

## SPATIAL AND TEMPORAL VARIATIONS OF TOTAL AND METHYLMERCURY CONCENTRATIONS IN PLANKTON FROM A MERCURY-CONTAMINATED AND EUTROPHIC RESERVOIR IN GUIZHOU PROVINCE, CHINA

QING WANG,<sup>†</sup>§ XINBIN FENG,<sup>\*†</sup> YUFENG YANG,<sup>†</sup>§ and HAIYU YAN<sup>‡</sup><sup>†</sup>Institute of Hydrobiology, Jinan University, Guangzhou, China<sup>‡</sup>State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang, China<sup>§</sup>Key Laboratory of Aquatic Eutrophication and Control of Harmful Algal Blooms, Guangdong Higher Education Institutes, Guangzhou, China

(Submitted 25 May 2011; Returned for Revision 4 July 2011; Accepted 7 September 2011)

**Abstract**—Total mercury (THg) and methylmercury (MeHg) concentrations in four size fractions of plankton from three sampling stations in the Hg-contaminated and eutrophic Baihua Reservoir, Guizhou, China, were investigated for biomagnification and trophic transfer of Hg at different sites with various proximity to the major point sources of nutrients and metals. Total Hg concentrations in plankton of the various size fractions varied from 49 to 5,504 ng g<sup>-1</sup> and MeHg concentrations ranged from 3 to 101 ng g<sup>-1</sup>. The percentage of Hg as MeHg varied from 0.16 to 70%. Total Hg and MeHg concentrations in plankton samples differed among the three sampling stations with different proximities from the major point sources. The plankton from the site closest to the dam contained the highest concentrations of MeHg. The successive increase of the ratios of MeHg to Hg from seston to macroplankton at all sites indicated that biomagnification is occurring along the plankton food web. However, biomagnification factors (BMF) for MeHg were low (1.5–2.0) between trophic levels. Concentrations of THg in seston decreased with an increase of chlorophyll concentrations, suggesting a significant dilution effect by the algae bloom for Hg. Eutrophication dilution may be a reason for lower MeHg accumulation by the four size classes of plankton in this Hg-contaminated reservoir. Environ. Toxicol. Chem. 2011;30:2739–2747. © 2011 SETAC

**Keywords**—Mercury Plankton Biomagnification Eutrophication Site-specific

## INTRODUCTION

Primary consumers assimilate organic and inorganic compounds from their diet for their own biosynthesis, somatic growth, reproduction, and repair. In doing so, they also incorporate contaminants that serve no beneficial physiological purpose [1]. One potentially toxic and ubiquitous contaminant is mercury (Hg). The methylated form (MeHg), a neurotoxin, is bioavailable to aquatic organisms and can cause severe health problems, including neurological [2] and cardiovascular damage [3,4] to humans. Since the late 1970s, Hg contamination in the biota of many freshwater aquatic systems has received great attention worldwide. Mercury distribution in biota is governed by a number of physical, chemical, and biological processes [4].

Because they are the entry point of Hg into the aquatic food web, plankton play a key role in Hg bioaccumulation [5,6]. Moreover, the planktonic food web plays an important ecological and ecotoxicological role in conveying both nutrients and contaminants to organisms at higher trophic levels [7]. Previous studies have suggested that diet is the major pathway that conveys MeHg to zooplankton [8,9] and fish [10]. Methylmercury concentrations increase as the body size of zooplankton increases [4,11]. As a result, macrozooplankton, the preferred prey size for planktivorous fish [12], transfers the highest MeHg concentrations to the higher trophic levels.

Until now, most field studies of the trophic transfer of MeHg in aquatic systems have been conducted in North America and

Europe [13–18]. Furthermore, many previous studies were conducted in oligotrophic waters that have low primary productivity and concentrations of phosphorus, nitrogen, and chlorophyll *a* [7,19]. Because of rapid economic growth, human activities have released a large amount of Hg and nutrients into the environment to the degree that some aquatic systems are seriously eutrophic and contaminated with Hg. A limited number of studies exist on the transfer of Hg and/or MeHg in planktonic food web in aquatic systems in China. Therefore, it is urgent to investigate how MeHg is conveyed through the planktonic food web, especially in aquatic systems in China that have serious Hg contamination.

To investigate the pattern of MeHg concentrations with an increase of plankton size under different hydrodynamic conditions, a field study was performed in a eutrophic reservoir in a karst region. The aim was to describe the accumulation patterns of total Hg (THg) and MeHg in the planktonic food web as well as to provide details on their association with increased body size of plankton relative to eutrophic conditions and proximity to pollution sources.

## MATERIALS AND METHODS

*Study area*

Guizhou province (N24°30′–29°13′, E103°1′–109°30′) in southwestern China is one of the most significant sources of Hg in the world [20,21]. It is also a typical subtropical karst region. The Baihua Reservoir, located 16 km northwest of Guiyan City, the capital of Guizhou Province (Fig. 1, location: N26°35′–26°42′, E106°27′–106°34′), has a surface area of approximately 14.5 km<sup>2</sup>, a maximum water storage capacity of 1.82 × 10<sup>8</sup> m<sup>3</sup>, with a maximum depth of approximately 45 m and a mean depth of 12.5 m [22]. It is long and narrow (length

All Supplemental Data may be found in the online version of this article.

\* To whom correspondence may be addressed

(fengxinbin@vip.skleg.cn).

Published online 20 September 2011 in Wiley Online Library  
(wileyonlinelibrary.com).

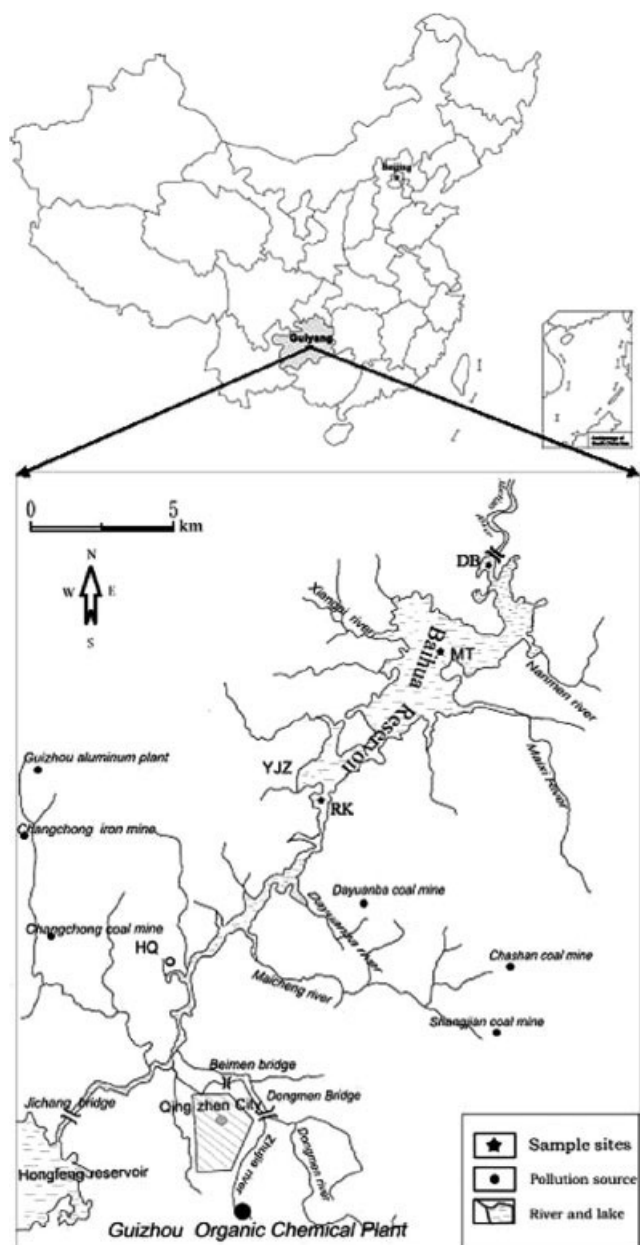


Fig. 1. Location of the sampling sites in the Baihua Reservoir, China (RK [upstream], MT [middle], and DB [downstream]). The direction of the water flow is from RK to DB. Guizhou Organic Chemical Plant, Guizhou aluminum plant, one iron mine, and seven small coal mines are situated at the upper reaches of the reservoir. RK is located at the inlet of the reservoir; MT is located in the middle part of the reservoir; DB is located near the dam of the reservoir.

