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High Mercury Concentrations of European Perch (*Perca fluviatilis*) in Boreal Headwater Lakes with Variable History of Acidification and Recovery

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Abstract High dissolved organic carbon and low pH are often associated with elevated mercury content of fish in boreal lakes, but less is known about the fish mercury dynamics in lakes recovering from acidification stress. We measured total mercury concentration (THg) in muscle of European perch (*Perca fluviatilis*) and evaluated the overall correlation with environmental and growth variables in 24 boreal headwater lakes in the 2010s. We found negative correlations of length-corrected perch THg with lake pH and perch growth, but no correlation with dissolved organic carbon. The main emphasis in the present study was focused to a subset of ten lakes

in southern Finland with known perch THg during severe acidification in the 1980–1990s. The comparison of perch THg concentrations in the 2010s with values determined in the 1980–1990s showed a sharp increase in most acidic lakes where the perch populations suffered from severe acid stress in the 1980s. This increase was attributed to growth condensation caused by sharp decrease in perch growth during recovery of reproduction capacity and the consequent increases in population densities of perch. Our results highlight the importance of perch growth rate and population density for understanding the variability of fish Hg in boreal headwater lakes, where recovery from acidification can lead to higher mercury concentration of fish in certain cases.

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1 Introduction

Acidification of poorly buffered boreal and alpine lakes in the 1900s due to air pollutants resulted in population declines of several sensitive fish species in northern Europe, the USA and Canada (Almer et al., 1974; Baker et al., 1993; Hesthagen et al., 1999). Tammi et al. (2003) estimated with a postal survey based on the Fennoscandian water chemistry data (Henriksen et al., 1998) that acidification-induced

reproduction failures and subsequent declines and extinctions of fish populations occurred in more than 10,000 Nordic lakes (Finland, Norway, Sweden). The estimate for Finland was 1850 fish populations, out of which 410 were affected perch populations. This is in line with estimates of critical levels of acidification for common fish species and modelled number of lakes that acidified over those levels in Finland (Rask et al., 1995). A gillnet survey of 80 lakes indicated low fish status in strongly acidified lakes of southern Finland (Rask & Tuunainen, 1990). These observations were ascertained with more detailed studies, including mark and recapture studies of perch populations in lakes in different degree of acidification (Lappalainen et al., 1988) and field and laboratory experiments on survival of perch embryos in acidic waters (Rask et al., 1990; Vuorinen et al., 1992). Studies of growth responses in remaining perch in affected lakes indicated exceptionally high growth rate in some lakes in conditions of abundant macroinvertebrate food availability with no food competition in the scarce perch populations (Nyberg et al., 1995; Raitaniemi et al., 1988; Rask et al., 2014a). High mercury concentrations have been often reported in fish from acidified lakes (Greenfield et al., 2001; Richman et al., 1988), but several studies have shown that the impacts of acidification or sulphate deposition on the dynamics of mercury are complex (Bloom et al., 1991; Wyn et al., 2010; Braaten et al., 2020; Watras et al., 2020).

International agreements to reduce sulphur emissions led to decreased acid deposition and a subsequent gradual increase of lake buffering capacity and pH (Stoddard et al., 1999; Vuorenmaa & Forsius, 2008) and associated recovery of fish reproduction (Fjellheim et al., 2007; Keller et al., 2007; Nyberg et al., 1995). As a consequence of decreased sulphur deposition, acidity of soil and ionic strength of soil water have decreased acting through chemically-controlled organic matter solubility in catchment soils, and dissolved organic carbon (DOC) loads from catchments to lakes have increased resulting in higher DOC concentrations in lake waters (Monteith et al., 2007; Vuorenmaa et al., 2006). Recent changes in climatic conditions, such as increased precipitation and discharge, are exerting a greater influence on variation and increasing DOC concentrations in surface waters (de Wit et al., 2016; Zwart et al., 2017).

Increasing DOC levels, often termed lake browning, have several important consequences in lake ecosystems including decreased light penetration in water, increased bacterial activity, reduced fish production and increased hypolimnetic anoxia (e.g. Arvola et al., 2014; Creed et al., 2018; Rask et al., 2014b). Because DOC compounds form complexes with metals, including Hg, the mercury load from catchments to lakes has increased which may result in increased mercury in fish in some lakes (Braaten et al., 2014a; Driscoll et al., 1995; Hongve et al., 2012). However, the mercury content of fish muscle is the outcome of a combination of deposition, environmental and biotic variables, and those are often region- and lake-specific. Recovery of boreal lakes from acidification provides an opportunity to evaluate the underlying factors behind mercury content of fish muscle (Braaten et al., 2020; Hrabik & Watras, 2002; Watras et al., 2006).

Atmospheric mercury originates from both natural and anthropogenic sources, of which the latter has been increasingly dominant since the industrial revolution (Chen et al., 2018; Pacyna et al., 2010; Streets et al., 2011). After the Minamata Bay accident, there was increasing interest in measuring fish mercury content globally, including in the industrial regions of northern Europe and North America. In Finland, fish mercury concentrations up to 5–6 mg kg⁻¹ (ww) were recorded due to industrial pollution in the late 1960s (Häsänen & Sjöblom, 1968). Since then, the fish Hg concentrations in Nordic countries have decreased due to the ban of the use of Hg as a slimeicide in the pulp and paper industry and due to the considerable reductions in Hg discharges from chloralkali plants (e.g. Åkerblom et al., 2014; Lodenius, 1991; Verta, 1990a). In a Finnish fish mercury survey in the early 2000s, a significant decreasing trend of pike (*Esox lucius* L.) mercury was recorded in lakes subject to industrial pollution in earlier decades (Paloheimo, 2005). However, in headwater lakes, where mercury mainly originates from atmospheric deposition to the catchment and to the lake surface, the Hg concentrations of pike had remained at similarly high levels as recorded in earlier surveys (Paloheimo, 2005). Taking into account the decrease in mercury emissions from European sources during the early 1990s (Travnikov et al., 2012), records of the decreases in surface sediment Hg concentrations (Mannio, 2001) and reductions in mercury deposition since the early 1990s

(Ilyin et al., 2015; Ruoho-Airola et al., 2014; Wängberg et al., 2010), lowered fish mercury concentrations could have been expected (Braaten et al., 2019).

