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Environmental and biological factors are joint drivers of mercury biomagnification in subarctic lake food webs along a climate and productivity gradient

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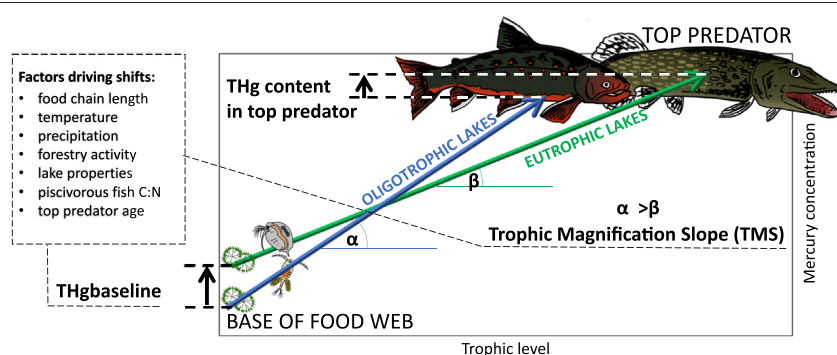
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HIGHLIGHTS

- Biomagnification of THg was evident in the food webs of 19 subarctic lakes.
- Trophic magnification slope (TMS) of THg was negatively related to food chain length.
- TMS and THg baseline were related to food chain length, environment, and predators.
- Forestry related eutrophication elevated THg baseline and lowered TMS.
- THg content of the top predator (northern pike) increased with lake productivity.

GRAPHICAL ABSTRACT



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ABSTRACT

Subarctic lakes are getting warmer and more productive due to the joint effects of climate change and intensive land-use practices (e.g. forest clear-cutting and peatland ditching), processes that potentially increase leaching of peat- and soil-stored mercury into lake ecosystems. We sampled biotic communities from primary producers (algae) to top consumers (piscivorous fish), in 19 subarctic lakes situated on a latitudinal (69.0–66.5° N), climatic (+3.2 °C temperature and +30% precipitation from north to south) and catchment land-use (pristine to intensive forestry areas) gradient. We first tested how the joint effects of climate and productivity influence mercury biomagnification in food webs focusing on the trophic magnification slope (TMS) and mercury baseline (THg baseline) level, both derived from linear regression between total mercury ($\log_{10}\text{THg}$) and organism trophic level (TL). We examined a suite of environmental and biotic variables thought to explain THg baseline and TMS with stepwise generalized multiple regression models. Finally, we assessed how climate and lake productivity affect the THg content of top predators in subarctic lakes. We found biomagnification of mercury in all studied lakes, but with variable TMS and THg baseline values. In stepwise multiple regression models, TMS was best explained by negative relationships with food chain length, climate-productivity gradient, catchment properties, and elemental C:N ratio of the top predator (full model $R^2 = 0.90$, $p < 0.001$). The model examining variation in THg baseline values included the same variables with positive relationships ($R^2 = 0.69$, $p = 0.014$). Mass-

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standardized THg content of a common top predator (1 kg northern pike, *Esox lucius*) increased towards warmer and more productive lakes. Results indicate that increasing eutrophication via forestry-related land-use activities increase the THg levels at the base of the food web and in top predators, suggesting that the sources of nutrients and mercury should be considered in future bioaccumulation and biomagnification studies.

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1. Introduction

Mercury (Hg) is a toxic metal monitored worldwide due to the fact it bioaccumulates in organisms and biomagnifies in food webs. Mercury occurs naturally in the environment in low concentrations, however, increased concentrations of environmental mercury since the Industrial Revolution has negatively impacted wildlife and human health (Streets et al., 2011). Most anthropogenic emissions consist of elemental mercury, which can be transported long distances and deposited even in the most sparsely populated Arctic regions (Pacyna et al., 2010).

While Arctic regions are typically considered the least impacted by pollution, many regions have relatively high concentrations of mercury in the environment and biota (Macdonald et al., 2005; Schindler and Smol, 2006; AMAP, 2011; Obrist et al., 2017). The majority of this mercury is transported via long-distance air masses from industrial and populated areas, condensed and deposited (by both wet and dry deposition) in cold Arctic regions in ice, snow, soil and water (Barrie et al., 1992; Pacyna et al., 2010; Streets et al., 2011; Amos et al., 2013). Mercury leaching to lakes is continuously observed to occur during spring thawing of snow and ice (Douglas and Blum, 2019), but far less is known on how land-use activities may contribute to the mercury leaching.

Arctic areas are warming faster than the global average (IPCC, 2014), with annual air temperature and precipitation predicted to rise by 3–8 °C and 30–60% respectively by the end of this century (IPCC, 2014). In response to this, faster thawing of ice and snow has been detected in Arctic areas (Sharma et al., 2016). These changes in the environment are causing mercury storage in permafrost and frozen grounds to leach into lakes in spring and summer (Schuster et al., 2018). Early ice melt in spring exposes lakes to solar radiation enhancing productivity, while snowmelt and ground thawing elongate the growing season resulting in increased length of the growing season and support overall vegetation growth and a northward extension of the tree-line in subarctic regions (Serreze et al., 2000; AMAP, 2011; Stern et al., 2012). In addition, intensifying anthropogenic activities such as deforestation, ditching of peatlands, oil drilling, road construction and mining all increase leaching of mercury from the terrestrial to aquatic systems (Schindler and Smol, 2006; Huntington et al., 2007; Jussila et al., 2014; Cott et al., 2015). In combination, the joint effects of warming, increased intensity of precipitation events and land-use activities increase the flux of mercury to lakes from the surrounding environment (Schroeder and Munthe, 1998; Sanei et al., 2010; Hudelson et al., 2019). These processes expose soil, increasing the runoff of carbon, nitrogen and phosphorus, with dissolved organic carbon (DOC) acting as a carrier of mercury from the catchment to the aquatic environment (Watras et al., 1998; Kolka et al., 1999; Poste et al., 2019).

Once deposited in anoxic conditions, such as peatland or lake sediments, Hg can be methylated to organic methylmercury (MeHg) via bacterial activity (Ullrich et al., 2001; Bravo et al., 2017). Methylmercury is the most toxic form of Hg, it bioaccumulates in organisms and biomagnifies in food webs (Watras and Bloom, 1992; Watras et al., 1998). Primary producers including both pelagic phytoplankton and benthic periphyton uptake different forms of mercury from lake water via active and passive processes (e.g. Morel et al., 1998; Douglas et al., 2012). Therefore, the baseline level of THg (i.e. the amount of THg observed in primary producers, hereafter THg baseline) is determined by the THg level in the waterbody, to which primary producers are

exposed (van der Velden et al., 2013). From primary producers onwards Hg is transported through the food web via dietary sources (Hall et al., 1997). The proportion of MeHg increases from primary producers (c. 15% of THg) to primary consumers (c. 30%), and finally to adult fish muscle, where most of THg (>90%) is composed of MeHg (Watras and Bloom, 1992; Watras et al., 1998; Lescord et al., 2018). The rate of mercury transfer in food webs can be described with trophic magnification slope (TMS), i.e. a linear regression between \log_{10} THg and trophic level (TL) of organisms (Borgà et al., 2012; Lavoie et al., 2013). High TMS indicates great magnification, while low TMS designate poor magnification within the food web.

