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Invited paper

Temperature sensitivity of gaseous elemental mercury in the active layer of the Qinghai-Tibet Plateau permafrost☆

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ABSTRACT

Soils represent the single largest mercury (Hg) reservoir in the global environment, indicating that a tiny change of Hg behavior in soil ecosystem could greatly affect the global Hg cycle. Climate warming is strongly altering the structure and functions of permafrost and then would influence the Hg cycle in permafrost soils. However, Hg biogeochemistry in climate-sensitive permafrost is poorly investigated. Here we report a data set of soil Hg (0) concentrations in four different depths of the active layer in the Qinghai-Tibet Plateau permafrost. We find that soil Hg (0) concentrations exhibited a strongly positive and exponential relationship with temperature and showed different temperature sensitivity under the frozen and unfrozen condition. We conservatively estimate that temperature increases following latest temperature scenarios of the IPCC could result in up to a 54.9% increase in Hg (0) concentrations in surface permafrost soils by 2100. Combining the simultaneous measurement of air–soil Hg (0) exchange, we find that enhanced Hg (0) concentrations in upper soils could favor Hg (0) emissions from surface soil. Our findings indicate that Hg (0) emission could be stimulated by permafrost thawing in a warmer world.

1. Introduction

Mercury (Hg) is a ubiquitous and persistent pollutant in the global environment that is transported over long distances (Selin, 2009). In aquatic ecosystems, divalent Hg is substantially transformed to neurotoxic methylmercury (Hammerschmidt et al., 2006; Hollweg et al., 2009; Schartup et al., 2013; Jonsson et al., 2014), which strongly bioaccumulates and biomagnifies along the food web, resulting in elevated exposure to wildlife and humans (Mason et al., 2012). Global change is altering the regional and global Hg cycle (Obrist, 2007; Fisher et al., 2012; Stern et al., 2012; Hararuk et al., 2013; Yang et al., 2016), but it remains a large uncertainty on understanding of the link between the Hg cycling and global change drivers (e.g., climate warming) (Krabbenhoft and Sunderland, 2013; Haynes et al., 2017).

Elemental Hg (Hg (0)) plays the dominant role in the global Hg cycle due to its long atmospheric lifetime (Selin, 2009). Surface soils contribute roughly 30% of global Hg (0) emission flux to the atmosphere (Agnan et al., 2015; Enrico et al., 2016; Obrist et al., 2017). The combination of a mass-balance analysis of atmospheric Hg deposition and the measurement of Hg stable isotopic compositions show that the Hg (0) is the dominant form of Hg deposition in the Arctic tundra (Obrist et al., 2017). Similar finding was also reported in peat bogs (Enrico et al., 2016). Current knowledge suggests that soil property (such as soil Hg level and soil organic carbon) is the dominant factor for soil Hg (0) concentrations (Wallschläger et al., 2002; Sigler and Lee, 2006), and a variety of environmental variables, such as soil temperature and moisture and microbial activity, redox potential, determine the fluctuation of soil Hg (0) concentration in a short time scale (Moore and Castro, 2012; Pannu et al., 2014; Obrist et al., 2014). Studies highlight the difference of soil Hg (0) dynamics among various ecosystems (Obrist et al., 2014), and also show the possible linkage between Hg (0) dynamics in upper soils and air–soil Hg (0) flux (Wallschläger et al., 2002; Sigler and Lee, 2006).

Permafrost and seasonal frozen soils account for about 70% of terrestrial area of Earth (NSIDC, 2016). Permafrost is potentially important for global Hg cycle (Schuster et al., 2018). Climate warming results in permafrost thaw. Expert assessment suggests that the global permafrost could decline by between 11 and 18% (RCP 2.6) and 48–63% (RCP 8.5) in 2100 (Schuur et al., 2013). One of the most significant consequences of permafrost thaw is the

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positive feedback to climate change since the warming accelerates the decomposition of soil carbon to release green house gasses to the atmosphere (Schuur et al., 2015). Recently, the sensitivity of soil Hg storage and speciation to the climate change have received increasing attention (Obrist, 2007; Obrist et al., 2010; Stern et al., 2012; Yang et al., 2016). Studies report that thawing permafrost is expected to be an important Hg source to surrounding environments (Rydberg et al., 2010; Fisher et al., 2012; Schartup et al., 2015). Thawing permafrost also concurrently stimulates the release of organic matter and nutrients to promote the biological activity in soil matrix (Frey et al., 2007; Abbott et al., 2015), which can provide important substrates and favorable environmental conditions to alter the Hg transformations in permafrost ecosystem (MacMillan et al., 2015). However, existing limited studies on soil Hg biogeochemistry mainly focus on temperate regions in warm seasons (i.e., soil temperature > 0 °C) (Sigler and Lee, 2006; Obrist et al., 2014) or in laboratory (Pannu et al., 2014; Yang et al., 2016; Walters et al., 2016). The sensitivity of soil Hg speciation in climate-sensitive permafrost remains poorly quantified in field (Barkay et al., 2011).

