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Mercury speciation and distribution in a glacierized mountain environment and their relevance to environmental risks in the inland Tibetan Plateau

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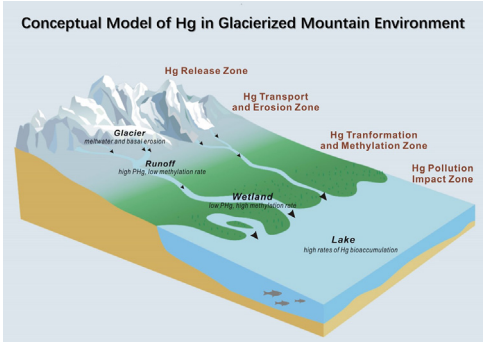
HIGHLIGHTS

- Different kinds of water samples were collected and analyzed for Hg speciation and distribution.
- Hg speciation and distribution in meltwater and runoff water samples differed from those in wetland.
- A conceptual model was proposed to illustrate Hg distribution and relevant environmental risks.
- Wetlands are a vital zone for investigating the environmental effects of Hg in glacierized basins.

GRAPHICAL ABSTRACT



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ABSTRACT

Glacierized mountain environments can preserve and release mercury (Hg) and play an important role in regional Hg biogeochemical cycling. However, the behavior of Hg in glacierized mountain environments and its environmental risks remain poorly constrained. In this research, glacier meltwater, runoff and wetland water were sampled in Zhadang-Qugaqie basin (ZQB), a typical glacierized mountain environment in the inland Tibetan Plateau, to investigate Hg distribution and its relevance to environmental risks. The total mercury (THg) concentrations ranged from 0.82 to 6.98 $ng \cdot L^{-1}$, and non-parametric pairwise multiple comparisons of the THg concentrations among the three different water samples showed that the THg concentrations were comparable. The total methylmercury (TMeHg) concentrations ranged from 0.041 to 0.115 $ng \cdot L^{-1}$, and non-parametric pairwise multiple comparisons of the TMeHg concentrations showed a significant difference. Both the THg and MeHg concentrations of water samples from the ZQB were comparable to those of other remote areas, indicating that Hg concentrations in the ZQB watershed are equivalent to the global background level. Particulate Hg was the predominant form of Hg in all runoff samples, and was significantly correlated with the total suspended particle (TSP) and not correlated with the dissolved organic carbon (DOC) concentration. The distribution of mercury in the wetland water differed from that of the other water samples. THg exhibited a significant correlation with DOC as well as TMeHg, whereas neither THg nor TMeHg was associated with TSP. Based on the above findings and the results from previous work, we propose a conceptual model illustrating the four Hg

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distribution zones in glacierized environments. We highlight that wetlands may enhance the potential hazards of Hg released from melting glaciers, making them a vital zone for investigating the environmental effects of Hg in glacierized environments and beyond.

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1. Introduction

Mercury (Hg) is a toxic heavy metal element that can remain in the air for long periods of time and can be transported through the atmosphere across long distances; thus, Hg is recognized as a global pollutant (Fisher et al., 2012; Krabbenhoft and Sunderland, 2013). In some remote areas, Hg accumulates in surface ecosystems through dry and wet deposition. Inorganic Hg in aquatic ecosystems (rivers, lakes, wetlands, etc.) can be transformed into methylmercury (MeHg) by the methylation process, becoming a highly toxic neurotoxin (Krabbenhoft and Sunderland, 2013) and ultimately possibly causing health impacts or even death in humans via bioaccumulation and biomagnification in food webs. In high-latitude and high-elevation regions, glaciers play a significant role in the retention of atmospherically deposited Hg and represent an active environmental medium in Hg biogeochemical cycling (Nagorski et al., 2014; Paudyal et al., 2017; Sun et al., 2017; Zhang et al., 2012; Zheng, 2015). Studies have revealed enhanced Hg deposition in polar snow packs (Durnford and Dastoor, 2011) and glaciers in alpine regions (Huang et al., 2014; Kang et al., 2016). Snow packs and glaciers act as transitional environments connecting the atmospheric reservoir and ecosystem uptake. During the ablation period, the majority of Hg can be remobilized into the melt water, posing potential environmental threats to organisms in downstream ecosystems (Dommergue et al., 2010; Zhang et al., 2014).

