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Effects of damming on the distribution and methylation of mercury in Wujiang River, Southwest China

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HIGHLIGHTS

- Total Hg concentrations decreased from upstream to downstream of the Wujiangriver as the result of the intercepted particulate Hg.
- Net Hg methylation and the output of MeHgfrom reservoirs increased with progressive increase of trophic stage in reservoir.
- Net Hg methylation and discharge of MeHgfrom hypolimnion resulted in the elevation of MeHgin several sections of the river.

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ABSTRACT

Newly built reservoirs are regarded as sensitive ecosystem for mercury (Hg) methylation. A comprehensive study was conducted to understand the influence of damming on the distribution and methylation of Hg within a river-reservoir ecosystem in Wujiang River Basin (WRB), Southwest China. Hg species in inflow-outflow rivers of six cascade reservoirs were analyzed each month during 2006. Mean concentrations of total Hg (THg) and methylmercury (MeHg) in river water in WRB were 3.41 ± 1.98 ng L^{-1} and 0.15 ± 0.06 ng L^{-1}, respectively. THg and particulate Hg (PHg) concentrations in outflow rivers of reservoirs significantly decreased after dam construction, suggesting that a considerable amount of PHg was intercepted by way of sedimentation. However, the influence of damming on the distributions of dissolved Hg (DHg) and reactive Hg (RHg) in rivers was less pronounced. MeHg concentrations in outflow rivers of the older reservoirs significantly increased compared to inflow rivers with the maximum increasing factor of 92%, indicating the active net Hg methylation in the reservoirs. However, the difference between MeHg in inflow rivers and outflow rivers were less pronounced in the newly constructed reservoirs, indicating that these reservoirs were not active sites of Hg methylation. The construction of the cascade reservoirs resulted in the elevation of MeHg in several sections of the Wujiang River, which attributed to the net Hg methylation in reservoirs and discharge of MeHg from hypolimnion. MeHg-enriched water in outflow rivers from hypolimnetic water could be transported to downstream, posing potential threat to the aquatic food web and human health.

1. Introduction

Mercury (Hg) as an extremely persistent, bioaccumulative, and toxic pollutant, has received considerable attention in the past decades (Lindqvist et al., 1991; Stein et al., 1996). Inorganic Hg (IHg) can undergo chemical and microbial transformations to methylmercury (MeHg), which can be accumulated by aquatic biota and biomagnified along the food chains (Watras et al., 1995), posing a potential threat to wildlife and human health (Hammerschmidt et al., 2006). Consequently, fish consumption is regarded as the main route of MeHg exposure to humans (Clarkson, 1993).

It is generally accepted that reservoirs are sensitive ecosystems for Hg methylation and subsequent contamination of MeHg to food web of aquatic ecosystem (Lucotte et al., 1999; St. Louis et al., 2004). The risk of elevated MeHg concentrations in fish is one of the most important concerns in newly constructed reservoirs. With the rapid development of dams in river systems over the world, the scientific communities are paying more and more attention to the impacts of dam construction on transportation and transformation of Hg in

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river-reservoir systems (Kelly et al., 1997; Heyes et al., 2000).

Elevated levels of MeHg in fish have been widely reported in recently impounded hydroelectric reservoirs in North American and North Europe, where fish consumption advisories were commonly issued by local health departments (Lucotte et al., 1999; Abernathy and Cumbie, 1977; Bodaly et al., 1984). In China, Hg concentrations in fish from reservoirs along the Wujiang River and the Yangze River were also significantly elevated (Jin and Xu, 1997; Li et al., 2013). Enhanced MeHg production in water from newly built reservoirs following impoundment may persist for up to 10 years (St. Louis et al., 2004; Kelly et al., 1997). Hg methylation is largely facilitated by a subset of sulfate-reducing bacteria (SRB), iron-reducing bacteria (IRB), methanogens, and firmicutes in natural environment (Gilmour and Henry, 1991; Gilmour et al., 2013; Schaefer et al., 2014). Recently, Bravo et al. (2014) explored that both SRB and IRB are potential methylators for MeHg production in reservoirs. Furthermore, the decomposition of flooded vegetation and organic matter in soils may stimulate the Hg methylation (Lucotte et al., 1999). The net Hg methylation rate decreases with the increase of the reservoir age as the result of the continual decomposition of organic matter in submerged soil (Lucotte et al., 1999; St. Louis et al., 2004). Active Hg methylation were also observed in old reservoirs in North America and Europe (e.g. Falls Creek Reservoir in Idaho, USA and Babeni Reservoir in Romanian) (Gray and Hines, 2009; Bravo et al., 2014).

The total amount of large dams in China accounted for more than 50% of the total globally since 1982, and many more new dams are being created rapidly (The World Commission on Dams, 2000). The Wujiang River is the largest tributary of the upper Yangtze River. Since 1970s, numerous large cascade reservoirs have been or are being constructed along Wujiang River Basin (WRB), Southwest China (Fig. 1), such as Wujiangdu Reservoir (WJD, impoundment in 1979), Dongfeng Reservoir (DF, 1994), Puding Reservoir (PD, 1994), Yingzidu Reservoir (YZD, 2003), Suofengying Reservoir (SFY, 2003), Hongjiadu Reservoir (HJD, 2004). In addition, a number of reservoirs were also built in the tributaries of Wujiang River, such as Baihua Reservoir (BH, 1966), Hongfeng Reservoir (HF, 1966), and Aha Reservoir (AH, 1960).

