



Mercury in fish from Norwegian lakes: The complex influence of aqueous organic carbon



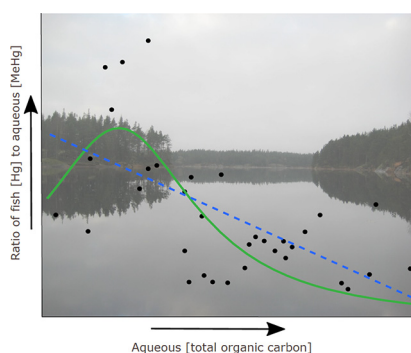
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HIGHLIGHTS

- Aqueous [TOC], [Hg] and [MeHg], and fish [Hg] studied in 34 boreal lakes
- Aqueous [TOC] is positively correlated to [Hg] in water and fish
- Fish BAFs (as [fish Hg]/[aqueous MeHg]) are lower in high [TOC] lakes
- Potential spurious correlation complicates relationship between TOC and BAF
- Increases in TOC affect Hg transport, bioavailability and food web accumulation

GRAPHICAL ABSTRACT



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ABSTRACT

Mercury (Hg) concentrations in water and biota are often positively correlated to organic matter (OM), typically measured as total or dissolved organic carbon (TOC/DOC). However, recent evidence suggests that higher OM concentrations inhibit bioaccumulation of Hg. Here, we test how TOC impacts the Hg accumulation in fish in a synoptic study of Methyl-Hg (MeHg) in water and total Hg (THg) in perch (*Perca fluviatilis*) in 34 boreal lakes in southern Norway. We found that aqueous MeHg ($r^2 = 0.49, p < 0.0001$) and THg ($r^2 = 0.69, p < 0.0001$), and fish THg ($r^2 = 0.26, p < 0.01$) were all positively related with TOC. However, we found declining MeHg bioaccumulation factors (BAF_{MeHg}) for fish with increasing TOC concentrations. The significant correlation between fish THg concentrations and aqueous TOC suggests that elevated fish Hg levels in boreal regions are associated with humic lakes. The declining BAF_{MeHg} with increasing TOC suggest that increased OM promotes increased aqueous Hg concentrations, but lowers relative MeHg bioaccumulation. A mechanistic understanding of the response from OM on BAF_{MeHg} might be found in the metal-complexation properties of OM, where OM complexation of metals reduces their bioavailability. Hence, suggesting that MeHg bioaccumulation becomes less effective at higher TOC, which is particularly relevant when assessing potential responses of fish Hg to predicted future changes in OM inputs to boreal ecosystems. Increased browning of waters may affect fish Hg in opposite directions: an increase of food web exposure to aqueous Hg, and reduced bioavailability of Hg species. However, the negative relationship between BAF_{MeHg} and TOC is challenging to interpret, and carries a great deal of uncertainty, since this relationship may be driven by the underlying correlation between TOC and MeHg (i.e. spurious correlations). Our results suggest that the trade-off between Hg exposure and accumulation will have important implications for the effects of lake browning on Hg transport, bioavailability, and trophodynamics.

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1. Introduction

Mercury (Hg) can undergo long-range transport in the atmosphere, and nearly two centuries of elevated Hg deposition from anthropogenic activities (Streets et al., 2011) have led to considerable stores of Hg in catchment soils, even in remote locations (Fitzgerald et al., 1998). Inorganic forms of Hg can be methylated into the neurotoxic Methyl-Hg (MeHg), which is biomagnified in aquatic food webs with potential harmful effects on aquatic organisms (Wolfe et al., 1998) and their consumers (Scheulhammer et al., 2007), including humans (Driscoll et al., 2013; Zahir et al., 2005). Although the toxic effects of Hg have been known for more than half a century (Kurland et al., 1960), researchers still struggle to understand and describe many of the complex processes involved in the biogeochemical cycling of the element, including mechanisms controlling accumulation and biomagnification of Hg in aquatic food webs.

Throughout the boreal zone, Hg concentrations in freshwater fish often exceed national and international dietary advisory limits, typically defined from 0.5–1.0 ppm (FAO UN, 1995; UNEP, 2002; Depew et al., 2012). Elevated concentrations are confirmed for both North American (e.g. Gandhi et al., 2014) and Scandinavian lakes (e.g. Åkerblom et al., 2014; Braaten et al., 2017), with levels posing a potential risk to ecosystem and human health. Recent studies have documented widespread increases in concentrations of aqueous organic matter (OM), normally measured as dissolved organic carbon (DOC) or total organic carbon (TOC), in freshwater lakes throughout the boreal forest zone (Monteith et al., 2007), and concentrations are predicted to change markedly with future climate change (de Wit et al., 2016, i.e. altered precipitation patterns). Browning of lakes may have strong impacts on Hg transport, availability and bioaccumulation and a better understanding of the interactions between climate, biogeochemistry and bioaccumulation is needed. In particular there is a need to increase our knowledge about the most decisive factors and processes for Hg accumulation in fish, and how these processes interact. This knowledge gap limits our ability to predict future levels of Hg in fish under various environmental changes.