The trophic magnification slope can be affected by various environmental and biotic factors (Lavoie et al., 2013; Clayden et al., 2013; Lescord et al., 2015), but there is a general need for detailed studies along many environmental gradients to understand the role of different drivers. Lavoie et al. (2013) conducted a global study of mercury biomagnification in aquatic food webs and found TMS increasing towards higher latitudes, but no significant relationship with total phosphorus. However, other studies have reported that climatic conditions and nutrients may show positive, negative or no relationships with TMS (Gantner et al., 2010; Kidd et al., 2012; Clayden et al., 2013; Sumner et al., 2019). These contrasting results from large and smaller spatial scales suggest that the mechanisms affecting mercury biomagnification in freshwater food webs are complex and likely driven by multiple factors including both within lake and catchment processes. Here, lake size, depth and catchment to lake size ratio are likely important, as they may affect pelagic and benthic energy and Hg pathways as well as residence time and flux of Hg runoff (Lee and Iverfeldt, 1991; Hammerschmidt et al., 2006; Gantner et al., 2010; Kidd et al., 2012; Thomas et al., 2016). Land cover type (i.e. peatland, forest, barren), prevailing Hg deposition rate and the degree of methylation processing in soils, in addition to land-use activities, change the structure of soils exposing historical Hg from deeper layers to water and air interactions (Schindler and Smol, 2006; Jussila et al., 2014; Cott et al., 2015; Ahonen et al., 2018). Furthermore, the joint effects of latitude, lake size and productivity are connected to species richness and food chain length, which may affect THg biomagnification rate e.g. via a varying number of trophic links, trophic levels and energy pathways (Post et al., 2000; Hillebrand, 2004; Takimoto and Post, 2013). Long-lived predatory fish accumulate THg over time, their presence may steepen THg biomagnification slope of a whole food web (Gantner et al., 2010; Ahonen et al., 2018).

We used detailed spatial, environmental and biotic data to test the potential changes in the THg baseline levels and TMS in a subarctic watercourse spanning a pronounced climatic (+3.2 °C temperature and +30% precipitation) and productivity (+45 $\mu\text{g L}^{-1}$ of total phosphorus [TP]) gradient. The climatic gradient is primarily determined by latitude and elevation, whereas productivity is primarily related to land-use change, ranging from pristine headwater lakes to intensive forestry managed catchments in southern reaches (Hayden et al., 2017; Ahonen et al., 2018). Tributary lakes of this watercourse include oligotrophic, mesotrophic to eutrophic lakes with biotic communities ranging from cold to warm water adapted taxa (Hayden et al., 2017). This subarctic watercourse represents an excellent environment to study joint effects of climate and productivity on THg baseline and TMS in a food web. We tested three main predictions: With the ongoing climate change and intensification of land-use activities, the mercury cycle is

expected to change due to intensified mercury run off from soil and biomagnify in food web. Thus, we first predicted P1) that increased temperature and productivity would elevate the THg baseline via increasing forestry-related leaching of mercury and decrease TMS due to the increasing complexity of the food web (Hayden et al., 2019; Keva et al., 2021). Secondly, we predicted (P2) that “best fit” regression models of THg baseline and TMS would include temperature and productivity as well as food chain length and number of fish species in each lake. Finally, we predicted (P3) that, leaching of mercury with intensified forestry-related land-use would result in an increase of THg concentrations in top predators.

2. Material and methods

2.1. Study area and sampling

We sampled the food webs of nineteen tributary lakes in the subarctic Tornio-Muoniojoki watercourse located on a latitudinal climate-productivity gradient from 69.0°N to 66.5°N on the Swedish-Finnish border region in August–September 2009–2013 (Fig. 1). All study lakes are situated north of the Arctic circle and considered subarctic in the current study (AMAP, 1998). The studied lakes are numbered according to climate-productivity gradient (roughly following latitudinal north-south direction), where the northernmost, oligotrophic lakes (TP: $\leq 10 \mu\text{g L}^{-1}$) are located in the mountain birch forest area with low anthropogenic activity and limited land-use focusing mainly on reindeer herding and nature tourism. Mesotrophic lakes (TP: $10\text{--}30 \mu\text{g L}^{-1}$) are located in the increasingly forested area mostly below the northernmost distribution lines of Scots pine (*Pinus sylvestris*) and Norway spruce (*Picea abies*) facing an increased human activity,

including forestry (Table S1). Southernmost lakes are turbid, eutrophic systems (TP: $\geq 30 \mu\text{g L}^{-1}$) with high nutrient content, densest human populations and intensive land-use activity focusing on commercial forestry and related activities (clear-cutting, site preparation, peatland ditching and dense forest road network) (Jussila et al., 2014; Hayden et al., 2019). Ditching of peatland for forestry activities has been very intense in the southern range of the study region, where circa 50–100 cm deep ditches are dug with caterpillars and excavators leading to very high ditch density in the drainage area network (e.g. Nieminen et al., 2018). Such activities are the main sources of nutrients and carbon (e.g. Finér et al., 2021) as there are very few agricultural areas or sewage water leaching to lakes in the study area. Permafrost is absent from the catchment areas of all study lakes.

Fish were collected with series of eight gillnets of $30 \times 1.8 \text{ m}$ size and varying knot-to-knot mesh sizes (12, 15, 20, 25, 30, 35, 45 and 60 mm) and one Nordic multi-mesh gillnet of $30 \times 1.5 \text{ m}$ size (mesh sizes: 5–55 mm) with 12 equidistant panels of 2.5 m (Hayden et al., 2017). Due to the low density and gill net susceptibility of some predator fish species such as pike (*Esox lucius*) and burbot (*Lota lota*), these samples were supplemented by angling. Nets were set overnight (10–12 h) in each habitat (pelagic, littoral and profundal) of each lake, with a minimum of three nets per habitat in each lake. Sampling proceeded through 3–5 consecutive nights to get representative samples of the fish community. Fish were removed from nets, euthanized with a cerebral concussion and stored in ice for transport to the laboratory.

Biofilm was collected from littoral rocks and plants by scraping the green surface. Pelagic algae samples were restricted to the sampling of blue-green algal blooms in some of the lakes with a $50 \mu\text{m}$ mesh net using horizontal hauls. Due to sampling difficulties caused by weather conditions and late growing season, algae were collected only in nine

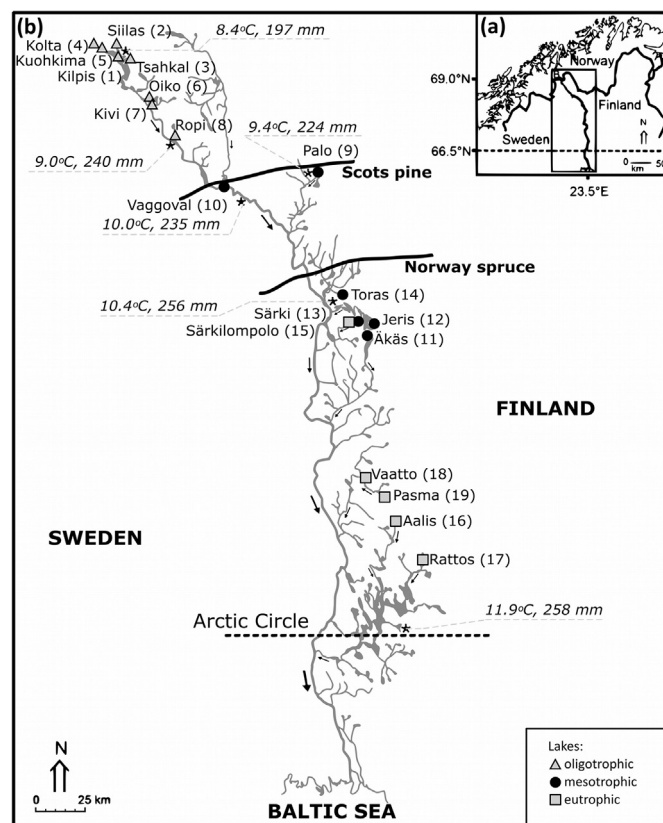


Fig. 1. Northern Fennoscandia indicating (a) the location of Tornio-Muonio watercourse on the border of Sweden and Finland (b). Studied tributary lakes are numbered (1–19) corresponding to lake numbers in Table 1. Constant lines indicate the occurrence of coniferous tree lines, asterisks indicate meteorological stations with values of mean temperature (°C) and precipitation (mm) of open water season (June–September) in 1981–2010 and arrows indicate flowing direction of the watercourse (modified from Hayden et al., 2017). Symbol legend in the lower right corner indicates lake trophic state.

