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Seasonal distributions of mercury species and their relationship to some physicochemical factors in Puding Reservoir, Guizhou, China

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ABSTRACT

A comprehensive study was conducted in July 2006, January 2007 and March 2007 to determine the impacts of some major physicochemical parameters on the level of mercury (Hg) in Puding Reservoir, Guizhou, China. The concentrations of Hg species in the summer campaign were significantly higher ($p<0.01$, generally 2 to 3 times higher) than those in the winter and spring campaigns, and no statistical differences were found between the same parameters for the latter two campaigns ($p>0.05$). Ancillary parameters including suspended particulate matter (SPM), dissolved organic carbon (DOC), temperature (T), dissolved oxygen (DO), pH, nitrate (NO_3^-) and chloride (Cl^-) were also measured. During the sampling campaign in July 2006, average values for SPM, DOC, T, and NO_3^- were all higher compared to the other two campaigns, which suggested a similar seasonal trend between these parameters and Hg species. Seasonal variability may be related to increased runoff. High runoff volume due to abundant precipitation in the summer carried Hg-laden particulates into the reservoir, whereas there was less precipitation in the winter and spring when Hg levels were lower. Increased agricultural activity in the summer season also increased Hg levels in Puding Reservoir.

1. Introduction

Mercury (Hg) is a persistent, bioaccumulative, and toxic pollutant (USEPA, 1997; UNEP, 2002). Consumption of contaminated fish is regarded as the main exposure route to methylmercury (MeHg), which is one of the most toxic forms of Hg (Clarkson et al., 2003; Tchounwou et al., 2003) and therefore consumption of contaminated fish poses a significant risk to human health (Meili, 1991). A great number of studies reported the creation of hydroelectric reservoirs resulted in elevated concentrations of Hg in biota (e.g. Bodaly et al., 1984; Tremblay et al., 1998; Montgomery et al., 2000). However, the distribution, transformation and accumulation mechanisms of Hg in reservoirs are still poorly understood because of the complexity in the geochemical cycling of Hg.

The chemical composition of water depends on the geologic characteristics of the watershed, vegetation types and climate. Water quality is also related to anthropogenic activities within the watershed from both point and non-point source inputs (Lyons et al., 2006). Previous investigations clearly demonstrated that non-point runoff of Hg from anthropogenically-altered landscapes, such as agricultural and urban surfaces, are of great importance (Balogh et al., 1998; Mason and Sullivan, 1998; Warner et al., 2005).

Puding Reservoir is located in the Wujiang River watershed (Fig. 1), which is considered to be highly vulnerable to stresses within the watershed. In this area, the dominant land use is agricultural, and agricultural runoff is the primary source of contaminants to the reservoir (Shi and Xiang, 2007; Zhang, 2009). Soil erosion is also a serious problem in the area, which is caused by the cultivation activities, steep plateau gradient and stony desertification (Zhu, 2005). More chemical fertilizers are used in the farming compared to other areas in China, to balance the loss of nutrients by soil erosion.

In Puding Reservoir, the physicochemical water parameters associated with anthropogenic activities may help to reveal the source or distribution characteristics of Hg. The objective of this research was to measure the spatial and temporal variation of Hg species with the water column of Puding Reservoir and to identify the mechanisms associated with the distribution of Hg.

2. Materials and methods

2.1. Site description and sampling

Wujiang Watershed located upstream of Changjiang River, which is the longest river in China, has developed a number of hydroelectric reservoirs in the past decades. Puding Reservoir (26°23′N, 105°48′ E) was created in 1995, and is primarily fed by Saicha River and Boyu

