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Mercury speciation and spatial distribution in surface waters of the Yarlung Zangbo River, Tibet

ZHENG Wei[1,2], KANG ShiChang[1,3*], FENG XinBin[4], ZHANG QiangGong[1] & LI ChaoLiu[1]

[1] *Laboratory of Tibetan Environment Changes and Land Surface Processes, Institute of Tibetan Plateau Research, Chinese Academy of Sciences, Beijing 100085, China;*
[2] *Academy for Environmental Planning, Ministry of Environmental Protection, Beijing 100012, China;*
[3] *The State Key Laboratory of Cryospheric Sciences, Lanzhou 730000, China;*
[4] *The State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, China*

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The Yarlung Zangbo River is the highest river in the world. It flows from west to east through the southern part of Tibet. The mercury (Hg) speciation and distribution in surface waters and soils near the bank of the Yarlung Zangbo River and its two tributaries, the Lhasa and Niyang Rivers, were investigated in June 2007. Simultaneously, major water quality parameters were also measured at the same stations. Total Hg (THg) and total methylmercury (TMeHg) concentrations in surface waters of the Yarlung Zangbo River ranged from 1.46 to 4.99 ng/L and from 0.06 to 0.29 ng/L, respectively, representing the background levels in river systems of the Tibetan Plateau. Particulate Hg (PHg) accounted for 69% of the THg, and the two Hg species had a significant relationship (r=0.990, P<0.01). Approximately 61% of the spatial distribution of THg was controlled by the concentrations of total suspended particles (TSP). Reactive Hg (RHg) concentrations ranged from 0.10 to 0.36 ng/L, and this fraction may play a weak role in terms of the transport and fate of Hg in surface waters. Dissolved methylmercury (DMeHg) constituted 71% of the TMeHg and was significantly correlated with TMeHg (r=0.746, P<0.01). The spatial distribution of TMeHg is not strongly affected by environmental factors such as THg, RHg, temperature, pH, dissolved organic carbon (DOC), and TSP. In addition, the inflow of both the Lhasa and Niyang Rivers probably influences the concentrations of THg in surface waters of the mainstream, but such an effect is not obvious for TMeHg.

Yarlung Zangbo River, mercury, speciation, spatial distribution, Tibet

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Mercury (Hg) is a toxic and nonessential heavy metallic element and is considered to be a global pollutant [1,2]. The global biogeochemical cycle of Hg has drawn a great attention in the domain of environmental science. Understanding the biogeochemical cycle of Hg in aquatic environments is significant in application and theory, because the consumption of aquatic product, especially fish, is a major route through which humans are exposed to Hg, and the processes of transport, transformation, fate and bioavailability of Hg in aquatic systems are extremely complex. It not only has been one of the focuses of global investigation for a long term, but also is a challenge scientists must face in future studies. River systems are readily accessible for utilization of human and are susceptible to anthropogenic activities. Therefore, riverine Hg biogeochemistry is an important issue in studying the biogeochemical cycle of Hg in aquatic environments.

The Tibetan Plateau is a sensitive region that is driving global environment changes; therefore, it has received a great deal of attention [3,4]. As part of the Tibetan Plateau, Tibet has the largest number of rivers in China. However, relatively few investigations have focused on Hg cycling in

*Corresponding author (email: shichang.kang@itpcas.ac.cn)

river systems of Tibet due to limitations of geographical conditions and analytical techniques. On the other hand, the emissions of Hg in Asia are increasing [5], which may have a significant effect on riverine Hg biogeochemistry in Tibet. The Yarlung Zangbo River (hereafter referred to as the YZ River) is the largest river in Tibet and serves as Tibetan main freshwater source and water vapor channel. Moreover, it is also an important international river throughout China and other countries in South Asia. Therefore, it is necessary to characterize the speciation and spatial distribution of Hg in surface waters of the YZ River and to identify their controlling factors. The results presented here provide a scientific basis for the effective protection and reasonable utilization of the YZ River Basin, new knowledge for understanding the behavior of Hg in aquatic environments from China, and basic data for the future researches on the global biogeochemical cycle of Hg.