18 km, mean width 0.8 km) with a single main input from the river and only one output. The reservoir is used for agricultural irrigation, fishing, and recreation. The Baihua Reservoir, which is drained by a 1,895-km<sup>2</sup> catchment area, was built in 1962. The residence time of the reservoir is approximately 37 d. The climate in this region represents a typical subtropical rainy monsoon with an annual mean temperature of 13.8°C. The highest water temperature was 28°C with an average temperature of 23°C in summer, while the lowest water temperature was 4°C with an average temperature of 10°C in winter. The average water temperatures in spring and autumn are 16°C and 14°C, respectively. The annual average precipitation is approximately 1,175 mm. The average total nitrogen and total

phosphorus concentration is 2.01 mg L<sup>-1</sup> and 0.02 mg L<sup>-1</sup>, respectively. Sulfate concentration in surface water is 76.8 mg L<sup>-1</sup>. The dissolved oxygen in surface water is 6.9 to 9.4 mg L<sup>-1</sup>, and less than 4.0 mg L<sup>-1</sup> in the bottom. An anoxic condition occurs in the bottom water between May and September because of algae bloom.

The terrain around the Baihua Reservoir is highly vegetated. The Guizhou Organic Chemical Plant (GOCP), located at the upper reach of the reservoir, was the only one in China that used metallic Hg as a catalyst to produce acetic acid (Fig. 1). Between 1971 and 1997, the reservoir experienced serious Hg contamination from GOCP. From 1971 to 1985, the plant consumed 573 metric tons of Hg [22]; the wastewater was directly discharged into Zhujia River, which flows into the Baihua Reservoir, and seriously contaminated it. At present the THg concentrations in Zhujia river still range from 250 to 1,000 ng L<sup>-1</sup> [22]. Because of the development of industries and the increase in population, the Baihua Reservoir receives a considerable amount of untreated domestic and industrial sewage. In addition, an iron mine, the Guizhou Aluminum Plant, and seven small coal mines are situated at the upper reaches of the reservoir and may be further sources of Hg contamination [22].

Mercury concentrations in the sediment of the Baihua Reservoir are high. Total mercury (THg) and MeHg concentrations range from 0.26 to 38.9 mg kg<sup>-1</sup> with an average concentration of 12.9 mg kg<sup>-1</sup> (dry wt) and from 0.5 to 27.5 µg kg<sup>-1</sup> with mean 6.1 µg kg<sup>-1</sup> (dry wt), respectively [22]. The average concentrations and ranges of Hg species in the water column in the Baihua Reservoir are 22.4 ng L<sup>-1</sup> and 7.1 to 152.8 ng L<sup>-1</sup> for THg, 2.52 ng L<sup>-1</sup> and 0.3 to 12.2 ng L<sup>-1</sup> for reactive Hg, 1.29 ng L<sup>-1</sup> and 0.11 to 10.2 ng L<sup>-1</sup> for total MeHg [23]. In the water column and sediment profiles, the distribution of THg showed decreasing trends with increasing distance from the pollution source [22,23]. However, fish have a low level of THg concentration, with an average of 28 ng g<sup>-1</sup> for carp [24]. Among the fish species collected, the minimum mean concentration of THg (21 ng g<sup>-1</sup>) was observed in silver carp (*Hypophthalmichthys molitrix*), a planktivorous species. The maximum mean concentration (47 ng g<sup>-1</sup>) was observed in bighead carp (*Aristichthys nobilis*), a planktivorous species. The range of THg in the two species was 17 to 143 ng g<sup>-1</sup> and the range of the body size was 23.5 to 50.0 cm. No significant correlation existed between THg concentration and body size [24]. This is simply because all fish sampled were aquaculture fish; because these are nonpredatory, they have a simple food chain and a low bioaccumulation rate of Hg [23,24].

#### Sample collection

Samples were taken from November 2008 to January 2009 from three sites, designated RK (upstream), MT (middle), and DB (downstream). They are located at varying distances from the wastewater discharge sources via the water channel, the closest to RK, which connects with the Hongfeng Reservoir (Fig. 1). Samples were taken twice a month at these sites during the daytime using a conical net of 64 µm mesh. The organisms were rinsed with filtered (0.45 µm porosity) reservoir water to remove adhered particulate matter and filtered through Nitex sieves of 610, 216, 108, and 38 µm mesh size to obtain four size fractions: 38 to 108, 108 to 216, 216 to 610, and >610 µm. Samples collected for zooplankton identification as well as for Hg and MeHg determination were separated into four fractions as described above. Samples for zooplankton identification were stored in vials and fixed in 4% formalin solution, while

samples for THg and MeHg analyses were kept frozen until lyophilization, then again stored frozen for analysis.

Temperature and pH profiles were measured using a probe (pIONeer 65). To quantify changes in trophic status, algal biomass (chlorophyll *a* = Chl *a*) was measured during each sampling campaign [25].

#### Methyl and THg analysis

Bulk plankton samples were freeze-dried and then ground to powder with a glass rod directly into a vial to reduce handling and the risk of Hg contamination. Total mercury concentration was determined by cold vapor atomic fluorescence spectrophotometry [26]. Briefly, 1 to 5 mg (dry weight) plankton aliquots were transferred to glass tubes and digested in 1 ml of a 10-HNO<sub>3</sub>:1 HCl mixture for 6 h at 120°C to ensure that all Hg was converted into and kept in the nonvolatile oxidized state (Hg<sup>2+</sup>). Mercury was then reduced to elemental Hg (Hg<sup>0</sup>) vapor by a stannous chloride (SnCl<sub>2</sub>) solution in a reaction vessel and analyzed by cold vapor atomic fluorescence spectrophotometry. Methylmercury was measured using GC-CVAFS detection (Glas-Col TM568; Tekran Model 2500). For MeHg analysis, freeze-dried plankton aliquots (5–10 mg) were homogenized and digested in 0.5 ml of a KOH/MeOH (1 g 4 ml<sup>-1</sup>) solution for 8 h at 68°C [26].

The detection limits for our methods were approximately 13 and 2 pg g<sup>-1</sup> of THg and MeHg, respectively. Precision and accuracy were tested by triplicate analysis of the standard reference material (TORT-2, National Research Council of Canada). The average values of THg and MeHg concentrations for TORT-2 were 0.274 ± 0.005 μg g<sup>-1</sup> and 0.155 ± 0.007 μg g<sup>-1</sup>, respectively, which are comparable to the certified values 0.27 ± 0.06 μg g<sup>-1</sup>, 0.152 ± 0.013 μg g<sup>-1</sup> for TORT-2. Blank runs contained no detectable Hg concentrations.