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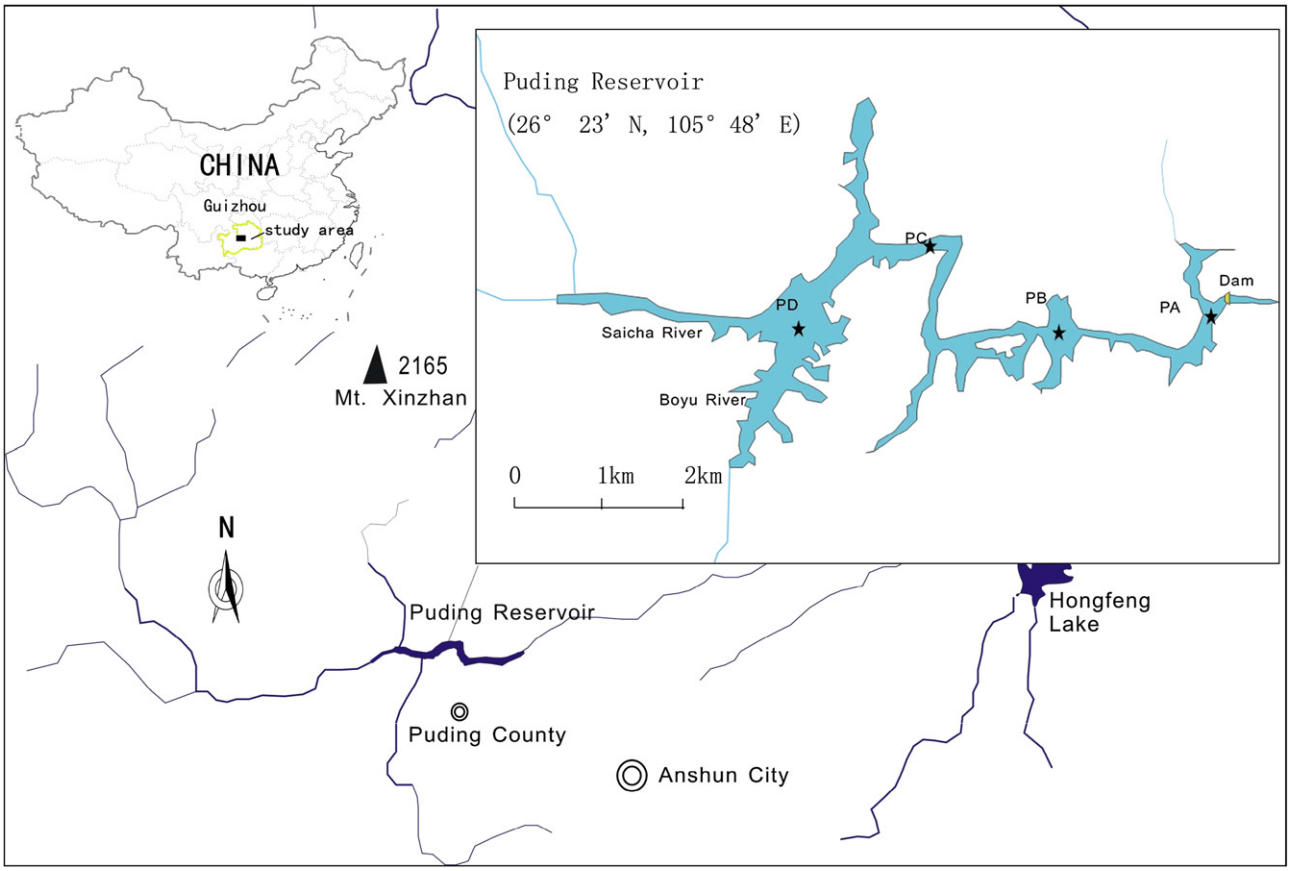

Fig. 1. Map of the Puding Reservoir, China, and the locations of the sampling sites. Station PD: upstream, station PC & PB: midstream, station PA: downstream.

River (Fig. 1). Less than 10% of water input is estimated to be from other tributaries according to the GWRD report (2008). It was constructed mainly for the purpose of hydroelectric power generation, flood control, tourism and to supply drinking water. The primary source of contaminants to the reservoir is from agricultural sources (Shi and Xiang, 2007; Zhang, 2009). The catchment of Puding Reservoir is 5871 km^2 at an altitude of 1400–2400 m. The climate is a typical subtropical humid monsoon and the rainy period extends from May to October, which yields about 75% of the annual precipitation (~1000 mm) (Zhu, 2005). Local agricultural activity mainly occurs during the period from April to September. The major soil types in the catchment are characterized by limestone soil and ultisol. The information on land use and reservoir parameters is presented in Table 1 (Liu and Chen, 2006; Zhang, 2009).

Table 1
Hydrological characteristics of the Puding Reservoir and land use of the catchment area.

Parameter	Unit	Value
Surface area	km^2	19.25
Drainage area	km^2	5871
Water capacity	$10^6 m^3$	420
Average water volume	$10^6 m^3$	248
Maximum depth	m	45
Mean depth	m	13
Theoretical residence time	days	27
Arable land coverage of the catchment area	%	35.9
Dry land coverage of the arable land	%	80.0
Forest coverage of the catchment area	%	21.7
Slope land (>8°) coverage of the catchment area	%	>80
Steep slope land (>25°) coverage of the arable land	%	11.6
Soil erosion area coverage of the catchment area	%	48.0

Four sampling locations were selected within the reservoir (Fig. 1). One site was located in the upstream section of the reservoir (PD), two sites were located in the middle of the reservoir (PB and PC) and one site was located in the downstream section of the reservoir 500 m from the dam (PA). The field sampling campaigns were carried out in July 2006, January 2007 and March 2007, representing the summer, winter and spring seasons, respectively. Sampling site PA is close to the dam, and therefore water depths fluctuate extensively; water depth was as high as 45 m in the summer (rainy season), and decreased to 28 m in the spring (dry season).

2.2. Cleaning and sampling procedures

Water sampling equipment was processed using Hg-contamination-free techniques (Baeyens, 1992; Montgomery et al., 1995). All borosilicate glass bottles, 100 mL in volume, were cleaned by acid-washing, rinsing with ultrapure deionized water (18.2 MΩ cm, Milli-Q) and heated to remove Hg at 500 °C, and filled with Milli-Q water and 0.5 mL sub-boiling distilled HCl, and then double-bagged until use. At each sampling site, the bottles were emptied and rinsed several times with reservoir water, then filled and preserved with sub-boiling distilled HCl to yield 0.5% acid solutions, and tightly sealed and double-bagged and stored in a wood box. After transport to the laboratory, the samples were stored at 4 °C in the dark until the analysis.

