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Trophic transfer of mercury and methylmercury in an aquatic ecosystem impacted by municipal sewage effluents in Beijing, China

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Abstract

Gaobeidian Lake, located in Beijing, China, serves as a recipient lake for effluents from a large municipal sewage treatment plant (MSTP). In order to evaluate the effects of discharging MSTP effluent on the mercury contamination of the local aquatic ecosystem, sediment cores, water, plankton, fish, and turtle samples were collected from Gaobeidian Lake for mercury speciation analysis. High concentrations of total mercury (T-Hg) were detected in sediment cores (5.24–17.0 μg/g dry weight (dw), average: 10.1 μg/g). The ratio of methylmercury (MeHg) to T-Hg was less than 0.3% in sediments and ranged from 35% to 76% in biota samples. The highest level of T-Hg and MeHg were found in aquatic bryophyte and crucian carp (3673 and 437 ng/g dw, respectively). The relative contents of MeHg were significantly correlated with trophic levels ($R^2 = 0.5506$, $p < 0.001$), which confirmed that MeHg can be bio-transferred and biomagnified via food chain in this aquatic ecosystem.

Key words: methylmercury; total mercury; trophic levels; aquatic ecosystem; municipal sewage treatment plants

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Introduction

Mercury is a toxic element and persistent contaminant in environment. Inorganic mercury can be methylated to methylmercury (MeHg) through both biotic and abiotic pathways (UNEP, 2002). Mercury contamination is not only a problem to aquatic ecosystems but also to terrestrial animals occupying higher trophic levels and may eventually lead to an imbalance of adjacent ecosystems (Cristol et al., 2008). MeHg can enter and be accumulated in food webs, which results in high concentrations in fish and other top predators such as piscivorous birds (Langston 1990; Orihel et al., 2007). High concentrations of mercury/MeHg in birds can affect immune, detoxification, nervous systems, and even their reproductive capacity (Clarkson 1990; Harada, 1995; Morel et al., 1998).

The applications of mercuric compounds have decreased significantly during the last 30 years, but they are still used in many household goods such as skin-lightening creams and germicidal soaps (Weldon et al., 2000). The residuals of mercury in these products can eventually reach the municipal sewage treatment plants (MSTP) via the disposal of domestic wastewater. Wastewater from hospitals is another possible source of mercury to MSTPs (Arenholt-Bindslev and Larsen, 1996; Lan et al., 2007). Besides, atmospheric deposition such as dust from combustion of coal in power plants may be a potential source of Hg contamination (Pacyna et al., 2006).

MSTP can efficiently remove a large amount of pollutants, especially biodegradable organics and solid wastes. However, some persistent contaminants in wastewater, such as persistent organic pollutants (POPs) and heavy metals, can be readily concentrated into the sewage sludge and effluents and re-enter to the environment via the disposal of effluents and sewage sludge from MSTPs. Previous studies have proved that about 4% of polybrominated diphenyl ethers (PBDEs) and Hg entering the treatment plant was discharged to the ambient environment through the effluent disposal (North, 2004; Balogh and Liang, 1995). Other studies also pointed out that MSTP effluents could be a potential point source of these persistent contaminants in the recipient water and might pose a threat to the nearby ecosystem (Karvelas et al., 2003; Barber et al., 2006; Katsoyiannis and Samara, 2007).

There are around 400 sewage treatment plants that have so far been built in China. The large amount of MSTP effluents discharged to ambient environment may pose ecological risks to aquatic ecosystems. Wang et al. (2007) found that PCBs and PBDEs accumulated in biota samples from this studied Gaobeidian area. Most researchers emphasize on the health effects of MSTP outputs and only a few studies investigated the heavy metal pollution in the effluent recipient water column (Chang et al., 2007).

In the present work, we analyzed the mercury speciation in lake water, sediment cores, and a section of the aquatic

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food web (including plankton, fish and turtle) samples collected from Gaobeidian Lake. The effect of discharging MSTP effluent on the bioaccumulation and biomagnifications of mercury in the local aquatic ecosystem was evaluated and discussed. To our knowledge, this is the first study focusing on Hg concentrations and speciation distribution in a large MSTP effluent recipient lake.

