



## Review

# The importance of bioconcentration into the pelagic food web base for methylmercury biomagnification: A meta-analysis



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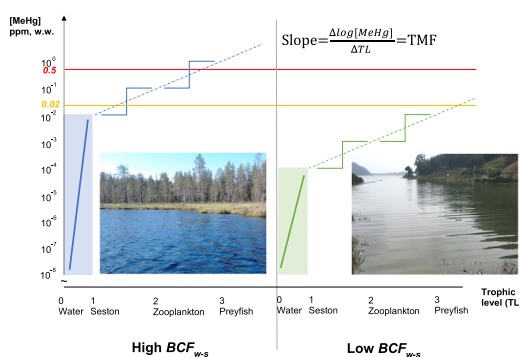
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## HIGHLIGHTS

- Extensive MeHg bioaccumulation literature review and meta-analysis
- MeHg bioconcentration factors at food web base predict preyfish MeHg.
- DOC and trophic status influence MeHg bioconcentration factors.
- MeHg concentration in water column does not predict MeHg in biota.
- Conceptualization of a MeHg BCF elevator leading to a BMF staircase

## GRAPHICAL ABSTRACT



## ARTICLE INFO

## Article history:

Received 9 March 2018

Received in revised form 23 July 2018

Accepted 23 July 2018

Available online 24 July 2018

Editor: Yolanda Picó

## Keywords:

Hg  
MeHg  
Meta-analysis  
Seston  
Fish  
Bioconcentration factors  
Biomagnification factors

## ABSTRACT

Methylmercury (MeHg) transfer from water into the base of the food web (bioconcentration) and subsequent biomagnification in the aquatic food web leads to most of the MeHg in fish. But how important is bioconcentration compared to biomagnification in predicting MeHg in fish? To answer this question we reviewed articles in which MeHg concentrations in water, plankton (seston and/or zooplankton), as well as fish (planktivorous and small omnivorous fish) were reported. This yielded 32 journal articles with data from 59 aquatic ecosystems at 22 sites around the world. Although there are many case studies of particular aquatic habitats and specific geographic areas that have examined MeHg bioconcentration and biomagnification, we performed a meta-analysis of such studies. Aqueous MeHg was not a significant predictor of MeHg in fish, but MeHg in seston i.e., the base of the aquatic food web, predicted 63% of the variability in fish MeHg. The MeHg bioconcentration factors (i.e., transfer of MeHg from water to seston;  $BCF_{w-s}$ ) varied from 3 to 7 orders of magnitude across sites and correlated significantly with MeHg in fish. The MeHg biomagnification factors from zooplankton to fish varied much less ( $\log BMF_{z-f}$   $0.75 \pm 0.31$ ), and did not significantly correlate with fish MeHg, suggesting that zooplanktivory is not as important as bioconcentration in the biomagnification of fish MeHg across the range of ecosystems represented in our meta-analysis. Partial least square (PLS) and linear regression analyses identified several environmental factors associated with increased BCF, including low dissolved organic carbon, low pH, and oligotrophy. Our study reveals the widespread importance of MeHg bioconcentration into the base of the aquatic food web for MeHg at higher trophic levels in aquatic food webs, as well as the major influences on the variability in this bioconcentration.

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## 1. Introduction

Methylmercury (MeHg) is known for its high degree of bioaccumulation in aquatic food webs, with MeHg concentrations in predatory fish that can be six orders of magnitude higher than in surface water (Lindqvist et al., 1991; McMeans et al., 2015). The stepwise biomagnification of MeHg from one trophic level to the next results in MeHg concentrations ([MeHg]) in aquatic biota that can pose health risks to humans as well as wildlife (Clarkson et al., 2003; Sheehan et al., 2014). Reducing environmental exposure of humans and wildlife to MeHg could save the European Union (EU) up to €8–9 billion per year (Bellanger et al., 2013). To protect human health, the World Health Organization (WHO) has set an environmental quality standard (EQS) in fish of 0.5 µg MeHg g<sup>-1</sup> wet weight (w.w.) (FAO/WHO, 2015). In the European Union, the EQS for total mercury (Hg) in fish (0.02 µg Hg g<sup>-1</sup> w.w.) is >20 times lower compared to the WHO EQS since the EU seeks to better protect secondary consumers in aquatic ecosystems (i.e. piscivorous birds and mammals) (EC, 2013; Vignati et al., 2013). The WHO EQS is already exceeded in >50% of Swedish lakes (Åkerblom et al., 2014) and is also often exceeded in piscivorous fish species in Northern America (Gandhi et al., 2014). Regionally, the more stringent EU EQS is often exceeded, especially in boreal freshwaters, including almost all those in Fennoscandia (Nguetseng et al., 2015).

