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ENVIRONMENTAL  
SCIENCES  
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# Distribution and transportation of mercury from glacier to lake in the Qiangyong Glacier Basin, southern Tibetan Plateau, China

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## ARTICLE INFO

### Article history:

Received 15 July 2015

Revised 10 September 2015

Accepted 28 September 2015

Available online 23 January 2016

### Keywords:

Mercury

Glacier

River water

Lake water

Transportation

Tibetan Plateau

## ABSTRACT

The Tibetan Plateau is home to the largest aggregate of glaciers outside the Polar Regions and is a source of fresh water to 1.4 billion people. Yet little is known about the transportation and cycling of Hg in high-elevation glacier basins on Tibetan Plateau. In this study, surface snow, glacier melting stream water and lake water samples were collected from the Qiangyong Glacier Basin. The spatiotemporal distribution and transportation of Hg from glacier to lake were investigated. Significant diurnal variations of dissolved Hg (DHg) concentrations were observed in the river water, with low concentrations in the morning (8:00 am–14:00 pm) and high concentrations in the afternoon (16:00 pm–20:00 pm). The DHg concentrations were exponentially correlated with runoff, which indicated that runoff was the dominant factor affecting DHg concentrations in the river water. Moreover, significant decreases of Hg were observed during transportation from glacier to lake. DHg adsorption onto particulates followed by the sedimentation of particulate-bound Hg (PHg) could be possible as an important Hg removal mechanism during the transportation process. Significant decreases in Hg concentrations were observed downstream of Xiao Qiangyong Lake, which indicated that the high-elevation lake system could significantly affect the distribution and transportation of Hg in the Qiangyong Glacier Basin.

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## Introduction

Mercury (Hg), a highly toxic trace metal, is considered to be a global pollutant due to its long-range transport via the

atmosphere and biomagnification in ecosystems (Selin, 2009). Inorganic Hg species in aquatic systems can be converted into toxic methylated forms, such as methylmercury (MeHg), a bioaccumulative and neurotoxic form of Hg, which poses a

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potential threat to human health after bioaccumulation and biomagnification through food chains (Li et al., 2015a, b; Zhang et al., 2010a). The consumption of aquatic products, especially fish, is an important pathway of Hg exposure for humans and wildlife around the world (Zhang et al., 2014; Mergler et al., 2007). Hg contamination and its biogeochemical cycles in the aquatic ecosystem have become a hot topic in scientific research.

The Tibetan Plateau, or “Third Pole”, is one of the most remote and pristine areas in the world, with an area of approximately 2,500,000 km<sup>2</sup> and an average elevation of more than 4000 m above sea level (a.s.l.) (Qiu, 2008). This region is the most glacier-concentrated region at low- and mid-latitudes (Kang et al., 2010a; Yao et al., 2012). Known as the “Water Tower of Asia”, the Tibetan Plateau is the source region of the fresh water for more than one-third of the world’s population through its 10 largest rivers, including the Yangtze River, Yellow River, Yarlung Tsangpo (Brahmaputra), Salween River, and Indus River (Huang et al., 2014; Immerzeel et al., 2010). However, in recent decades, several studies have shown that pollutants, such as Hg, originating from outside the Tibetan Plateau could be transported over a long range via atmospheric circulation and eventually be deposited onto the glaciers, endangering ecosystems and human health in the downstream regions of the glacier-fed river basins (Huang et al., 2014; Huang et al., 2012a; Zhang et al., 2014). This becomes more significant for the downstream ecosystems in the light of ongoing global warming, as the accumulated pollutants will largely be released due to accelerated glacier melting (Bogdal et al., 2009; Zhang et al., 2015). The study of Hg distribution and its transportation processes from the glacier to its downstream on the Tibetan Plateau is thus imperative for further understanding its potential downstream impact. However, due to the difficulty of sampling, there have been few studies to date on the subject, and therefore, the transportation process of Hg from the glaciers to its downstream areas in glacier basins in the Tibetan Plateau is still poorly understood. Currently, very limited studies on Hg distribution have been conducted in the aquatic systems of Nam Co Basin, which lies at the foot of the Nyainqêntanglha Mountain in the southern Tibetan Plateau, and is the second largest saline lake on the Tibetan Plateau with an area of approximately 1960 km<sup>2</sup>; and in the Yarlung Zangbo River, which is the highest river in the world with an average elevation of over 4000 m a.s.l., originates from the Gyaimanezong Glacier in the northern foothills of the Himalayas, flows from west to east through the southern part of Tibetan Plateau, and has a total length of 2057 km and a drainage area of 239,228 km<sup>2</sup> (Wang et al., 2012b; Zheng et al., 2010). A previous study on the Hg distribution and transportation in the Zhadang Glacier Basin on the south shore of Lake Nam Co, southern Tibetan Plateau, has revealed that total mercury (THg) in the melt water was dominated by PHg, and decreased significantly during the transportation from the supraglacial streamwater to the downstream. Moreover, a significant positive relationship was found between THg concentrations and total suspended particulates (TSPs) in the meltwater, indicating that particulates played an important role during the transportation process (Guo, 2012).

The Qiangyong Glacier Basin is situated on the southern part of the Tibetan Plateau, and consists of glaciers, rivers and lakes. Glacier meltwater is the main water supply for the