1 Materials and methods

1.1 Sampling site and sample collection

Gaobeidian Lake is located in the east of Beijing, China (Fig. 1). Its water source is mostly from Gaobeidian MSTP effluent. The Gaobeidian MSTP is the biggest MSTP in Beijing, which can treat approximate one million tons of wastewater per day. About 30% of Gaobeidian MSTP effluent was discharged into the recipient water lake. The temperature of this lake water was relatively high because part of the water input of this lake were cooling water from a thermoelectric power plant (from May to October in 2006 the temperature was constantly above 30°C with the highest 45°C). Some other physicochemical and water quality parameters for Gaobeidian Lake were as follows: chemical oxygen demand (COD_{Cr}) (46.3 ± 6.3) mg/L, pH 7.7 ± 0.1, suspended solid (SS) (16.0 ± 3.4) mg/L, total phosphorus (TP) (2.3 ± 0.7) mg/L, and total nitrogen (TN) (27.8 ± 4.4) mg/L (Wang et al., 2008).

In this study, sediments, water and a part of the aquatic food web samples were collected from Gaobeidian Lake during September and December, 2006. Two sediment cores (S1 and S2) were taken from Gaobeidian Lake using a special sediment core sampler. Cores S1 and S2 were collected about 36 cm and 40 cm in depth, respectively. The cores were sectioned at 4-cm intervals using a stainless steel blade. Sectioned sediment samples were packed by aluminum foil in sealed plastic bags. Biota samples included primary producers spirogyra and march brown (*Limnodrilus hoffmeisteri*), zooplankton (*Monia rectirostris*, *Monia micrura*, and *Monia macrocopa* as the dominant species), common carp (*Cyprinus carpio*) (*n* = 4), crucian carp (*Carassius auratus*) (*n* = 5), leather catfish (*Silurus meridionalis*) (*n* = 4), java tilapia (*Tilapia nilotica*) (*n* = 3), and Chinese softshell turtle (*Chinemys reevesii*) (*n* = 3). Aquatic species were stored in ice-box after collection and immediately transported to the laboratory. The weight, length and age of biota samples were measured and identified.

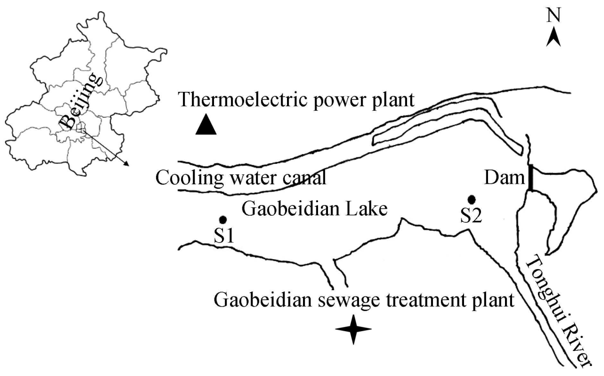

Fig. 1 Location and surroundings of Gaobeidian Lake.

The muscle tissue of aquatic samples was separated and homogenized by an analytical mill. Both the sediment and biota samples were dried at –52°C and a pressure less than 5×10^3 Pa for 48 hr, and subsequently grinded to fine powder. The samples were then sealed in polyethylene bottles and stored in a refrigerator until analysis. All contents in this study are expressed on a dry weight (dw) basis.

1.2 Trophic levels calculation

The trophic levels (TLs) of the selected aquatic organisms were measured based on nitrogen isotope ratios. Because of the preferential excretion of lighter nitrogen isotopes, $\delta^{15}N$ has been suggested to be a useful empirical measure of trophic status, as $\delta^{15}N$ increases with trophic levels (Minawaga and Wada, 1984; Cabana and Rasmussen, 1994). Nitrogen stable isotopes analysis followed previously established method (Hu et al., 2005), and $\delta^{15}N$ was calculated from the following formula:

$$\delta^{15}N = ({}^{15}N/{}^{14}N_{sample}/{}^{15}N/{}^{14}N_{standard} - 1) \times 1000‰ \quad (1)$$

where, ${}^{15}N/{}^{14}N_{standard}$ values are based on atmospheric N_2.