1 Study area

The YZ River is the highest large volume river in the world with an average elevation of over 4000 m. It originates from the Gyaimanezong Glacier in the northern foothills of the Himalayas, flows from west to east through the southern part of Tibet, and has a total length of 2057 km and a drainage area of 239228 km^2. Based on the hydrology and valley terrain characteristics, the YZ River can be divided into three segments, the upper reaches (from the source to Litse), middle reaches (from Litse to Pai), and lower reaches (from Pai to Baxika). The YZ River has many tributaries, among which there are the Dogxung Tsangpo, Nianchu River, Lhasa River, and Parlung Zangpo with a drainage area of over 10000 km^2. The Lhasa River, located on the left bank of the middle reaches of the YZ River and draining into the mainstream at Qushui, has the largest drainage area among the five major tributaries. The amount of water in the Niyang River, located on the left bank of the middle-lower reaches of the YZ River and draining into the mainstream at Luding, is only less than that in the Parlung Zangpo in all of the tributaries. Climate regimes in the YZ River Basin vary in different segments due to their geographic location and topography. The climate pattern in the upper, middle, and lower reaches is temperate grassland, temperate forest steppe, and humid temperate climate, respectively. The annual flow flux of the YZ River is dominated by precipitation, with its high flow period covering from June to September (maximum flow occurring in July or August) and low flow period covering from November to next April [6].

2 Materials and methods

2.1 Sample collection

(i) Surface waters. Surface water samples (20 cm beneath the surface) were collected along the upper and middle reaches of the YZ River (Sites 1 to 17) and from its two tributaries (the Lhasa and Niyang Rivers) from 15 to 29 June, 2007 (Figure 1). Surface water samples were collected against the water current to avoid disturbing the water and bottom sediments. Unfiltered and filtered water samples for Hg species analysis [7–9] were filled in pre-cleaned [10] 100-mL borosilicate glass bottles and acidified with 0.4% (*v*/*v*) HCl (MOS grade) on site. The filtered samples were collected by passing the water through filters (0.45-μm pore size, 47-mm diameter, Millipore) within 24 hours of sampling. In addition, water samples for dissolved organic carbon (DOC) and total suspended particle (TSP) analysis were taken using 30-mL brown glass bottles and 1500-mL polyethylene bottles, respectively.

(ii) Surface soils. While sampling water samples, surface soil samples (0–10 cm) near the river bank were collected using a random sampling method and sealed in polyethylene bags.

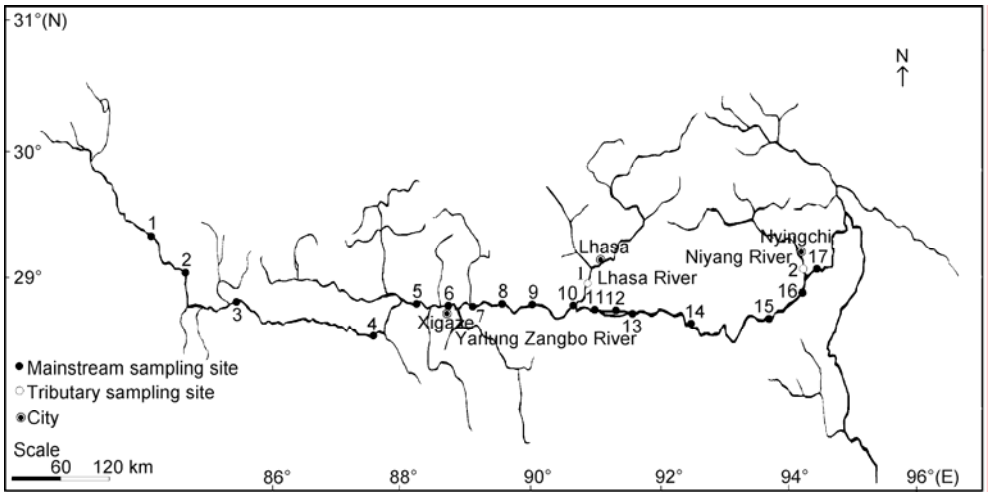

Figure 1 Location of water sampling sites along the YZ River Basin.

