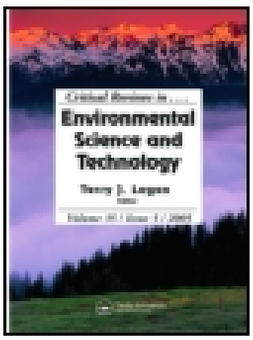

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Mercury biogeochemistry over the Tibetan Plateau: An overview

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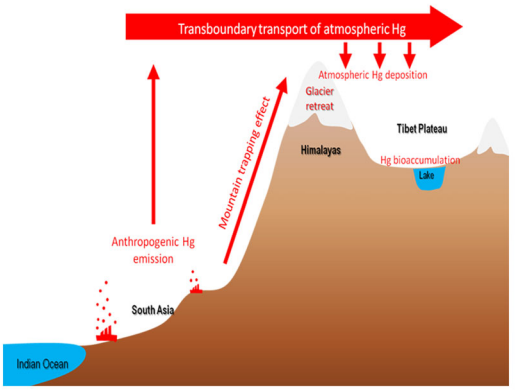
ABSTRACT

Since the beginning of the 21st century, our knowledge of the sources and fate of Hg over the Tibetan Plateau (TP), the "Roof of the World," has been greatly enhanced with growing literature. Here, we reviewed the available literature to obtain a comprehensive understanding of Hg biogeochemistry over the TP. The biogeochemical Hg cycling is characterized by the following features: (1) There are existing but limited local emission sources of anthropogenic Hg in the TP. The Indian Summer Monsoon is an important transporter of atmospheric Hg pollution into the inland TP; (2) "Cold trapping effect" plays an important role in the atmospheric Hg deposition over the TP. Glacier, vegetation, and soil act as important "sinks" of atmospheric Hg pollution; (3) Enhanced anthropogenic activities around the TP, climate warming and glacier melting have the potential impacts to affect the behavior and distribution of Hg; (4) Significant bioaccumulation of MeHg (>100 ng/g) has been found in the Tibetan aquatic food chains. Considering that transboundary transport is responsible for the widespread Hg pollution in the TP, international/regional collaborations regarding Hg emission regulations are needed to reduce the migration of Hg and to mitigate adverse Hg pollution impacts on the TP.



KEYWORDS Mercury; Tibetan Plateau; transboundary pollution

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1. Introduction

Mercury (Hg) is a contaminant of primary concern in the global environment because the atmosphere plays a crucial role in the transport and geochemical cycling of Hg (Driscoll, Mason, Chan, Jacob, & Pirrone, 2013; Beckers & Rinklebe, 2017). Hg is emitted into the environment through natural sources (e.g., volcanic/geothermal activities, wildfires, and biomass burning) and anthropogenic activities (e.g., fossil fuel combustion, non-ferrous metal mining) (Pirrone et al., 2010). Anthropogenic activities have largely promoted the emission of Hg by a factor of 3-5 since the Industrial Revolution in the 1800s (Drevnick et al., 2016; Kang et al., 2016; Schuster et al., 2002). The majority of Hg emitted into the atmosphere is in the form of gaseous elemental Hg(0). This Hg(0) species have a long atmospheric lifetime (0.5-2 years), which allows hemispheric to global transport and dispersion prior to deposition (Amos, Jacob, Streets, & Sunderland, 2013). Hg(0) is removed from the atmosphere through (1) oxidation by ozone, hydroxyl radical and halogen radicals as Hg(II) (Schroeder & Munthe, 1998), or (2) uptake by vegetation (Obrist et al., 2017; Wang, Bao, Lin, Yuan, & Feng, 2016). Once deposited to terrestrial and aquatic ecosystems, Hg is susceptible to transformation into methyl-Hg (MeHg), a neurotoxin, which can bioaccumulate into humans and wildlife (Gilmour, Henry, & Mitchell, 1992). High levels of MeHg have been observed in food chains of various aquatic environments, and fish consumption is the route of MeHg exposure to the global population (Morel, Kraepiel, & Amyot, 1998).

At present, even the world's most remote sites (e.g., polar regions) are facing a great challenge to Hg pollution (Bargagli, Monaci, Sanchez-Hernandez, & Cateni, 1998; Dietz, Outridge, & Hobson, 2009). A substantial amount of Hg is carried into the North Pole (Arctic) and South Pole (Antarctic) via long-range transport by air and water currents from regions of intensive anthropogenic activities located at lower latitudes (Cole et al., 2013; Temme, Einax, Ebinghaus, & Schroeder, 2003). The polar regions are thought to be an important global sink for atmospheric Hg, in particular, during the atmospheric depletion events (AMDEs) in the springtime (Ariya et al., 2004; Schroeder & Munthe, 1998). During the AMDEs, a series of photochemical reactions between ozone and halogens create reactive halogen species such Br radicals that oxidize gaseous Hg(0) to reactive Hg(II), which is quickly removed from the atmosphere and results in large deposition fluxes onto snow surface and tundra lands (Ebinghaus et al., 2002). Hg in the polar environment also enters food chains, resulting in high levels of MeHg in Arctic top predators (e.g., polar bears, birds, whales) and MeHg biomagnification from lower trophic food chains (Atwell, Hobson, & Welch, 1998). This poses serious health risks for Arctic indigenous peoples, who rely on subsidence hunting and fishing (Dewailly et al., 2001).

The Tibetan Plateau (hereafter, TP), known as the "Third Pole" and "Water tower of Asia," has one of the most imposing topographic features on the surface of the Earth. The TP covers ~26% of the land ($\sim 2.5 \times 106\,km^2$) and <1% of the total population of China, with an average elevation of >4000 m above sea level (a.s.l.) (Kang et al., 2019; Yao et al., 2012). Therefore, TP has long been believed as a remote, isolated, and fragile ecosystem. Unlike the polar regions, the TP is situated in mid-latitudes and located in the immediate vicinity of some of the most polluted areas in the world, such as South Asia, East Asia, and Southeast Asia. In the last few decades, rapid industrialization in these areas has released a massive amount of Hg and other air pollutants (e.g., black carbon, persistent organic pollutants (POPs)) into the atmosphere (Yang, Zhang, Li, Jiang, & Jing, 2013). In the past, the remoteness and high elevation of the TP were believed to protect the TP from air pollution that haunts the densely populated regions of China and South Asia. However, owing to its volatility, Hg can migrate from relatively lower and warmer regions to higher and colder regions through "grass-hopping" and "cold trapping" effects (Huang, Kang, Zhang, Jenkins, et al., 2012; Loewen et al., 2005; Zhang, Yin, et al., 2013). Moreover, the atmospheric circulation pattern of the TP was characterized by the Indian monsoon in the summer and the westerlies in the winter (Figure 1). Such a climate regime could exert a profound impact on the transport of atmospheric Hg to the TP. Growing evidence has indicated that the TP may act as a sink of atmospheric pollutants (Kang et al., 2019), but there are still emerging scientific issues that need to be addressed as follow. What types of environmental and anthropogenic factors control the transport of Hg into the TP? What is the extent of atmospheric Hg deposition to the TP? What are the present and future environmental impacts of Hg in the TP?

