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Mercury cycling in grasslands: deposition, plant uptake, and biomass-burning emissions

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In order to evaluate the effectiveness of the Minamata Convention on mercury, understanding the air–surface mercury exchange in grasslands is important as they cover 20%–40% of the Earth's land surface. An in-depth quantitative understanding of the processes of the mercury cycle, such as dry and wet depositions, evasion from soil, plant uptake, and natural and prescribed biomass burning, is essential to explore the mercury cycle in these regions; however, only a limited number of studies are available on these processes, and many questions regarding them still remain. In this mini-review, the key emission and sinking processes occurring in natural and semi-natural grasslands and the potential of stable mercury isotope measurements for tracing studies of mercury origin(s) in grasslands are discussed.

KEYWORDS

air–surface exchange of mercury, mercury emissions from prescribed grassland burning, atmospheric mercury deposition, plant uptake of atmospheric mercury, stable mercury isotope ratio, *Miscanthus sinensis*, *Pleioblastus chino* var. *viridis*, mercury flux

1 Introduction

Mercury (Hg) is a toxic heavy metal with the unique characteristics of existing in a liquid state and evaporating slowly, even at room temperature and atmospheric pressure. Because of this characteristic, evaporated elemental Hg or gaseous elemental Hg (GEM) disperses into the atmosphere and spreads globally. Therefore, Hg is found in various substrates on the terrestrial surface and even in wildlife in pristine areas. Atmospheric Hg is generally categorized into four forms: GEM, gaseous oxidized Hg (GOM), particulate-bound Hg (PBM), and Hg in precipitation. For a convenience of sampling, GEM and GOM are sometime combined and treated as total gaseous mercury (TGM). Methylmercury in the atmosphere is typically almost negligible. GEM is the dominant atmospheric form of Hg (>90%–95% of the sum of GEM, GOM, and PBM concentrations) (Schroeder and Munthe, 1998; Hang et al., 2004; Sprovieri et al., 2010; Gay et al., 2013; Gustin et al., 2015; Sun et al., 2019). These forms can interconvert in the environment, especially between GEM and GOM. In a complex process, anthropogenic Hg used in the past (known as legacy Hg) remains and cycles through the terrestrial environment along with newly introduced Hg through chemical and physical processes such as evasion (e.g., legacy Hg volatilization) from soil (Wallschläger et al., 1999; Wang et al., 2003; Frescholtz and Gustin, 2004; Poissant et al., 2004; Moore and Carpi, 2005; Ericksen et al., 2006; Choi et al., 2009; Moore and Castro, 2012; Park et al., 2014), dry deposition (Fritsche et al., 2008a; Fritsche et al., 2008b; Zhang et al., 2009; Baya and Van Heyst, 2010; Rutter et al., 2011; Zhang et al., 2012; Fang et al., 2013; Zhang et al., 2016; Hao et al., 2021; Ahmed et al., 1987), wet deposition (Butler et al., 2008; Guo et al., 2008; Prestbo and Gay, 2009; Seo et al., 2012; Marumoto and