2.2 Sample analysis

(i) Water quality parameters. Temperature (T) and pH were measured using a pocket meter (HANNA, Italy) on site. DOC and TSP were analyzed by the high temperature catalytic oxidation [11] and gravimetric method, respectively.

(ii) Hg speciation in water samples. All Hg species were measured by cold vapor atomic fluorescence spectrometry (CVAFS) (Tekran 2500, Canada). Total Hg (THg) and dissolved Hg (DHg) were determined following the analytical procedure of BrCl oxidation-$SnCl_2$ reduction-trap preconcentrating-CVAFS [7,9]. Particulate Hg (PHg) was determined using the following equation: PHg=THg–DHg. Reactive Hg (RHg) was determined following the analytical procedure of $SnCl_2$ reduction-trap preconcentrating-CVAFS [9]. Total methylmercury (TMeHg) and dissolved methylmercury (DMeHg) were determined using the standard distillation-ethylation-GC separation-CVAFS technique [8,12]. Particulate methylmercury (PMeHg) was determined using the following equation: PMeHg=TMeHg–DMeHg.

Quality control for the determination of THg was conducted with replicate samples and field blanks, which were collected at 20% and 15% of the sampling sites, respectively. The coefficient of variation for the replicate samples was 4±2% (4 groups). The field blanks (0.29, 0.25, 0.24 ng/L) wereless than 20% of the minimum concentrations for all samples.

(iii) THg in soil samples. THg_{soil} was determined using aqua regia digestion in a hot water bath followed by a BrCl oxidation-$SnCl_2$ reduction-trap preconcentrating-CVAFS [13]. Quality control for the determination of THg_{soil} was conducted with standard reference materials (SRMs) (GSS-5, GBW-07405, Ministry of Geology and Mineral Resources of China). The mean testing error of the 4 SRMs was 1.2±0.6%, with a coefficient of variation of 0.6%.

2.3 Data analysis

Statistical analysis was performed using Microsoft Office Excel 2003 and SPSS 16.0. All statistical results were considered significant at $P<0.05$.

3 Results and discussion

3.1 Distribution of Water quality parameters

Figure 2 shows the distributions of the main water quality parameters including T, pH, DOC, and TSP in the YZ River and its two tributaries. The mean T was 16.5±2.0°C (mean±SD, n=17), which is comparable to that in summer reported by Liu [6]. The T of the YZ River is relatively low for its high elevation, low temperature, and considering that its major source is glacial meltwater. The mean pH was 8.4±0.1 (n=17), with a tight range of spatial variation. The pH values of the Lhasa and Niyang Rivers were lower than

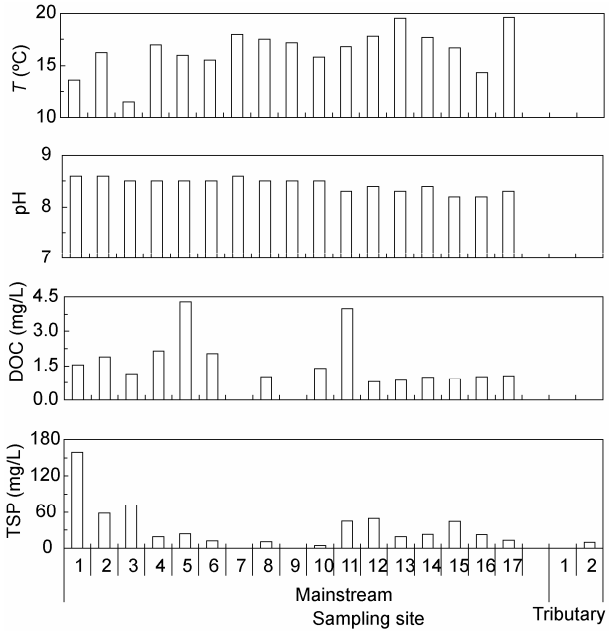

Figure 2 Water quality parameters measured concomitantly with Hg determinations in the YZ River Basin.

those of the YZ River. The average concentration of DOC was 1.67±1.09 mg/L (n=15), which is close to the lower limit of DOC of the global natural rivers (2–25 mg/L) [14]. The average concentration of TSP was 38±39 mg/L (n=15), which is much lower than the global average riverine level (150 mg/L) [15]. One order magnitude of difference in TSP at different cross sections of the river was observed. The TSP concentrations at the upstream section of the river from Site 1 (29.76°N, 83.94°E) to 3 (29.32°N, 85.26°E) were distinctly higher, possibly due to dust fallout from the arid regional characteristics.

