Influence of Eutrophication on the Distribution of Total Mercury and Methylmercury in Hydroelectric Reservoirs

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摘要 (概要)

The decomposition of flooded vegetation and organic carbon in soils may also stimulate Hg methylation (Furutani and Rudd, 1980; Lucotte et al., 1999). [...] some studies have pointed out the role of primary productivity in the biota system in the accumulation and biomagnification of MeHg (Cleckner et al., 1998; Pickhardt et al., 2002). [...] the popularity of cage aquaculture appears to be the key factor in the high primary productivity in WX and WD, which would actually significantly change the conditions promoting net MeHg production to produce the large difference between the WJD and the YZD as well as between WS, WZ and WX, WD in terms of MeHg distribution in water.

全文文献

Headnote
The distribution of mercury (Hg) and the characteristics of its methylation were investigated in Wujiangdu (WJD) and Yinzidu (YZD) reservoirs in Guizhou province, China. The two reservoirs are characterized by high and low levels of primary productivity, respectively. Mercury species in water samples from depth profiles in both reservoirs and from interface water in the WJD were analyzed each season during 2007. The concentrations of total Hg (HgT^sub unf^) and methylmercury (MeHgT^sub unf^) in unfiltered water samples from the WJD varied from 3.0 to 18 pmol dm^sup -3^ and from 0.17 to 15 pmol dm^sup -3^, respectively; ranges were 2.0 to 9.5 pmol dm^sup -3^ for HgT^sub unf^ and 0.14 to 2.2 pmol dm^sup -3^ for MeHgT^sub unf^ in the YZD. Elevated methylmercury concentrations in water samples from the bottom water and water-sediment interface demonstrated an active net Hg methylation in the downstream reach of the WJD. There was no discernable Hg methylation occurring in the YZD, nor in the upstream and middle reaches of the WJD. The results suggest that high primary productivity resulting from cage aquaculture activities in the WJD is an important control on Hg methylation in the reservoir, increasing the concentrations of MeHg in water in the Wujiang River basin, southwestern China.

Abbreviations: CVAFS, cold vapor atomic fluorescence spectroscopy; DO, dissolved oxygen; DOC, dissolved organic carbon; GC, gas chromatography; HgT^sub f^, filtered total mercury; HgT^sub unf^, unfiltered total mercury; MeHg, methylmercury; MeHgT^sub f^, filtered methylmercury; MeHgT^sub unf^, unfiltered methylmercury; PHg, particulate mercury; PMeHg, particulate methylmercury; RHg , reactive mercury; T, temperature; TN, total nitrogen; TP, total phosphorus; TSS, total suspended solid; WJD, Wujiangdu Reservoir; YZD, Yinzidu Reservoir.

MERCURY (Hg) is an extremely toxic pollutant, and has received considerable attention due to its methylation, accumulation, and biomagnification in aquatic food chains (Lindqvist et al., 1991). Elevated levels of methylmercury (MeHg) have been observed in fish in newly constructed reservoirs compared with fish in adjacent lakes (Hecky, 1991; Abernathy and Cumbie, 1997; Bodaly, 1997). Methylmercury production in water from new reservoirs following impoundment may persist for up to 10 yr (St. Louis et al., 2004; Hall et al., 2005). Subsequently, the increased MeHg levels in fish from reservoirs may last for up to 30 yr (Mailman et al., 2006). A previous study showed that biological activity, nutrient availability, pH, temperature, and redox potential, as well as the presence of inorganic and organic complexing agents, exert an influence on net MeHg production (Ullrich et al., 2001). The decomposition of flooded vegetation and organic carbon in soils may also stimulate Hg methylation (Furutani and Rudd, 1980; Lucotte et al., 1999). Moreover, some studies have pointed out the role of primary productivity in the biota system in the accumulation and biomagnification of MeHg (Cleckner et al., 1998; Pickhardt et al., 2002).

China has abundant water resources, and the Yangtze River basin is a major watershed. With the implementation of the Chinese Western Development Policy, numerous large reservoirs (e.g., Hongjiadu, Puding, Yinzidu, Dongfeng, Suofengying, and Wujiangdu) have been or are being constructed in Guizhou province along the Wujiang River, the largest tributary of the upper Yangtze River (Fig. 1). Wujiangdu (WJD) and Yinzidu (YZD) reservoirs are located in the lower and upper portions of the Wujiang River basin, respectively.

Recent studies have shown that the WJD is a net sink for total Hg (HgT^sub unf^) but a net source of MeHg to the downstream ecosystems, with the maximum level of MeHg in water occurring in summer (Feng et al., 2009a). These authors reported that an extended water residence time increased the net MeHg production in the reservoirs. Feng et al. (2009b) indicated that high primary productivity (the synthesis and storage of organic molecules during the growth and reproduction of photosynthetic organisms) in reservoirs may be a favorable factor for the net MeHg production in sediment. He et al. (2008a) suggested that hypereutrophication in Hongfeng Reservoir results in low dissolved oxygen (DO) and high dissolved organic carbon (DOC), accelerating the formation of MeHg in the hypolimnion, especially during summer. Associated studies showed that age, water volume, water residence time, trophic level, and nutrient characteristics have significant effects on net Hg methylation (Jiang, 2005; He, 2007). To date, the vertical distribution and spatial variation of Hg species in the cited reservoirs have not been investigated, and the influence of primary productivity on the biogeochemical processing of Hg in the WJD and the YZD is unclear.