Matsuyama, 2014; Xu et al., 2014; Gichuki and Mason, 2014; Qin et al., 2016; Fang et al., 2018; Du and Fang, 1983), and plant uptake and emission (Fres et al., 2003; Lodenius et al., 2003; Zhang et al., 2005; Millhollen et al., 2006; Poissant et al., 2008; Baya and Van Heyst, 2010; Converse et al., 2010; Niu et al., 2011; Graydon et al., 2012; Niu et al., 2013; Cui et al., 2014; Niu et al., 2014; Blackwell and Driscoll, 2015; Assad et al., 2016; Chiarantini et al., 2016; Luo et al., 2016; Fu et al., 2016; Canário et al., 2017; Gustin et al., 2008). Furthermore, Hg can be converted to methylmercury, a highly toxic and bioaccumulative form of Hg in vertebrates (Kidd et al., 2012), which can then enter the ecosystem (Liu et al., 2012). This complex dynamic makes it challenging to trace the origins of Hg, and our current understanding of air–surface Hg exchange remains unclear despite extensive scientific efforts over the past five decades. Advancing this understanding will directly contribute to the evaluation of the Minamata Convention on Mercury, the United Nations-implemented global treaty regulating anthropogenic mercury use. Recently, GEM uptake by plants has received attention as it was not considered in the Global Mercury Assessment 2018 of the UN reports (Global Mercury Assessment, 2018) despite its potential as a major sink for atmospheric GEM (Mason et al., 2005; Jiskra et al., 2018). To date, most flux studies over vegetated fields have focused on Hg cycling in forest ecosystems; in contrast, reports on Hg cycling in grasslands remain limited (Erickson et al., 2006; Fritsche et al., 2008a; Fritsche et al., 2008b; Converse et al., 2010; Niu et al., 2011; Graydon et al., 2012; Niu et al., 2013; Millhollen et al., 2006; Stamenkovic et al., 2008).

Globally, grasslands cover a substantial portion (20%–40%) of the land surface of the Earth (White et al., 2000; Conant, 2010). Thus, results from the Hg cycle studies in grasslands can provide valuable information on the atmospheric–surface Hg exchange and will improve the model predictions for the exchange. Regarding Hg flux in vegetated fields, grasslands have certain advantages; they typically host a limited number of dominant short plant species, which makes gaining overall biomass samples that represent the grassland vegetation more feasible (Irei et al., 2023) than obtaining samples representing forested fields (Friedli et al., 2007). This simplifies the interpretation of vegetated field data. Moreover, in the case of semi-natural grasslands that are periodically burnt by humans to prevent forest encroachment and control pests, controlled burning can reset the above-ground Hg pool, thus offering unique opportunities to study air–surface Hg exchange. Such semi-natural grasslands are found throughout Japan. In addition, the significance of biomass burning as a domestic Hg emission source has not yet been evaluated in Japan, where wildfires are rare events. In this study, features of Japanese semi-natural grasslands, key processes observed or likely occurring in Japanese and other grasslands, and the potential use of stable isotope ratio measurements in such tracing studies are discussed.

2 Semi-natural grasslands in Japan

Most of the semi-natural grasslands across Japan are small (<30 ha), while large semi-natural grasslands are found in national parks and the training fields of the Ministry of Defense of Japan. The burned area of each grassland, as reported by the local

municipalities, ranges between 30 and 16,000 ha, which are typically dominated by few grass species with scattered trees. The major plant species are *Miscanthus sinensis* (*M. sinensis*) and *Pleiblastus chino* (*P. chino*) var. *viridis*, which are perennial C₄ and C₃ plants, respectively, and grow to heights of 1–3 m and under 1 m. Both species grow from spring through fall and wither in winter. After withering, *M. sinensis* retains most of its leaves on the stem, whereas *P. chino* var. *viridis* sheds its leaves in late fall. These plant species are commonly found throughout Japan (e.g., on the roadside, in mountainous areas, and in grasslands) and are widely distributed across East Asia.

3 Hg flux in grasslands

3.1 Dry/wet deposition

In areas far from Hg sources, the atmosphere is the primary supplier of Hg to the terrestrial surface. GEM, GOM, and PBM in the air are transported downward and eventually collide with and deposit onto the terrestrial substrates. In this study, we define this abiotic Hg accumulation as dry deposition and distinguish it from plant uptake, which is sometimes also considered dry deposition. Hg deposited via precipitation is defined as wet deposition. Understanding the contributions of GEM, GOM, and PBM depositions to the fate of atmospheric Hg remains a challenging aspect of Hg-cycle studies.