We collected water samples at different depths of the water column by using Van-Dorn water sampler from a boat in the middle of the reservoir. Water samples were collected at depth intervals of 5–20 m depending on the depth of water. Both filtered and unfiltered water samples were collected. The filtered samples were prepared by using a 0.45-μm filter (Millipore) on site, and all samples were filled in borosilicate glass bottles. Additional samples for

ancillary chemistry analysis were collected using appropriate bottles and preserved as necessary (e.g., Welch et al., 1996; Carey et al., 2005).

2.3. Analytical methods

Hg species such as THg, dissolved Hg (DHg), total methylmercury (TMeHg), and dissolved methylmercury (DMeHg), were analyzed for each sample. The analytical methods used for Hg speciation in water have been described in detail elsewhere (Bloom and Fitzgerald, 1988; Bloom, 1989; Horvat et al., 1993; U.S. EPA, 2001, 2002; Yan et al., 2003; He et al., 2008), which were all based on cold vapor atomic fluorescence spectrometry (CVAFS). Hg associated with the particulate matter (PHg), is defined as the THg minus the DHg.

Samples for THg and DHg analysis were digested by BrCl oxidation. After oxidation, $NH_2OH{\cdot}HCl$ was added to destroy the free halogens before adding stannous chloride ($SnCl_2$) to convert Hg(II) to volatile Hg(0). The resulting sample was then purged with Hg-free N_2 and Hg(0) was absorbed onto a gold trap and analyzed by CVAFS (Gill and Fitzgerald, 1987; Bloom and Fitzgerald, 1988; Yan et al., 2003).

TMeHg and DMeHg concentrations in water were determined using the standard distillation–ethylation–GC separation–CVAFS technique (Bloom, 1989; US EPA, 2001). The distillation of a 45-mL acidified aliquot was carried out at 125 °C under Hg-free N_2 flow until approximately 35 mL of water was collected in the receiving vessel. The sample after being pH adjusted and ethylated was then purged with N_2 onto a Tenax trap. The trapped ethyl analog of CH_3Hg, $CH_3CH_3CH_2Hg$ was thermally desorbed, separated in GC column, and then decomposed to Hg (0) and analyzed by CVAFS.

Quality assurance and quality control of the analytical processes were performed by using field blanks, system blanks, spike recoveries and sample duplicates. Field blanks and duplicates were taken regularly (>10%) throughout each sampling campaign (Yan et al., 2005). Detection limits were estimated as three times the standard deviation of the blank measurement and were 0.10 ng/L for total Hg (THg), 0.029 ng/L for MeHg, respectively. The reproducibility for THg had an analytical precision (coefficient of variation) of 2% to 6%, and was <11.0% for MeHg. Recoveries for matrix spikes were in the range of 88.1%–111.0% for THg analysis, and 88.5%–110.9% for MeHg analysis.

Water quality parameters such as temperature (T), pH and dissolved oxygen (DO) were measured in-situ using portable field test kit (plONneer 65). DO was measured using the test kit after calibration in 100% air saturation and the DO value was temperature and pressure compensated. The accuracy of DO measurement was within ±0.2 mg/L. Dissolved organic carbon (DOC) was measured by the high-temperature combustion method (Cosovic et al., 2000). Suspended particulate matter (SPM) was measured in laboratory using gravimetric method (China EPA, 1989). Water anions such as chloride (Cl^-), nitrate (NO_3^-) were detected by ion chromatography (Dionex).

3. Results and discussion

3.1. Temporal and spatial trends

The statistical summary of the concentrations of THg, DHg, PHg, TMeHg, DMeHg and PMeHg for sampling conducted during July 2006, January 2007 and March 2007 (the summer, winter and spring season respectively) are presented in Fig. 2 and Table 2.

The concentrations of all Hg species in the rainy summer season were significantly higher ($p<0.01$) than those during the dry winter and spring seasons, while there was no statistical differences between the latter two seasons ($p>0.05$). Some discernable differences were found in the spatial distribution (between upstream and downstream) as well within the water column.

As shown in Table 2, during the campaign of July 2006, the average values of SPM, DOC, T, NO_3^- were higher compared to the other two campaigns, which suggested a similar seasonal trend governed Hg species and these ancillary parameters. However, DO, pH and Cl^- did not follow this seasonal trend.

In order to understand the stratification status of the reservoir, we presented the distribution of the DO, water temperature and pH in the reservoir as shown in Fig. 3. pH showed significant differences in the vertical profiles in the summer while no significant distribution patterns were found for the other two seasons. DO decreased in the water column in the summer and spring and remained stable in