## RESULTS

### Reservoir characteristics, plankton species composition

The mean (±SD) water temperatures were 18.8°C (±0.9), 16.2°C (±0.2) in November 2008; 15.0°C (±0.6), 12.2°C (±0.1) in December 2008; and 10.3°C (±0.1) in January 2009. The water was alkaline (pH >7). Chlorophyll *a* concentrations, used as a proxy for algal biomass, were not significantly (*p* > 0.05) different among the three sites (Table 1). The dominant species and groups in the plankton community are listed in Table 2 according to the four size fractions.

### THg and MeHg concentrations in plankton

Mean THg and MeHg concentrations in the plankton from the three sites are presented in Table 3. The THg concentrations in plankton collected at the RK site were higher than at the other two sites (Fig. 2), whereas MeHg concentrations at DB were higher than at the other two sites (Fig. 2). The THg concentrations in seston were found to be lowest at the DB site and highest at the RK site. The THg concentrations in seston (38–108 μm) were four to 40 times greater than those in macrozooplankton (>610 μm) at the RK site (Fig. 3). Methylmercury concentrations in macrozooplankton at DB were the highest among the three sites. The MeHg concentrations in macrozooplankton (>610 μm) were two to 12 times greater than in seston (38–108 μm) at the DB site (Fig. 3). Plankton samples from the RK site had significantly higher THg concentrations than samples from the other sites (*p* < 0.05), whereas THg concentrations at MT and DB did not differ significantly from each

Table 1. Water quality characteristics in Baihua Reservoir, China (RK [upstream], MT [middle], and DB [downstream]) between November 2008 and January 2009

Site		T (°C)	pH	Chl <i>a</i> (μg L <sup>-1</sup> )	Secchi depth (m)
RK	Range	10.2–17.8	7.67–8.28	4.64–10.10	0.6–1.9
	Mean ± SD	14.08 ± 3.02	7.94 ± 0.26	7.51 ± 2.54	1.48 ± 0.51
MT	Range	10.3–19.4	7.75–8.13	6.23–14.84	1.5–2.1
	Mean ± SD	14.62 ± 3.55	7.93 ± 0.16	10.48 ± 3.90	1.86 ± 0.25
DB	Range	10.4–19.2	7.71–8.14	6.19–11.28	1.8–2.1
	Mean ± SD	14.74 ± 3.46	7.92 ± 0.16	9.58 ± 2.07	1.94 ± 0.15

T = temperature; Chl *a* = chlorophyll *a*; ± standard deviation (SD).

other (*p* > 0.05). Linear regression analysis showed that THg concentrations in lower trophic levels could be used successfully to predict THg concentrations in higher trophic levels. The THg concentrations (ng g<sup>-1</sup> dry wt) decreased significantly from seston to macrozooplankton (*R* = -0.91, *p* < 0.0001; *R* = -0.83, *p* < 0.0001) at RK and MT sites, respectively (Fig. 4). However, the negative correlation between THg concentration and plankton size was not found at the DB site.

Methylmercury concentrations (ng g<sup>-1</sup> dry weight) increased significantly from seston (14 ± 6), to micro- (24 ± 8), to meso- (47 ± 20), to macrozooplankton (67 ± 20, Fig. 3) at the DB site. The MeHg concentrations and plankton size were log-transformed, which resulted in a significant improvement in the ability to predict log-MeHg concentrations from plankton size (*R* = 0.85, *p* < 0.0001, DB site; Fig. 4). However, the mean concentrations of MeHg increased only from seston (8 ± 1) to microzooplankton (11 ± 8), and decreased gradually in the mesozooplankton size fraction (10 ± 4) and macrozooplankton (8 ± 4) at the RK site. Similarly, MeHg concentrations increased from seston (10 ± 8) to microzooplankton (15 ± 12), but decreased in the mesozooplankton (14 ± 6) and macrozooplankton (12 ± 2; Fig. 3) at the MT site. Results from linear regression analysis showed that MeHg concentrations at lower trophic levels could be used to predict MeHg concentrations at higher trophic levels.

Mercury was found to bind strongly to all particles within the planktonic food web. The particle-water partition coefficients (log<sub>10</sub>*K*<sub>d</sub>) averaged 4.42 ± 0.07, 4.68 ± 0.08, and 5.32 ± 0.30 for MeHg and averaged 5.43 ± 0.52, 4.76 ± 0.21, and 4.80 ± 0.06 for THg at sites RK, MT, and DB, respectively. The seston partition coefficient at RK was highest for THg and lowest for MeHg among the three sites (Table 3). By quantifying Hg concentrations in four ecologically relevant size fractions, we were able to determine the importance of Hg scavenging (the sum of Hg accumulation by particles due to surface adsorptive, transmembrane uptake, and ingestion by consumers) processes by each particle size among the planktonic food web. Seston made up the majority of the particulate Hg pool in site RT and had the highest average Hg concentrations (mean 3,441 ± 1,513 ng g<sup>-1</sup> dry weight).

### MeHg/THg concentration ratios in plankton

The percentage of THg as MeHg was low, except at the site closest to the dam. At RK, the percentage increased significantly from seston (0.27% ± 0.07%) to microzooplankton (1.23% ± 0.61%), mesozooplankton (2.15% ± 0.66%), and macrozooplankton (5.09% ± 1.03%); at MT, the percentage increased significantly from seston (4.06% ± 1.87%) to microzooplankton (8.34% ± 2.91%), mesozooplankton (11.36% ± 3.33%), and macrozooplankton (13.1% ± 2.23%); at DB, the

Table 2. Size fraction and dominant species composition of plankton in Baihua Reservoir, China

Size fractions	Dominant species and groups
Macrozooplankton > 610 $\mu\text{m}$	Calanoid: <i>Neodiantomus schmackeri</i> Cyclopoid: <i>Mesocyclops thermocyclopoides</i> , <i>Thermocyclops taihokuensis</i> Cladoceran: <i>Daphnia cucullata</i>
Mesozooplankton 216–610 $\mu\text{m}$	Cyclopoid: <i>M. thermocyclopoides</i> , <i>T. taihokuensis</i> Cladoceran: <i>Diaphanosoma brachyurum</i> , <i>Ceriodaphnia cornuta</i> , <i>Bosmina longirostris</i> , <i>Bosminopsis deitersi</i> Early instars of cladocerans and copepods
Microzooplankton 108–216 $\mu\text{m}$	Copepod nauplii Cladoceran: <i>B. longirostris</i> , <i>C. cornuta</i> , <i>B. deitersi</i> Rotifer: <i>Asplanchna brightwelli</i> , <i>Trichocerca</i> spp. Phytoplankton: <i>Melosira granulata</i>
Seston 38–108 $\mu\text{m}$	Rotifer: <i>Keratella cochlearis</i> , <i>Polyarthra vulgaris</i> Phytoplankton: <i>Pediastrum</i> spp., <i>Oocystis</i> spp., <i>Microcystis</i> spp., <i>Anabeana</i> spp., <i>Melosira</i> spp.

percentage increased significantly from seston (11.99%  $\pm$  1.73%) to microzooplankton (24.57%  $\pm$  1.99), mesozooplankton (34.49%  $\pm$  6.59%), and macrozooplankton (50.47%  $\pm$  4.90%). Although MeHg concentrations did not increase significantly with the increase of plankton size at RK and MT, the percentage of MeHg in the planktonic food web increased significantly from seston to macrozooplankton (Fig. 5). Variations of the measured plankton size fractions could significantly predict the ratios of MeHg/THg at all sites ( $R = 0.75$ ,  $p < 0.001$ , RK;  $R = 0.54$ ,  $p = 0.015$ , MT;  $R = 0.84$ ,  $p < 0.0001$ , DB). Concentration ratios (MeHg:THg) in macroplankton were significantly higher than those of the corresponding lower size fractions.

