



## Geochemical processes of mercury in Wujiangdu and Dongfeng reservoirs, Guizhou, China

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Surface sediment in the reservoirs is the active mercury methylating sites in the systems.

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### ABSTRACT

The geochemical processes of mercury in Dongfeng (DF) and Wujiangdu (WJD) reservoirs, which were constructed in 1992 and 1979, respectively in Wujiang River, which is the upper branch of Yangtze River were investigated. One sampling site was chosen upriver of 1 km from the dam for each reservoir. Three sampling campaigns were conducted at these sampling sites in December 2003, April 2004 and July 2004, respectively. The distributions of different mercury species in the water column, sediment, and sediment pore water were studied. We found that the sediment is the net source of both inorganic and MeHg to the water column for both reservoirs. The MeHg diffusion fluxes in WJD reservoir at all sampling campaigns were significantly higher than those in DF reservoir. Our study demonstrated that the high primary productivity in the reservoir produced elevated organic matter content that would favor the methylmercury production in sediment.

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### 1. Introduction

One of the important environmental consequences of creating reservoirs for various purposes, such as hydroelectric generation, flood control, irrigation, fisheries production, and recreation, is the contamination of methylmercury (MeHg) to the food web of the aquatic system (Smith et al., 1974; Abernathy and Cumbie, 1977; Cox et al., 1979; Lucotte et al., 1999; St. Louis et al., 2004; Hall et al., 2005). For instance, MeHg concentrations in predatory fish harvested from northern boreal reservoirs in Manitoba (i.e. Bodaly et al., 1984), Québec (Brouard et al., 1994; Schetagne, 1999), and Newfoundland (Scruton et al., 1994), Canada, as well as in Finland (Lodenius et al., 1983), often exceed  $0.5 \text{ mg kg}^{-1}$  wet mass, which is the food advisory limits for many countries, for more than 20 years after initial flooding. Mercury concentrations in fish from Gezhouba reservoir, Yangtze River were also elevated (Jin and Xu, 1997). MeHg is an important neurotoxin to human because it crosses without hindrance the blood–brain and placental barriers to reach its principal target tissue, the brain, creating irreversible damages to the nervous system (Clarkson, 1993).

The decomposition of organic carbon in flooded soils in reservoirs is believed to fuel the microbial methylation of inorganic Hg to MeHg (Compeau and Bartha, 1983; Kelly et al., 1997; Hall et al.,

2004). The latest study (Hall et al., 2004) demonstrated that the amount of organic carbon stored in a reservoir prior to flooding is not a good indicator of the extent of future MeHg increases. However, the Hg methylation rates will decrease with the increase of the ages of the reservoirs as the result of the decomposition of organic carbon in flooded soils (e.g. St. Louis et al., 2004; Lucotte et al., 1999; Hall et al., 2005). Apart from the ages of reservoirs, many other factors may also govern the net MeHg production rate in reservoirs. Soil types including organic carbon and Hg concentrations in flooded soil, the ratio of flooded area and water volume, water chemistry, water temperature, and water residence time in reservoirs (Therriault and Schneider, 1998; Montgomery et al., 2000; St. Louis et al., 2004) are important parameters that may control Hg methylation rates in reservoir systems. In order to better understand and model Hg transportation from the aquatic systems to food chains in reservoir, the geochemical processes of Hg (both THg and MeHg) in reservoir system need to be fully understood. However, this kind of information is still limited, especially in China, where a great number of new reservoirs have been created.

China started to build reservoirs since 1949 mainly for flood control and power generation, and up to 2000 the total number of large dams reached 24,119, which is more than half of the total number of large dams over the world (The World Commission on Dams, 2000). Many more dams have been created since 2001 in western part of China. However, studies of the biogeochemical cycling in reservoirs in China are very limited and information to assess the eco-environmental impacts of methylmercury

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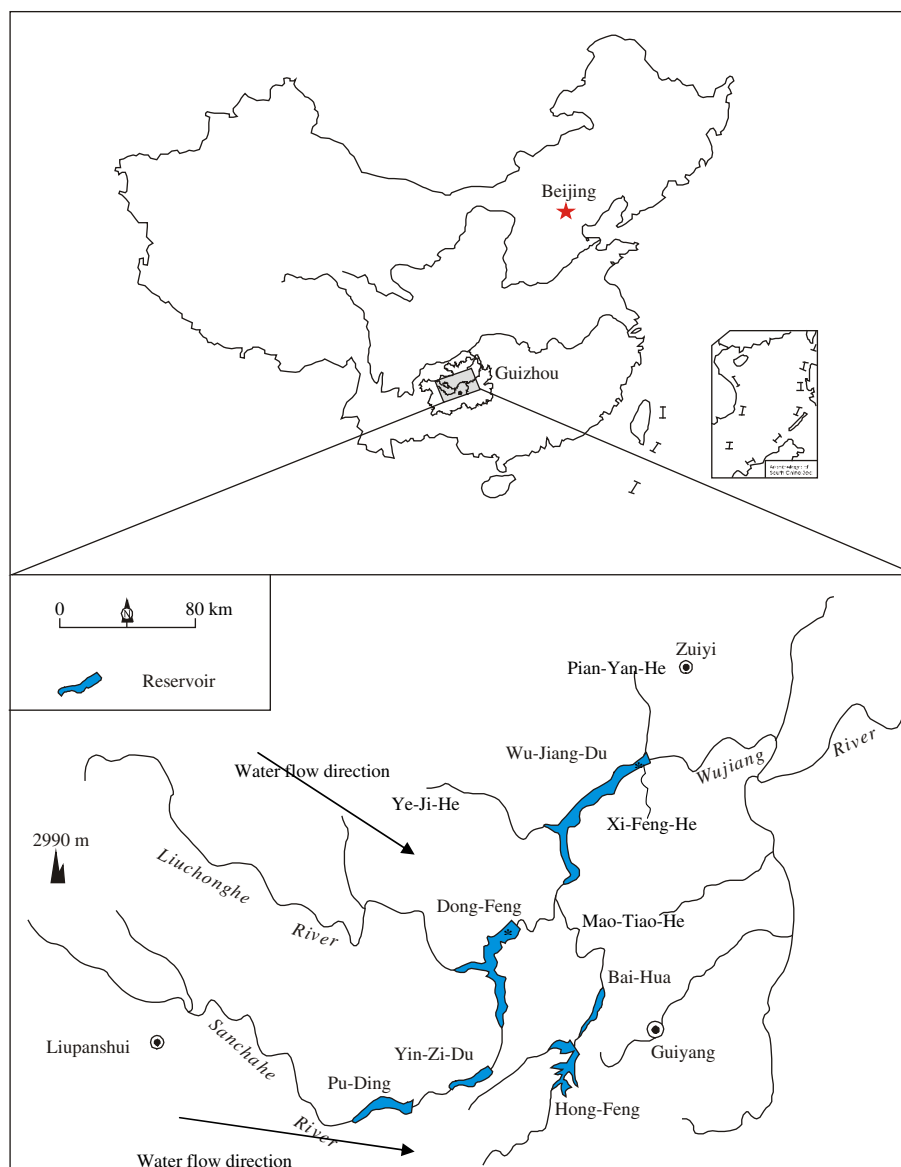


Fig. 1. Location of study area in Wujiang river, Guizhou, China.

contamination in newly built reservoirs in China is largely unavailable. From October 2003 to September 2004, we for the first time conducted an intensive study on mercury biogeochemical cycling in Wujiangu (WJD) and Dongfeng (DF) Reservoirs which are adjacent reservoirs created on Wujiang River. In a companion paper (Feng et al., 2009), we reported the mass balance study of both THg and MeHg in these two reservoirs, and we found that both reservoirs are the net sinks for THg, but net sources for MeHg. Furthermore, we observed that the MeHg yield in WJD reservoir ( $140.9 \text{ g MeHg km}^{-2} \text{ yr}^{-1}$ ) was much higher than that of DF reservoir ( $32.9 \text{ g MeHg km}^{-2} \text{ yr}^{-1}$ ) (Feng et al., 2009). In this paper we studied the biogeochemical processes of Hg in two reservoirs to better understand the controlling factors of methylmercury production in reservoirs created in Wujiang catchments.

## 2. Materials and methods

### 2.1. Site description

The detailed description of WJD and DF reservoirs were given by Feng et al. (2009). Briefly, WJD and DF reservoirs are located on the Wujiang River, which is

a branch of Yangtze River, in Guizhou Province, Southwestern China (Fig. 1). The outlet of DF reservoir flows into WJD reservoir. Before flooding, there were agriculture farmlands distributed along the valleys. The reservoirs lie on the Yunnan-Guizhou Plateau with altitudes varying from 700 to 1200m above sea level. The bedrocks of the watershed of the two reservoirs mainly consist of limestone and dolomite. Its climate represents a typical subtropical humid monsoon with an average temperature of  $13.4^\circ\text{C}$  and an average annual rainfall of 1130 mm. The rainy season covers from May to October, and more than 70% of the annual precipitation occurs in this period of time.

