s (Vuorenmaa et al., 2006; Arvola et al., 2010; Arzel et al., 2020). Questions related to the potential effects of these long-term environmental changes include the following: (1) did the pike mercury decrease due to lower Hg deposition (Ruoho-Airola et al., 2014) or due to reduced methylation activity of sulphate-reducing bacteria after decreased sulphate deposition (Hrabik & Watras, 2002; Braaten et al., 2020)? Or (2) did pike mercury concentrations increase in the course of lake browning due to increased dissolved organic matter load from catchments to the lakes accompanied by increased load of historical mercury (Hongve et al., 2012; Braaten et al., 2018; Moslemi-Aqdam et al., 2022)?

In addition, some of our study lakes have been subject to two important local changes that could affect the overall Hg dynamics of lakes. First, beavers have occupied several lakes over the years (Vehkaoja et al., 2015) with a subsequent increase in DOC fluxes and availability of mercury in the ecosystem (Painter et al., 2015; Ecke et al., 2017). We here address the question, (3) did beaver floods increase pike mercury in these lakes? Second, in a lake-scale experimental research on sustainable fishery management (Lehtonen et al., 2017), some lakes were subject to effective fishing and here the question is, (4) did intensive pike removal decrease pike mercury due to growth dilution (cf. Lavigne et al., 2010; Moslemi-Aqdam et al., 2022)?

2 Material and Methods

2.1 Study Lakes

We sampled pike from 12 natural headwater lakes for THg analyses at circa 10-year intervals between 1983 and 2013. The lakes are located in the Evo forest area, southern Finland (Fig. 1), which is part of the Lammi Long-Term Ecological Research area (LTER, Rask et al., 2014a). The catchment areas of the lakes are 0.04–62 km² and mainly covered by coniferous boreal forest on peatland, sandy or rocky landscape. The lakes are small with surface area between 1 and 45 ha, maximum depth 6–14 m, and altitude 126–156 m above sea level (Table 1). All 12 lakes are inhabited by pike and perch (*Perca fluviatilis* L.) and 10 of them also by roach (*Rutilus rutilus* L.). Other species like ruffe (*Gymnocephalus cernuus* L.), bream (*Abramis brama* L.) and bleak (*Alburnus alburnus* L.) were present to a lesser extent (Olin et al., 2017a). A diet study on three of the lakes indicated that perch and roach clearly dominated the diet of pike (Tiainen, 2017). Water quality data for 1983 were from Arvola et al. (1990) and Rask and Metsälä (1991), and for the years 1993, 2002 and 2013 from the databases of the Lammi Biological Station, University of Helsinki (for details, see Arvola et al., 2010). Briefly, the lakes were acidic with a pH range 4.9–6.7 and alkalinity from 0 to 0.25 mmol L⁻¹. Water colour ranged from very clear to highly humic (7–495 mg Pt L⁻¹), and the nutrient levels of lakes varied from oligotrophic to eutrophic (tot P 2–73 µg L⁻¹ and tot N 218–1107 µg

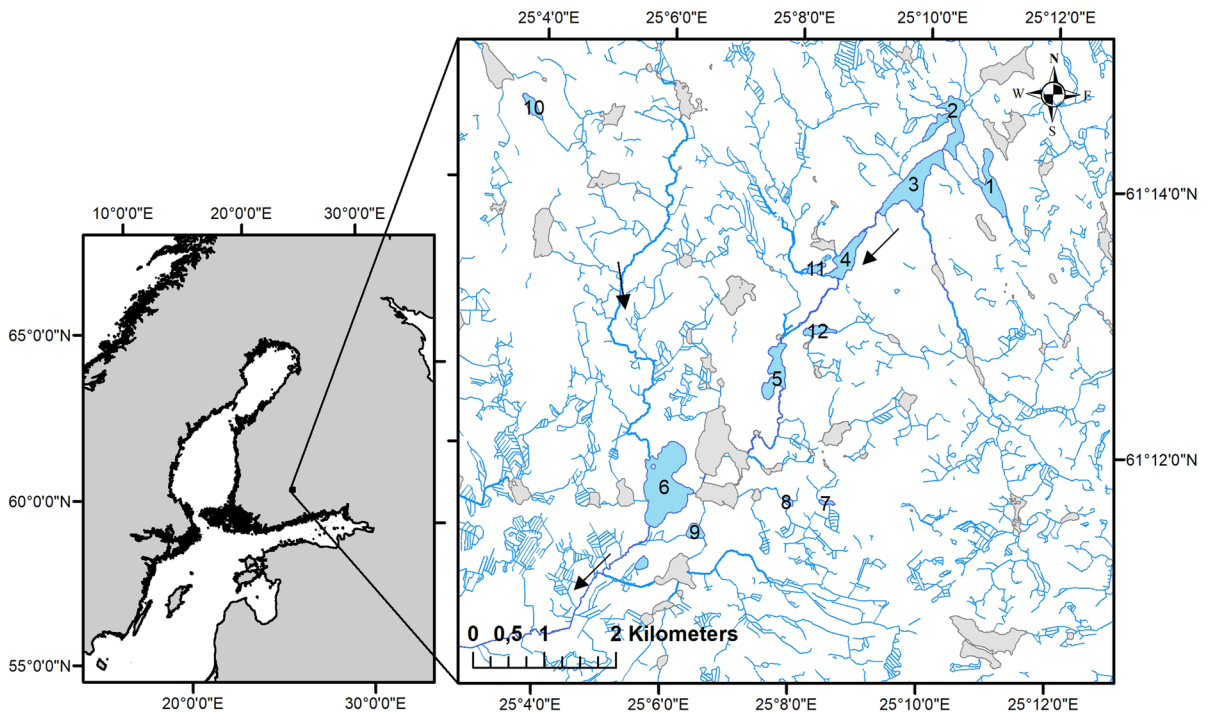


Fig. 1 The location of the 12 study lakes in southern Finland and in the Evo area. The lake numbers are as given in Table 1; chain lakes 1–6 and headwater lakes 7–12

L^{-1}). Ca ranged between 0.7 and 6.6 $mg L^{-1}$ and iron from 0.1 to 1.6 $mg L^{-1}$ (Table 1).