Numerous studies were conducted to investigate the biogeochemical cycling of Hg in river-reservoir ecosystem in WRB, Southwest China. Early studies have confirmed that the newly constructed reservoirs, such as the YZD, SFY, and HJD, within WRB were not active sites for Hg methylation given the low organic matter content in submerged soil (Meng et al., 2010; Yao et al., 2011). However, the older reservoirs within WRB, such as the WJD, PD, and DF, were characterized by a much more active net Hg methylation compared to the newly constructed reservoirs (Meng et al., 2010, 2016; Yao et al., 2011; Feng et al., 2009a, b). These observations implied that Hg methylation will be promoted with the increase of trophic stage in reservoir. Unfortunately, most of the research related to Hg cycling in river-reservoir system have focused on the unique biogeochemistry within reservoirs, while

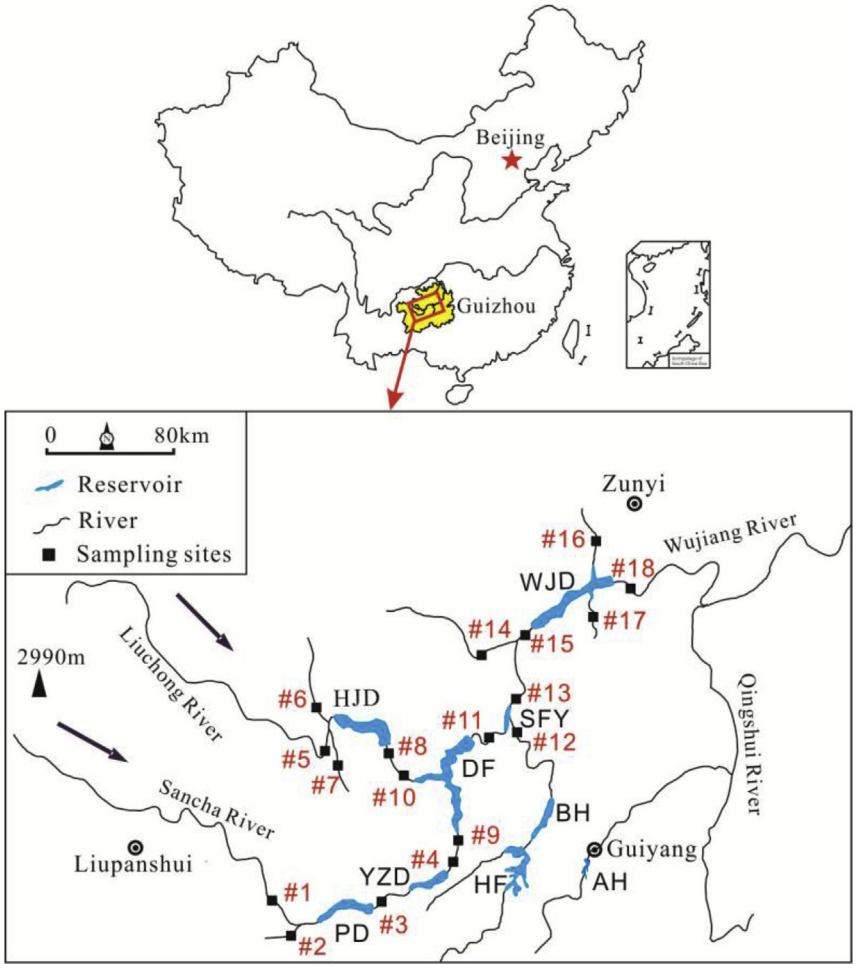

Fig. 1. Map of the study area and sampling sites of inflow-outflow rivers of reservoirs in Wujiang River Basin, Guizhou Province, China. (Hongjiadu Reservoir, HJD; Yinzidu Reservoir, YZD; Suofengying Reservoir, SFY; Puding Reservoir, PD; Dongfeng Reservoir, DF; Wujiangdu Reservoir, WJD; Aha Reservoir, AH; Hongfeng Reservoir, HF; Baihua Reservoir, BH).

little attention has been paid to inflow-outflow rivers of reservoirs. Inflow-outflow rivers provide a link between the watershed and reservoir environment, and therefore, understanding the influence of dam construction on the distribution of Hg species in inflow-outflow rivers of reservoir has significant implication for a large scale Hg biogeochemical cycles.

To better understand the influence of dam construction on the biogeochemical cycling of Hg in river-reservoir ecosystems, we measured concentrations of Hg species over a 12-months period from January to December 2006 in the inflow-outflow rivers of six cascade reservoirs in WRB, Southwest China. The primary objectives of this study were 1) to investigate spatial and temporal variations of Hg species in inflow-outflow rivers of the reservoirs; and 2) to reveal the influence of cascade reservoirs on the distributions and methylation of Hg in rivers-reservoir ecosystem.

2. Materials and methods

2.1. Study area and sites description

The Wujiang River is the largest tributary of the upper Yangtze River, mainly flowing in a karstic environment in Guizhou Province, Southwest China (Fig. 1). Eighteen sampling stations recognized as the inflow and outflow of the six cascade reservoirs were chosen, as depicted in Fig. 1 and Table 1. The six selected reservoirs, including PD, HJD, YZD, SFY, DF, and WJD, are located at the mainstream of the Wujiang River, Southwestern China (Fig. 1). Basic parameters of six selected reservoirs in WRB are shown in Table 2. On the basis of the trophic state of the specific reservoir, the six selected reservoirs in WRB are classified as the oligotrophic-mesotrophic stage (YZD, SFY, and HJD), the mesotrophic-eutrophic stage (PD and DF), and the hyper eutrophic stage (WJD) (Table 2). More information concerning the Wujiang River, the six selected reservoirs, and sampling sites is shown in Text S1, Supplementary Material.

Table 1
Summary of inflow-outflow rivers to reservoirs in Wujiang River Basin, Guizhou province, China.

Reservoir	Inflow into reservoir	Outflow of reservoir
PD[a]	SC (#1) and BY (#2)	Discharge water (#3)
YZD[a]	Outflow of PD (#3)	Discharge water (#4)
HJD[a]	LCH (#5), LJ (#6), MK (#7)	Discharge water (#8)
DF[a]	mainstream (#10) and YZD outflow (#9)	Discharge water (#11)
SFY[a]	Outflow of DF (#11), MT river (#12)	Discharge water (#13)
WJD[a]	YJ river (#14), Mainstream of WJD (#15), PY river (#16), XF river (#17)	Discharge water (#18)

[a] Puding Reservoir, PD; Yinzidu Reservoir, YZD; Hongjiadu Reservoir, HJD; Dongfeng Reservoir, DF; Suofengying Reservoir, SFY; Wujiangdu Reservoir, WJD.