Currently, the atmospheric dry deposition rates of GEM under the background atmospheric concentrations are poorly understood because these rates cannot be directly measured using existing sampling techniques, which cannot distinguish between air-suspended and depositing GEM. Instead, the dry deposition rate of GEM is estimated using theoretical calculations that apply observed vertical concentration gradients (also referred to as flux measurements) and estimated deposition velocities (Zhang et al., 2009; Zhang et al., 2016). However, Hg flux measurements themselves are also difficult because of the bidirectional transport of GEM, which involves both dry deposition and evasion (Gustin et al., 2008).

GEM both deposits onto and evades from the boundary of the substrate surface. Wang et al. (2003) observed a strong correlation between GEM and soil Hg concentrations, along with a positive relation between elevated Hg levels in the air or soil and the proximity to factory emission sources, indicating the dry deposition of high-concentration GEM. The concentration ratios of atmospheric GEM and soil Hg at the closest receptor site to that at the most distant receptor site were 5 and 40, respectively. The difference, however, cannot be explained by the dry deposition of GEM only, which is proportional to the atmospheric concentration of the species of interest. The contribution of GOM produced during the 4 km transport might be part of the explanation, but the production of GOM during such a short transport time remains uncertain. Atmospheric oxidation rates of GEM are under debate (Ariya et al., 2015). Laboratory studies exposing soil to 0 ng m⁻³–170 ng m⁻³ GEM air also demonstrated that GEM dry deposition was nonlinear with respect to GEM concentration (Xin et al., 2007) and that deposition may not occur under low GEM concentrations. Hg flux studies in the U.S. grasslands

(Ericksen et al., 2006) reported net Hg emissions, supporting the above laboratory findings. In contrast, long-term studies of GEM deposition on the prairie soil showed seasonal variations in dry deposition: deposition during winter and emission during other seasons (Obrist et al., 2005; Stamenkovic et al., 2008). Dry deposition on soils in grasslands across global regions may vary, and further investigation is needed to identify the controlling factors in addition to seasonal dependence.

GOM is an ionic form (mostly Hg^{2+}) and is, therefore, believed to readily form chemical bonds with other substances and ions, making it prone to adhere to many substrate surfaces. Owing to this nature and its low vapor pressure, GOM deposition is considered unidirectional unless GOM is reduced to GEM. GOM is typically measured using the Tekran 2537A/1130/1135 mercury speciation analyzer unit (Tekran Instruments Corp., Toronto, Canada), which also sequentially analyzes GEM and PBM (Gustin et al., 2019). The measurement results are then used to estimate the dry deposition of GOM. Lyman et al. (2007) evaluated this approach by conducting *in situ* indoor (greenhouse) air measurements using the mercury speciation unit and the offline analysis of water-soluble Hg in water used to rinse the leaf surface of two plant species, aspen and sagebrush, which were raised in the greenhouse for several months. The results demonstrated almost insignificant deposition rates of GOM, while the modeled deposition based on airborne GOM measurements showed significant GOM deposition. Some reports have shown that GOM measurements using the speciation monitor tend to yield significantly lower concentrations than those determined via cation exchange membrane filter sampling (Gustin et al., 2019; Gustin et al., 2015), implying that the discrepancy between theoretical and observed GOM depositions reported by Lyman et al. might have been even larger had they used cation exchange membrane filters to monitor GOM concentrations. Further studies are needed to investigate the poorly understood mechanisms of GOM deposition.

PBM dry deposition is considered unidirectional. Its deposition processes are believed to be similar to those of other particulate matter, depending on gravitational settling, aerodynamic transport, and surface uptake (Seinfeld and Pandis, 1998). Among GEM, GOM, and PBM, PBM typically has the lowest concentration (5% or less), often near the measurement uncertainty. Consequently, PBM dry deposition is generally regarded as having a minor impact on total dry deposition, with the dry deposition of atmospheric Hg mostly being governed by GEM.