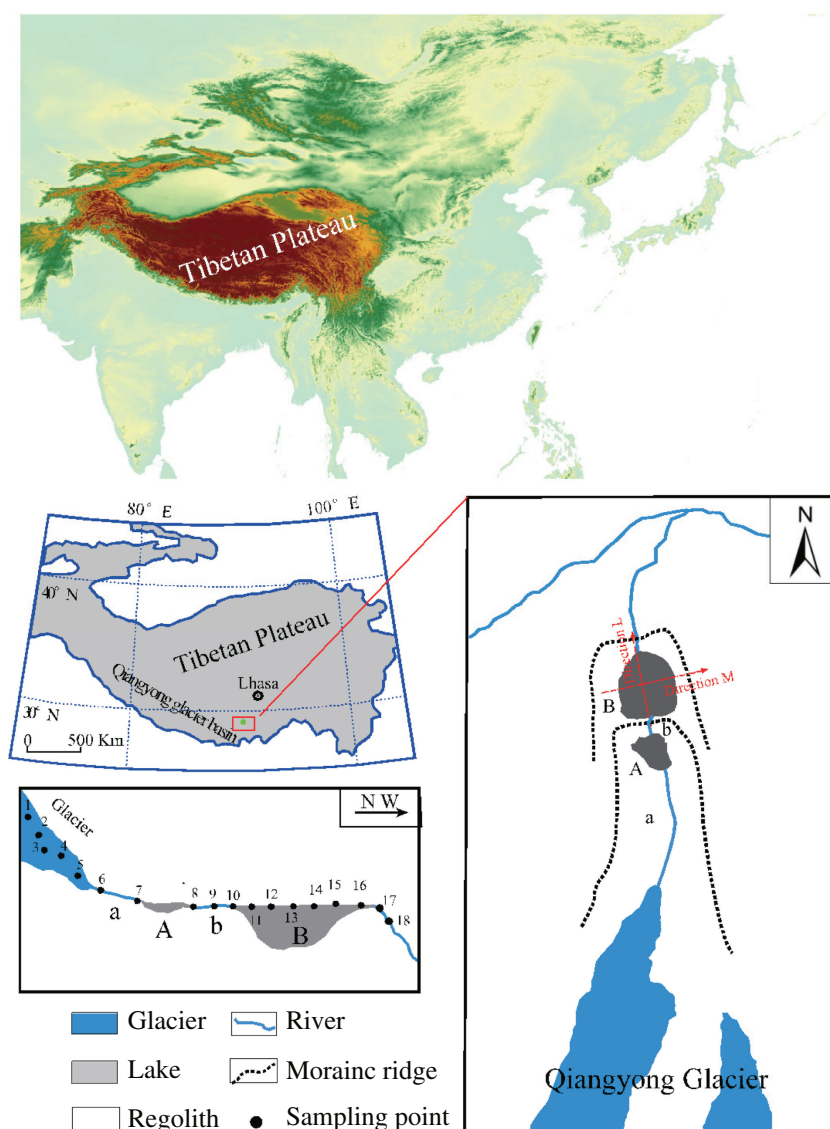
lakes: the meltwater from Qiangyong Glacier flows toward the glacial terminus river and supplies Da Qiangyong Lake directly, which finally forms a semi-enclosed lake system (Luo et al., 2003; Owen et al., 2005). Therefore, it is considered an ideal region to investigate Hg transportation from the glacier to its downstream areas. In this study, we conducted an extensive sampling campaign and obtained successive snow, glacier melt stream water, and lake water samples in this region. The spatiotemporal distribution and transportation process of Hg from glacier to lake over the Qiangyong Glacier Basin were investigated. Moreover, the dominant factors controlling the Hg distribution and its transportation process were also discussed. This study will not only enhance our understanding of the Hg distribution and transportation mechanisms in the high-elevation glacier basins, but also be important for assessing the potential impact of Hg on downstream ecosystems as well as its environmental risks.

## 1. Materials and methods

### 1.1. Study area and sampling location

The Qiangyong Glacier Basin (28°53′N, 90°13′E) is located between the Himalayan ranges and the Yarlung Zangbo River in the southern Tibetan Plateau (Fig. 1). The annual mean air temperature in this area is approximately 2.4°C, with the lowest temperature in January at −5.5°C and the warmest in July at 10°C (Luo et al., 2003). The annual mean precipitation in the region is approximately 370 mm, with 90% of the precipitation occurring in the summer season (Li et al., 2011). The atmospheric circulation in our study region is mainly influenced by westerlies and the Indian monsoon, which prevails in the summer season from June to September (Tian et al., 2007; Tian et al., 2001; Zhang et al., 2012). Qiangyong Glacier is located on the north slope of the Himalayas, with a length of 4.6 km, a maximum width of 2.8 km and a total area of 7.7 km<sup>2</sup> (Luo et al., 2003). The average equilibrium line altitude of Qiangyong glacier is approximately 5600 m a.s.l. (Luo et al., 2003).

A proglacial lake is defined as a lake system that is formed not far from the glacier terminus during glacial melting and retreat and is directly supplied by glacial meltwater (Karlén, 1981). Situated at an altitude of approximately 4780 m a.s.l., Qiangyong Lake is the proglacial lake of Qiangyong Glacier and is less than 1 km from the glacier tongue (Fig. 1) (Li et al., 2011; Owen et al., 2005). Qiangyong Glacier Basin consists of two lakes: Da Qiangyong Lake, with an area of approximately 0.1 km<sup>2</sup> and a maximum water depth of approximately 18.5 m, and Xiao Qiangyong Lake, with an area of approximately 0.03 km<sup>2</sup> and a maximum water depth of approximately 4.5 m. The hydrological conditions of the two lake systems are directly affected by the melting intensity of Qiangyong Glacier. Glacier meltwater in the summer is the main water supply for the two lakes through the glacial terminus river and a river formed between the Da Qiangyong Lake and Xiao Qiangyong Lake (hereafter, the middle river). These rivers and lakes in Qiangyong Glacier Basin form a typically semi-enclosed lake system over the southern Tibetan Plateau (Owen et al., 2005). However, an outflow river formed beneath the basin of Da



**Fig. 1** – The map showing the location of the Tibetan Plateau, Qiangyong Glacier Basin and the sampling sites in the Qiangyong Glacier Basin (A: Xiao Qiangyong Lake; B: Da Qiangyong Lake; a: Glacial terminus river; b: middle river).

Qiangyong Lake due to accelerated glacial melting, and the lake system reached a temporally open status during the summer season (Li et al., 2011).

The lithology distributed in the Qiangyong Glacier Basin is mainly made up of slate, siltstone, arenaceous shale, mud shale, basalt and conglomerate. The bedrocks that Qiangyong Glacier covers are mesozoic metamorphic sandstone, shale and slate (Luo et al., 2003). The major chemical component of the bedrock is  $\text{SiO}_2$  (52% of the total mass) followed by  $\text{Al}_2\text{O}_3$  (21%) and  $\text{Fe}_2\text{O}_3$  (10%) (Luo et al., 2003). The residual moraine formed by the quaternary glaciations and loose deposits formed via the weathering action of the running water and frost are widely distributed in The Valley of Qiangyong Glacier Basin (Li et al., 1983).