Mercury bioaccumulates in organisms and biomagnifies in aquatic food webs in the form of methylmercury (MeHg) which may enter lakes through precipitation, watershed runoff and in-lake and catchment methylation of inorganic Hg (Morel et al., 1998; Ullrich et al., 2001). The relative importance of these different sources varies depending on the amount of deposition, lake type and catchment hydrology (Rudd, 1995). Eckley et al. (2005) suggested that in-lake methylation of Hg can be more important than MeHg transport from the drainage basin, in spite of the dominance of wetlands in lake catchments. However, intensive forestry-related land use in boreal catchments may increase both methylation hot-spots as well as run-off of historical wetland mercury via machinery wheel tracks and ditching activities (Ahonen et al., 2018; Eklöf et al., 2018).

Mercury in water is predominately methylated by sulphate reducing bacteria (SRB) under anoxic conditions (Choi et al., 1994; Ullrich et al., 2001). SRB occur commonly in the chemocline of oxygen-stratified headwater lakes in Finland (Arvola et al., 1992; Karhunen et al., 2013) resulting in increased MeHg concentrations below the oxycline of the lakes (Verta et al., 2010). The biomagnification potential of MeHg in aquatic food webs is much higher than that of total mercury (THg), illustrated by an increasing proportion of MeHg along food chains from phytoplankton c. 15%, to zooplankton c. 30% to fish c. 90–95% (Watras et al., 1995, 1998). In fish muscle tissue, THg can be efficiently used as a proxy for MeHg concentration (Downs et al., 1998; Lescord et al., 2018).

In addition to differences in lake and catchment characteristics and in-site specific mercury dynamics (Parkman & Meili, 1993; Richman et al., 1988; Simonin et al., 2008), the structure and function of food webs affect the bioaccumulation of mercury into fish (Futter, 1994; Rask et al., 1996; Yu et al., 2011). Increasing trophic position and pelagic energy sources are often related to higher mercury content of fish (Kahilainen et al., 2016; Karimi et al., 2016). This is the case with the European perch, *Perca fluviatilis* L. (Braaten et al., 2014b; Kraemer et al., 2012), which is a generalist carnivore characterized by ontogenetic diet shift from zooplankton to macroinvertebrates

and fish (Allen, 1935; Estlander et al., 2010). Given the rapid changes in food web structure in recovering acidified lakes, including population and growth responses of fish (Keller et al., 2007; Nyberg et al., 2010; Rask et al., 2014a), further increase in the variability in fish mercury concentrations can be expected.

We studied water chemistry, perch growth and perch mercury content in 24 small acid-sensitive boreal headwater lakes in southern Finland to test the main underlying correlations. We predicted a negative correlation of fish THg content with pH and growth rate, but a positive correlation with DOC (Driscoll et al., 1995; Watras et al., 1995). In 10 of the lakes, perch mercury concentrations had been measured during the 1980–1990s (Metsälä & Rask, 1989), and from these lakes with variable history of acidification and recovery, we predicted decreased THg in perch muscle as pH has increased and deposition of sulphate and mercury decreased (Ruoho-Airola et al., 2014).

2 Material and Methods

2.1 Study Area

For the present study, we sampled 24 natural headwater lakes for perch THg. All the lakes are in southern Finland, mainly in conservation or nature management areas with limited land-use activity. The catchment areas of the lakes are relatively small and comprise a mixture of coniferous and deciduous boreal forest and peatland or rocky landscape. The lakes are small with surface area between 1 and 43 ha, maximum depth 3–19 m, and altitude 16–156 m above sea level (Table 1). Water quality data were obtained from the databases of the Department of Environmental Sciences and Lammi Biological Station, University of Helsinki, and from the Finnish Environment Institute (http://www.syke.fi/open_data). The lakes are acidic with a pH-range 4.6–6.4 and alkalinity from –0.05 to 0.1 mmol l⁻¹ during the autumn turn over. Water colour and DOC ranged from 15–395 mg Pt l⁻¹ and 2.9–24.5 mg l⁻¹, respectively (Table 1). Sixteen of the 24 lakes have a known history of acidification and recovery from acidification (Forsius et al., 1990; Mannio, 2001; Vuorenmaa & Forsius, 2008), browning (Vuorenmaa et al., 2006), fish responses to acidification and recovery (Lappalainen et al., 1988;

Table 1 Location, lake area, maximum depth and water quality of the 24 study lakes in the 2010s