2.2. Sample collection

Surface water samples (20-cm depth) from each sampling site were collected once a month during a one year period from January to December in 2006. All samples were collected following ultra-clean sample-handling protocols. Filtration of water samples were conducted within 24 h after sample collection using a 0.45 μm filter (Millipore), and subsequently analyzed for dissolved Hg (DHg), dissolved MeHg (DMeHg) and dissolved organic carbon (DOC). Total Hg (THg), reactive Hg (RHg), total MeHg (TMeHg), and total suspended solids (TSS) in water samples were analyzed in each of the unfiltered samples. A detailed description of sample collection and preparation is given in the Text S2, Supplementary Material.

2.3. Sample analyses

THg, DHg, and RHg were quantified using the dual-stage gold amalgamation method coupled with cold vapor atomic fluorescence spectroscopy (CVAFS, Tekran 2500, Tekran Inc. Canada) following approved methodologies (Bloom and Fitzgerald, 1988; USEPA, 2002). Particulate Hg (PHg) was calculated as the difference between the concentration of THg and DHg in the sample. Water samples were quantified for MeHg using CVAFS (Brooks Rand Model III, Brooks Rand Labs, U.S.A.) following distillation, aqueous phase ethylation, and isothermal gas chromatographic (GC) separation (Liang et al., 1994; USEPA, 2001). Particulate MeHg (PMeHg) was calculated as the difference between the concentrations of TMeHg and DMeHg in the sample.

TSS was determined gravimetrically by filtering an aliquot of water (typically 1500 mL) through a pre-weighed 0.45 μm pore-size, 47 mm (diameter) polycarbonate membrane filter. Water quality parameters such as pH, temperature (T), Dissolved oxygen

Table 2
Basic parameters of six selected reservoirs in Wujiang River Basin, Guizhou province, China.

Parameters	HJD[a]	YZD[a]	SFY[a]	PD[a]	DF[a]	WJD[a]
Construction time/year	2004	2003	2005	1993	1994	1979
Height of dam/m	180	136	122	75	162	165
Total water volume/10^8 m^3	49.47	4.55	2.01	4.2	10.3	23.0
Surface area of the reservoir/km^2	80.5	15.0	5.7	19.3	19.1	47.8
Annual runoff volume/10^8 m^3	47.0	44.2	124.6	33.8	109	158
Normal water level/m	1140	1086	837	1145	970	760
Distance from river source/km	268	289.9	372	238.4	333	443
Average annual flow/m^3 s^{-1}	155	141	395	123	355	502
Water residence time/day[b]	380	44	7	45	33	53
Regulation mode	Annual	Seasonal	Daily	Seasonal	Seasonal	Seasonal
Total nitrogen/mg L^{-1}[c]	5.21	4.38	7.57	6.25	8.81	8.46
Total phosphorus/mg L^{-1}[c]	0.019	0.003	<0.001	0.019	0.035	0.468
Chl.a/μg L^{-1}[c]	2.25	3.73	1.34	10.36	3.35	8.99
Trophic level index[c]	49	42	40	57	56	72
Trophic stage[c]	I	I	I	II	II	III

I, oligotrophic-mesotrophic; II, mesotrophic-eutrophic; III, hyper eutrophic.
[a] Hongjiadu Reservoir, HJD; Yinzidu Reservoir, YZD; Suofengying Reservoir, SFY; Puding Reservoir, PD; Dongfeng Reservoir, DF; Wujiangdu Reservoir, WJD.
[b] Data obtained from Guo (2008).
[c] Data obtained from Yu (2008).

(DO) were measured in-situ using a portable analyzer (plONneer 65 Portable Multi-parameter Instrument, France). DOC was qualified in the laboratory by using a high-temperature combustion technique with TOC analyzer (Elementar High TOC II).

2.4. Quality assurance and quality control

Quality control for the THg and MeHg determinations was conducted by field blanks, method blank, matrix spikes, and duplicate samples. Specially, the relative standard deviations for duplicate sample analyses were 4.5% and 5.4% for THg and MeHg. Recoveries for matrix spikes (within the range of samples concentrations) ranged from 88 to 108% for the MeHg analysis, and from 93 to 110% for the THg analysis. It should be noted that the mean concentrations of Hg species in inflow/outflow rivers were calculated through the arithmetic averaged concentrations of multiple inflow/outflow rivers throughout all the sampling campaigns. A detailed description of quality control measures was described in Text S3, Supplementary Material, including field blanks, method blank, matrix spikes, duplicate samples, and statistical analysis of the resulting analytical data.

3. Results and discussion

3.1. General water quality characteristics

The distribution of monthly pH, T, DO, DOC, TSS, and conductivity in inflow and outflow rivers of six studied reservoirs in WRB are shown in Table S1 and Table S2, Supplementary Material. The surface water temperature exhibited expected seasonal patterns, with the maximum and minimum values occurring in the summer and winter, respectively. However, the seasonal trend of surface water pH was not clear, though pH in the surface water was slightly alkaline in all of the samples (annual average value of 8.15 ± 0.36). The annual average concentration of DOC in the surface water in the Wujiang River was 0.91 ± 0.42 mg L^{-1}, which was lower than that observed in the Yangtze River, as well as in most rivers globally (Duan, 2000). The high pH and low DOC in surface water can be explained by the karstic geology in WRB. Moreover, DOC showed obvious seasonal patterns, with the highest average DOC values in the summer and lowest values in the winter, which may be caused by the inner source of natural organic matter (NOM) produced in situ (phytoplankton) to the DOC.

Slightly elevated values of TSS in the surface water of WRB were observed in May, June, July, and October. Seasonal patterns of DO were pronounced and well correlated with TSS and T. Elevated DO in surface water during the periods from late May to August compared to the remainder months of the year were explained by algal blooms in some sections of reservoirs, such as the downstream of WJD (Meng et al., 2010; Zhu, 2005). Conductivity showed slightly seasonal distributions that were opposite to that of DOC and TSS, with a decreased average conductivity value in rainy seasons (during the period from August to October). This was believed to be the result of dilution of the river water by rainfall during the rainy season.

3.2. Spatial and temporal variations of total mercury in river water

Temporal distributions of THg, DHg, PHg, and RHg concentrations in surface water in inflow and outflow rivers of the six studied reservoirs are shown in Fig. 2, Fig. S1, Fig. S2 and Fig. S3, Supplementary Material, and summary data are shown in Table S3 and Table S4, Supplementary Material.