The basic characteristics of the reservoirs are listed in Table 1. Both reservoirs are large dams according to the definition given by the World Commission on Dams

Table 1  
Basic parameters of Dongfeng and Wujiangdu reservoirs.

	unit	Dongfeng	Wujiangdu
Construction time		1992	1979
Watershed area	$\text{km}^2$	18,161	27,790
Average annual flow	$\text{m}^3 \text{ s}^{-1}$	355	502
Flow of total suspended solid	$10^6 \text{ t a}^{-1}$	12.6	15.3
Height of dam	m	168	165
Total water volume	$10^6 \text{ m}^3$	1025	2300
Surface area of the reservoir	$\text{km}^2$	19.1	47.8
The average water residence time	day	33.4	53.0

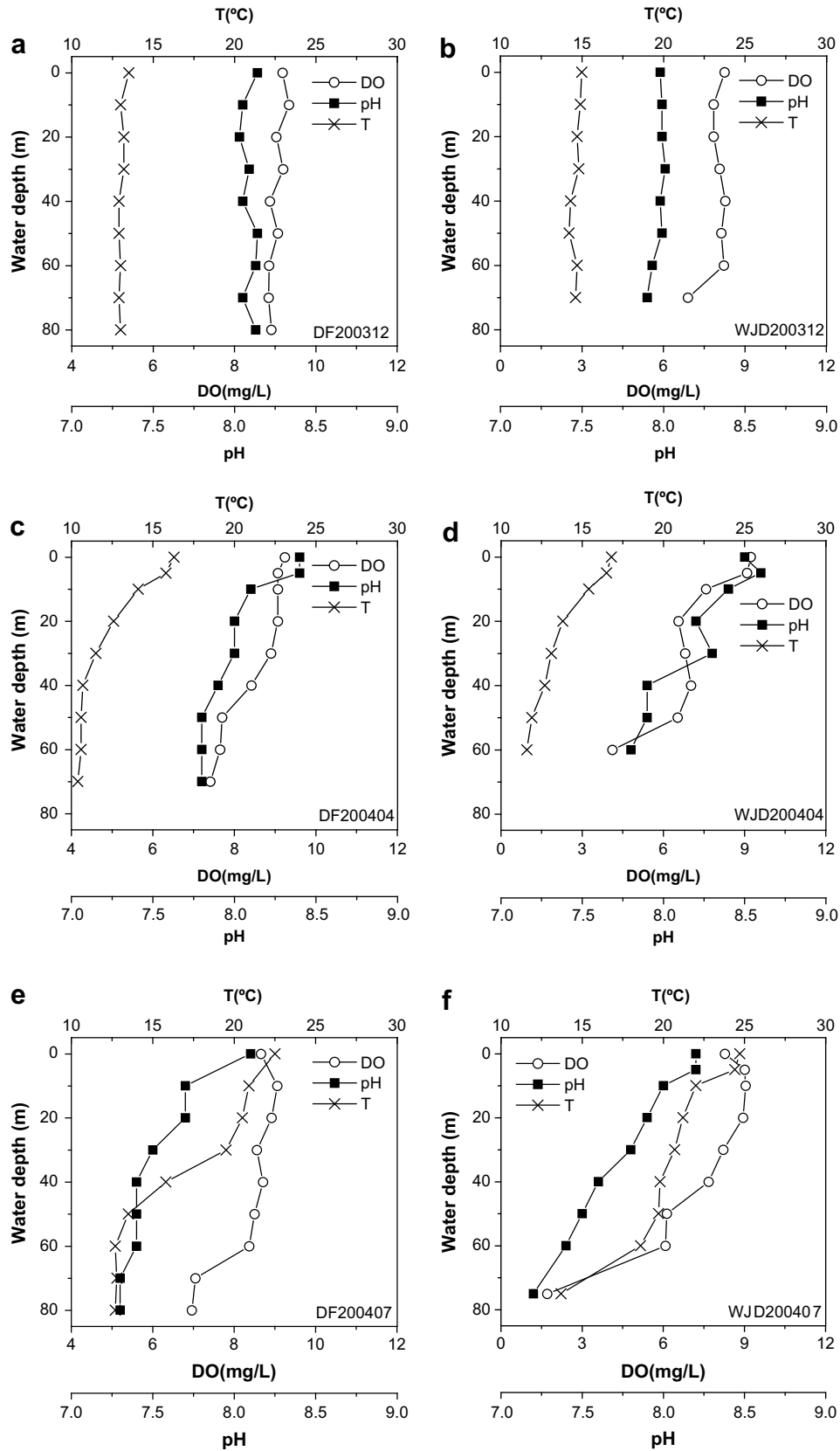


Fig. 2. The water temperature, pH and dissolved oxygen (DO) distribution patterns in water columns of DF and WJD reservoirs in different seasons. a) the sampling campaign conducted in Winter (December 2003) in DF reservoir; b) the sampling campaign conducted in Winter (December 2003) in WJD reservoir; c) the sampling campaign conducted in Spring (April 2004) in DF; d) the sampling campaign conducted in Spring (April 2004) in WJD; e) the sampling campaign conducted in Summer (July 2004) in DF; f) the sampling campaign conducted in Summer (July 2004) in WJD. Please be noted that the scales of DO axes are not always the same.

(2000). The maximum water depths of both reservoirs are almost identical to be about 90 m. In order to compare the Hg biogeochemical processes in two reservoirs, we selected one sampling site about 1 km from the dam in each reservoir as shown in Fig. 1, where the water depths are about 85 m during rainy season. Three sampling campaigns were conducted at these sampling sites in December 2003, April 2004 and July 2004, which representing winter and dry season, spring and dry season and summer and wet season, respectively, to investigate the distribution of different Hg species in water columns and sediment profiles.

## 2.2. Sampling methods and analytical techniques

Water samples at different depths of both reservoirs were collected using acid-cleaned, Teflon lined, 10-L Nisiki sampler on a wooden boat. Both filtered (0.45  $\mu\text{m}$  Millipore membrane filter) and unfiltered water samples for Total Hg (THg) and MeHg analysis were immediately filled in pre-cleaned 100 mL borosilicate glass bottles and acidified upon collection to 0.5% v/v sub-boiling distilled ultra-pure HCl acid within 48 h for storage until subsequent processing or analysis. The water samples were preserved in a refrigerator at 4 °C immediately after being transported to the laboratory. The borosilicate glass bottles were acid-cleaned followed by baking in a Muffle furnace at 450 °C for 1 h. In addition, an aliquot of 300 ml water samples was immediately after collection transferred into an extensively cleaned borosilicate glass impinger which was wrapped with black paper to prevent sunlight, and purged with mercury free argon with a flow rate of 300 ml  $\text{min}^{-1}$  for 30 min and dissolved gaseous mercury (DGM) was collected on a pre-blanked gold trap in the field. Mercury collected on the gold traps was analyzed using dual-stage

amalgamation coupled with AFS detection (Feng et al., 2002). Total and dissolved Hg (filtered with 0.45  $\mu\text{m}$  Millipore membrane filters) in water samples were analyzed within 28 days after sampling using dual stage gold amalgamation method and CVAFS detection according to the method described by Qiu et al. (2006). Total MeHg in unfiltered water and dissolved MeHg (DMeHg) in filtered waters (0.45  $\mu\text{m}$ ) were analyzed using distillation and ethylation processes and GC–CVAFS detection followed US EPA Method 1630 (2001) (He et al., 2008), and particulate MeHg (PMeHg) was the difference between Total MeHg and DMeHg.

Parameters, such as water temperature, pH and dissolved oxygen (DO) were measured using a portable multi-meter (Henna, Italy) immediately after sampling. 1 L water sample was filtered using 0.45  $\mu\text{m}$  (polyvinylidene difluoride) immediately after sampling and the filter was used to measure the chlorophyll-a content using ethanol extraction coupled with spectrophotometry (Jin and Tu, 1990). Coincidentally with Hg sampling, we collected water samples in Nalgene polypropylene bottles for analyses of DOC after filtration using 0.45  $\mu\text{m}$  glass fiber filter. Following appropriate pretreatment and preservation, samples were analyzed using high temperature combustion method with a TOC analyzer (Jiang, 2005).

Mason et al. (1998) compared three commonly used methods to extract pore water from sediments, namely, (i) sediment core sectioning followed by separation of pore water by centrifugation and filtration; (ii) squeezing of the core using gas pressure to extract the pore water; and (iii) use of an in situ dialysis membrane device. They concluded that centrifugation was the most reliable method for determination of Hg and MeHg in estuarine porewaters. Therefore, we used centrifugation method to extract pore water in our study. 30 cm long undisturbed sediment cores were collected using SWB-1 which is a custom designed sediment