More importantly, Hg is a redox-sensitive metal, and its biogeochemical cycling is particularly susceptible to environmental changes (Krabbenhoft & Sunderland, 2013; Point et al., 2011; Stern et al., 2012; Yin, Feng, Hurley, Krabbenhoft, Lepak, Kang, et al., 2016). Growing evidence suggested that montane regions are experiencing more rapid environmental changes than those in lower elevations (Song, Huang, Richards, Ke, & Phan Hien, 2014). For instance, the TP has undergone a dramatic rise in temperature, which is as twice as faster than the environment at lower elevations (Zhang, Yao, et al., 2014), and the annual precipitation rate in the TP has also significantly increased (Li et al., 2013; Liang, Chen, Jin, Wan, & Gong, 2007). Climate warming has promoted glacier melting in the TP (Song et al., 2014), which may facilitate the release of legacy Hg from glaciers into lakes and rivers. As these aquatic ecosystems are hotspots for Hg methylation and bioaccumulation (Gilmour et al., 1992; Morel et al., 1998), the bioaccumulation of Hg in the TP food webs is expected to change, and the risk of Hg exposure to the TP population may suffer from these changes.

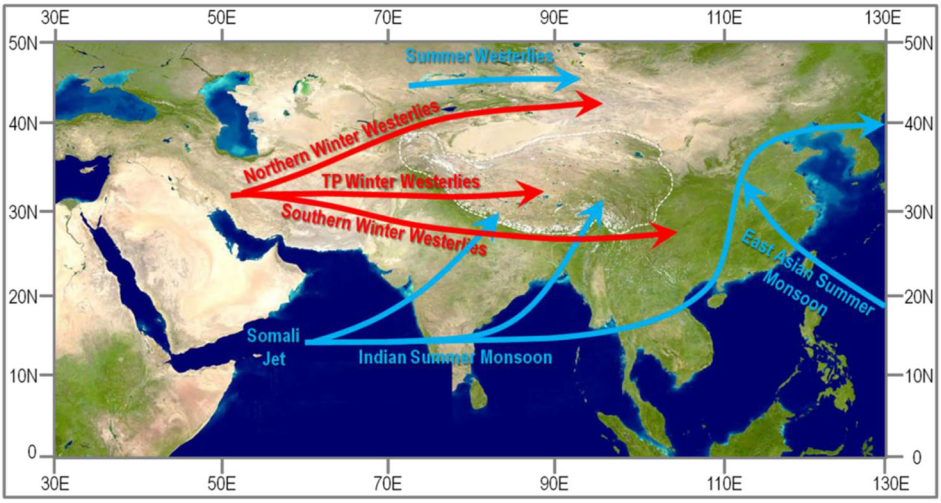

Figure 1. The terrain of the Tibetan Plateau and its surroundings. Major atmospheric circulation systems around the Plateau are shown, with blue arrows for summer and red arrows for winter (modified after Yang et al. (2014)).

Since the beginning of this century, our knowledge of the geochemical fate of Hg over the TP has been greatly enhanced with the growing literature. To our knowledge, more than 70 studies have investigated the fate of Hg in a variety of environmental samples in the TP and produced a tremendous amount of data over the last decade, providing an opportunity to obtain a comprehensive understanding of Hg biogeochemistry of the TP. Therefore, this work aims to integrate the published data and already-acquired knowledge and to critically review the biogeochemical Hg cycling in the fragile ecosystems of TP. This study highlights the environmental risk of Hg in the TP, which could facilitate targeted assessments and effective mitigation strategies regarding the enhanced Hg pollution on the TP.

2. Sources of Hg to the TP

2.1. Local anthropogenic Hg sources

Due to its sparse human population and minimal industrial activities, the local anthropogenic Hg emission was thought to be limited. However, a recent study by Liu et al. (2017) showed that the per capita release of total Hg from municipal sewage in Tibet is the highest in China. The highest Hg levels of municipal sewage in the TP ($> 10\,\mu g\ L^{-1}$) were later ascribed to the common use of traditional medicines (TTMs) by Tibetans. TTMs have been shown to contain large quantities of inorganic Hg (10^3 to $10^5\,\mu g\ g^{-1}$) because pharmacists have intentionally added metallic Hg into medicine as therapeutic ingredients. For $>$ 1000 years, Tibetan nobilities have ingested daily Hg containing pills, and now, most TP populations have access to these expensive pills.

Inorganic Hg tends to be excreted rapidly from the human body, which may lead to high levels of inorganic Hg in the municipal sewage in Tibetan urban areas (Liu, He, Baumann, et al., 2018). Although there is no MeHg data on Tibetans, a recent study has estimated that a total of ~3.6 tons of Hg was released from the human body into the local environment as a result of TTMs ingestion in the year 2015 alone (Liu, He, Ge, et al., 2018).

Biomass burning (mainly yak feces) is another local Hg emission source in the TP. For centuries, nomadic people have herded yaks. As pack animals, yaks not only supply meat, milk, and fiber for fabrics but also generate heating fuel in the form of feces. Even nowadays, yak feces is the major fuel for Tibetans. According to Long, Wiener, Jianlin, and Ruijun (2003), an adult yak consumes 17.5 kg dry matter (DM) per day and yield 5.25 kg DM per day or 1916 kg per year. A family in the TP can use about 22 000 kg of dry yak feces per year (Long et al., 2003). Yak feces combustion could emit dangerous levels of aerosols into the atmosphere, and especially in the indoor areas. Chen, Kang, Bai, Sillanpää, and Li (2015) reported that aerosols emitted from yak feces combustion are characterized by enriched heavy metals (e.g., Cd, Zn, and Pb). Although the total amount and distribution of Hg emitted from yak feces combustion in TP remain poorly studied, the influence of local Hg emission from yak feces combustion cannot be ignored. During combustion, it is presumed that a substantial amount of Hg is released in conjunction with other heavy metals (Chen et al., 2015). Further studies on Hg concentration in yak feces and Hg emission factors are necessary to estimate the emission of Hg during the combustion of yak feces.

Due to its chalcophilic nature, Hg is found in abundance in sulfide minerals (e.g., galena, sphalerite, and chalcopyrite) in hydrothermal deposits (Yin et al., 2012; Yin, Feng, Hurley, Krabbenhoft, Lepak, Hu, et al., 2016). Since 1999, Chinese geologists have discovered more than 600 new sites of copper, iron, lead and zinc ore deposits on the TP, and preliminary estimates show the plateau has 30 million to 40 million tons of reserves for copper, 40 million tons of lead and zinc and billions of tons of iron (Kezia, 2011). However, these mineral deposits have been rarely explored and mined to date, due to China's priority for environmental protection in the TP. Hg emitted from potential local mining and refining is still very limited in the TP.

