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Mercury mass balance study in Wujiangdu and Dongfeng Reservoirs, Guizhou, China

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Reservoirs are the sink of total mercury but source of methylmercury to the aquatic systems.

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ABSTRACT

From October 2003 to September 2004, we conducted a detailed study on the mass balance of total mercury (THg) and methylmercury (MeHg) of Dongfeng (DF) and Wujiangdu (WJD) reservoirs, which were constructed in 1992 and 1979, respectively. Both reservoirs were net sinks for THg on an annual scale, absorbing 3319.5 g km^{-2} for DF Reservoir, and 489.2 g km^{-2} for WJD Reservoirs, respectively. However, both reservoirs were net sources of MeHg to the downstream ecosystems. DF Reservoir provided a source of 32.9 g MeHg km^{-2} yr^{-1}, yielding 10.3% of the amount of MeHg that entered the reservoir, and WJD Reservoir provided 140.9 g MeHg km^{-2} yr^{-1}, yielding 82.5% of MeHg inputs. Our results implied that water residence time is an important variable affecting Hg methylation rate in the reservoirs. Our study shows that building a series of reservoirs in line along a river changes the riverine system into a natural Hg methylation factory which markedly increases the %MeHg in the downstream reservoirs; in effect magnifying the MeHg buildup problem in reservoirs.

1. Introduction

Reservoirs are world-widely created for various purposes including the production of hydroelectricity, irrigation, flood control, fisheries production, and recreation (The World Commission on Dams, 2000). Many environmental and socio-economic consequences result from reservoir development (Dionne and Therien, 1997). Since early 1970s, scientists have observed that methylmercury (MeHg) concentrations in fish from newly created reservoirs are much elevated compared to fish from adjacent natural lakes (Smith et al., 1974; Abernathy and Cumbie, 1997; Cox et al., 1979). MeHg is produced from inorganic mercury by bacterial activity, which is enhanced by the decomposition of flooded vegetation and organic carbon in soils (Lucotte et al., 1999). From a toxicological perspective, MeHg is the most important form of Hg because it bioaccumulates in food chains and is a strong neurotoxin for human and wildlife. It is well known that reservoirs have mercury contamination problems in their fisheries that last for several decades after flooding (Bodaly et al., 1984; Verdon et al., 1991). It was recently estimated that there are now over 1.5 million km^2 of reservoir surface area globally (St Louis et al., 1994), making this a widespread environmental and socio-economic problem for populations or individuals that rely on reservoir fisheries for subsistence and fish production. Although many studies have been conducted in North America and Europe to understand the process leading to high MeHg concentrations in fish in newly created reservoirs (Kelly et al., 1997; St Louis et al., 1994; Lucotte et al., 1999; Porvari, 1998), there remains a long way to go to fully understand the mechanism.

The total number of large dams in China has exceeded 50% of the total number all over the world since 1982, and the increase rate of the total number of large dams in China is much faster than that of the rest of the world (The World Commission on Dams, 2000), which is obviously due to the rapidly increasing demands for energy needs resulting from economic development. The famous "three Gorge Dam" will be the central concern for the Chinese scientific community in the near future in terms of the environmental consequences. Obviously, one of the most important issues is the possible MeHg contamination in fish. Unfortunately, studies related to mercury biogeochemical cycling in reservoirs in China are extremely limited in number, although it is reported that mercury concentrations in fish from Gezhouba Reservoir have exceeded those allowed marketing and human consumption which is 0.3 mg kg^{-1} in China (Jin and Xu, 1997).

Southwestern China is the most abundant region for water resources in China due to distinct climate conditions. With the implementation of the "Go West" policy, a great number of large reservoirs have been and are being constructed along a dozen of

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large rivers in this region (e.g. Wujiang, Jinshajiang, Yalongjiang, Mingjiang, Daduhe, Lanchangjiang, Nujiang, and Hongshuihe Rivers) for electricity production. Meanwhile, Southwestern China is also the most contaminated area in terms of Hg due to special geological background features and anthropogenic emissions (Feng and Qiu, 2008). First, Southwestern China is situated in the Circum-Pacific global mercuriferrous belt, and dozens of Hg and other metals mines are distributed in this region. As a result, the background Hg concentrations ($\sim$0.1 mg kg^{-1}) in soil in this region are significantly elevated compared to the average background soil Hg concentration in China, which is 0.038 mg kg^{-1} (Environmental Monitoring Center, 1992; Feng et al., 2006). Both Hg and other metal mining activities have released a great amount of Hg to surface waters in this region (Feng et al., 2004a; Qiu et al., 2005, 2006). Moreover, acid rain caused by coal combustion emissions is also regarded as an important environmental concern in Southwestern China, especially Guizhou province (Feng et al., 2002; Hao et al., 2001). Hg concentrations in coals produced in this region are much higher than that in other provinces in China (Feng et al., 2002; Feng and Qiu, 2008), probably due to the special geological background. As a result, Guizhou Province is regarded as one of the largest mercury emission source regions in China (Streets et al., 2005), and the total Hg deposition rates in this region reached 28.0–195 μg m^{-2} month^{-1} (Tan et al., 2000), which were significantly elevated compared to the wet deposition rates reported in Europe and North America (e.g. Vanarsdale et al., 2005; Lindqvist, 1991). Coal combustion from both domestic and industrial sectors, artisanal zinc smelting processes, and mercury mining activities are the major anthropogenic Hg emission sources in the province (Feng et al., 2002, 2004a; Feng and Qiu, 2008). Meanwhile Hg emissions