out of nineteen lakes (Table S2). For lakes with missing data, average values of carbon and nitrogen stable isotopes and THg were calculated from the nearest similar lake i.e. for the oligotrophic lakes above Scots pine distribution line (lakes no 1–8), mesotrophic lakes below Scots pine distribution line (9–14) and eutrophic lakes near the Arctic circle (15–19) (Table S2, Table S4). Pelagic zooplankton were collected with a 50 µm mesh net using vertical hauls (max 0–20 m) from the deepest sampling point in each lake (Hayden et al., 2019). Benthic macroinvertebrates were collected from the lake shoreline (depth 0–0.5 m) using handpicking from stones and vegetation and kick-net sampling (500 µm mesh). In addition, we collected a benthic soft-sediment transect from littoral to deepest part of each lake (depths from 1, 2, 3, 5, 10, 15, 20, 30 and 40 m, if maximum depth allows) with Ekman grab (surface area 272 cm²) by using three replicates in each depth (Hayden et al., 2017). Samples were stored separately in clean 2 ml polypropylene tubes and transported to the laboratory for identification and sorting.

2.2. Environmental variables

The climate measurements (mean air temperature [°C] and precipitation [mm]) in open-water season (June–September) were obtained from long-term archives (1981–2010) of the Finnish Meteorological Institute and Klein Tank et al. (2002) (total of six meteorological stations in the study area Fig. 1). Lake habitat characteristics (compensation depth [m], mean depth [m], lake volume [m³] and area [m²]), altitude [m a.s.l.], total nitrogen [µg L⁻¹], and total phosphorus [µg L⁻¹] were measured on-site and catchment properties (forest percentage [%], tree volume in a forested area [m³ ha⁻¹], sparse vegetation [%], catchment area [km²], peatland area [km²], and ditch in catchment area [km km⁻²]) and catchment-lake area ratio were obtained from own sampling (lake morphology), Lapland Centre for Economic Development, Transport and Environment (nutrients), Finnish Environment Institute (catchment land-use) and Land Survey of Finland (lake altitude) (for details; Hayden et al., 2017, 2019; Ahonen et al., 2018). A principal component analysis (PCA) was conducted on environmental variables of all studied lakes (Tables S1 and S3, Fig. S1) to reduce the number of explanatory variables into the following three principal components (PC1–PC3). The first principal component (PC1) explained 54.8% of

variation in the lake environmental data set and spanned from north to south describing a climate-productivity gradient including altitude [m a.s.l.], air temperature [°C], precipitation [mm], forested area [%], tree volume in a forested area [m³ ha⁻¹], sparse vegetation [%], total phosphorus [µg L⁻¹], total nitrogen [µg L⁻¹], compensation depth in a lake [m] and ditch length [km km⁻²] at the end with lower importance (Fig. S1, Table S1, Table 1). PC2 explained 16.8% of the total variance in lake environmental data and was determined by catchment characteristic related variables (catchment area [km²], peatland area [km²] and the ratio of catchment and lake areas) (Fig. S1, Table S3, Table 1). The third principal component (PC3) explained 16.1% of the total variance and represented lake morphometry (lake area [km²], lake volume [m³] and lake mean depth [m]) (Fig. S1, Table S3, Table 1).

2.3. Fish

Nineteen fish species were sampled in the studied area including nine-spined stickleback (*Pungitius pungitius*), common sculpin (*Cottus gobio*), dace (*Leuciscus leuciscus*), Alpine bullhead (*Cottus poecilopus*), roach (*Rutilus rutilus*), grayling (*Thymallus thymallus*), ide (*Leuciscus idus*), minnow (*Phoxinus phoxinus*), brown trout (*Salmo trutta*), bleak (*Alburnus alburnus*), European whitefish (*Coregonus lavaretus*), smelt (*Osmerus eperlanus*), perch (*Perca fluviatilis*), ruffe (*Gymnocephalus cernua*), burbot, vendace (*Coregonus albula*), peled whitefish (*Coregonus peled*), Arctic charr (*Salvelinus alpinus*) and pike (Table S2). Whitefish occur in two morphs i.e. large sparsely rakered whitefish (LSR) and densely rakered whitefish (DR) in Lake Ropi (8) (Hayden et al., 2013). Four lakes (11, 13, 14, 15) contain whitefish × vendace hybrids, which have distinct diet and life-history traits. Both whitefish morphs and hybrids are ecologically divergent groups and have a significant role in energy and mercury flows in subarctic lake food webs potentially also affecting biomagnification (Thomas et al., 2016; Kahilainen et al., 2017). Salmonids dominate in the northernmost lakes of the study region, percids in the mesotrophic lakes and cyprinids in the eutrophic lakes (Hayden et al., 2017).

Each fish individual was identified to species (whitefish were identified to morph and hybrid level based on head morphology and a number of gill rakers in the first left gill arch (Kahilainen

Table 1

Biomagnification regression intercepts (±SE) i.e. THg baseline and slopes (±SE) i.e. TMS of log₁₀THg (ng g⁻¹ d.w.) versus trophic level for 19 lakes numbered by PC1 value in Tornio-Muoniojoki watercourse with adjusted coefficient of determination (adj. r²), probability value (p), trophic magnification factor (TMF) and THg level at the base of food web (basal THg) (ng g⁻¹ d.w.). The linear regressions were calculated from average values for each group (fish species, algae, zooplankton and littoral benthos). Number of fish species in each lake (n_{spc}), total number of samples (n_{sample}), average THg content of 1 kg pike (ng g⁻¹ d.w.) (Pike THg_{adj}) and variables selected by backward stepwise model for GLM (climate-productivity (PC1), catchment properties (PC2), lake morphometrics (PC3), food chain length (FCL), top predator age (P_{age}), top predator C:N ratio (P_{C:N}) and zooplankton C:N ratio (Z_{C:N})). For more detailed samples sizes in each group see Table S5 and for GLM details see Section 3.3. In pike column, NA indicates species is not present in lake or data is too limited to calculate value for 1 kg pike. Lakes trophy are marked with a grey triangle for oligotrophic lakes, black circles for mesotrophic lakes and grey square for eutrophic lakes corresponding to Fig. 1.