The annual mean concentration of THg in inflow-outflow rivers of reservoirs in WRB was 3.41 ± 1.98 ng L^{-1} (ranging from 1.43 ng L^{-1} to 19.04 ng L^{-1}) throughout all the sampling sites. The annual average concentration of THg in the inflow rivers of the reservoirs was 3.90 ± 2.21 ng L^{-1}, with the highest value observed in XF (#17) and the lowest seen in the inflow river of SFY (Outflow of DF, #11). The elevated levels of THg in XF (#17) could be attributed to the point source of Hg from local factories (Xifeng Phosphorus and Calcium Factory) (Text S1 Supplementary, Material). The average concentration of THg in surface water in WRB are comparable with the values that are recognized as uncontaminated rivers of North American and Europe (<5 ng L^{-1}) (Ullrich et al., 2001). Moreover, the mean THg concentration in surface water in WRB is significantly below the Chinese surface water standard of 50 ng L^{-1} (Ministry of Environmental Protection, 2002), and also below the 12 ng L^{-1} standard for THg recommended by the USEPA to protect against adverse chronic effects on aquatic life (USEPA, 1992).

The seasonal distribution patterns and mean concentrations of THg in inflow and outflow rivers of reservoirs in WRB were illustrated in Fig. 2. No discernable difference in the THg concentration in inflow and outflow rivers of reservoirs was observed among the spring, summer, fall and winter (K-W test, $p > 0.05$, $n = 39$), with the inflow river water in PD and HJD as exceptions. The mean concentrations of THg in inflow rivers in PD (5.21 ± 1.96 ng L^{-1}) and HJD (4.22 ± 1.73 ng L^{-1}) were significantly higher than those in the four other reservoirs (2-way ANOVA, $p < 0.01$). The statistical analysis further observed that concentrations of THg in inflow river of PD and HJD in rainy seasons (summer and fall) were significantly

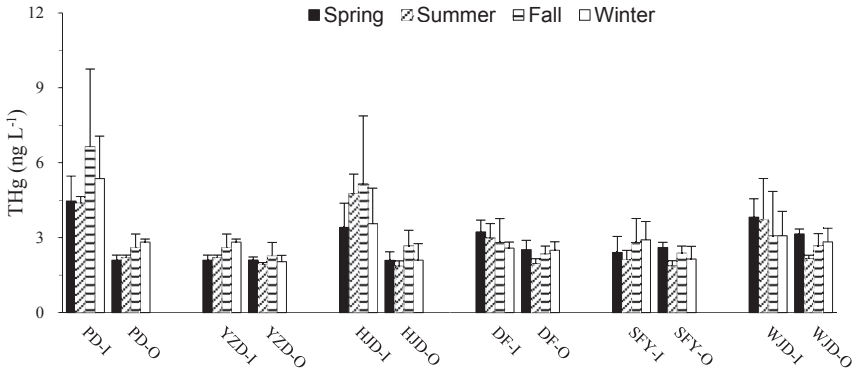

Fig. 2. Annual mean concentrations of total mercury (THg) in inflow and outflow rivers of six reservoirs in Wujiang River Basin, Guizhou Province, China. (PD-I, inflow of PD; PD-O, outflow of PD; YZD-I, inflow of YZD; YZD-O, outflow of YZD; HJD-I, inflow of HJD; HJD-O, outflow of HJD; DF-I, inflow of DF; DF-O, outflow of DF; SFY-I, inflow of SFY; SFY-O, outflow of SFY; WJD-I, inflow of WJD; WJD-O, outflow of WJD).

higher than those in dry season (spring and winter) (T-test, $p < 0.01$).

As shown in Fig. 1, PD and HJD are located at the upper end of WRB. With the increase of rainfall and rainfall intensity during the rainy season, more Hg in surface runoff and sediment re-suspension entered the rivers (Guo, 2008), which resulted in the elevated THg concentration compared with the dry season. When the river water flows into the reservoir, the water flow rate decrease significantly. Therefore, concentrations of TSS in outflow river of PD (2.05 ± 0.81 mg L^{-1}) and HJD (1.28 ± 0.84 mg L^{-1}) were also significantly decreased when compared with that in inflow rivers of PD (3.37 ± 1.12 mg L^{-1}) and HJD (3.14 ± 1.47 mg L^{-1}) (T-test, $p < 0.01$, both for PD and HJD). Consequently, a considerable amount of Hg bound to particulate matter was intercepted by way of sedimentation (Guo, 2008), which explains the lower THg concentrations in outflow rivers of reservoirs compared with that in inflow rivers in PD and HJD. As shown in Fig. 2, THg concentrations in outflow rivers of reservoirs were lower than that in the corresponding inflow river throughout the six reservoirs (T-test, $p < 0.05$), which was primarily caused by the sedimentation of PHg after the construction of reservoirs. Previous studies observed that the construction of the reservoir reduced the concentration of Hg in river water (Bonzongo et al., 1996; Lawson et al., 2001; Quemerais et al., 1999), which is consistent with our results.

The statistical analysis showed that significant positive correlations were observed between PHg and TSS in inflow rivers throughout the six reservoirs during the sampling periods (Table 3), indicating that the Hg bound to particulate matter from runoff (riverbank and riverbed) was the primary source of PHg in inflow rivers. Furthermore, concentrations of THg in inflow rivers were significantly correlated with PHg and TSS, with correlation coefficients of 0.98 and 0.62, respectively, indicating that the level of THg in inflow rivers was controlled by PHg and TSS. However, no correlation was observed between TSS and THg, PHg in outflow rivers of reservoirs during the sampling periods (TSS *vs.* THg: $r = -0.04$, $p = 0.71$, $n = 72$; TSS *vs.* PHg: $r = -0.07$, $p = 0.58$, $n = 72$), which could be explained by the dissolution of PHg in reservoir and contribution of planktonic derived NOM to TSS. Our results were consistent with observations by previous studies (Lawson et al., 2001; Guentzel et al., 2007; Paraquetti et al., 2004).