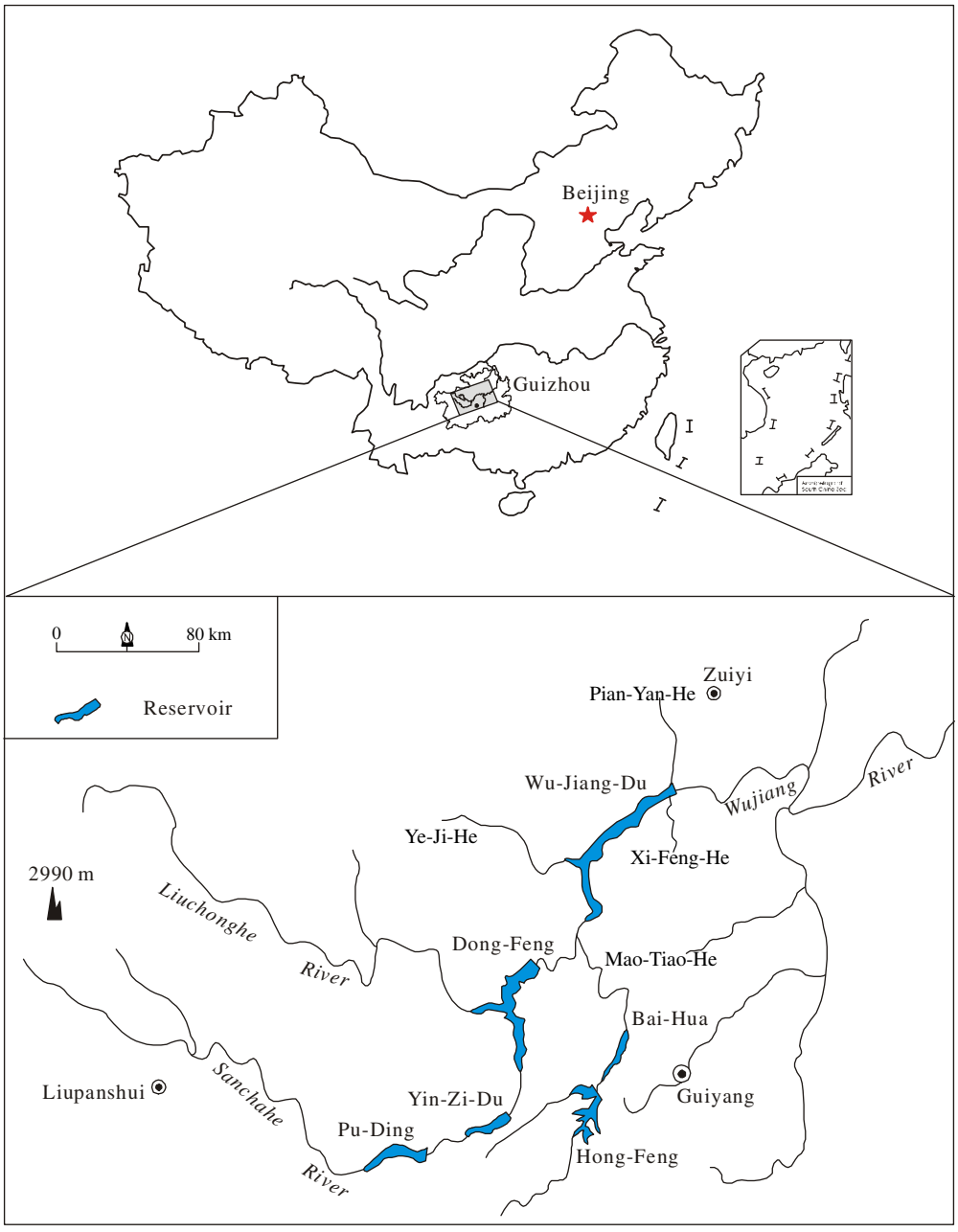


Fig. 1. Location of study area in Wujiang river, Guizhou, China.

from Hg enriched soils in the Province are also considered to be important regional atmospheric Hg emission sources (Feng et al., 2005; Wang et al., 2005, 2007a,b). Consequently, TGM concentrations in ambient air of Guiyang, the capital city of the province were significantly elevated (averaged at 8.4 ng m^{-3}) compared to the global background values (1.5–2.0 ng m^{-3}) (Feng et al., 2004c; Ebinghaus et al., 2002). Due to the special geological background and anthropogenic Hg emissions, the surface water in this region is contaminated with Hg. For example, THg concentrations in surface water of Wujiang River ranged from 10.9 to 329.6 and 2.6 to 125.7 ng L^{-1} during rainy season and dry season, respectively (Jiang et al., 2004), which were significantly elevated compared to the values reported in Europe and North America (Lindqvist, 1991; St Louis et al., 1994), which were generally much less than 10 ng L^{-1}. From October 2003 to September 2004, we for the first time conducted an intensive study on the mercury balance in Wujiangdu (WJD) and Dongfeng (DF) Reservoirs which are adjacent reservoirs created on Wujiang River.

2. Materials and methods

2.1. Site description

WJD and DF Reservoirs are located on the Wujiang River, which is a branch of Yangtze River, in Guizhou Province, Southwestern China (Fig. 1). Before flooding, there were agriculture farmlands distributed along the valleys. The reservoirs lie on the Yunnan-Guizhou Plateau with altitudes varying from 700 to 1200 m 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 °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 predominant inflow into DF Reservoir is from Liuchonghe (LCH) and Sanchahe (SCH) rivers (Fig. 1). There are five inflows into WJD Reservoir, and the largest inflow is from the outflow of DF Reservoir. Mao-Tiao-He (MTH), Ye-Ji-He (YJH), Pian-Yan-He (PYH), and Xi-Feng-He (XFH) are the other 4 water inflows to WJD Reservoir (Fig. 1). Beside the inflows from rivers, groundwater and runoffs from the upland also flow into the reservoirs. The basic characteristics of the reservoirs are listed in Table 1.

2.2. Sampling methods and analytical techniques

All hydrological inputs and outputs to the two study reservoirs were sampled during one year period from October 2003 to September 2004 (Table 2, Figs. 2 and 3). Since the two reservoirs are adjacent, we set up one precipitation sampling site beside WJD Reservoir to represent the precipitation samples for both. The design of the precipitation collector followed the Swedish style (Lindqvist, 1991), and the funnel is made of borosilicate glass. It is a bulk collection sampler with the funnel open all the time. The precipitation sample was stored in a pre-cleaned 500 mL borosilicate bottle in which 10 mL concentrated HCl with low mercury blank was added to prevent adsorption of mercury to the surface of the container. The precipitation samples were collected bi-weekly. Landis and Keeler (1997) showed that the volume-weighted averages for the bulk and the wet-only event samples were comparable, so that our two-weekly bulk collections represented predominantly wet deposition. Both filtered (0.45 μm Millipore membrane filter) and unfiltered water samples for MeHg and total Hg (THg) were taken in pre-cleaned borosilicate glass bottles. Water samples from SCH, LCH, outflow from DF Reservoir and WJD Reservoir were collected monthly. Water samples from MTH, YJH, XFH, PYH rivers, runoff and groundwater were only collected in December 2003 for the dry season and in July 2004 for the rainy season. All collection samples were preserved by adding 0.5% (v/v) of sub-boiling distilled ultra-pure HCl acid within 48 h. Water samples were preserved in a refrigerator at 4 °C. Water samples for dissolved gaseous mercury (DGM) concentration analysis in surface water of both reservoirs were taken in December 2003, April 2004 and July 2004, respectively. 300 mL of surface water was transferred immediately after collection into an extensively cleaned borosilicate glass impinger which was wrapped with black paper to exclude sunlight, and purged with mercury free argon with a flow rate of 300 mL min^{-1} for 30 min. 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 in water samples were analyzed within 28 days after sampling using the dual-stage gold amalgamation method and CVAFS detection according to the method described by Qiu et al. (2006). MeHg in waters were analyzed using distillation and ethylation processes and GC-CVAFS detection followed US EPA Method 1630 (2001). MeHg could be detected at concentrations above 0.01 ng L^{-1} at a blank level of 0.045 ng L^{-1}. The detection limit for THg was 0.2 ng ng L^{-1} at a blank level of 0.3 ng L^{-1}. Recoveries on matrix spikes of MeHg in water samples were in the range of 88.2–108.4%.

Table 1
Basic parameters of Dongfeng and Wujiangdu Reservoirs.

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

2.3. Mass balance budget calculations

Input–output calculations were done as described in St Louis et al. (1994, 2004). The basic equation used in the input–output budget calculations for each of the reservoirs (St Louis et al., 1994, 2004) was

$$\mathrm{Net}_{(\mathrm{MeHg,THg,Water})} = \sum \mathrm{O}_{(\mathrm{MeHg,THg,Water})} - \sum \mathrm{I}_{(\mathrm{MeHg,\ THg,\ Water})}$$

where for MeHg, THg, or water, $\sum \mathrm{I}_{(\mathrm{MeHg,\ THg,\ Water})}$ was the monthly or annual sum of all inputs to the reservoirs, $\sum \mathrm{O}_{(\mathrm{MeHg,\ THg,\ Water})}$ was the monthly or annual sum of all outputs from the reservoirs (Table 2). Mass inputs to both reservoirs originated in rivers inflow, direct precipitation, direct runoff from upland, and groundwater. For THg, dry deposition was also considered as one of the mass inputs. Previous studies have found that dry deposition of THg is about 46% of total deposition in our study area (Guizhou Province) (Tan et al., 2000). We used this ratio to estimate total deposition inputs of THg at both reservoirs (Tables 6 and 7; Figs. 2 and 3). The mass output for MeHg and water was based on the monthly or annual output from reservoir outflow (Table 2). For THg outputs, evaporation of Hg^0 from the water surface to the atmosphere is also included because studies showed that natural water surfaces are important atmospheric Hg emission sources (Feng et al., 2004b, 2008; Lindberg and Zhang, 2000; Wängberg et al., 2001). The evaporation flux of Hg from water surface was estimated using the thin film gas-exchange model, according to the following equation (1):

Table 2
Summary of inputs of water, MeHg, THg, and total suspended solid to Dongfeng and Wujiangdu reservoirs.