Dust particles deposited on the Qiangyong Glacier were produced by long-range transport of fine atmospheric particulate matter generated in the vast arid and semi-arid regions

in the hinterland and to the west of the Tibetan Plateau (Wake et al., 1994; Huang et al., 2012a), and were the main source of river/lake sediments for the Qiangyong Glacier Basin (Li et al., 2011), whereas particles eroded from the watershed and the river banks contributed less to the river/lake sediments (Li et al., 2011). The grain size of the particulate matter deposited on the glaciers over the Tibetan Plateau ranges from  $1\ \mu\text{m}$  to  $13\ \mu\text{m}$  (Li et al., 2000). The sediments in Da Qiangyong Lake and the subglacially-formed debris in deposits on the bedrock surface of Qiangyong Glacier also showed fine-grain characteristics. To be specific, for the lake sediments in the lake cores, the median grain size varied from 6 to  $35\ \mu\text{m}$ . The major component of the lake sediments was fine silt ( $4\text{--}20\ \mu\text{m}$ ), which accounts for 35%–65% of the total mass. Clay mineral ( $<5\ \mu\text{m}$ ) and coarse-grained clastic ( $>20\ \mu\text{m}$ ) contents occupies 10%–45% and 5%–60% respectively (Li et al., 2011). For the debris in chemical deposits on the bedrock surface, the mode size is  $8\ \mu\text{m}$ . Silt and clay

**Table 1 – Detailed information of sampling sites.**

Sampling point	Sampling site	Longitude/latitude	Altitude (m a.s.l.)	Sample numbers	Sampling time
1–5	Surface snow	–	5101–5597	7	August 5–6
6	Glacial terminus river	28°53′12.4″N/90°13′58.6″E	–	17	August 7–9
7	Inlet of Xiao Qiangyong Lake	28°53′11.4″N/90°13′34.1″E	4882	17	14:00, August 10
8	Outlet of Xiao Qiangyong Lake	28°53′18.6″N/90°13′33.5″E	4881	1	14:00, August 10
9	Middle River	28°53′19.5″N/90°13′33.5″E	4908	1	August 7–9
10	Inlet of Da Qiangyong Lake	28°53′20.7″N/90°13′33.3″E	4891	1	14:00, August 10
11–16 (Direction L)	Da Qiangyong Lake	–	4770	12	August 10
17	Outlet of Da Qiangyong Lake	28°53′18.6″N/90°13′33.5″E	4881	1	14:00, August 10
18	Outflow river	28°53′33.5″N/90°13′30.8″E	4869	1	14:00, August 10

(<50  $\mu\text{m}$ ) accounts for 98% of the total mass and clay occupies 32%. Suspended particulate matter in the glacial meltwater and debris in the chemical deposits on the bedrock surface have the same source. The main chemical components of the suspended particulate matter in the glacial meltwater were  $\text{SiO}_2$  (31% of the total mass) followed by  $\text{Al}_2\text{O}_3$  (21%),  $\text{CaO}$  (14%), and  $\text{Fe}_2\text{O}_3$  (9%) (Luo et al., 2003).

## 1.2. Sample collection

Surface snow (0–5 cm), river and lake water samples were collected from the Qiangyong Glacier Basin in this study (Fig. 1 and Table 1). Seven surface snow samples were collected at altitudes ranging from 5101 m to 5597 m (Table 2), including 3 surface snow samples for THg analysis and 4 surface snow samples for DHg analysis. A total of 34 surface river water samples for DHg analysis were collected from the two rivers. The river water samples were collected at a time interval of 2 hr during the daytime (8:00 am–20:00 pm) and 4 hr during the nighttime. These river water samples were not analyzed for THg because all of the river water samples were filtered through a 0.45- $\mu\text{m}$  membrane (Durapore, Millipore) in the field. The river water samples collected at the inlets (sampling points 7 and 10) and outlets (sampling points 8 and 17) of the two lakes were analyzed for THg. A total of 12 surface lake water samples of Da Qiangyong Lake were collected at a regular distance interval from the shore to the center of the lake in two directions (directions L and M). Some physico-chemical parameters, including pH, temperature, and conductivity, were measured using a HACH Hydrolab multi-parameter probe *in situ*.

All of the samples were collected in replicates strictly following “clean hands–dirty hands” sampling protocols (Fitzgerald, 1999). Polypropylene clean room suits, polyethylene gloves and masks were worn at all times during sampling. The snow samples were sampled using high density

polyethylene (HDPE) scoops. To prevent contamination, the scoops were soaked in 20%  $\text{HNO}_3$  for 24 hr and rinsed three times with ultrapure Millipore® water before use. All of the water samples were collected approximately 20 cm beneath the water surface using 50-mL polypropylene Falcon® centrifuge tubes. When river/lake water was collected, the sampling person faced the flow direction to prevent disturbance of the water flow and river/lake deposits. The 50-mL polypropylene BD Falcon® centrifuge tubes were rinsed 3 times with river/lake water before sampling. All of the samples were stored in 50-mL polypropylene BD Falcon® centrifuge tubes and immediately doubly packed in Ziploc bags. Field blanks filled with ultrapure water were opened during sample collection and were handled as samples. 100  $\mu\text{L}$  of BV-III grade (CMOS) HCl (Beihua Chemical, China) was spiked into all water samples *in situ* (for lake/river water samples) or within 24 hr (for surface snow samples) after sampling where field conditions precluded the onsite spike. The water samples for DHg determination were filtered through a 0.45- $\mu\text{m}$  membrane (Durapore, Millipore) first using a 500 mL borosilicate glass filtering apparatus. The filtered water was then transferred to a new 50-mL polypropylene Falcon tube and acidified to 0.5% (V/V) with HCl for preservation. All of the samples were stored at 4°C until analysis.