No	Lake name	THg baseline; Intercept ± SE	TMS; Slope ± SE	Adj. r ²	p	n _{spc}	n _{sample}	basal THg	TMF	Pike THg _{adj}	PC1	PC2	PC3	FCL	P _{age}	P _{C:N}	Z _{C:N}
1	Kilpis ▲	0.583 ± 0.22	0.602 ± 0.08	0.87	<0.001	8	277	3.8	4.0	490	-4.36	3.14	5.11	3.80	6.65	3.43	5.13
2	Siilas ▲	0.775 ± 0.17	0.633 ± 0.07	0.92	<0.001	6	70	6.0	4.3	NA	-4.16	-0.16	-0.95	2.83	4.17	3.40	6.34
3	Tsahkal ▲	0.959 ± 0.36	0.579 ± 0.08	0.79	0.017	4	164	9.1	3.8	NA	-3.96	-0.38	-0.45	3.18	6.83	3.26	6.58
4	Kolta ▲	0.509 ± 0.33	0.862 ± 0.14	0.86	0.007	6	112	3.2	7.3	670	-3.47	-0.79	-1.18	2.64	5.44	3.23	5.80
5	Kuohkima ▲	0.630 ± 0.14	0.627 ± 0.09	0.97	<0.001	6	152	4.3	4.2	1080	-3.26	-0.04	-1.60	3.51	6.50	3.18	5.35
6	Oiko ▲	1.050 ± 0.20	0.499 ± 0.11	0.88	<0.001	9	264	11.2	3.2	1925	-2.15	-0.94	-0.72	3.86	6.72	3.20	4.58
7	Kivi ▲	0.973 ± 0.17	0.670 ± 0.10	0.95	<0.001	9	271	9.4	4.7	1570	-2.15	0.45	-1.10	3.26	8.24	3.23	5.40
8	Ropi ▲	0.888 ± 0.16	0.533 ± 0.08	0.90	<0.001	11	308	7.7	3.4	1020	-1.63	0.12	-0.63	3.81	8.06	3.24	4.78
9	Palo ●	0.268 ± 0.20	0.814 ± 0.16	0.93	<0.001	8	260	1.9	6.5	555	-0.03	0.21	-1.01	3.11	4.47	3.21	4.59
10	Vaggoval ●	0.605 ± 0.19	0.655 ± 0.13	0.92	<0.001	7	188	4.0	4.5	1075	0.30	-1.57	-0.35	3.17	4.41	3.16	4.18
11	Äkäs ●	0.688 ± 0.25	0.477 ± 0.14	0.82	<0.001	8	328	4.9	3.0	705	1.32	-0.65	0.91	3.85	4.83	3.25	3.77
12	Jeris ●	0.873 ± 0.11	0.431 ± 0.06	0.96	<0.001	9	229	7.5	2.7	NA	1.73	-0.38	1.77	3.99	4.00	3.21	4.76
13	Särki ●	0.983 ± 0.12	0.310 ± 0.06	0.93	<0.001	6	279	9.6	2.0	NA	1.80	-1.32	0.77	4.41	3.00	3.23	4.85
14	Toras ●	0.876 ± 0.18	0.459 ± 0.08	0.87	<0.001	10	325	7.5	2.9	NA	2.03	0.16	0.04	3.92	5.67	3.21	5.57
15	Särkilompolo □	0.806 ± 0.26	0.451 ± 0.12	0.80	<0.001	8	295	6.4	2.8	1215	2.07	-1.20	0.25	4.23	5.29	3.19	4.51
16	Aalis □	0.932 ± 0.24	0.638 ± 0.13	0.85	<0.001	9	332	8.6	4.3	1485	3.77	-0.72	0.61	3.25	6.81	3.21	4.50
17	Rattos □	0.937 ± 0.19	0.508 ± 0.09	0.88	<0.001	8	321	8.6	3.2	1630	3.83	-0.97	0.46	3.86	6.54	3.22	4.70
18	Vaatto □	0.984 ± 0.24	0.553 ± 0.14	0.84	<0.001	8	289	9.6	3.6	2300	4.12	5.59	-2.51	3.80	7.46	3.20	4.57
19	Pasma □	1.096 ± 0.24	0.513 ± 0.12	0.80	<0.001	8	261	12.5	3.3	1875	4.21	-0.55	0.58	3.50	8.38	3.21	4.55

et al., 2011a, 2011b), and measured in total length (± 1 mm) and weight (± 0.1 g). For stable isotopes and total mercury analyses, a piece of white dorsal fish muscle was frozen at -20 °C, freeze-dried for 48 h at -50 °C and powdered with a glass rod. The age of top predatory fish was determined by inspection of clear and burned otoliths for Arctic charr and burbot, whereas ageing of pike was determined from cleithrum bone and scale impressions (Thomas et al., 2016; Ahonen et al., 2018).

2.4. Lower trophic levels

Algal and biofilm samples were viewed under a preparation microscope and any inorganic or organic non-plant material was removed. The remaining material was stored in 2 ml polypropylene tubes. Zooplankton composite samples containing rotifers, cladocerans, and copepods were concentrated on 50 μ m mesh and stored in 2 ml polypropylene tubes. Benthic macroinvertebrates were sorted to genus or family level and stored in 2 ml polypropylene tubes directly, except for Mollusca that was first dissected from their shells to avoid the effect of carbonate on stable isotope analyses. Due to the small amount of biomass of benthic and pelagic primary producers available in each lake, biofilm and pelagic phytoplankton were combined for subsequent statistical analyses, hereafter referred to as algae. All samples were first frozen to -20 °C followed by freeze-drying (-50 °C for 48 h). Freeze-dried samples were homogenized to a fine powder using a glass rod and maintained at -20 °C until further analyses.

2.5. Stable isotopes

Subsamples of fish, invertebrates and primary producers were weighed (1 ± 0.1 mg) into tin cups for analyses of elemental carbon and nitrogen ratio (C:N ratio) and nitrogen stable isotope ratio. Elemental composition and $\delta^{15}\text{N}$ were determined using an elemental analyzer coupled to a continuous-flow isotope ratio mass spectrometer. Laboratory specific standards were calibrated against atmospheric nitrogen used for $\delta^{15}\text{N}$. The analytical error was 0.2‰ for the $\delta^{15}\text{N}$. The trophic level of each fish was calculated from $\delta^{15}\text{N}$ by using formula (Post, 2002b):

$$\text{TL}_{\text{consumer}} = \frac{\delta^{15}\text{N}_{\text{consumer}} - \delta^{15}\text{N}_{\text{baseline}}}{\Delta^{15}\text{N}} + \lambda \quad (1)$$

where λ is the trophic level of the baseline organism (TL = 1 for primary producers), $\text{TL}_{\text{consumer}}$ is the trophic level of a given consumer, $\delta^{15}\text{N}_{\text{consumer}}$ and $\delta^{15}\text{N}_{\text{baseline}}$ are the nitrogen stable isotope values of a given consumer and baseline organism (i.e. mean of algae) in a given lake, respectively, and $\Delta^{15}\text{N}$ is a trophic fractionation factor which was set to 3.4‰ per trophic level (Post, 2002b). We acknowledge that trophic fractionation can be variable from primary producers to top consumers (Bunn et al., 2013), but without more detailed evidence from these lakes, we adopted 3.4‰ trophic fractionation (van der Velden et al., 2013). We tested the sensitivity of our approach by calculating trophic levels for fish using one-source algae and zooplankton (TL_{alg} and TL_{zpl}) or two source-model of zooplankton and littoral benthic macroinvertebrates (TL_{zpl&bmi}) (Fig. S2). All models provided similar results (one-way ANOVA between all groups, p -value = 0.126, F -value = 2.16, df = 2; paired t -test for TL_{alg} vs TL_{zpl}, p -value = 0.328, t -value = 1.00, df = 18; for TL_{alg} vs TL_{zpl&bmi}, p -value = 0.198, t -value = -1.34 , df = 17; for TL_{zpl} vs TL_{zpl&bmi}, p -value = 0.092, t -value = 0.09, df = 17) (Fig. S2) and we therefore chose a one source model with algae as the baseline as this allowed us to assess TMS across four trophic levels in the subsequent analyses. For each lake, we identified a top consumer species with the highest mean trophic level which was also used to define food chain length (FCL) (Cabana and Rasmussen, 1996; Post et al., 2000).