The annual mean concentration of PHg in rivers (inflow and outflow rivers) of the reservoirs in WRB was 2.05 ± 1.73 ng L^{-1} (ranging from 0.62 ng L^{-1} to 17.1 ng L^{-1}) throughout all the sampling sites. The annual mean concentrations of PHg in inflow river and outflow rivers were 2.46 ± 1.95 ng L^{-1} and 1.15 ± 0.26 ng L^{-1}, respectively. As shown in Fig. S1, Supplementary Material, concentrations of PHg in inflow rivers in the rainy seasons (summer and fall) were generally higher than those in the dry seasons (winter and spring) (T-test, $p < 0.05$), especially at PD and HJD, which was similar to the seasonal trend of THg. Furthermore, the PHg concentration in outflow rivers was significantly lower than that in inflow rivers throughout the six reservoirs during the sampling periods (T-test, $p < 0.05$). Due to the relative low primary productivity (Chlorophyll *a*) of the inflow rivers of reservoirs in WRB (Zhu, 2005), the adsorption of Hg by phytoplankton was very limited. Our previous study showed that concentrations of PHg in the bottom water from upstream to downstream of WJD were all significantly higher than the corresponding overlying water, confirming the input of particulate Hg from sediment resuspension (Meng et al., 2010). When compared to reservoir, inflow river represents a relatively strong hydrodynamic condition. Hence, it is reasonable to speculate that the PHg in inflow river water of WRB was primarily from runoff together with sediment re-suspension (Hurley et al., 1998; Meng et al., 2010).

PHg represented $56.1 \pm 10.1\%$ of THg in the river water. The statistical analysis showed that the ratio of PHg to THg (PHg/THg) in inflow rivers (59%) was significantly higher than that in outflow rivers (49%) throughout the six reservoirs during the sampling periods (T-test, $p < 0.05$), which can be explained by the interception effect of reservoirs to particulate materials but not the dissolution of PHg in reservoir. However, the ratios of PHg/THg in inflow rivers of WRB were much lower than the observations in inflow rivers (Fox river) of Lake Michigan in North American (93.6%) (Hurley et al., 1998). In this study, the average ratio of PHg/THg in the inflow rivers of reservoirs during rainy season (61%) was slightly higher than that during dry season (57%) (T-test, $p > 0.05$). Although the temporal/seasonal trend of PHg/THg was not clear, the spatial distribution of PHg/THg in inflow rivers was clearly observed in this study (Fig. S4, Supplementary Material). In detail, the average ratios of PHg/THg in inflow rivers of PD (63%) and HJD (66%), which are located in the upstream section of WRB, were significantly higher than those in inflow rivers of downstream reservoirs (e.g. YZD, 44%; DF, 55%; SFY, 53%) (T-test, $p < 0.05$). The clear spatial distribution patterns of PHg/THg in inflow rivers of WRB further confirmed that particulate matter from soil erosion was the potential sources of PHg in the inflow rivers of the upstream reservoirs in WRB (Zhang et al., 2009; Meng et al., 2010; Yao et al., 2011).

The annual mean concentration of DHg in river water (inflow and outflow rivers) in WRB was 1.36 ± 0.44 ng L^{-1} (ranging from 0.57 ng L^{-1} to 3.1 ng L^{-1}) throughout all the sampling sites. The annual mean concentration of DHg in inflow rivers (1.44 ± 0.48 ng L^{-1}) was slightly higher than that in outflow rivers (1.18 ± 0.28 ng L^{-1}). However, no discernable spatial and seasonal trends of DHg in the rivers was observed throughout the six reservoirs during the sampling periods (Fig. S2, Supplementary Material), which was different from that for THg and PHg. The Partition Coefficient (K_d) between Hg in the particulate phase (solid) and the dissolved phase (liquid) was calculated based on the measured concentrations of PHg, DHg, and TSS, as described in Hurley et al. (1998) and Covelli et al. (2006). Our calculated data showed that the average $\log K_d$ in rivers of WRB was 5.8 ± 0.20, with values in inflow rivers and outflow rivers of 5.8 ± 0.2 and 5.9 ± 0.3, respectively. The $\log K_d$ observed in this study was slightly higher than that in other rivers globally, such as the inflow rivers of the Chesapeake Bay (5.06–5.52) (Lawson et al., 2001) and the Fox River (5.4–6.0, Hurley et al., 1998). However, our results showed similar levels of $\log K_d$ when compared to the Isonzo River (5.5–6.2, Covelli et al., 2006) and the Lot-Garonne River (5.2–6.2, Schafer et al., 2006).

The annual mean concentration of RHg in river water (inflow and outflow rivers) in WRB was 0.24 ± 0.11 ng L^{-1} (ranging from

Table 3
Pearson's Correlation Matrix, giving the Linear Correlation Coefficients (r) among the concentrations of Hg species and total suspended solid (TSS) in inflow rivers of reservoirs in Wujiang River Basin, Guizhou Province, China (n = 168).

	THg	PHg	DHg	RHg	TMeHg	PMeHg	DMeHg	TSS
THg[a]	1							
PHg[a]	0.98[c]	1						
DHg[a]	0.61[c]	0.45[c]	1					
RHg[a]	0.49[c]	0.50[c]	0.23[c]	1				
TMeHg[a]	0.48[c]	0.50[c]	0.17[b]	0.26[c]	1			
PMeHg[a]	0.58[c]	0.61[c]	0.16[b]	0.37[c]	0.93[c]	1		
DMeHg[a]	0.15	0.14	0.14	0.02	0.76[c]	0.49[c]	1	
TSS[a]	0.62[c]	0.62[c]	0.31[c]	0.31[c]	0.47[c]	0.53[c]	0.21[c]	1

[a] THg, total mercury; PHg, particulate mercury; DHg, dissolved mercury; RHg, reactive mercury; TMeHg, total methylmercury; PMeHg, particulate methylmercury; DMeHg, dissolved methylmercury; TSS, total suspended solid.
[b] Correlation is significant at the 0.05 level (2-tailed).
[c] Correlation is significant at the 0.01 level (2-tailed).