Wet deposition of Hg is also considered unidirectional over short temporal scales, and its wet deposition rate is determined by analyzing Hg concentrations in precipitation collected using a funnel with a defined horizontal cross-sectional area over a specific period. Hg in precipitation exists mostly in oxidized forms (Hg^{2+}), typically with only a few percent or less as methylmercury (Ahmed et al., 1987; Guo et al., 2008; Marumoto and Matsuyama, 2014; Qin et al., 2016). These ions often form complexes with organic and inorganic ligands and can be reduced to elemental Hg under sunlight (Amyot et al., 1997), necessitating the use of chemical stabilizers during precipitation sampling (Ahmed et al., 1987). In addition, post-sampling changes in precipitation volume due to water evaporation can alter Hg concentration measurements. Wet deposition is an important pathway in the fate of atmospheric Hg and a major source of soil Hg in remote

areas. The wet deposition rates of Hg measured at various locations far from Hg sources have exhibited both negative (Fang et al., 2013) and positive (Prestbo and Gay, 2009) correlations between Hg concentrations and precipitation amounts, possibly reflecting the proximity of the receptor sites to stationary emission sources. However, redox chemistry in the atmosphere, particularly the conversion process of elemental to oxidized Hg, is likely involved during the long-range transport of elemental Hg. To the best of our knowledge, this remains unclear and is a major research theme that scientists continue to investigate.

3.2 Evasion from soil

Soil is the largest reservoir of Hg in pristine ecosystems (Gustin et al., 2008; Krabbenhoft et al., 2005; Mason et al., 1994; Lindqvist, 1991). However, under certain conditions, this largest reservoir can become a source of atmospheric Hg emissions. Many flux studies over bare soils (Choi et al., 2009), wetlands (Poissant et al., 2004), deserts (Ericksen et al., 2006), forest floors (Ericksen et al., 2006), grasslands (Ericksen et al., 2006), and rice paddies (Zhang et al., 2024) have demonstrated the overall flux of TGM emissions from the soil. Studies comparing Hg emissions from sterilized and non-sterilized soils exhibited only small differences, indicating that bacterial activity in these specific soils has a limited influence on Hg emission (Choi et al., 2009). To date, reported factors determining the direction of Hg flux at the soil surface include soil temperature (Poissant et al., 2004; Choi et al., 2009; Moore and Castro, 2012), air temperature (Stamenkovic et al., 2008), soil moisture (Wallschläger et al., 1999; Frescholtz and Gustin, 2004; Ericksen et al., 2006; Park et al., 2014; Xin et al., 2007), light irradiation (Frescholtz and Gustin, 2004; Moore and Carpi, 2005; Choi et al., 2009; Park et al., 2014; Xin et al., 2007; Poissant and Casimir, 1998; Schlüter, 2000), the extent of litter cover (which suppresses direct sunlight exposure) (Choi et al., 2009; Stamenkovic et al., 2008), and plant canopy greenness above the soil (Stamenkovic et al., 2008). These specific factors are seasonally dependent; therefore, long-term monitoring of the net air-surface exchange might be appropriate to evaluate Hg fluxes in grasslands.

3.3 Plant uptake and emission

Hg flux studies over vegetated fields indicate Hg emissions (Luo et al., 2016; Lindberg et al., 1979; Lindberg et al., 2002; Yuan et al., 2019) and, at times, net Hg emission and a sink of zero (Fritsche et al., 2008b). However, multiple reports document Hg uptake by plant species, such as Hg detection in foliage (Millhollen et al., 2006; Assad et al., 2016), crop and grass leaves (Niu et al., 2013; Yin et al., 2013; Meng et al., 2018), and depositional flux over the vegetated fields (Lodenius et al., 2003; Poissant et al., 2008; Niu et al., 2011). Measurements of GEM and CO_2 concentrations at multiple locations at the same time in the Northern Hemisphere clearly demonstrate seasonal variations in GEM concentrations corresponding to CO_2 cycles driven by plant photosynthesis (Jiskra et al., 2018). Diel variations in atmospheric GEM observed in a temperate forest in China also provide strong evidence that plant uptake and efflux are involved in these