## 1.3. Analytical methodology

The analysis of Hg was performed according to US EPA Method 1631 (version E) (EPA, 2002). The Hg concentration was quantified via cold vapor atomic fluorescence spectroscopy (CVAFS) using an Analytik-Jena Hg analyzer (Analytik-Jena Corporation Inc., Jena, Germany) in a Class 1000 metal-free laboratory clean room at the State Key Laboratory of Cryospheric Sciences, Chinese Academy of Sciences (CAS). The analytical protocol for Hg was described elsewhere (Huang et al., 2012b). Briefly, each sample was first oxidized by  $\text{BrCl}$  (0.5 mL/100 mL sample). After oxidization,  $\text{NH}_2\text{OH}\cdot\text{HCl}$  (0.2 mL/100 mL sample) was added to

**Table 2 – Detailed information of sampling sites on Qiangyong glacier.**

Sampling site	Longitude/latitude	Altitude (m a.s.l.)	Sample types	Sample number
1	28°52′43.2″N/90°13′19.5″E	5597	DHg	2
2	28°51′51.1″N/90°13′15.9″E	5500	THg/DHg	2
3	28°51′52.5″N/90°13′12.4″E	5498	THg	1
4	28°52′34.6″N/90°13′19.5″E	5231	DHg	1
5	28°52′43.9″N/90°13′45.6″E	5101	THg	1



the sample to remove the remaining BrCl.  $\text{SnCl}_2$  was then added to the sample to reduce  $\text{Hg}^{2+}$  to  $\text{Hg}^0$ , which was quantified by the fluorescence spectrophotometer. All samples were analyzed within 2 weeks after collection.

Quality assurance and quality control of the analysis were conducted using replicates, method blanks, field blanks and ongoing precision and recovery (OPR) standards. The method detection limit (MDL), defined as 3 times the standard deviation of 10 replicate measurements of a blank solution, was less than 0.2 ng/L for THg and DHg. The relative standard deviations (RSDs) for the replicate samples were <5% for DHg and THg. An OPR standard of 5 ng/L and a method blank were interspersed in every 10 samples of THg and DHg to check instrument operation. The Hg concentrations of the method and field blanks were always found to be below the MDL, indicating minimal contamination during sampling, transportation and treatment for this study. The recovery percentage was 95%–105% of the certified values for DHg and THg. Additionally, Falcon® centrifuge tubes filled with ultra-pure water were randomly tested for Hg concentration before being used for sampling; they were always below the MDL.

#### 1.4. Flow measurement

A diurnal observation in the middle river was conducted at the hydrological cross-section (Table 1), where the river water was sampled at the same time. The time interval of the flow observation was 2 hr during the daytime (8:00 am–20:00 pm) and 4 hr during the nighttime. The flow measurement method was described in detail elsewhere (Hersch, 1995). Briefly, 5 flow measurement perpendiculars at regular distance intervals were chosen for flow measurement according to the river width ( $\leq 5$  m) during the observation period. The flow velocity, water depth and distance between the observation verticals were measured using a Propeller Flow Velocity Meter (LS1206B), a sounding pole and a measuring tape, respectively. The discharge of the middle river was estimated by using the mean-section method (Hersch, 1998).

#### 1.5. Statistical analysis

All of the data processing and statistical analyses were conducted using Microsoft Office Excel 2007, IBM SPSS Statistics 20 and Origin Pro 8, including data-normality tests, correlation analysis, and regression analysis. The upper limit of the significance level ( $p$ ) was 0.05, and the correlations were considered statistically significant when  $p < 0.05$ .

## 2. Results

### 2.1. Concentration levels of THg concentration in surface snow

Fig. 2 shows that the THg concentrations in surface snow samples ranged from 7.6 to 10.6 ng/L with an average of  $8.8 \pm 0.5$  ng/L ( $n = 3$ ). A previous study showed that the THg concentrations in glacier snow over western China ranged

from <1 to 43.6 ng/L (Zhang et al., 2012), which were comparable to those observed from the Qiangyong Glacier. Moreover, the THg concentrations in the Qiangyong Glacier were also at the lower end of the THg concentrations in the remote regions of the Arctic (Kang et al., 2010b), but were significantly lower than the concentrations reported for snow samples in heavily polluted cities, such as Beijing (106–162 ng/L) (Wang et al., 2004).

The DHg concentrations in surface snow samples ranged from 1.0 to 4.5 ng/L with an average of  $2.5 \pm 0.5$  ng/L ( $n = 4$ ) (Fig. 2), which accounted for 28.1% of THg and indicated that Hg in surface snow was dominated by PHg. Previous studies revealed that particulate matter played an important role in atmospheric Hg transportation and deposition over western China (Huang et al., 2012a; Huang et al., 2012c; Zhang et al., 2012) and that the main source of Hg deposited on the Plateau was primarily associated with particulate matters (Loewen et al., 2007; Huang et al., 2012c). Similar to what has been found in previous studies, the higher PHg% in our study reconfirmed the distribution and deposition mechanisms of atmospheric Hg over western China.

### 2.2. Concentration levels of Hg in lake water

The THg concentrations in lake water samples ranged from 0.4 to 1.8 ng/L, with an average of  $0.9 \pm 0.4$  ng/L ( $n = 12$ ) (Hg). A recent study on 38 lakes in the Tibetan Plateau revealed that the THg concentrations in the surface lake waters ranged from <1 to 40.3 ng/L. The THg concentration levels in the surface water of Da Qiangyong Lake lie within this range (Li et al., 2015a, b). Moreover, the concentration levels of THg in surface water of Da Qiangyong Lake were comparable to those determined in uncontaminated freshwaters worldwide (<5 ng/L) (Bloom, 1989) and natural lakes in the Antarctic (0.4–1.9 ng/L) (Watras et al., 1995), but were significantly lower than those observed in heavily polluted areas (Table 3), as in Guizhou Province, which is recognized as a heavily Hg-polluted area in China due to the presence of two key anthropogenic Hg emission sources (i.e., coal combustion and metals smelting), resulting in high levels of Hg in local ecosystems and serious Hg contamination to the local environment (Feng et al., 2008; Horvat et al., 2003; Jiang et al., 2006). As a result, the THg concentrations in the water of Da Qiangyong Lake represented the background concentration levels of THg in natural lake water over the Tibetan Plateau.

### 2.3. Concentration levels of Hg in river water

The DHg concentrations in the river water samples ranged from 0.2 to 7.7 ng/L with an average of  $1.3 \pm 1.5$  ng/L ( $n = 34$ ) (Fig. 2). Specifically, the DHg concentrations ranged from 0.2 to 7.7 ng/L with an average of  $1.7 \pm 2$  ng/L ( $n = 17$ ) in water samples taken from the glacial terminus river. The DHg concentrations were lower in the water samples taken from the middle river, which ranged from 0.2 to 2.1 ng/L with an average of  $0.8 \pm 0.5$  ng/L ( $n = 17$ ). The concentration levels of DHg in the river waters over the Qiangyong Glacier Basin were comparable with those determined in Yalu Tsangpo River and Yenisei River, but significantly lower than those determined in heavily polluted areas (Table 4).