Parameter	Comments
Inputs to Dongfeng Reservoir	
Deposition	Concentrations measured in bulk deposition; rainfall data collected from the local meteorological station
Inflow from SCH, LCH rivers	Concentrations measured; water volume data collected from a hydrological station nearby
Runoff from uplands	Concentrations measured; water data collected from a hydrological station nearby
Groundwater	Concentrations measured; water data collected from a hydrological station nearby
Outputs to Dongfeng Reservoir	
Outflow	Concentrations measured; water volume data collected from a hydrological station nearby
Evasion of Hg^0 from water surface	Hg^0 evasion fluxes were calculated based on DGM measurement
Inputs to Dongfeng Reservoir	
Deposition	Concentrations measured in bulk deposition; rainfall data collected from the local meteorological station
Inflow from Dongfeng reservoir	Concentrations measured; water volume data collected from a hydrological station nearby
Inflows from MTH, YJH, XFH, and PYH rivers	Concentrations measured; water volume data collected from a hydrological station nearby
Runoff from uplands	Concentrations measured; water data collected from a hydrological station nearby
Groundwater	Concentrations measured; water data collected from a hydrological station nearby
Outputs to Dongfeng Reservoir	
Outflow	Concentrations measured; water volume data collected from a hydrological station nearby
Evasion of Hg^0 from water surface	Hg^0 evasion fluxes were calculated based on DGM measurement

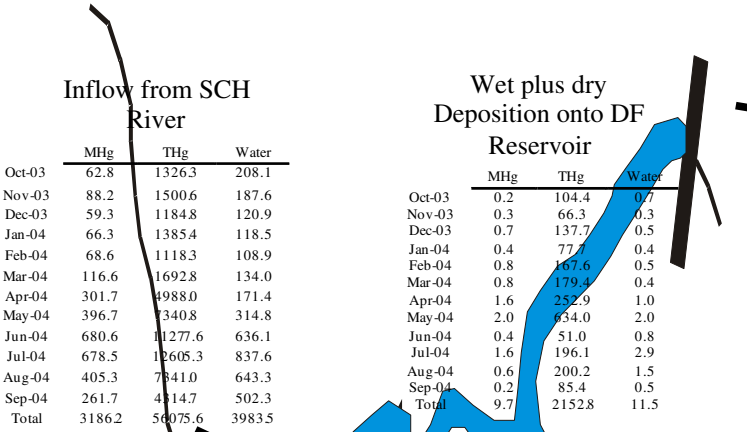
$$F = k_w(\text{DGM-TGM}/H') \quad (1)$$

where F is the evasion flux (ng m^{-2} h^{-1}), the term k_w, is the gas transfer velocity of Hg^0 in the water–air interface, TGM is the total gaseous Hg^0 level (ng m^{-3}) which we applied an average value of 2.9 ng m^{-3} in rainy season and an average value of 8.4 ng m^{-3} in dry season according to measurement conducted in Hong-Feng Reservoir area (Wang et al., 2005), H is the Henry's Law coefficient which is corrected for temperature (Sanemasa, 1975; Miles and Fink, 1998), and DGM is the dissolved gaseous mercury concentration in surface water (ng L^{-1}). The k_w values were estimated using Wanninkhof's approach (Wanninkhof, 1992),

$$k_w = 0.31u^2(S_c/600)^{-0.5} \quad (2)$$

where k_w is in cm h^{-1}, u is the wind speed in m s^{-1} (using the average wind speed recorded from meteorological station nearby), and S_c is the Schmidt number (=ν/D, where ν = kinematic viscosity, D = air molecular diffusitivity of Hg^0). The S_c number is corrected for temperature according to Lindberg and Zhang (2000) and Wängberg et al. (2001). We measured DGM concentrations in December 2003, April 2004 and July 2004 in surface water during daytime (Table 3). We assume that the December data are representative of winter months (December, January, and February), April data represent both spring and fall (March, April, May, September, October, and November), and July data represent summer (June, July, August). It is well documented that there is a diurnal pattern of DGM concentrations in surface water with DGM concentrations higher during daytime than at night (Feng et al., 2004b; O'Driscoll et al., 2003; Lindberg and Zhang, 2000). Therefore, our calculations of monthly Hg^0 emission fluxes according to DGM data measured during daytime may be the upper limit of Hg^0 emission flux. Nevertheless, Hg^0 evasion from water surfaces only constituted a minor portion of THg outputs as shown in Figs. 2 and 3.

The annual budgets were calculated from the monthly data from October 2003 to September 2004. A negative values for $Net_{(MeHg,\ THg,\ Water)}$ indicates that the reservoir was a net sink, while a positive value indicates the reservoir was a net source of MeHg, THg, or water.

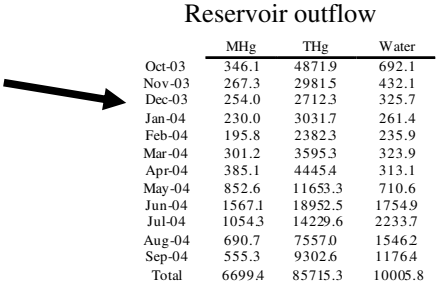


Inflow from SCH River

	MHg	THg	Water
Oct-03	62.8	1326.3	208.1
Nov-03	88.2	1500.6	187.6
Dec-03	59.3	1184.8	120.9
Jan-04	66.3	1385.4	118.5
Feb-04	68.6	1118.3	108.9
Mar-04	116.6	1692.8	134.0
Apr-04	301.7	4988.0	171.4
May-04	396.7	7340.8	314.8
Jun-04	680.6	11277.6	636.1
Jul-04	678.5	12605.3	837.6
Aug-04	405.3	7841.0	643.3
Sep-04	261.7	4514.7	502.3
Total	3186.2	56075.6	3983.5

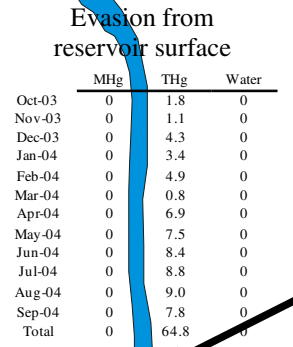
Wet plus dry Deposition onto DF Reservoir

	MHg	THg	Water
Oct-03	0.2	104.4	0.7
Nov-03	0.3	66.3	0.3
Dec-03	0.7	137.7	0.5
Jan-04	0.4	77.7	0.4
Feb-04	0.8	167.6	0.5
Mar-04	0.8	179.4	0.4
Apr-04	1.6	252.9	1.0
May-04	2.0	634.0	2.0
Jun-04	0.4	51.0	0.8
Jul-04	1.6	196.1	2.9
Aug-04	0.6	200.2	1.5
Sep-04	0.2	85.4	0.5
Total	9.7	2152.8	11.5



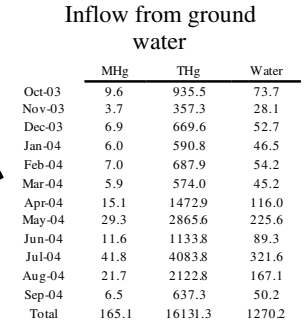
Reservoir outflow

	MHg	THg	Water
Oct-03	346.1	4871.9	692.1
Nov-03	267.3	2981.5	432.1
Dec-03	254.0	2712.3	325.7
Jan-04	230.0	3031.7	261.4
Feb-04	195.8	2382.3	235.9
Mar-04	301.2	3595.3	323.9
Apr-04	385.1	4445.4	313.1
May-04	852.6	11653.3	710.6
Jun-04	1567.1	18952.5	1754.9
Jul-04	1054.3	14229.6	2233.7
Aug-04	690.7	7557.0	1546.2
Sep-04	555.3	9302.6	1176.4
Total	6699.4	85715.3	10005.8