0.09 ng L^{-1} to 1.01 ng L^{-1}) throughout all the sampling sites. The annual mean concentrations of RHg in inflow rivers and outflow rivers were 0.25 ± 0.12 ng L^{-1} and 0.22 ± 0.08 ng L^{-1}, respectively. There was no significant difference in RHg concentrations between inflow rivers and outflow rivers because RHg in water primary exists in a dissolved phase. (T-test, $p > 0.05$) (Fig. S3, Supplementary Material). However, the ratio of RHg/THg in inflow rivers (7.4%) was significantly lower than that in outflow rivers (9.7%) (T-test, $p < 0.05$). Compared with inflow rivers, concentrations of THg in outflow rivers were significantly decreased due to the interception effect of the reservoir. Therefore, the relatively higher RHg/THg in outflow rivers (compared with inflow rivers) can be explained by the decreased THg in outflow rivers (compared with inflow rivers) together with the stable RHg concentrations between inflow rivers and outflow rivers.

3.3. Spatial and temporal variations of methylmercury in river water

The annual mean concentration of TMeHg in river water (inflow and outflow rivers) in WRB was 0.15 ± 0.06 ng L^{-1} (ranging from 0.07 ng L^{-1} to 0.70 ng L^{-1}) throughout all the sampling sites. The annual mean concentrations of TMeHg in inflow rivers and outflow rivers were 0.14 ± 0.06 ng L^{-1} and 0.17 ± 0.06 ng L^{-1}, respectively. Seasonal distribution patterns and concentrations of TMeHg in inflow rivers and outflow rivers are illustrated in Fig. 3. The statistical analysis showed that TMeHg concentrations in inflow rivers in the summer were significantly higher than those in the three other seasons (winter, spring, and fall) (T-test, $p < 0.05$). The elevated TMeHg concentrations in inflow rivers during the summer can be explained by the increased surface runoff due to the sufficient precipitation (Zhang et al., 2009; Meng et al., 2010), which subsequently resulted in the increase of MeHg entering into river water through surface runoff and sediment re-suspension pathways. Furthermore, another reason could be the higher water temperature together with the increase amount of algal derived NOM during summer, which can stimulate bacterial activity and Hg methylation (Bravo et al., 2017).

A significantly positive correlation between TMeHg and TSS in inflow rivers was observed in this study, with a correlation coefficient of 0.47 ($p < 0.001$, $n = 168$) (Table 3). Moreover, TMeHg concentrations in inflow rivers were positively correlated with PMeHg ($r = 0.93$, $P < 0.001$, $n = 168$). The significant correlations between TMeHg and TSS as well as PMeHg in inflow rivers implied that river erosion and surface runoff could be the main sources of MeHg in the inflow rivers of the six reservoirs in WRB. The outflow river water was mainly from discharged water of the reservoirs. In this study, concentrations of TMeHg in outflow rivers in warm seasons (especially during the periods from July to September) were significantly higher than those in the dry seasons (T-test, $P < 0.01$). Furthermore, TMeHg concentrations in outflow rivers was also significantly higher than that in inflow rivers especially in rainy seasons (T-test, $P < 0.05$). These observations implied the active Hg methylation in reservoirs during warm seasons. Canavan et al. (2000) observed that concentrations of MeHg in the discharged water of a reservoir in New Mexico were approximately six times higher from late summer to early fall. This observation shown above were consistent with the results in this study.

The average ratio of TMeHg to THg (TMeHg/THg) in inflow-outflow rivers throughout the six reservoirs was 5.3 ± 2.9%. The ratios of TMeHg/THg in inflow rivers throughout the six reservoirs exhibited the following distribution patterns (Fig. S5, Supplementary Material): summer (5%)>spring, fall (4%)>winter (3%), with the corresponding trend of summer (11%) > fall (7%) > spring, winter (6%) in outflow rivers. Furthermore, the ratio of TMeHg/THg in outflow rivers (7.4%) was significantly higher than that in inflow rivers (4.3%) (T-test, $p < 0.05$), which further confirmed the net Hg methylation in the reservoirs.

The annual mean concentration of DMeHg in river water (inflow and outflow rivers) in WRB was 0.08 ± 0.03 ng L^{-1} (ranging from 0.03 ng L^{-1} to 0.26 ng L^{-1}) throughout all the sampling sites. The annual mean concentrations of DMeHg in inflow rivers and outflow rivers were 0.07 ± 0.03 ng L^{-1} and 0.10 ± 0.04 ng L^{-1}, respectively. Concentrations of DMeHg in outflow rivers were significantly higher than those in inflow rivers (T-test, $p < 0.05$). The higher DMeHg concentrations in outflow rivers can be attributed to the discharge of water from the hypolimnion with high levels of MeHg. Muresan et al. (2008) observed that MeHg concentrations in the hypolimnion of reservoirs were approximately 3 times higher than those in the upper water column, which support our results.

The seasonal distribution patterns of DMeHg in the inflow-outflow rivers of six reservoirs are illustrated in Fig. S6, Supplementary Material. There were no discernable differences in the DMeHg of inflow rivers among spring, summer, fall and winter (K-W test, $p > 0.05$). However, DMeHg concentrations in outflow rivers was significantly higher than that in inflow rivers especially in rainy seasons (T-test, $P < 0.01$). Furthermore, DMeHg concentrations in outflow rivers of the six reservoirs in rainy seasons (summer and fall) were significantly higher than those in dry seasons (spring and winter) (T-test, $p < 0.05$), which was consistent with the seasonal trend of TMeHg in outflow rivers. Recently, we observed that both the newly constructed reservoir (e.g. YZD) and old reservoir (e.g. WJD) in WRB were completely stratified during rainy seasons, especially at the downstream section of the