#### Biomagnification factors for plankton

Biomagnification factors (BMFs) for MeHg were calculated as the quotients of MeHg concentrations between the various plankton size classes and between macrozooplankton and the fish, *Aristichthys nobilis* [27], collected from the reservoir. A BMF greater than 1 indicates that MeHg is biomagnified through the food chain; if the BMF is less than 1, it is not. Methylmercury BMFs were the smallest from mesozooplankton to macroplankton (Fig. 6). The mean ( $\pm$ SD) BMFs between microzooplankton and mesozooplankton at DB sites was the highest (2.27  $\pm$  0.67) and the lowest (0.91  $\pm$  0.27) was at RK.

The statistical results showed that MeHg in each size-fraction of plankton was not significantly correlated with pH

( $p > 0.05$ ), but THg concentrations in seston were negatively correlated to the mean Chl *a* concentrations ( $R = -0.51$ ,  $p = 0.034$ ).

#### DISCUSSION

Mercury contamination in lakes, reservoirs, and other bodies of water is a worldwide problem. Although it is known that large amounts of Hg are emitted in China, relatively few studies have been conducted there on the bioaccumulation and biomagnification of Hg in the planktonic food web [28]. The Hg contamination in the Baihua Reservoir originated from discharge from a combination of point and nonpoint sources and from atmosphere deposition. The primary point sources are located upstream of the reservoir where sewage and other contaminants from industrial sources flow in. Moreover, the amount of Hg emitted from coal combustion in Guizhou Province is more than 10 tons a year, at least 50% of which is deposited in the environment of the province [29]. Atmospheric deposition is likely an additional minor source of Hg contamination into the reservoir.

#### THg and MeHg in the planktonic food web

The major Hg pollution source (GOCP) is located upstream (Fig. 1) of the canyon-shaped Baihua Reservoir. Since most Hg in wastewater is bound to particulate matter [22], higher concentrations of THg in both water and sediment are observed

Table 3. Total mercury (THg) and methylmercury (MeHg) concentrations (ng g<sup>-1</sup> dry wt) in plankton from three sampling sites in the Baihua Reservoir, China (RK [upstream], MT [middle], and DB [downstream])

Site	Size fraction	MeHg	THg				
RK	Range	Mean $\pm$ SD	Log $K_d^a$				
	38–108	6–10	8 $\pm$ 1	4.4	1680–5504	3441 $\pm$ 1513	6.0
	108–216	3–25	11 $\pm$ 8	4.5	683–2032	1169 $\pm$ 514	5.6
	216–610	6–15	10 $\pm$ 4	4.5	281–913	594 $\pm$ 285	5.3
	> 610	4–14	8 $\pm$ 4	4.4	81–390	196 $\pm$ 125	4.8
	Mean $\pm$ SD		4.42 $\pm$ 0.07			5.43 $\pm$ 0.52	
MT	Range	Mean $\pm$ SD	Log $K_d^a$				
	38–108	3–22	10 $\pm$ 7	4.6	167–525	297 $\pm$ 142	5.0
	108–216	5–35	15 $\pm$ 12	4.8	138–208	177 $\pm$ 26	4.8
	216–610	6–20	14 $\pm$ 6	4.7	87–158	138 $\pm$ 3	4.7
	> 610	9–15	12 $\pm$ 2	4.7	67–112	93 $\pm$ 18	4.5
	Mean $\pm$ SD		4.68 $\pm$ 0.08			4.76 $\pm$ 0.21	
DB	Range	Mean $\pm$ SD	Log $K_d^a$				
	38–108	9–24	14 $\pm$ 6	5.0	50–172	121 $\pm$ 45	4.8
	108–216	15–32	24 $\pm$ 8	5.2	49–128	100 $\pm$ 33	4.7
	216–610	15–67	47 $\pm$ 20	5.5	123–159	135 $\pm$ 15	4.8
	> 610	53–101	67 $\pm$ 20	5.6	110–155	133 $\pm$ 20	4.8
	Mean $\pm$ SD		5.32 $\pm$ 0.30			4.80 $\pm$ 0.06	

<sup>a</sup>  $K_d$ : particle-water partition coefficient, [particulate MeHg or THg (ng kg<sup>-1</sup>)]/[dissolved MeHg or THg (ng L<sup>-1</sup>)]. The dissolved MeHg and THg concentrations at the three sites were from unpublished data measured in 2009.

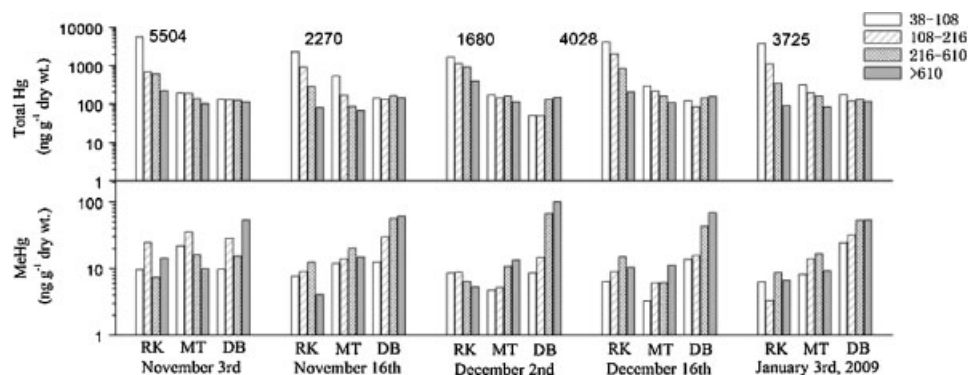


Fig. 2. Total mercury (THg) and methylmercury (MeHg) concentrations ( $\text{ng g}^{-1}$  dry wt) in plankton collected from the three sites in the Baihua Reservoir, China (RK [upstream], MT [middle], and DB [downstream]).

upstream. Given its proximity to the sources, the RK site was expected to have the highest concentrations of Hg in both water and biota. This pattern was observed in the Baihua Reservoir, where a declining trend was observed in plankton THg concentrations from upstream to downstream. The THg concentrations in the same size fraction of plankton at RK and MT sites were in general higher than those at the DB site. The highest concentrations were found at the RK site, which was heavily affected by wastewater discharged from GOCP. Results for THg distribution clearly indicate that the THg concentrations in plankton rapidly decrease with increasing distance from the chemical plant. The highest concentrations for THg were recorded at the RK site, but the highest MeHg values were observed at the DB site, which is an indication of active methylation of Hg in the reservoir. The concentrations of MeHg increased with the distance from the source of pollution, demonstrating that inorganic Hg is being transformed to MeHg. This may well explain the high MeHg values at the DB site that is closest to the dam and where the MeHg values are the highest. This site is deepest near the dam and the hydrodynamic condition is stable, similar to a lake environment, which will easily result in an anoxic condition in the sediment and be favorable to net Hg methylation. At the inlet of the reservoir, however, the RK site is shallow and the environment is similar to a river environment, which will not generate an anoxic condition (because of agitation in the sediment) and not be favorable to net Hg methylation [30]. The MT site close to a dock was in

the transition section impacted by ship navigation. Similar results were also observed in Clear Lake in California [31].