To better understand the controlling factors of MeHg production in reservoirs, we measured Hg concentrations in the WJD and the YZD, two reservoirs having vastly different levels of primary productivity. The objectives of this study were (i) to determine the spatial and temporal distributions of Hg species and the characteristics of Hg methylation in these reservoirs; and (ii) to reveal any relationships between primary productivity and Hg methylation.

Materials and Methods

Study Area

The Wujiang River is the largest tributary of the upper Yangtze River, mainly flowing in a karstic environment in Guizhou province, southwest China. The total area of the watershed is about 87,920 km^sup 2^. The Wujiang River basin is characterized by a humid subtropical monsoon climate with

an average annual rainfall of 900 to 1400 mm and an annual mean temperature of 14.6°C. The WJD was built in 1979, and has a watershed of 27,790 km^sup 2^ and a volume of 23 × 10^sup 8^ m^sup 3^; it is located in the lower Wujiang River basin (Fig. 1). The average water residence time in the WJD is 53.0 d. The YZD, constructed in 2001 with a watershed of 6422 km^sup 2^ and a volume of 5.27 × 10^sup 8^ m^sup 3^, is located in the upper Wujiang River basin (Fig. 1). The average water residence time in the YZD is 43.7 d. At present, there are no significant point source discharges in the drainage basins of these reservoirs.

Concentrations of NO^sub 3^^sup -^ and total nitrogen (TN) in the WJD were much higher than those in the YZD during April 2006 to January 2007, and concentrations of PO^sub 4^^sup -3^ and total phosphorus (TP) in the WJD were about 10 to 100 times higher than those in the YZD (Wang et al., 2008; Wang et al., 2009; Yu, 2008). Wang et al. (2008) and Yu (2008) found that the chlorophyll content in surface water of the WJD was significantly higher than that of the YZD. Concentrations of TN, TP, chlorophyll, and phytoplankton cell density/biomass in the WJD all increase from upstream to downstream (Dang, 2008). These investigations showed that a hyper state of eutrophication exists in the WJD, while the YZD is oligotrophic-mesotrophic, indicating a much higher level of primary productivity in the WJD than in the YZD (Kimmel and Groege, 1984).

Sample Collection

Four sampling stations in each reservoir, spatially distributed from upstream to downstream, were chosen as depicted in Fig. 1. Stations WS (27°13'27.8" N, 106°31'20.6" E) and YS (26°27'34.7" N, 106°01'25.3" E) are at the upper ends of the reservoirs. WZ (27°15'50.3" N, 106°36'04.1" E) and YZ (26°29'12.2" N, 106°06'33.4" E) are located in the middle parts of the reservoirs. WX (27°18'38.8" N, 106°42'44.2" E) and YX (26°32'21.1" N, 106°08'07.9" E) are situated in the lower parts of reservoirs. WD (27°19'07.5" N, 106°44'54.6" E) and YD (26°34'47.6" N, 106°08'15.8" E) are located adjacent to the dams (within about 500 m). The WJD and the YZD are located in the karstic environment of the Wujiang River basin, which is a typical deep-valley, high-mountain gorge. The water columns are deeper and deeper from upstream to downstream of both reservoirs. Cage aquaculture activities conducted by local residents are popular in the lower end of the WJD, but are absent elsewhere in the WJD and in the YZD. No formal quantitative data on cage aquaculture activities are available.

Water samples from each site were collected each season in 2007 using an acid-washed, Teflon-coated, 10-L Niskin sampler (Model 1010X series, General Oceanics Inc., Miami, FL). Samples were collected from five to eight different depths at each sampling station. All samples were collected following ultraclean sample handling protocols. Water samples were transferred from the sampler into acid-cleaned borosilicate glass bottles. These bottles had been rigorously cleaned before use by immersion in 10% HNO^sub 3^sub (v/v), followed by rinsing with ultrapure deionized water (18.2 MΩ Milli-Q) and heating for 1 h in a muffle furnace at 500°C; they were then double-bagged and stored in a sealed box. Each bottle was rinsed three times with reservoir water before sample collection. Filtered samples were collected on-site using a 0.45-μm filter (Millipore), and subsequently analyzed for dissolved total Hg (HgT^sub f^) and dissolved MeHg (MeHgT^sub f^). Total Hg (HgT^sub unf^), reactive Hg (RHg), total MeHg (MeHgT^sub unf^), and total suspended solid (TSS) were analyzed in each of the unfiltered samples.

In addition, undisturbed interface water samples (5-10 cm above the water-sediment interface) and sediment cores were collected from the WJD using a custom-designed sampler (Wang et al., 1998). The interface water samples were filtered on-site into cleaned borosilicate glass bottles and subsequently analyzed for HgT^sub f^ and MeHgT^sub f^. The sediments were immediately sliced into 1-cm sections and collected in acid-cleaned centrifuge tubes (Supplementary Fig. S1). Sediment samples were freeze-dried, homogenized with a mortar, and analyzed for organic matter. Due to the absence of a sediment layer, no interface water and sediment were available in the YZD. Therefore, we only collected water samples at the bottom of this reservoir.