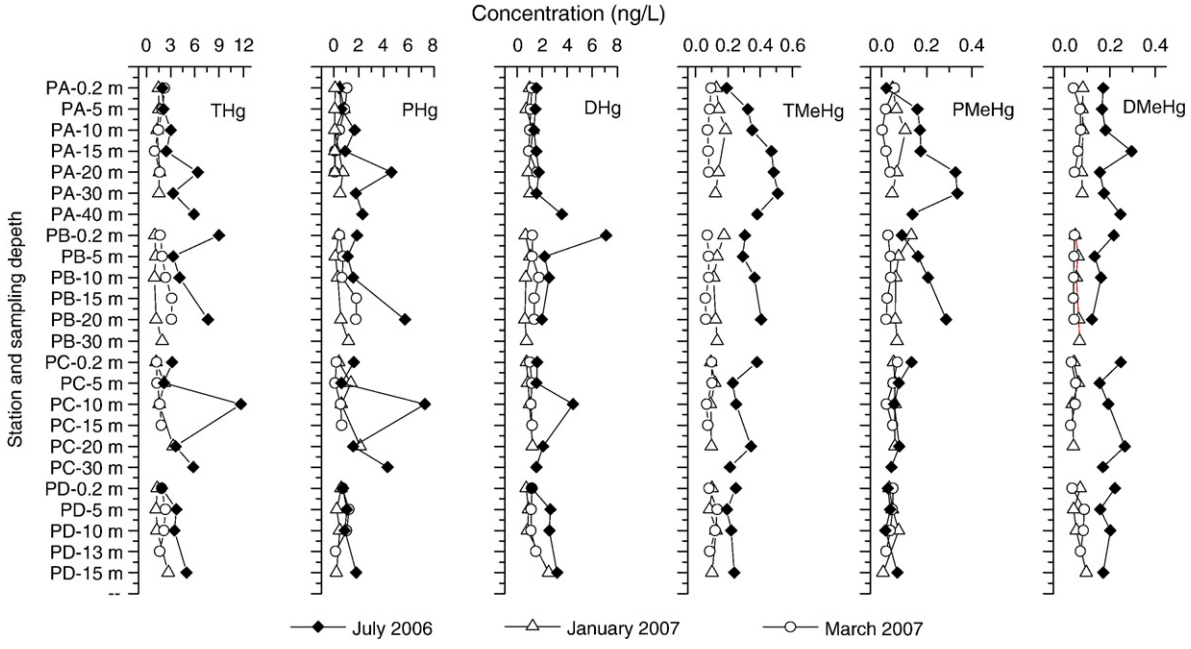

Fig. 2. Concentrations of THg, PHg, DHg, TMeHg, PMeHg and DMeHg in the PA, PB, PC, PD stations of the Puding Reservoir. Sampling was conducted during July 2006, January 2007 and March 2007.

Table 2
The statistical summary of the concentrations of different mercury species and the ancillary parameters for the Puding Reservoir sampled during July 2006, January 2007 and March 2007.

	PHg(ng/L)	DHg(ng/L)	PMeHg(ng/L)	DMeHg(ng/L)	SPM(mg/L)	DOC(mg/L)	T(°C)	DO(mg/L)	pH	NO_3^-(mg/L)	Cl^-(mg/L)
July 2006 number = 20											
Mean ± SD	2.11 ± 1.86	2.37 ± 1.40	0.13 ± 0.10	0.19 ± 0.05	5.45 ± 1.84	1.86 ± 1.83	23.7 ± 2.5	8.38 ± 2.86	7.83 ± 0.26	13.8 ± 1.17	3.12 ± 0.53
Min–Max	0.43–7.27	1.17–7.11	0.02–0.33	0.12–0.29	1.43–8.75	0.38–8.15	20.7–28.7	4.14–13.5	7.45–8.31	12.3–17.6	2.58–4.26
January 2007 number = 18											
Mean ± SD	0.62 ± 0.50	0.97 ± 0.43	0.06 ± 0.03	0.06 ± 0.02	3.74 ± 1.15	1.12 ± 0.41	9.3 ± 0.6	8.58 ± 2.17	7.76 ± 0.43	11.1 ± 0.58	4.58 ± 0.88
Min–Max	0.09–2.12	0.61–2.52	0.01–0.13	0.03–0.10	1.23–5.90	0.65–2.05	7.90–10.30	6.57–13.06	6.53–8.53	9.43–12.2	3.26–6.25
March 2007 number = 18											
Mean ± SD	0.70 ± 0.52	1.23 ± 0.21	0.04 ± 0.02	0.05 ± 0.02	3.38 ± 0.86	0.90 ± 0.06	16.3 ± 1.7	5.34 ± 1.26	8.05 ± 0.33	6.16 ± 2.42	3.49 ± 1.38
Min–Max	0.09–1.80	0.92–1.73	0.00–0.07	0.03–0.09	2.08–4.85	0.81–1.01	14.0–20.6	3.07–7.47	7.37–8.78	2.53–8.68	1.35–4.94

winter, except for the DO in surface water due to atmospheric aeration. Water temperatures, DO and pH trends indicated there was an isotherm in the winter, with stratification beginning in early spring and was fully stratified in summer.

3.2. Distribution of total, particulate and dissolved Hg

Spatial and temporal distributions of THg, PHg and DHg in each sampling site are presented in Fig. 2. THg aqueous concentrations ranged from 1.00 to 11.74 ng/L with a mean (±SD) concentration of 2.74 ± 2.07 ng/L, which were comparable with levels reported for reservoirs in North America (e.g. Brigham et al., 2002). The levels of THg and PHg were relatively higher in the summer compared with those in the winter and spring (Figs. 2 and 4). The highest average concentrations of THg and PHg were observed in July at the midstream sites PB (6.01 ng/L) and PC (3.06 ng/L), respectively. Similarly, highly elevated concentrations of SPM also occurred in July at PB (average = 6.26 mg/L) and 5.78 mg/L at PC. The lowest averages of THg (1.26 ng/L) and PHg (0.38 ng/L) were also found at the midstream sites (PB in the winter and PC in the spring respectively), which corresponded to lower SPM averages of 3.64 and 3.74 mg/L respectively. In addition, both THg and PHg peak values matched the highest concentrations of SPM.