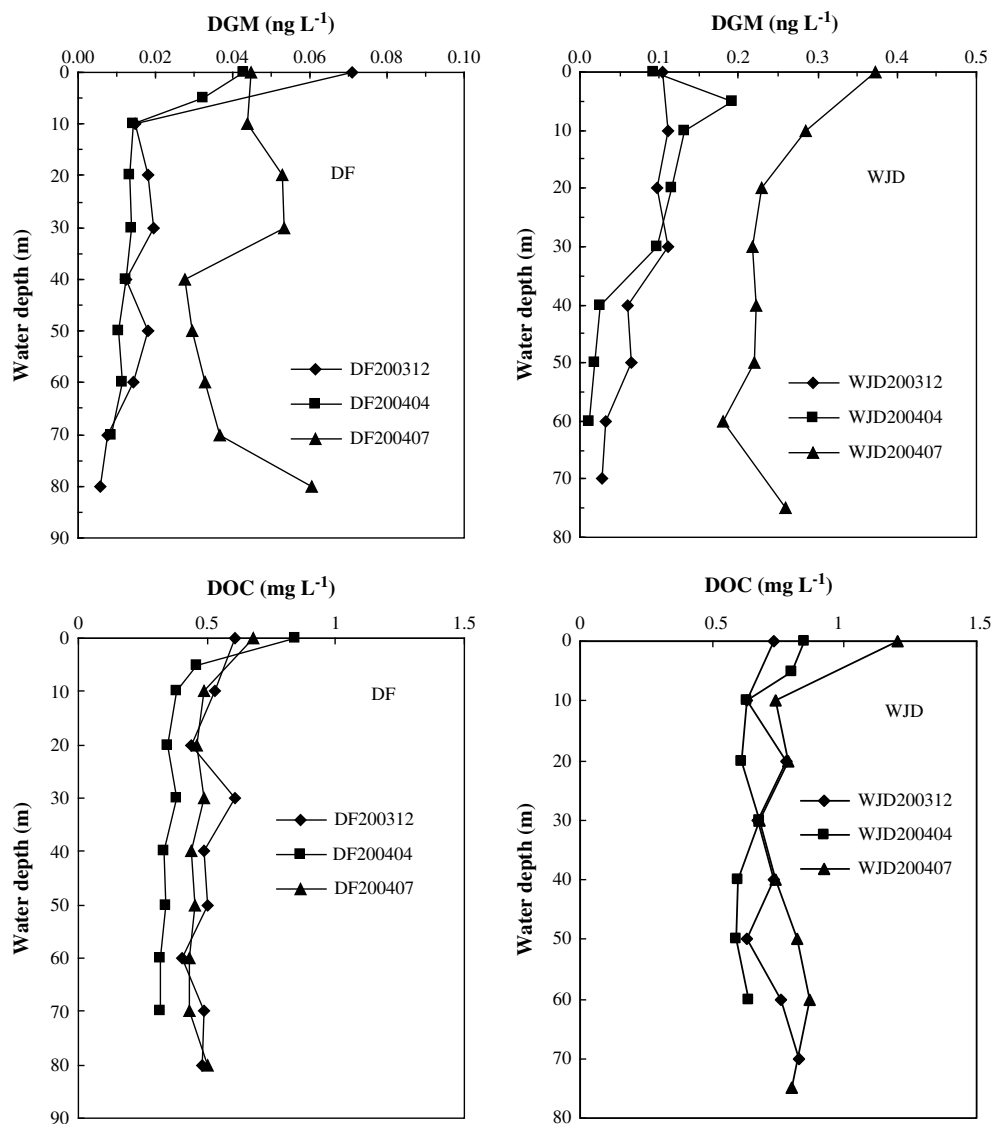


Fig. 3. The distribution of dissolved gaseous mercury (DGM) and dissolved organic carbon (DOC) in water columns of DF and WJD reservoirs in different seasons. Please be noted that the scales of DGM axes are not always the same.

core sampler (Wang et al., 1998). The sediment cores were immediately transferred in a glove box under nitrogen, and sliced into 1 or 2 cm intervals using a plastic cutter and collected in acid-cleaned 50 ml plastic centrifuge tubes. Samples were centrifuged for 30 min to separate the pore water under nitrogen in a gloverbox immediately after being transported to the laboratory. The pore water was then filtered through 0.45  $\mu\text{m}$  disposable polycarbonate filter units, which had been acid-washed prior to use, to remove any remaining particulate and acidified to 0.5% with sub-boiling distilled ultra-pure HCl acid. At each reservoir, at least two sediment cores were collected, and one for THg concentrations in pore water analysis, and one

for MeHg analysis. The pore water samples were immediately acidified to 0.5% v/v sub-boiling distilled ultra-pure HCl acid, and stored in Teflon bottles cold until analysis. The sediment samples after extracting pore water were freeze-dried for solid phase THg and MeHg analysis. Total Hg in pore water was determined using standard techniques (Qiu et al., 2006), including preoxidation by BrCl, reduction by  $\text{NH}_2\text{OH}\cdot\text{HCl}$  and  $\text{SnCl}_2$ , pre-concentration of  $\text{Hg}^0$  onto a Au trap with an aspirator, and analysis by cold vapor atomic fluorescence spectrometry (CVAFS) with a Tekran model 2500 detector. MeHg in pore water was determined after distillation to liberate the MeHg from the matrix (Horvat et al., 1993). The distillates were analyzed

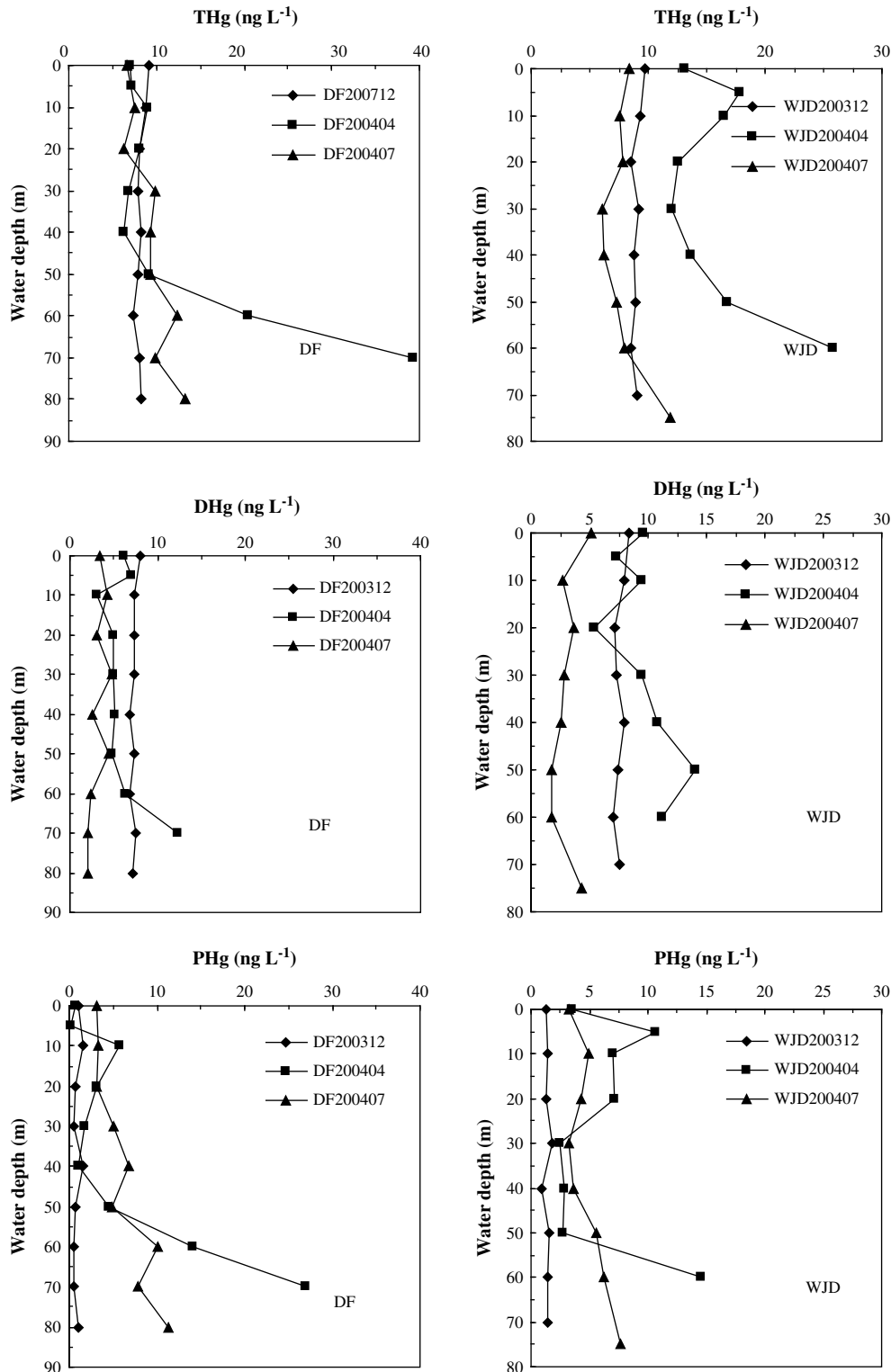


Fig. 4. The distribution of THg, DHg, and PHg in water columns of DF and WJD reservoirs in different seasons. The scales of X- and Y-axes are different for DF and WJD panels.

**Table 2**

THg concentrations and the percentage in DHg and PHg species in water column of WJD reservoir.

Dec-2003				Apr-2004				Jul-2004			
Depth (m)	THg (ng L <sup>-1</sup> )	DHg (%)	PHg (%)	Depth (m)	THg (ng L <sup>-1</sup> )	DHg (%)	PHg (%)	Depth (m)	THg (ng L <sup>-1</sup> )	DHg (%)	PHg (%)
0	9.7	86.6	13.4	0	13.1	73.4	26.6	0	8.3	61.7	38.3
10	9.4	84.9	15.1	5	17.9	40.6	59.4	10	7.6	35.6	64.4
20	8.5	84.6	15.4	10	16.4	57.2	42.8	20	7.9	46.4	53.6
30	9.2	79.5	20.5	20	12.6	43.4	56.6	30	6.1	47.5	52.5
40	8.8	89.9	10.1	30	12.0	78.9	21.1	40	6.2	42.2	57.8
50	9.0	83.1	16.9	40	13.7	79.5	20.5	50	7.3	24.1	75.9
60	8.5	83.3	16.7	50	16.8	84.0	16.0	60	8.0	22.0	78.0
70	9.1	83.8	16.2	60	25.8	43.5	56.5	75	11.9	36.3	63.7
average	9.0	84.5	15.5		16.0	62.6	37.4		7.9	39.5	60.5

using aqueous phase ethylation, trapping on Tenax trap, isothermal GC separation, and CVAFS detection. THg in sediment was determined by acids (1:3 HCl + HNO<sub>3</sub>) digestion followed by CVAFS detection method (Qiu et al., 2006). MeHg in sediment was determined using HNO<sub>3</sub> leaching/CH<sub>2</sub>Cl<sub>2</sub> extraction, ethylation, trapping on Tenax trap, isothermal GC separation, and CVAFS detection method (Liang et al., 2004). The concentrations of organic matter in the sediment samples were analyzed using KCr<sub>2</sub>O<sub>7</sub> oxidation coupled with volumetric technique (Jiang, 2005).