All water samples were acidified on-site to 0.5% (v/v) using concentrated hydrochloric acid (HCl); the sample bottles were then sealed, double-bagged, and transported to the lab on ice, within 24 h. They were stored in a refrigerator at +4°C in the dark until analysis.

Sample Analyses

The analyses of Hg species in water samples were conducted within 28 d after sampling. The HgT^sub unf^, HgT^sub f^, and RHg were quantified using dual amalgamation cold vapor atomic fluorescence spectroscopy (CVAFS) (Tekran 2500, Tekran Inc., Toronto, ON, Canada) following approved methodologies (Bloom and Fitzgerald, 1988; USEPA, 2002). Samples for HgT^sub unf^ and HgT^sub f^ analysis were oxidized with 0.5% (v/v) BrCl (bromine chloride). Excess BrCl was reduced with hydroxylammonium chloride before adding SnCl2 (stannous chloride) to convert HgII to volatile Hg0. The Hg0 was trapped by gold amalgamation (Bloom and Fitzgerald, 1988; USEPA, 2002). Particulate Hg (PHg) was calculated as the difference between the concentration of HgT^sub unf^ and HgT^sub f^ in the sample. Reactive Hg was determined by the addition of 20% (v/v) SnCl2 solution to unfiltered and acidified samples, followed by the purgeand- trap CVAFS method indicated above.

Water samples were quantified for MeHg using CVAFS (Brooks Rand Model III, Brooks Rand Labs, Seattle, WA) following distillation, aqueous phase ethylation, and isothermal gas chromatographic (GC) separation (Liang et al., 1994; USEPA, 2001). A 45-mL aliquot of acidified sample was distilled at 125°C for 3 to 4 h to transfer MeHg into a clean water matrix. The distillate was adjusted to pH 4.9 with 2 M sodium acetate and ethylated by addition of 0.1 mL 1% (v/v) NaB(C^sub 2^H^sub 5^)^sub 4^ (sodium tetraethylborate). Ethylated Hg species were purged onto a Tenax trap using N^sub 2^. These species were then desorbed from the trap by gentle heat in an Ar stream, isothermally separated by gas chromatography, pyrolytically decomposed to Hg^sup 2^, and detected by atomic fluorescence spectroscopy. Particulate MeHg (PMeHg) was calculated as the difference between the concentrations of MeHgT^sub unf^ and MeHgT^sub f^ in the sample.

Total suspended solid was determined gravimetrically by filtering an aliquot of water (typically 1500 mL) through a preweighed polycarbonate membrane filter, 0.45-μm pore size, 47 mm in diameter. Water quality parameters such as pH and temperature (T) were measured in situ using a portable analyzer (PD- 501, Shanghai San-Xin Instrumentation Inc., Shanghai, China). Dissolved oxygen (DO) in water was monitored using an in situ DO probe (HI 7042S, HANNA Instruments Inc., Padova, Italy). The concentrations of organic matter in the surface sediment samples were analyzed using KCr^sub 2^O^sub 7^ (potassium dichromate) oxidation coupled with volumetric analysis (Jiang, 2005).

Quality Assurance

Quality control for HgT^sub unf^ and MeHgT^sub unf^ determination was conducted by field blanks, matrix spikes, and duplicate samples. The method detection limit (3 × σ) was 0.10 pmol dm^sup -3^ for HgT^sub unf^ and 0.050 pmol dm^sup -3^ for MeHgT^sub unf^. The method blank was in each case less than the detection limit. Field blanks were 0.70 ± 0.20 pmol dm^sup -3^ for HgT^sub unf^ and 0.060 pmol dm^sup -3^ for MeHgT^sub unf^. The relative standard deviations for duplicate sample analyses were <8.5% for MeHgT^sub unf^ and HgT^sub unf^. Recoveries for matrix spikes ranged from 87 to 113% for MeHgT^sub unf^ analysis, and from 91 to 108% for HgT^sub unf^ analysis.

Statistical evaluation was performed using SPSS 11.5 software (SPSS China Ltd., Shanghai, China). Relationships between covariant sets of data were analyzed by regression analysis. Correlation coefficients (r) and significance probabilities (p) were computed for the linear regression fits. Significant differences were declared at $p < 0.05$.

Results and Discussion

General Water Quality Characteristics

Vertical profiles for T, pH, and DO from the WJD and the YZD are displayed in Fig. 2 and 3, and summary data are shown in Supplementary Table S1.

Water temperature profiles exhibited expected seasonal and spatial patterns, with temperatures in the WJD and the YZD ranging from 12 to 25°C and from 10 to 24°C, respectively. The reservoirs were well mixed in fall and winter, and stratified in spring and summer, with stronger trends at the downstream stations of the reservoirs. Total suspended solid showed a seasonal pattern that was opposite of T, with the highest average TSS values in dry seasons (spring and winter) and the lowest values in wet seasons (summer and fall).