Half of the lakes (numbers 1–6 in Table 1) belong to the main lake chain of the Evo area and are subsequently referred to as “chain lakes”, whereas the rest (lakes 7–12) are seepage lakes or uppermost drainage lakes with an outlet, but no inlet, and designated as “headwater lakes”. The chain lakes are larger (12–45 ha) than the headwater lakes (0.9–4.2 ha) and are therefore less vulnerable to lake-specific changes due to direct human (forestry, fishing) or beaver (flooding) activities in their catchments. Most of the lakes are mainly fed by surface runoff, but two of them (L. Syrjäälunnen and L. Iso Valkjärvi) are groundwater lakes with clear water. The headwater lakes L. Iso Valkjärvi and L. Valkea-Kotinen acidified to pH levels of <5 during the 1980s (Rask & Metsälä, 1991) but have recovered from the acidification since the early 1990s.

Introduced beavers (*Castor canadensis*) are common in the lakes of the study area (Vehkaoja et al., 2015; Kivinen et al., 2020) and have temporarily caused floods on the shores of certain lakes (L.

Haarajärvi, L. Haukijärvi, L. Majajärvi, L. Huhmari), potentially affecting the Hg dynamics in the lake-catchment systems. During 2005–2013, three lakes in the present study (L. Haarajärvi, L. Haukijärvi, L. Majajärvi) were included in the project “Ecologically sustainable fishing” (Lehtonen et al., 2017). This project offered an opportunity to assess the effect of intensive pike fishing (Tiainen et al., 2017) on mercury concentration.

2.2 Sampling of Fish

Fish for mercury analyses were caught in May–September 1983/1984, 1993, 2002 and 2013 with wire traps, fyke nets and hook and line (5–21 pike per lake and per year, 26 to 60 per lake in total and 490 in the whole study, Table 2). Samples from the years 1983 ($n = 108$) and 1984 ($n = 45$) were pooled together to represent the pike THg level of the early 1980s. For 2002, missing samples from L. Valkea-Kotinen were replaced with those taken in 1999. No samples from L. Huhmari were available in 2013.

Table 1 Lake characteristics and water quality given as range of autumn turnover surface water sampling in the study years (LA lake area, CA catchment area, Alt altitude, Depth maximum depth, Alk alkalinity)

Lake	Lake number	LA (ha)	CA (ha)	Alt (masl)	Depth (m)	pH	Alk ($\mu\text{eq L}^{-1}$)	Colour (mg Pt L^{-1})	Ntot ($\mu\text{g L}^{-1}$)	Ptot ($\mu\text{g L}^{-1}$)	Ca (mg L^{-1})	Fe (mg L^{-1})
Haarejärvi ^{a,b}	1	15	155	141	14	6.1–6.5	0.07–0.14	100–193	402–553	9.0–18.0	3.0–3.7	0.3–1.1
Sorsajärvi	2	19.2	1080	134	13	5.9–6.3	0.07–0.13	138–367	421–664	12–19	3.5–3.8	0.3–1.3
Savjärvi	3	27.3	2190	134	12	5.9–6.4	0.08–0.12	165–370	457–696	21–32	3.5–4.0	0.3–1.2
Rahtijärvi	4	12.9	2922	131	13	6.2–6.4	0.09–0.15	160–331	562–685	22–52	3.6–4.4	0.4–1.4
Pitkäniemen-järvi	5	13.9	3340	127	10	6.4–6.5	0.12–0.15	111–296	329–619	17–22	3.5–4.4	0.7–1.5
Alinen Rautjärvi	6	49.9	6198	125	12	6.5–6.6	0.07–0.14	61–270	427–596	17–21	2.4–4.3	0.3–1.2
Syrjänalunen	7	1	4	137	8	6.5–6.6	0.21–0.22	7–18	218–622	2–9	4.7–5.5	0.05–0.1
Huhmari ^a	8	1.1	14	128	8	6.4–6.6	0.18–0.25	85–140	310–972	23–73	5.2–6.6	0.8–1.4
Iso Valkjärvi	9	3.9	13	126	8	4.9–6.0	0–0.03	15–85	389–1107	11–59	0.7–1.2	0.07–0.16
Valkea-Kotinen	10	4.2	9	156	6	4.9–5.5	0–0.02	157–250	450–545	12–24	1.7–3.0	0.3–0.82
Haukijärvi ^{a,b}	11	2.2	580	131	8	5.8–6.7	0.07–0.19	170–495	393–691	22–42	3.8–5.9	0.4–1.6
Majajärvi ^{a,b}	12	3.9	186	133	12	5.6–6.1	0.05–0.1	205–412	624–650	13–27	3.5–4.2	0.6–0.8

^aLakes occupied by beavers during the study period (Vehkaoja et al. 2015)

^bLakes in the project “Ecologically sustainable fishing” during 2005–2013, including intensive pike removal (Lehtonen et al. 2017, Tiainen 2017)

Table 2 Lake-specific mean total length, weight, age, growth, length at age 5 years and measured THg concentration of pike