2.6. Total mercury

Total mercury content (ng g^{-1} dry weight) of algae, invertebrates and fish white dorsal muscle was analyzed with direct mercury analyzer (Milestone DMA-80, Italy). For controlling the variability, two duplicates of each sample (20–30 mg) were analyzed when the sample amount was not limiting. Only duplicates with <10% of percentage difference were accepted for further analyses. All sets were analyzed with blank control and DORM-4 certified reference material (National Research Council Canada, Canada, powdered fish protein, mean THg concentration \pm SD, 410.0 ± 55.0 ng g^{-1}) at the beginning and ending of each set run (mean \pm SD, 403.7 ± 18.6 ng g^{-1} , mean recovery = 98.5%, n = 635) corrected by blanks (mean \pm SD, 2.0 ± 2.9 ng g^{-1}). In this study, the sample size (n) for THg analyses was 4718.

2.7. Statistical analysis

Biomagnification of THg in food webs was evaluated with the slope (b coefficient) from the $\log_{10}\text{THg}$ and trophic level linear regression Eq. (2) and is hereafter called TMS (ng g^{-1} d.w.) (Borgå et al., 2012; Lavoie et al., 2013). From the same equation, the intercept (a coefficient) indicates the THg exposure level of primary producers and is hereafter called THg baseline (ng g^{-1} d.w.) (Borgå et al., 2012). Additionally, Trophic Magnification Factor (TMF) was calculated from the TMS (3), to establish an increase of THg content per trophic level (Fisk et al., 2001; Borgå et al., 2012). Furthermore, we converted THg baseline back to a linear scale (4) to estimate the difference of basal THg intake by primary producers between lakes independently of the original exposure level (ng g^{-1} d.w.) (van der Velden et al., 2013):

$$\log_{10}\text{THg} = a + b \cdot \text{TL} \quad (2)$$

$$\text{TMF} = 10^b \quad (3)$$

$$\text{basal THg} = 10^a \quad (4)$$

In mercury biomagnification calculations, we used average values for each group to avoid bias in different sample sizes among the groups (Table 1). Two general multiple regression models with stepwise backward direction model selection based on AIC score were built to test the role of environmental (climate-productivity gradient [PC1], catchment properties [PC2], lake morphometrics [PC3]) and ecological variables (food chain length [FCL], number of fish species in the lake [n_{sp}] as a proxy for food web complexity, top predator age [P_{age}] as a proxy for predator size and longevity, and C:N ratio as a lipid content proxy (Kiljunen et al., 2006; Fagan et al., 2011) for top predator and zooplankton [$P_{C:N}$ and $Z_{C:N}$, respectively]) potentially explaining TMS and THg baseline in lakes. The full model takes the form:

$$\text{TMS/THg baseline} \sim \text{PC1} + \text{PC2} + \text{PC3} + \text{FCL} + n_{sp} + P_{age} + P_{C:N} + Z_{C:N} \quad (5)$$

Pike was the top predator in most lakes, except for three cases in the north, where the top predator was Arctic charr (Lake Kilpis [1]) or burbot (Lake Tshakal [3] and Lake Ropi [8]) (Table S4). Zooplankton refers to a bulk sample containing both copepods and cladocerans, where the former are more lipid-rich than later. THg content in 1 kg pike, commonly used as standardization of mercury values (see Åkerblom et al., 2014), was calculated with linear regression of THg content and weight in each lake. In all statistical analyses, a significance limit of $\alpha = 0.05$ was used. Statistical analyses were conducted with R version 1.1.463 (R Core Team, 2019) and by using FactoMineR package (Lê et al., 2008) for PCA analysis and MASS package (Venables and Ripley, 2002) for regression models.

3. Results

3.1. Prediction 1: TMS and THg baseline of food web

THg content was positively correlated with trophic level in all lakes and the fitted linear regression slopes were statistically significant, collectively indicating that biomagnification occurs in all measured food webs (Fig. 2). Mean ± SD of TMS was 0.81 ± 0.21 (range: 0.27–1.10) and the mean ± SD of the adjusted coefficient of determination (r^2) was 0.57 ± 0.13 (range: 0.31–0.86). TMS was negatively related to climate-productivity gradient (PC1; $r^2 = 0.20$; p-value = 0.055), as oligo- and mesotrophic lakes tended to have steeper slopes than eutrophic lakes (Fig. 3). TMS was negatively related to food chain length ($r^2 = 0.83$, $p < 0.001$), as oligotrophic lakes had simpler and shorter food chains (mean ± SD FCL = 3.36 ± 0.43), than mesotrophic (FCL = 3.87 ± 0.46) or eutrophic (FCL = 3.73 ± 0.33) lakes. Shallow oligotrophic and mesotrophic lakes with only littoral habitat tended to have shorter food chains (Table 1, Table S1).

Shallow oligotrophic lakes had the highest TMF of all lakes with an average of 4.36 (Table 1). The lowest TMF scores were found in mesotrophic lakes (mean value 3.43) and slightly higher TMF values in eutrophic lakes (mean value 3.44) (Table 1). THg baseline in food webs in the subarctic Tornio-Muoniojoki watercourse had a mean ± SD value of 0.81 ± 0.21 ng g⁻¹ d.w. (Table 1). The THg baseline was positively, but not significantly related to PC1 ($r^2 = 0.14$; p-value = 0.113) and FCL ($r^2 = 0.20$, p-value = 0.052).

However, when examined by lake type, eutrophic lakes tended to have higher basal THg values (9.14 ng g⁻¹ d.w.) and longer food chains (FCL = 3.73) compared to oligotrophic lakes (basal THg = 6.53 ng g⁻¹ d.w., FCL = 3.36) (Table 1, Fig. 3).

3.2. Prediction 2: Regression model of TMS and THg baseline

The full GLM model for TMS and THg baseline initially had eight explanatory variables (Eq. (5)). The final and most parsimonious TMS model was TMS ~ PC1 + PC2 + PC3 + FCL + P_{C:N}, explaining 90% of the variation, and for THg baseline the final model was THg baseline ~ PC1 + PC2 + PC3 + FCL + P_{C:N} + P_{age} which explained 69% of variation (Table 2; Table S5). Individual variable linear regressions with TMS and THg baseline are presented in Table S6.

3.3. Prediction 3: THg content of top predator of food web in climatic-productivity gradient

Top predator THg content was calculated for 1 kg pike in each study lake, where this species exists (Table 1). There was a significant positive relationship between THg content of 1 kg pike and climate productivity index (PC1) (Fig. 3e). In the average 1 kg pike, adjusted THg content of all the study lakes was 1209 ng THg g⁻¹ d.w., with mean ± SD ranged from 890 ± 219 ng g⁻¹ d.w. in mesotrophic, 1126 ± 494 ng g⁻¹ d.w. in oligotrophic to 1658 ± 368 ng g⁻¹ d.w. in eutrophic lakes.