Trophic levels of aquatic organism were calculated using the equation described by Fisk et al (2001):

$$TL_{consumer} = (\delta^{15}N_{consumer} - \delta^{15}N/zooplankton)/3.8 + 2 \quad (2)$$

where, $TL_{consumer}$ is the trophic level, and the trophic level of zooplankton was assumed to be 2.

1.3 Chemical analysis

The total mercury (T-Hg) and MeHg content in biota and sediment samples were analyzed following the method described by Liang et al. (2003) and Shi et al. (2005), respectively. T-Hg in water was detected by direct injection into atomic fluorescence spectrometer, and pretreatment of MeHg analysis in water was similar to biota sample without digestion. T-Hg was detected using an AF-620 non-dispersive atomic fluorescence spectrometer (Beijing Rayleigh Analytical Instrument Co., China) after hydride generation. MeHg content analysis was conducted by an HPLC-UV-AFS system.

Through the analytical procedure, reagent blanks were processed simultaneously to deduce the error and analyses of spiked matrix and certified reference materials (DORM-2) was also processed for validation. Recoveries of T-Hg and Me-Hg in the spiked sediments were in the range of 80%–110%. The determined results of T-Hg and MeHg in DORM-2 were (4.71 ± 0.40) and (4.45 ± 0.34) μg/g dw, respectively, which was in agreement with the certified values ((4.64 ± 0.26) μg/g dw for T-Hg and (4.47 ± 0.32) μg/g dw for MeHg), confirming the feasibility of the analytical process.

2 Results and discussion

2.1 TLs of organisms from Gaobeidian Lake

TLs of the samples were converted from $\delta^{15}N$ values according to Eq. (2). TLs of these biota samples ranged from 2.00 to 3.97, and the Chinese softshell turtle samples ranged from 3.33 to 3.84. According to their food habits, java tilapia, common carp and crucian carp belong to omnivorous fish whereas leather catfish and Chinese softshell turtle are piscivorous, thus the latter two species should have higher TLs than other species. The results, however, showed that java tilapia had the highest TLs (average 3.94), while common carp had the widest range of TLs (2.00–2.96) despite of their similar age (all about 3 years old). The abnormal TLs indicated that in addition to their food habits, the MSTP effluents may also influence the metabolism of these aquatic species. Muir et al. (2003) presumed that $\delta^{15}N$ can be influenced by municipal waste sources with individuals differing by up to 0.3% in $\delta^{15}N$.

2.2 Mercury concentration and speciation in sediment

T-Hg concentrations in sediment cores ranged from 5.24 to 17.0 μg/g dw with an average concentration of 10.1 and 10.2 μg/g dw, respectively, for core S1 and S2 (Fig. 2). The results are just at the critical level (10.0 μg/g dw) set by the European Union. The vertical distributions of T-Hg showed no obvious correlation with the sediment depth (Fig. 2). The concentration of MeHg ranged from 1.40 to 18.1 ng/g in S1, and < limit of detection (LOD, 1 ng/g) to 6.35 ng/g in S2, with the average of 5.34 ng/g and 2.96 ng/g, respectively. Most MeHg concentrations in different metrics were 0.1% or less, except for a few individual samples which have relative higher MeHg concentrations but accounted for no more than 0.3% of T-Hg. Although some researchers reported higher percentage (37%) of MeHg in sediment (Houserová et al., 2007), the present result was similar to MeHg levels in River Nura measured by Ullrich et al (2007) and those from many other areas in the world (Kannan et al., 1998; Bloom et al., 1999; Horvat et al. 2003). Based on the investigation by Lan et al. (2007), Hg concentration in influent of this MSTPs fell in the range of 0.1–4.0 ng/mL. The referred study pointed out that most Hg in the influent was transferred to the sludge, and the concentration in the effluents of this study was below LOD (< 0.05 ng/mL). However, it is inevitable that a small amount of Hg would still be present in the effluent and enter the receiving water body through the drainage of effluent. This might be the most important source of Hg in the sediment of Gaobeidian Lake. Because combustion of coal in power plants is one of the largest anthropogenic sources of Hg (Pacyna et al., 2006), the power plant located nearby Gaobeidian MSTP may be an another source of Hg.