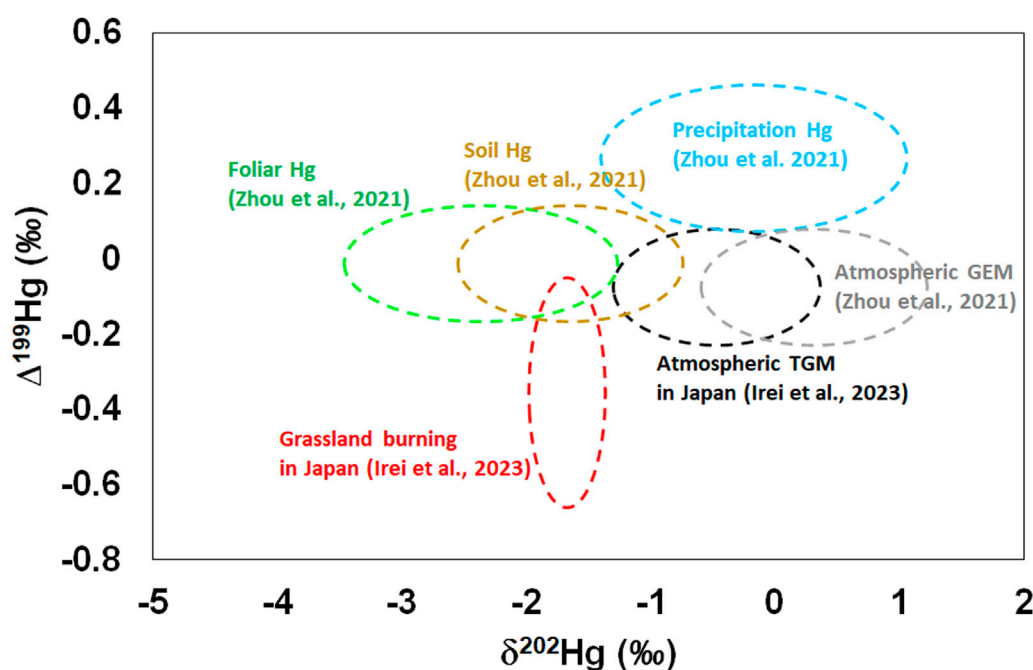


FIGURE 1

Variations of $\delta^{202}\text{Hg}$ and $\Delta^{199}\text{Hg}$ reported for Hg from different sources. Ellipses represent Hg from foliar (green), soil (brown), precipitation (blue), atmospheric GEM (gray), atmospheric TGM (black), and grassland burning in Japan (red). Data reported by Zhou et al., 2021, and Irei et al., 2023 were adopted.

variations (Fu et al., 2016). These observations were made at forest sites, and to the best of our knowledge, such apparent trends have not yet been observed in grassland studies. Plant uptake of atmospheric GEM likely relies on the plant species and leaf age. Identifying plant species that assimilate atmospheric GEM and their spatial distribution is critical for advancing our understanding of the Hg cycle.

3.4 Biomass burning

During biomass burning, Hg assimilated into and adhered to the surfaces of standing plants and litterfall is re-emitted into the atmosphere. Moreover, Hg on the ground surface may volatilize back into the air. Nriagu and Pacyna (1988) first reported the importance of Hg emissions from biomass burning. Since then, Hg emissions from forest fires have been reported at various locations, such as the Amazon region (Veiga et al., 1994; Albernaz et al., 2010), the São Paulo–Santiago area, South America (Ebinghaus et al., 2007), Cape Peninsula, Africa (Brunke et al., 2001), forests in Ontario (Friedli et al., 2003) and Québec, Canada (Sigler et al., 2003), near the Rocky Mountains, United States (Biswas et al., 2007), the Mediterranean region (Cinnirella and Pirrone, 2006), and long-range transported biomass-burning plumes along the Pacific coast, United States (Weiss-Penzias et al., 2007). As estimated, during 1997–2006, biomass burning globally accounted for 8% of the total atmospheric Hg emissions (anthropogenic and natural sources) (Friedli et al., 2008). Recently reported TGM emissions from prescribed burning in Japanese grasslands (Irei, 2022) were not included in this estimation.