2.2. Transboundary transport of Hg

Due to the rapid economic development in the past decades, South Asian and Southeast Asian countries have become one of the world's largest anthropogenic sources of air pollutants (Burger Chakraborty, Qureshi, Vadenbo, & Hellweg, 2013; Mukherjee, Bhattacharya, Sarkar, Zevenhoven, & Bodaly, 2009). As shown in Figure 1, the large-scale atmospheric

circulation patterns over the TP are dominantly governed by the Indian summer monsoon (ISM, June to September) and the westerlies (October to May). Since East Asia is located in the downwind of the Indian monsoon and the Westerlies (Figure 1), anthropogenic Hg emissions from this region are believed to have a very limited impact on the TP. The ISM not only brings massive moisture but also carries a substantial supply of atmospheric contaminants from South Asia and Southeast Asia to the TP, as supported by the increased transport of black carbon (Cong et al., 2013; Li et al., 2016) and Persistent Organic Pollutants (Yang, Xie, et al., 2016) during the ISM season. However, the westerlies are thought to bring dry air and relatively fewer contaminants as they originate and travel far away from the upwind sources such as Europe and the Middle East (Huang et al., 2019).

We recognize that transboundary transport plays an important role in regulating the distribution of atmospheric Hg in the TP. The total gaseous Hg (TGM) concentrations at 6 stations over the TP, as shown in Figure 2, were reported by several studies (Fu, Feng, Zhu, Wang, & Lu, 2008; Fu et al., 2012; Lin et al., 2019; Yin et al., 2018; Zhang, Fu, et al., 2015; Zhang et al., 2016). The average TGM concentrations were 2.55 ± 0.73 ng m^{-3} at Shangri-La (Zhang, Yao, et al., 2015), 2.09 ± 0.63 ng m^{-3} at Mt Ailao (Zhang et al., 2016), 1.98 ± 0.98 ng m^{-3} at Waliguan (Fu et al., 2012), 1.42 ± 0.37 ng m^{-3} at Qomolangma (Lin et al., 2019) and 1.33 ± 0.24 ng m^{-3} at Namco (Yin et al., 2018), which are within or higher than the global background TGM concentrations (1.1-1.3 ng m^{-3} and 1.5-1.7 ng m^{-3} in the Northern and Southern Hemisphere, respectively). At 4 stations (Qomolangma, Shangri-La, Mt Ailao and Namco) located more closely to South Asia and Southeast Asia, there was an increase in TGM concentrations during the ISM. Compared to the non-ISM period, TGM concentrations during the ISM period increased by 12% and 10% at Mt. Ailao and Mt. Qomolangma, respectively (Lin et al., 2019; Zhang et al., 2016). At Shangri-La, three high TGM events (2-6 ng m^{-3}) were observed during the ISM season (Zhang, Fu, et al., 2015). At Namco, the seasonal variation of TGM was characterized by higher concentrations during warm seasons and lower concentrations during cold seasons, decreasing in the following order of summer (1.50 ± 0.20 ng m^{-3}) > spring (1.28 ± 0.20 ng m^{-3}) > autumn (1.22 ± 0.17 ng m^{-3}) > winter (1.14 ± 0.18 ng m^{-3}) (Yin et al., 2018). Overall, the close relationship between TGM and the ISM together with the difference in seasonal TGM patterns clearly indicated that the transboundary transport of Hg from South Asian and Southeastern Asian countries to the TP. At the Waliguan station, which is located at the Northeastern TP, the TGM concentrations were also thought to be influenced by the winds from South Asia (Fu et al., 2012), although this station was reported to receive some Hg from inland China. As shown in Figure 2, the airmass

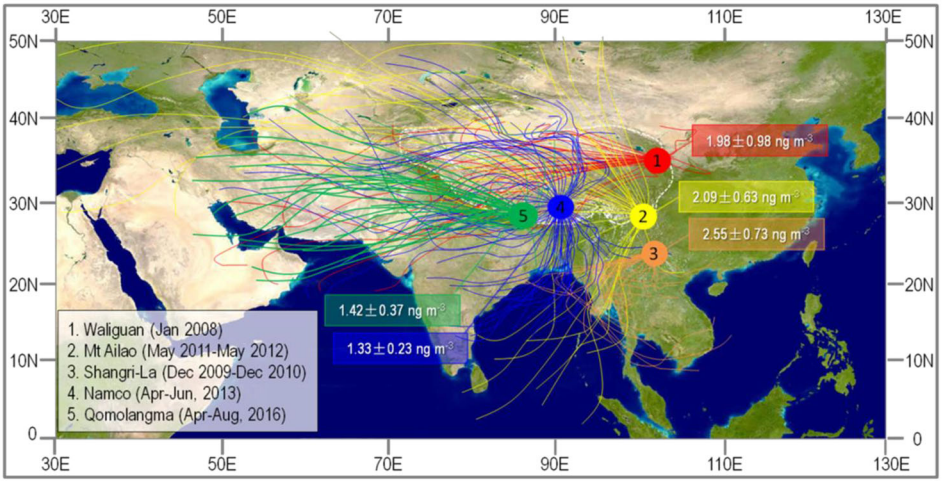

Figure 2. TGM concentrations and backward trajectories ended at 5 stations in the Tibetan Plateau (Fu et al., 2012; Lin et al., 2019; Yin et al., 2018; Zhang, Fu, et al., 2015; Zhang et al., 2016).

back-trajectories analysis clearly suggested that the transboundary transport of Hg from South Asia and Southeast Asia may be responsible for both the transport and deposition (discussed further below) of atmospheric Hg at all these stations (Fu et al., 2012; Lin et al., 2019; Yin et al., 2018; Zhang, Fu, et al., 2015; Zhang et al., 2016). Westerlies in wintertime may also bring Hg into TP. However, no information is available at this time regarding the transport of Hg by Westerlies.

3. Atmospheric Hg deposition

Atmospheric Hg consists of gaseous elemental Hg (GEM), gaseous oxidized Hg (GOM), and particulate-bound Hg (PBM). GEM is the predominant species and usually accounts for $>$ 95% of total atmospheric Hg. GOM and PBM are found in much lower concentrations, often not more than 1–5% of the total atmospheric Hg (Schuster et al., 2002). Mercury can be removed from the atmosphere mainly via wet and dry deposition (Zhang, Fu, et al., 2013; Jiskra et al., 2018). Wet deposition of Hg is the air-to-surface flux in the form of precipitation, which scavenges mainly GOM and PBM from the atmosphere. Dry deposition is Hg flux in the absence of precipitation and is believed to include all three Hg phases. Here, we synthesized the current understandings on the distribution of Hg in precipitation and glacier snow over the TP, which further supports a significant influence of transboundary atmospheric Hg transport over the TP.