This seasonal distribution of Hg could be a result of agricultural runoff. In July, high runoff volume due to abundant precipitation carried Hg-containing particulates into the reservoir, whereas there was less precipitation in the winter and spring. This observation was verified by the positive relationship between THg and SPM ($r = 0.47$, $p < 0.01$) and PHg and SPM ($r = 0.49$, $p < 0.001$), which suggested SPM may play an important role in the distribution of THg and PHg. The highest THg and SPM concentrations found in the middle of the reservoir (instead of the upstream section) may be explained by specific catchment topography. In the middle section of the reservoir, the catchment topography is generally steeper than the upstream and downstream sections. In addition, agricultural activities in the steep slopes accelerated soil erosion to the middle section, where THg and SPM concentrations were higher.

The DHg concentrations varied from 0.61 to 7.11 ng/L and showed a strong positive relationship with concentrations of THg ($r = 0.80$, $p < 0.001$). Although a significant correlation ($r = 0.31$, $p < 0.05$) was found between DHg and SPM, it was still insufficient to believe SPM was the source of DHg. DHg accounts for a major fraction of THg during each

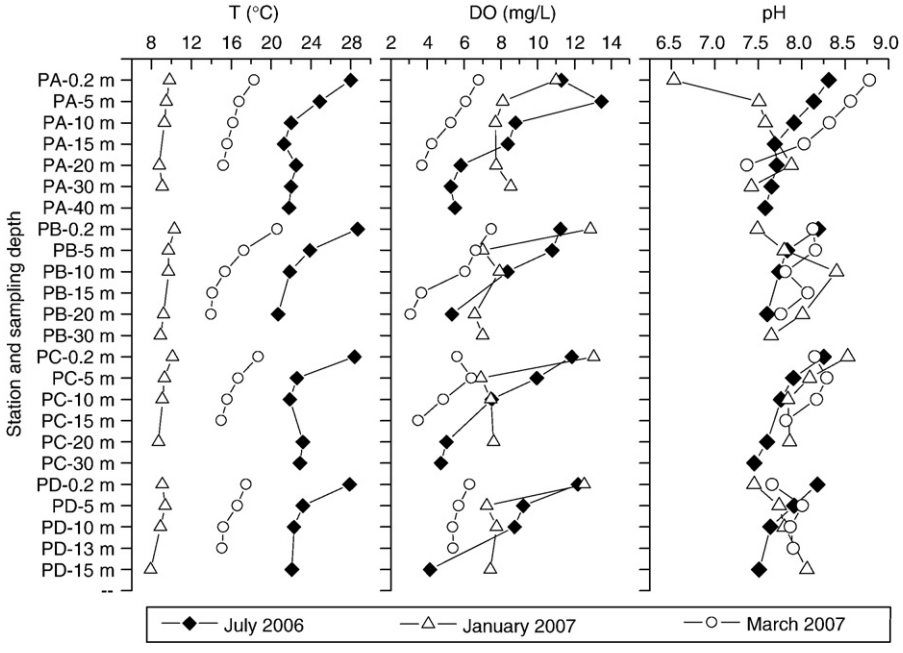

Fig. 3. Seasonal distribution of DO, pH and water temperature (T) in the Puding Reservoir.