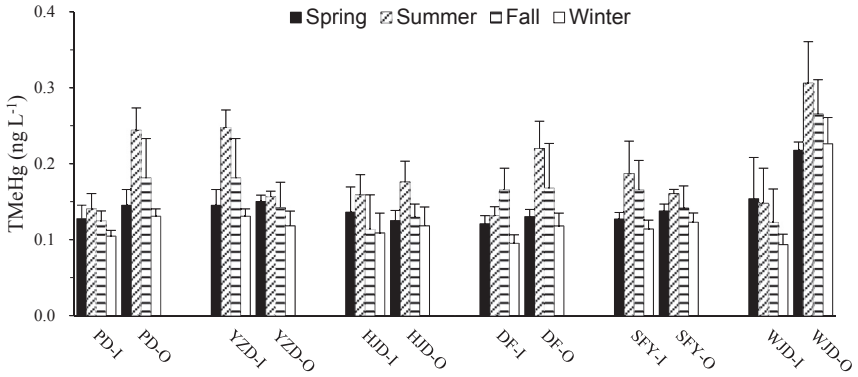

Fig. 3. Annual mean concentrations of total methylmercury (TMeHg) in inflow and outflow rivers of six reservoirs in Wujiang River Basin, Guizhou Province, China. (PD-I, inflow of PD; PD-O, outflow of PD; YZD-I, inflow of YZD; YZD-O, outflow of YZD; HJD-I, inflow of HJD; HJD-O, outflow of HJD; DF-I, inflow of DF; DF-O, outflow of DF; SFY-I, inflow of SFY; SFY-O, outflow of SFY; WJD-I, inflow of WJD; WJD-O, outflow of WJD).

reservoirs (Meng et al., 2010). Furthermore, MeHg concentrations and the ratios of MeHg/THg in hypolimnion of reservoirs were highly elevated compared with overlying water, indicating the active net Hg methylation in the stratification zone of water column (Meng et al., 2010). A previous study confirmed that the elevated DMeHg in hypolimnion of reservoirs during summer and fall can result in the increase of DMeHg concentrations in outflow rivers (Watras et al., 1995). Choe and Gill (2003) reported that the DMeHg bound to organic colloids can pass through the 0.45 μm size of filter membrane and exist in liquid phase. These observations shown above support our results.

The average ratio of DMeHg to TMeHg (DMeHg/TMeHg) in inflow-outflow rivers throughout the six reservoirs was 50.7 ± 7.7%. Specially, the ratio of DMeHg/TMeHg in outflow rivers (55%) was slightly higher than that in inflow rivers (48%), but was not significantly different (T-test, $p > 0,05$), which could be attributed to the dissolution of PMeHg in water body along the reservoirs. We further calculated the ratios of DMeHg to DHg (DMeHg/DHg) in the river water of the six reservoirs. Our results showed that the average ratios of DMeHg/DHg in inflow rivers (5.2 ± 2.7%) were significantly lower than those in outflow rivers (8.4 ± 4.3%) (T-test, $p < 0.05$), which could be explained by the net Hg methylation in reservoirs. Coquery et al. (2003) further observed that the levels of DMeHg/DHg in surface water of reservoirs were similar to inflow rivers; however, the ratios of DMeHg/DHg in the hypolimnion of reservoirs were approximately 10 times higher than those in surface water, indicating active Hg methylation in the hypolimnion.

4. Influence of damming on the distribution and methylation of mercury in Wujiang River

Current study observed the clear sedimentation of Hg in reservoirs within WRB (Fig. 4). Similar to the spatial distribution patterns of TSS in inflow-outflow rivers of reservoirs, concentrations of THg also gradually decreased from upstream to downstream of WRB (Fig. 4). For example, the annual mean THg concentrations in the inflow rivers of PD (SC) and HJD (LC) were 4.65 ± 1.28 ng L^{-1} and 4.50 ± 2.13 ng L^{-1}, respectively. Under the sedimentation processes of Hg in PD and HJD, concentrations of THg (annual mean concentration) in the outflow rivers (reservoir discharge) of PD and HJD significantly decreased (approximately 2 times), to 2.43 ± 0.40 ng L^{-1} and 2.18 ± 0.53 ng L^{-1}, respectively. These results indicated that a great quantity of Hg was intercepted by the reservoirs and stored probably in the sediment (Text S4, Supplementary Material). On the other hand, reservoir's scavenging effect on Hg was clearly weakened in the four other reservoirs, which were located in the downstream portions of WRB (Text S4, Supplementary Material). It is generally acceptable that sediment and/or hypolimnion are very important site for Hg methylation. Therefore, the dam intercepted Hg can be transformed into MeHg in reservoir, posing a potential threat to river-reservoir ecosystems.

Previous studies showed that dam construction significantly changed the concentrations and spatial distribution patterns of MeHg in river (Canavan et al., 2000) and subsequently resulted in the increase of MeHg levels in downstream river. Canavan et al. (2000) further observed that the concentrations of MeHg were still very high in the outflow river of the reservoir, approximately 22 km away from the dam. To better understand the influence of dam construction on the distributions of MeHg in the Wujiang River, the six selected reservoirs in WRB are classified as the oligotrophic-mesotrophic stage (YZD, SFY, and HJD), the mesotrophic-eutrophic stage (PD and DF), and the hyper eutrophic stage (WJD) based on the trophic state of the specific reservoirs (Table 2). We further observed the distributions of TMeHg and DMeHg in inflow-outflow rivers of different trophic stages of