Due to the karstic geology of the Wujiang River basin, the water was slightly alkaline in most samples (WJD: pH = 7.7 ± 0.47; YZD: pH = 7.7 ± 0.47) but slightly acidic in the bottom stratum at WX in summer (6.8) and WD in winter (6.7) and summer (6.9), which may result from the formation of organic acids in the sediment (He et al., 2008b). The pH reached distinct peaks of 9.5 and 9.7 in the surface water at WX and WD during spring, coinciding with maxima in DO (12 mg dm^sup -3^ at both sites) during algal blooms (He et al., 2008b). No significant variations of pH with depth were observed at WS, WZ, or at any site in the YZD during this study.

Vertical profiles of DO were pronounced and well correlated with thermal stratification. Spatial and seasonal differences in DO levels were observed in the WJD but were absent in the YZD. In the WJD, explicit defi- ciencies of DO were persistent in the bottom waters of WX and WD throughout the seasons without thermal stratification but were absent at WS and WZ. In summer, low DO concentrations (WS: 7.1 mg dm^sup -3^; WZ: 6.5 mg dm^sup -3^; WX: 1.2 mg dm^sup -3^; WD: 1.6 mg dm^sup -3^) in the hypolimnion of the WJD were believed to be the result of intensive bacterial decomposition of settled degradable organic matter (He et al., 2008b). The spring maximum of DO at WX and WD at the surface was explained by algal blooms, as previously mentioned (He et al., 2008b). However, elevated DO in surface water was not observed at WS and WZ during spring, possibly indicating the absence of an algal bloom there. For the YZD, there was a slightly decreasing trend in DO near the bottom in summer, showing the effect of stratification.

The WJD and the YZD were completely stratified in summer, especially at the downstream sites, but abundant algae were only present at WX and WD (Zhu et al., 2006; Dang, 2008). The more pronounced chemical stratification at WX and WD compared with WS, WZ, and throughout the YZD is due to higher primary productivity levels at WX and WD. Hence, primary productivity is a main factor controlling water column characteristics in stratified reservoirs.

Organic Matter Contents in the Surface Sediments of the WJD

The organic matter contents in surface sediments at downstream sites in the WJD (range: 8.76-11.4%) were higher compared with those at upstream sites (3.74-5.47%; see Table 1). Cage aquaculture activities are popular at the lower end of the WJD but absent in the upper reservoir. The levels of TN, TP, phytoplankton cell density/biomass, and chlorophyll in the WJD were all elevated at the lower end of the reservoir compared with upstream (Dang, 2008). Owing to the popularity of cage aquaculture on the lower WJD, fish feed and fish feces were potentially significant sources of organic matter input to sediments there, contributing to the higher primary productivity compared with upstream. This phenomenon was not found in the YZD.

We believe river channel erosion and surface runoff were the main sources of sediment organic matter at the upper end of the WJD (WS) during our sampling. The difference in organic matter contents in surface sediment between spring (5.29%) and summer (3.74%) at WS was probably due to differences in the intensity of river erosion and surface runoff between spring and summer.

Mercury in Reservoir Waters

Unfiltered Total Mercury

Spatial and seasonal distributions of HgT^sub unf^, HgT^sub f^, RHg , and PHg concentrations in the reservoirs are shown in Fig. 4, 5, and Supplementary Table S2. Annual mean concentrations of HgT^sub unf^ were 6.5 ± 2.8 pmol dm^sup -3^ in the WJD (range: 3.0-18 pmol dm^sup -3^) and 5.0 ± 1.5 pmol dm^sup -3^ in the YZD (range: 2.0- 9.5 pmol dm^sup -3^). The HgT^sub unf^ concentrations in these reservoirs were significantly below the Chinese surface water standard of 250 pmol dm^sup -3^ (Environmental Quality Standards for Surface Water; GB3838-2002), and also below the 60 pmol dm^sup -3^ standard for HgT^sub unf^ recommended by the USEPA to protect against adverse chronic effects on aquatic life (USEPA, 1992)

The HgT^sub unf^ concentrations in the water column of the WJD were significantly higher than those of the YZD (K-S test, $p < 0.001$). Peak levels of HgT^sub unf^ in the WJD were observed during spring in surface water at stations WX (18 pmol dm^sup -3^) and WD (16 pmol dm^sup -3^), coinciding temporally and spatially with peaks of TSS (WX: 5.4 mg dm^sup -3^; WD: 3.0 mg dm^sup -3^) and PHg (WX: 9.2 pmol dm^sup -3^; WD: 6.0 pmol dm^sup -3^). These elevated levels of HgT^sub unf^ in the WJD may be attributed to seasonal increases in anthropogenic activities and the appearance of algal blooms (Zhu et al., 2006; Dang, 2008). Algae have the capacity to bind Hg (Hurley et al., 1991), and the higher HgT^sub unf^ in the WJD than in the YZD could be due to the higher level of primary productivity in the WJD. Elevated levels of HgT^sub unf^ observed during summer in the bottom water of stations WS (10 pmol dm^sup -3^), WZ (13 pmol dm^sup -3^), WX (17 pmol dm^sup -3^), and WD (13 pmol dm^sup -3^) coincided with elevated levels of PHg at these sites, perhaps suggesting that sediment resuspension was significant at that time.

