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Differential bioaccumulation of mercury by zooplankton taxa in a mercury-contaminated reservoir Guizhou China☆

Sheng-Xing Long [a, c], Paul B. Hamilton [b], Yang Yang [a, *], Sai Wang [a], Wen-da Huang [a], Chuan Chen [c], Ran Tao [a]

[a] Institute of Hydrobiology, College of Life Science and Technology, JiNan University, Guangzhou, PR, 510632, China
[b] Research and Collections, Canadian Museum of Nature, P.O. Box 3443, Station D, Ottawa, Ontario, K1P 6P4, Canada
[c] Guizhou Normal University, Guiyang, Guizhou, 550001, China

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ABSTRACT

Mercury (Hg) contamination in aquatic systems remains a global concern with the biomagnification of methylmercury (MeHg) through primary consumers (zooplankton) to fish and humans. In this study, total mercury (THg) and MeHg concentrations were analyzed in zooplankton collected from Baihua reservoir (Guizhou Province, China). Our results demonstrated that THg and MeHg concentrations were strongly correlated to zooplankton community and biomass composition. The THg concentration was significantly higher in micro-zooplankton compared to meso-zooplankton and macro-zooplankton, and MeHg concentration increased significantly as body size increased. Hg increases in zooplankton were influenced by the numbers of calanoid copepods and *Daphnia* present relative to phytoplankton and zooplankton biomass. Many zooplankton taxa in the three size-fractions were affected by THg exposure. The biomasses of *Bosmina longirostris*, *Thermocyclops brevifurcatus, Asplanchna priodonta* and *Cyclops vicinus vicinus* were positively correlated with Hg accumulation, while *Daphnia hyalina*, and *Phyllodiaptomus tunguidus* had a negative association. THg and MeHg bioaccumulation factors were correlated with phosphorus and total nitrogen concentration, zooplankton biomass, and chlorophyll-*a* concentration. Phosphorus loading was associated with increased THg and MeHg accumulation in the zooplankton highlighting biomagification with eutrophication. Chlorophyll-*a* levels were not correlated to THg and MeHg accumulation in zooplankton when phytoplankton densities were $>10^7$ cells L^{-1} and chlorophyll-*a* concentrations <9 μg L^{-1}. This finding contradicts the idea of MeHg biodilution with increased algae biomass. However, changes in the phytoplankton species and biomass altered the availability of food for zooplankton, particularly micro-zooplankton and macro-zooplankton. Ultimately, the bioaccumulation of MeHg and THg across lower trophic levels was based more on the availability of preferred food resources than on total biological productivity.

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1. Introduction

Public health concerns about neurotoxicity, relating to the trophic transfer and bioaccumulation of methylmercury (MeHg) in aquatic food chains, are increasing around the world (He et al., 2008; Liu et al., 2012; Todorova et al., 2015). Increased food consumption containing unknown levels of Hg has also been a concern, resulting in the development of guidelines for fish consumption in many countries (e.g. Clarkson and Magos, 2006; Todorova et al., 2015). Numerous toxicity studies have shown that MeHg bioaccumulation and trophic transfer in lakes is related to physical, chemical, and ecological factors, including inorganic Hg, dissolved organic carbon, sulfide (S^{2-}), sulfate (SO_4^{2-}), dissolved oxygen, temperature, pH, phytoplankton and zooplankton (e.g. Driscoll et al., 2007; Feyte et al., 2012; Yan et al., 2013). Trophic transfer and bioaccumulation of MeHg is often positively related to total mercury (THg) (Long et al., 2017). However, one study, documented that increased eutrophication in the water column had a negative relationship to Hg bioaccumulation; this response was associated with the dilution of Hg through increased phytoplankton densities (Baihua reservoir, in Southwest China) (Liu et al., 2012). Results

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* Corresponding author.
E-mail address: Yangyang@scies.org (Y. Yang).

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from past research also suggest that eutrophic reservoirs have lower Hg concentrations in fish (top trophic level) compared to oligotrophic reservoirs; this negative relationship has been in part attributed to higher phytoplankton biomass (MeHg dilution per cell) and the partitioning of Hg into biomass size fractions across trophic levels (Pickhardt et al., 2002; Chen and Folt, 2005). Chen et al. (2012) also found that THg concentrations in zooplankton and predators were negatively correlated with chlorophyll-*a* (Chl-*a*) concentration because of bio-dilution linked to eutrophication. These results are aligned with observed high Hg burdens observed in fish from more pristine (lower productivity) lakes and reservoirs in North America and northern Europe (Munthe et al., 2007; Larssen, 2010).

Zooplankton are ubiquitous, often abundant in freshwater and can play a key role in the biogeochemical cycling of Hg in freshwater systems (e.g. Todorova et al., 2015; Long et al., 2017). Variability in community composition (including body size) may determine the upward trophic migration of Hg (Chen et al., 2012; Todorova et al., 2015; Gosnell et al., 2017), and like phytoplankton, higher zooplankton densities can potentially decrease Hg transfer to fish through zooplankton density dilution (Chen and Folt, 2005). At present, it is not clear how Hg accumulates when there are changes in eutrophic state. This year long temporal and spatial study focused on changes in THg and MeHg concentrations in zooplankton from Baihua Lake reservoir (southwest China). Baihua reservoir has experienced elevated concentrations of Hg due to direct discharges from an acetic acid plant. The reservoir has also received substantial loads of nitrogen and phosphorus as a result of untreated domestic inputs. Recent decreases in nitrogen and phosphorus in the reservoir are attributed to wastewater treatment and reduced loading; wastewater dumping of Hg has been prohibited since 1997. Thus Hg pollution, nutrient loads, and primary production have changed markedly over the last 50 years in Baihua reservoir and documentation of this pollution makes the system a good in situ study case. The objective of the current study is to investigate Hg and MeHg bioaccumulation by zooplankton in relation to Hg availability and phytoplankton biomass. This study tested the hypothesis that differences in biomass and species composition of zooplankton can account for a significant amount of the variation in Hg accumulation. Bioaccumulation theory was used (in the context of the changing nutrient status of karst plateau reservoirs in China) along with changes in the zooplankton community to provide an alternative perspective on the potential effects of food chain composition on bioaccumulation of Hg and MeHg.