Lake	Latitude	Longitude	Altitude (masl)	Lake area (ha)	Max. Depth (m)	pH	Alk (mmol L ⁻¹)	Colour (mg Pt L ⁻¹)	DOC mg L ⁻¹	Tot. N (µg L ⁻¹)	Tot. P (µg L ⁻¹)	Ca (mg L ⁻¹)	Acidification history
Pöksynhaara	60.319	24.584	80	2.7	9	4.6	-0.034	140	14.0	740	8.3	0.38	
Teinipussi	60.134	23.343	64	7	5	4.7	-0.021	160	15.6	790	18.0	0.51	
Valkojärvi	60.129	23.345	67	2.8	7	4.9	-0.018	15	3.1	270	6.0	0.43	1, 2
Vähä Valkjärvi	61.185	25.095	126	2.3	3	4.9	-0.046	36	5.2	455	8.0	0.3	1, 2, 5
Alinen Mustajärvi	61.208	25.114	129	0.7	7	5.0	0.005	156	8.9	595	15.0	1.2	1
Suolikas	60.329	24.573	77	28.9	10	5.0	-0.01	80	9.3	550	7.0	0.39	1, 4
Pieni Valkojärvi	60.127	23.349	66	1.1	4	5.1	-0.008	100	10.6	690	18.0	0.47	
Ruuhijärvi	60.314	24.566	73	29	19	5.2	-0.001	55	7.7	480	6.6	0.51	1
Valkealampi	60.324	24.575	84	2.5	> 5	5.3	-0.004	25	4.5	570	8.2	0.28	
Pieni Lehmälampi	60.346	24.593	89	1.9	8	5.3	-0.006	20	5.7	274	8.0	0.72	1, 2, 4, 5
Iso Valkjärvi	61.19	25.108	126	4.2	8	5.3	0.003	47	5.6	410	11.0	0.7	1
Valkea-Kotinen	61.242	25.063	156	3.4	6	5.3	0.01	185	13.8	476	20.0	2.3	1
Hauklampi	60.317	24.609	76	2.5	8	5.4	-0.004	15	4.6	312	6.0	0.72	1, 2, 5
Orajärvi	60.307	25.588	87	22	3	5.4	0.005	20	4.3	300	9.0	0.6	1, 2, 5
Horkkajärvi	61.214	25.159	143	1.1	13	5.4	0.032	390	18.2	714	21.0	3.7	1
Yli-Falkkilanjärvi	60.144	23.453	63	2.4	> 5	5.5	0.018	50	8.5	580	5.5	0.9	
Pitkäjärvi	60.129	23.305	47	37.1	14	5.5	0.014	50	8.2	540	6.0	0.73	1, 4
Pitkälampi	60.217	23.606	80	15.1	10	5.6	0.013	40	8.4	530	6.8	0.69	
Horsmajärvi	60.153	23.467	56	42.8	18	5.7	0.023	35	6.6	490	6.5	0.91	
Kattilajärvi	60.302	24.614	62	33.9	9	5.9	0.04	40	6.9	490	5.5	0.85	1, 3, 5
Taka-Haukilampi	60.218	23.598	50	4.4	> 5	5.9	0.051	65	9.7	640	8.9	0.98	
Rahtijärvi	61.225	25.145	131	13.2	12	6.0	0.12	257	12.0	562	17.0	4.2	1
Viisjön	59.963	23.315	16	28.6	13	6.4	0.068	30	6.8	330	7.0	1.6	1, 3, 5
Valkea Mustajärvi	61.22	25.117	135	13.9	11	6.4	0.075	40	5.3	385	7.0	2.5	1

Acidification history for 16 of the lakes is given as records of fish responses: (1) known acidification history, (2) perch reproduction failures and consequent population and growth responses, (3) roach population responses, (4) roach extinct, (5) growth and population responses recovered

Tammi et al., 2004) and growth responses of perch (Nyberg et al., 2010; Raitaniemi et al., 1988; Rask et al., 2014a). For ten lakes, perch mercury concentrations were also available from the years of strong acidification in 1980s and 1990s (Metsälä & Rask, 1989). Four of these lakes suffered from severe acidification in those years, including perch population and growth responses (Raitaniemi et al., 1988; Rask et al., 2001), whereas the rest of the lakes acidified only slightly with no perch population responses (Metsälä & Rask, 1989).

2.2 Sampling of Fish

Fish for mercury analyses were caught during 2009–2014 with wire traps, gillnets, and hook and line (11 to 56 perch per lake, 664 in total, Table 2). Total length (mm) and weight (g) were measured for each fish. Samples for mercury analyses were taken

from dorsal white muscle and frozen ($-20\text{ }^{\circ}\text{C}$) until analysed. Opercular bones of all fishes were taken for age determination. As fish growth rate is known to affect mercury bioaccumulation (Göthberg, 1983; Rask et al., 1996; Verta, 1990b), a simple proxy of perch growth for each lake was calculated by dividing the mean length of sampled perch by their mean age (mean annual length increment). For the ten lakes with perch mercury data from two periods, 1980–1990s and 2010s, the Hg concentrations in the 2010s were related to the changes in the mean annual length increment of perch.

2.3 Mercury Analyses

THg concentrations of the perch muscle samples were determined in the Department of Environmental Sciences, University of Helsinki, using a Milestone

Table 2 Lake-specific mean total length, weight, age, growth and measured THg values of perch in the 24 lakes of the study

Lake	Length (cm)	Weight (g)	Age (yr)	Growth (cm yr ⁻¹)	THg measured ($\mu\text{g g}^{-1}$)		THg ($\mu\text{g g}^{-1}$) 15 cm stand	N of perch	N of fish species
					average	range			
Pöksynhaara	16.3	47.6	8.3	2.0	0.91	0.36–1.63	0.88	31	2
Teinipussi	14.6	34.9	6.0	2.5	0.52	0.10–1.13	0.54	33	2
Valkojärvi	19.2	94.6	6.4	3.0	1.23	0.54–2.65	1.07	35	1
Vähä Valkjärvi	17.4	42.7	9.0	1.9	0.85	0.57–1.17	0.73	11	1
Alinen Mustajärvi	12.3	18.5	3.0	4.1	0.21	0.12–0.27	0.19	12	2
Suolikas	17.1	66.1	6.9	2.5	0.9	0.36–2.50	0.76	56	3
Pieni Valkojärvi	19.4	84.5	8.3	2.3	0.67	0.13–1.76	0.74	41	1
Ruuhijärvi	17.2	57.0	5.6	3.1	0.42	0.20–0.77	0.36	36	3
Valkealampi	17.7	63.6	5.3	3.3	0.86	0.23–1.71	0.66	31	1
Pieni Lehmälampi	17.8	58.8	6.0	3.0	0.81	0.27–1.39	0.58	12	1
Iso Valkjärvi	14.4	32.2	4.0	3.6	0.18	0.12–0.39	0.18	12	2
Valkea-Kotinen	14.6	32.7	4.0	3.7	0.31	0.16–0.61	0.28	12	2
Hauklampi	22.1	123.0	8.0	2.8	0.86	0.48–1.55	0.74	12	1
Orajärvi	15.7	35.8	6.0	2.6	0.71	0.43–0.87	0.66	10	1
Horkkajärvi	12.4	20.9	9.0	1.4	0.56	0.23–0.80	0.76	12	1
Yli-Falkkilanjärvi	17.3	58.3	5.4	3.2	0.81	0.36–1.41	0.73	32	2
Pitkäjärvi	18.3	75.5	5.9	3.1	0.88	0.33–1.78	0.69	47	3
Pitkälampi	20.8	113.4	5.3	4.0	0.47	0.18–1.15	0.21	50	3
Horsmajärvi	17.4	77.5	4.7	3.7	0.32	0.14–0.84	0.26	45	4
Kattilajärvi	14.5	31.9	4.4	3.3	0.33	0.06–0.76	0.35	30	5
Taka-Haukilampi	15.9	54.2	5.9	2.7	0.51	0.16–1.39	0.47	33	5
Rahtijärvi	13.0	29.9	4.0	3.3	0.34	0.19–0.71	0.35	12	6
Vitsjön	20.5	116.6	6.7	3.1	0.68	0.15–2.01	0.38	45	4
Valkea Mustajärvi	15.6	50.5	3.0	5.2	0.21	0.15–0.25	0.20	12	2