Quality control for Hg and methyl-Hg determinations was addressed with method blanks, blank spikes, matrix spikes, certified reference materials of sediment (GBW07405; CRM580), and blind duplicates. MeHg could be detected at concentrations above 0.01 ng L<sup>-1</sup> at a blank level of 0.045 ng L<sup>-1</sup> in water samples. The detection limit for THg in water samples was 0.2 ng L<sup>-1</sup> at a blank level of 0.3 ng L<sup>-1</sup>. Limits of determination were 0.01 ng g<sup>-1</sup> for total Hg and 0.003 ng g<sup>-1</sup> for methyl-Hg in sediment samples, respectively. The average total Hg concentration of the geological standard of GBW07405 was 0.30 ± 0.01 μg g<sup>-1</sup> (n = 5), which is comparable with certified value of 0.29 ± 0.04 μg g<sup>-1</sup>. Average methyl-Hg concentration of 70.6 ± 0.6 ng g<sup>-1</sup> (n = 7) was obtained from CRM580 with certified value of 70.2 ± 3 ng g<sup>-1</sup>. Recoveries on matrix spikes of MeHg in water samples were in the range of 88.2–108.4%. The relative percentage difference was <8.5% for total Hg in sediment and water samples.

### 2.3. Calculating the diffusive flux of inorganic Hg (IHg) and MeHg from sediment pore water

The diffusion flux of IHg and MeHg from sediment pore water to the water column, in the absence of biological irrigation, is usually calculated based on Fick's first law as described in the following equation (Gill et al., 1999; Hammerschmidt et al., 2004; Holmes and Lean, 2006; Goulet et al., 2007; Rothenberg et al., 2008):

$$F = \frac{\phi D_w}{\theta^2} \frac{\partial C}{\partial x} \quad (1)$$

where F is the diffusive flux of IHg or MeHg at the sediment–water interface,  $\theta$  is the tortuosity (dimensionless),  $\phi$  is the sediment porosity, and  $D_w$  is the diffusion coefficient of IHg or MeHg in water without the presence of the sediment matrix. A relationship between tortuosity and porosity has recently been proposed (Boudreau, 1996) and will be used for all flux calculations made herein:

$$\theta^2 = 1 - \ln(\phi^2) \quad (2)$$

The diffusion coefficients of IHg and MeHg in water were estimated to be  $9.5 \times 10^{-6}$  and  $1.3 \times 10^{-5}$  cm<sup>2</sup> s<sup>-1</sup> at 25 °C, respectively (Gill et al., 1999; Covelli et al.,

1999). Temperature corrections to the diffusion coefficients at 25 °C were made when necessary using the relationship (Lerman, 1979)

$$D_{T1} = D_{T2}(1 + 0.048\Delta t) \quad (3)$$

where  $\Delta t$  is the temperature difference in degrees Centigrade.

The porosity is computed using the following equation

$$\phi = 1 - [G/(Vd)] \quad (4)$$

where G is dry weigh of the sediment (g), V is volume of the fresh sediment (cm<sup>3</sup>), and d is the density of the dry sediment (g cm<sup>-3</sup>).

The concentration gradient was calculated from the IHg (THg–MeHg) or MeHg concentrations in the bottom waters and porewaters collected from the first sediment interval.

## 3. Results and discussion

### 3.1. The distribution of Hg species in water column

The water temperature, pH and DO distribution patterns in water columns at both reservoirs in December 2003, April 2004, and July 2004 sampling campaigns are illustrated in Fig. 2. Both reservoirs are alkaline because the bedrocks in the watersheds of both reservoirs are limestone and dolomite. The water columns in both reservoirs were well mixed in December, but stratification occurred in April and July campaigns (Fig. 2). Generally, the maximum DGM concentrations occurred in surface water (0–5 cm), and DGM concentrations decreased gradually with the depth (Fig. 3). Photo-induced reduction of divalent Hg is believed to be the major contribution of DGM formation in surface water (Poullain et al., 2004; Amyot et al., 1997; O'Driscoll et al., 2004; Feng et al., 2004, 2008). The increase of DGM concentrations in the bottom of the water column in both reservoirs during July campaign may indicate that DGM could also be produced during demethylation processes in the sediment. In general, DGM concentrations decreased in the order of July > April > December for WJD reservoir, and in the order of July > December > April for DF reservoir.

**Table 3**

THg concentrations and the percentage in DHg and PHg species in water column of DF reservoir.

Dec-2003				Apr-2004				Jul-2004			
Depth (m)	THg (ng L <sup>-1</sup> )	DHg (%)	PHg (%)	Depth (m)	THg (ng L <sup>-1</sup> )	DHg (%)	PHg (%)	Depth (m)	THg (ng L <sup>-1</sup> )	DHg (%)	PHg (%)
0	9.1	87.8	12.2	0	7.0	88.8	11.2	0	6.7	52.4	47.6
10	8.9	82.2	17.8	5	7.2	96.6	3.4	10	7.5	56.5	43.5
20	8.1	91.8	8.2	10	8.9	35.2	64.8	20	6.3	49.9	50.1
30	7.9	92.3	7.7	20	8.0	61.6	38.4	30	9.8	48.7	51.3
40	8.3	82.1	17.9	30	6.8	74.1	25.9	40	9.3	27.5	72.5
50	7.9	91.9	8.1	40	6.2	82.6	17.4	50	9.3	48.1	51.9
60	7.3	93.7	6.3	50	9.2	51.3	48.7	60	12.4	19.3	80.7
70	8.0	92.6	7.4	60	20.4	30.7	69.3	70	9.9	20.8	79.2
80	8.2	87.1	12.9	70	39.3	31.2	68.8	80	13.3	15.9	84.1
average	8.2	89.1	10.9		12.5	61.3	38.7		9.4	37.7	62.3

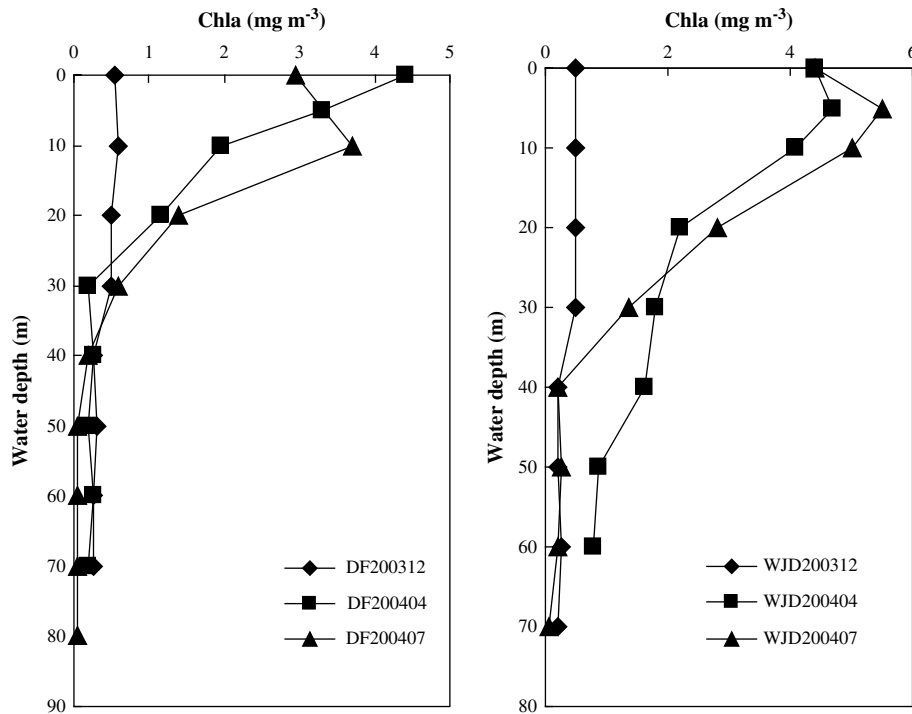


Fig. 5. The distribution of chlorophyll-a in water columns of DF and WJD reservoirs in different seasons.

Apart from photo-induced reduction, DGM could also be formed by biological and chemical processes (Poulain et al., 2004). Moreover, the DGM formation by photo-induced reduction may also be mediated by the structure and concentrations of DOC in water (O'Driscoll et al., 2004). DOC concentrations in water of WJD reservoir were generally higher than those of DF reservoir (Fig. 3). Meanwhile, Wang (2003) and Zhu (2005) demonstrated that the origins of DOC in both reservoirs were different, and the major source of DOC in DF reservoir was allochthonous, while the main contribution of DOC in WJD reservoir was autochthonous. The structures of DOC of different origins were unfortunately not studied, but they may differ significantly. Therefore, we observed that DGM concentrations in WJD were much higher than those in DF (Fig. 3).