No discernable difference in the annual mean concentration of HgT^sub unf^ was observed among WS, WZ, WX, WD (K-W test, $p = 0.99$). The concentrations of HgT^sub unf^ during the cold and dry seasons (spring and winter) in the WJD were higher than those in hot and wet seasons (summer and fall) (K-S test, $p = 0.001$).

In the YZD, the concentration of HgT^sub unf^ at YS was significantly higher than those at the other sampling sites (K-S test, $p = 0.004$) during all the seasons. Similarly, the TSS concentration at YS was much higher than those at YZ, YX, and YD (K-S test, $p < 0.001$). This implies that particulate matter and sedimentation were important factors controlling the distribution of HgT^sub unf^ in the YZD. However, no discernable vertical trends in the distribution of HgT^sub unf^ were observed in the water column of the YZD during our sampling. This results from a lack of local point sources and the absence of an algal bloom in this reservoir.

Filtered Total Mercury

The HgT^sub f^ concentrations ranged from 1.7 to 15 pmol dm^sup -3^ (mean = 4.3 ± 2.1 pmol dm^sup -3^) in the WJD, and from 1.1 to 5.0 pmol dm^sup -3^ (mean = 2.5 ± 0.70 pmol dm^sup -3^) in the YZD. The HgT^sub f^ represented 18 to 99% (mean = 67 ± 17%) of HgT^sub unf^ in the WJD and 17 to 93% (mean = 53 ± 17%) of HgT^sub unf^ in the YZD. Regression analysis revealed a significant positive correlation between HgT^sub f^ and HgT^sub unf^ in the WJD ($r = 0.80$, $p < 0.0001$, $n = 110$), suggesting that HgT^sub f^ is the main Hg fraction present in the water column of the WJD. For the YZD, no relationship between HgT^sub f^ and HgT^sub unf^ was found ($r = 0.12$, $p = 0.25$, $n = 100$), implying that the Hg speciation in the YZD is presumably dictated by other factors.

Elevated concentrations of HgT^sub f^ were observed in bottom water in the WJD at WX (15 pmol dm^sup -3^) and WD (8.0 pmol dm^sup -3^) during summer, probably due to the diffusion of Hg from sediment (Feng et al., 2009b). In addition, spring surface water samples at WX and WD were enriched in HgT^sub f^. This could be explained by atmospheric inputs combined with stratification of water column. Except for these cases, none of the other sites exhibited spatial or seasonal trends in HgT^sub f^ (Fig. 5 and Supplementary Table S2).

Particulate Mercury

The PHg concentrations ranged from 0.013 to 10 pmol dm^sup -3^ (mean = 2.2 ± 1.7 pmol dm^sup -3^) in the WJD, and from 0.31 to 7.0 pmol dm^sup -3^ (mean = 2.6 ± 1.6 pmol dm^sup -3^) in the YZD. Regression analysis revealed a significant positive correlation between PHg and HgT^sub unf^ in the YZD ($r = 0.88$, $p < 0.0001$, $n = 100$), implying there is partitioning of Hg to particulate matter in the YZD.

Concentrations of both HgT^sub unf^ and PHg were elevated in surface water at WX and WD in spring. The elevation of HgT^sub unf^ concentrations resulted from increased levels of PHg. Concentrations of TSS were also enhanced (WX: 5.4 mg dm^sup -3^; WD: 3.0 mg dm^sup -3^), primarily due to algal bloom formation in the surface water. The elevated PHg levels may be due to an increased fraction of organic particles (Zhu et al., 2006; Dang, 2008), which can adsorb more Hg than inorganic particles (Hurley et al., 1991). Concentrations of PHg in the bottom water at stations WS (summer), WZ (summer), WX (spring and fall), and WD (summer) were all significantly higher than the corresponding overlying water, indicating the input of particulate Hg from sediment resuspension. In contrast, no discernible spatial or seasonal trends in PHg were observed in the YZD because algal blooms and bottom sediments were absent there.

Reactive Mercury

Reactive Hg concentrations ranged from 0.18 to 3.1 pmol dm^sup -3^ (mean = 0.75 ± 0.46 pmol dm^sup -3^) in the WJD, and from 0.18 to 3.9 pmol dm^sup -3^ (mean = 0.95 ± 0.75 pmol dm^sup -3^) in the YZD. There were no discernable differences in RHg concentrations between the WJD and the YZD stations during the sampling seasons (K-S test, $p = 0.27$). Furthermore, we failed to observe any spatial or seasonal variation in RHg concentrations for either the WJD or the YZD. The RHg concentration was at a maximum in the bottom water of the WJD (3.1 pmol dm^sup -3^) during the summer. These elevated levels of RHg are possibly explained by diffusion of RHg from sediment, heightened by the lack of mixing. This explanation is supported in that no such maximum was observed in the YZD, where a sediment layer is absent.