Length corrected (to 15 cm) THg values, number of sampled perch and number of fish species per lake are also given

DMA-80 Direct Mercury Analyzer (Milestone GmbH, Germany) and results given as THg (mg kg^{-1}) in wet weight of fish (ww). Certified reference materials, DORM-4 fish protein (National Research Council, Canada, mercury concentration of 0.410 mg kg^{-1}) or cod muscle, CRM 422 (EC Community Bureau of Reference, mercury concentration of 0.559 mg kg^{-1}), were used for quality assurance of the analyses at the beginning and end of each run. Two replicate subsamples (ww 0.1–0.25 g) from each fish sample were analysed, and the mean concentrations of duplicates were used in subsequent data analyses.

For comparisons of lakes in 2009–2014, the measured THg concentrations of individual perch ($n > 30$) were standardized to 15 cm total length using lake-specific linear regression models. For lakes with a smaller fish sample ($n \leq 30$), a simple linear relationship between mercury concentration and fish length was assumed. For 10 lakes, historical mercury data during acidification had been measured with a Perkin Elmer Coleman MAS-50 analyzer (Metsälä & Rask, 1989). For quality control, see Verta (1990a) and Rask et al. (2007).

The effects of time period (1983–1994 vs. 2013–2014), lake and perch length (covariate) and their interactions on THg concentration was tested with ANCOVA. To normalize variances, Hg concentration and perch length were ln-transformed and results presented as geometric mean values. Tukey–Kramer’s test was used for pairwise comparisons. Results were considered significant, when $p < 0.05$.

3 Results

3.1 Perch THg Content and Correlations with Environmental and Biotic Variables During the 2010s

The measured average concentration of THg in perch from 24 lakes varied between 0.18 and 1.23 mg kg^{-1} (ww) (Table 2). Individual perch THg ranged from 0.06 to $2.65 \text{ mg Hg kg}^{-1}$ and concentrations $> 1 \text{ mg Hg kg}^{-1}$ were detected from 14 lakes. The highest THg concentrations of $> 2 \text{ mg kg}^{-1}$ were measured from four large (total length 31–36 cm) and old (10–11 years) individuals in three different lakes (Table 2).

Relating the length-corrected average THg concentrations of perch to lake water quality parameters resulted in significant correlation with pH (Fig. 1a) and alkalinity, but not with water colour, DOC (Fig. 1b), total nutrients or calcium (Table 3). THg had a significant inverse relation to the average annual length increment of perch ($r = 0.7$, $p < 0.001$, Fig. 1c).

Among the 16 lakes with known fish population responses to acidification, the highest perch THg concentrations were recorded in the lakes that had suffered from severe fish population damage in the 1980s, including reproduction failures and changes in population structure and growth of perch and/or disappearance of roach (*Rutilus rutilus* L.). The present data contained seven such lakes with mean perch THg concentration $0.71\text{--}1.07 \text{ mg kg}^{-1}$. In the nine lakes of known fish population status without acidification-induced perch population responses in the 1980s, the mean THg concentration was significantly lower, between 0.21 and 0.68 mg kg^{-1} ($t = -6.92$, $df = 14$, $p < 0.001$, Fig. 1d).

3.2 Perch THg Content During Acidification and Recovery

A comparison of perch THg concentrations in the 2010s with those measured in ten lakes in the 1980–1990s (Table 4) indicated increased levels in seven out of ten examined lakes. The four clearest lakes Hauklampi, Pieni Lehmälampi, Orajärvi and Vähä Valkjärvi (water colour $< 50 \text{ mg Pt L}^{-1}$) showed the highest increase in perch THg, up to $> 3\times$ from the 1980–1990s to the 2010s (Fig. 2). These four lakes suffered from severe acidification-induced perch population crash in the 1980s (Fig. 3) but have since recovered with a pH increase of ca. 0.5 units, whereas in three humic lakes, the alkalinity even decreased due to increased organic acidity in the course of browning of the lakes (Table 5). In five lakes, the increase in perch mercury was statistically significant (Tukey–Kramer’s test, Table 6), and only one of them (Valkea-Kotinen) was humic (water colour $> 50 \text{ mg Pt L}^{-1}$). Clear water Lake Iso Valkjärvi was the only lake with a statistically significant decrease in perch THg.