This work revealed that T-Hg concentration were relatively constant (mostly between 8.00 and 12.0 μg/g) along the depth profile of the sediment cores (Fig. 2). This may reflect that Hg input and deposition in this lake was relatively stable during recent years. The relationship between percentage of MeHg and concentration of MeHg in the two sediment cores was also investigated and the results revealed a linear increasing trend (R^2 = 0.9101, $p <$ 0.001) (Fig. 3), which implied that MeHg was the primary factor of MeHg relative contents in the sediments. This was in agreement with the finding by Kannan et al. (1998). No significant correlation was found between T-Hg and MeHg concentration in sediments ($p > 0.05$), which reflects a fact that T-Hg concentration is not the only factor influencing Hg methylation. Factors affecting mercury methylation are decisive for the MeHg concentration in sediments, of which sulfate-reducing bacteria is the most important one. However, SRB communities are usually not uniformly distributed in sediments because of environment conditions such as nutrient and redox conditions. Because of the speciality of this lake, there also might have disturbance during recent years, therefore the explanation of mercury vertical distribution in sediment cores was complex.

2.3 Mercury speciation in biota

Ethyl- and phenyl-mercury are currently used in industry. Ethyl-mercury is one of the metabolites of thiomersal, which is used as a preservative in some vaccines. Phenylmercuric acetate is an organo-mercury compound used as a fungicide. MSTP effluent is a rather complex matrix, and we tried to determine whether these species existed in this ecosystem. However, among the all samples, only organic mercury species found was MeHg, which was in

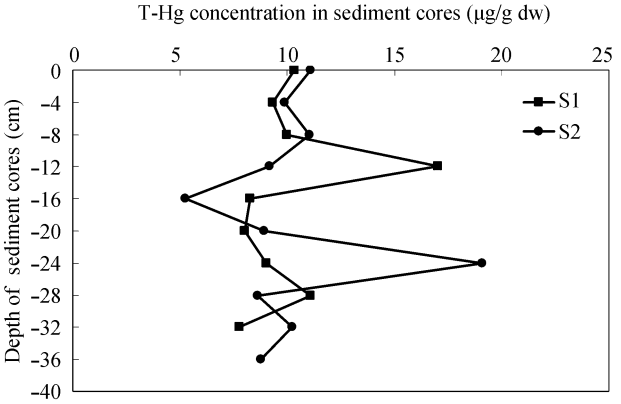

Fig. 2 Vertical distribution of total mercury concentrations in sediment cores from Gaobeidian Lake.

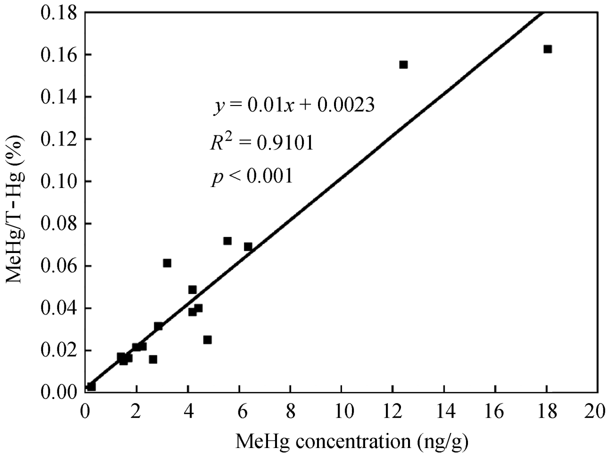

Fig. 3 Relation between the percentage of MeHg to MeHg in sediments.

consistent with previous studies (Houserová et al., 2007; Liang et al., 1994). The highest level of T-Hg was found in aquatic bryophyte (3673 ng/g). A possible reason is that bryophytes are phytoplanktons that have a body with high surface-to-volume ratio and are frequently absence of cuticle, and thus has a high capacity to accumulate metals. Previous work also proved that bryophytes can be used as bioindicators of environmental pollution (Carballeira et al., 2001). The MeHg concentration in aquatic bryophyte and zooplankton was below LOD (5 ng/g). Our work also implies that aquatic bryophyte is a good indicator for T-Hg concentration in this aquatic ecosystem but not for MeHg.