Total mercury tends to be bound to particulates, which are much higher in concentration in nonfiltered water than filtered water [20]. The high THg concentrations observed in the smaller size fraction of plankton at RK and MT sites may be attributed to Hg binding to algal exudates or other organic colloids (particulate Hg), which was the major form of Hg in natural aquatic systems [22], and controlled the partitioning of Hg in the water column [32]. The nature of Hg binding to phytoplankton also possesses the potential to significantly affect trophic transfer of Hg [33]. The inlet (RK) and transition sections (MT) of the reservoir, which are unstable hydrodynamically, had more suspended or resuspended particulate matter that were difficult to rinse off using the small mesh size during sampling. The highest concentrations of THg found in seston contained the major contaminant load in this reservoir.

The THg concentrations in the four size fractions of the planktonic communities from the three sites ranged from 49 to 5,504  $\text{ng g}^{-1}$  dry weight (Fig. 2). This range was similar to that in a eutrophic lake in the North China Plain [28] and much higher than in northern Québec lakes, which are mainly affected only by atmospheric deposition of Hg (Table 4). Total mercury concentrations at the RK site were even higher than in Grande Marsh, a highly Hg-contaminated marsh from gold mining in northern Colombia [34]. However, THg concentrations in plankton at the DB site were similar to that found in northern

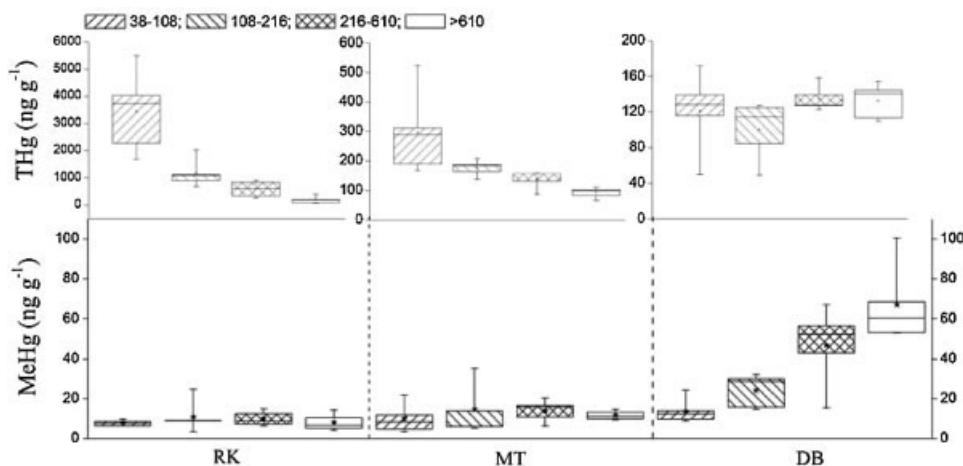


Fig. 3. Total mercury (THg) and methylmercury (MeHg) concentrations ( $\text{ng g}^{-1}$  dry wt) in plankton collected from the Baihua Reservoir, China (RK [upstream], MT [middle], and DB [downstream]) as a function of net mesh size.

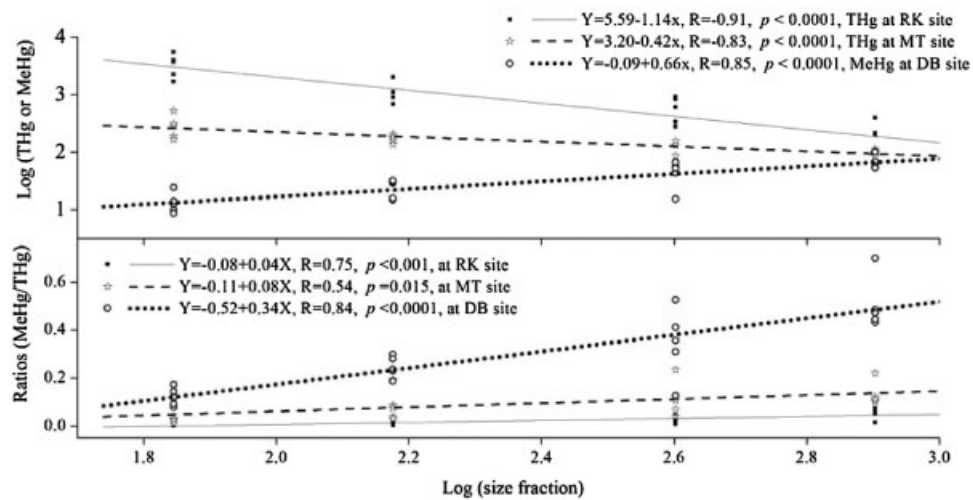


Fig. 4. Linear regressions of total mercury (THg) in the Baihua Reservoir, China at RK (upstream) and MT (middle); methylmercury (MeHg) at DB (downstream); and THg:MeHg ratios at the three sampling sites with size fractions (log transformation).

Québec lakes (Table 4). Methylmercury levels in plankton of the Baihua Reservoir are within the range of relatively uncontaminated lakes in the USA, northern Québec, and Eastern Europe (Table 4). Compared to the uncontaminated and contaminated waters in North America and Europe, THg concentrations in plankton were greatly elevated but MeHg concentrations were slightly lower in the Baihua Reservoir. The low MeHg concentration in plankton was also indicated by the fact that MeHg concentrations in carp fish samples in this reservoir were also low (average value of  $28 \text{ ng g}^{-1}$ ) compared to yellow perch in other pristine lakes [22,24,27]. Plankton is the diet of planktivorous fish and serves as the primary entry point of Hg into the aquatic food web [5,6].

Studies have concluded that the majority of Hg biomagnified through the food chain is in the form of MeHg [35]. Copepods assimilate MeHg much more efficiently than inorganic Hg. The relative assimilation efficiency of MeHg to inorganic Hg is 2.0 [36]. Methylmercury concentrations in zooplankton can increase many fold compared to MeHg concentrations in their algal diet [13,16]. The planktonic food web plays a key role in MeHg bioaccumulation in organisms at higher trophic levels

[1,19], and eventually in humans. Concentrations of MeHg in the four size fractions of the planktonic communities at the three sites ranged from 3 to  $101 \text{ ng g}^{-1}$  dry weight. The concentrations are in the lower range of concentrations observed in northern Québec lakes (Table 4). In almost all samples, the smaller fractions ( $38\text{--}108 \mu\text{m}$ ) had lower MeHg concentrations than the larger fractions ( $108\text{--}216 \mu\text{m}$ ). The fact that the smallest fractions were mainly composed of algae and the larger fractions of predatory species is symptomatic of a biomagnification of MeHg in the planktonic food chain. Increasing MeHg concentrations with increasing plankton size classes follows the concept of MeHg bioaccumulation in aquatic organisms and is consistent with previous studies from temperate lakes [4,7,11]. These studies report that MeHg bioaccumulation patterns were independent of the taxonomic composition of planktonic food webs and were related to somatic growth of planktonic organisms [7]. However, the concentrations of MeHg in plankton at the RK and MT sites increased only from seston to microzooplankton and decreased thereafter from mesozooplankton to macrozooplankton, different from that at the DB site. One reason may be that the unstable hydrological conditions may