Considering the countless small-scale prescribed burnings worldwide and the recent large-scale wildfires triggered by global warming, the proportion of these emissions may increase in the future.

4 Tracing the origin of Hg using the stable Hg isotope ratio

Naturally occurring Hg contains seven stable isotopes: ^{196}Hg , ^{198}Hg , ^{199}Hg , ^{200}Hg , ^{201}Hg , ^{202}Hg , and ^{204}Hg . Hg isotope ratios are expressed as follows:

$$\delta^x\text{Hg} (\%) = \left[\frac{\left(\frac{x\text{Hg}}{^{198}\text{Hg}} \right)_{\text{Sample}}}{\left(\frac{x\text{Hg}}{^{198}\text{Hg}} \right)_{3133}} - 1 \right] \times 1000,$$

where x denotes the stable mercury isotope of mass x and the bracketed isotope ratios with subscripts “sample” and “3133” refer to the stable mercury isotope ratios of mass x relative to mass 198 for the sample and NIST SRM 3133, respectively. $\delta^x\text{Hg}$ values are then converted to $\Delta^x\text{Hg}$ values using the following equation to evaluate mass-independent fractionation of Hg isotopes (Blum and Bergquist, 2007):

$$\Delta^x\text{Hg} (\%) \approx \delta^x\text{Hg} - (\delta^{202}\text{Hg} \times \beta_x),$$

where β_x is the mass-dependent fractionation factor for the isotope of mass x .

$\delta^{202}\text{Hg}$ and $\Delta^{199}\text{Hg}$ of Hg in the air, rain, soil, and plant foliage are distinct (Demers et al., 2013; Yu et al., 2016; Zhou et al., 2021;

Jiskra et al., 2019). The mechanisms behind their isotope fractionations are intriguing, but remain unknown. Despite this, the distinct isotope signatures hold potential for tracing the origins of Hg. Figure 1 shows an example plot of the ranges observed for $\delta^{202}\text{Hg}$ and $\Delta^{202}\text{Hg}$ for TGM in background air and plumes from prescribed grassland burning in Japan, together with literature values (Zhou et al., 2021) for Hg in precipitation, soil, and atmospheric GEM observed. The majority of $\delta^*\text{Hg}$ studies of Hg in precipitation and atmospheric GEM or TGM consistently show similar $\delta^*\text{Hg}$ and $\Delta^*\text{Hg}$ distributions (Demers et al., 2013; Yu et al., 2016; Zhou et al., 2021; Jiskra et al., 2019; Zheng et al., 2016; Jiskra et al., 2015), likely due to a homogeneous GEM mixture in the atmosphere of the Northern Hemisphere. $\delta^*\text{Hg}$ of total gaseous Hg collected from the prescribed grassland burning showed distinct values, which reflected Hg in plants and/or Hg deposited on the surface. The combination of flux studies with isotope ratio measurements provides insights into air–surface Hg exchange.

5 Future perspectives

Grasslands remain understudied in terms of the Hg cycle despite their extensive terrestrial coverage. Previous studies combining Hg flux measurements and stable Hg isotope analysis are even scarcer than forest studies. The ease of above-ground biomass surveys and the considerably uniform canopy heights make terrestrial Hg flux studies in grasslands valuable for examining the physical deposition and plant uptake of atmospheric Hg. These studies provide insights that can help enhance our understanding of processes that Hg undergoes in the natural environment and evaluate the effectiveness of the Minamata Convention on Hg.

Author contributions

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