Methylmercury

The distribution patterns and concentrations of MeHgT^sub unf^, PMeHg, and MeHgT^sub f^ in the reservoirs are illustrated in Fig. 6, 7, and Supplementary Table S2. The mean ratios of MeHgT^sub f^/ MeHgT^sub unf^ are comparable in the two reservoirs (69 ± 22% in the WJD and 61 ± 23% in the YZD). Statistical analyses yielded significant positive correlations between MeHgT^sub f^ and MeHgT^sub unf^ in the WJD ($r = 0.80$, $p < 0.0001$, $n = 111$) and in the YZD ($r = 0.61$, $p < 0.0001$, $n = 101$). Furthermore, statistically significant differences of MeHgT^sub unf^ and MeHgT^sub f^ levels were found between the WJD and the YZD throughout the four sampling campaigns (K-S test, $p = 0.016$ and $p < 0.001$ for MeHgT^sub unf^ and MeHgT^sub f^, respectively). Annual average MeHgT^sub unf^ concentrations showed statistically significant differences among the four sampling stations (WS, WZ, WX, WD) in the WJD (K-W test, $p = 0.05$). In addition, annual average MeHgT^sub unf^ concentrations at WX and WD were signifi- cantly higher than those at WS and WZ (K-S test, $p = 0.01$). This observation suggests that considerably different levels of net Hg methylation exist between the upper and lower parts of the WJD. In comparison, the relative spatial variation of MeHgT^sub unf^ among the four sampling stations in the YZD is less pronounced (K-W test, $p = 0.92$). The MeHgT^sub unf^ concentrations in both reservoirs were significantly higher in summer than in other seasons (K-S test, $p = 0.001$ and $p < 0.001$ for the WJD and the YZD, respectively).

There are two possible sources of MeHg in the reservoirs: (i) in situ production being controlled by redox chemistry and/ or settling particulate matter containing MeHg (Meili, 1997; Eckley et al., 2005; He et al., 2008b), and (ii) diffusion or resuspension, or both, of MeHg from underlying sediments (Mason and Sullivan, 1999; Lawson et al., 2001). Gilmour and Henry (1991) showed that low pH and negative redox potential, not only increase methylation rates but also decrease demethylation rates, resulting in net production of MeHg.

In summer, MeHgT^sub unf^, MeHgT^sub f^, and PMeHg were all low in the mixed layer but increased sharply to maxima in the low-oxygen region at downstream sites in the WJD. Similar observations were made in a stratified estuary in New England (Mason et al., 1993) and in a reservoir of southwestern China (He et al., 2008b). Our results indicate that active Hg methylation occurred at WX and WD during summer, in support of the conclusion by Ullrich et al. (2001) that high temperatures favor the Hg methylation process. In contrast, summer maxima of MeHg in bottom waters were not observed in the upper end of the WJD nor in any part of the YZD, plausibly ruling out sustained Hg methylation at those sites.

Feng et al. (2009a) reported that net annual Hg methylation was significantly higher in the WJD than in Dongfeng Reservoir because of the longer water residence time in the WJD. The water residence time in the WJD and the YZD is similar. However, the production of MeHg was much lower in the YZD. Hence, in this study the key indicator for Hg methylation must not be the water residence time but rather the primary productivity. Another interesting outcome of this study is that Hg methylation was much more active in the lower end of the WJD than in the upstream sites because of the primary productivity gradient, which supports the above conclusion.

The highest MeHgT^sub unf^ concentrations during the summer stratification occurred at 90 and 95 m at WX and WD, respectively, where DO was lowest (WX: 1.6 mg dm^sup -3^; WD: 1.2 mg dm^sup -3^). Optimum conditions for Hg methylation were apparently reached at this depth during summer. The speciation and biochemical availability of Hg as well as environmental factors (such as DO, pH, temperature, redox potential) here favored net Hg methylation (cf. Ullrich et al., 2001).

Regression analyses yielded inverse correlations when plotting MeHgT^sub unf^ and MeHgT^sub f^ vs. DO in the WJD, with Pearson correlation coefficients of -0.55 ($n = 111$, $p < 0.001$) and -0.40 ($n = 111$, $p < 0.001$), respectively. Similar relationships were also reported by He et al. (2008a). Reactive Hg and pH were significantly correlated with MeHgT^sub unf^ (RHg : $r = 0.40$, $n = 111$, $p < 0.001$; pH: $r = -0.31$, $n = 111$, $p < 0.001$) as well as with MeHgT^sub f^ (RHg : $r = 0.50$, $n = 111$, $p < 0.001$; pH: $r = -0.24$, $n = 111$, $p = 0.01$). This suggests that low DO and pH and high RHg are prerequisites for elevated MeHg concentrations in the WJD. Hence, elevated MeHg in the bottom water may be plausibly attributed to in situ methylation and/ or transfer from sediment to overlying waters; this is supported by the previous observation that sediment is the net source of MeHg to the water column (Feng et al., 2009b). No correlations were observed between MeHg and DO, pH, RHg in the YZD during any of the sampling seasons.

Spatial and seasonal distributions of PMeHg in the WJD showed little variation, with the exception of marked maxima in the bottom water at WX (12 pmol dm^sup -3^) and WD (4.9 pmol dm^sup -3^) in summer. These maxima may result from the release of MeHg from sediment resuspended into the water column. These results agree with previous observations that sediment resuspension may act as an additional MeHg source to water bodies (e.g., Mason and Sullivan, 1999; Lawson et al., 2001).