3.2 Distribution of Hg

(i) Hg concentrations and speciation. THg and TMeHg in surface waters of the YZ River averaged 2.79±1.05 ng/L (n=17) and 0.12±0.05 ng/L (n=17), respectively. TMeHg constituted 5±2% (n=17) of the THg. Dissolved and particulate Hg accounted for 31% (0.77±0.16 ng/L, n=15) and 69% (2.01±1.05 ng/L, n=15) of the THg, and 71% (0.08 ±0.03 ng/L, n=15) and 29% (0.04±0.04 ng/L, n=15) of the TMeHg. The partition coefficient (K_d) is the particulate Hg concentration divided by the dissolved Hg concentration (reported as $\log K_d$). The $\log K_d$ varied from 4.5 to 5.6 for THg and from 3.1 to 5.1 for TMeHg in surface waters of the YZ River. The concentrations of both THg and TMeHg in surface waters of the YZ River are similar to those observed in the unpolluted Savannah River of the United States, but much lower than for rivers impacted by anthropogenic activities (Table 1). The percentage of TMeHg expressed as a fraction of THg in the YZ River is higher than that observed in the Savannah River, reaching up to 3.4%. The $\log K_d$ values

Table 1 Comparison of THg and TMeHg concentrations in surface waters of the YZ River with other rivers (unit: ng/L)

	Location	Period	THg	TMeHg	Pollution source	Reference
China	YZ River (Tibet)	2007	1.46–4.99	0.06–0.29	None	this study
	Songhua River (Jilin & Heilongjiang)	2005	21–173		Historical acetaldehyde production	[16]
		1990–1992		0.11–0.83	Historical acetaldehyde production	[17]
	Wujiang River (Guizhou)	2002	2.6–329.6		Mining, coal combustion and rock weathering	[18]
	Huangpu River (Shanghai)	2002	120–1580		Urban sources	[19]
US	Scioto River (Ohio)	2004	1.2–12		Flow through urban and agricultural land	[20]
	Savannah River (Georgia)	2000–2002	2.59	0.085	None	[21]
	Carson River (Nevada)	1994	4.28–2107	0.3–7.2	Mining	[22]
	Patuxent River	1993& 1995–1996	<0.5–6	0.025	None	[23]
	Nura River (Kazakhstan)	2001–2005	<2–4300		Historic acetaldehyde production	[24]
	Pra River (Ghana)	2002–2003	28.70–462.10	<0.028–19.640	Gold mining	[25]

of THg and TMeHg in the YZ River are similar to those observed in the unpolluted Patuxent River (4.8–5.7 and 3.8–4.0), but lower than for the Carson River impacted by mining activities (4.2–6.7 and 4.0–5.5).

RHg represents the Hg substrate available for methylation, Hg^0 formation, and other conversion processes [26]. Its concentrations in surface waters of the YZ River ranged from 0.10 to 0.36 ng/L with an average level of 0.20 ng/L, which are much lower than those observed in rivers impacted by anthropogenic activities such as the Wujiang River (1.5 ng/L) [18] and Carson River (0.4–12.28 ng/L) [22]. RHg comprised 8±4% (n=17) of the THg, indicating that only a very small fraction of Hg found in surface waters of the YZ River is Hg^{2+}. This is likely due to the composition and properties of TSP in the river.