Globally, there are large regional differences between Hg concentrations ([Hg]) in both water and fish across regions (Selin et al., 2007; Selin, 2009). Even within regions there is a considerable variability in fish [Hg], which generally does not relate well to [Hg] or MeHg concentrations [MeHg] in water (Rolfhus et al., 2011; Liu et al., 2012). An indicator to better understand the transfer of Hg from water into biota, and the subsequent biomagnification in fish, could improve the environmental policies to reduce fish [Hg]. Chen et al. (2014) have already shown the significance of using water column particulate MeHg to predict fish MeHg through an investigation of 10 estuary sites in Northeastern USA. This would be valuable since the environmental response to policies such as Hg emission reductions can be slow, often on a time-scale of decades to centuries (Kumamoto Prefecture, 1998; Meili et al., 2003; Kindaichi and Matsuyama, 2005; Danielsson et al., 2011), and fraught with uncertainties.

While Hg in fish poses a threat to consumers regardless of its chemical speciation, MeHg is the Hg species that bioaccumulates most efficiently (Clarkson et al., 2003). Many studies that analyzed [MeHg] in aquatic biota with a focus on MeHg bioaccumulation in piscivorous wildlife and its relation to Hg exposure have shown that [MeHg] increases steadily with each trophic level (Mason et al., 1995; Munthe et al., 2007; Clayden et al., 2013). A comprehensive review by Lavoie et al. (2013) on [Hg] and [MeHg] along the aquatic food chains across

the globe has put much of the existing data in perspective. They found that biomagnification patterns above the base of the food web were relatively similar with some consistent differences between regions. Lavoie's review, however, used longer-lived benthic mussels/snails as the trophic baseline. Thus, the review did not address the entry of Hg into the base of the aquatic food web, i.e., the step from aqueous [MeHg] to organisms at the lowest trophic level. Using seston (i.e., suspended particles, mostly composed of small algae and bacteria) as the base of the pelagic food web, a study of the western Great Lakes region found that dissolved organic matter (DOM) limited MeHg uptake from water by seston, zooplankton, and fish, yielding lower bioconcentration factors (BCF, defined as the ratio between the MeHg concentrations in biota and water), while the trophic magnification factor (TMF) from the base of the food web upwards remained stable across different food webs (Rolfhus et al., 2011). This finding is an indication of the complexity of Hg bioconcentration from water into the biota due to the influence of different environmental factors, but also the importance of the variability of bioconcentration into the base of the pelagic food web for the biomagnification to higher trophic levels.

Correlations between catchment characteristics and fish [Hg] have also been utilized to examine the influence of environmental factors on [Hg] in aquatic biota. Kidd et al. (2011) have argued that physical and chemical characteristics, e.g. DOC and pH, reflect Hg bioavailability in the environment that may be a critical influence on Hg biomagnification. For example, low pH surface water (pH < 6) with higher [Hg] is associated with higher [Hg] in generally oligotrophic freshwater ecosystems (Le Faucheur et al., 2014). On the other hand, eutrophic aquatic ecosystems are associated with increased plankton biomass and consequently lower [Hg] in the aquatic biota (per unit biomass), which is described as “biodilution” (Pickhardt et al., 2002; Karimi et al., 2007). Some very eutrophic aquatic ecosystems in China and Eastern Europe have relatively high aqueous [MeHg] (>1 ng L<sup>-1</sup>), but rather little [MeHg] in biota (<0.2 µg g<sup>-1</sup> w.w.) (Farkas et al., 2000; Nguyen et al., 2005; Suchanek et al., 2008; Liu et al., 2012). This stands in contrast to higher [MeHg] in the biota of oligotrophic boreal ecosystems (>0.5 µg g<sup>-1</sup> w.w.) despite aqueous [MeHg] that are often <1 ng L<sup>-1</sup> or even <0.1 ng L<sup>-1</sup> (Meili, 1991; Watras and Bloom, 1992; Clayden et al., 2013). Some of these differences may derive from varying MeHg photodemethylation (PD) activity in freshwater lakes as a function of color (Lehnher and St. Louis, 2009). Nevertheless, the magnitude of the discrepancy between aqueous [MeHg] and fish [MeHg] across regional and global scales, but also within regions, points to the need for a better understanding of what controls the transfer of MeHg from water into the base of pelagic food webs. We are specifically interested in how the basal MeHg transfer influences the overall distribution of MeHg in aquatic ecosystems (i.e., lakes, reservoirs, and estuaries).

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