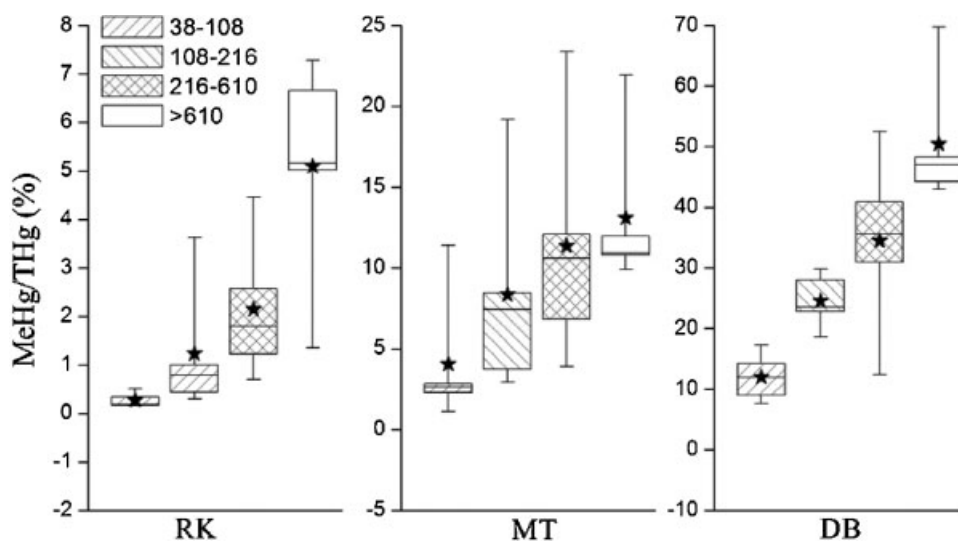


Fig. 5. The ratios of methylmercury to total mercury (MeHg:THg) in plankton collected from the Baihua Reservoir, China (RK [upstream], MT [middle], and DB [downstream]) as a function of net mesh size.

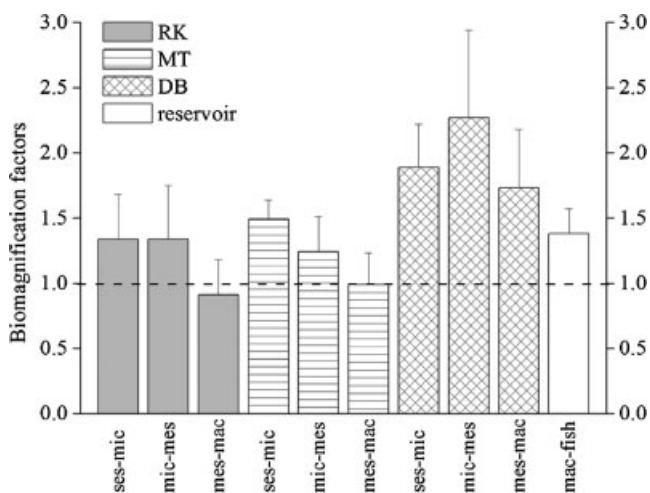


Fig. 6. Accumulation factors of methylmercury (MeHg) between different size classes of the planktonic food web (seston, ses; microzooplankton, mic; mesozooplankton, mes; and macrozooplankton, mac) and fish (*Aristichthys nobilis*) in the Baihua Reservoir, China (RK [upstream], MT [middle], and DB [downstream]). Fish data from Yan et al. [27].

affect the feeding behavior of mesozooplankton and macrozooplankton at RK and MT. Another reason may be that immature individuals were dominant in the zooplankton community at RK and MT, which was the opposite at the DB site. Watras et al. [19] observed that the MeHg concentrations and the ratios of MeHg to THg were higher in zooplankton than in microseston (phytoplankton and bacterioplankton) in several

pelagic food webs from some Wisconsin (USA) lakes. The bioaccumulation of MeHg in planktonic organisms has important toxicological implications for higher trophic levels [7]. Because MeHg concentrations increased with body size, macrozooplankton, as the preferred prey size for planktivorous fish, transfers the highest MeHg concentrations to the higher trophic levels [7].

The contrasting patterns between THg and MeHg concentrations suggested that MeHg bioaccumulation was independent of THg in the planktonic food web. The concentration of MeHg at RK was low, compared to DB, indicating different behaviors of Hg in the channel-shaped reservoir, possibly favoring demethylation over methylation under particular conditions. In general, no correlation exists between the concentrations of THg and MeHg in plankton. It is well known that THg concentration is not the only factor influencing Hg methylation [20].

Biomagnification was defined by Watras et al. [19] as the increasing concentration of a contaminant at successively higher trophic levels of a food web. For example, Mathews and Fisher [37] and Watras et al. [19] both suggested that MeHg biomagnifies from phytoplankton to fish. According to Reinfelder et al. [38], biomagnification of MeHg in aquatic food webs is well known and highest in large, long-lived, upper trophic level organisms. Most of the mean biomagnification factors for plankton in Baihua Reservoir were approximately 1.5 to 2.0 between adjacent trophic levels, which were smaller than the values calculated for hydroelectric reservoirs in northern Québec [13], but were in the same ranges obtained in northern Wisconsin lakes [19]. The biomagnification factors between trophic levels at the DB site are somewhat larger than those at the other two sites.

Table 4. Comparison of total mercury (THg) and methylmercury (MeHg) concentrations in different environmental matrices of plankton from different countries

Country	Geographical area	Size fraction	THg (ng g <sup>-1</sup> dry wt)	MeHg (ng g <sup>-1</sup> dry wt)	Reference	
Canada	Northern Quebec	>190	75–310	20–220	[11]	
Canada	Northern Québec Lake 1	53–100	81 ± 15	20 ± 6	[47]	
		100–200	68 ± 14	24 ± 6	[47]	
		200–500	88 ± 5	44 ± 7	[47]	
		>500	98 ± 7	65 ± 9	[47]	
	Northern Québec Lake 2	53–100	83 ± 10	21 ± 6	[47]	
		100–200	64 ± 7	23 ± 4	[47]	
		200–500	78 ± 4	38 ± 5	[47]	
		>500	109 ± 12	62 ± 6	[47]	
	Northern Québec Lake 3	53–100	68 ± 17	24 ± 9	[47]	
		100–200	54 ± 11	24 ± 5	[47]	
		200–500	71 ± 8	37 ± 6	[47]	
		>500	83 ± 9	54 ± 6	[47]	
		Central Quebec Lake 3	>200		17 to 377	[48]
		Lakes located in Quebec	>53	25 to 350	10 to 150	[4]
	Lake Superior	>153	22–133	15–46	[49]	
	La Grande hydroelectric complex	50–200	87 ± 20	27 ± 2.8	[50]	
	La Grande hydroelectric complex	200–950	132 ± 9.7	86 ± 6.6	[50]	
Hungary	Lake Balaton	>300	14–42	1–13	[51]	
Brazil	Tapajos River	Phytoplankton	17–125	2–25	[52]	
		Zooplankton	83–268	20–140	[52]	
Colombia	Grande Marsh, Cauca River basin <sup>a</sup>	0.45–64	520 ± 30		[34]	
		>150	940 ± 50		[34]	
USA	Lakes in northern Wisconsin	>153	33–206	6–161	[19]	
		Clear Lake, California	Plankton	10–90	1–3	[31]
			Zooplankton	10–70	3–7	[31]
China	Baihua Reservoir <sup>a</sup>	38–108	50–5504	3.3–24.4	Present study	
		108–216	49–2032	3.3–35.3	Present study	
		216–610	87–913	6.2–67.1	Present study	
		>610	67–391	4.1–100.6	Present study	

<sup>a</sup> Sites impacted by current or previous mercury contaminated.

