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Insights into mercury in glacier snow and its incorporation into meltwater runoff based on observations in the southern Tibetan Plateau

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ABSTRACT

The Tibetan Plateau (TP) is recognized as "Water Tower of Asia". Yet our understanding of mechanisms influencing migration of mercury (Hg) during snowmelt and its incorporation into freshwater in mountain glaciers on the TP remains quite limited. Extensive sampling of environmental matrices (*e.g.*, snow/ice) were conducted on the East Rongbuk (ER) glacier on Mt. Everest and Zhadang (ZD) glacier on Mt. Nyainqentanglha for Hg speciation analysis. Speciated Hg behaved quite different during snowmelt: a preferential early release of DHg (dissolved Hg) was observed at the onset of snowmelt, whereas PHg (particulate-bound Hg) and THg (Total Hg) become relatively enriched in snow and released later. Small fraction of Hg in snow was lost during a snowmelt day (18.9%–34.7%) with a large proportion (58.1%–87.3%) contributed by PHg decrease, indicating that the deposited Hg is most likely retained in glacier snow/ice. Furthermore, THg were positively correlated with PHg and crustal major ions (*e.g.*, Ca^{2+}, Mg^{2+}) during snowmelt, indicating that Hg is mainly migrated with particulates. The main pathway of Hg loss during snowmelt was most probably associated with release of PHg with meltwater, which was greatly influenced by ablation intensity of snow/ice. This should be paid particular concern as Hg preserved in mountain glaciers will mostly enter aquatic ecosystem as climate warms, impacting on downstream ecosystems adversely. Obvious decrease of THg during the downstream transport from glacier was observed with a large proportion contributed by PHg decrease. The main removal mechanism of Hg was associated with sedimentation of PHg during the transport process.

Introduction

Mercury (Hg) is well-known as a globally hazardous pollutant because of its potential for long-range transport *via* the atmosphere, persistence and biomagnification through food chains (Selin, 2009). Mercury mainly exists in atmosphere in three forms: gaseous elemental Hg (Hg^0), divalent reactive gaseous Hg (Hg(II)), and particulate-bound Hg (PHg). Reactive

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gaseous Hg and PHg have relatively high dry deposition velocity and water solubility and are more readily to be scavenged from atmosphere *via* wet/dry deposition. Gaseous elemental Hg is the predominant form of Hg in atmosphere (>90%) and has a long atmospheric residence time of 0.5–2 years, which enables Hg to undergo a long-range transport before being oxidized to Hg(II) and being deposited to remote areas (Schroeder and Munthe, 1998).

With an average elevation above 4000 m above sea level (a.s.l.), the Tibetan Plateau (TP, including the Himalayas), or the "Third Pole", is one of the most remote and pristine areas in the world and is the most glacier-concentrated region at low- and mid-latitudes (Yao et al., 2012). Pollutants originating from heavily-polluted areas surrounding it (*e.g.*, central and southern Asia) could be transported a long distance and eventually deposited onto glaciers over the TP (Beal et al., 2015; Kang et al., 2016). The accumulated pollutants will be largely released as glacier snow/ice melting accelerates due to global warming, impacting on human health and ecosystems of downstream regions adversely (Sun et al., 2016; Zhang et al., 2017). This issue is of particular concern as TP serves as the source region of Asia's major rivers (*e.g.*, Yangtze River, Indus and Brahmaputra) (Yao et al., 2012), and glacier melt in the TP is an important contributor to river discharge of these rivers (*e.g.*, 48% of annual river discharge for Indus river basin; Immerzeel et al., 2010). To date, some previous studies focused on spatiotemporal distribution and deposition of Hg in glacier snow on the TP have been conducted (Huang et al., 2012, 2012; Loewen et al., 2007; Q. Zhang et al., 2012). For instance, previous studies have revealed that concentration levels of Total Hg (THg) in glacier snow over western China (*i.e.*, Tibetan Plateau and northwest China) were higher in northern region compared to those from southern region, and higher in non-monsoon season than in monsoon season. The spatiotemporal variability of THg concentrations in glacier snow was consistent with that of atmospheric particulate loadings, suggesting that deposition of atmospheric Hg over western China was primarily associated with particulate matter and dust storm activity might provide the largest source of Hg deposition on the TP. However, it is still unclear Hg is transported long distance with dust or partitioning onto particulates nearby the deposition sites (Loewen et al., 2007; Q. Zhang et al., 2012). For spatial distribution of Hg in snow of an individual glacier, a previous study reported an increase in THg with increasing altitude in surface snow of four high-altitude glaciers over the TP, and proposed a possible altitude magnification effect on Hg deposition in alpine snow (Huang et al., 2012).