Aqueous OM is known to affect the cycling of Hg in aquatic environments through chemical complexation (Ravichandran, 2004), through direct and indirect impacts on photochemical degradation of MeHg (Lehnher and Louis, 2009; Klapstein et al., 2018), and through microbial production and de-methylation mechanisms and processes (Gilmour et al., 1998). In recent decades, many studies have shown significant, positive correlations between aqueous concentrations of OM and Hg, both in water and biota (Braaten et al., 2014a; Chasar et al., 2009; Driscoll et al., 1995; Meili et al., 1991). However, more recent investigations suggest that at higher OM concentrations, Hg may be less available for uptake into aquatic food webs (e.g. French et al., 2014; Jeremiason et al., 2016; Tsui and Finlay, 2011). French et al. (2014) showed reduced bioaccumulation of both total Hg (THg) and MeHg in aquatic invertebrates (amphipods of different size classes: 250–2000 μm and $>2000 \mu\text{m}$) in Arctic lakes at DOC concentrations $>8.6 \text{ mg C L}^{-1}$, while increased accumulation occurred at DOC $<8.6 \text{ mg C L}^{-1}$. Amphipods from lakes with higher DOC concentrations had lower Hg bioaccumulation factors (BAFs), defined as the ratio between the concentration of Hg in an organism and the concentration of Hg in its surrounding environment, i.e. water (French et al., 2014). A similar threshold value ($\approx 8 \text{ mg C L}^{-1}$) was proposed by Driscoll et al. (1994) from a limited data set of North American fish populations. French et al. (2014) suggest that the mechanism responsible for the change in BAF with OM concentrations relates to the complexation of Hg by OM, where high presence of humic acids make Hg species less bioavailable.

Herein, we wanted to investigate the relationship between aqueous concentrations of MeHg and TOC in 34 boreal lakes in southern Norway and assess the effects of TOC concentrations on Hg in perch (*Perca fluviatilis*). We explore various modelling approaches, including a BAF approach similar to that of French et al. (2014), and discuss the

limitations and potential future environmental implications of our results. Hg concentrations in aquatic food webs depend strongly on the uptake of Hg at the base of the food web (Chasar et al., 2009), implying that a potential TOC threshold for Hg bioaccumulation in fish prey will also be reflected as a threshold response in fish Hg. We hypothesise that reduced bioavailability of MeHg occurs at higher TOC concentrations, leading to lower BAFs for Hg in fish.

2. Experimental

2.1. Study sites and field sampling

Our investigation is based on 34 lakes from southeast Norway, located between 58.84° N and 60.51° N, 7.96° E and 12.51° E (WGS84, see Fig. 1 and Supporting Information for details). All lakes are located within the boreal forest ecotone, dominated by coniferous forest and wetlands, without direct influence from agricultural activities. Lake-specific information on geographical location is attached as Supporting Information (Table S1), while water chemistry (THg, MeHg and TOC) and fish Hg concentrations are shown in Table 1

Samples were collected in the early autumn (August–September) between 2008 and 2012. Perch (*Perca fluviatilis*) is the most abundant fish species in all lakes, and was collected using gill nets composed of different mesh sizes for a broad distribution of fish size. For further details regarding sampling and sample handling we refer to Braaten et al. (2014b). To be included in our calculations, fish data from each of the 34 lakes had to comprise of at least $n = 10$ specimens, with the mean (\pm one standard deviation) being $n = 23 \pm 7$ fish (Table 1).

Water sampling for chemical analysis was conducted as described in Braaten et al. (2014a). In short, samples of surface water for Hg speciation were collected using 250 mL fluoropolymer bottles, following ultraclean sampling procedures to avoid contamination (USEPA, 1996). All bottles were previously unused, and water for determination of THg and MeHg was sampled in separate bottles to avoid errors resulting from loss of Hg during preservation (Creswel et al., 2016; Braaten et al., 2014c). Samples for TOC determination were collected at the same time as the samples for Hg analysis in an high-density polyethylene (HDPE) bottle.

2.2. Analytical methods

Hg concentrations in fish were determined as THg. Wet samples of muscle tissue were analysed by thermal decomposition and direct atomic absorption spectrophotometry (Lumex Mercury Analyser RA915). Quality assurance and quality control (QA/QC) are described in detail in Braaten et al. (2014b). Relative standard deviation (RSD) of sample duplicates was $<10\%$ and recovery of certified reference material (DORM-3 fish protein) within 90–110%.

The analytical method for MeHg in water was based on USEPA method 1630 (USEPA, 1998) by distillation, aqueous ethylation, purge and trap, and cold vapor atomic fluorescence spectrometry (CVAFS). The method for THg in water followed USEPA method 1631 (USEPA, 2002) by oxidation, purge and trap, and CVAFS. Method detection levels (MDLs) were 0.02 and 0.1 ng L^{-1} for MeHg and THg, respectively. For both Hg species, automated systems were used for analysis (Brooks Rand Instruments MERX). QA/QC are described in detail in Braaten et al. (2014a). RSD of sample duplicates was $<10\%$ and $<20\%$ for THg and MeHg, respectively. Recoveries of blank spikes and matrix spikes were 80–120% for MeHg and 90–110% for THg. Both THg and MeHg were determined on unfiltered water to allow for comparison with levels of TOC.

TOC was measured by infrared spectrophotometry according to Norwegian and European Standard NS-EN1484 with a measurement uncertainty of $\pm 20\%$ and a MDL of 0.1 mg L^{-1} .

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