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Assessing correlations between monomethylmercury accumulation in fish and trophic states of artificial temperate reservoirs

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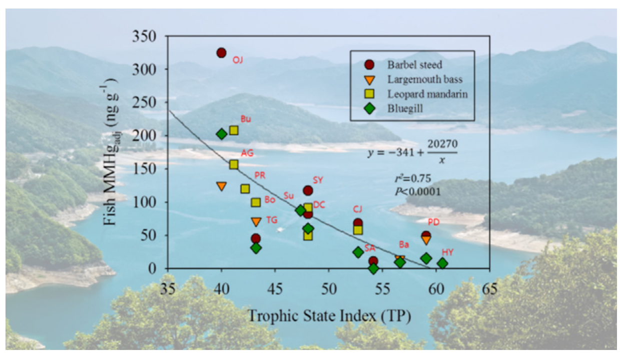
HIGHLIGHTS

- Fish MMHg showed a negative correlation with lake trophic state index.
- It might be a result of particle dilution of MMHg at the base of food chains.
- Measuring trophic state is a practical tool to predict MMHg bioaccumulation.

GRAPHICAL ABSTRACT



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ABSTRACT

We investigated monomethylmercury (MMHg) concentrations in 448 samples of four common fish species (barbel steed, largemouth bass, leopard mandarin, and bluegill) and the trophic states of 14 artificial reservoirs in South Korea in order to understand how trophic states of reserviors affect MMHg concentrations in fish. The trophic state index (TSI) of each reservoir was determined using empirical equations based on the monthly chlorophyll-a, total phosphorus, and Secchi depth, collected over a three-year period. The length-normalized MMHg concentrations in fish showed a negative correlation with the TSI based on chlorophyll-a ($r^2 = 0.90$) and total phosphorus ($r^2 = 0.75$) that may be a result of particle dilution of MMHg in surface waters. This study revealed that MMHg accumulation in fish was better correlated with TSI than MMHg in water, therefore, we suggest that the measurement of TSI based on chlorophyll-a and total phosphorus is an effective way to predict MMHg bioaccumulation across diverse reservoirs.

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1. Introduction

Monomethylmercury (MMHg) concentrations can differ in top predator fish from lakes with similar inorganic Hg(II) inputs due to the physicochemical and biological properties of the lakes, which

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cause significant variations in MMHg production and bioaccumulation (Dittman and Driscoll, 2009; Greenfield et al., 2001; Lithner et al., 2000; Watras et al., 1998). In general, eutrophication promotes in situ MMHg production rate as a result of enhanced organic input in the bottom sediment or hypolimnion (Gray and Hines, 2009; Noh et al., 2016). High algal biomass scavenging Hg(II) sinks to the sediment, leading to the active decomposition of organic matter and producing favorable conditions for microbial Hg(II) methylation (Barsanti and Gualtieri, 2005; Chen et al., 2000; Chen et al., 2007). Studies have reported higher MMHg concentrations in fish from eutrophic lakes compared to those that are oligotrophic, and this has been explained by increased net production of MMHg in eutrophic lakes (Lithner et al., 2000; Stone et al., 2011; Verburg et al., 2014).

The biological factors of the fish, such as size, age, feeding type, ingestion rate, and growth rate, are important for determining the ultimate MMHg concentrations in their tissues. MMHg concentration in fish often increases with the size and age of the fish due to cumulative bioaccumulation and/or prey selection shift (Chen et al., 2007; Driscoll et al., 1994). The positive correlation between total Hg (THg) concentration in fish and body size has been attributed to shifts in diet to a higher trophic position, as evidenced by $\delta^{15}N$ of the fish tissues (Power et al., 2002; Xu et al., 2016).

Algal blooms can reduce MMHg uptake into the food web through a biomass dilution effect (Chen and Folt, 2005; Pickhardt et al., 2002) or a somatic dilution effect caused by disproportionate increases in biomass gain relative to Hg gain (Karimi et al., 2007). Somatic dilution occurs when the activity or respiration rates of organisms are comparatively low or when their food quality is high, as observed in eutrophic lakes (Karimi et al., 2007). These observations contradict those of previous studies, which show higher fish MMHg concentrations in eutrophic lakes than in oligotrophic lakes (Lithner et al., 2000; Stone et al., 2011; Verburg et al., 2014), indicating that there are uncertainties with the consequence of eutrophication regarding MMHg accumulation in fish.

South Korea has about 18,000 reservoirs, most of which were constructed after the 1970s, to prevent water shortages in dry seasons (Kim et al., 2001). These reservoirs have diverse geographical characteristics and trophic states, ranging from oligotrophic to eutrophic, depending on the catchment land use and hydrodynamic conditions (Noh et al., 2016). We carried out preliminary studies on the site variation in aqueous MMHg in these reservoirs, and we demonstrated that algal production was a critical predictor of the variance of the fraction of MMHg over Hg (%MMHg) in lake waters (Noh et al., 2016). However, a higher MMHg production in algal-enriched reservoirs does not guarantee greater bioaccumulation of MMHg in fish, according to the available literature (Chen et al., 2005; Clayden et al., 2013; Pickhardt et al., 2002). Therefore, further studies are needed to understand the relationship between the eutrophication of reservoir waters and MMHg accumulation in fish.