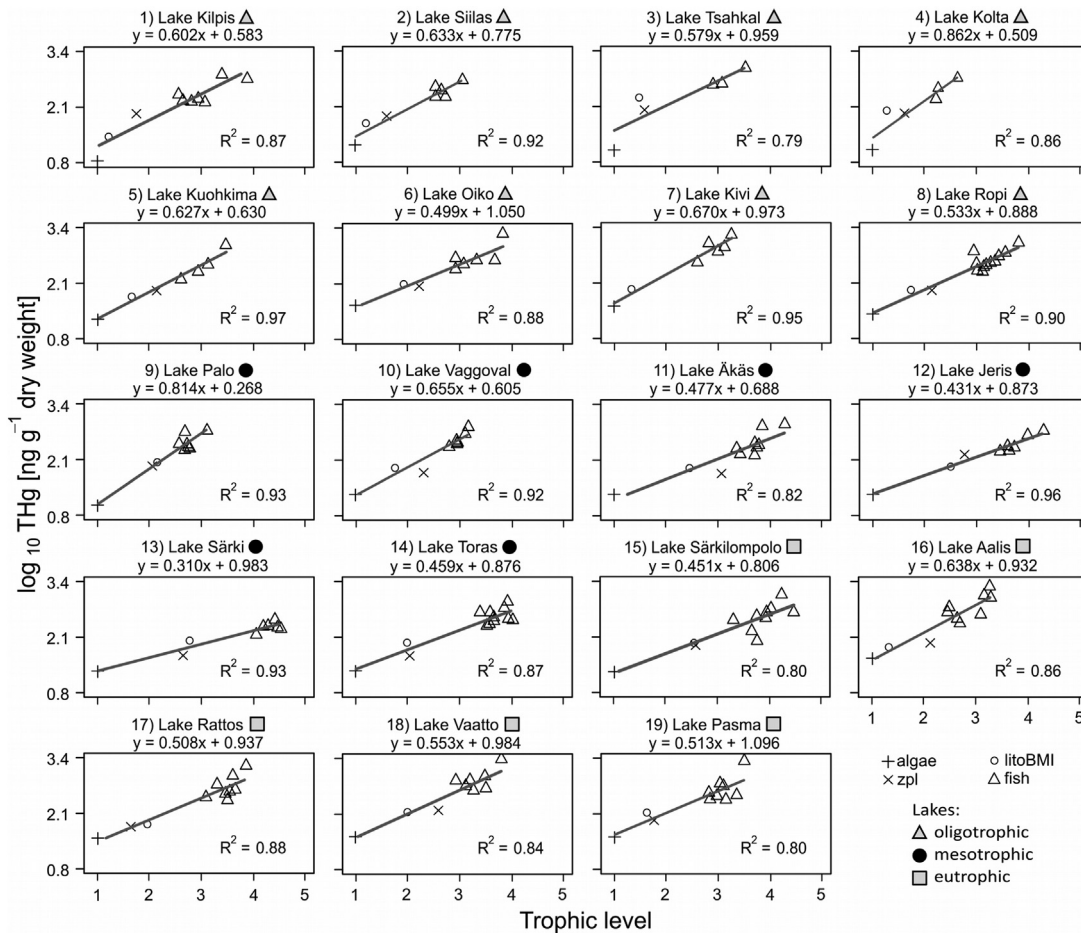


Fig. 2. Linear regression of average THg (log₁₀THg) (ng g⁻¹ d.w.) and trophic level (TL) for each group from primary producers (algae) to top fish consumers in each lake. Lakes are ordered with increasing climate-productivity gradient (PC1 value). For equation coefficients and statistical details, see Table 1 and further info on sample sizes, THg content and combined TMS plot (Table 2S, Table S4, Fig. S3, respectively). Abbreviations: litoBMI - littoral benthic macroinvertebrates; zpl - zooplankton.

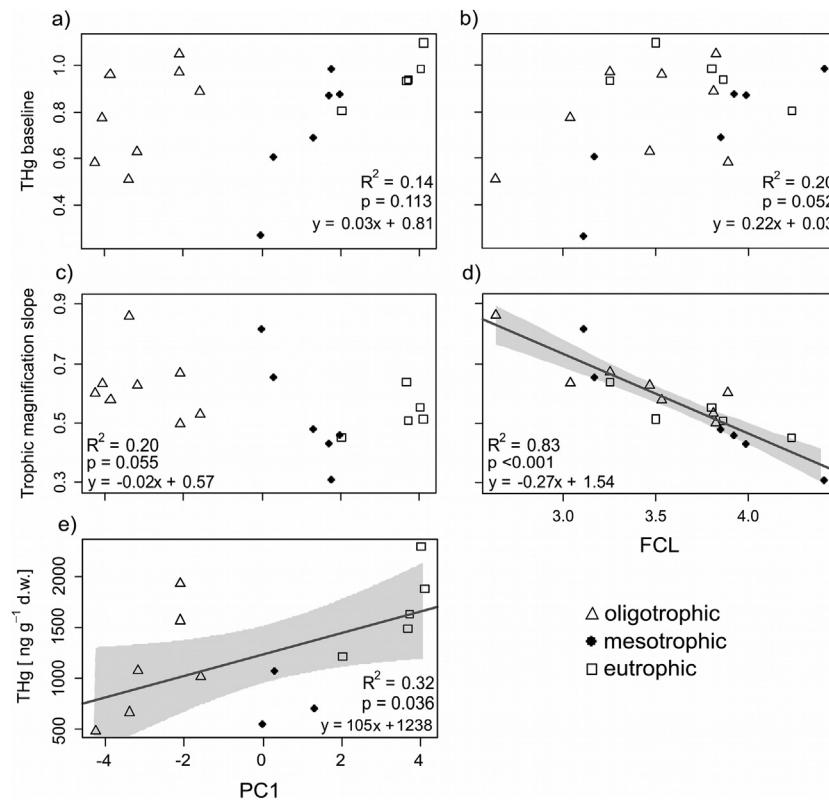


Fig. 3. The relationship of THg baseline (ng g⁻¹ d.w.) (a, b) and trophic magnification slopes (TMS) (ng g⁻¹ d.w.) (c, d) along climate-productivity gradient (PC1) and food chain length (FCL) with 95% confidence interval for significant relationships. Average THg content [ng g⁻¹ d.w.] of 1 kg pike in 14 lakes along climate-productivity gradient (e).

4. Discussion

4.1. Main results

Mercury biomagnification in food webs was found in all 19 lakes studied. Our predictions that increasing temperature and productivity would decrease the trophic magnification slope (TMS) and increase mercury baseline levels (THg baseline), were not statistically supported by the data. Instead, the ecological processes underlying these relationships were more complex and final explanatory models for trophic magnification slope and THg baseline included both biotic (food chain length, top predator age, lipid content of primary and top consumers) and abiotic (climate-productivity, catchment properties and lake morphometry) factors. Additionally, mercury content in a size standardized 1 kg pike, the apex predator in the lakes, increased towards warmer and more productive lakes.