Lake	Length (cm)		Weight (g)	Age (yr)	Growth (cm yr ⁻¹)	L of 5 yr (cm)	THg measured (mg kg ⁻¹)		N pike
	(Mean)	(Range)					Average	Range	
Haarajärvi	50.5	23.0–97.0	915	5.9	8.5	34.5	0.85	0.73–2.87	45
Sorsajärvi	52.2	12.0–86.2	1231	6.4	7.8	39.5	0.92	0.36–1.83	37
Savijärvi	41.8	14.0–54.6	473	5.5	8.4	40.4	0.81	0.18–1.57	36
Rahtijärvi	46.3	22.0–84.5	739	6.1	7.4	38.9	0.81	0.27–1.40	40
Pitkäniemenjärvi	44.6	31.0–79.3	641	6.9	6.7	34.6	0.71	0.33–2.01	36
Alinen Rautjärvi	45.6	28.0–74.3	595	5.3	9.4	43.3	0.58	0.21–1.26	35
Syrjälanunen	37.1	18.0–70.7	488	5.1	7.8	39.1	0.19	0.05–0.46	43
Huhmari	40.2	20.0–64.0	500	5.3	7.9	41.0	0.54	0.19–1.02	26
Iso Valkjärvi	34.0	16.0–70.0	413	5.4	7.3	35.0	0.50	0.12–1.40	64
Valkea-Kotinen	45.5	22.5–78.0	777	7.4	n.d.	n.d.	0.60	0.20–1.74	40
Haukijärvi	50.6	20.0–99.5	1102	6.1	8.2	41.2	1.13	0.15–2.67	38
Majajärvi	40.4	27.0–70.3	576	5.9	7.7	36.5	1.15	0.67–1.95	49

Total length and weight were measured with an accuracy of 1 cm and 1 g for each fish, and sex was determined when possible (117 females and 95 males). Samples for mercury analyses were taken from dorsal white muscle and frozen (−20 °C) until analysed. Cleithrum bones of fish were taken for age determination (age data available for 93 females and 77 males) and back-calculation of growth (Cas-selman, 1996). As fish growth rate is known to affect mercury bioaccumulation (Göthberg, 1983; Verta, 1990), a simple proxy of pike growth for each lake was calculated by dividing the length of sampled fish by their age (mean annual length increment, Table 2).

2.3 Mercury Analyses

The mercury content of pike samples was determined in the Department of Biological and Environmental Sciences, University of Helsinki. In 1983–2002, a cold vapour atomic absorption spectrophotometric technique and a PerkinElmer/Bacharach Coleman 50b MAS analyser (detection limit 0.01 µg Hg g⁻¹) was used. Prior to and after the analyses, the calibration of the instrument was checked with a mercury standard solution, and acid-permanganate solution was used as blank samples during the analyses (Rask & Metsälä, 1991; Rask et al., 2007). Duplicate samples (0.1–0.25 g, ww) from each fish were analysed, and the mean content of duplicates was used in subsequent analyses. The values of fish mercury concentrations are

given as THg, mg kg⁻¹ of fresh muscle tissue (wet weight, ww). For ascertaining the quality of analyses, two common reference materials were used: dogfish muscle (National Research Council Canada, Certified Reference Material DORM-1, mercury content 0.794 ± 0.074 mg kg⁻¹) and cod muscle (European Commission Community Bureau of Reference, BCR-422, mercury content 0.559 ± 0.016 mg kg⁻¹).

In 2013, an atom absorption spectrometer direct mercury analyser (Milestone DMA-80, Sorisole, Italy) was used. Certified reference materials DORM-4 fish protein (National Research Council, Canada, mercury concentration 0.410 ± 0.055 mg kg⁻¹) and cod muscle (European Commission Community Bureau of Reference, BCR-422, mercury content 0.559 ± 0.016 mg kg⁻¹) were used for quality assurance of the analyses at the beginning and end of each run.

2.4 Statistical Analyses

The effects of year (1983, 1993, 2002 and 2013), lake (12 lakes), fish sex, fish length (covariate) and their interactions on THg concentration was analysed with a general linear mixed model (GLMM).

The study design is a four-timepoint (year) repeated measurement structure. Within each timepoint, 4–21 THg measurements from random samples of pike of different sizes (12–100 cm, $n = 490$) were obtained from 12 lakes. The fish length was regarded

as a continuous variable, and therefore, the data were analysed with a random slope model in which a separate slope was fitted for each year and lake. A general linear mixed model was fitted using a normal distribution (identity link) (Stroup, 2013), and the Kenward-Roger approximation was used for analysis of degrees of freedom. The distribution of pike THg values was right skewed and therefore ln-transformation was applied. All analyses were conducted in log scale, and the results were back transformed with an exponential function and presented in the original scale.

The symmetricity of residual distribution and model fit was checked from the shape of Pearson residuals and from the observed vs predicted plots. The effect of outliers was checked by reanalysing datasets in which observations with Pearson residual $> |2.5|$ were excluded. Results were not changed, and therefore, the original dataset was used for analysis. The modelling was performed by the GLIMMIX procedure of the SAS/STAT software (version 9.4, SAS Institute, 2018).

The THg model structure for lake and year effects is:

$$\begin{aligned} \log(\text{THg}) = & \text{year}(F) \\ & + \text{lake}(F) \\ & + \text{year} \times \text{lake}(F) + \text{length}(C) \\ & + \text{length}(R \text{ slope}) \end{aligned}$$

where F stands for fixed effect, C for covariate and R for random effect. Because the fish for each lake and year were random samples from the fish population, the data were analysed with a random slope (fish length) model in which a separate slope was fitted for each year and lake.

To estimate the effects of sex, growth (mean annual length increment) and environmental variables (lake depth, water colour, alkalinity, total phosphorus,

total nitrogen, Fe and Ca) on pike THg, the parameters were separately added to the linear model as an additional covariate. Inter-correlation among the environmental variables (Table 3) was not taken into account.

The model-fitted year THg estimates are covariate-adjusted, and they answer to the question about association between pollution, climate and natural factors and THg concentration. The slope estimates of covariates show the direction (decrease, increase) and strength of the effect. The association cannot be directly seen because of different age and size distributions of fish in different lakes but needs to be modelled.