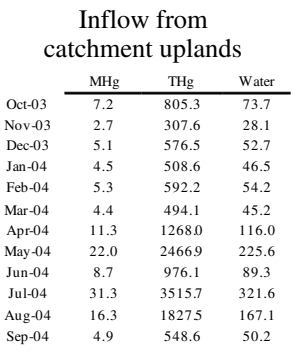
Evasion from reservoir surface

	MHg	THg	Water
Oct-03	0	1.8	0
Nov-03	0	1.1	0
Dec-03	0	4.3	0
Jan-04	0	3.4	0
Feb-04	0	4.9	0
Mar-04	0	0.8	0
Apr-04	0	6.9	0
May-04	0	7.5	0
Jun-04	0	8.4	0
Jul-04	0	8.8	0
Aug-04	0	9.0	0
Sep-04	0	7.8	0
Total	0	64.8	0



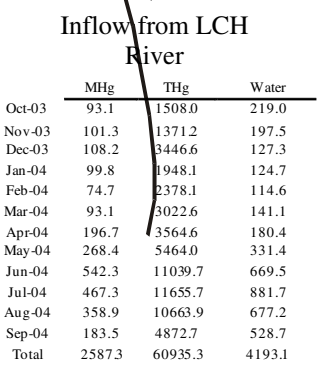
Inflow from ground water

	MHg	THg	Water
Oct-03	9.6	935.5	73.7
Nov-03	3.7	357.3	28.1
Dec-03	6.9	669.6	52.7
Jan-04	6.0	590.8	46.5
Feb-04	7.0	687.9	54.2
Mar-04	5.9	574.0	45.2
Apr-04	15.1	1472.9	116.0
May-04	29.3	2865.6	225.6
Jun-04	11.6	1133.8	89.3
Jul-04	41.8	4083.8	321.6
Aug-04	21.7	2122.8	167.1
Sep-04	6.5	637.3	50.2
Total	165.1	16131.3	1270.2



Inflow from catchment uplands

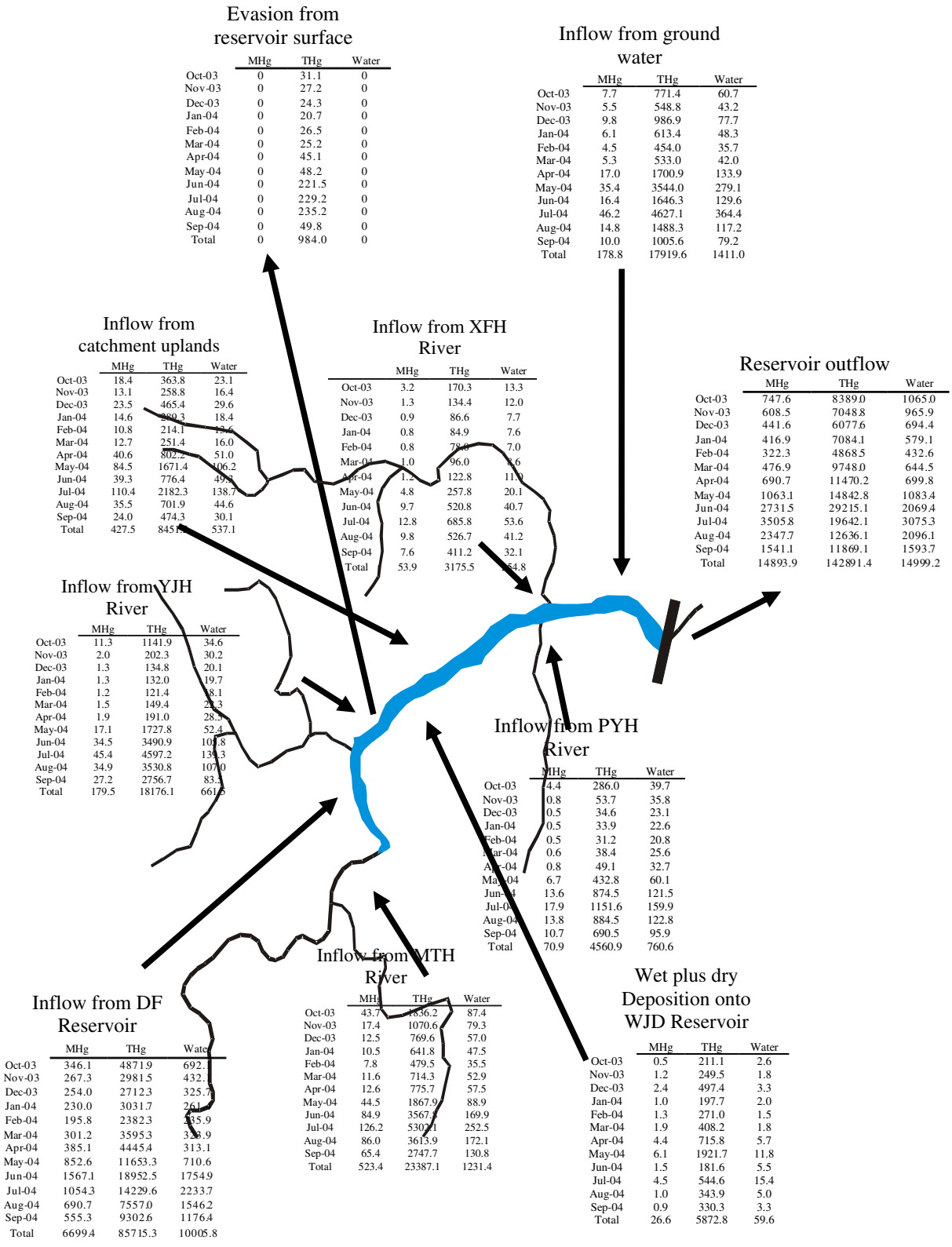
	MHg	THg	Water
Oct-03	7.2	805.3	73.7
Nov-03	2.7	307.6	28.1
Dec-03	5.1	576.5	52.7
Jan-04	4.5	508.6	46.5
Feb-04	5.3	592.2	54.2
Mar-04	4.4	494.1	45.2
Apr-04	11.3	1268.0	116.0
May-04	22.0	2466.9	225.6
Jun-04	8.7	976.1	89.3
Jul-04	31.3	3515.7	321.6
Aug-04	16.3	1827.5	167.1
Sep-04	4.9	548.6	50.2
Total	123.6	13887.1	1270.2



Inflow from LCH River

	MHg	THg	Water
Oct-03	93.1	1508.0	219.0
Nov-03	101.3	1371.2	197.5
Dec-03	108.2	3446.6	127.3
Jan-04	99.8	1948.1	124.7
Feb-04	74.7	2378.1	114.6
Mar-04	93.1	3022.6	141.1
Apr-04	196.7	3564.6	180.4
May-04	268.4	5464.0	331.4
Jun-04	542.3	11039.7	669.5
Jul-04	467.3	11655.7	881.7
Aug-04	358.9	10663.9	677.2
Sep-04	183.5	4872.7	528.7
Total	2587.3	60935.3	4193.1

Fig. 2. The monthly input and output of MeHg, THg and water to Dongfeng reservoir (MeHg and THg in g; water in 10^6 m^3).