The concentrations of T-Hg and MeHg in fish samples were in the range of 125–580 ng/g dw and 48.7–438 ng/g dw, respectively. Significant correlations were obtained between T-Hg and MeHg concentrations in biota samples except for aquatic bryophyte and zooplankton ($R^2 = 0.9194$, $p < 0.001$) (Fig. 4). The highest levels of T-Hg and MeHg were found in a crucian carp sample (579 and 438 ng/g, respectively). On a wet weight basis, the average T-Hg and MeHg concentration in fish muscles were 99.5 and 59.1 ng/g, respectively. None of the T-Hg concentrations in these samples exceeded the maximum permissible concentration of Hg in fresh water fish in both China and Europe (300 ng/g wet weight). MeHg concentrations also did not exceed the Chinese tolerance limit of MeHg in fish (200 ng/g wet weight) (GB2762-94). Compared to other similar Hg contaminated areas, T-Hg and MeHg contents in fish in this lake were at relatively low levels (Ullrich et al., 2007). Sorensen et al. (1990) also reported that T-Hg in sediments from unstratified lakes did not correlate with fish Hg concentrations. Mercury accumulation in biota depends on bioavailable Hg in their living environment (water, sediments) which is influenced by factors such as organic carbon, sulfide, aerobic/anaerobic conditions and acidity of the water bodies. Sulfide and organic contents in sediment have a strong affinity for heavy metals. Hg has a high affinity for sulfide, with which HgS can be formed. Once HgS is formed, the bioavailability of sedimentary Hg sharply decreased. The organic contents in sediment could form chelate complexes with Hg which also decrease the bioavailability of sedimentary Hg. Wang et al. (2007) found high level of sulfide contents in these two cores by gel permeation chromatography. The average total organic carbon content in collected sediment cores was 11%. Furthermore, the pH of Gaobeidian Lake was 7.7 ± 0.1. Haines et al. (1992) reported that alkaline surface water can decrease the uptake of MeHg. In addition, the water temperature of Gaobeidian Lake was relatively high (between 12°C and 45°C, 5–10°C higher than ambient temperature). As well known, high temperature of the surrounding environment would enhance the metabolism of the biota, thus Hg and MeHg would be more rapidly eliminated from biota. From the results of this study, we could conclude that Hg contamination in Gaobeidian Lake aquatic biota was not very significant. However, the aquatic Hg could be transferred to terrestrial food webs by emergent aquatic insects and might pose risks to terrestrial ecosystems (Cristol et al., 2008). The sediment was a potential source of Hg, and should also be paid special attention.

Muscle tissue of piscivorous biota samples (leather catfish and Chinese softshell turtle) contained higher contents of T-Hg and MeHg than omnivorous fishes (common carp, java tilapia) except for crucian carp (Fig. 5), because Hg can accumulate through food chain. Age and growth rates of the fish could also affect Hg concentration in fish muscle. We measured the age of fish samples and found they are in the range of 1–4 years. No obvious relationship was found between age and the content of Hg in muscle. It is known that common carp, java tilapia, and leather catfish grow much faster than crucian carp, and Hg concentration can be diluted because of faster growth rates. Jin et al. (2006) found that mercury concentration in crucian carp was significant higher than that in common carp collected from Ya-er Lake (423 ng/g vs. 78.9 ng/g in T-Hg and 185 ng/g vs. 39.3 ng/g in MeHg), which is in consistent with our result.

Ratio of MeHg to T-Hg in the muscle of biota samples ranged from 35% to 76%, and this result was in the range of previous works (Jin et al., 2006; Houserová et al., 2007; Ecosystem Health, 2002). In all biota samples, the relative