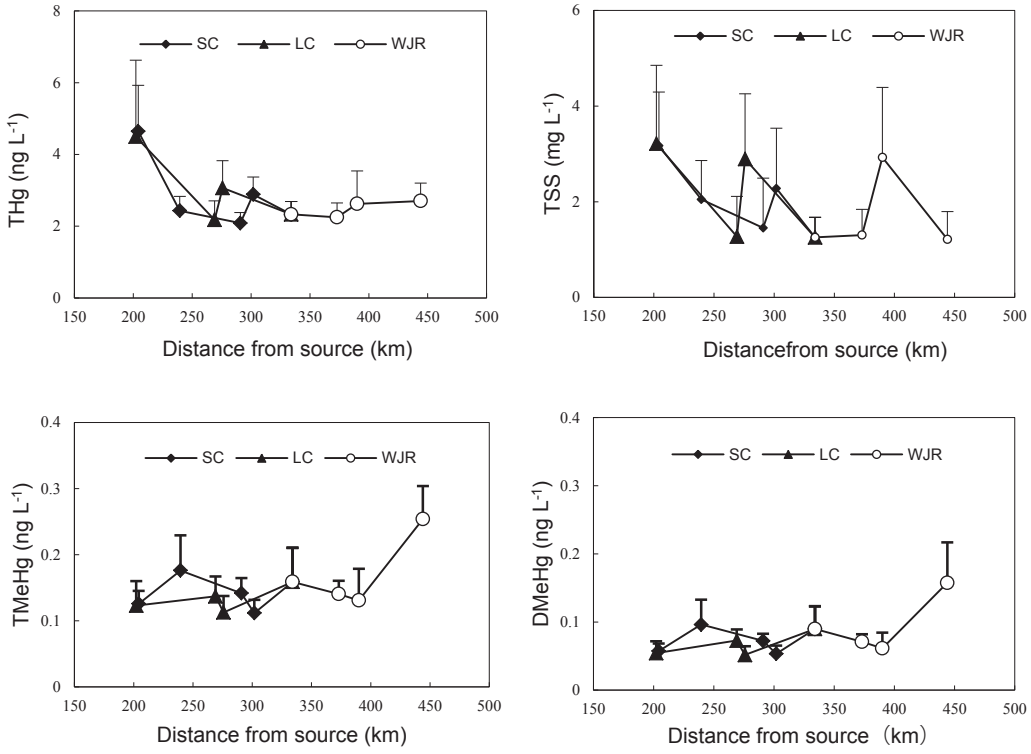

Fig. 4. Spatial distributions of total Hg (THg), total suspended solid (TSS), total methylmercury (TMeHg), and dissolved methylmercury (DMeHg) in different sections of Wujiang River, Guizhou Province, China. (SC, Sancha River; LC, Liuchong River; WJR, Mainstream of Wujiang River; the source of Wujiang River is Xianglu mountain in Weining county, Guizhou Province, China).

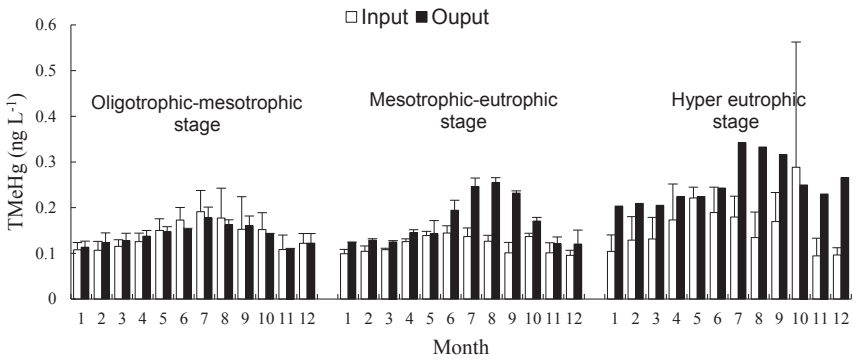

Fig. 5. Monthly mean concentrations of total methylmercury (TMeHg) in inflow-outflow rivers in different trophic stages of reservoirs in Wujiang River Basin, Guizhou Province, China. (YZD, SFY, and HJD with oligotrophic-mesotrophic stage; PD and DF with mesotrophic-eutrophic stage; WJD with hyper eutrophic stage).

reservoirs (Fig. 5 and Fig. S7, Supplementary Material). As shown in Fig. 5, the TMeHg concentrations in outflow rivers of PD, DF, and WJD significantly increased by the factors of 46%, 53%, and 92% compared with the data in the inflow rivers of these three reservoirs, respectively.

Previous studies showed that apart from the age of the reservoir, many other factors including flooded soil type (e.g. organic matter content and Hg concentration), water volume, water residence time, water temperature, water chemistry, water depth in reservoir, may also exert an influence on net MeHg methylation in reservoirs (St. Louis et al., 2004; Ullrich et al., 2001). As listed in Table 2, the average water residence time in YZD (44 days) was 6.3 times higher than that in SFY (7 days), but was an order of magnitude lower than that in HJD (380 days). Furthermore, the total water volume in YZD was ~2 times higher than that in SFY, but was ~11 times lower than that in HJD. However, the distribution patterns of MeHg within inflow-outflow rivers (elevated from inflow rivers to outflow rivers of reservoirs) were less pronounced in the newly constructed reservoirs in WRB (e.g., YZD, SFY, and HJD). In contrast, both the water residence time and total water volume in YZD, PD, DF, and WJD was similar. However, the statistical analysis showed that concentrations of TMeHg and DMeHg in the outflow rivers in the hyper eutrophic stage reservoirs (WJD) were significantly higher than those in oligotrophic-mesotrophic (YZD, SFY, and HJD) and mesotrophic-eutrophic (PD and DF) stage reservoirs (K-S test, $p < 0.01$, $n = 18$), indicating that the net Hg methylation and the output of MeHg from reservoirs increased with the increase of trophic stage in reservoir ecosystem (Text S4, Supplementary Material). The six cascade reservoirs which are located in the karstic environment of WRB, are typical deep-valley, high-mountain gorges. Hence, it was reasonable that the selected reservoirs represented similar geological background (e.g. water depth of reservoirs, organic matter content and Hg concentrations in submerged soil) (Meng et al., 2010; 2016; Yao et al., 2011; Feng et al., 2009a, b). Therefore, current study jointly with the previous observations implied that the primary indicator for Hg methylation could not be the water residence time, total water volume, and basis geological background in reservoirs. In comparison, our observations illustrated that primary productivity (trophic state) could be one of the most important factors controlling the net Hg methylation in reservoirs in WRB. The increase in planktonic derived NOM could be the possible reason to explain the enhanced MeHg production in the reservoirs in WRB (Bravo et al., 2017). Furthermore, the construction of the cascade reservoirs resulted in the elevation of MeHg in several sections of the Wujiang River (Fig. 4). For example, the MeHg concentration exhibited the highest level in the section that was approximately 445 km distant from the source of the Wujiang River (Fig. 4). The elevated MeHg levels in specific sections of Wujiang River can be attributed to the net Hg methylation in reservoirs and discharge of MeHg into the downstream areas of the Wujiang River. MeHg-enriched water in outflow rivers could be transported to downstream areas, posing potential threat to the aquatic food web and human health.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.chemosphere.2017.07.077.

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