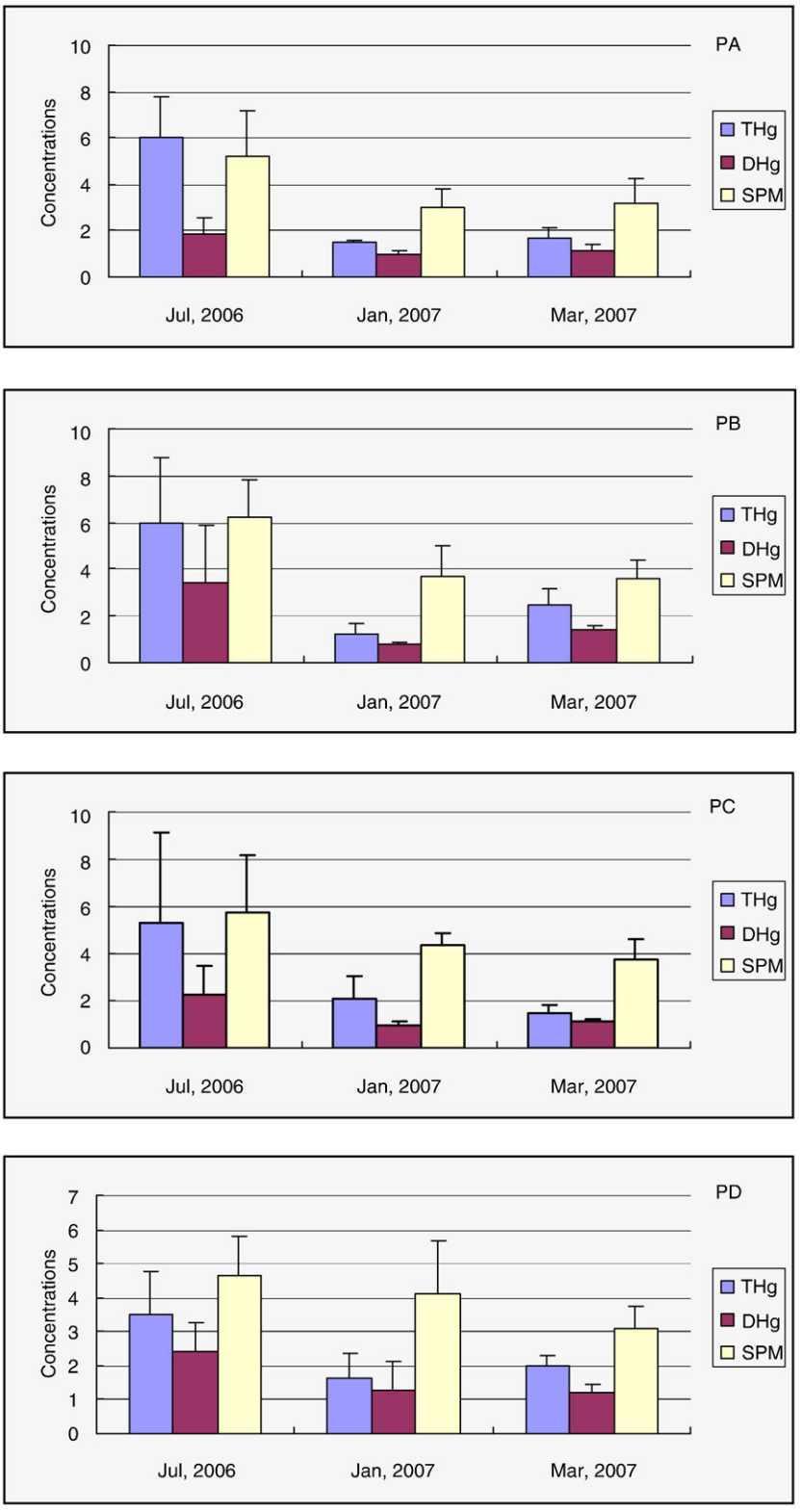

Fig. 4. Seasonal comparisons of the average concentrations of THg, DHg and SPM at each site in the Puding Reservoir from July 2006 to March 2007. The unit for the THg and DHg is ng/L, and for SPM is mg/L.

sampling period. Mean DHg/THg ratio in July was 56.9%, while the ratios were elevated to 63.1 and 67.5% in winter and spring, respectively. These ratios are similar to previous research in Hongfeng Reservoir (He et al., 2008), one of the reservoirs in the Wujiang watershed. The proportion of DHg increased in the winter and spring probably because of the reduced precipitation, which resulted in reduced particles (or SPM).

3.3. The distribution of total, particulate and dissolved methylmercury

The seasonal distributions of MeHg in water column are shown in Fig. 2. MeHg species showed statistically significant differences ($p<0.01$) between the summer campaign and the other two campaigns. TMeHg in July ranged from 0.19 to 0.51 ng/L with an

average (±SD) of 0.32±0.10 ng/L, which was higher than those in the winter and spring (Table 2). The highest value of 0.51 ng/L occurred at PA and was 2.74 times higher than the highest value in the other seasons. The spatial and temporal distributions of TMeHg (Fig. 2) showed that TMeHg increased significantly in the hypolimnion in July, especially at the PA site, where TMeHg increased from 0.19 ng/L at the surface to 0.51 ng/L in the hypolimnion.

Many studies have shown that increased methylation in water column would be expected to occur in anoxic conditions because sulfur reducing bacteria (SBR) will be active when oxygen is depleted (Ullrich et al., 2001; Eckley and Hintelmann, 2006). However in our research, no obvious anoxic conditions were found in the hypolimnion in July (Fig. 3), and DMeHg, which may well reflect this methylation, showed no visible increase trend in the hypolimnion (Fig. 2).Though TMeHg showed the increasing trend in July, it is not convincible to say the hypolimnion methylation commonly happened. In July, the linear correlation coefficient between TMeHg and PMeHg was higher ($r=0.89$, $p<0.001$) compared to TMeHg and DMeHg ($r=0.23$, $p<0.001$), which suggested PMeHg in July likely determined the distribution of MeHg. Bloomed phytoplankton in July can absorb MeHg from water, which may explain the higher PMeHg concentrations (Hurley et al., 1994).

DMeHg concentrations ranged from 0.12 to 0.29 ng/L with an average (±SD) of 0.19±0.05 ng/L in the summer, which was significantly higher than those in the winter and spring (Table 2). DMeHg accounted for a major fraction of TMeHg during each sampling period. The highest average fraction (63.8%) was found in the rainy season compared with 50.4% in winter and 59.5% in spring. During the dry seasons (winter and spring) the majority of THg in the reservoir was in the dissolved phase, however during the rainy season a higher proportion of THg was bound to particles, which suggested the source of the Hg was from erosion within the watershed. The increase in MeHg in the dissolved phase during the summer suggested that MeHg was produced within the reservoir as opposed to being transported into the reservoir from the watershed.