3.1. Dry deposition

The dry deposition flux of Hg in the TP is difficult to quantify due to the lack of direct and accurate measurements. Only one study has demonstrated that the dry deposition flux of Hg can be up to 35.2 μg $m^{-2}yr^{-1}$ in Lhasa, the largest city of Tibet, indicating that dry deposition may act as an important process for the atmospheric Hg loadings to the TP. To date, the exact source of dry deposited Hg in the TP remains poorly understood. The ISM has been proven as the main driver delivering black carbon from the South Asian continent to the TP (Li et al., 2016). Similarly, it is believed that monsoons can transport a substantial amount of PBM from surrounding regions (e.g., South Asia and Southeast Asia) to the TP, as much higher dry deposition flux of Hg was observed in the Kathmandu Valley of Himalayan foothill region (135 μg $m^{-2}yr^{-1}$, according to Guo et al. (2017)).

3.2. Precipitation

The total Hg (THg) concentrations of precipitation collected at several sites (Figure 3) have been previously investigated (Huang, Kang, Zhang, Yan, et al., 2012; Huang et al., 2013, 2015; Tripathee et al., 2019a, 2019b). The THg and MeHg concentrations were variable at Namco (THg: 4.8 ng L^{-1}; MeHg: 0.031 ng L^{-1}), Lhasa (THg: 24.8 ng L^{-1}), the SET station (THg: 4.0 ng L^{-1}; MeHg: 0.112 ng L^{-1}) and four stations in Nepal such as Kathmandu (THg: 19.8 ng L^{-1}), Dhunche (THg: 8.0 ng L^{-1}), Dimsa (THg: 7.1 ng L^{-1}) and Gosainkunda (THg: 6.5 ng L^{-1}). These values are much lower than those observed in the polluted regions (e.g., metropolitan cities). The majority of Hg in precipitation from these stations is in the form of particulate-Hg (PHg) (47.4-77%), indicating that the scavenging of particulate-Hg from the atmosphere is an important mechanism contributing Hg to precipitation. Biomass and fossil fuel combustion in South Asia and Southeast Asia can contribute to atmospheric particulate-Hg to the TP (Guo et al., 2017; Zhang, Fu, et al., 2014).

The wet THg deposition fluxes were calculated to be 1.75 μg $m^{-2}yr^{-1}$ at Namco, 8.2 μg $m^{-2}yr^{-1}$ at Lhasa, 3.9 μg $m^{-2}yr^{-1}$ at the SET station, 34.9 μg $m^{-2}yr^{-1}$ at Kathmandu, 15.9 μg $m^{-2}yr^{-1}$, respectively. As shown in Figure 3, ~60-80% of wet THg deposition fluxes at these stations occurred during the ISM season (Huang et al., 2012, 2013, 2015; Tripathee et al., 2019a, 2019b). These results can partly be explained by the fact that most of the rainfall occurs during the ISM season, and partly by the influence of transboundary transport of Hg from South Asia and Southeast Asia. Indeed, during the ISM season of 2013, atmospheric water vapor samples collected from the TP showed a mean THg concentration of 12.5 ng L^{-1} (range: 2.5 to 73.7 ng L^{-1}, Huang, Kang, Zhang, Yan, et al.,

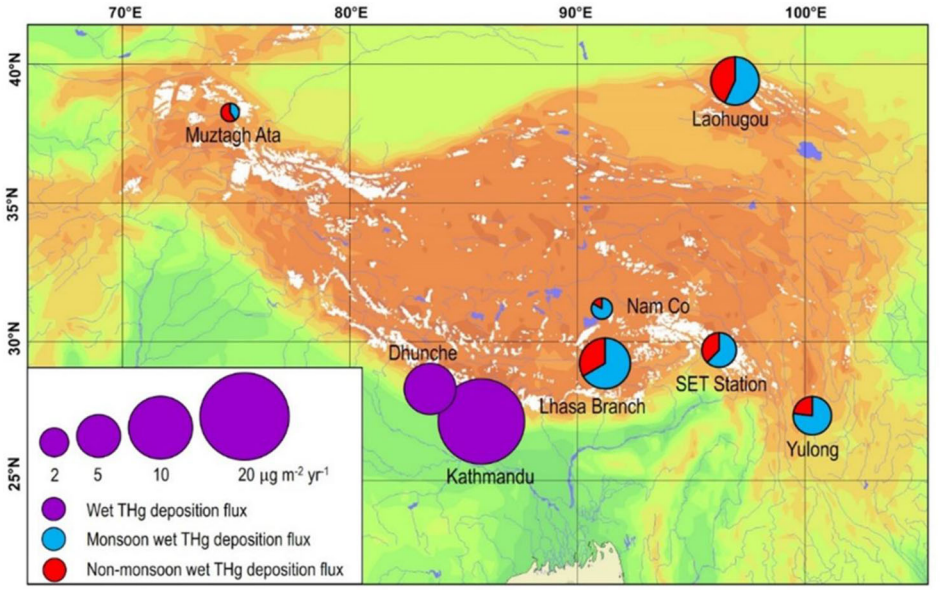

Figure 3. Spatial distribution of wet THg deposition fluxes over the Tibetan Plateau (Huang, Kang, Zhang, Yan, et al., 2012; Huang et al., 2013, 2015; Tripathee et al., 2019a, b).

2016), which is higher than that observed in other mountains such as Mt. Bamboo in Taiwan (9.6 ng L^{-1}; Sheu & Lin, 2011).

3.3. Snow and glacier

The TP is home to the largest aggregate of glaciers outside the polar regions. The concentration of Hg in glacier snow samples collected from several sites in the TP (Figure 4) were investigated by several studies (Huang, Kang, Zhang, Yan, et al., 2012; Huang, Kang, Zhang, Jenkins, et al., 2012; Huang et al., 2016, 2019; Paudyal et al., 2017, 2019; Zhang et al., 2012). In general, the glacier snow Hg concentrations were largely different in East Rongbuk glacier from the Mt. Everest (2.22 ± 0.56 ng L^{-1}), Zhadang glacier from the Mt. Nyainqêntanglha (0.90 ± 0.29 ng L^{-1}), Guoqu (GQ) glacier from the Mt. Geladaindong (3.63 ± 1.12 ng L^{-1}), Muztagata (MZ) glacier from the Mt. Muztagata (8.56 ± 3.08 ng $L^{-}1$). These values are comparable with those observed in surface snow collected from polar regions (Douglas & Sturm, 2004). Most of Hg (48%) in the Tibetan glacier snow was associated with particulate matter (Zhang et al., 2012), indicating that particulate bound Hg could accumulate to a significant amount in glacier snow. This can be proven by the high THg concentrations in glacier cryoconite (a type of dust which is deposited and built upon glaciers) over the TP (17.9 to 114.5 ng g^{-1}, according to Huang et al. (2019)). Moreover,