Neither the increased MeHg in bottom water nor the seasonal and vertical patterns of MeHgT^sub unf^, MeHgT^sub f^, and PMeHg observed in the WJD were observed in the YZD. Particulate MeHg maxima (with corresponding PMeHg/MeHgT^sub unf^ ratios at 83 and 91% for YZ and YD,

respectively) were observed in the YZD during summer (in the bottom water at YZ, and at 40 m at YD). Previous observations showed that dry/wet deposition of MeHgT^sub unf^ to the Wujiang River basin in wet seasons was much lower than in dry periods (Guo et al., 2008a,b). Therefore, neither atmospheric inputs nor in situ methylation can explain the maxima in PMeHg concentrations in the YZD. A more likely explanation is that terrestrial processes, such as erosion triggered by intense runoffepisodes during summer, were responsible for these observations.

Concentrations of MeHgT^sub unf^ in wet seasons were much higher than those in dry seasons in the YZD. A significantly positive correlation between MeHgT^sub unf^ and PMeHg/TSS was observed in all tributary rivers of the YZD (Guo, 2008; Guo et al., 2008c), indicating that river erosion and surface runoff are the main sources of elevated MeHgT^sub unf^ during summer in the YZD (Guo, 2008; Guo et al., 2008b). Annual average concentrations of MeHgT^sub unf^, MeHgT^sub f^, and PMeHg in the WJD were much higher than those in the YZD. This indicates that these two reservoirs, characterized by different levels of primary productivity, exhibit widely different strengths of Hg methylation and demethylation. A similar difference also appears to exist within the WJD, where only downstream stations (WX and WD) demonstrate high MeHgT^sub unf^ concentrations. Additional work is needed to demonstrate that the sediment is a source of Hg to water column, including analyses of Hg and MeHg in sediment cores and pore waters.

Previous studies have implied the presence of active MeHg production in newly constructed reservoirs and concluded that an enhanced methylation of Hg may last for >30 yr after impoundment (St. Louis et al., 2004; Hall et al., 2005; Lucotte et al., 1999). Studies in North America showed that Hg methylation rates decrease with the age of the reservoir as a result of the decomposition of organic carbon in flooded soils (St. Louis et al., 2004; Hall et al., 2005; Lucotte et al., 1999). However, the observations obtained from the WJD and the YZD tell a different story. The WJD (an old reservoir) is characterized by a much more active net Hg methylation compared with the YZD (a new reservoir). Furutani and Rudd (1980) found that organic material stimulates Hg methylation, hence diluting organic carbon concentrations in surface sediment by supplementing with inorganic material lowers MeHg production and bioaccumulation rates. Other studies have examined the differences in MeHg cycling in experimentally flooded wetland and upland catchments with varying carbon contents (Kelly et al., 1997; Bodaly et al., 2004).

Given the karstic environment of the Wujiang River basin, the organic carbon content (range: 2-5%) in submersed soils was very low (Jiang, 2007). A recent study indicated that most of the organic matter in the YZD is derived from the watershed, with little autochthonous material evident (Jiang, 2007). Primary productivity in the YZD is currently much lower than that in the WJD, apparently due to the absence of cage culture fishing in the YZD. In addition, lower organic carbon concentrations in the upland soils of the YZD may inhibit methylating microorganisms from colonizing the YZD or at least decrease their rate of metabolism. Hence, the low methylation rates in the YZD are a result of the low organic carbon content in submersed soils and/or low primary productivity. However, it seems clear that Hg methylation will increase with increases in primary productivity.

Filtered Total Mercury and Filtered Methylmercury in the Deep Interface Water of the Wujiangdu Reservoir and in Bottom Water of the Yinzidu Reservoir

The spatial and seasonal patterns of HgT^sub f^ and MeHgT^sub f^ in the interface water of the WJD are shown in Fig. 8 and 9. No discernable difference in HgT^sub f^ and MeHgT^sub f^ levels was observed in the YZD between the bottom water and the overlying water during the entire sampling seasons (Fig. 5 and 7). The concentration of HgT^sub f^ and MeHgT^sub f^ in the bottom water in the YZD is significantly lower than in the WJD (Supplementary Table S3).

Interface water HgT^sub f^ concentration downstream of the WJD was elevated compared with the upper stream of the WJD. The interface water HgT^sub f^ is plausibly perpetrated by diffusion of HgT^sub f^ from sediment water due to a continuous concentration gradient (Jiang, 2005; He et al., 2008a).

There was no discernible difference in MeHgT^sub f^ levels between the interface water and the overlying water at WS and WZ sites. The levels of MeHgT^sub f^ in the interface water represent maxima concentrations in seasonal vertical profiles at WX and WD sites, with the summer data of WD as an exception. Again, the baseline MeHg levels in interface water are probably due to the diffusion of MeHg from the surface layer of sediment (Furutani and Rudd, 1980; Mason et al., 1993). However, the enhancement in MeHgT^sub f^ at WX during the entire period of sampling and at WD during winter, fall, and spring demand an additional source, such as the in situ net Hg methylation and the diffusion of MeHg from the surface layer of sediment. To gain further support for this hypothesis, more detailed work is required to establish the temporal and spatial variability of Hg species in the sediment core and pore water with a spatial coverage from upstream to downstream of the reservoir.