4.2. Prediction 1: Climate and productivity relationship with THg biomagnification

Subarctic watercourses provide model systems to understand climate and productivity drivers of THg biomagnification in lake food webs and predict potential future with warming climate and land-use. In a global study of TMS in lakes, Lavoie et al. (2013) observed a positive relationship between TMS and latitude, but we did not find this trend in our smaller regional scale study. However, we cannot directly compare TMS values of these studies as they used $\delta^{15}N$ values and mercury content as wet weight values (Lavoie et al., 2013). Our results are more comparable with studies from Canadian Arctic and subarctic lakes, where TMS (calculated from $\log_{10}THg$ [ng g⁻¹ d.w.] against $\delta^{15}N$ derived trophic level in each lake) range between 0.30 and 0.72 (mean 0.48) (van der Velden et al., 2013). The slightly lower mean values in the Canadian lakes compared to our study lakes likely relate to

Table 2

Selection of backward stepwise model for generalized linear multiple regression analysis based on minimum AIC values. The final selected models are bolded (further details in Table S5). Climate-productivity gradient (PC1), catchment properties (PC2), lake morphometrics (PC3), food chain length (FCL), number of fish species in lake (n_{spc}), top predators age (P_{age}) and C:N ratio of top predator and zooplankton ($P_{C:N}$, $Z_{C:N}$).

Model	r ²	AIC	Deviance	Final model statistics
Trophic magnification slope (TMS)				
FCL + PC1 + PC2 + PC3 + P _{C:N} + Z _{C:N} + P _{age} + n _{spc}	0.91	-106.06	0.03	r ² = 0.90, RSE = 0.05,
FCL + PC1 + PC2 + PC3 + P _{C:N} + Z _{C:N} + P _{age}	0.91	-108.05	0.03	F _{5,13} = 24.48,
FCL + PC1 + PC2 + PC3 + P _{C:N} + Z _{C:N}	0.91	-109.92	0.03	p < 0.001,
FCL + PC1 + PC2 + PC3 + P_{C:N}	0.90	-110.67	0.03	AIC = -110.67
Intercept (THg baseline)				
FCL + PC1 + PC2 + PC3 + P _{C:N} + P _{age} + Z _{C:N} + n _{spc}	0.71	-65.23	0.24	r ² = 0.69, RSE = 0.15,
FCL + PC1 + PC2 + PC3 + P _{C:N} + P _{age} + Z _{C:N}	0.71	-67.23	0.24	F _{6,12} = 4.42,
FCL + PC1 + PC2 + PC3 + P_{C:N} + P_{age}	0.69	-67.86	0.26	p = 0.014, AIC = -67.86

regression calculations, as we used average THg values for different groups. In the Canadian Arctic and subarctic lakes, there was no clear latitudinal trend in either TMS or THg baseline (Gantner et al., 2010; van der Velden et al., 2013) corresponding to our results. The presence of Arctic charr as the dominant species in all the study lakes in both of these studies suggests that those lakes are oligotrophic, which may explain the less pronounced latitudinal pattern compared to our study, which includes lakes of a range of different trophic states.

Lake productivity is often related to biodilution of THg in organisms (Pickhardt et al., 2002; Chen and Folt, 2005; Borgå et al., 2012; Poste et al., 2015; Razavi et al., 2015); increased productivity results in longer and more complex food webs with increased biomass at each trophic level, and THg is distributed across this biomass decreasing biomagnification in the food web. However, no statistical support for this was found in the global lake comparisons (Lavoie et al., 2013), in Arctic lakes (Gantner et al., 2010) or subarctic lakes in the current study. Thus inconsistency in the relationship between eutrophication and mercury biomagnification may originate from different sources of nutrients i.e. natural or anthropogenic sources. Verburg et al. (2014) studied natural, geothermal lakes with different nutrient levels in New Zealand and found that biomagnification was most elevated in a eutrophic lake. In contrast, increasing level of eutrophication via agricultural and sewage sourced eutrophication decrease biomagnification in subtropical and tropical lakes via biodilution in algae (Poste et al., 2015; Razavi et al., 2015). In the eutrophic lakes analyzed here, increased productivity is derived from both increasing nutrient levels, eutrophication and browning via intensive forestry activities, especially peatland ditching. Such catchment modification likely induces considerable leaching of sequestered mercury from the peatland, elevating THg in primary producers and subsequent trophic levels. Our THg baseline values (mean \pm SD; 0.81 ± 0.20) are comparable to Canadian Arctic and subarctic studies, where THg baseline varied between 0.32 and 2.14 (mean $1.55 \log_{10}$ THg ng g⁻¹ d.w.) (van der Velden et al., 2013). In contrast, TMS was positively correlated with total phosphorus and negatively correlated with THg baseline along a gradient of the Canadian boreal lakes, with similar nutrient values to our study (Kidd et al., 2012). The difference may originate from much larger lakes in the Canadian study supporting more complex food webs. Nutrients (total nitrogen [TN], TP) explained a relatively small amount of variation in TMS (mean 0.23) in Canadian boreal lakes (Lescord et al., 2019), but a joint nutrient (TN, TP, total organic carbon [TOC]) and cation (calcium [Ca], chlorine [Cl]) PCA axis explained 30% of TMS variance (mean 0.17) in coastal Nova Scotian lakes (Clayden et al., 2013). Observed variation is likely due to regional disparities, where catchment, lake properties and nutrient sources are very different (Riget et al., 2000; Eloranta et al., 2015). Furthermore, contrasting results of productivity effects on TMS are likely linked to very drastic changes in biotic communities and food webs often present along our studied climate-productivity gradients (Hayden et al., 2017, 2019). For future THg biomagnification studies, we suggest that mercury TMS should be calculated as a function of trophic level instead of $\delta^{15}\text{N}$ values as this makes both spatial and literature comparisons possible.

A change to the food web structure in southern lakes caused TMS to have a stronger relationship with food chain length (FCL), than with climate-productivity index (PC1). Here, TMS decreased (83% variance explained) and THg baseline increased (20%) with increasing FCL. While FCL is well-known to elevate contaminant content (e.g. Rasmussen et al., 1990; Cabana et al., 1994), there is a little landscape-level evidence on how food chain length relates to TMS and THg baseline. Food chain length is positively correlated with ecosystem size, where larger lakes tend to have also more habitats and species (Post et al., 2000; Post, 2002a; Vander Zanden and Fetzer, 2007; Takimoto and Post, 2013). While the biodilution process is more robust in highly productive lakes, other variables such as ecosystem size, lake chemistry or food source preference can cause deviation in THg biomagnification and mask the biodilution in long food webs (Ouédraogo et al., 2015).

Kidd et al. (2012) found a positive relationship between lake size and TMS, while the opposite was true with THg baseline. The very pronounced differences among regions highlight the need for comprehensive explanatory models including different abiotic and biotic variables to explain THg biomagnification in different lake types.

4.3. Prediction 2: Explanatory models of THg biomagnification

The best fit model for TMS included PC1, PC2, PC3, FCL, and $P_{C:N}$, which together explained 90% of total variation in TMS. The THg baseline model included the same variables, but with inverse relationships, and top predator age (P_{age}). While many studies have used regression models to explain TMS and THg baseline in food webs (e.g. Kidd et al., 2012; Clayden et al., 2013; Lescord et al., 2015), we are not aware of any studies which included the same set of variables we used here. Below, we overview how each variable in both models contributes to the observed variation.