In order to further investigate MeHg production, we calculated %MeHg (the percentage of MeHg to THg) used by several studies as a surrogate measurement for methylation activity (e.g. Gilmour et al., 1998; Mitchell et al., 2008). In the dry seasons (winter and spring), %TMeHg and %DMeHg were 6.8±3.3% and 3.5±1.5% respectively, which increased to 9.0±4.5%, 5.5±3.0% in the summer, indicating the MeHg production was taking place within the reservoir. In many studies, these seasonal changes have been most commonly attributed to increased methylation — in the water column as well as the sediment (e.g Gilmour and Henry, 1991). Since the water column was not anoxic, seasonally increased MeHg production in the reservoir probably occurred in anoxic bottom sediments. Both re-suspension of sediment and diffusion of MeHg from sediment pore water can bring MeHg to water column (Feng et al., 2009). In addition, increased temperature in July possibly contributed to increased methylation in anoxic bottom sediments, which produced higher %MeHg in the water column in July. An early study (Guo, 2008) measured monthly inflows and outflows of MeHg in Puding Reservoir in 2006, which showed that ~2 times more MeHg was exported in the summer than was imported, and this was ~2 times higher than in the other seasons. In addition Guo (2008) reported the net MeHg flux was 69.4 g/a. Results from the present study confirmed that MeHg was produced within the reservoir, especially in the summer season.

3.4. Hg and physicochemical factors

The distribution and occurrence of Hg species in aquatic environments are regulated by many chemical and biological parameters (Gill and Bruland, 1990; Bloom and Etfler, 1990). MeHg content in water is influenced by a wide variety of environmental factors, such as the total and reactive Hg content, water temperature, redox potential, pH, the inorganic and organic solutes and microbial activity in waters (Ullrich et al., 2001). For the purpose of investigating possible controlling factors, the correlations between Hg species and seven ancillary water quality parameters (SPM, DOC, DO, T, pH, Cl^-, NO_3^-) were studied for all data sets during all 3 sampling campaigns. The correlation matrix is showed in Table 3. SPM, T and NO_3^- were found to be more correlated with the distribution of Hg species in the reservoir, while DOC, DO and pH were less important. Cl^- was also significantly negative correlated with a few Hg species.

Nitrate, which can be used as a measure of watershed runoff, ranged from 2.53 to 17.6 mg/L in the Puding Reservoir. The highest concentration was observed in the summer, which was much higher than levels observed in the Scioto River, where Hg levels were elevated due to the agricultural activities and urbanization (up to 11 mg/L) (Lyons et al., 2006). From Table 3, nitrate concentrations were significantly positively related to different Hg species. This could be explained by agricultural activities. In the summer season, agricultural activities were associated with increased usage of chemical fertilizer, including nitrate. Higher runoff from precipitation transported nitrate (from fertilizer) together with Hg-containing particulate matter (from erosion) to the reservoir. NO_3^- and Hg concentrations were lower in the winter or spring when both the precipitation and agricultural activities decreased. The concentrations of Hg species varied exponentially (Fig. 5) with the concentrations of NO_3^- which roughly reflected the frequency of agricultural activities.

Table 3
Pearson's correlation matrix, giving the linear correlation coefficients (r) between the variables.

	THg	DHg	PHg	TMeHg	DMeHg	PMeHg	SPM	DOC	T	DO	pH	NO_3^-	Cl^-
THg	1												
DHg	0.81***	1											
PHg	0.89***	0.45***	1										
TMeHg	0.57***	0.48***	0.50***	1									
DMeHg	0.41**	0.46**	0.39**	0.74***	1								
PMeHg	0.37**	0.16	0.45***	0.85***	0.41**	1							
SPM	0.47**	0.31*	0.49***	0.45***	0.27*	0.48***	1						
DOC	0.04	0.15	−0.05	0.22	0.23	0.07	0.10	1					
T	0.52***	0.55***	0.36**	0.72***	0.57***	0.33**	0.16	0.28*	1				
DO	−0.08	0.06	−0.18	0.18	0.29*	0.08	0.18	0.32*	0.14	1			
pH	−0.13	−0.04	−0.17	−0.15	−0.29*	−0.21	0.05	0.07	0.18	0.06	1		
NO_3^-	0.39**	0.36**	0.32*	0.67***	0.73***	0.46***	0.25*	0.20	0.34**	0.33**	−0.28*	1	
Cl^-	−0.31*	−0.23	−0.30*	−0.39**	−0.22	−0.24*	−0.24*	−0.17	−0.49***	0.03	0.07	0.21	1

* $p<0.05$.
** $p<0.01$.
*** $p<0.001$.