(ii) Spatial distribution of Hg. The spatial distributions of Hg species in surface waters of the YZ River are presented in Figure 3. THg showed a decreasing trend from Sites 1 to 6 (29.34°N, 88.83°E), an increasing trend from Sites 6 to 11 (29.30°N, 91.07°E), and a clear decreasing trend from Sites 11 to 17 (29.49°N, 94.58°E). Statistical analysis showed that THg, DHg, and PHg followed normal distributions (P=0.307, 0.119, 0.257, n=15), that DHg had no significant correlation with THg, and that PHg was significantly related to THg (r=0.990, P<0.01, n=15) (Figure 4). These results indicate that THg levels are dependent on PHg, and that 98% of the spatial variation of THg can be explained by the linear relationship between THg and PHg. Moreover, TSP that followed a normal distribution excluding an extreme value from Site 1 (P=0.127, n=14) had a significant relationship with THg in surface waters (Figure 4). This indicated that 61% of the spatial variation of THg was controlled by TSP, while the rest was due to other factors. Therefore, Hg in surface waters of the YZ River primarily combines with particles. This finding is in accordance with that reported for the Carson or Minnesota Rivers [22,27]. Most suspended particles in river water come from the erosion of weathered rocks and soils within the river basin, while a small portion are the organic and mineral materials produced in the water [28]. No significant relationship was found between THg in surface waters and soils near the bank in this study; however, the results presented here are not sufficient to assess the impact of soil erosion on the distribution of particles containing Hg in surface waters of the YZ River, and more detailed studies are needed to better quantify this conclusion. The peak values of THg, PHg, and

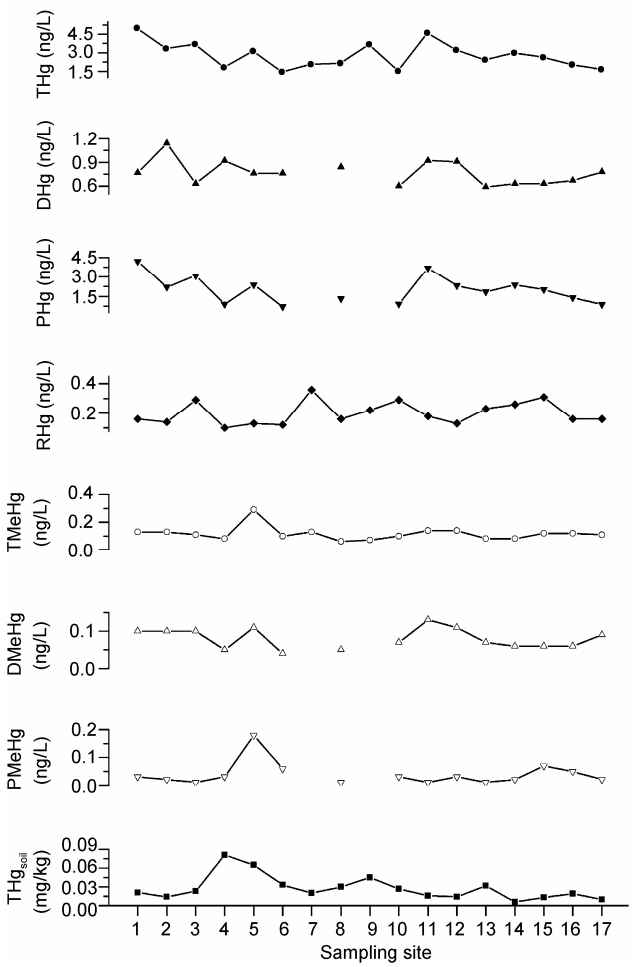

Figure 3 Spatial distribution of Hg species in surface waters and soils near the bank of the YZ River.