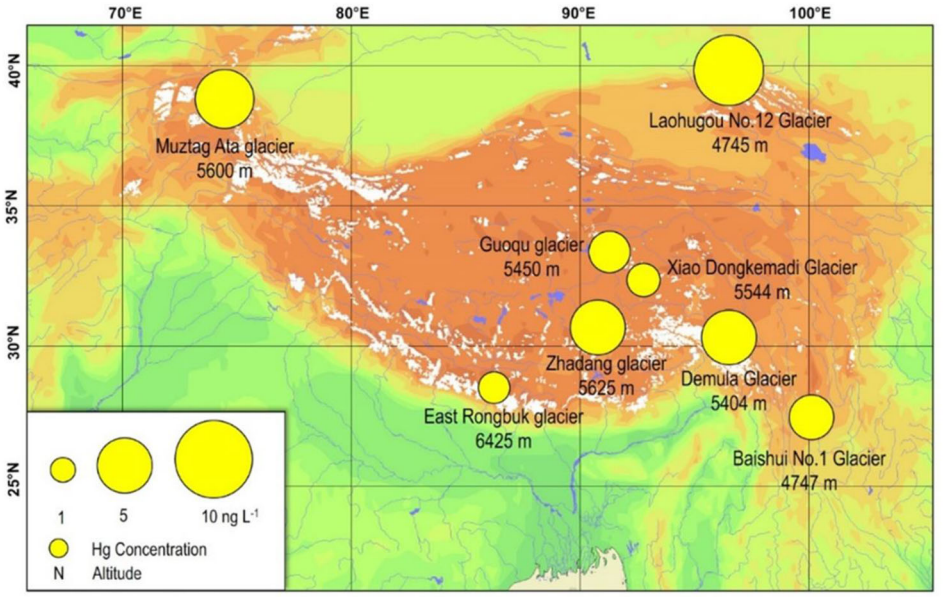

Figure 4. Spatial distribution of snow THg concentration over the Tibetan Plateau (Huang, Kang, Zhang, Yan, et al., 2012; Huang, Kang, Zhang, Jenkins, et al., 2012; Huang et al., 2016; Paudyal et al., 2017, 2019; Zhang et al., 2012).

at several glaciers in the TP (altitude: 4000-5000 m a.s.l.), the concentrations of THg in glacier snow increased significantly with increasing altitude (Huang et al., 2014), demonstrating that atmospheric Hg is cold-trapped and magnified toward low temperature and/or high-elevation glaciers over the TP. Estimated deposition fluxes of atmospheric Hg to the TP glaciers ranged from 0.74 to 7.89 μg $m^{-2} yr^{-1}$, according to Zhang et al. (2012).

3.4. Atmospheric Hg deposition history revealed by glacier ice core and sediment core

Lake sediments and ice cores serve as natural archives for documenting the historical changes of atmospheric Hg deposition. In the last decade, analysis of Hg variations in age-dated lake sediments and glacier ice cores yielded comprehensive Hg reconstructions, showing clearly enhanced deposition flux in the last century (Kang et al., 2016; Wang et al., 2010; Yang et al., 2010; Yin, Feng, Hurley, Krabbenhoft, Lepak, Kang, et al., 2016).

Studies on sediment cores indicated that enhanced Hg deposition flux by a factor of 2-40 over the TP started at the onset of the global Industrial Revolution in the 19th century. The most significant increase in Hg deposition flux began since the 1950s, which reflects the enhanced anthropogenic Hg emission from Asia (Kang et al., 2016; Wang et al., 2010; Yang et al., 2010; Yin, Feng, Hurley, Krabbenhoft, Lepak, Kang, et al., 2016). Kang

et al. (2016) and Yin, Feng, Hurley, Krabbenhoft, Lepak, Kang, et al. (2016) reconstructed the historical variations of atmospheric Hg deposition by several lake sediments in the TP, which provides a detailed understanding of the atmospheric Hg deposition history (Figure 5). They pointed out that the Hg accumulation rates in the Himalayas (average: 112.1 μg $m^{-2} yr^{-1}$), which is located at the border between South Asia and TP, were significantly greater than those determined in the inland TP (average: 14.6 μg $m^{-2} yr^{-1}$).

Kang et al. (2016) also retrieved a deep ice core (length: 147 m) in the Mt. Geladaindong from the inner TP. In general, the Hg accumulation rate calculated from the Geladaindong ice is representative of the net retention of atmospheric Hg, which was 1 to 2 orders of magnitude lower than the lake sediments in the TP. However, its temporal trend agrees very well with the trend observed from the lake sediments (Figure 5). The depositional chronologies of atmospheric Hg in the ice core showed that the Hg deposition rate increased during the onset of the Industrial Revolution, followed by a dramatic increase after World War II. The increasing trend continues to the present-day in most of the records, reflecting the continuous increase in anthropogenic Hg emissions from South Asia (Kang et al., 2016).

4. Hg in aquatic ecosystems

4.1. Hg in glacier meltwater, river water, and lake water

Glacier acts as a temporary reservoir for atmospheric Hg over the TP. Hg stored in glacier snow can be released to river and lake waters when the glaciers are subject to melting with climate warming. Climate warming in the TP is twice faster than the global average in the past several decades (Zhang, Yao, et al., 2015), which has been a particular concern for glacier mass change over the TP (Yao et al., 2018), because high mountain glaciers are more sensitive to climate change than polar glaciers. As a result of climate warming, the accumulated Hg in mountain glaciers in the TP may be released by accelerated glacier thinning, and retreat may endanger ecosystems and human health in the glacier-fed downstream regions. There are more than 1500 lakes in the TP, most of which are fed by the meltwater of glaciers (Song, Huang, & Ke, 2013).