*THg and MeHg concentrations in plankton*

Bioaccumulation of metal contaminants in aquatic food webs varies with metals and ecological factors, such as the trophic level, abundance of the organism within a trophic level, and their feeding strategies [28]. Metal bioaccumulation within a particular trophic level is also related to nutrient status, algal densities, and growth rates [39,40]. Nutrient enrichment in lakes could dilute the transfer of metals to higher trophic levels via blooms and/or density dilution in which the increase in algal biomass reduces the mass specific concentrations of metal and slows the rate of trophic transfer to zooplankton [6,28,39]. Eutrophication may play an important role in Hg transport and immobilization [41]. Eutrophication produces algal blooms, with a high growth rate and large capacity to bind Hg [42]. When algae moves by currents and/or settles to sediment, the distribution of Hg changes. More important, eutrophication affects the chemical forms and bioavailability of Hg in water [43]. Furthermore, when the algal bloom is diluted, the concentration of Hg as in MeHg per cell decreases, resulting in a lower dietary input to the zooplankton grazer and reducing bioaccumulation in algal-rich eutrophic systems [39]. Therefore, the interaction of nutrient and metal contaminants in Chinese lakes could have important influences on the actual bioaccumulation and trophic transfer of metals in aquatic food webs that received nutrient inputs from sewage discharge and aquaculture, and industrial contaminants from factories in the nearby cities [28].

Concentrations of THg in seston were negatively correlated with Chl *a* concentrations due to a short-lived and algal-induced dilution effect [32]. Lower Hg concentrations in plankton are a function of high nutrient and algal densities because of bio-dilution effects [6,28,39,44]. Unlike patterns observed in previous studies, THg bioaccumulation did not consistently increase with the trophic level in the Baihua Reservoir [19,45,46]. The decrease of THg in the food web, apart from hydrodynamic conditions, may be the result of high levels of eutrophication, which caused a biomass dilution of metal in the food web as seen in other productive lakes and in controlled experiments [6,28,39,45]. The highest THg and lowest MeHg concentrations found in the seston were probably related to the eutrophic conditions with elevated suspended particles and unstable hydrodynamic conditions. In this situation, Hg at RK and MT could be strongly complexed to or adsorbed by organic particulate matter or phytoplankton, causing a large dilution of Hg and then reducing Hg residence time in the water column, thus decreasing its availability to organisms. Therefore, we observed high THg concentrations at both RK and MT sites and low THg concentrations at DB. Consequently, the eutrophication of the reservoir affected Hg uptake, toxicity, and trophic transfer. Clearly, more study is urgently needed to elucidate the impact of eutrophication on the uptake of different Hg species by aquatic organisms.

**CONCLUSIONS**

Temporal and spatial comparisons of THg and MeHg concentrations in plankton showed differences among the three sampling sites from upstream to downstream of the Baihua Reservoir. These site-specific accumulation patterns of THg and MeHg reflect the unique hydrodynamics in reservoirs that regulate assimilation of Hg among all plankton size classes. The increasing ratios of MeHg to Hg at successively higher trophic levels of the planktonic food web suggest that biomagnification occurs throughout this food web. The high THg and low MeHg

concentrations found in the planktonic food web in the reservoir were probably related to the eutrophic conditions of this ecosystem, elevated content of suspended particles, and unstable hydrodynamic conditions. Patterns of THg and MeHg concentrations within the planktonic food web were constant throughout the study period. We attribute this to the minor seasonal variation of zooplankton species composition (unpublished data). The THg concentrations in plankton rapidly decreased as distance from the chemical plant increased. The present study shows the need for further investigation of the bioaccumulation of Hg in such contaminated aquatic ecosystems, which serve as an important source of fish production and drinking water to local human populations.

**SUPPLEMENTAL DATA****Figs. S1–S3. (1.02 MB DOC).**

*Acknowledgement*—The present study received financial support from the Chinese Academy of Sciences through a Sino-Swiss collaboration (GJHZ0903), the Natural Science Foundation of China (40873085, 40973083, 41173079, 41040030), and by the State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences. We thank Mingyan Gu and Runsheng Yin for assistance in field sampling, and Xin Zhao, Bo Meng, and Heng Yao for help in the MeHg analyses. We thank Larry Liddle for help with the article.

**REFERENCES**

1. Kainz M, Arts MT, Mazumder A. 2008. Essential versus potentially toxic dietary substances: A seasonal comparison of essential fatty acids and methyl mercury concentrations in the planktonic food web. *Environ Pollut* 155:262–270.
2. Castoldi AF, Coccini T, Ceccatelli S, Manzo L. 2001. Neurotoxicity and molecular effects of methylmercury. *Brain Res Bull* 55:197–203.
3. Yoshizawa K, Rimm EB, Morris JS, Spate VL, Hsieh C-c, Spiegelman D, Stampfer MJ, Willett WC. 2002. Mercury and the risk of coronary heart disease in men. *New Engl J Med* 347:1755–1760.
4. Masson S, Tremblay A. 2003. Effects of intensive fishing on the structure of zooplankton communities and mercury levels. *Sci Total Environ* 304:377–390.
5. Herrin RT, Lathrop RC, Gorski PR, Andren AW. 1998. Hypolimnetic methylmercury and its uptake by plankton during fall destratification: A key entry point of mercury into lake food chains? *Limnol Oceanogr* 43:1476–1486.
6. Chen CY, Folt CL. 2005. High plankton densities reduce mercury biomagnification. *Environ Sci Technol* 39:115–121.
7. Kainz M, Telmer K, Mazumder A. 2006. Bioaccumulation patterns of methyl mercury and essential fatty acids in lacustrine planktonic food webs and fish. *Sci Total Environ* 368:271–282.
8. Montgomery S, Lucotte M, Cournoyer L. 2000. The use of stable carbon isotopes to evaluate the importance of fine suspended particulate matter in the transfer of methylmercury to biota in boreal flooded environments. *Sci Total Environ* 261:33–41.
9. Paterson MJ, Rudd JWM, St. Louis V. 1998. Increases in total and methylmercury in zooplankton following flooding of a peatland reservoir. *Environ Sci Technol* 32:3868–3874.
10. Hall BD, Bodaly RA, Fudge RJP, Rudd JWM, Rosenberg DM. 1997. Food as the dominant pathway of methylmercury uptake by fish. *Water Air Soil Pollut* 100:13–24.
11. Kainz M, Lucotte M, Parrish CC. 2002. Methyl mercury in zooplankton—the role of size, habitat, and food quality. *Can J Fish Aquat Sci* 59:1606–1615.
12. Brooks JL, Dodson SI. 1965. Predation, body size, and composition of plankton. *Science* 150:28–35.
13. Tremblay A, Lucotte M, Schetagne R. 1998. Total mercury and methylmercury accumulation in zooplankton of hydroelectric reservoirs in northern Québec (Canada). *Sci Total Environ* 213:307–315.
14. Porvari P. 1998. Development of fish mercury concentrations in Finnish reservoirs from 1979 to 1994. *Sci Total Environ* 213:279–290.
15. Gorski PR, Cleckner LB, Hurley JP, Sierszen ME, Armstrong DE. 2003. Factors affecting enhanced mercury bioaccumulation in inland lakes of Isle Royale National Park, USA. *Sci Total Environ* 304:327–348.