Fig. 4 shows the distributions of THg, dissolved Hg (DHg) and particulate Hg (PHg) concentrations in water columns of both reservoirs during 3 sampling campaigns. It is clearly seen that THg, DHg and PHg were almost evenly distributed in water columns of both reservoirs in winter campaign (December 2003) when the water columns were well mixed according to the observation data of water temperature, pH and DO (Fig. 2). Meanwhile DHg was the dominant Hg species in water columns of both reservoirs in winter campaign (Tables 2 and 3). It is the dry season in winter in the watershed area of Wujiang River, and water inflows to both reservoirs are the lowest (Feng et al., 2008), resulting in the low load of allochthonous particulate matters to both reservoirs. Due to the low water temperatures, the biological activities were not active in winter, which was indicated by the chlorophyll-a distribution

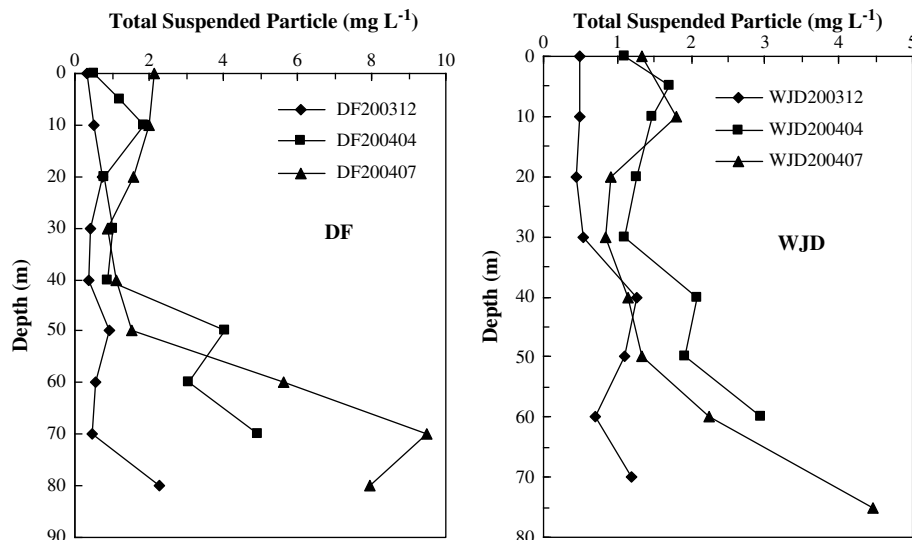


Fig. 6. The distribution of total suspended particles in water columns of DF and WJD reservoirs in different seasons.

patterns in water (Fig. 5). This may also engender the low production rate of autochthonous particulate matters in reservoirs. We can obviously see that the total suspended particulate (TSP) concentrations were the lowest among the three campaigns in both reservoirs (Fig. 6).

The distributions of THg, DHg and PHg in water columns in both reservoirs were not uniform in April and July sampling campaigns when the stratification occurred (Fig. 4). In general, THg concentrations in water column of WJD reservoir were higher than those

in DF reservoir except in July. In April, it was still in dry season, and the allochthonous particulate loading from the inlet rivers of both reservoirs were relatively low. The surface water temperatures, however, arose, triggering the blooming of biological activities, especially in surface water as shown in Fig. 5. As a result, TSP concentrations in water columns of both reservoirs increased, and we also saw that the re-suspension of sediments could occur in April at both reservoirs because the TSP concentrations in bottom water increased significantly (Fig. 6). The THg peak at water depths

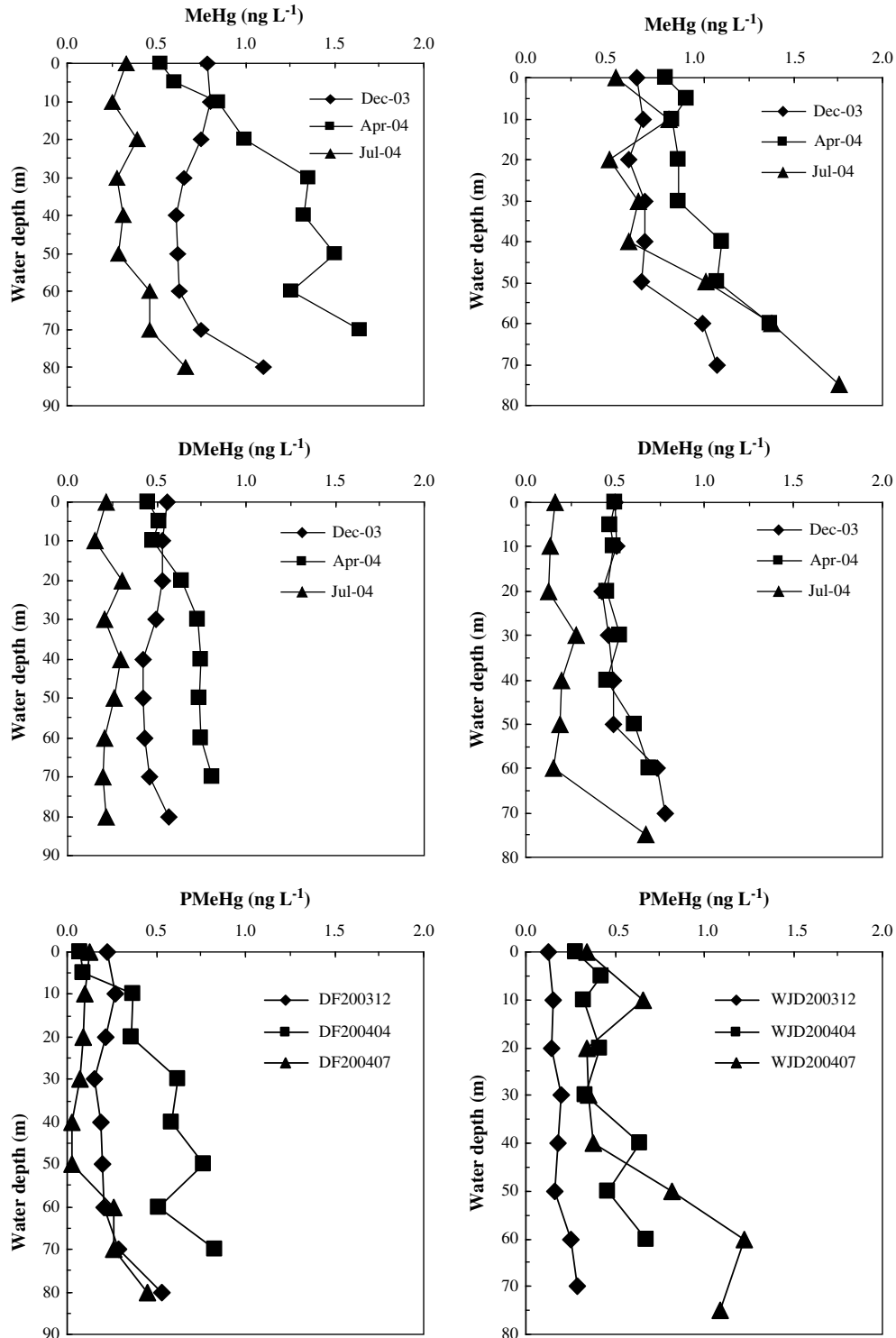


Fig. 7. The distribution of MeHg, DMeHg, and PMeHg in water columns of DF and WJD reservoirs in different seasons.



**Table 4**  
MeHg concentrations and the percentage in DMeHg and PMeHg species in water column of WJD reservoir.

Dec-2003				Apr-2004				Jul-2004			
Depth (m)	MeHg (ng L <sup>-1</sup> )	DMeHg (%)	PMeHg (%)	Depth (m)	MeHg (ng L <sup>-1</sup> )	DMeHg (%)	PMeHg (%)	Depth (m)	MeHg (ng L <sup>-1</sup> )	DMeHg (%)	PMeHg (%)
0	0.62	80.4	19.6	0	0.78	64.6	35.4	0	0.50	32.0	68.0
10	0.66	77.2	22.8	5	0.90	52.5	47.5	10	0.80	17.5	82.5
20	0.58	74.7	25.3	10	0.82	60.4	39.6	20	0.47	26.4	73.6
30	0.66	69.4	30.6	20	0.86	52.3	47.7	30	0.63	44.7	55.3
40	0.67	73.7	26.3	30	0.86	61.7	38.3	40	0.57	34.5	65.5
50	0.65	74.8	25.2	40	1.10	41.7	58.3	50	1.01	18.7	81.3
60	0.99	74.9	25.1	50	1.07	57.2	42.8	60	1.38	11.3	88.7
70	1.07	73.0	27.0	60	1.37	50.4	49.6	75	1.76	38.1	61.9
average	0.74	74.8	25.2		0.97	55.1	44.9		0.89	27.9	72.1

from 5 to 10 m in WJD reservoir in April (a small peak also observed in DF reservoir) (Fig. 4e, f) may be resulted from the blooming of phytoplankton or/and zooplankton, induced by the elevated PHg concentrations in water because Chla contents in water column peaked at this water depth as shown in Fig. 5. On the basis of that fact that both THg and PHg were elevated in the bottom water of both reservoirs, we suggest that these peaks in Hg species were the result of re-suspension of surface sediments. An early study (Zhu, 2005) showed that the primary productivity in WJD reservoir were much higher than that in Dongfeng reservoir. This explained the much elevated THg concentrations in water column in WJD reservoir in April.

During the July campaign, THg concentrations in bottom water increased in both reservoirs, which may also result in the re-suspension of surface sediments (Fig. 4). It is interesting to note that the percentages of THg presented as in the form of PHg increased in the order of December < April < July for both reservoirs (Tables 2 and 3), which demonstrated that the transformation of Hg species occurred with the changes of seasons. From December, April to July, the water temperatures constantly increase and biological activities (algae blooms) which can produce organic particulate matters will also increase as shown from the Chla contents in water column (Fig. 5). Organic particulate matters could absorb more ionic Hg than inorganic particulates and this may explain the seasonal pattern of the percentages of THg presented as in the form of PHg in water. In WJD reservoir, THg concentrations in water columns increased in the order of July < December < April. However, there were no large and significant seasonal variations of the distribution of THg concentrations in water column in DF reservoir.