Food chain length displayed a strong negative relationship with the climate-productivity gradient, and was the most important factor in the TMS model, explaining 83% of total variation. The main process behind FCL is likely biodilution that was found in multiple studies (Chen and Folt, 2005; Keva et al., 2021). Subsequently, THg is distributed among greater variety and biomass of species at each trophic level (Poste et al., 2015; Razavi et al., 2015). Increased FCL in the more productive lakes is also related to the dominance of pike at the top of food chain as well as an increasing number of fish species, especially in the pelagic habitats (Ahonen et al., 2018).

Increasing lake productivity is related to changing catchment properties (PC2), where sparsely populated areas change towards increasingly modified catchments with forest clear-cutting, site preparation, and ditched peatlands (Jussila et al., 2014). Ditching of peatlands for forestry use releases historically accumulated mercury storages of the catchment. Site preparation and ditching activities are known to increase mercury methylation hotspots in lake catchments elsewhere (Eklöf et al., 2016; Ukonmaanaho et al., 2016), and this may contribute to our estimated THg baselines in the southern lakes. An intensive drainage network of the whole catchment will likely transport a large amount of nutrients and historical mercury into these lakes, elevating THg baseline. While anthropogenic eutrophication via agricultural fertilizers and sewage is generally considered to lower mercury concentrations in the base of the food web and organisms, it does not necessarily bring much mercury into the system. This difference between these ultimate sources of eutrophication should be considered when interpreting mercury bioaccumulation and biomagnification. Moreover, catchment derived nutrients and dissolved organic matter in southernmost murky lakes probably enhance anoxic conditions, which are known to promote methylation processes within lakes (Ullrich et al., 2001; Bravo et al., 2017). Increased dissolved organic matter also increases bacterial primary production in subarctic lakes and may further elevate THg baseline (Forsström et al., 2013; Lescord et al., 2018) and also THg content in top predators. Additionally, smaller lakes have a greater THg concentration in the water, which, according to our model, also elevates THg baseline.

On the contrary, TMS model showed a positive correlation with PC3, indicating that THg biomagnification was lower in small lakes, but this trend is far from clear. Both food web structure and catchment properties are highly dependent on climatic factors such as air temperature and precipitation (Hayden et al., 2017; Obrist et al., 2018). Thus, PC3 alone might not have a significant correlation with TMS or THg baseline (Fig. 3), even though it has a significant impact when included in the multiple regression model (an increase of r^2 in the model of 1% for TMS and 7% for THg baseline) (Table S5). In general, biotic fauna shift from cold-adapted lipid-rich species (e.g. copepods, salmonid fishes) to warmer adapted lean species (e.g. cladocerans, cyprinid and percid fishes) along the lake gradient (Hiltunen et al., 2016; Kahilainen et al., 2016; Hayden et al., 2017; Keva et al., 2021). This was observed as a

higher C:N ratio of top predator and zooplankton in the northern oligotrophic lakes and a lower C:N ratio in the eutrophic lakes (Table 1). In our study, Arctic charr and burbot were at the top of food chain only in the northernmost oligotrophic lakes, while pike was the top predator in all meso- and eutrophic lakes, and indeed in several oligotrophic lakes too. While all these species are piscivorous, pike has the ability to use a much wider range of all types of fish prey including cannibalism (Kahilainen and Lehtonen, 2003; Kahilainen et al., 2019). Arctic charr is more lipid-rich fish (mean C:N ratio 3.43) than pike (3.22) or burbot (3.25), which likely explains why predator C:N ratio was included in the model (one-way ANOVA test of C:N ratio between predator species p -value = 0.003, F -value = 8.12, df = 2).

THg baseline models included top predator age (P_{age}) as a significant variable (in final model, t -value = 3.65, p -value = 0.003) (Table S5). Top predator age itself is positively correlated to THg baseline (r^2 = 0.24, p -value = 0.033), but there is no correlation to TMS (r^2 = 0.01, p -value = 0.636) (Table S6). Age, length and mass of fish are directly related to THg bioaccumulation. While predator constantly accumulates THg with age and size, there is little impact of detoxication process (Kasper et al., 2009; Dang and Wang, 2012; Wang, 2012). THg baseline indicates THg input to the aquatic ecosystem, which later biomagnifies in top predators, thus our explanatory model used P_{age} to indicate greater THg input to the aquatic environment, however, this conclusion might be misleading, as fish in stable environmental conditions (i.e. predators from one lake) likewise bioaccumulate THg with time (Simoneau et al., 2005).

4.4. Prediction 3: Top predator THg in food webs along climate-productivity gradient

Top predator fish such as Arctic charr, burbot, and pike are an important component in the indigenous people diet in this region throughout the year (Sjölander, 2011; Thomas et al., 2016). According to health standards set by United Nations, the maximum level for mercury acceptable in fish muscle tissue for human consumption is 2500 ng g^{-1} d.w. for non-predatory fish and 5000 ng g^{-1} d.w. for predatory fish, based on the consumption of two fish meals per week (UNEP, 2008). We found several pike (n = 5) and perch (n = 12) which exceeded these limits from oligotrophic (Lake Oiko [6] and Kivi [7]) and eutrophic (Lake Aalis [16], Rattos [17] and Vaatto [18]) lakes. On the average, those individuals were old and large (perch average values: age = 13, total length = 32.0 cm; pike average values: age = 16, total length = 90.8 cm). We found that THg content of a standardized 1 kg pike increased with increasing temperature and productivity. This indicates potential adverse health effects for local fish consumers in the Fennoscandia contingent on the progressing climate and land-use change towards the end of this century.

5. Conclusions

TMS was negatively related to increasing temperature, productivity and food chain length, whereas the opposite was evident for THg baseline. This indicates that mercury biomagnification will be lowered in warmer and more productive conditions most likely due to an increased number of pelagic species and more complex food webs where mercury is biodiluted. THg baseline models indicate that the same variables increase the mercury content at basal resources. This may contribute to the overall higher mercury content in different trophic levels as we found increasing mercury content of a standardized 1 kg pike towards warmer and more productive lakes. The studied watercourse gradient corresponds to conservative future climatic scenarios for this region, but in addition, included a strong land-use gradient from near-pristine to intensive forestry catchments. Based on the current data, we cannot directly point out how much both the TMS and THg baseline will shift with future climate as land-use has to be also considered. Accordingly, results generally indicate the direction of change in the future rather

than the accurate prediction of change with certain climatic and land-use conditions.

Our results were opposite relative to the vast majority of previous studies and showed that eutrophication can increase THg baseline and THg content of top predator. However, it is very important to define the sources of nutrients as in most cases eutrophication originates from leaching agricultural fertilizers or direct sewage inputs, whereas in our case nutrients and carbon originate from ditched peatlands and peatland forests those leaching their historical mercury storages into lakes. In such conditions, leaching mercury from ditched catchment to the lakes elevates THg content of different trophic levels from baseline to top predators. Based on the results, we suggest that the sources of eutrophication should be always considered in the mercury bioaccumulation and biomagnification studies.

CRediT authorship contribution statement

Natalia Kozak: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing – original draft, Visualization. **Salla A. Ahonen:** Investigation, Data curation, Writing – review & editing. **Ossi Keva:** Methodology, Investigation, Writing – review & editing. **Kjartan Østbye:** Writing – review & editing. **Sami J. Taipale:** Resources, Writing – review & editing. **Brian Hayden:** Resources, Writing – review & editing. **Kimmo K. Kahilainen:** Conceptualization, Methodology, Validation, Investigation, Resources, Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2021.146261>.

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