Evasion from reservoir surface

	MHg	THg	Water
Oct-03	0	31.1	0
Nov-03	0	27.2	0
Dec-03	0	24.3	0
Jan-04	0	20.7	0
Feb-04	0	26.5	0
Mar-04	0	25.2	0
Apr-04	0	45.1	0
May-04	0	48.2	0
Jun-04	0	221.5	0
Jul-04	0	229.2	0
Aug-04	0	235.2	0
Sep-04	0	49.8	0
Total	0	984.0	0

Inflow from ground water

	MHg	THg	Water
Oct-03	7.7	771.4	60.7
Nov-03	5.5	548.8	43.2
Dec-03	9.8	986.9	77.7
Jan-04	6.1	613.4	48.3
Feb-04	4.5	454.0	35.7
Mar-04	5.3	533.0	42.0
Apr-04	17.0	1700.9	133.9
May-04	35.4	3544.0	279.1
Jun-04	16.4	1646.3	129.6
Jul-04	46.2	4627.1	364.4
Aug-04	14.8	1488.3	117.2
Sep-04	10.0	1005.6	79.2
Total	178.8	17919.6	1411.0

Inflow from catchment uplands

	MHg	THg	Water
Oct-03	18.4	363.8	23.1
Nov-03	13.1	258.8	16.4
Dec-03	23.5	465.4	29.6
Jan-04	14.6	289.3	18.4
Feb-04	10.8	214.1	13.6
Mar-04	12.7	251.4	16.0
Apr-04	40.6	802.2	51.0
May-04	84.5	1671.4	106.2
Jun-04	39.3	776.4	49.3
Jul-04	110.4	2182.3	138.7
Aug-04	35.5	701.9	44.6
Sep-04	24.0	474.3	30.1
Total	427.5	8451.6	537.1

Inflow from XFH River

	MHg	THg	Water
Oct-03	3.2	170.3	13.3
Nov-03	1.3	134.4	12.0
Dec-03	0.9	86.6	7.7
Jan-04	0.8	84.9	7.6
Feb-04	0.8	78.9	7.0
Mar-04	1.0	96.0	8.6
Apr-04	1.2	122.8	11.0
May-04	4.8	257.8	20.1
Jun-04	9.7	520.8	40.7
Jul-04	12.8	685.8	53.6
Aug-04	9.8	526.7	41.2
Sep-04	7.6	411.2	32.1
Total	53.9	3175.5	254.8

Reservoir outflow

	MHg	THg	Water
Oct-03	747.6	8389.0	1065.0
Nov-03	608.5	7048.8	965.9
Dec-03	441.6	6077.6	694.4
Jan-04	416.9	7084.1	579.1
Feb-04	322.3	4868.5	432.6
Mar-04	476.9	9748.0	644.5
Apr-04	690.7	11470.2	699.8
May-04	1063.1	14842.8	1083.4
Jun-04	2731.5	29215.1	2069.4
Jul-04	3505.8	19642.1	3075.3
Aug-04	2347.7	12636.1	2096.1
Sep-04	1541.1	11869.1	1593.7
Total	14893.9	142891.4	14999.2

Inflow from YJH River

	MHg	THg	Water
Oct-03	11.3	1141.9	34.6
Nov-03	2.0	202.3	30.2
Dec-03	1.3	134.8	20.1
Jan-04	1.3	132.0	19.7
Feb-04	1.2	121.4	18.1
Mar-04	1.5	149.4	22.3
Apr-04	1.9	191.0	28.5
May-04	17.1	1727.8	52.4
Jun-04	34.5	3490.9	103.8
Jul-04	45.4	4597.2	136.3
Aug-04	34.9	3530.8	107.0
Sep-04	27.2	2756.7	83.5
Total	179.5	18176.1	661.5

Inflow from PYH River

	MHg	THg	Water
Oct-03	4.4	286.0	39.7
Nov-03	0.8	53.7	35.8
Dec-03	0.5	34.6	23.1
Jan-04	0.5	33.9	22.6
Feb-04	0.5	31.2	20.8
Mar-04	0.6	38.4	25.6
Apr-04	0.8	49.1	32.7
May-04	6.7	432.8	60.1
Jun-04	13.6	874.5	121.5
Jul-04	17.9	1151.6	159.9
Aug-04	13.8	884.5	122.8
Sep-04	10.7	690.5	95.9
Total	70.9	4560.9	760.6

Inflow from MTH River

	MHg	THg	Water
Oct-03	43.7	1336.2	87.4
Nov-03	17.4	1070.6	79.3
Dec-03	12.5	769.6	57.0
Jan-04	10.5	641.8	47.5
Feb-04	7.8	479.5	35.5
Mar-04	11.6	714.3	52.9
Apr-04	12.6	775.7	57.5
May-04	44.5	1867.9	88.9
Jun-04	84.9	3567.8	169.9
Jul-04	126.2	5302.1	252.5
Aug-04	86.0	3613.9	172.1
Sep-04	65.4	2747.7	130.8
Total	523.4	23387.1	1231.4

Inflow from DF Reservoir

	MHg	THg	Water
Oct-03	346.1	4871.9	692.1
Nov-03	267.3	2981.5	432.1
Dec-03	254.0	2712.3	325.7
Jan-04	230.0	3031.7	261.6
Feb-04	195.8	2382.3	235.9
Mar-04	301.2	3595.3	338.9
Apr-04	385.1	4445.4	313.1
May-04	852.6	11653.3	710.6
Jun-04	1567.1	18952.5	1754.9
Jul-04	1054.3	14229.6	2233.7
Aug-04	690.7	7557.0	1546.2
Sep-04	555.3	9302.6	1176.4
Total	6699.4	85715.3	10005.8

Wet plus dry Deposition onto WJD Reservoir

	MHg	THg	Water
Oct-03	0.5	211.1	2.6
Nov-03	1.2	249.5	1.8
Dec-03	2.4	497.4	3.3
Jan-04	1.0	197.7	2.0
Feb-04	1.3	271.0	1.5
Mar-04	1.9	408.2	1.8
Apr-04	4.4	715.8	5.7
May-04	6.1	1921.7	11.8
Jun-04	1.5	181.6	5.5
Jul-04	4.5	544.6	15.4
Aug-04	1.0	343.9	5.0
Sep-04	0.9	330.3	3.3
Total	26.6	5872.8	59.6

Fig. 3. The monthly input and output of MeHg, THg and water to Wujiangdu reservoir (MeHg and THg in g; water in 10^6 m^3).

Table 3
Dissolved gaseous mercury concentrations in surface water of Wujiangdu and Dongfeng Reservoirs (ng L^{-1}).

Sampling time	December, 2003	April, 2004	July, 2004
WJD	0.105	0.094	0.372
DF	0.071	0.043	0.045

Table 4
Annual percentage input from each source and annual total inputs to Dongfeng Reservoir.

		Average annual % of total inputs from each source				
	Annual total input	Precipitation	SCH	LCH	Groundwater	Direct runoff
THg	149 000 g	1.4	37.6	40.8	10.9	9.3
MeHg	6070 g	0.2	52.5	42.6	2.7	2.0
Water	10 300 × 10^6 m^3	0.2	38.8	40.8	12.4	7.9

Table 5
Annual percentage input from each source and annual total inputs to Wujiangdu Reservoir.

		Average annual % of total inputs from each source							
	Annual total input	Precipitation	DFR	MTH	YJH	XFH	PYH	Ground water	Direct runoff
THg	167 000 g	3.5	51.2	14.0	10.9	1.9	2.7	10.7	5.1
MeHg	8160 g	0.3	82.1	6.4	2.2	0.7	0.9	2.2	5.2
Water	14 900 × 10^6 m^3	0.4	67.3	8.3	4.4	1.7	5.1	9.5	3.3

3. Results and discussion

3.1. Hydrology of the reservoirs

The largest annual inputs of water to DF Reservoir came from SCH and LCH rivers which constituted 38.8% and 40.8% of the total inputs, respectively (Table 4, Fig. 2). Groundwater and direct upland runoff contributed 12.4% and 7.9% of the total inputs of water to DF Reservoir, respectively (Table 4, Fig. 2). Wet deposition onto the DF Reservoir accounted for 0.2% of the total inputs of water to the reservoir (Table 4, Fig. 2). The largest annual input of water (67.3%) to WJD Reservoir came from the outlet of DF Reservoir (Table 5, Fig. 3). MTH, YJH, XFH, and PYH rivers supplied 8.3%, 4.4%, 1.7% and 5.1% of the total inputs of water to the reservoir (Table 5, Fig. 3). Groundwater and direct upland runoff contributed 9.5% and 3.3% of the total inputs of water to WJD Reservoir, respectively (Table 5, Fig. 3). Wet deposition onto the WJD Reservoir accounted for only 0.4% of the total inputs of water to the reservoir (Table 5, Fig. 3)

Water outputs and inputs were well balanced annually for both reservoirs, but there are large variations of monthly water yields (Tables 6 and 7). Generally reservoirs were net sinks for water in the rainy season, while they were net sources in the dry season (Tables 6 and 7).