The distributions of MeHg concentrations in water columns in both reservoirs are shown in Fig. 7. In general, average MeHg concentrations in water column increased in the order of July < December < April for DF reservoir, and December < July < April for WJD reservoir, respectively. We observed that PMeHg concentrations in water columns determined the distribution pattern of MeHg in water columns, since DMeHg concentrations did not vary tremendously in the water columns in all sampling campaigns for both reservoirs (Fig. 7). In April, the phytoplankton started to bloom, resulting in high PMeHg concentrations in water columns in both reservoirs since phytoplankton can absorb MeHg from water (Hurley et al., 1994). We observed that both DMeHg and PMeHg concentrations in the bottom of water columns were elevated in all sampling campaigns in both reservoirs indicating that sediments were MeHg source to the water. Both re-suspension of sediment and diffusion of MeHg from sediment pore water can bring MeHg to water column.

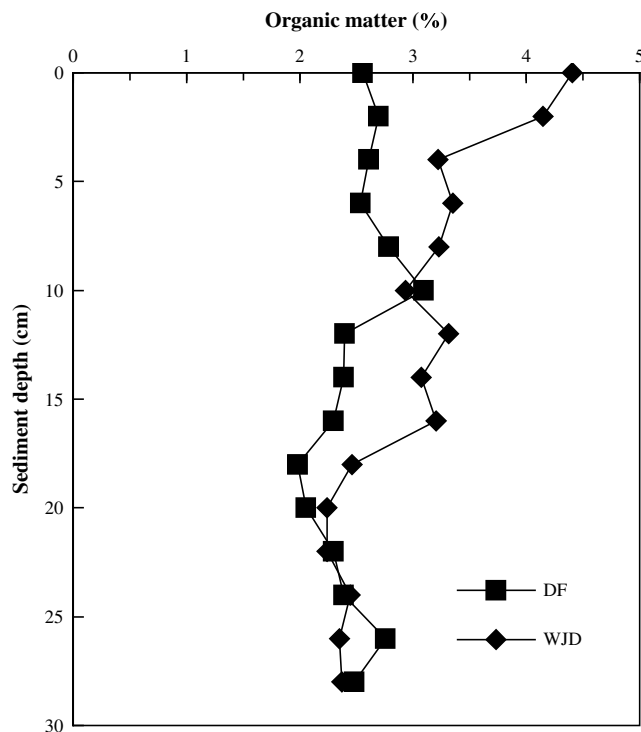
The average percentages of MeHg presented as in the form of PMeHg in water column of WJD reservoirs increased in the order of December < April < July (Table 4). However, the percentage of MeHg presented as in the form of PMeHg in water column of DF

reservoir did not change significantly among different sampling times (Table 5). The mechanism behind the difference in speciation of MeHg in water between two reservoirs is not clear yet. This may be related to the difference of primary productivity between both reservoirs. Much study is needed to elucidate the discrepancy.

### 3.2. Distribution of Hg species in sediment profiles

The organic contents in sediment cores collected in July campaign were analyzed and the organic matter contents in the top of the sediment in WJD reservoir were much higher than those in DF reservoir (Fig. 8). This supported the fact that the primary productivity in DF reservoir was less than WJD reservoir (Zhu, 2005) because the organic matters with autochthonous are produced in the reservoir while those with allochthonous origin are from the input from the catchments.

Fig. 9A and B showed the distribution of THg concentrations in sediment profiles of WJD and DF reservoir during three sampling campaigns conducted in December, April and July, respectively.



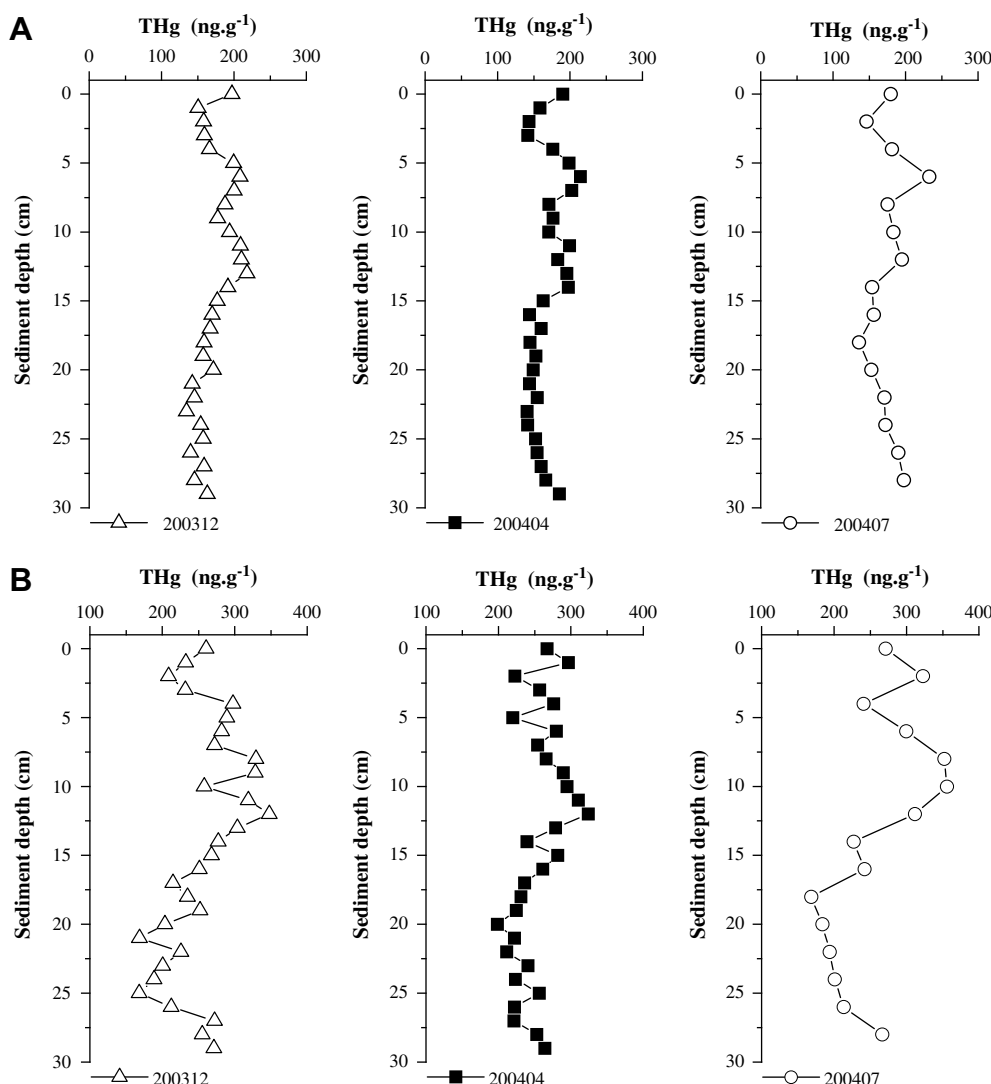
**Fig. 8.** The distribution of organic matter in sediment profiles of DF and WJD reservoirs.

**Table 5**  
MeHg concentrations and the percentage in DMeHg and PMeHg species in water column of DF reservoir.

Dec-2003				Apr-2004				Jul-2004			
Depth (m)	MeHg (ng L <sup>-1</sup> )	DMeHg (%)	PMeHg (%)	Depth (m)	MeHg (ng L <sup>-1</sup> )	DMeHg (%)	PMeHg (%)	Depth (m)	MeHg (ng L <sup>-1</sup> )	DMeHg (%)	PMeHg (%)
0	0.78	71.7	28.3	0	0.52	85.9	14.1	0	0.34	63.3	36.7
10	0.80	66.7	33.3	5	0.60	85.4	14.6	10	0.25	61.8	38.2
20	0.75	70.7	29.3	10	0.84	56.4	43.6	20	0.39	77.5	22.5
30	0.65	75.9	24.1	20	0.99	63.9	36.1	30	0.28	72.7	27.3
40	0.62	68.8	31.2	30	1.36	54.0	46.0	40	0.32	92.4	7.6
50	0.62	68.3	31.7	40	1.33	56.1	43.9	50	0.29	90.3	9.7
60	0.63	67.9	32.1	50	1.50	49.3	50.7	60	0.47	44.0	56.0
70	0.75	60.9	39.1	60	1.26	59.3	40.7	70	0.46	43.5	56.5
80	1.10	51.7	48.3	70	1.64	49.3	50.7	80	0.66	32.5	67.5
average	0.74	67.0	33.0		1.12	62.2	37.8		0.38	64.2	35.8

Sediment cores from DF reservoir included sediment and soil. No significant variations of THg distribution in sediment profiles were observed for both reservoirs. Moreover, no significant seasonal variations of THg distributions in sediment profiles were displayed for both reservoirs. The average THg concentrations in sediment profiles of WJD reservoir were 254.2 ng/g, 254.2 ng/g, and 256.7 ng/g in December, April and July campaigns, respectively. The average THg concentrations in sediment profiles of DF reservoir were

172.4 ng/g, 167.8 ng/g, and 167.8 ng/g in December, April and July campaigns, respectively. However, totally different distribution patterns in sediment profiles were observed for MeHg (Fig. 10). Generally, MeHg concentrations were enriched in the uppermost part of the sediment profiles, and decreased with depth. The peak MeHg concentrations in sediment profile of WJD reservoir in all three sampling campaigns and in sediment profile of DF reservoir in July campaign occurred at the first 1–2 cm of sediment, while the



**Fig. 9.** The distribution of THg in sediment profiles of DF and WJD reservoirs in different seasons. A) DF reservoir; B) WJD reservoir.