Glacier ablation can increase the streamflow of glacier-fed rivers and may simultaneously result in the release and transport of Hg to downstream ecosystems (Huang et al., 2014; Li et al., 2015; Sharma et al., 2015; Sun et al., 2016, 2018; Zhang et al., 2012). THg concentrations in the water of glacier outlets (i.e., proglacial meltwater) were found to be about 5 times higher than those in the surface snow (Huang et al., 2014). THg levels in

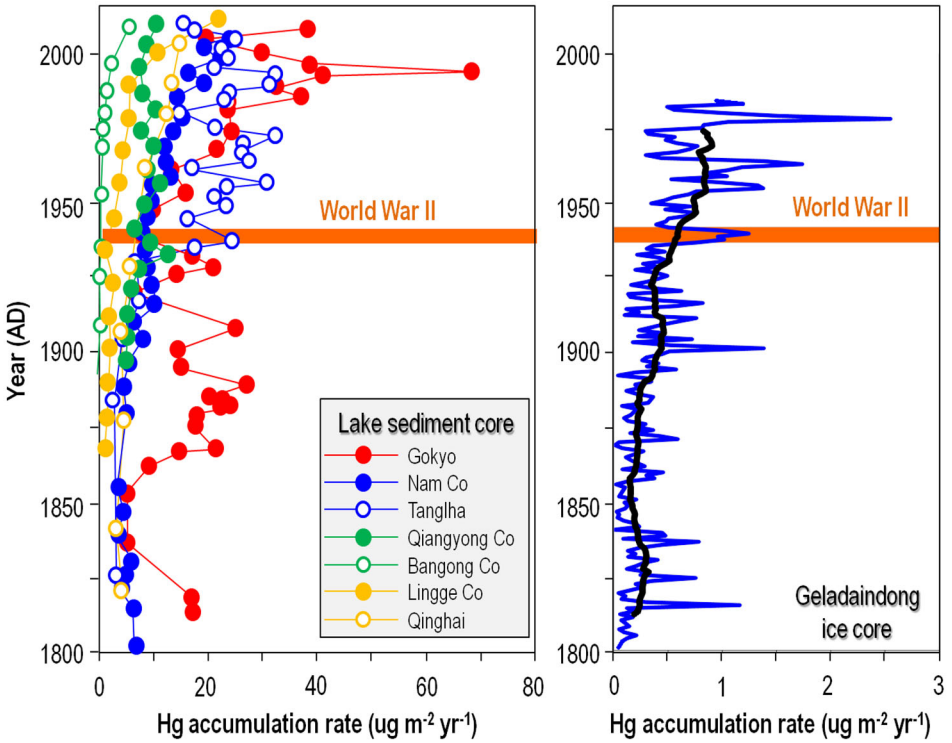

Figure 5. Historical trends of Hg accumulation rate reconstructed from sediment and ice cores from the Tibetan Plateau (Yin, Feng, Hurley, Krabbenhoft, Lepak, Kang, et al., 2016; Kang et al., 2016).

glacial meltwater in the TP (1.18 to 5.54 ng L^{-1}) were found to be comparable with that of glacier snow (Huang et al., 2014). With ongoing climate warming, glacier retreat is expected to continue and the predicted reduction of glacier volume in the TP. More historically accumulated glacier mass will melt and accordingly the total amount of released Hg will increase.

TP rivers receive water from glacier meltwater and precipitation. Previous studies have shown that THg levels in the TP rivers were highly variable (1.46-4.99 ng L^{-1}, according to Zheng, Kang, Feng, Zhang, and Li (2010)), but were lower than that of glacier meltwater and precipitation. As PHg was found to be the predominant form of THg in these rivers, the low THg concentrations have been explained by the sedimentation of PHg (Sun et al., 2016). Moreover, Li et al. (2016) have reported the concentration of Hg in waters of 38 Tibetan lakes (range: <1-40.3 ng L^{-1}; mean: 3.82 ng L^{-1}) and found Hg is strongly positively correlated with total dissolved solids, exhibiting an increasing trend along the southeast-northwest transect. The total Hg concentration in saline lakes reaches up to 40.3 ng L^{-1}, indicating enhanced accumulation of Hg by water evaporation in salt

lakes. In aquatic ecosystems, Hg is readily transformed into MeHg via microbial processes. There is a lack of MeHg dataset at this time, preventing us from understanding the extent as well as factors controlling Hg methylation in the aquatic ecosystems of the TP. However, the relatively higher THg in the 38 Tibetan lakes (mean: 3.82 ng L^{-1}, Li et al., 2016) compared to other rural/remote lakes (e.g., Plitvice Lakes in Croatia (1.22 ng L^{-1}, Vukosav et al. (2014)), Great Lakes in North America (1.0 ± 2.05 ng L^{-1}, Dove et al. (2012)), 60 Lakes in Northern Canada (3.64 ± 2.52 ng L^{-1}, Vaidya, Howell, & Leger, 2000) and 101 lakes in Western US (1.07 ± 1.41 ng L^{-1}, Krabbenhoft et al., 2002)), coupled with the ongoing climate warming, may greatly facilitate the production of MeHg in the TP, similar to that found in the pristine region of the Arctic (Yang, Xie, et al., 2016). With accelerated warming in the TP, the MeHg production rates are reasonably expected to increase in the TP ecosystems. Further studies on MeHg levels in the environmental matrix of TP such as aquatic ecosystems are therefore urgently needed.

4.2. Hg levels in Tibetan fish

The concentrations of THg and MeHg in fish from the TP were investigated by a number of studies (Pandey et al., 2017; Shao et al., 2016; Sharma et al., 2013; Thapa, Sharma, Kang, & Sillanpää, 2014; Xu, Zhang, & Wang, 2016; Yang, Jing, Zhang, & Jiang, 2013; Yang et al., 2011; Zhang, Pan, et al., 2014). In summary, a total of 517 fish individuals belong to 18 species were collected from 18 different regions. Regarding the reported data, THg concentrations ranged from 48.6 to 1684.8 ng g^{-1} (wet weight, ww), and MeHg concentration ranged from 26.2 to 1269.8 ng g^{-1}(ww). The MeHg/THg ratios generally exceeded 70%, suggesting that MeHg is the dominant form of Hg in fish muscles (Zhang, Yao, et al., 2014). As shown in Figure 6, through a careful analysis of the reported data, ~94% of the fish contained MeHg of $>$ 100 ng g^{-1} (ww) which exceeded the U.S. Fish and Wildlife Service Criteria of 100 ng g^{-1} of ww for the protection of fish-eating birds and wildlife (USFWS, 2005); ~62% of the fish exceeded the 300 ng g^{-1} of ww set for human health by the U.S. Environmental Protection Agency (USEPA, 2001).