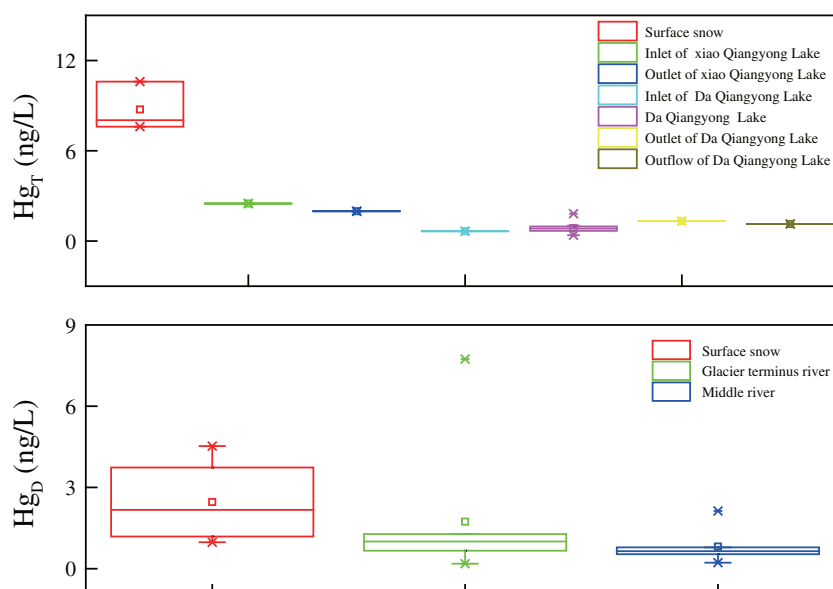


Fig. 2 – Spatial distribution of THg and DHg concentrations over the Qiangyong Glacier Basin. Each boxplot represents one sampling point. The boxes mark the 25th and 75th percentiles; The hollow points are the average concentrations; the horizontal lines inside the box are the median values; the short sidelines out of boxes are the maximum (top) and minimum (bottom); the crosses mark the 99th (top) and 1st (bottom) percentiles. DHg: dissolved Hg.

### 3. Discussion

#### 3.1. Spatial distribution of THg in lake water

The water depth of the sampling sites increased as the distance from the shore to the center of the lake increased according to our field measurement data (i.e., the deeper the water depth, the longer the distance from sampling sites to the lake shore). As shown in Fig. 3, the THg distribution showed significant spatial differences in Da Qiangyong Lake. Generally, the Hg concentrations showed a decrease from the shore to the center of Da Qiangyong Lake. The THg concentrations decreased to the minimum value at the sampling site where the water depth was approximately 18 m. The decreasing trend of the THg concentrations from the shore to the center of the lake was further supported by the correlation analysis (Fig. 3). There was a significant negative relationship between the water depth and the THg concentrations ( $R^2 = 0.6$ ,  $p < 0.05$ ).

Previous studies on the THg distribution in the surface water of a high-elevation lake, Nam Co, southern Tibetan Plateau,

revealed that the THg concentrations in the lake surface water were dominated by the PHg, and showed significant spatiotemporal variability, with “higher/lower in the monsoon/non-monsoon” for the temporal distribution and “higher/lower in the near-shore/central” for the spatial distribution, respectively. This phenomenon was consistent with the distribution pattern of TSP, which was significantly affected by the wind waves, indicating that the wind waves could significantly affect the THg distribution in the lake waters over the Tibetan Plateau (Wang, 2012b; Wang et al., 2012b; Zheng, 2009). Similarly, the spatial distribution of THg in Da Qiangyong Lake could also be explained by the fact that the hydrodynamic conditions were more intense near the shore than in the center of the lake, attributed to the disturbance by the wind waves. The TSP content in the near-shore water of Da Qiangyong Lake was significantly affected by the wind waves according to our field observation. Previous studies revealed that sediments can act both as sinks and as secondary sources of Hg (Li et al., 2006; Ullrich et al., 2001). The sedimentation of Hg, especially PHg, was considered an important Hg removal mechanism in the waters, whereas the resuspension of sediments releases the Hg

Table 3 – Comparison of the THg concentrations in water from Da Qiangyong Lake and other lakes worldwide.

Region	Site	THg (ng/L)	Main pollution sources	Reference
China	Da Qiangyong Lake, Tibetan Plateau	0.4–1.8	Non	This study
	Baihua Lake, Guizhou, southeastern China	20.3–336.0	Industrial drainage	Hou et al. (2004)
	Aha Reservoir, Guizhou, southeastern China	2.1–19.1	Mine drainage	Bai et al. (2006)
	Dianchi Lake, Yunnan, southwestern China	18–46 ng L	Industrial/domestic drainage	Wang et al. (2012a)
	Nam Co, Tibetan Plateau	0.3–4.3	Non	Wang et al. (2012b)
Global	Wisconsin Lake, USA	0.4–4.8	Non	Benoit et al. (2001)
	Lake Gordon, Australia	2.3 ± 0.4	Non	Bowles et al. (2003)
	Lake Balaton, Hungary	1.5–6.5	Low pollution region	Nguyen et al. (2005)
	Kodai, India	356–465	Thermometer factory	Karunasagar et al. (2006)

**Table 4 – Comparison of DHg concentrations in the river waters of the Qiangyong Glacier Basin and other areas worldwide.**

Study sites	DHg (ng/L)		References
	Mean	Range	
Riverwaters in Qiangyong Glacier Basin, Tibetan Plateau	1.3 ± 1.5	0.2–7.7	This study
Wujiang River, Southwestern China	7.1	1–49.2	Jiang et al. (2003)
Huangpu River, Eastern China	270 ± 420	50–1490	Ding et al. (2005)
Yalu Tsangpo River, Tibetan Plateau	0.77 ± 0.16		Zheng et al. (2010)
Patuxent River estuary, USA		0.2–1.5	Benoit et al. (1998)
Carson River, USA		2.5–46.1	Bonzongo et al. (1996)
Yenisei River, Siberia	1.5 ± 0.7	0.8–2.1	Coquery et al. (1995)
Ob River, Siberia		2.4–3.2	Coquery et al. (1995)

once sunk into the sediments (Li et al., 2006; Ullrich et al., 2001). The strong disturbance of sediments by wind waves led to the resuspension of the sediments, which accelerated the release of PHg once sunk into the sediments (Li et al., 2006; Ullrich et al., 2001). This was further supported by the fact that higher turbidity was observed in the near-shore surface lake water and decreased significantly from the shore to the center of the lake according to our field observations. Moreover, the input of Hg adsorbed on particulates carried by the river could also lead to a higher PHg concentration near the shore (Wang et al., 2012b), leading to higher THg concentrations in the near-shore areas.