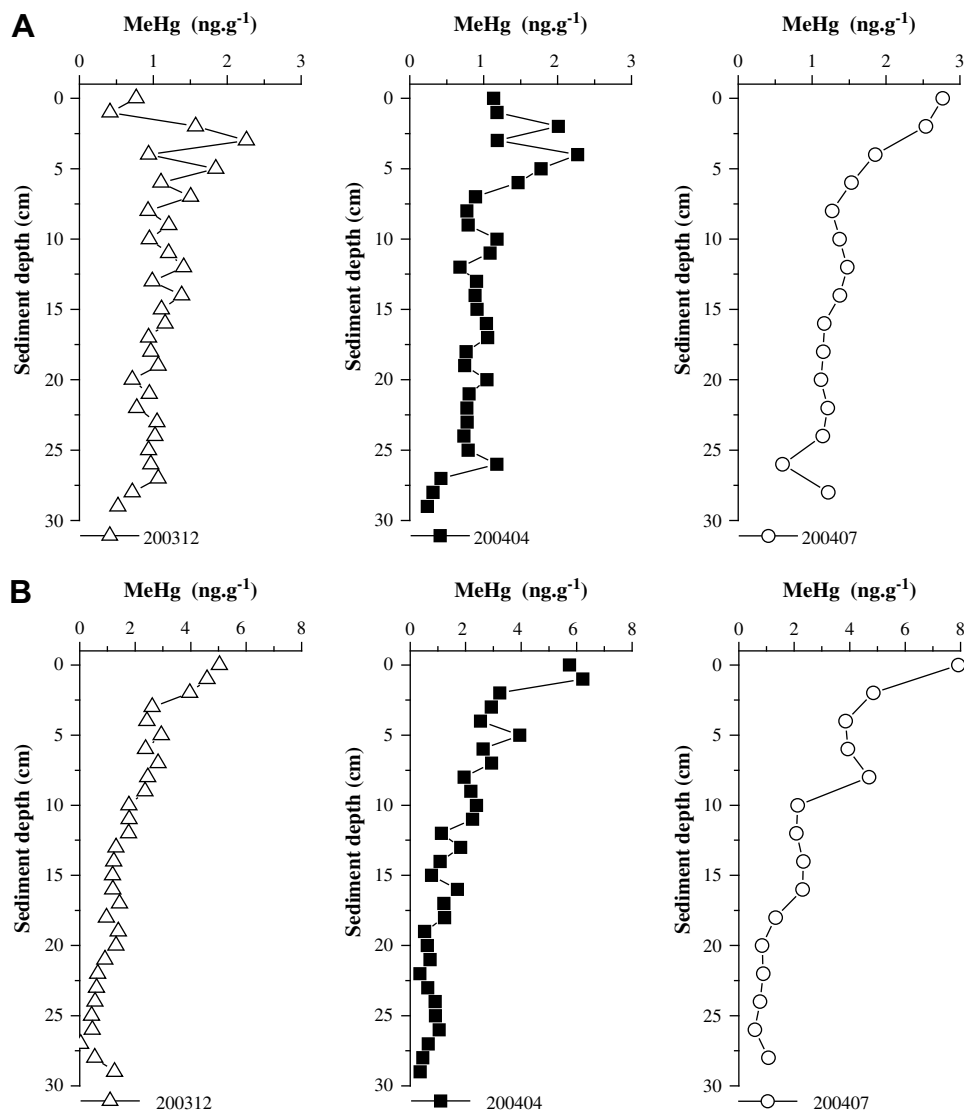


Fig. 10. The distribution of MeHg in sediment profiles of DF and WJD reservoirs in different seasons. A) DF reservoir; B) WJD reservoir.

maximum concentrations appeared at the 4–5 cm of the sediment profile of DF reservoir in December and April campaigns. This demonstrated that much faster methylation processes than the demethylation processes mainly occurred at the sediment surface in WJD reservoir, while much faster methylation processes than the demethylation processes occurred at the depth of 4–5 cm of the sediment profiles in DF reservoir. A discussion of the reasons for the discrepancy will be given in the following section. For both reservoirs, MeHg concentrations in sediment profiles were the highest in July campaign and the lowest in the December campaign. This demonstrated that much higher mercury methylation rates occurred in sediment in warm seasons than cold seasons, which supported the conclusion that high temperatures favor mercury methylation process in sediment (Ullrich et al., 2001). It is interesting to note that the maximum MeHg concentrations in sediment profiles in WJD reservoir at each sampling campaigns were all much higher than the values at the corresponding sampling campaign in DF reservoir (Fig. 10). This illustrated that the MeHg production rates in WJD reservoir were much higher than DF reservoir, which can explain that the MeHg yield in WJD reservoir ( $140.9 \text{ g MeHg km}^{-2} \text{ yr}^{-1}$ ) was much higher than that of DF reservoir ( $32.9 \text{ g MeHg km}^{-2} \text{ yr}^{-1}$ ) (Feng et al., 2009).

### 3.3. Distribution of Hg species in sediment pore water and diffusion flux of Hg species to water column

The distribution patterns of pore water MeHg in both reservoirs were completely different from those of pore water inorganic Hg (IHg) (THg–MeHg) as shown in Figs. 11 and 12. The pore water MeHg profiles of both reservoirs at all sampling campaigns showed elevated concentrations close to or at the sediment surface and generally decreased at greater depth (Fig. 11). We point out that our data cannot address the potential demethylation processes occurring in these sediments. Therefore it is not possible to discount the fact that Hg methylation processes are occurring throughout the sediment profile and that demethylation processes producing inorganic Hg are simply more active at depth and due to changes in sediment environment are less active closer to the surface sediment and therefore producing the observed pattern. It is interesting to note that the peak MeHg in pore water in WJD reservoir occurred at the sediment and water interface, but at about 2 cm in the sediment in DF reservoir. The pore water MeHg distribution patterns were quite similar with those of sediment MeHg. It is obvious that MeHg concentrations in pore water, especially in pore water at the surface sediment were much higher than those of

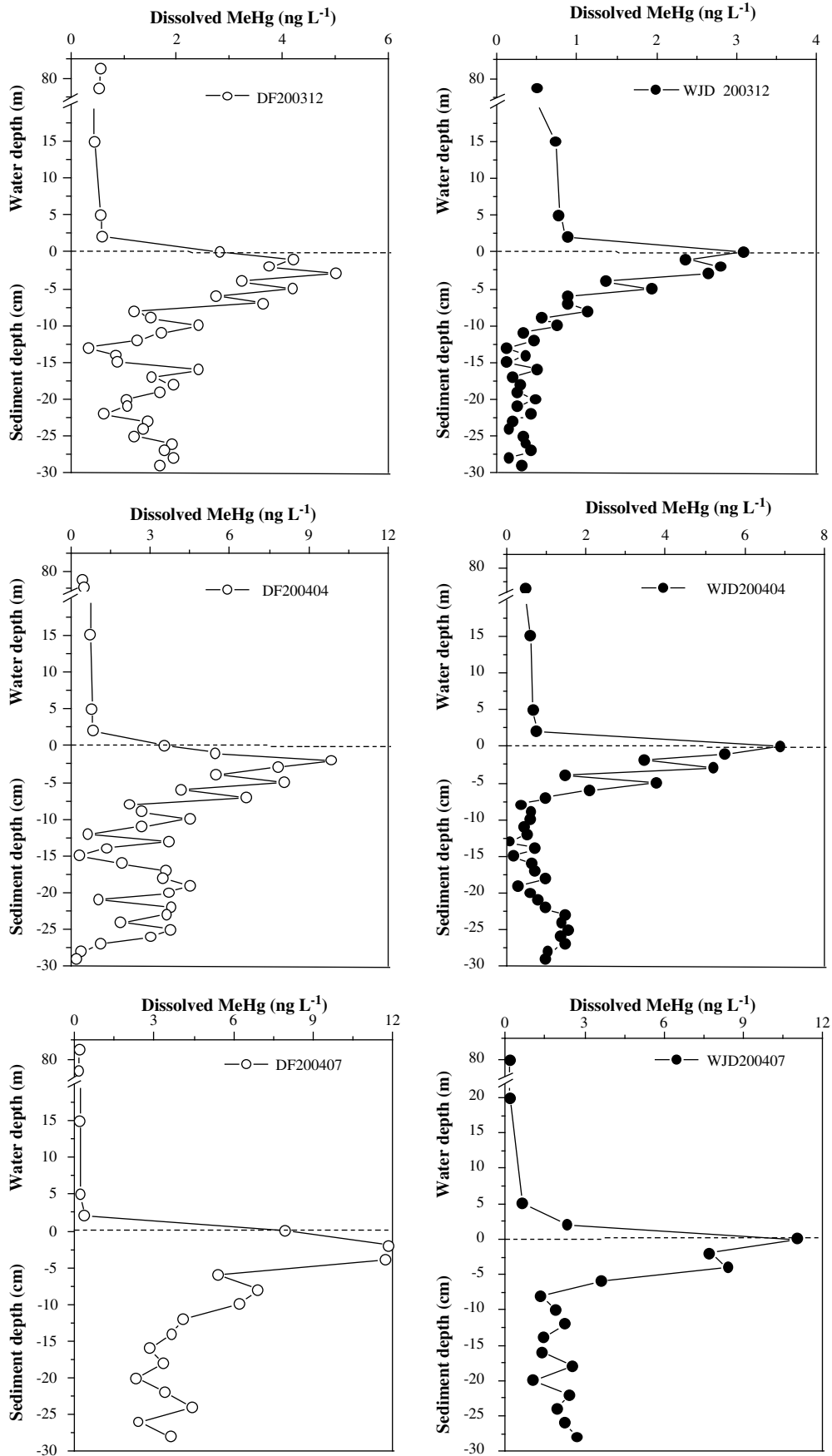


Fig. 11. The distribution of dissolved MeHg in water column and sediment pore water of DF and WJD reservoirs in different seasons.