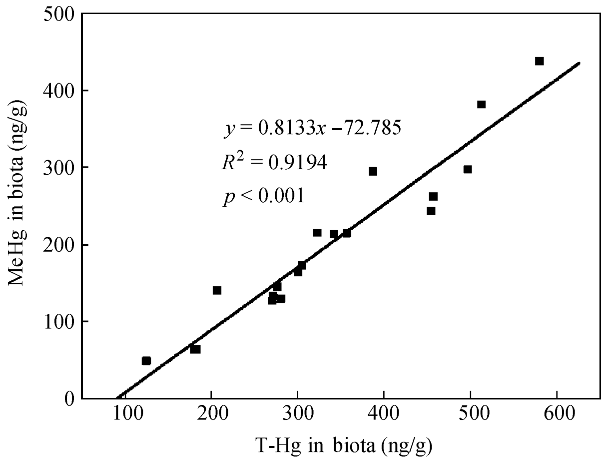

Fig. 4 Relation between the concentrations of total mercury and methyl mercury in fish muscles.

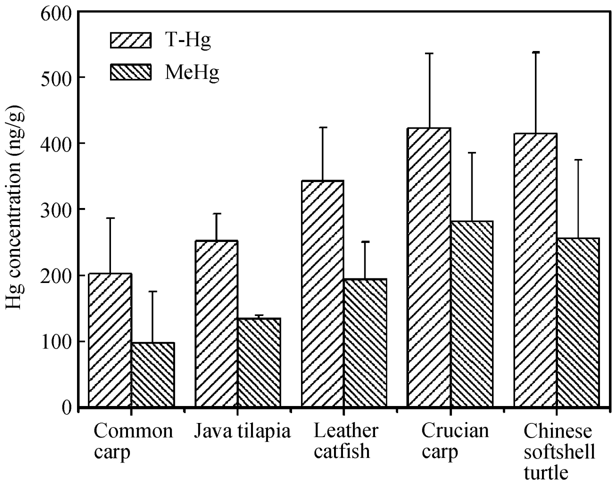

Fig. 5 Content of total mercury and methylmercury in the fish muscles.

contents of MeHg was positively correlated with trophic levels (R^2 = 0.5506, p < 0.001) (Fig. 6). The similar phenomenon in a river ecosystem affected by mercury mining was observed by Žižek et al. (2007). It indicating that MeHg accumulated along with trophic levels in this aquatic ecosystem.

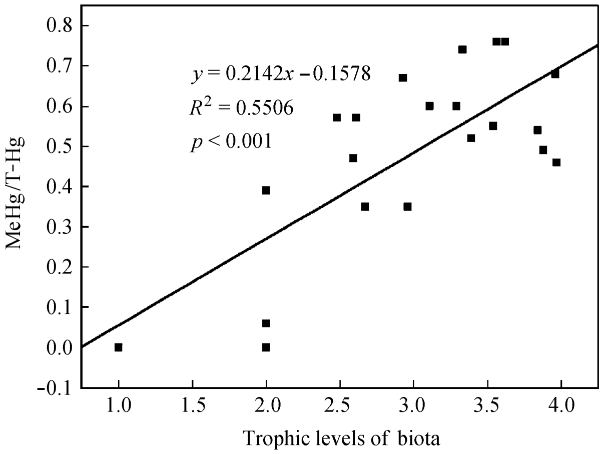

Fig. 6 Relation between the relative methylmercury content and trophic levels in biota samples.

3 Conclusions

The speciation of mercury in sediment cores and a part of food web from Gaobeidian Lake were investigated. MeHg was found in biota, while ethyl- or phenyl-mercury were undetectable. Relatively high Hg contents in sediments indicated that the sediment of Gaobeidian Lake was probable contaminated by the effluent from the MSTP. The highest T-Hg level lead aquatic bryophytes a good biomarker for T-Hg in aquatic ecosystem but not for MeHg. There were significant positive correlations between T-Hg and MeHg concentrations in fish samples. Although Hg and MeHg in Gaobeidian Lake sediments had very low bioavailability in the aquatic ecosystem under the recent circumstance (high sulfide contents, high TOC, high temperature), MeHg can still be accumulated along with trophic levels in this aquatic ecosystem. It also demonstrated that under some circumstance, the sedimentary Hg was not correlated with Hg in fish muscle. Although current Hg contamination in Gaobeidian Lake aquatic biota is not serious, aquatic Hg can move into terrestrial ecosystems through the food chain and high Hg contents in the sediment should still be paid attention to.

Acknowledgments

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