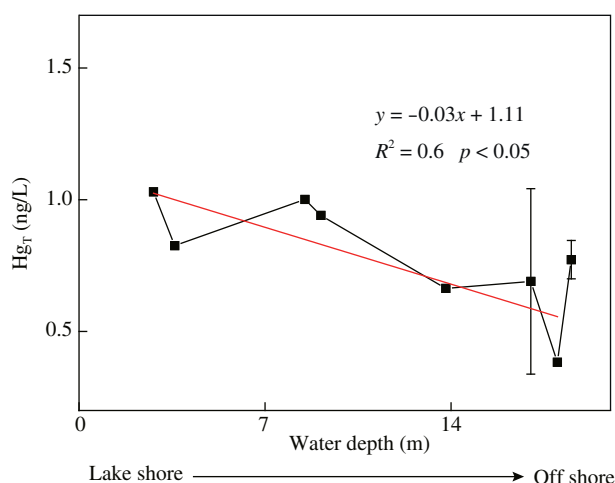
### 3.2. Diurnal variation of DHg in river water

The DHg concentrations in the rivers showed significant diurnal variations in Qiangyong Glacier Basin (Fig. 4). Generally, the DHg concentration was observed to be lowest from 8:00 am to 14:00 pm. A dramatic increase of DHg concentration was observed at approximately 14:00 pm, and the DHg

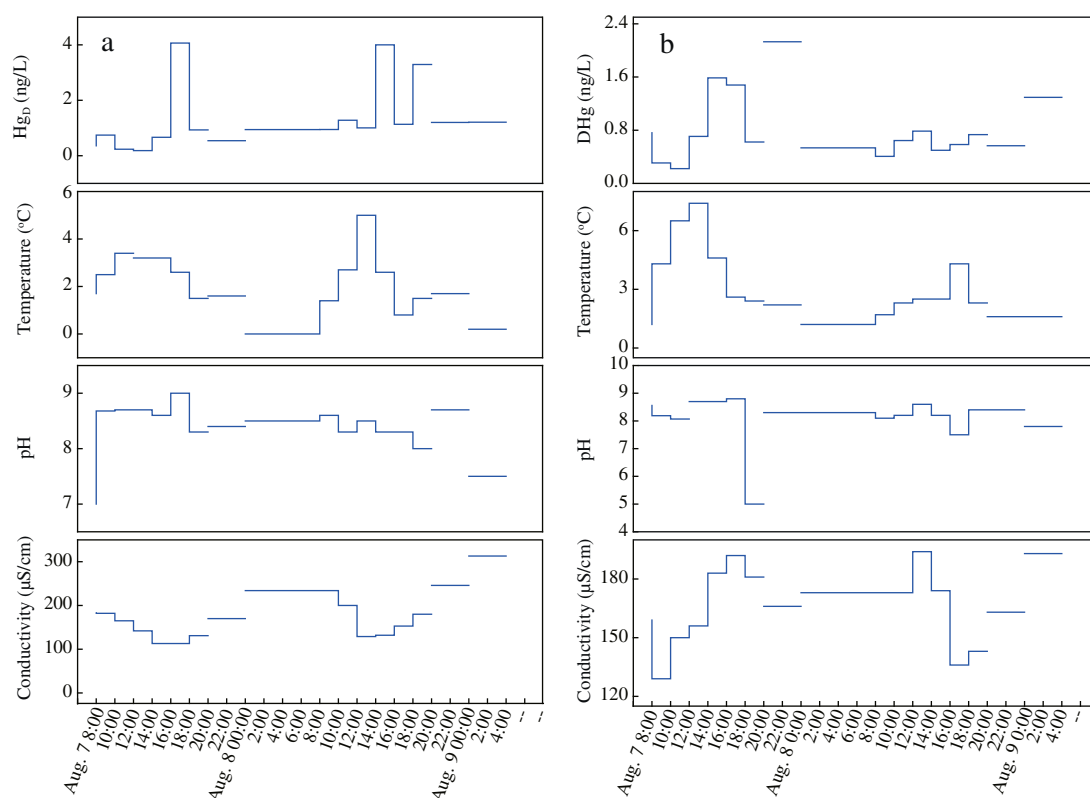
reached its peak concentration between 16:00 pm–18:00 pm. Accordingly, the highest DHg concentrations in the glacial terminus river (from 0.7 to 4.1 ng/L with an average of 2.3 ng/L) and in the middle river (from 0.5 to 1.6 ng/L with an average of 0.9 ng/L) were found from 16:00 pm to 20:00 pm. Relatively higher DHg concentrations were found at night compared with those determined from 8:00 am to 14:00 pm in the rivers.

Significant diurnal variation was also observed in the runoff of the middle river (Fig. 5). Runoff of the middle river ranged between 1.5–3.3 m<sup>3</sup>/sec with an average of 1.9 m<sup>3</sup>/sec. The runoff showed the lowest values (1.6–1.8 m<sup>3</sup>/sec) in the morning (i.e., 8:00 am–12:00 pm) and highest values (2.1–3.3 m<sup>3</sup>/sec) in the afternoon (i.e., 16:00 pm–20:00 pm), which was consistent with that of DHg. Previous studies have revealed that Hg in the glacier-fed meltwater/river water in glacierized regions over Tibetan Plateau presented significant diurnal variations, with high Hg concentrations during high flow periods. The diurnal variations of Hg were consistent with that of the runoff, suggesting that runoff played an important role in the temporal distribution of Hg, and glacier ablation intensity had a strong influence on the Hg concentration in river water by dominating the release of Hg from the glacier to its downstream areas (Guo, 2012; Wang et al., 2012b). Consistent with previous studies, our results may further support the previous finding that runoff was the dominant factor affecting the DHg concentrations in the glacier-fed river water in the Tibetan Plateau (Fig. 5). Namely, when the runoff increased, the glacier meltwater flushed accumulated Hg from the glacier surface into its downstream areas (Fain et al., 2008). Moreover, the disturbance of river sediments by the runoff would result in the resuspension of particulates and accelerate the release of DHg from sediment pore waters (Ullrich et al., 2001), which lead to the relatively higher DHg concentrations in the river water in our study.