In total, 10 different THg models were fitted and are presented in Table 4 (models 1–10). For year and lake comparisons (Table 4, model 1), the model-estimated averages were obtained from contrast specification which consists of model effects (year, lake, length) and G-side random effects (length) and their coefficients (ESTIMATE statement in SAS PROC GLIMMIX). The pairwise comparisons of years and years within lakes were calculated with linear contrasts including fixed effects, covariates and random slope effects (ESTIMATE statement). Due to multiple statistical tests, p -values and confidence intervals in each test set were adjusted by using a simulation-based correction in a step-down fashion (Westfall, 1997, SAS 9.4 documentation: PROC GLIMMIX, ESTIMATE statement). This is a less conservative approach compared to the Bonferroni method. Conclusions from statistical comparisons are based on adjusted p -values and confidence intervals when applicable.

For year, lake and sex comparisons (Table 4, models 1 and 2), the fish length was adjusted to 54.0 cm, which corresponds to a 1-kg fish (Rask & Metsälä, 1991). With the rest of the THg models

Table 3 Pearson correlation coefficients for environmental variables; significant correlations in bold

	Depth	Alk	Colour	Ntot	Ptot	Ca	Fe
Depth	1	0.12	0.36	-0.02	-0.06	0.06	0.33
Alk	0.12	1	-0.08	-0.20	-0.14	0.84	0.28
Colour	0.36	-0.08	1	0.28	0.39	0.29	0.71
Ntot	-0.02	-0.20	0.28	1	0.64	-0.04	-0.02
Ptot	-0.06	-0.14	0.39	0.64	1	-0.05	0.28
Ca	0.06	0.84	0.29	-0.04	-0.05	1	0.39
Fe	0.33	0.28	0.71	-0.02	0.28	0.39	1

Table 4 General linear mixed model structure for all statistical models; the target variable describes the main interest for each model, and the target describes the model outcome from which the conclusions are drawn (UN unstructured, Vc variance component)

Model	Response variable	Fixed variable	Covariate	Random variable	Subject	Covariance structure	Target variable	Target
1	ln(Hg)	Year, lake Year × lake	Length	Length	Lake × year	Vc	Year, lake Year × lake	Averages Differences
2	log(Hg)	Year, sex	Length	Intercept Length	Lake × year	UN	Sex	Averages Difference
3	log(Hg)	Year	Growth Length	Intercept Length Growth	Lake × year	UN	Growth	Trend
4	log(Hg)	Year	Depth Length	Intercept Length	Lake × year	UN	Depth	Trend
5	log(Hg)	Year	Alkalinity, Length	Intercept Length	Lake × year	UN	Alkalinity	Trend
6	log(Hg)	Year	Colour Length	Intercept Length	Lake × year	UN	Colour	Trend
7	log(Hg)	Year	tot N Length	Intercept Length	Lake × year	UN	tot N	Trend
8	log(Hg)	Year	tot P Length	Intercept Length	Lake × year	UN	tot P	Trend
9	log(Hg)	Year	Ca Length	Intercept Length	Lake × year	UN	Ca	Trend
10	log(Hg)	Year	Fe Length	Intercept Length	Lake × year	UN	Fe	Trend
11	Growth	Year, sex Year × sex	Length	Intercept Length	Lake × year	UN	Year, sex Year × sex	Averages Differences
12	Growth	Year, sex Year × sex	Age	Intercept Age	Lake × year	UN	Year, sex Year × sex	Averages Differences

(Table 4), the adjustment was done for a 43.9-cm length (data average). For details of the THg model calculations, see the supplementary information (Tables SI 1 and SI 2).

The effects of year (1983 and 2013), lake (12 lakes), fish sex, fish length or age (covariate) and their interactions on growth were analysed with a GLMM in the same manner as above. Two different growth models were fitted (Table SI 3), and statistical models are presented in Table 4 (models 11 and 12).

3 Results

3.1 No Consistent Change in the Mean Pike THg During 1983–2013

The model-estimated annual average pike THg of all 12 lakes, length-corrected to 54 cm, was 0.72–0.88 mg kg⁻¹ (ww) during 1983–2013. The highest concentrations occurred in 2002 and were significantly higher than those in 1983 (GLMM, $t = 4.41$, $p < 0.0001$) and 2013 (GLMM, $t = 2.86$, p