There was a significant negative correlation between the perch THg concentrations in the 2010s and the change in average annual length increment

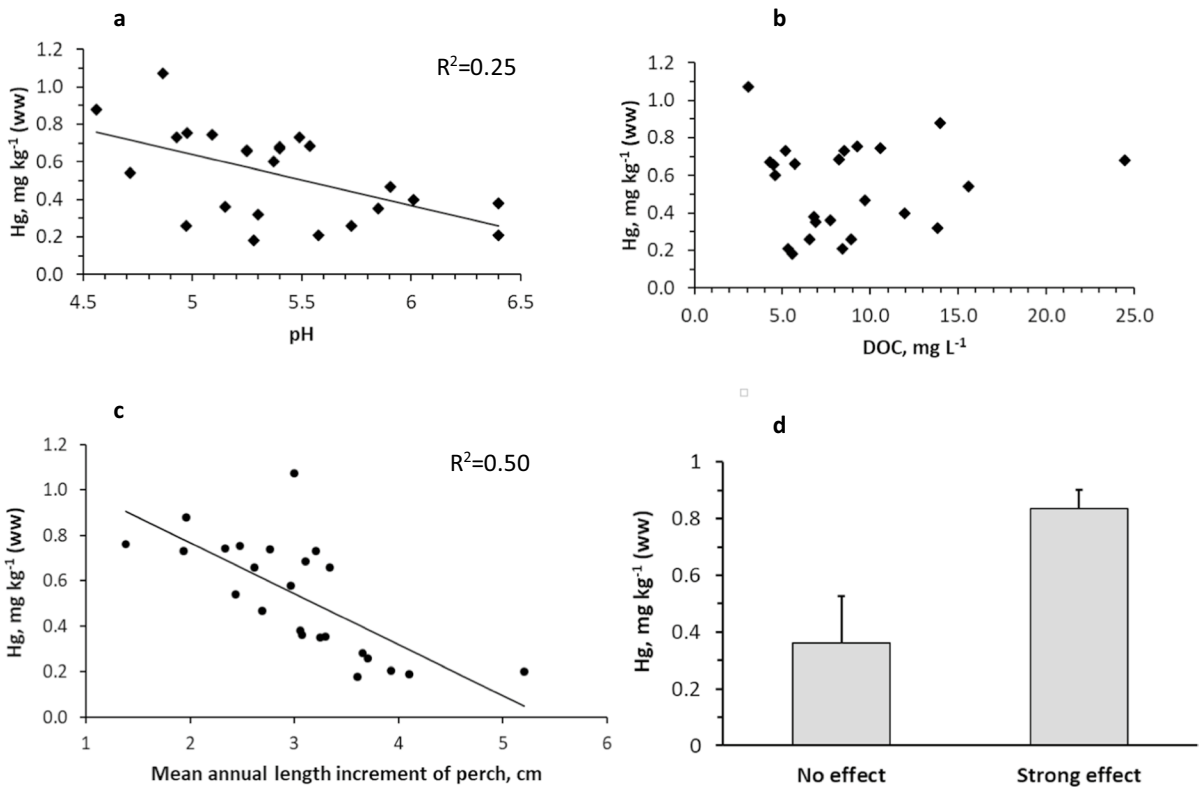


Fig. 1 The relation of 15 cm length-standardized THg concentration of perch to **a** lake pH, **b** DOC and **c** mean annual length increment of the fish in 24 lakes. **d** The perch THg (*mean* ± *SD*, 15 cm length-standardized) in lakes with known

history of acidification-induced fish responses. No effect=no perch population responses to acidification recorded (*n*=9). Strong effect=reproduction failures and changes in population structure and growth of perch or disappearance of roach (*n*=7)

Table 3 Correlations (*r*) between length-corrected THg values of perch and water quality parameters in the 24 study lakes

	Perch THg	pH	Alk	Colour	DOC	Tot. N	Tot. P	Ca
Perch THg	1.00							
pH	-0.52	1.00						
Alk	-0.50	0.88	1.00					
Colour	-0.01	-0.11	0.25	1.00				
DOC	0.01	-0.20	0.09	0.87	1.00			
Tot. N	0.06	-0.30	-0.07	0.60	0.79	1.00		
Tot. P	-0.11	-0.19	0.10	0.83	0.74	0.51	1.00	
Ca	-0.30	0.51	0.76	0.73	0.47	0.10	0.55	1.00

Significant correlations in bold

between the two sampling periods, 1980–1990s and 2010s (*r*=0.7, *p*<0.05, Fig. 4). In those lakes, where the annual length increment had decreased, THg was 0.3–0.8 mg kg⁻¹, whereas THg was below 0.3 mg kg⁻¹ in the lakes where the annual length increment had increased.

4 Discussion

The present study clearly demonstrated high perch mercury concentrations in poorly buffered headwater lakes in southern Finland. The concentrations recorded in the present study were 4–5 fold higher than in eight eutrophicated lakes in the same region

Table 4 ANCOVA results to test the effects of time period (1983–1994 vs. 2013–2014), lake and perch length (covariate) and their interactions on perch THg-concentration

Effect	No. df	Den df	F Value	$p > F$
Lake	9	182	3.32	<0.001
Time period	1	182	61.89	<0.001
Perch length	1	182	23.21	<0.001
Lake x Time period	9	182	18.09	<0.001
Perch length x Lake	9	182	4.31	<0.001

(Marttila & Roikonen, 2016) with known fish status (Olin et al., 2006). Furthermore, in the present study, the perch mercury concentrations exceeded the EQS (Ecological Quality Standard) limit of the European Union (EU) Water Framework Directive ($0.20\text{--}0.25 \mu\text{g g}^{-1}$ (ww) in Finland depending on the water colour) in 21 out of 24 lakes (88%) and the upper limit for human consumption of fish in EU ($0.5 \mu\text{g g}^{-1}$) in 15 lakes (63%). Perch THg content was negatively correlated with pH, whereas there was no correlation with DOC. In fact, growth rate seems to be a more important determinant of THg content in perch than any single environmental variable. This highlights the importance of understanding fish life-history traits and population dynamics when explaining changes in THg content.