The influence of trophic states of reservoirs on MMHg accumulation in fish was investigated here by performing a field study of 14 reservoirs in South Korea from 2013 to 2015. The geochemical characteristics of the surface water, including dissolved oxygen, pH, electric conductivity (EC), Secchi depth (SD), suspended solids (SS), total organic carbon (TOC), chlorophyll a (chl-a), total phosphorus (TP), and total nitrogen (TN), were obtained from the national networking systems as monthly data. The Hg species concentrations in water and fish, together with supplementary water chemistry data, were determined from the sampling campaigns. We used these data to assess the relationship between reservoir trophic state and MMHg concentrations in fish.

2. Experimental methods

2.1. Sample collection

The 14 test reservoirs are located in South Korea from 35 to 38° N and 126 to 129° E within generally forested catchments without wetlands (Fig. 1). South Korea has a temperate climate, and the mean air temperature is 22 to 25 °C in the summer and −5 to −2 °C in the winter (Kim et al., 2001).

A total of 448 fish were collected using trap netting from 14 reservoirs between 2013 and 2015. The target fish were barbel steed (*Hemibarbus labeo*), largemouth bass (*Micropterus salmoides*), leopard mandarin (*Siniperca scherzeri*), and bluegill (*Lepomis macrochirus*), because these species are common in South Korea. Five to 52 fish were collected from each reservoir. In total, 98 barbel steeds were collected from seven reservoirs (Paldang, Chungju, Daecheong, Soyang, Okjeong, Togyo, and Seonam), 91 largemouth bass were collected from five reservoirs (Paldang, Soyang, Okjeong, Togyo, and Banwol), 100 leopard mandarins were collected from seven reservoirs (Chungju, Paro, Daecheong, Soyang, Boryeong, Buan, and Angye), and 159 bluegills were collected from eight reservoirs (Paldang, Chungju, Daecheong, Okjeong, Hoeya, Sueo, Togyo, and Banwol). After collection, the fish were weighed, measured for length, and then filleted. Fillet samples were lyophilized, pulverized, and homogenized using a stainless steel blender. The general habitat and feeding type of each species are summarized in Table S1, and mean lengths and weights are presented in Table S2.

Surface water samples were collected from 14 reservoirs in the summers (June–September) from 2013 to 2015. Surface water samples were collected from a water depth of 0.5 m at each reservoir using thorough "clean hands and dirty hands" protocols to avoid Hg contamination. Unfiltered water was collected by a peristaltic pump through Teflon® tubing from the water surface. Dissolved THg and MMHg samples were filtered on-site through 0.45 μm capsule filters (polyether sulfone membrane, Millipore®, Germany) connected to the peristaltic pump. Hg samples were collected in pre-cleaned Teflon® bottles with diluted nitric acid (20%) and hydrochloric acid (15%). Samples were preserved with hydrochloric acid (0.4% v/v) within a few hours of collection and kept at 4 °C until analysis. To avoid sample contamination, field and equipment blanks were prepared with Milli-Q® water at each reservoir. The concentrations of particulate THg and MMHg were determined from the differences between the unfiltered THg (or unfiltered MMHg) and dissolved THg (or dissolved MMHg), normalized to the SS concentration.

Surface water measurements (i.e., temperature, conductivity, and dissolved oxygen) were carried out on-site using a portable water quality probe (YSI Inc., USA). The unfiltered water samples were stored in polyethylene bottles so that pH, SS, and chl-a concentrations could be determined in the laboratory. Filtered water samples for measuring dissolved organic carbon (DOC) and sulfate concentrations were immediately transferred into acid-cleaned and pre-combusted borosilicate glass bottles and were frozen until analysis.

2.2. Analysis of SS, chl-a, DOC, and sulfate

The SS, chl-a, DOC, and sulfate in the water samples were analyzed as described by Noh et al. (2016). The water samples were filtered gently through dried and pre-weighed 0.4 μm Whatman® polycarbonate membranes to determine SS concentration and through 0.7 μm Whatman® GF/F filters to determine chl-a concentration. Particle-bearing membranes were dried at 105 °C for 2 h, and the SS loads were measured. The chl-a retained on the GF/F filters was extracted in 90% acetone for 12 h in dark conditions and sonicated for 10 min. The solution containing extracted chl-a was centrifuged to remove glass filter debris, and then the absorbance of the supernatant was measured at wavelengths of 750, 665, 645, 630, and 480 nm using an ultraviolet-visible spectrophotometer (Optizen, Korea). The DOC samples were acidified and sparged with ultrapure oxygen gas to remove inorganic carbon, after which DOC was determined with a TOC analyzer (Elementar, Germany). Sulfate concentration was determined by ion chromatography (Dionex, Thermo Scientific, USA).

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