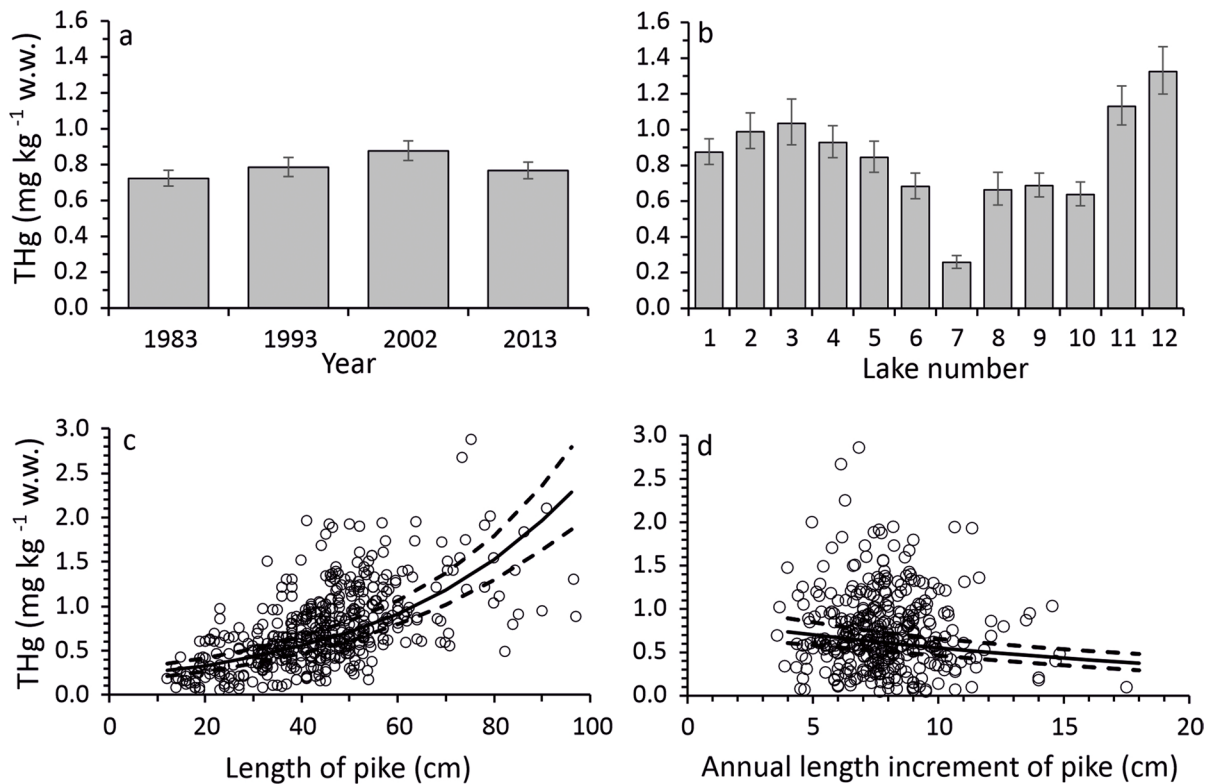


Fig. 2 Average ($\pm 95\%$ CI) model-estimated and length-corrected (54 cm) pike THg **a** of all study lakes in the four sampling campaigns and **b** in single lakes over the study period of

four decades, with lake numbers as in Table 1 and Fig. 1, and the relation of pike THg to **c** fish length and **d** growth (=average annual length increment) of pike

< 0.0231, Fig. 2a, Table SI 1). Average estimated THg concentrations of different lakes over the four-decade study period varied between 0.26 and 1.33 mg kg⁻¹ (ww) (Fig. 2b, Table SI 1), and concentrations measured from individual pike ranged from 0.05 to 2.87 mg kg⁻¹ (Table 2). The lake, year and pike length (Fig. 2c) statistically significantly affected the pike THg concentration (GLMM, Table SI 2). Length was always the most important variable in all GLMM.

There was a statistically significant difference in the model-estimated average pike THg between the sexes (GLMM, $t = -2.94$, $p < 0.0038$), the concentrations (mean $\pm 95\%$ CI) being higher in males, 0.66 ($n = 95$, 0.48–0.90) mg kg⁻¹ (ww) than in females 0.59 ($n = 117$, 0.43–0.81) mg kg⁻¹ (Table SI 1).

The growth of pike was slow, with the mean total length of 5-year-old fish ranging between 34.5 and 43.5 cm among the different lakes (Table 2). Nevertheless, the growth, given as the average annual

length increment (Fig. 2d), still had a negative effect on the THg concentration of pike (GLMM, $F = 32.35$, $p < 0.0001$, Table SI 2), with a 4.7 % (95 % CI 3.1–6.3) decrease per 1-cm increase in the annual growth of pike. The average growth was significantly higher for females than males, 8.1 and 7.5 cm per year, respectively (GLMM, $t = 3.07$, $p = 0.0025$, fish age as a covariate (Table SI 3)).

3.2 Pike THg and Lake Characteristics

Lake depth statistically significantly (GLMM, $F = 7.01$, $p < 0.0116$) affected the pike THg, with an average increase of 0.05 mg kg⁻¹ (ww) THg per 1-m increase in the maximum depth of lakes (Fig. 3a, Tables 5 and SI 2). Among water quality parameters, pike THg concentrations increased significantly with increases in DOC-related water colour (GLMM, $F = 26.91$, $p < 0.0001$, Fig. 3b), total phosphorus, total nitrogen and iron (Tables 5

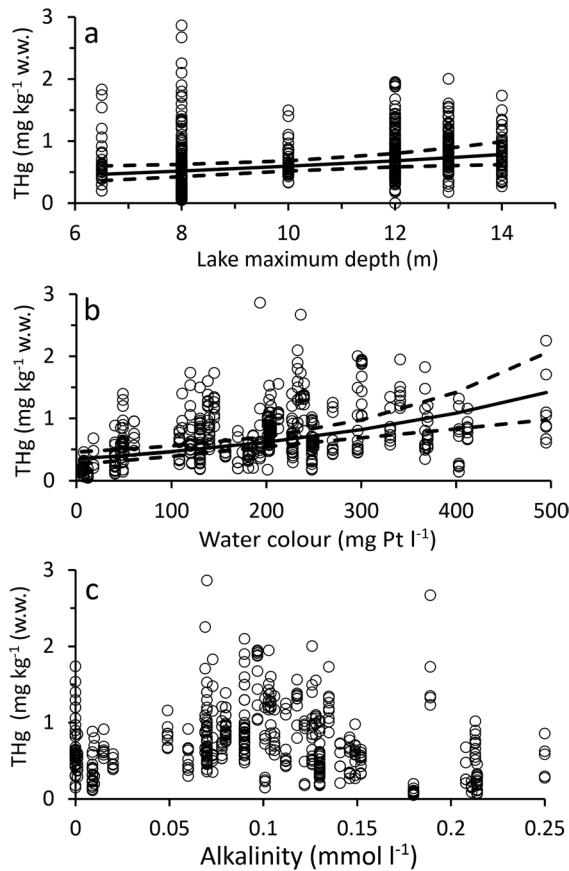


Fig. 3 The relation of pike THg concentration to **a** the maximum lake depth, **b** lake water colour and **c** lake alkalinity

and SI 2). Pike THg was not significantly related to the acidification-related parameters alkalinity (GLMM, $F = 2.50$, $p = 0.123$, Fig. 3c) and calcium (Tables 5 and S2).