Although significantly higher runoff (3.3 m<sup>3</sup>/sec) was observed at approximately 18:00 pm compared to that measured at 16:00 pm (2.1 m<sup>3</sup>/sec), the average DHg concentrations during the two sampling periods were almost equivalent (1 ng/L). The increase of DHg concentration with increasing runoff appeared to have been hindered by the high flow of the runoff, which was further supported by correlation analysis between the two parameters (Fig. 6). The DHg concentrations in the river water of the middle river and the runoff were exponentially correlated during the daytime ( $R^2 = 0.7$ ,  $p < 0.01$ ). This phenomenon can be explained by the fact that elevated runoff accelerated the release of particulates from the glacier, which led to a higher amount of suspended particulates in the river water and enhanced the



**Fig. 3 – Spatial distribution of THg in the surface water of Da Qiangyong Lake and correlation between THg concentrations and water depth of the sampling sites, displaying the decreasing trend from the shore to the center of Da Qiangyong Lake. The trendline was derived from linear fitting by the data of all sampling points. The error bars are the standard errors (SEs) of THg concentrations in sampling points with the same water depth.**

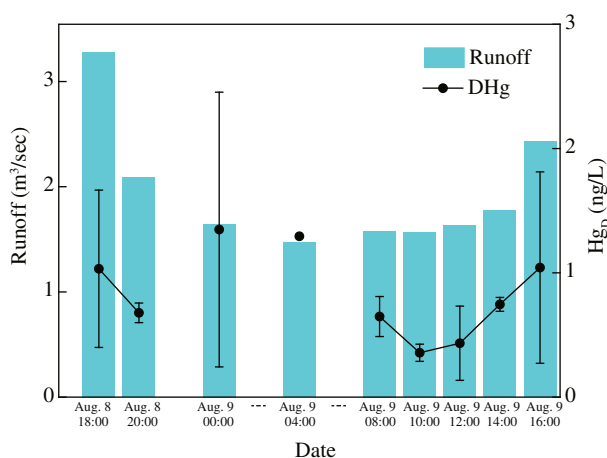


**Fig. 4 – Diurnal variations of DHg concentrations and some physico-chemical parameters (i.e., pH, temperature, conductivity) in the river water over Qiangyong Glacier Basin (a: Glacial terminus river; b: Middle river), showing low DHg concentrations in the morning (8:00 am–14:00 pm) and high DHg concentrations in the afternoon (16:00 pm–20:00 pm). DHg: dissolved Hg.**

adsorption of Hg under high flow conditions (Bonzongo et al., 1996; Ullrich et al., 2001).

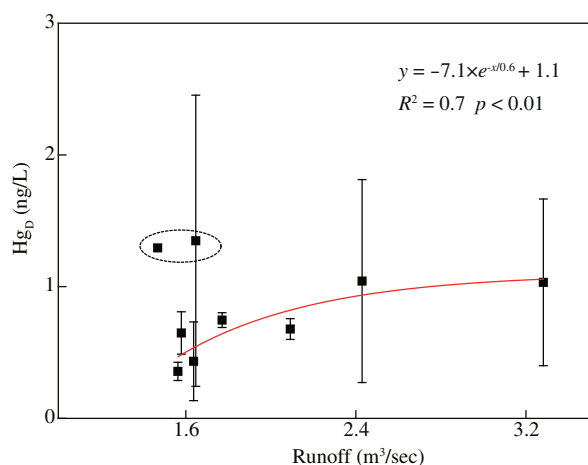
Previous studies on the Hg distribution in glacier-fed river water over the Tibetan Plateau have shown that PHg dominated the Hg species in the river water, and the proportion of PHg to THg increased with the elevation of

runoff, which indicated that the adsorption of Hg onto the particulates was enhanced under high flow conditions (Guo, 2012; Wang, 2012b; Zheng et al., 2010). TSP content in the glacier-fed melt/river waters in glacierized areas over the Tibetan Plateau showed significant diurnal variations, with high contents in the periods when the glacier dramatically melted. The high TSP contents were consistent with those of elevated discharge and THg concentrations in the river water, indicating that the meltwater could flush more particulates from the glacier surface and bring them downstream when the glacier dramatically melted (Guo, 2012; Wang, 2012b). Similarly, high TSP content most probably occurred in the glacier-fed river water in the Qiangyong Glacier Basin under high flow conditions given the fact that dust particles deposited on the Qiangyong Glacier were the main sources of river sediments in Da Qiangyong Lake (Li et al., 2011) and would be released to its downstream under high flow conditions. Moreover, the resuspension of the sediments on the riverbed also contributed larger amounts of particulates to the river water under high flow conditions. As Hg has a high tendency to be adsorbed onto surfaces, large amounts of Hg would be adhered to the particulates, which would scavenge the DHg in the river water (Jiang et al., 2003; Paraquetti et al., 2004; zhang et al., 2010b). This mechanism may be particularly significant during the Hg transportation in the Qiangyong Glacier Basin, as Hg is preferentially adsorbed in the neutral to alkaline pH range onto Fe oxides- and clay mineral-rich particulate matter, which has been found in river/lake water in the Qiangyong Glacier Basin (Ullrich et al., 2001).