However, a knowledge gap in this region was the lack of information on the fate of Hg following its deposition onto glacier snow/ice. For instance, a part of deposited Hg could probably be remitted into atmosphere *via* a sunlight–induced mechanism (Lalonde et al., 2003); Some part of Hg might be released with meltwater and contaminate aquatic ecosystem (Dommergue et al., 2003). To date, it's still unclear what the main pathway of Hg loss and mechanisms influencing migration process of speciated Hg during snowmelt in glacier snow on the TP is. Three major loss mechanisms of Hg in snow have been proposed during post-depositional processes, included 1) percolation, 2) settling of PHg and 3) photo-induced reduction of Hg(II), which may then be emitted back to the atmosphere as Hg^0 (Lalonde et al., 2002). Previous studies in the Arctic (Poulain et al., 2004; Dommergue et al., 2009) and some mid-latitude sites (Lalonde et al., 2002, 2003; Poulain et al., 2007) have revealed that a significant fraction of deposited Hg might probably not enter the aquatic/terrestrial environment due to the subsequent photoreduction of Hg(II) in snow, which could result in >40% loss of THg concentration in surface snow within 24 hr (Lalonde et al., 2003). However, deposition of Hg over the TP is driven primarily in the form of PHg (Loewen et al., 2007). The deposited PHg in glacier snow/ice over the TP could be far less reducible than deposited Hg(II) in redox reactions, and thus less influenced by the photoreduction process (Durnford and Dastoor, 2011; Loewen et al., 2007). This could be evidenced by most high THg concentration peaks corresponded to high particulate loads in snowpits, and significant correlations between insoluble particles and THg concentrations in snowpits and ice core of the TP (Q. Zhang et al., 2012; Kang et al., 2016). Consequently, post-depositional release of Hg *via* the photoreduction and reemission should be not significant in glacier snow of the TP. Instead, Hg stored in glacier snow of the TP would be most likely released with meltwater after its deposition due to snow/ice melt. Studies on post-depositional fate of Hg were required in order to understand the relationship between Hg deposition onto snow and its incorporation into snowmelt and freshwater, which is essential for assessing its potential downstream impact and biogeochemical cycles of Hg in high-elevation glacier basins on the TP.

In this study, concentrations of speciated Hg in various environmental matrices (*i.e.*, glacier snow/ice, lake/stream water) are presented. Migration process of speciated Hg during snowmelt and downstream transport of Hg from glacier were discussed. This study is an attempt to provide insights for assessing post-depositional fate of Hg in snow and potential environmental risk of Hg release by accelerated glacier melt to glacial-fed downstream ecosystems on the TP as climate warms.

1. Material and methods

1.1. Study area and sampling location

The Himalayas is the highest mountain ranges on Earth acts as a natural barrier between the TP and Indian subcontinent. The East Rongbuk (ER) glacier (28.02°N, 86.96°E) is located on the north-eastern slope of Mt. Everest in the central Himalayas (Fig. 1), with a length of 14.0 km, a width of 0.8 km and a total area of 48.45 km^2. The altitude of glacial terminus is approximately 5540 m a.s.l. (T. Zhang et al., 2012). The equilibrium line altitude (ELA) of ER glacier is estimated to be 6419 m a.s.l. (King et al., 2017). According to air temperature records at 6000 m a.s.l of ER glacier, the annual mean temperature is −5.4°C with the extreme high temperature of 18.6°C and extreme low temperature −29.3°C. The daily mean temperature is above 0°C from June to August (Yang et al., 2011). The mass balance of ER glacier between 2000 and 2015 was −0.44 ± 0.19 m water equivalent (w.e.)/yr with the mean surface lowering rates of −1.04 ± 0.18 m/yr in glacier ablation zones (King et al., 2017). Landscape in the glacier basin consists of glaciers, rivers and lakes (Fig. 1). A proglacial lake formed at 5214 m a.s.l. and is approximately 3 km from the

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