4.1 Perch THg Is Mainly Related to pH and Growth Rate

Increased concentrations of mercury in water (Braaten et al., 2018; Watras et al., 1995), sediments (Kainz & Lucotte, 2006; Mannio, 2001) and fish (Braaten et al.,

2020; Porvari, 1998) have been linked to amount of dissolved organic matter in lakes. Consequently, the recent rise of DOC concentrations of lakes has been considered to be the major reason for increased mercury concentrations in the fish of formerly acidified lakes (Gandhi et al., 2014; Hongve et al., 2012; Wyn et al., 2010). In the present study, this is not the case as the increases in perch mercury were highest in the clearest lakes that also remained clear ($\text{DOC} < 5 \text{ mg L}^{-1}$) from the 1980s to the 2010s. The lack of significant correlations between perch THg and DOC was not surprising in the acid-sensitive headwaters of the present study, whereas significant negative correlations between lake pH or alkalinity and fish mercury concentrations have been reported in many related studies (Björklund et al., 1984; Greenfield et al., 2001; Verta et al., 1990).

One possible explanation for increased perch THg might have been enhanced bioavailability of mercury through increased methylation by sulphate-reducing bacteria (SRB) in anoxic conditions (Eckley et al., 2005; Ullrich et al., 2001). The importance of oxygen stratification to MeHg dynamics and perch mercury concentrations was shown in a lake-scale thermocline manipulation experiment during 2004–2007 (Arvola et al., 2017; Forsius et al., 2010) where deepening of the thermocline and consequent decrease in the area of oxycline of a small lake resulted in a decrease of epilimnetic MeHg concentration in the water (Verta et al., 2010), followed by a decrease of THg in small perch (Rask et al., 2010). However, as two of the four lakes with highest perch mercury increase between the 1980s and 2010s (Orajärvi and Vähä Valkjärvi) are shallow (maximum depth 4.5 and 3 m, respectively) and lack an anoxic hypolimnion,

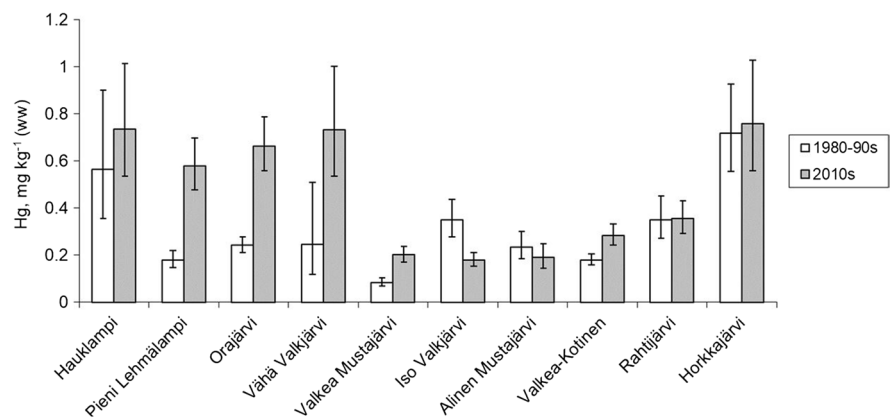
Fig. 2 Length-standardized (15 cm) THg concentrations (geometric mean \pm 95% c.l.) of perch in ten lakes in the 1980–1990s and in the 2010s. Lakes listed along increasing water colour measured in the 2010s (Table 5)

Fig. 3 a Length frequency distribution of perch in L. Orajärvi during 1985–2007 derived from standard gill-net catches. Black symbols refer to the years of highest acidification impact on the populations, open symbols to the years of recovery. **b** Back-calculated growth of the year-classes 1964–2006 for perch in L. Orajärvi ($n = 641$). The earliest year-classes 1964–1968, indicated with solid grey line, were born before the highest acidification impact. The year classes 1972–1990 with broken line are from the years of highest impact on perch population, and those from 1991 and onwards with solid black line are from years of recovery. Redrawn from Rask et al. (2014a)