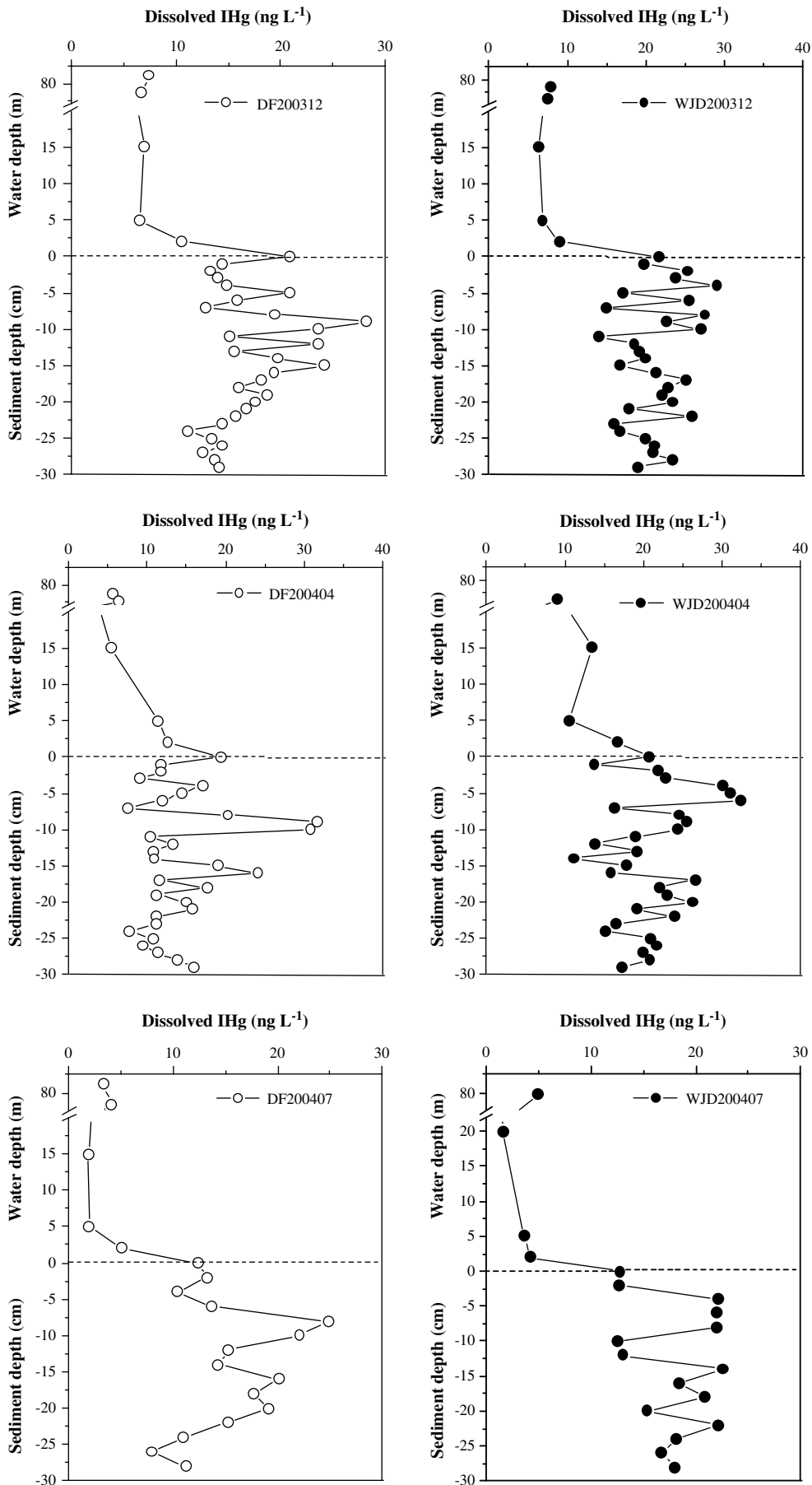


Fig. 12. The distribution of dissolved inorganic mercury (IHg) in water column and sediment pore water of DF and WJD reservoirs in different seasons.

**Table 6**

Estimated diffusion fluxes of inorganic Hg and MeHg of Wujiangdu (WJD) and Dongfeng (DF) Reservoirs in different seasons (in  $\text{ng m}^{-2} \text{day}^{-1}$ ).

		December	April	July
IHg	WJD	64.9	49.1	44.3
	DF	62.5	60.3	41.4
MeHg	WJD	15.6	39.4	65.6
	DF	14.4	17.8	48.0

water column at all sampling seasons. This implies that surface sediment is a strong MeHg source in the reservoirs (Hammerschmidt et al., 2004; Holmes and Lean, 2006; Rothenberg et al., 2008). However, the distribution trend of pore water DIHg is not clear, though DIHg concentrations in pore water were generally higher than those in the column (Fig. 12). Sediment is also a source of IHg to water column.

The diffusion fluxes of both MeHg and IHg from sediment to water column in different seasons for both reservoirs were calculated and listed in Table 6. IHg is defined as the difference between THg and MeHg. We can clearly see that sediment in both reservoirs were net sources of both MeHg and IHg in the reservoirs. The IHg diffusion fluxes decreased in the order: December > April > July for both reservoirs. In contrast, however, MeHg diffusion fluxes displayed a completely different temporal pattern. The maximum MeHg fluxes occurred in July and the minimum fluxes occurred in December for both reservoirs. This demonstrated that high temperatures in summer favor net mercury methylation (Ullrich et al., 2001). The summer maximum MeHg flux in WJD reservoir explained the maximum yield of MeHg to downstream of WJD reservoir (Feng et al., 2009). Even though the maximum MeHg flux occurred in summer, but the maximum yield of MeHg in DF reservoir did not occur in summer (Feng et al., 2009). This is simply because in summer DF reservoir is a sink for water for flood control and the total water outflow was less than the total inflow (Feng et al., 2009).

It is clearly that the MeHg diffusion fluxes in WJD reservoir at all sampling campaigns were significantly higher than those in DF reservoir. This explained the fact we observed in the mass balance study (Feng et al., 2009) that the MeHg yield from WJD is much higher than that of DF reservoir. However, in order to quantify the overall MeHg diffusion fluxes from the sediment to water column, more sampling sites from upstream to downstream of both reservoirs are needed. It is obvious that more study is needed to quantify the total MeHg diffusion fluxes from sediments to water column from both reservoirs.

The elevated organic matter contents in sediment of WJD reservoir compared to DF reservoir as shown in Fig. 8 explain the higher MeHg diffusion fluxes observed in Table 6 (Lucotte et al., 1999). It is generally believed that high levels of organic matter promote reducing conditions (Callister and Winfrey, 1986; Regnell et al., 1996), which favor sulphate reduction that in turn promotes mercury methylation which is predominately linked to the activities of sulphate-reducing bacteria (Compeau and Bartha, 1985; King et al., 2000). The high levels of organic matter contents in sediment of WJD reservoir may have resulted in the favorable conditions for mercury methylation at the surface sediment so that the maximum MeHg concentrations in pore water of the sediment occurred at the surface sediment. However, the low level of organic matter contents in DF reservoir may have induced the favorable redox conditions for mercury methylation occurred at 3–4 cm in depth of the sediment, which resulted in the lower MeHg diffusion flux from the sediment because only the MeHg in pore water of the surface sediment (1 cm in depth) can easily diffuse to the water column provided that a gradient of MeHg concentrations between pore

water of the sediment and the water column existed (i.e. Holmes and Lean, 2006). However, we are measuring net MeHg production and that the trends we are seeing may be related trends in both methylation and demethylation processes.

Studies in North America, have shown that the age of the reservoirs is a key factor to govern mercury methylation rates, and with increase of the age of the reservoirs, the organic contents in flooded soil decreased with the decomposition processes and mercury methylation rates will decrease (i.e. St. Louis et al., 2004; Schetagne et al., 2000). However, our study showed that the old reservoir (WJD) produced much more methylmercury than the young reservoir (DF). We suggest that this unexpected trend results from the fact that soils flooded by the formation of the reservoir in the Wujiang River were initially characterized by low levels of organic matter (less than 2%) (Jiang, 2005) and that as the reservoirs aged they accumulated allochthonous and autochthonous organic matter that ultimately increased the level of organic matter in the sediment and stimulated net mercury methylation rates in surface sediments. The primary productivity in a given reservoir can increase organic matter contents in sediments (Wang, 2003), and the relatively small increase in organic matter from 2% to 4% would actually significantly change the conditions promoting net MeHg production to produce the large difference between the WJD and DF reservoirs.

#### 4. Conclusions

In this paper, we studied the biogeochemical processes of Hg in Wujiangdu and Dongfeng reservoirs to better understand the controlling factors of methylmercury production in reservoirs created in Wujiang catchments. Taken as a whole, our data indicate that:

- The sediment is the net source of both inorganic and MeHg to the water column for both reservoirs,
- The MeHg diffusion fluxes in WJD reservoir at all sampling campaigns were significantly higher than those in DF reservoir, and the elevated organic matter contents in sediment of WJD reservoir compared to DF reservoir may explain the higher MeHg diffusion fluxes,
- The high primary productivity in the reservoir resulted in high organic matter contents in the sediment may favor the net methylmercury production in sediment.

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