3.3 Lake-Specific Responses in Pike THg

The model-estimated average pike THg in the chain lakes (lakes 1–6 in Table 1 and Fig. 1) was 0.56–0.84 mg kg^{-1} in the first sampling campaign of 1983–1984 (Fig. 4a). In all chain lakes, the highest average pike THg occurred in 2002, with the highest levels of 1.41 mg kg^{-1} (ww) in L. Savijärvi and the lowest in L. Alinen Rautjärvi (0.82 mg kg^{-1} ww). In 2013 the average pike THg of chain lakes was 0.80–1.01 mg kg^{-1} (ww). The pattern of pike THg was fairly similar throughout the lake chain (Figure 4a), and statistically significant differences in pairwise comparisons of decades were found in each lake (Table 6).

In the headwater lakes, the variation in pike THg was wider than that in the chain lakes (Fig. 4b), from measured concentrations of 0.05–0.46 mg kg^{-1} (ww) in the clear groundwater lake L. Syrjälanunen to 0.15–2.67 mg kg^{-1} (ww) and 0.67–1.95 mg kg^{-1} (ww) in the brown water lakes L. Haukijärvi and L. Majajärvi, respectively (Table 2).

In the acidified lakes L. Iso Valkjärvi and L. Valkea-Kotinen, significantly higher pike THg concentrations were measured in 1983 (1.19 and 0.81 mg kg^{-1} , respectively) than in the following sampling campaigns (GLMM, $p < 0.001$, Table 6), coinciding with the years of lowest pH and alkalinity of the lakes (Table 1). Since then, the pike THg in these lakes varied between 0.51 and 0.77 mg kg^{-1} (ww), with a slight decrease in the clear water lake L. Iso Valkjärvi and with an increasing trend in the brown water lake L. Valkea-Kotinen (Fig. 4b).

The outlets of the chain lake Haarajärvi and the headwater lakes Huhmari, Haukijärvi and Majajärvi were dammed by beavers temporarily in the 1980s, resulting in rises of floodwater into the surrounding

Table 5 GLMM results to test the effects of lake characteristics on pike THg concentrations; significant effects in bold

Effect	Log scale	Std. err.	DF	t value	Pr > t	95% Conf. int.		Data scale change/effect unit		
	Estimate					Lower	Upper	Estimator	Lower	Upper
Max. depth	0.069	0.026	39.19	2.65	0.0116	0.016	0.122	1.072	1.016	1.130
Alk	−1.892	1.197	40.47	−1.58	0.1218	−4.311	0.527	0.151	0.013	1.694
Colour	0.0028	0.00054	38.27	5.19	<.0001	0.0017	0.0039	1.0028	1.0017	1.0039
Ntot	0.0021	0.00067	37.12	3.10	0.0037	0.00072	0.0034	1.0021	1.00072	1.0034
Ptot	0.024	0.0073	36.88	3.22	0.0026	0.0088	0.038	1.024	1.0088	1.039
Ca	−0.035	0.061	31.98	−0.57	0.5732	−0.159	0.090	0.966	0.853	1.094
Fe	0.566	0.154	32.27	3.67	0.0009	0.252	0.879	1.760	1.286	2.409

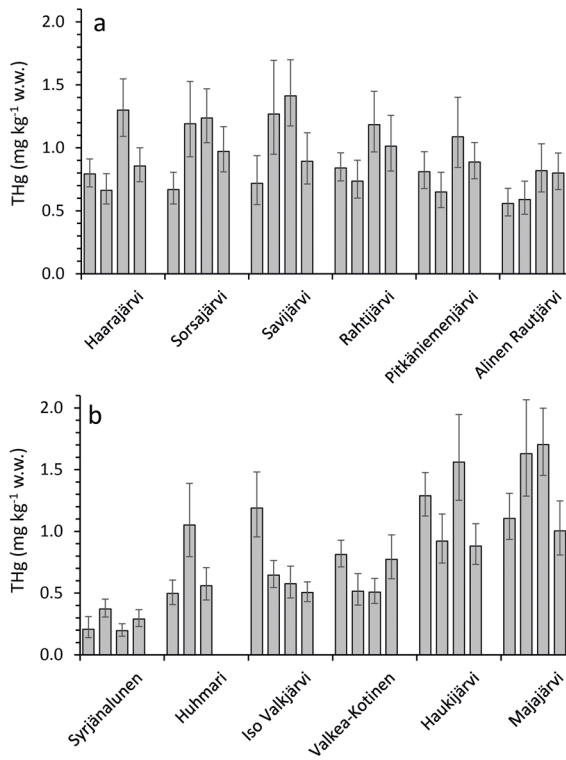


Fig. 4 Length-corrected (54 cm) pike THg (geometric mean \pm 95 % CI) in **a** chain lakes and **b** headwater lakes during the sampling campaigns of four decades from the 1980s to the 2010s

riparian forest and wetland. The beaver floods were followed by high variation in the pike THg of these lakes (Fig. 4b). The clearest example of beaver effect was L. Huhmari, where the first beaver flood took place in 1990 and doubled the pike THg content from 0.50 mg kg⁻¹ (ww) in 1983 to 1.05 mg kg⁻¹ (ww) in 1993 (GLMM, $p = 0.0001$). After disappearance of beavers, the pike THg dropped again, to 0.56 mg kg⁻¹ (ww) in 2002 (GLMM, $p = 0.0027$ (Tables 6 and SI 1).