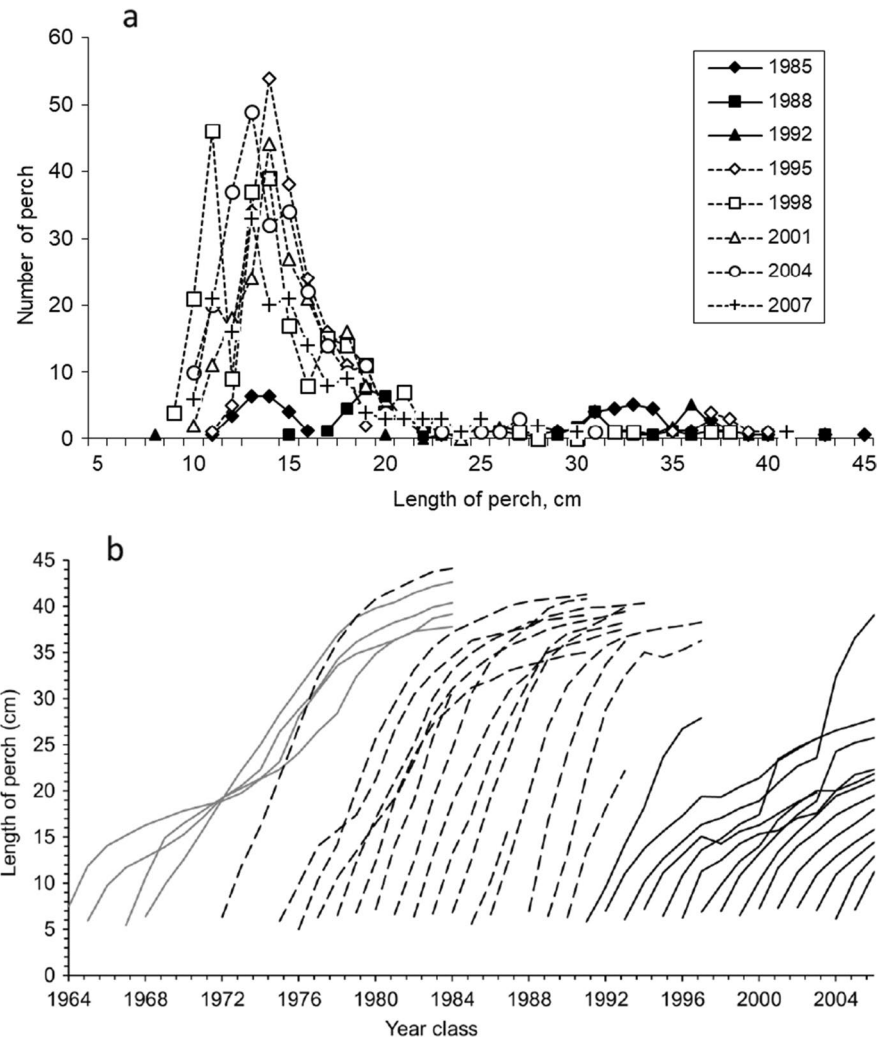


Table 5 Water quality in ten lakes sampled for perch mercury in two time periods, 1980-90 s and 2010s

Lake	pH		Alkalinity ($\mu\text{eq L}^{-1}$)		Colour (mg Pt L^{-1})		DOC (mg L^{-1})		Ptot ($\mu\text{g L}^{-1}$)		N of perch	
	1980s	2010s	1980s	2010s	1980s	2010s	1980s	2010s	1980s	2010s	1980s	2010s
Vähä Valkjärvi	4.47	4.93	-46	-9	5	36	4.3	5.2	8.0	8.0	7	11
Pieni Lehmälampi	4.67	5.25	-16	-6	15	20	4.6	5.7	7.0	8.0	10	12
Hauklampi	4.72	5.37	-11	-4	12.5	15	4.5	4.6	9.0	6.0	5	12
Orajärvi	4.9	5.4	-30	5	5	20	1.5	4.3	6.0	9.0	25	10
Iso Valkjärvi	4.98	5.28	2	3	77	47	6.5	5.6	18.0	11.0	6	12
Valkea-Kotinen	5.3	5.3	5	10	122	185	11.3	13.8	20.0	20.0	20	12
Alinen Mustajärvi	5.45	4.97	20	5	90	156	6.9	8.9	22.0	15.0	5	12
Horkkajärvi	5.54	5.4	58	32	290	390	13.0	18.2	18.0	21.0	5	12
Rahtijärvi	5.99	6.01	120	104	216	257	10.7	12.0	26.0	17.0	5	12
Valkea Mustajärvi	6.44	6.4	50	75	40	40	5.3	5.3	10.0	7.0	7	12

Number of fish samples also given

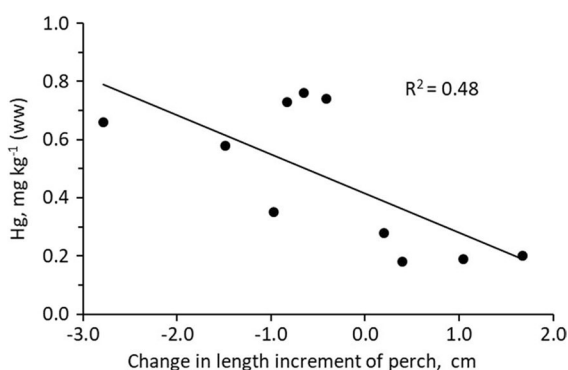
Table 6 Results of Tukey–Kramer’s test on within-lake differences in perch mercury concentrations between the two time periods

Lake	Year 1	Year 2	Estimate	SE	DF	t Value	Pr> t
Hauklampi	1985	2013	0.264	0.159	182	1.65	0.100
Pieni Lehmälampi	1986	2013	1.165	0.119	182	9.82	< 0.001
Orajärvi	1994	2014	1.004	0.111	182	9.07	< 0.001
Vähä Valkjärvi	1986	2013	1.098	0.260	182	4.22	< 0.001
Valkea Mustajärvi	1983	2013	0.881	0.134	182	6.6	< 0.001
Iso Valkjärvi	1983	2013	-0.667	0.143	182	-4.67	< 0.001
Alinen Mustajärvi	1983	2013	-0.210	0.190	182	-1.1	0.271
Valkea Kotinen	1992	2013	0.458	0.103	182	4.46	< 0.001
Rahtijärvi	1983	2013	0.010	0.178	182	0.06	0.954
Horkkajärvi	1983	2013	0.057	0.174	182	0.33	0.744

Significant differences in bold

the occurrence and role of SRB in MeHg dynamics is probably marginal in these two lakes. Further, reduced sulphate deposition, which has resulted in decreases of lake SO_4 concentrations (Nyberg et al., 2010), may have limited the activity of SRB in lakes (Braaten et al., 2020). Thus, changes in in-lake methylation of mercury by SRB do not explain the increases of perch mercury concentrations in our lakes.

The significant inverse relation between the length increment and mercury concentration of perch in the present study emphasizes the importance of growth patterns of fish. Fast growing individuals are known to exhibit growth dilution of Hg in fish, and thus effective fishing has been suggested as a method to decrease the Hg levels in fish and the fish consumption risk of humans (Göthberg, 1983; Verta, 1990b).

**Fig. 4** The relation between perch THg in the 2010s (15 cm length-standardized) and the change of average annual length increment of perch from the 1980–1990s to the 2010s in ten lakes

4.2 Perch THg Content Increased During the Recovery of Strongly Acidified Lakes

In some of our strongly acidified lakes, the growth of perch was very fast in the 1980s due to very low population densities caused by repeated reproduction failures and relaxing of resource competition (Lappalainen et al., 1988; Raitaniemi et al., 1988). This was particularly evident in Lake Orajärvi with 3-year-old perch of >20 cm and 5-year-olds close to 30 cm in total length (Nyberg et al., 1995; Rask et al., 2014a).