3.2. THg concentrations and input–output budgets

Concentrations of THg in the precipitation samples collected in a one year period varied from 17.9 to 124.3 ng L^{-1} with an overall volume-weighted average of 51.8 ng L^{-1} ($n = 18$). It is apparent that THg concentrations in precipitation samples collected in our study area were much elevated compared to those reported in relatively pristine areas in North America and Europe, which were generally lower than 10 ng L^{-1} (e.g. St Louis et al., 2004; Lindqvist et al., 1991; Hall et al., 2005a,b; Steding and Flegal, 2002). The elevation of THg in precipitation is attributed to the contamination of Hg in the regional ambient air due to both anthropogenic and natural Hg emissions. Even though THg concentrations in precipitation samples are elevated, annual inputs of THg from wet and dry depositions accounted for only 1.4% of the THg inputs to DF Reservoir, and 3.4% to WJD Reservoir, respectively (Table 4 and Fig. 2).

Due to the special geological background and anthropogenic Hg emissions in the study area, the THg concentrations in surface water were significantly elevated. Therefore, inputs from rivers contributed most of the Hg to both reservoirs. SCH and LCH rivers provided 37.6% and 40.8% of the THg entering the reservoir, respectively (Table 4 and Fig. 2). Groundwater and direct runoff only provided 10.9% and 9.3% of the THg inputs, respectively. Outflow from DF Reservoir contributed most of the hydrological inputs to WJD Reservoir, and meanwhile it was also the largest contributor (51.2%) of the total inputs of THg (Table 5 and Fig. 3). MTH, YJH, XFH and PYH rivers provided 14.0%, 10.9%, 1.9%, and 2.7% of the total inputs of THg to the reservoir, respectively (Table 5 and Fig. 3). Groundwater and direct runoff supplied 10.7% and 5.1% of the total inputs of THg (Table 5 and Fig. 3).

Both reservoirs were net sinks for THg on an annual scale, absorbing 3319.5 g km^{-2}, or 42.5% of the total inputs for DF Reservoir, and 489.2 g km^{-2}, or 14.0% of inputs for WJD Reservoir, respectively (Tables 6 and 7). It is clearly seen from Fig. 4 that THg concentrations in SCH and LCH river water, which are the major inflows to the reservoir were generally higher that those in the

Table 6
Net yields of MeHg, THg, IHg, and the net entrapment of water from Dongfeng Reservoir.

Month	MeHg yield		THg yield		IHg yield		Water yield	
	g km^{-2}[a]	% of input	g km^{-2}	% of input	g km^{-2}	% of input	10^6 m^3 km^{-2}	% of input
Oct-04	9.1	100.2	10.2	4.1	1.1	0.5	7.52	26.1
Nov-04	3.7	36.3	−32.5	17.2	−36.2	20.3	0.02	0.1
Dec-04	3.9	41.1	−172.7	54.8	−176.6	57.8	−0.50	2.9
Jan-05	2.8	29.9	−77.3	32.7	−80.0	35.3	−3.1	18.3
Feb-05	2.1	25.1	−133.9	51.7	−135.9	54.2	−4.0	24.7
Mar-05	4.2	36.4	−123.9	39.7	−128.1	42.6	−1.4	7.4
Apr-05	−7.4	26.8	−371.4	61.4	−364.0	63.1	−12.1	42.4
May-05	7.0	18.7	−372.3	37.9	−379.3	40.1	−16.2	30.3
Jun-05	16.9	26.0	−288.9	22.5	−305.8	25.1	15.8	20.8
Jul-05	−8.7	13.6	−932.9	55.6	−924.2	57.2	−0.9	0.8
Aug-05	−5.9	14.0	−763.8	65.8	−758.0	67.8	−2.6	3.2
Sep-05	5.2	21.6	−60.1	11.0	−65.3	12.5	3.3	5.6
Total	32.9	10.3	−3319.5	42.5	−3352.3	44.7	−14.2	2.6

[a] km^{-2} refers to the surface area of reservoir.

Table 7
Net yields of MeHg, THg, and the net entrapment of water from the Wujiangdu Reservoir.

Month	MeHg yield		THg yield		IHg yield		Water yield	
	g km^{-2}[a]	% of input	g km^{-2}	% of input	g km^{-2}	% of input	10^6 m^3 km^{-2}	% of input
Oct-04	6.5	71.8	−25.8	12.8	−23.2	12.0	2.4	11.9
Nov-04	6.3	97.1	33.0	28.7	33.2	30.5	6.6	48.7
Dec-04	2.9	44.8	8.7	7.3	12.2	10.8	3.2	28.2
Jan-05	3.2	57.4	43.5	41.4	45.9	46.1	3.2	36.0
Feb-05	2.1	44.7	18.1	21.4	20.6	25.9	1.4	17.9
Mar-05	3.0	42.0	83.4	68.9	87.5	76.7	3.2	31.1
Apr-05	4.8	49.0	56.7	30.8	61.7	35.4	1.5	11.3
May-05	0.2	1.1	−171.2	35.5	−149.5	32.4	−5.0	17.9
Jun-05	20.2	54.6	−12.0	1.9	4.8	0.8	−6.4	12.8
Jul-05	43.7	147.3	−281.4	40.4	−295.4	44.3	−5.7	8.1
Aug-05	30.6	164.8	−120.8	31.0	−132.9	35.8	−1.2	2.6
Sep-05	17.6	119.8	−121.3	32.7	−124.2	34.9	−0.7	2.2
Total	140.9	82.5	−489.2	14.0	−459.4	13.8	2.6	0.8

[a] km^{-2} refers to the surface area of reservoir.

outflow water of DF and WJD Reservoirs. In Fig. 4, the THg concentrations vary from around 6 to max 30 ng L^{-1}, hence in the very low range of the ranges of THg concentrations in Wujiang river (Jiang et al., 2004). This is simply because a few new reservoirs were built on the upper reach of WJD and DF Reservoirs as shown in Fig. 1, which trapped mercury in these reservoirs and resulted in the decrease of THg concentrations in river water compared to the river in the upper reach of Wujiang River.

However, the monthly THg yields varied significantly for both reservoirs. For DF Reservoir, it was a net source for THg in October 2003, mainly because a 26.1% greater volume of water flowed out the reservoir than entered, whereas during the remainder of the year, the reservoir was a net sink for THg (Table 6). A large portion of the increased THg export in October 2003 was actually MeHg (see below, Table 6), suggesting that methylation processes in the reservoir changed the chemical speciation and bioavailability of the incoming Hg. The reservoir was a net source of THg from November 2003 to April 2004 for WJD Reservoir, when a greater volume of water flowed out of the reservoir than entered (Table 7). For the remainder of the year, the reservoir was a net sink of THg.

PHg constituted a large portion of THg in surface water as shown in Table 8. Especially during rainy season, PHg was more than 50% of THg in the LCH and SCH rivers, and the percentages of THg as PHg in water samples from these rivers, which are the major hydrological inputs to the reservoir, were generally higher than those of outflows from DF and WJD Reservoirs, especially during the rainy season (Table 8). This demonstrated that the sedimentation of particulate matter in reservoir is one of the most important removal mechanisms of THg from the reservoir's waters.

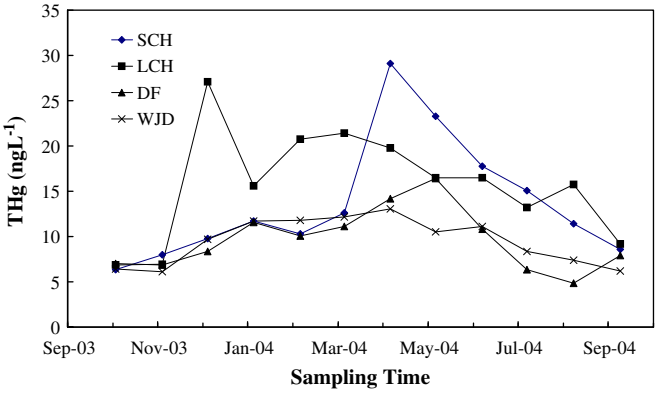


Fig. 4. The distribution of THg concentrations in river water of LCH and SCH, and water in reservoir of DF and WJD.