Zhang, Yao, et al. (2014) proposed that trophic transfer might not be the main factor for the enrichment of MeHg in the TP fish. In the TP lakes, it was suggested that the nutrient-poor environments and low temperatures may inhibit the efficient elimination of Hg in fish (Lavoie, Jardine, Chumchal, Kidd, & Campbell, 2013). According to Zhang, Yao, et al. (2014), fish in the TP lakes were not located at a high trophic position, and the benthic algae and zooplankton consumed by fish were often the main

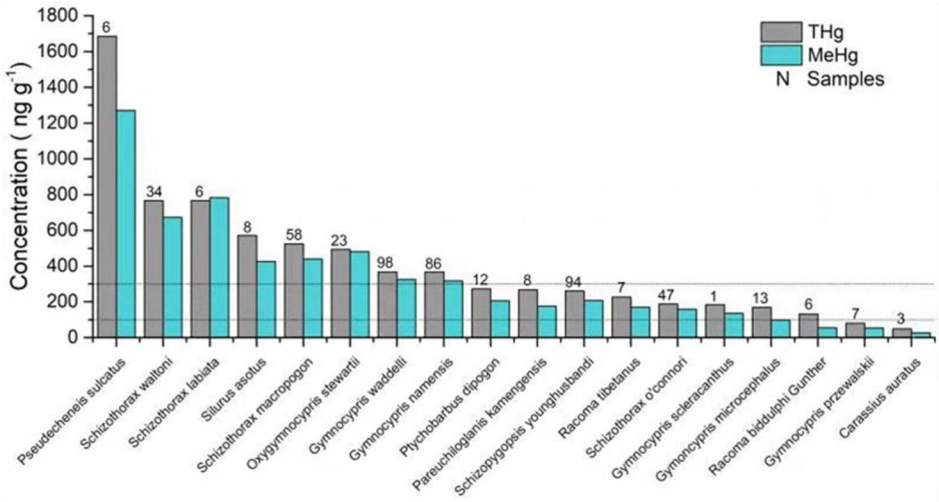

Figure 6. Variations of THg concentration in fishes from Tibetan Plateau (Yang et al., 2011; Yang, Jing, et al., 2013; Sharma et al., 2013; Shao et al., 2016; Zhang, Yao, et al., 2014; Thapa et al., 2014; Xu et al., 2016; Pandey et al., 2017).

producers. Moreover, as there are few natural fish predators living in TP, most fish in the TP lakes and rivers had a long life span (several to tens of years, according to Zhang, Yao, et al. (2014)). Therefore, the high MeHg concentrations in the TP fish were largely attributed to fish's slow growth, long life span, and lack of efficient Hg elimination in fish.

5. Hg in vegetation and surface soil

Vegetation and soil act as the primary sink for atmospheric Hg in the TP (Huang et al., 2020; Wang, Luo, et al., 2016). In a previous study by Shao et al. (2017), the THg concentrations of mosses and lichens collected from different altitudes of the TP (1983-5147 m a.s.l.) ranged 13.1-273.0 ng/g (n = 130) and 20.2-345.9 ng/g (n = 52), respectively, and were not correlated with the THg in soils ($p > .05$). This is not surprising because moss and lichen got much nutrients from the air not the soil. According to Shao et al. (2017), a significantly positive correlation between Hg contents in mosses and the altitudes of sampling sites was observed, indicating the mountain trapping of atmospheric Hg in the alpine areas of the TP. Although moss and lichen may not be good representatives for plants in general, they have been used as an indicator of atmospheric Hg concentration (Estrade, Carignan, & Donard, 2010). In fact, vegetation receives a substantial amount of Hg from the atmosphere due to the stomata uptake of gaseous Hg(0) (Yin, Feng, & Meng, 2013). Some plant species (e.g., *Androsace tapete*) have been used as an indicator for atmospheric Hg

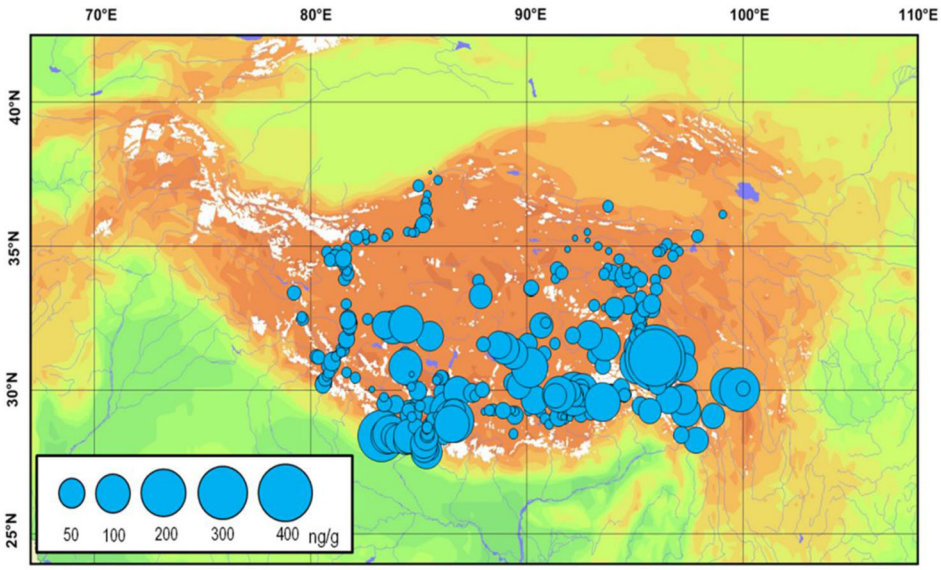

Figure 7. Spatial distribution of soil THg in the Tibetan Plateau (Sheng et al., 2012; Gong et al., 2014; Xie et al., 2014; Liu et al., 2016; Sun, Wang, et al., 2017; Wang, Luo, et al., 2016; Tripathee, Guo, Kang, Paudyal, Huang, Sharma, Gyawali, 2019; Huang et al., 2020).

concentration in the TP (Tong et al., 2016). In a recent study at Mt. Ailao (Wang et al., 2019), it was proved that precipitation can enhance litterfall Hg deposition by increasing biomass production, reduce litter decomposition rate, and facilitate short-term Hg uptake to decomposing litter.

The THg concentrations of surface soil (mostly 0-5 cm depth) collected from the TP showed a wide range of 1 to 418 ng g^{-1} (Figure 7), according to a number of studies (Gong, Wang, Xue, Xu, & Yao, 2014; Huang et al., 2020; Liu et al., 2016; Sheng, Wang, Gong, Tian, & Yao, 2012; Sun, Kang, et al., 2017; Tripathee et al., 2019a, 2019b; Wang, Luo, et al., 2016; Xie et al., 2014). No clear correlation was found between soil Hg and soil organic carbon (SOC) in the Tibetan soils, indicating that SOC is not the major driver for the spatial variations in soil THg (Huang et al., 2020). The spatial variation of soil Hg may be explained by the difference of THg concentrations in basement rocks. However, this seems unlikely because two recent studies demonstrated significant anomalies in mercury isotope ratios, which is used to fingerprint geological, natural, and anthropogenic sources of Hg, in Tibetan soil (Wang, Luo, et al., 2016; Huang et al., 2020), which is different from the Hg isotope ratios in basement rocks (Xu et al., 2018). Bare soils primarily receive atmospheric Hg through litterfall or direct atmospheric deposition. It is interesting that the soil THg concentrations showed a clear decreasing trend from the south to the north of the TP (Figure 7). In particular, the highest soil Hg concentrations were observed in the southern TP, which is more close to South Asia and Southeast Asia.