Alpine glaciers are sensitive to climate change (Beniston, 2003) and have changed dramatically in recent decades (Kang et al., 2015; Zemp et al., 2015; Zhang et al., 2015), likely causing the export of legacy contaminants within glacierized mountain environments (Sharma et al., 2015; Steinlin et al., 2016; Zhang et al., 2017). In alpine glaciers, Hg has been found predominately in the form of particulate-bound Hg, which is conducive to Hg preservation (Huang et al., 2014; Paudyal et al., 2017; Sun et al., 2017). During glacier ablation, the proportion of Hg emitted into the atmosphere is very low (Paudyal et al., 2017), and the vast majority of Hg stored in snow and ice enters the water and is then transported downstream in the runoff. In addition, the rapid recession of alpine glaciers has not only accelerated the release of Hg preserved in snow and ice but has also promoted the release of Hg in periglacial areas via the erosion of soils by glacier-fed runoff in glacierized mountain environments. Mountain glaciers are considered to have a higher Hg release efficiency than polar glaciers (Sun et al., 2017) and act as active components in Hg cycling in mountain environments. In addition to the release and mobilization of Hg, receding alpine glaciers can affect the hydrology of downslope ecosystems, leading to terrain changes within glacierized watersheds. For example, melting glaciers are recognized to have significant effects on wetland areas in glacierized basins (Dangles et al., 2017; Polk et al., 2017).

To date, although some works have quantified the amount of Hg released by melting glaciers and revealed related mechanisms, underscoring the efficient and important role of melting glaciers in mobilizing Hg within glacierized watersheds, knowledge of the behavior of Hg released in glacierized environment, particularly its potential environmental risks, remains limited. This represents a major gap for further assessment of the environmental effects of Hg derived from the melting of alpine glaciers. In this study, three different kinds of water samples, including glacier meltwater, glacier-fed runoff and wetland water, were collected in a typical glacierized catchment in the inland Tibetan Plateau. The samples were analyzed for Hg species and selected water chemical parameters. The goal of this research is to elucidate Hg distribution in the waters and the related influencing factors in glacierized environments in order to reveal the environmental risks of Hg associated with the melting of mountain glaciers.

2. Materials and methods

2.1. Study area

The study area, the Zhadang-Qugaqie Basin (ZQB), is located to the east of the Nyainqentanglha Mountains (NM) in the south-central Tibetan Plateau. As a transition zone from maritime glaciers in southeastern Tibet to continental glaciers in the northern Tibet (Gao et al., 2015), the NM region represents one of the highest concentrated regions of modern glaciers in China. Covering an area of 59.6 km^2, the ZQB is located on the northeastern side of the summit of the NM and contains the Zhadang glacier and the Qugaqie River, which is 15 km in length. The Qugaqie River is fed by the Zhadang glacier and drains into Lake Nam Co. The Zhadang glacier, in the upstream drainage area, covers an area of 1.9 km^2 (Li et al., 2014). The temperature and precipitation of this region exhibit significant seasonal variation and are influenced by the continental climate and the Indian monsoon (Li et al., 2014). Precipitation is mainly concentrated in summer and autumn and is relatively limited in winter and spring. >90% of precipitation occurs between June and September, when the ablation is greatest (Zhang et al., 2013). In the context of global warming, the glacier retreat in the NM area has been dramatic, and between the 1970s and 2007, the glacier area in the NM region decreased by 37.1 km^2, accounting for 18.2% of the total glacier area (Chen et al., 2009).

The ZQB mainly consists of morainic, periglacial and aeolian landforms. The glacial peripheral area is dominated by morainic landform where freezing and thawing influences are very strong in the downriver area. Due to the effects of frost processes and gravity, periglacial landforms are widely developed and cover 60% of the land surface in the ZQB (Li et al., 2014). The aeolian landforms, consisting of fine and medium sands, are found near the lake coast because of glacier retreat and the consequent increase in newly exposed surfaces. The land cover is composed of bare ground (50.0%), grassland (34.9%), wetland (6.6%), water body (0.1%) and glacier (8.4%) (Li et al., 2014; Ran et al., 2009). Wetlands exist along the Qugaqie river and feature vegetation coverages of >70% (Tian et al., 2009), producing a large amount of organic matter in the basin.

2.2. Sample collection

Glacier meltwater, runoff and wetland water samples were collected from the ZQB in this research (Fig. 1, Table 1, $n = 56$). The glacier meltwater samples included both supraglacial and proglacial river water samples. The runoff samples were collected at upstream and downstream sites along the Qugaqie River. The wetland water samples were collected from midstream and downstream sites in the ZQB. Total mercury (THg) and total methylmercury (TMeHg) samples were collected with polypropylene BD Falcon® centrifuge tubes spiked with 250 μL of BV-III grade (CMOS) HCl (Beihua Chemical, China). Sampling personnel used polyethylene gloves and strictly followed the "Clean Hands-Dirty Hands" protocol during the sampling (Fitzgerald, 1999; Sun et al., 2017). Falcon® polypropylene centrifuge tubes were rinsed >3 times during sampling, and surface water was collected to avoid touching the mud at the bottom. Additionally, 10% of the samples were field blanks inserted into sampling process; for these blank samples, the same volume of prepared ultra-pure water was poured into

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