A major water input to WJD Reservoirs is from the outflow of DF Reservoir. DF Reservoir in effect acted as a filter of large particulates upstream from WJD, so that WJD waterborne particulates were in a much smaller size distribution than DF. This will result in a lower sedimentation rate in WJD Reservoir. Therefore, we observed that THg yield in WJD Reservoir was much less than that in DF Reservoir (Tables 6 and 7).

Both reservoirs were also net sinks of inorganic Hg (IHg; or THg minus MeHg) annually (Tables 6 and 7). Since constituting the majority of THg in water samples, IHg almost behaved the same way in both reservoirs as THg (Tables 6 and 7).

3.3. MeHg concentrations and input–output budgets

The volume-weighted average concentration of MeHg in wet deposition was 0.43 ng L^{-1} (n = 16). Annual wet deposition of MeHg onto DF and WJD Reservoirs was 9.7 and 26.6 g km^{-2}, respectively (Figs. 2 and 3), and contributed 0.2% and 0.3% of the total MeHg inputs to DF and WJD Reservoirs, respectively (Tables 4 and 5).

Concentrations of MeHg in direct runoff to the reservoirs were somewhat higher than precipitation (0.87 ng L^{-1}, n = 2), and annual MeHg inputs from this source accounted for 2.0% of the total MeHg inputs to DF Reservoir (Table 4), and 5.2% of total MeHg inputs to WJD Reservoir, respectively (Table 5). Concentrations of MeHg in groundwater were relatively low (0.13 ng L^{-1}, n = 2). This source only provided annually 2.7% and 2.2% of total MeHg to DF and WJD Reservoirs, respectively (Tables 4 and 5).

Inputs from SCH and LCH rivers had much higher MeHg concentrations than other inputs to DF Reservoir (0.3–1.76 ng L^{-1} for SCH river and 0.35–1.09 ng L^{-1} for LCH river, Fig. 5). SCH and LCH rivers together annually contributed 95.1% of total MeHg inputs to DF Reservoir (Table 4, Fig. 2). We note that even though SCH river provided a little less total water inputs to DF Reservoir than LCH, it contributed 9.9% more MeHg inputs to the reservoir than LCH river (Table 4, Fig. 2). We can see from Fig. 1 that two reservoirs (Pu-Ding and Yin-Zi-Du) are located on the upper stream of DF Reservoir on SCH river. This suggests that MeHg production and transportation from these two reservoirs contributed to the elevated MeHg inputs from SCH river to DF Reservoir.

Due to high MeHg concentrations in the outflow water of DF Reservoir (0.45–1.23 ng L^{-1}, Fig. 5), it provided 82.1% of the total MeHg inputs to WJD Reservoir (Table 5, Fig. 3). MeHg

Table 8
Percentage of THg in water presented as particulate Hg (PHg).

	SCH		LCH		DF-X		WJD-X	
	THg (ng L^{-1})	PHg/THg %	THg (ng L^{-1})	PHg/THg %	THg (ng L^{-1})	PHg/THg %	THg (ng L^{-1})	PHg/THg %
Oct-03	6.4	7.8	6.9	8.8	7.0	18.7	7.9	17.4
Nov-03	8.0	28.3	6.9	14.4	6.9	18.2	7.3	28.0
Dec-03	9.8	21.3	27.1	75.3	8.3	13.1	8.8	15.4
Jan-04	11.7	20.6	15.6	32.1	11.6	19.4	12.2	30.9
Feb-04	10.3	11.4	20.7	50.9	10.1	8.3	11.3	12.1
Mar-04	12.6	21.4	21.4	51.0	11.1	18.6	15.1	37.7
Apr-04	29.1	78.5	19.8	59.8	14.2	41.5	16.4	36.4
May-04	23.3	73.1	16.5	55.8	16.4	53.7	13.7	41.6
Jun-04	17.7	63.2	16.5	55.8	10.8	28.7	14.1	42.8
Jul-04	15.0	60.1	13.2	51.9	6.4	40.4	6.4	56.2
Aug-04	11.4	36.1	15.7	69.9	4.9	14.4	6.0	15.1
Sep-04	8.6	23.2	9.2	43.6	7.9	8.1	7.4	25.0
Average	13.7	37.1	15.8	47.4	9.6	23.6	10.6	29.9

concentrations in MTH (0.36 ng L^{-1}, $n = 2$), YJH (0.2 ng L^{-1}, $n = 2$), XFH (0.16 ng L^{-1}, $n = 2$) and PYH (0.07 ng L^{-1}, $n = 2$) were relatively low, and supplied 6.4%, 2.2%, 0.7%, and 0.9% of the total MeHg inputs to WJD Reservoir, respectively (Table 5, Fig. 3).

The average MeHg concentrations in the outflows of DF (0.77 ng L^{-1}) and WJD (0.89 ng L^{-1}) reservoirs were higher than the average MeHg concentrations in all inputs to the corresponding reservoirs. The percentage of THg that is MeHg (%MeHg) is a good relative indicator of MeHg production rates in ecosystems (St Louis et al., 1994, 2004; Rudd, 1995; Gilmour et al., 1998). The %MeHg of outflow from WJD Reservoir was generally higher than from DF Reservoir, which in turn was higher than in the inflows (SCH and LCH) to DF Reservoir (Fig. 6). This study shows that building a series of reservoirs in line along a river changes the riverine system into a natural Hg methylation factory which markedly increases the %MeHg in the downstream reservoirs; in effect magnifying the MeHg buildup problem in reservoirs. A peak in %MeHg in outflow of WJD Reservoir, which followed the summer temperature peak was observed (Fig. 7). This pattern of elevated %MeHg during the summer period was not observed in DF Reservoir.

Overall, both reservoirs were net sources of MeHg to the downstream ecosystems (Tables 6 and 7). DF Reservoir provided 32.9 g MeHg km^{-2} yr^{-1}, yielding 10.3% of the amount of MeHg that entered the reservoir, and WJD Reservoir provided 140.9 g MeHg km^{-2} yr^{-1}, yielding 82.5% of MeHg inputs (Tables 6 and 7). The maximum MeHg yields occurred in summer (July, August and September) in WJD Reservoir (Table 7), which agreed with the pattern of elevated %MeHg during the summer period in WJD Reservoir. Such a seasonal MeHg yield pattern was not observed in DF reservoir (Table 6). Moreover, DF Reservoir was a net sink of MeHg in April, July and August (Table 6). We found that MeHg concentrations in both SCH and LCH rivers were quite high, and particulate MeHg was the predominate species (65.1–85.2% of MeHg in SCH river and 81.9–89.8% in LCH river presented as particulate MeHg, Table 9) from April to August 2004. However, the percentages of MeHg that was in particulate form were significantly lower in the outflow of DF and WJD Reservoirs (27.0–51.7% in outflow of DF, and 37.2–69.5% in outflow of WJD) at the same period of time (Table 9). This implied that a large quantity of MeHg went to

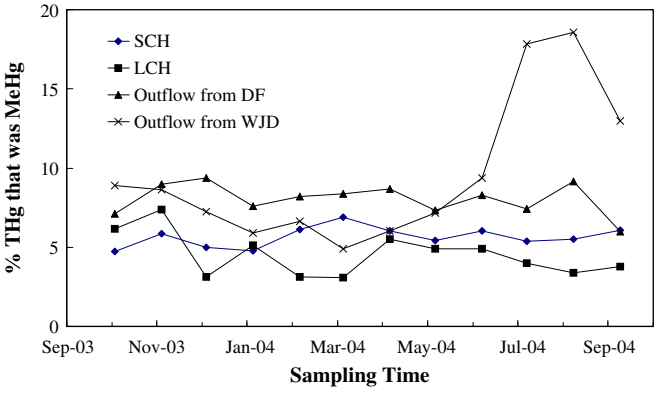


Fig. 6. The %MeHg in river water of LCH and SCH as well as the outflow water from DF and WJD Reservoirs.