The Qinghai-Tibet Plateau (QTP) is the highest plateau in the world with a mean altitude of 4000 m above sea level (a.s.l.) and wide distribution of seasonal frozen soils and permafrost (Cheng and Wu, 2007; Li et al., 2008). Increasing evidences show that the QTP is one of the most sensitive regions to the climate warming (Li et al., 2008; Wu and Zhang, 2008; Wu et al., 2012). The dramatic climate change and the huge temperature variation associated with the thaw-freeze cycle of the active layer result in the great variation of soil structure, moisture, heat balance and chemical and biological processes of carbon cycle (Peng et al., 2015a; Wu et al., 2015), which could have potentially important consequences for the soil Hg biogeochemistry (Ci et al., 2016a; Yang et al., 2016; Sun et al., 2017).

Here we measured Hg (0) concentrations in four different depths (5 cm, 10 cm, 20 cm and 40 cm) of a typical active layer of the central QTP continuous permafrost during four seasonal campaigns in 2014–2015. The goal of this study is to characterize the temporal and vertical variation of soil Hg (0) concentration and investigate the effect of environmental factors on soil Hg (0) dynamics in the active layer of permafrost. We follow the latest temperature scenarios presented by the IPCC RCP (Representative Concentration Pathways) scenarios (IPCC in Climate Change, 2013) to explore the consequence of global warming scenarios for Hg (0) concentrations in the active layer of permafrost. Combining synchronous measurement results of air–soil Hg (0) exchange in the same soil plot (Ci et al., 2016a), we further discuss the potential effect of climate change on Hg (0) emission in a warmer world.

2. Materials and methods

2.1. Research station

The field measurements were conducted at the Beiluhe Permafrost Engineering and Environmental Research Station affiliated to the Northwest Institute of Eco-Environment and Resources, Chinese Academy of Sciences (NIEER–CAS). The station (34° 49′ 45″ N, 92° 56' 06" E) is located in the central QTP with an elevation of about 4700 m a.s.l. (Fig. 1). The Beiluhe region experiences a typical alpine semiarid climate with cold winter (to −30 °C) and warm summer (to 20 °C), inducing the dramatic variation of surface temperature about 50 °C (Peng et al., 2015a). The annual mean surface air temperature is about −2 to −3 °C. The annual potential evaporation (ca. 1300 mm) is greatly higher than the annual precipitation (ca. 300 mm), and mostly precipitation occurs during May to October (Peng et al., 2015a). The Beiluhe station lies in the ice-rich continuous permafrost zone with an active layer thickness

Fig. 1. Sampling location. Beiluhe station is located in the central Qinghai-Tibet plateau with an elevation of 4700 m above sea level.

(the maximum thawing depth during summer time) of 2.0–3.2 m and a permafrost thickness of 60–200 m (Peng et al., 2015a; Wu et al., 2015). The surface soils generally start to freeze in September and thaw in May (Peng et al., 2015a). The active layer thickness around the Beiluhe station is increasing at a rate of ca. 3–4 cm y^{-1} over the past two decades due to the climate warming (Wu et al., 2015).

2.2. Soil Hg(0) measurement

A bare soil plot of 2 × 2 m was separated to two soil plots of 1 × 2 m to observe soil Hg (0) concentrations and air–soil Hg (0) flux, respectively (Ci et al., 2016a). Soil Hg (0) concentrations were measured for 6–13 days in the unsaturated condition during four campaigns (June, September, and December 2014 and May 2015). The method of Moore et al. (2011) was slightly modified to collect and determine Hg (0) in different soil depths. Briefly, Hg (0) in soil air was collected using the fluted borosilicate funnel with a top diameter of 90 mm and stem length of 100 mm. The funnels were buried top down at four sampling depths: 5, 10, 20 and 40 cm. Teflon tube was used to connect the glass stem to the gold trap. Funnels were separated 40 cm laterally to minimize disturbance during installation and to reduce the risk of depleting of Hg (0) pool in the soil matrix. Hg (0) in soil air was collected on gold traps by pumping soil air out of each funnel for 2–4 h at 25 ml min^{-1}. The flow rate has been proved no disturb the soil gas profile (Sigler and Lee, 2006; Fang and Moncrieff, 1998; Moore et al., 2011). Hg (0) on gold traps was quantified with a cold vapor atomic fluorescence spectrometry (CVAFS, Model III, Brooks Rand, USA) using the two-stage gold amalgamation, as shown in previous studies (Ci et al., 2011; Ci et al., 2016a,b). The analysis system was calibrated using a gas-tight microsyringe to inject Hg (0)-saturated air from a water bath and the analytical precision was 3%. The detection limit of system was 0.02 ng m^{-3} based on three times standard deviation of system blanks (Ci et al., 2011a,b). Soil Hg (0) concentrations were calculated with the equation: soil Hg (0) concentration (ng m^{-3}) = Hg (0) (ng)/flow rate (m^3 min^{-1})/the duration of sampling time (min).

2.3. Atmospheric Hg(0) measurement

The concentrations of atmospheric Hg (0) were monitored by a similar gold trap system (Ci et al., 2016a). Hg (0) in the ambient air was collected on gold traps by pumping ambient air at 0.50 L min^{-1} (0.03 m^{-3} h^{-1}). The height of atmospheric Hg (0) measurement was 3 cm above the ground. More details can be found in the study of Ci et al. (2016a).