In the three lakes (L. Haarajärvi, L. Haukijärvi, L. Majajärvi) targeted for intensive pike fishing during 2008–2012, the pike THg decreased from 2002 to 2013 more than in any other study lake, excluding L. Savijärvi (Fig. 4). In L. Haarajärvi, the uppermost lake of the lake chain, the decrease in pike THg from 2002 to 2013 was 0.44 mg kg⁻¹ (ww) whereas the decrease was 0.02–0.26 mg kg⁻¹ in other chain lakes except L. Savijärvi with a 0.52 mg kg⁻¹ (ww) drop. In the headwater lakes L. Haukijärvi and L. Majajärvi, the decrease in pike THg from 2002 to 2013 was 0.68 and 0.70 mg kg⁻¹ (ww), respectively. The three fish-manipulated lakes, L. Haarajärvi, L. Haukijärvi and L. Majajärvi, together with L. Savijärvi, were the only ones with a statistically significant decrease of pike THg from 2002 to 2013 (GLMM, $p = 0.0019$, 0.0007, 0.0007 and 0.0095, respectively, Tables 6 and SI 1).

Table 6 Statistical significance of between-year differences in pike THg concentrations in the study lakes with p -values adjusted for multiple comparisons within lake level (significant differences in bold); detailed results in Table SI 1

Lake	1983–1993	1983 vs 2002	1983 vs 2013	1993 vs 2002	1993 vs 2013	2002 vs 2013
Haarajärvi	0.2200	<.0001	0.4789	<.0001	0.0898	0.0019
Sorsajärvi	0.0015	<.0001	0.0188	0.8078	0.3066	0.1448
Savijärvi	0.0158	0.0002	0.3839	0.5380	0.1483	0.0095
Rahtijärvi	0.4860	0.0250	0.3443	0.0063	0.1068	0.4860
Pitkämäenjärvi	0.3026	0.1929	0.4608	0.0126	0.0940	0.3179
Alinen Rautjärvi	0.9200	0.0519	0.0394	0.1129	0.1129	0.9200
Syrjäälunnen	0.0406	0.8049	0.2513	0.0005	0.2513	0.0822
Huhmari ^a	0.0001	0.6900		0.0027		
Iso Valkjärvi	<.0001	<.0001	<.0001	0.5140	0.0931	0.5140
Valkea-Kotinen	0.0059	0.0007	0.9150	0.9280	0.0453	0.0218
Haukijärvi	0.0269	0.2753	0.0051	0.0038	0.7635	0.0007
Majajärvi	0.0243	0.0013	0.7432	0.7592	0.0114	0.0007

^a Year 2013 missing

4 Discussion

Our study clearly demonstrates that mercury concentrations of pike in small boreal headwater lakes of southern Finland have remained high despite the ca. 50 % decrease of mercury concentration in deposition from $11 \mu\text{g L}^{-1}$ in the late 1980s (Iverfeldt, 1991) to levels 4–6 $\mu\text{g L}^{-1}$ in the 2000s (Wängberg et al., 2010; Ruoho-Airola et al., 2014). Pike THg was strongly linked to biological characteristics like length, growth and sex of fish as observed in many earlier studies (e.g. Madenjian et al., 2014; Moslemi-Aqdam et al., 2022). Among environmental factors, water colour was important, as in most lakes the highest pike THg concentrations were recorded in 2002 after consistent browning of lakes during the 1990s due to decreased sulphate deposition (Arvola et al., 2010; Arzel et al., 2020), suggesting a dominant role of DOC in the lake water Hg dynamics.

Furthermore, the model-estimated average mercury concentrations exceeded the EU recommended health limit (1.0 mg kg^{-1} , ww) for human consumption of pike (EU, 2006) at least in one sampling campaign in 9 out of the 12 lakes. These concentrations are higher than those measured from large lakes and coastal brackish waters of Baltic Sea, from where the majority of pike for human consumption are caught (Ahonen et al., 2018; Airaksinen et al., 2018; Braaten et al., 2019).

4.1 Browning of Lakes

Pike THg content was significantly affected by water colour, emphasizing the importance of increased organic carbon load to mercury dynamics in catchment-lake systems (Porvari, 1998; Braaten et al., 2020; Moslemi-Aqdam et al., 2022). The occurrence of the highest pike THg levels during the 2002 sampling campaigns followed the most intensive period of browning of the lakes in the study area which took place during the 1990s (Arvola et al., 2010; Arzel et al., 2020; Moslemi-Aqdam et al., 2022). Significant positive relations of total P, total N and iron to the pike THg in the lakes of the present study were probably caused by their coupling with the humic substances as well (Kortelainen et al., 1986; Kozak et al., 2023). An increasing DOC concentration shifts lakes from phytoplankton primary production towards heterotrophic bacterial production (Forsström

et al., 2013) that may also promote both within-lake methylation processes and more efficient Hg transfer in the food chain (Moslemi-Aqdam et al., 2023). The rise of DOC concentrations in lakes has also been considered to be the major reason for recent increases in mercury concentrations in the fish of lakes recovered from acidification (Wyn et al., 2010; Hongve et al., 2012;), but in some cases, the growth dilution phenomenon has been shown to be more important (Keva et al., 2017; Moslemi-Aqdam et al., 2021; Rask et al., 2021).

4.2 Acidified Lakes

In our study, the two lakes with $\text{pH} < 5.0$ during the 1980s, L. Iso Valkjärvi and L. Valkea-Kotinen (Rask & Metsälä, 1991), were the only ones with the highest pike THg measured during the first sampling campaign. This supports the role of acidification as a reason for high pike THg concentrations in the 1980s during the years of the highest sulphate deposition (Verta et al., 1990; Rask et al., 2021). In L. Iso Valkjärvi, liming in the early 1990s (Rask et al., 1996) enhanced the decrease of pike THg in the lake but a decrease also took place in the control basin of the lake (Rask et al., 2007). Later, during 2007–2012, intensive fishing of perch (Olin et al., 2017b) may have contributed to the mercury dynamics and continuous decrease of pike THg in the lake, as happened also with the perch (Rask et al., 2007). In L. Valkea-Kotinen, the significant increase of pike THg up to the last sampling campaign in 2013 is apparently related to the browning of the lake (Vuorenmaa et al., 2014; Rask et al., 2021).