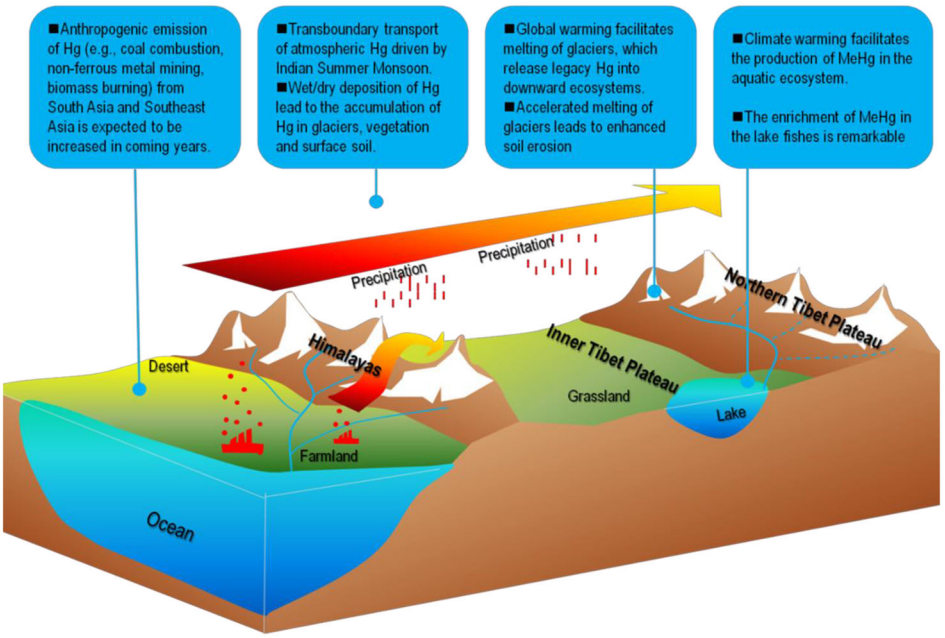

Figure 8. Illustration showing the current understanding of Hg over the Tibetan Plateau (Modified from Kang et al., 2019).

This agrees with the spatial patterns of atmospheric Hg deposition (as mentioned in Section 2.2, 3.1 and 3.2), reconfirming that transboundary transport of atmospheric Hg from South Asia and Southeast Asia is a major source of Hg in the TP soils.

6. Conclusions and implications

Through a careful literature review, this study outlines a comprehensive view of the sources and environmental fate of Hg in the TP. In Figure 8, we summarize the major highlights of the Hg biogeochemical cycle in the TP.

First, the local emission of Hg within the TP is thought to be less important due to the sparse human population and minimal industrial activities in the TP, although a substantial amount of Hg can be released from human sewage (~3.6 tons in 2015, Liu, He, Ge, et al. (2018)) and yak feces burning (the exact amount of Hg remains unknown). South Asia and Southeast Asia are the dominant emission sources of Hg deposited to the TP. Due to its high elevation and the dominance of ISM to this study region, the TP is believed to be sensitive to the transboundary transport of atmospheric Hg from South Asia and Southeast Asia. Atmospheric Hg emitted from these areas can traverse the Himalayas and reach the inland TP region and further accumulate in the surface environment due to the

"altitude effect" or "cold trapping effect." The vast cryospheric compartments such as glacier, vegetation, and surface soil over the TP act as major sinks of atmospheric Hg.

Second, climate warming in the TP has a profound impact on facilitating the release of legacy Hg from melting glacier to aquatic ecosystems. The cryospheric environments of the TP are particularly sensitive to global climate change. In particular, global warming has not only caused increased precipitation at an average annual rate of 10.9 mm per decade from 1961 to 2008 but also resulted in continuous increases of growing season ($\sim$1.04 day y^{-1}, according to Zhang, Zhang, et al. (2013)) and vegetation coverage (3961.9 $km^{-2} yr^{-1}$ during the past 2 to 3 decades, according to Liang et al. (2007)). Precipitation and vegetation are efficient scavengers of atmospheric Hg and play a vital role in the Hg inputs to surface soil (Loewen et al., 2007; Zhang, Zhang, et al., 2013). Increased precipitation and accelerated glacier melting have aquatic caused rapid input of Hg pollution in the aquatic ecosystems of the TP. In addition, climate warming promotes methylation of Hg to produce the more toxic MeHg (Yang, Xie, et al., 2016), thereby increasing the environmental risk of Hg pollution over the TP.

As the "water tower of Asia," the TP is the source of main Asian rivers (e.g., the Ganges, the Indus) which supply water for approximately 1/6 of the world's population (Immerzeel, Van Beek, & Bierkens, 2010). In this regard, it seems that the scenarios of Hg biogeochemical cycling in the TP, such as transboundary transport of atmospheric Hg to the TP, increased release of legacy Hg from melting glacier and increased Hg methylation due to climate warming, would cause adverse impacts to downstream regions of rivers, and thus potentially threaten the fragile ecosystems of the TP as well as many South Asian and Southeast Asian countries.

The human and environmental exposure to Hg depend on the quantities of Hg entering the environmental reservoirs. In general, current Hg levels in different environmental reservoirs over TP remain lower than those in regions with intensive human activity (e.g., metropolitan cities). However, as South Asia and Southeast Asia are the dominant emission sources of Hg deposited to the TP, transboundary transport of anthropogenic Hg from these areas is expected to be increased in the near future, considering the fact that the economy and industrialization in South Asian and Southeastern Asian countries will be rapidly growing in the coming decades. Fortunately, with the recently signed Minamata Convention that entered into force in 2017, more actions from these countries will also be carried out to reduce the emission of anthropogenic Hg. Therefore, it is believed that the risk of Hg pollution in the TP will be reduced ultimately.

Although studies conducted over the past two decades has improved our understanding of the biogeochemistry of Hg over the TP, many additional

efforts are still required to fill knowledge gaps, which can be can be summarized as follows: (1) As the transboundary transport is the major pathway of transporting Hg to the TP, international/regional collaborations (especially between China and South Asian countries) regarding Hg emission regulation are urgently needed to reduce the flows of Hg and mitigate adverse Hg pollution to the TP; (2) Although studies have been performed on the levels and bioaccumulation of Hg in the aquatic food webs of the TP, no data on terrestrial food chains have thus far been reported. Due to the close relation between terrestrial food chains and human life, more work on the terrestrial Hg bioaccumulation in the TP is urgently needed in the future; (3) Climate change and emissions regulation both play an important role in the Hg biogeochemistry in the TP. As the TP is highly sensitive to climatic factors but far away from direct emission sources, long-term monitoring of Hg deposition and assessment of transboundary Hg pollution to the TP are required in the future, to better understand the effects of both climate and Hg emission control; (4) Modeling is a powerful way to simulate the global transport of Hg, however, such work has not been done for the TP. It will be helpful and needed to develop Hg models that simulate the sources, fluxes and transportation processes of Hg in the TP in the future.

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