The maximum MeHgT^sub f^ were not positioned at interface water but at the bottom of water column (95 m) at WD station during summer. The implication is that MeHg in this region is not only due to diffusion of MeHg from surface sediment and/or the accumulation of settling particulate matter, but also from in situ methylation in anoxic water (Meili, 1997; Eckley et al., 2005; He et al., 2008b). Diffusion of MeHg from the anoxic region (95 m) into the deep water is implied in this case. However, the data which support the hypothesis are limited. Thus, to better understand this observation, further work needs to be done.

The MeHg data indicated that Hg methylation was present during all sampling seasons at WD station, which is in agreement with earlier observation (Guo, 2008; Guo et al., 2008b; Feng et al., 2009a).

The ratios of MeHgT^sub f^/HgT^sub f^ in interface water (spring: 79%; summer: 39%; fall: 94%; winter: 86%) at WD station were elevated compared with overlying water (18 ± 18%), with the exception of a summer maximum of 94% presenting at 95 m. The elevated proportions of MeHgT^sub f^/ HgT^sub f^ in interface water are probably a result of active Hg methylation and/or the dissolution of oxides and anaerobic decomposition of particulate organic matter (Eckley et al., 2005; He et al., 2008b).

Cage aquaculture activities started from 1999 downstream of the WJD. The scale of the cage aquaculture activities increased (32,698-37,219 m^sup 2^; from 1999 to 2001), where the annual average fish feed used for cage aquaculture was about 9409 t (Sun et al., 2005). The minimum loss rate of fish feed is about 15% (Zhong et al., 2004), and the maximum digestive rate of fish for fish feed is about 40% (Peng, 2000). If those rates stand, to date the total lost fish feed in the WJD was 6.84 × 10^sup 7^ kg. In addition, Sun et al. (2005) quantified that TN and TP discharged due to the cage aquaculture activities in the WJD can reach to 2.78 × 105 kg yr^sup -1^ and 5.44 t × 10^sup 4^ kg yr^sup -1^, respectively. Hence, it is obvious that the cage aquaculture activities were the key factors for the high primary productivity in the WJD.

Phytoplankton-derived organic matter was suggested as an important source of organic matter present in surface sediment (Xiong and Yuan, 2006). Even submersed soils from the WJD, just as in the YZD, contain very low organic matter, being a consequence of the karstic environment in the Wujiang River basin (Jiang, 2007). In this study, organic carbon content in surface sediment collected from WX and WD was all relatively high (8.76-11.4%), in comparison with sediments from WS, WZ (3.74-5.47%), and the submersed soils (2-5%) (Table 1). Hence, the popularity of cage aquaculture appears to be the key factor in the high primary productivity in WX and WD, which would actually significantly change the conditions promoting net MeHg production to produce the large difference between the WJD and the YZD as well as between WS, WZ and WX, WD in terms of MeHg distribution in water. In all, the high primary productivity resulting from cage aquaculture activities is the key factor that controls the Hg methylation in reservoir.

Comparisons with Other Reservoirs

The magnitude of HgT^sub unf^, HgT^sub f^, and PHg concentrations in water samples from the WJD and the YZD was compared with data from the literature (listed in Table 2). Compared with other Chinese lakes/reservoirs, the concentrations of HgT^sub unf^, HgT^sub f^, and PHg in the WJD and the YZD are significantly lower. Therefore, the WJD and the YZD are apparently less impacted by local pollution sources.

It is also interesting that concentrations of MeHgT^sub unf^ and MeHgT^sub f^ in the water samples from downstream of the WJD appear higher than those reported for the other reservoirs and lakes worldwide, including Hongfeng reservoir, Caohai Lake, and Aha reservoir, which are located in the same region. Nevertheless, there is no discernable difference in MeHg levels between in the upstream and the middle of the WJD as well as the entire the YZD and uncontaminated global baseline waters. This further confirms that Hg methylation was amplified in the downstream of the WJD but not in the upstream and the middle of the WJD as well as the YZD.

Conclusions

The levels of HgT^sub unf^, HgT^sub f^, and PHg in water samples from the WJD and the YZD were similar to those reported for other lakes and reservoirs as well as those in remote areas in Europe and North America. The primary productivity that resulted from the cage aquaculture activities affected the degree to which net Hg methylation occurred in the reservoirs studied. Elevated MeHg concentrations at the bottom of the water column-interface water demonstrated the active Hg methylation in the downstream (WX and WD) of the WJD, which was caused by a high level of primary productivity. In contrast, no discernable Hg methylation was observed in the upstream (WS) and middle (WZ) of the WJD. Neither was there Hg methylation occurring in the YZD, which belongs to a low level of primary productivity. Our study demonstrated that primary productivity is an important factor that controls the potential of net Hg methylation and the distribution of mercury species in water.

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