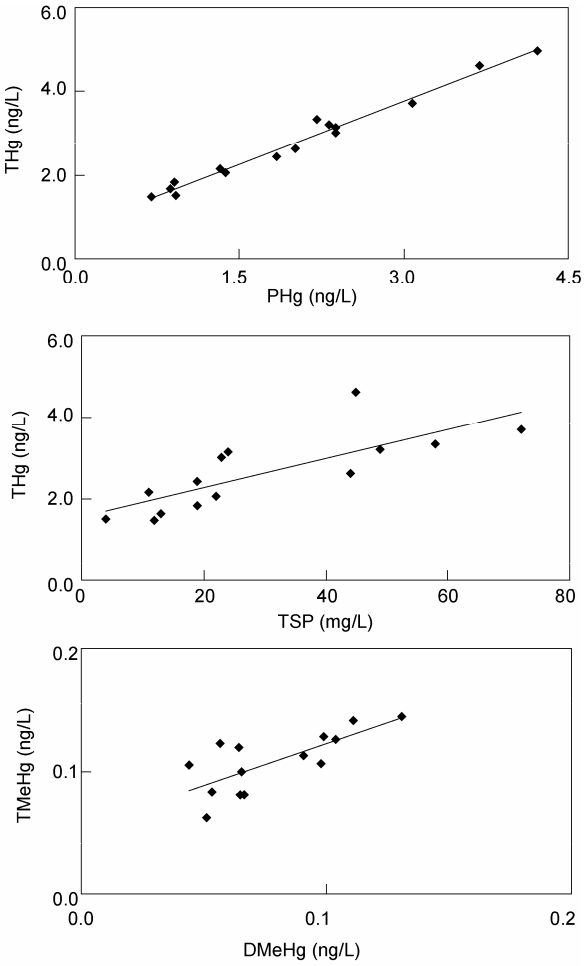

Figure 4 Relationships between PHg and THg, TSP and THg, and DMeHg and TMeHg in surface waters of the YZ River.

TSP were observed at Site 1 of the upper reaches of the river, where there is little human activity, but higher surface wind speed when compared with the other sampling sites. These findings can probably be attributed to sediment resuspension and dust settling. In addition, many studies have shown that there is a positive relationship between DOC and DHg [29,30]. However, the spatial variation of DHg is not controlled by DOC in surface waters of the YZ River, which is similar to the Wujiang River in China [31] and the Carson River in the United States [22]. This is probably because the alkaline water and low concentration of DOC in surface waters of the YZ River result in little complexation of Hg to DOC. Therefore, it can be assumed that a large fraction of DHg in the studied system is available for conversion processes and uptake by aquatic organisms.

The spatial variation trend of RHg was not obvious, and it had a wavy distribution. Statistical analysis showed that RHg followed a normal distribution (P=0.107, n=17), and that there was no significant relationship between RHg and THg, suggesting that only a very small fraction of Hg found in surface waters of the YZ River is available for conversion processes.

TMeHg decreased from Sites 1 to 4 (29.12°N, 87.58°E) in the upper reaches of the river, and fluctuated from Sites 5 (29.36°N, 88.13°E) to 17 in the middle reaches of the river, with the TMeHg levels at Site 5 being exceptionally high. Statistical analysis showed that TMeHg, DMeHg, and PMeHg followed or approximated normal distributions with the exception of an extreme value from Site 5 (P=0.412, 0.334, 0.023, n=14), that DMeHg was significantly related to TMeHg (r=0.746, P<0.01, n=14) (Figure 4), and that PMeHg had no significant correlation with TMeHg. These results indicate that DMeHg is the dominant species of MeHg, controlling 56% of the spatial variation of TMeHg. The spatial variation of TMeHg in river water is influenced by other Hg species and some water quality parameters, such as the total and reactive Hg, temperature, pH, DOC, suspended particles, redox potential (Eh), dissolved oxygen (DO), and microorganisms [32,33]. Moreover, the most important factors generally vary with the rock, bioclimatic condition, and human activity (land use, mining, industry, agriculture) in a river basin. Table 2 shows the Pearson correlation coefficients (r) between THg, RHg, T, pH, DOC, and TSP and TMeHg in surface waters of the YZ River. Only TSP and TMeHg were significantly correlated, but their correlation probability value (P) was lower than that between TSP and THg, indicating that the influence of TSP concentrations on TMeHg was relatively small, and that TMeHg was probably associated with other characteristics of particles. Some studies have shown that particulate organic matter may play an important role in mercury methylation [34]. As a result, TMeHg in surface waters of the YZ River is not strongly affected by THg, RHg, T, pH, DOC, and TSP, while its spatial distribution is likely controlled by other factors. The concentrations of both TMeHg and PMeHg were abnormally high at Site 5 without influence by anthropogenic activities, possibly due to the types and concentrations of particulate organic matter. In addition, in surface waters of the YZ River, the spatial variation of DMeHg is not controlled by DOC, which is similar to DHg, and the spatial variation of PMeHg is not dominated by TSP.