2. Materials and methods

2.1. Study sites

Baihua Lake reservoir (26° 35′–26° 42′ N, 106° 27′–106° 34′ E) is approximately 16 km northwest of Guiyang City, Guizhou Province, southwest China. The reservoir was built in 1962 and filled in 1966. Baihua reservoir provides hydroelectric power, water for irrigation, drinking water, and recreational resources to local communities and millions of people in the City of Guiyang. The reservoir is long and narrow with a mean water depth of 13 m and mean water residence time of one month (Liu et al., 2012). The reservoir is in a subtropical karst region with a typical subtropical rainy monsoon climate having an annual mean air temperature of 13.8 °C. The highest water temperature recorded was 28 °C, with a mean water warm season temperature of 23 °C. The lowest winter water temperature recorded was 4 °C, with a mean cold season temperature of 10 °C (Yan et al., 2013). Annual mean precipitation around Baihua reservoir is approximately 1175 mm (Wang et al., 2011). The highest total nitrogen (TN) and total phosphorus (TP) concentrations recorded in the reservoir in 1997 and 1998 were 5.34 and 1.16 mg L^{-1}, respectively. These concentrations were the result of inputs from a fish farming industry. The mean TN and TP concentrations have since decreased in 2012 to 1.70 and 0.03 mg L^{-1} respectively (Liu et al., 2012; Long et al., 2016).

High THg concentrations in the sediment and water, 12.9 mg kg^{-1} and 22.4 ng L^{-1}, respectively, were observed due to anthropogenic loading of Hg. Mercury was used as a catalyst for the production of acetic acid between 1971 and 1997 by the Guizhou Organic Chemical plant near the upper part of the reservoir. The chemical plant used 573 t of Hg between 1971 and 1985 (Yan et al., 2008). Untreated wastewater from the plant was discharged directly into the Zhujia River, which flows into Baihua reservoir. This caused the reservoir to become seriously contaminated with Hg (Zhang, 2000). More recently, Liu et al. (2012) reported THg concentrations in the sediment at 3677 ± 2596 ng g^{-1}, and THg in water column at 6.9 ± 4.5ng L^{-1}; THg concentrations in the water column are currently declining. In the present work, biogeochemical cycling of Hg in zooplankton was investigated at four study sites. S1, Yanjiaozhai is located near the Guizhou Organic Chemical Plant. A large number of zooplankton sample were collected here because of the water depth (8–12 m). S2, Qushuiko is located near the Maixi River, a source of domestic wastewater (8 m depth), S3, Bengfang is positioned near the water intake for the City of Guiyang (18 m depth) and S4, Daba is found nearby the dam (22 m depth). (S1 Yanjiaozhai, 26° 37′ 55″ N, 106° 29′ 57″ E; S2 Qushuiko 26° 39′ 08″ N, 106°32′02″ E; S3 Bengfang 26° 40′ 12″ N, 106° 33′ 04″ E; S4 Daba 26° 40′ 59″ N, 106° 32′ 40″ E) (Fig. 1).

2.2. Sample collection

2.2.1. Lake water sampling

Water samples were collected from Baihua reservoir for THg and MeHg analysis. Each water sample (a mixture from 0.5 m below the surface, 6 m deep, and 0.5 m above the bottom) was collected in a borosilicate glass bottle (120 mL). This sample mix allowed for direct correlations between THg and MeHg in water and THg and MeHg in zooplankton which were collected between the surface and 0.5 m above the bottom. Before collection, each bottle was cleaned by soaking in acid, rinsed with ultrapure deionized water, and baked for several hours at 500 °C in a muffle furnace. Each water sample was acidified with 0.5% HCl, placed in a clean zip-closed bag within another zip-closed bag, transported to the laboratory within 24 h, and stored at 4 °C in the dark until analyzed (Long et al., 2017). The water temperature, dissolved oxygen concentration, and pH were measured when each sample was collected using a 6600 multi-sensor sonde (Yellow Springs Inc., Yellow Springs, OH, USA). TN and TP concentrations in each sample were determined using the alkaline potassium persulfate oxidation method (Ma et al., 2014). Chlorophyll-*a* was analyzed by spectrophotometry (UV-2550, Japan) after extraction in 90% acetone (Ma et al., 2014). A phytoplankton sample was collected from the surface water at each sampling site for phytoplankton community composition, abundance, and biomass enumerations. Each phytoplankton sample was preserved with the addition of Lugol's iodine, then allowed to settle for 48 h and concentrated to a final volume of 30 mL. Cell density was measured using a Sedgwick–Rafter counting chamber with counts at magnifications between 200x and 400x. The species were identified as described by Hu et al. (1980). The total algal biovolume for each species was calculated

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