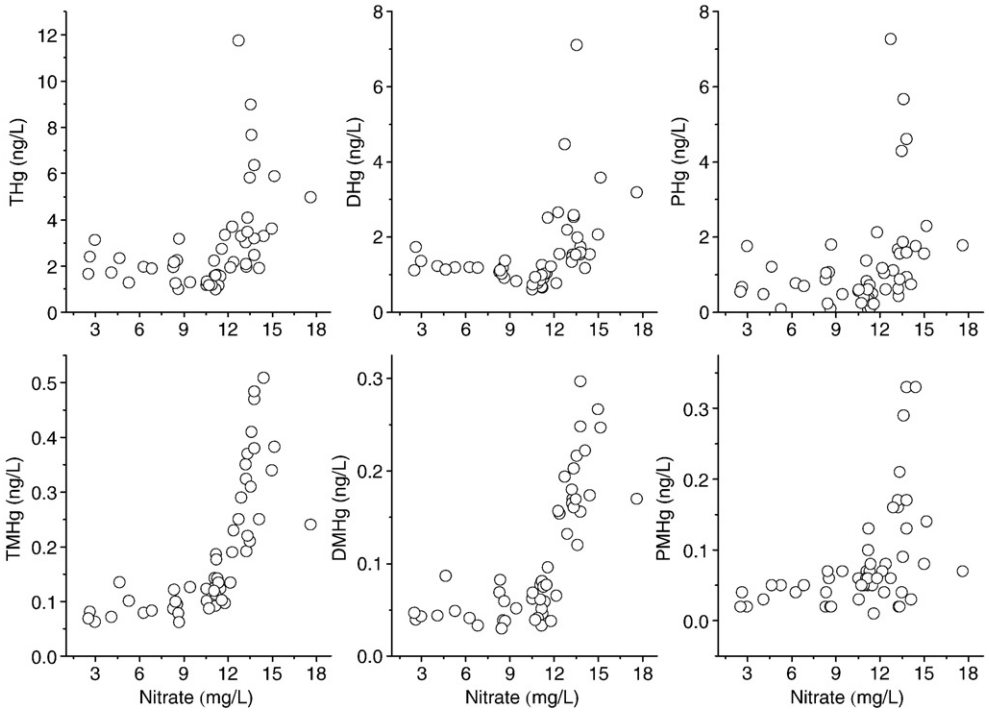

Fig. 5. The correlations between nitrate concentration and the concentrations of different Hg species.

These results suggest that the agricultural activity in the summer could increase Hg levels in the Puding Reservoir.

Warmer temperatures (T) may increase microbial Hg methylation (Hecky et al., 1991). In our investigation, the water temperatures were significantly and positively correlated with different Hg species as shown in Table 3. Though the seasonal differences observed for MeHg can be related to SPM, they are also likely influenced by water temperature. With the seasonal mean temperature differences of up to 14.4 °C between summer and winter, and 7.4 °C between summer and spring, significantly lower DMeHg and PMeHg in the winter and spring campaigns (Fig. 2) may be attributed to the reduced microbial methylation (Winfrey and Rudd, 1990; Bodaly et al., 1993; Ramlal et al., 1993).

Suspended particulate matter (SPM), which is significantly related to the Hg species (Table 3), appears to play a key role in the seasonal distribution of Hg species and has been fully discussed in the text. Though DO had a significant positive relationship ($r=0.29$, $p<0.05$) with DMeHg, this positive relationship is believed to be an accidental coincidence since decreased DO favors the methylation of Hg (He et al., 2008). The range of pH values was limited, from 6.53 to 8.78. A significant negative correlation was obtained between pH and DMeHg. Concentrations of DOC in the Puding Reservoir ranged from 0.38 to 8.15 mg/L (Table 2). However, DOC concentrations were poorly related to the dissolved fractions of Hg (Table 3), indicating that only a small fraction of DHg was complexed to dissolved organic ligands (Bonzongo et al., 1996). Therefore, it can be assumed that a large fraction of DHg in the studied area is available for conversion processes and uptake by aquatic organisms.

Chloride (Cl^-) concentration is considered to be higher in the more urbanized portions of water bodies due to domestic wastewater discharges (e.g. from Lyons et al. (2006), a peak Cl^- concentration of 52 mg/L was found downstream of sewage treatment facilities). For this study, the peak concentration of 6.25 mg/L was observed in the winter campaign, and in general, Cl^- averages were higher in the winter and spring and lower in the summer. The latter result may be due to the dilution effect from precipitation (since most runoff is from agricultural activities, not urban runoff). High precipitation or runoff in July diluted the Cl^- concentrations in the reservoir, while Hg levels were not diluted due to high SPM levels input to the reservoir. Hence, significantly negative correlations between Cl^- and some of Hg species were found as shown in Table 3.

4. Conclusions

We investigated the seasonal distribution of Hg species and some physicochemical factors in the Puding Reservoir in three seasons. Trends of Hg concentrations observed showed that the concentrations of all the Hg species (PHg, DHg, PMeHg and DMeHg) in the summer campaign are significantly higher ($p<0.01$, generally 2 to 3 times higher) than those in the winter and spring campaigns. We found that SPM, T and NO_3^- had significantly positive correlations with Hg species in the reservoir. During the rainy season in the summer, high runoff volume due to abundant precipitation carried Hg-containing particulates eroded from the watershed into the water body. It was also found that agricultural activity in the summer season potentially enhanced the Hg levels in the Puding Reservoir.

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