**Fig. 5 – Diurnal variations of runoff and its comparison with the DHg concentrations at the fixed hydrological observation cross-section in middle river. The error bars are the standard errors (SEs) of DHg concentrations at sampling points. DHg: dissolved Hg.**





**Fig. 6 – Correlation between DHg concentrations and runoff in middle river. The trendline was derived from exponential fitting of the measured data, except for the DHg concentration data in the dotted circle, which was collected during nighttime (i.e., 0:00 am and 4:00 am). The error bars for the points are the SEs for DHg concentrations at sampling times. DHg: dissolved Hg.**

The lowest runoff was observed at night (i.e., 0:00 am and 4:00 am); however, relatively high DHg concentrations were observed in rivers at night compared to those determined from 8:00 am to 14:00 pm. This phenomenon was especially true in the middle river, where the DHg concentration reached its highest value, which may attributed to the reduced adsorption of DHg by particulates and/or partly due to lower dilution of DHg in the river water during the low flow of runoff (Zhang et al., 2010b).

### 3.3. Spatial distribution and transportation of Hg in the Qiangyong Glacier Basin

Hg has a high tendency to be adsorbed on particulates (Andren and Harriss, 1975), and particles play an important role in the transportation of Hg in aquatic systems (Krabbenhoft et al., 1998; Lawson et al., 2001; Mason et al., 1999). The spatial distribution of Hg (THg and DHg) was investigated to study Hg transportation from the glacier to the lake in the Qiangyong Glacier Basin. THg decreased significantly during transportation from the glacier to the downstream areas (Fig. 2). THg concentration fell from 8.8 ng/L in the surface snow to 2.5 ng/L at the inlet of Xiao Qiangyong Lake, and further decreased to 1.1 ng/L in the outflow of Da Qiangyong Lake, which indicated that approximately 87.5% of THg found in the surface snow was removed during the transportation process. Significant decreases were also observed in DHg concentrations during transportation from the glacier to Da Qiangyong Lake. The DHg concentration decreased from 2.5 ng/L in the surface snow to 1.7 ng/L in the glacial terminus river. In the middle river, the DHg concentration decreased to 0.8 ng/L, which was approximately 32% less than that in the surface snow.

A pioneering study on the Hg distribution in glacier-fed melt water/river water in the Zhadang Glacier Basin revealed that THg concentration was dominated by PHg. Both Hg species

(i.e., THg, PHg and DHg) decreased significantly during the transportation process from the glacier to its downstream areas, whereas the proportion of PHg to THg increased during the transportation process, which indicated that DHg adsorption onto the particulates and the sedimentation of PHg were the main removal mechanisms of Hg during the transportation process in the glacier-fed basins (Guo, 2012). As described earlier, DHg adsorption onto particulates might result in the decrease of the DHg concentration in the river water over the Qiangyong Glacier Basin. Thus, the main removal mechanism of speciated Hg is likely attributed to DHg adsorption onto particulates followed by the sedimentation of PHg during the transportation process (Bonzongo et al., 1996; Jiang et al., 2005; Zhang et al., 2010b). The more particulates in the snow were released into the meltwater and the consequent increase in contact area of particulates with Hg when the snow melted, the more DHg was adsorbed onto the particulates (Li et al., 2006). The resuspension of sediments and the flush of meltwater onto the glacier surface would also evoke a high level of TSP, which would cause a decrease in the DHg concentration. Thus, both THg and DHg concentrations consequently decreased with the sedimentation of particulates during the transportation process.

Previous studies revealed that a lake/reservoir has a “trap effect” on the Hg species in water, and usually behaves as a terminal “sink” for Hg from upstream (Bai et al., 2006; Jiang et al., 2005; Lawson et al., 2001). The significant decreases of Hg concentrations observed in the water from the middle river may further support the significant “purification” and “retention” effects of the lake on Hg species in the Qiangyong Glacier Basin. Both the THg and DHg concentrations in the river water downstream of Xiao Qiangyong Lake are obviously lower than those determined upstream of the lake. The THg concentration decreased from 2.5 ng/L at the inlet to 2.0 ng/L at the outlet of Xiao Qiangyong Lake. The DHg concentration downstream of Xiao Qiangyong Lake (i.e., middle river) was approximately 50% less than that in the upstream (i.e., glacial terminus river). This phenomenon could be explained by the fact that when the inflow river emptied into the lake, the slowdown of the flow, which was attributed to increase of meltwater-carrying distance, reduced topographic slope and stable hydrodynamic conditions of the lake (Li et al., 2011), subsequently gave rise to the sedimentation of the particulates and field turbidity decreases according to our field observation. As Hg is prone to adsorb onto surfaces, a large proportion of Hg in water was adhered to particulates (Jiang et al., 2005). Thus, with the sedimentation of the suspended particulates, the “trap effect” of the lake would reduce both the DHg and THg concentrations (Jiang et al., 2005). To sum up, our study revealed that the high-elevation lakes play an important role in controlling the transportation and fate of Hg species in the Qiangyong Glacier Basin.

## 4. Conclusions

The average THg concentration in the surface snow on Qiangyong Glacier was 8.8 ng/L, which was comparable to those of other typical glaciers in the Tibetan Plateau. Analysis of Hg speciation in the surface snow on Qiangyong Glacier has

suggested that most of the Hg in surface snow was accounted for in PHg, reconfirming that atmospheric deposition of Hg over the Tibetan Plateau was primarily associated with particulate matter. Significant diurnal variations of DHg concentrations were observed in the river water, with low concentrations in the morning (8:00 am–14:00 pm) and high concentrations in the afternoon (16:00 pm–20:00 pm). The diurnal variation of Hg was exponentially correlated with that of the runoff, indicating that runoff was the dominant factor affecting the DHg concentrations in glacier-fed river water. Moreover, obvious decreases were also observed in different Hg species (i.e., THg and DHg) during transportation from the glacier to lake in the Qiangyong Glacier Basin. Hg adsorption onto particulates followed by the sedimentation of PHg during the transportation process was believed to be an important mechanism for the removal of Hg during the transportation process.

Significant decreases of DHg and THg concentrations were observed in the downstream of Xiao Qiangyong Lake, indicating that the lake could act as a “sink” to reduce the Hg concentration in the river water. The high-elevation lakes might play an important role in controlling the transportation and fate of Hg species in the Qiangyong Glacier Basin.

## Acknowledgments

This work was supported by the National Natural Science Foundation of China (Nos. 41121001, 41225002, 41571073), the Strategic Priority Research Program (B) of the Chinese Academy of Sciences (No. XDB03030504). The authors greatly appreciate two anonymous referees for their constructive comments and thoughtful suggestions. The authors also would like to thank Li Yawei and Qu Bing for the arduous sampling work.

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