Consequently, an apparent reason for relatively low perch THg in those years was the growth dilution, i.e. muscle grew faster than the fish accumulated mercury (e.g. Göthberg, 1983; Keva et al., 2017). The increased availability of macroinvertebrate food apparently enabled the increase in perch growth (Nyberg et al., 2010) and may also have contributed to the growth dilution of mercury. This is because littoral macroinvertebrates have been shown to be less efficient transporters of Hg up food chains than crustacean zooplankton (Kahilainen et al., 2016; Karimi et al., 2016). From the beginning of the 1990s, the reproduction of perch recovered and resulted in high population density followed by a steep decrease in perch growth (Nyberg et al., 1995; Rask et al., 2014a). This explained the increased perch THg concentration, possibly also contributed to by increased use of zooplankton food by perch of new abundant year-classes after a sharp decrease of macroinvertebrates (Nyberg et al., 2010). Thus, an “inverse growth dilution”, i.e. mercury condensation via starvation, took place (Keva et al., 2017). This also happened in two other formerly strongly acidified lakes (Lake Pieni Lehmälampi, Lappalainen et al., 1988 and Lake Vähä

Valkjärvi, Rask et al., 2001) although the decrease of perch growth was not as steep as in Lake Orajärvi. In Lake Hauklampi (Raitaniemi et al., 1988); there was almost no difference in perch growth between the two sampling periods and the difference in mercury concentrations remained nonsignificant. In L. Valkea Mustajärvi a significant increase in the perch THg took place, but this cannot be explained neither by water quality changes nor by growth responses.

In Lake Valkea-Kotinen, the only humic lake with significant increase in perch THg, the increase in organic carbon load (Vuorenmaa et al., 2014) may have contributed to the Hg load and the perch THg. In this lake, being a monitoring site in the International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems (UNECE ICP IM), the increased DOC was connected to the decrease of general productivity of the lake during a 20-year monitoring period (Arvola et al., 2014), but the growth responses of perch were slight in comparison to the strongly acidified lakes of the present study (Rask et al., 2014b). In L. Horkkajärvi, the most humic of the ten lakes, the slight increase in perch THg may be connected to the organic load but be as well due to natural variation.

Lake Iso Valkjärvi was the only lake with a statistically significant decrease in perch THg concentrations between the two sampling periods. However, there was a liming experiment in the lake in the early 1990s (Rask et al., 1996) and an experimental size-selective removal of perch in 2008–2011 (Olin et al., 2017) that both have probably affected the Hg dynamics in the lake. Further, a sudden collapse of most of the perch population in the reference half of the lake took place during the liming experiment in autumn 1992 (Rask et al., 1996) resulting in a five-fold increase in length growth of remaining perch and subsequent growth dilution of THg to half of that before the fish kill (Rask et al., 2007). The fish kill affected also lower trophic levels of the lake which was seen in increased abundance of macroinvertebrates like phantom midge larvae (*Chaoborus* sp.) and water boatmen (*Corixidae* sp.), but in decreased abundance of cladoceran zooplankton, apparently due to increased predation by macroinvertebrates (Rask et al., 1996). The increased availability of benthic macroinvertebrates apparently enabled the increase in perch growth and may have contributed to the growth dilution of perch THg also in this case. The

size-selective removal fishing in 2008–2011 may also have affected the decrease of perch THg in Iso Valkjärvi as it temporarily decreased the biomass and increased the growth-rate of perch, changed the population structure towards smaller and younger individuals (Olin et al., 2017), and increased the proportion of macroinvertebrates in the diet of perch (Nurminen et al., 2018).

Climate change has resulted in a 0.4 °C increase in mean air temperature per decade and a 17 day addition per decade in the duration of ice free period of lakes in the study region (Jylhä et al., 2014). This should have an overall positive growth and reproduction response for perch and should lower the mercury content via growth dilution. However, it seems that intra-specific density dependent processes and lowered water transparency of lakes due to browning are more important determinants of growth than warming (Rask et al., 2014b). Increasing precipitation and air temperature, followed by hydrological changes with increasing runoff and elevated leaching of substances from catchments to lakes, browning of lake waters and effects on lake temperature, stratification and oxygen dynamics (Couture et al., 2015; Vuorenmaa et al., 2014), will continuously affect the mercury dynamics in catchments and lakes (Ahonen et al., 2018). Time series analyses have shown that responses in fish Hg over decades are often non-linear in relation to linear decreases of mercury emissions and deposition (Gandhi et al., 2014; Braaten et al., 2019; Wang et al., 2019; Watras et al., 2019, 2020). This has been attributed to legacy mercury and changing biogeochemical processes. Climate change has become the most prevalent contributor to this divergence (Wang et al., 2019), and regular oscillation of water levels in lakes connected to large-scale atmospheric circulation patterns are also important (Watras et al., 2020). Taking into account all this, changes in the bioaccumulation of Hg and wide variations of mercury concentrations in biota of aquatic food webs up to fish are expected to continue in the future.

To conclude, the results of this study clearly indicated the occurrence of high perch mercury concentrations in poorly buffered boreal headwater lakes. The negative correlation of fish THg with lake pH and growth of perch was as predicted, but the lack of correlation with DOC was not. Given the complex effects of climate change processes and decreased atmospheric SO₄ deposition on mercury dynamics

in lakes and their catchments, we believe that chemical recovery of the lakes from acidification and consequent changes in the food web, including reproduction of perch, increase in population density and drastic decreases in growth of perch, were the main mechanisms leading to increases in perch mercury in formerly strongly acidified lakes of the present study, counter to our original prediction. Our results suggest that growth rate and population density of fish should always be taken account of when assessing changes in mercury content in muscle.

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

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