Furthermore, The Lhasa and Niyang Rivers were set as examples to preliminarily examine the influence of the inflow of tributaries on the spatial distributions of THg and TMeHg in surface waters of the YZ River. Figure 5 shows THg and TMeHg levels of the Lhasa River, Niyang River, and YZ River sites located just upstream or downstream

Table 2 Pearson correlation coefficients (r) between environmental factors and TMeHg in surface waters of the YZ River [a)]

	THg	RHg	T	pH	DOC	TSP
TMeHg	0.365 (n=16)	0.029 (n=16)	–0.182 (n=16)	–0.079 (n=16)	–0.022 (n=13)	0.616* (n=13)

a) Statistical significance of correlation coefficients indicated at the 0.05 (*) levels.

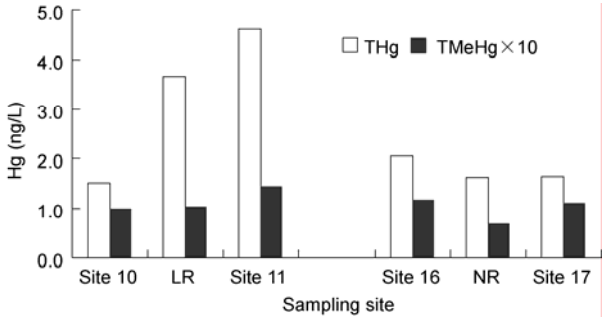

Figure 5 Influence of the inflow of the Lhasa River (LR) and Niyang River (NR) on THg and TMeHg levels in surface waters of the YZ River.

from each tributary. THg concentrations were in the order of Site 11 > the Lhasa River > Site 10 and the Niyang River ≈ Site 17 < Site 16, and TMeHg concentrations were Site 11 > the Lhasa River ≈ Site 10 and the Niyang River < Site 17 ≈ Site 16. These results suggest that the inflow of both the Lhasa and Niyang Rivers probably influences the concentrations of THg in surface waters of the mainstream, but such an effect is not obvious for TMeHg. In addition, the other reason for high levels of THg at Site 11 of the mainstream was that the portion of the river flowing through the Gongga County was strongly affected by anthropogenic activities. Moreover, the levels of Hg at the sampling site in the Lhasa River (29.48°N, 90.93°E), which was more heavily influenced by human activities, were higher than those observed at the sampling site in the Niyang River (29.47°N, 94.42°E). These findings suggest that anthropogenic activities may have a potential impact on the YZ River system.

4 Conclusions

In this study, Hg speciation and spatial distribution in surface waters of the YZ River were preliminarily investigated. The results revealed that the mean THg concentration in surface waters of the YZ River was 2.79 ng/L, which is comparable to that observed in some unpolluted rivers of the United States, lower than the global average riverine concentration of THg (5.0 ng/L) [35], and much lower than for rivers that have been strongly impacted by anthropogenic activities. Thus, the YZ River is less disturbed by anthropogenic Hg emissions, and THg concentrations in its surface waters can be taken to represent the background levels in river systems of the Tibetan Plateau. PHg accounted for 69% of the THg, and the $\log K_d$ value of THg was 4.9, indicating that TSP plays an important role in terms of the transfer, conversion and fate of Hg in surface waters of the YZ River. The spatial variation of THg was very similar to that of PHg, and its 61% was controlled by TSP. RHg has little influence on the transport and fate of Hg in surface waters of the YZ River, accounting for only 8% of the THg. The mean concentration of TMeHg was 0.12 ng/L, which is similar to the reported background value of approximately 0.1 ng/L for aquatic environments in China [36], but much lower than for polluted rivers. Thus, it can be taken to represent the riverine background concentration of the Tibetan Plateau. Moreover, the dominant component of TMeHg is dissolved. TMeHg is not strongly affected by THg, RHg, *T*, pH, DOC, and TSP, while its spatial distribution is likely dependent on other factors. In addition, the inflow of both the Lhasa and Niyang Rivers probably influences the concentrations of THg in surface waters of the YZ River, but such an effect is not obvious for TMeHg.

We are currently conducting studies to evaluate the concentration and distribution of THg and TMeHg in fish from the YZ River, which will provide some important basic data to enable a further understanding of the biogeochemical cycle of Hg in the YZ River.

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