4.3 Beavers

Beaver floods affect the water chemistry and biota of small headwater lakes (Vehkaoja et al., 2015; Nummi et al., 2018; Nummi et al., 2021). The results of Vehkaoja et al. (2015) showed that water colour increased especially during the first three beaver-impoundment years, while concentrations of dissolved oxygen simultaneously decreased. In the beaver lakes, increased leaching of mercury from riparian areas and low oxygen concentration in the hypolimnion of the lakes associated with the flooding may increase within-lake methylation processes (Eckley et al., 2005), jointly

promoting mercury biomagnification through the food web and culminates as increased pike THg (Moslemi-Aqdam et al., 2023). Our observations are in line with those by Painter et al. (2015), who showed that beaver impoundments can increase the availability and subsequent uptake of MeHg by basal food web organisms in beaver ponds. It has also been found that, during the comeback of beavers after extirpation, the pioneer inundation caused elevated MeHg concentrations in the system, but later re-floodings produced only negligible increases (Levanoni et al., 2015). Moreover, young beaver ponds, but not the old ones, tend to be a source of methylmercury in water (Ecke et al., 2017). In our study, the first beaver flood in L. Huhmari took place in 1990 and resulted in a sharp increase of pike THg.

4.4 Intensive Fishing

Fish removal has been suggested as a method to promote fish growth and hence decrease the Hg levels in remaining fish, and the risk to humans of fish consumption (Göthberg, 1983; Verta, 1990, Sharma et al., 2008). Three of our study lakes, L. Haarakjärvi, L. Haukijärvi and L. Majajärvi, were targeted for effective pike removal during 2008–2012, with 30–50 % of pike biomass removed annually (Tiainen et al., 2017). An increase in the growth of pike was recorded in all lakes, especially in L. Haarakjärvi, where the increase was statistically significant in all age groups (Tiainen, 2017). Thus, the growth dilution apparently had a role in explaining the decrease of pike THg in these lakes from 2002 to 2013. Apparently, this was also the reason for the difference in THg concentration between the sexes in our study, as female pike grew significantly faster than males (see also Casselman, 1996). The three pike manipulation lakes were also inhabited by beavers in certain years (Vehkaoja et al., 2015), but during the “Ecologically sustainable fishing” project 2005–2013, beaver floods took place only in L. Haukijärvi. The decrease of pike THg also in this lake from 2002 to 2013 emphasizes the importance of pike population for the mercury dynamics of the lakes (see Sharma et al., 2008).

4.5 Forest Management

Although the sampling programme for our study did not offer sufficient context for detailed assessment of the impacts of forest harvesting on pike THg, the forestry in the lake catchments likely has increased DOC and mercury load to the lakes and overall variation in the pike THg concentrations (Moslemi-Aqdam et al., 2022; Moslemi-Aqdam et al., 2023). An earlier study showed that catchment deforestation (46 % of the catchment area) and associated burning of logging remains resulted in changes in water quality, increases in algal primary production and decomposition of organic matter in one small uppermost lake in our study area (Rask et al., 1993). This was accompanied by an increase in THg in cladoceran zooplankton and fish (Rask et al., 1994). A Canadian study (Garcia & Carignan, 2000) reported an effect of forest clearcutting on fish THg, with Hg levels of northern pike in logged lakes twice as high as those in reference lakes. A Swedish study (Wu et al., 2018) reported an average increase of 26 % in Hg concentrations of large perch from lakes after clearcutting of their catchments.

4.6 Climate Change

Effects of climate change in our study area have been recorded as a statistically significant increase in annual and spring mean air temperature, thinning of annual mean snow depth and shortening of the lake-ice season (Jylhä et al., 2014). Dissolved organic carbon concentrations in lakes have increased, partly due to decreasing sulphate deposition and partly by climate change-induced hydrological changes with increasing runoff and leaching of substances from catchments to lakes (Arvola et al., 2010; Vuorenmaa et al., 2014). The consequent browning of lake waters has led to stronger stratification and lowered oxygen concentration (Vuorenmaa et al., 2014; Couture et al., 2015), which may elevate the mercury concentration of perch and pike (Rask et al., 2010; Ahonen et al., 2018; Kozak et al., 2021). The higher pike THg concentrations in our deeper study lakes may have been due to the relatively larger volume of anoxic hypolimnetic water in the deeper lakes. In L. Valkea-Kotinen, the primary production decreased during the 20-year period 1990–2009 mainly due to climatic forcing and consequent hydrological changes and increased DOC

loading (Arvola et al., 2014). The decrease of general productivity of the lake resulted in slower growth of perch (Rask et al. 2014b), which may have contributed to the increase in perch THg (Rask et al., 2010; 2021) and also explains the increase of pike THg (Moslemi-Aqdam et al., 2021) as recorded in the present study.

4.7 Concluding Remarks

The results of the present study indicated high pike mercury levels in pristine boreal headwater lakes from the 1980s to 2010s. The ca. 50 % decrease of mercury deposition during this period did not result in generally lower pike THg levels. The reasons for this, and even increased pike THg until the shift of the millennium, were most probably connected to the regional effects of decreased atmospheric SO₄ deposition on increased DOC mobility and leaching and on mercury dynamics in lakes and their catchments, together with the complex effects of climate change. Thus, the answer to our original study question 1 is no, as no regular decrease in pike THg took place over decades. The answer to question 2 is yes, as pike THg concentrations increased during the course of lake browning. The answer to questions 3 and 4 on local effects is yes, as beaver floods resulted in increases of pike THg, while intensive pike removal decreased pike THg. Taking into account deposition (mercury and other elements), environmental (lake and catchment) and biotic (size, sex, age and growth of fish) variables is thus crucial for disentangling the most important factors behind decadal trends in fish mercury.

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Data Availability The datasets generated and analysed during the current study are available from the corresponding author on reasonable request.

Declarations

Conflict of Interest The authors declare no competing interests.

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