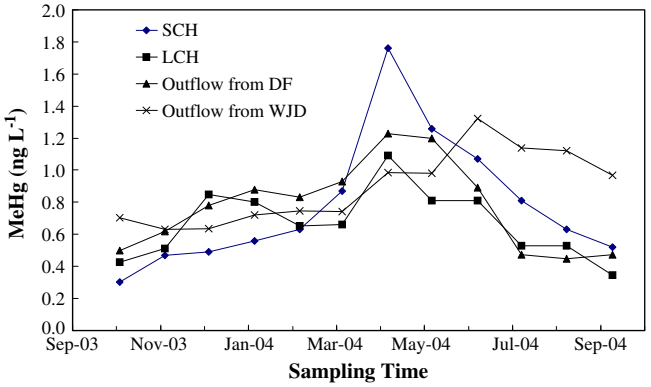


Fig. 5. The distribution of MeHg concentrations in river water of LCH and SCH as well as the outflow water from DF and WJD Reservoirs.

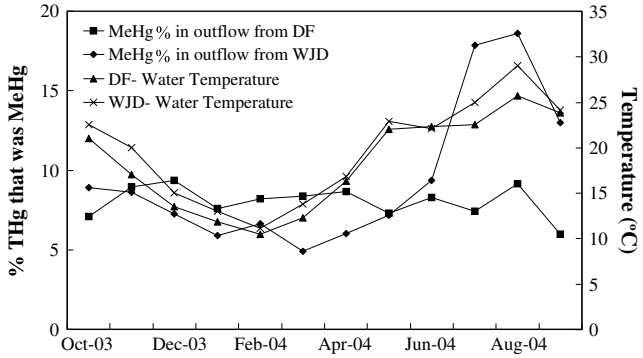


Fig. 7. The temporal variation of water temperature and the %MeHg in water of the outflow from DF and WJD reservoirs.

Table 9
Percentage of MeHg in water presented as particulate MeHg (PMeHg).

	SCH		LCH		Outflow from DF		Outflow WJD	
	MeHg (ng L^{-1})	PMeHg/MeHg (%)	MeHg (ng L^{-1})	PMeHg/MeHg (%)	MeHg (ng L^{-1})	PMeHg/MeHg (%)	MeHg (ng L^{-1})	PMeHg/MeHg (%)
Oct-03	0.30	15.2	0.43	12.5	0.50	3.8	0.70	25.2
Nov-03	0.47	1.1	0.51	2.5	0.62	10.5	0.63	12.1
Dec-03	0.49	7.1	0.85	83.5	0.78	14.5	0.64	16.9
Jan-04	0.56	6.6	0.80	89.6	0.88	22.7	0.72	10.1
Feb-04	0.63	6.3	0.65	57.1	0.83	15.7	0.75	24.4
Mar-04	0.87	39.0	0.66	46.5	0.93	29.5	0.74	21.6
Apr-04	**1.76**	**80.3**	***1.09***	***89.8***	***1.23***	***45.0***	***0.99***	***37.2***
May-04	**1.26**	**85.2**	***0.81***	***86.5***	***1.20***	***47.5***	***0.98***	***53.0***
Jun-04	**1.07**	**82.4**	***0.81***	***86.5***	***0.89***	***40.5***	***1.32***	***67.7***
Jul-04	**0.81**	**69.6**	***0.53***	***83.2***	***0.47***	***51.7***	***1.14***	***69.5***
Aug-04	**0.63**	**65.1**	***0.53***	***81.9***	***0.45***	***27.0***	***1.12***	***63.2***
Sep-04	0.52	65.5	0.35	5.5	0.47	31.0	0.97	56.1
Average	0.78	43.6	0.67	60.4	0.77	28.3	0.89	38.1

sediment during sedimentation process in DF Reservoir during the period from April to August 2004. As discussed previously, maximum THg entrapments were observed in DF Reservoir at the same period of time, due mainly to the sedimentation process (Table 6).

Previous studies demonstrated that the net Hg methylation rates in reservoir systems decreased with the increase of the ages of the reservoirs (e.g. St Louis et al., 2004; Lucotte et al., 1999; Hall et al., 2005a,b). Approximately 3 weeks after flooding, MeHg concentrations in the reservoir jumped 7-fold and reached 3 ng L^{-1}, which constituted about 80% of THg (St Louis et al., 2004). MeHg concentrations in the open water region of the reservoir decreased after the first 2 years of flooding to on average between 0.46 and 0.65 ng L^{-1} annually (St Louis et al., 2004). WJD Reservoir was constructed in 1979 (Table 1), and the net methylation rates in the reservoir were still quite high even at the age of 24 years old, further demonstrating that MeHg contamination problem could last for more than 24 years for certain large reservoirs. On the other hand, DF Reservoir was dammed in 1992, and it was much younger than WJD Reservoir. However, the net annual Hg methylation rate was significantly lower in DF Reservoir, implying that factors other than the age of reservoir actually also affected Hg methylation rate.

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 for 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. DF and WJD are adjacent reservoirs, and the geological background, soil type (including organic carbon and Hg concentrations in flooded soil), climate conditions of both reservoirs are almost identical. Moreover, the ratios of flooded area (the surface area of the reservoir) and water volume of the reservoir are almost the same for both reservoirs (53.7×10^6 m^3 km^{-2} for DF, and 48.1×10^6 m^3 km^{-2} for WJD, Table 1). The major difference between DF and WJD Reservoirs is probably the average water residence time (Table 1). Our results implied that a longer water residence time will increase the net MeHg production rate in the reservoir. The biogeochemical processes of Hg in both reservoirs will be evaluated in a companion paper (Feng et al., in press), which supported this conclusion.

Our study demonstrated that reservoirs are an important MeHg source to the downstream ecosystem. As the implementation of the "Go West" policy in China continues, more reservoirs are going to be built on Wujiang River for electricity generation purposes. By 2010, 12 large reservoirs in total will have been created in Wujiang River. The construction of reservoirs will not only produce a MeHg contamination risk to the reservoir system, which will eventually contaminate the fish through the aquatic food chains, but also seriously contaminate the downstream aquatic ecosystems. Wujiang River is an important upper branch of Three Gorge Dam, and it will be a large MeHg input source to the big dam when it is constructed by 2112.

4. Conclusions

In this study, we conducted a detailed study on the mass balance of THg and MeHg of Dongfeng (DF) and Wujiangdu (WJD) reservoirs, which were constructed in 1992 and 1979, respectively. Taken as a whole, our data indicate that:

- Both reservoirs were net sinks for THg on an annual scale, yielding −3319.5 g km^{-2}, or 42.5% of inputs for DF Reservoir, and −489.2 g km^{-2}, or 14.0% of inputs for WJD Reservoirs, respectively,
- Both reservoirs were net sources of MeHg to the downstream ecosystems. DF Reservoir provided a source of 32.9 g MeHg km^{-2} yr^{-1}, yielding 10.3% of the amount of MeHg that entered the reservoir, and WJD Reservoir provided 140.9 g MeHg km^{-2} yr^{-1}, yielding 82.5% of MeHg inputs,
- The maximum MeHg yields occurred in summer (July, August and September) in WJD Reservoir, which agreed with the pattern of elevated %MeHg during the summer period in WJD Reservoir. Such a seasonal MeHg yield pattern was not observed in DF Reservoir,
- Our results implied that factors other than the age of reservoir actually affected Hg methylation rate and that a longer water residence time will increase the net MeHg production rate in the reservoirs.

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