16. Kainz M, Mazumder A. 2005. Effect of algal and bacterial diet on methyl mercury concentrations in zooplankton. *Environ Sci Technol* 39:1666–1672.
17. Orihel DM, Paterson MJ, Blanchfield PJ, Bodaly RA, Gilmour CC, Hintelmann H. 2008. Temporal changes in the distribution, methylation, and bioaccumulation of newly deposited mercury in an aquatic ecosystem. *Environ Pollut* 154:77–88.
18. Sharma CM, Borgström R, Huitfeldt JS, Rosseland BO. 2008. Selective exploitation of large pike *Esox lucius*—Effects on mercury concentrations in fish populations. *Sci Total Environ* 399:33–40.
19. Watras CJ, Back RC, Halvorsen S, Hudson RJM, Morrison KA, Wentz SP. 1998. Bioaccumulation of mercury in pelagic freshwater food webs. *Sci Total Environ* 219:183–208.
20. Horvat M, Nolde N, Fajon V, Jereb V, Logar M, Lojen S, Jacimovic R, Falnoga I, Liya Q, Faganeli J, Drobne D. 2003. Total mercury, methylmercury and selenium in mercury polluted areas in the province Guizhou, China. *Sci Total Environ* 304:231–256.
21. Feng X, Qiu G. 2008. Mercury pollution in Guizhou, southwestern China — An overview. *Sci Total Environ* 400:227–237.
22. Yan H, Feng X, Shang L, Qiu G, Dai Q, Wang S, Hou Y. 2008. The variations of mercury in sediment profiles from a historically mercury-contaminated reservoir, Guizhou province, China. *Sci Total Environ* 407:497–506.
23. Yan H, Feng X, Shang L, Qiu G. 2006. A primary study on biogeochemical cycling characteristics of mercury in Baihua Reservoir in Guizhou. *Chin J Geochem* 25:104–104.
24. Yan H, Rustadbakken A, Yao H, Larssen T, Feng X, Liu T, Shang L, Haugen TO. 2010. Total mercury in wild fish in Guizhou reservoirs, China. *J Environ Sci* 22:1129–1136.
25. Lorenzen CJ. 1967. Determination of chlorophyll and pheo-pigments: spectrophotometric equations. *Limnol Oceanogr* 12:343–346.
26. Pichet P, Morrison K, Rheault I, Tremblay A. 1999. Analysis of total mercury and methylmercury in environmental samples. In Lucotte M, Schetagne R, Therien N, Langlois C, Tremblay A, eds, *Mercury in the Biochemical Cycle: Natural Environments and Hydroelectric Reservoirs of Northern Quebec (Canada)*. Springer, New York, NY, USA, pp 41–52.
27. Yan H, Feng X, Liu T, Shang L, Li Z, Li G. 2008. Present situation of fish mercury pollution in heavily mercury-contaminated Baihua reservoir in Guizhou. *Chin J Ecol* 27:1357–1361.
28. Chen C, Pickhardt P, Xu M, Folt C. 2008. Mercury and arsenic bioaccumulation and eutrophication in Baiyangdian Lake, China. *Water Air Soil Poll* 190:115–127.
29. Feng X, Sommar J, Lindqvist O, Hong Y. 2002. Occurrence, emissions and deposition of mercury during coal combustion in the province Guizhou, China. *Water Air Soil Pollut* 139:311–324.
30. Friedl G, Wüest A. 2002. Disrupting biogeochemical cycles — Consequences of damming. *Aquat Sci* 64:55–65.
31. Suchanek TH, Eagles-Smith CA, Harner EJ. 2008. Is Clear Lake methylmercury distribution decoupled from bulk mercury loading? *Ecol Appl* 18:A107–127.
32. Pickhardt PC, Folt CL, Chen CY, Klaue B, Blum JD. 2005. Impacts of zooplankton composition and algal enrichment on the accumulation of mercury in an experimental freshwater food web. *Sci Total Environ* 339:89–101.
33. Ng TY-T, Amiard-Triquet C, Rainbow PS, Amiard JC, Wang W-X. 2005. Physico-chemical form of trace metals accumulated by phytoplankton and their assimilation by filter-feeding invertebrates. *Mar Ecol Prog Ser* 299:179–191.
34. Marrugo-Negrete J, Benitez L, Olivero-Verbel J. 2008. Distribution of mercury in several environmental compartments in an aquatic ecosystem impacted by gold mining in northern Colombia. *Arch Environ Contam Toxicol* 55:305–316.
35. U.S. Environmental Protection Agency. 1997. Mercury Study Report to the Congress. EPA 452/R-97-0003. Washington, DC.
36. Lawson N, Mason R. 1998. Accumulation of mercury in estuarine food chains. *Biogeochemistry* 40:235–247.
37. Mathews T, Fisher N. 2008. Trophic transfer of seven trace metals in a four-step marine food chain. *Mar Ecol Prog Ser* 367:23–33.
38. Reinfelder JR, Fisher NS, Luoma SN, Nichols JW, Wang WX. 1998. Trace element trophic transfer in aquatic organisms: A critique of the kinetic model approach. *Sci Total Environ* 219:117–135.
39. Pickhardt PC, Chen CY, Klaue B, Blum JD. 2002. Algal blooms reduce the uptake of toxic methylmercury in freshwater food webs. *Proc Natl Acad Sci U S A* 99:4419–4423.
40. Karimi R, Chen CY, Pickhardt PC, Fisher NS, Folt CL. 2007. Stoichiometric controls of mercury dilution by growth. *Proc Natl Acad Sci U S A* 104:7477–7482.
41. Coelho JP, Pereira ME, Duarte A, Pardal MA. 2005. Macroalgae response to a mercury contamination gradient in a temperate coastal lagoon (Ria de Aveiro, Portugal). *Estuar Coast Shelf Sci* 65:492–500.
42. Radway JC, Wilde EW, Whitaker MJ, Weissman JC. 2001. Screening of algal strains for metal removal capabilities. *J Appl Phycol* 13:451–455.
43. He T, Feng X, Guo Y, Qiu G, Li Z, Liang L, Lu J. 2008. The impact of eutrophication on the biogeochemical cycling of mercury species in a reservoir: A case study from Hongfeng Reservoir, Guizhou, China. *Environ Pollut* 154:56–67.
44. Kamman NC, Lorey PM, Driscoll CT, Estabrook R, Major A, Pientka B, Glassford E. 2004. Assessment of mercury in waters, sediments, and biota of New Hampshire and Vermont lakes, USA, sampled using a geographically randomized design. *Environ Toxicol Chem* 23:1172–1186.
45. Chen CY, Stemberger RS, Klaue B, Blum JD, Pickhardt PC, Folt CL. 2000. Accumulation of heavy metals in food web components across a gradient of lakes. *Limnol Oceanogr* 45:1525–1536.
46. Power M, Klein GM, Guiguer KRRR, Kwan MKH. 2002. Mercury accumulation in the fish community of a sub-Arctic lake in relation to trophic position and carbon sources. *J Appl Ecol* 39:819–830.
47. Surette C, Lucotte M, Tremblay A. 2006. Influence of intensive fishing on the partitioning of mercury and methylmercury in three lakes of northern Quebec. *Sci Total Environ* 368:248–261.
48. Garcia E, Carignan R, Lean D. 2007. Seasonal and inter-annual variations in methyl mercury concentrations in zooplankton from boreal lakes impacted by deforestation or natural forest fires. *Environ Monit Assess* 131:1–11.
49. Back RC, Gorski PR, Cleckner LB, Hurley JP. 2003. Mercury content and speciation in the plankton and benthos of Lake Superior. *Sci Total Environ* 304:349–354.
50. Schetagne R, Doyon J-F, Fournier J-J. 2000. Export of mercury downstream from reservoirs. *Sci Total Environ* 260:135–145.
51. Nguyen HL, Leermakers M, Kurunczi S, Bozo L, Baeyens W. 2005. Mercury distribution and speciation in Lake Balaton, Hungary. *Sci Total Environ* 340:231–246.
52. Roulet M, Lucotte M, Guimarães JRD, Rheault I. 2000. Methylmercury in water, seston, and epiphyton of an Amazonian river and its floodplain, Tapajós River, Brazil. *Sci Total Environ* 261:43–59.