



Gaseous mercury fluxes from forest soils in response to forest harvesting intensity: A field manipulation experiment



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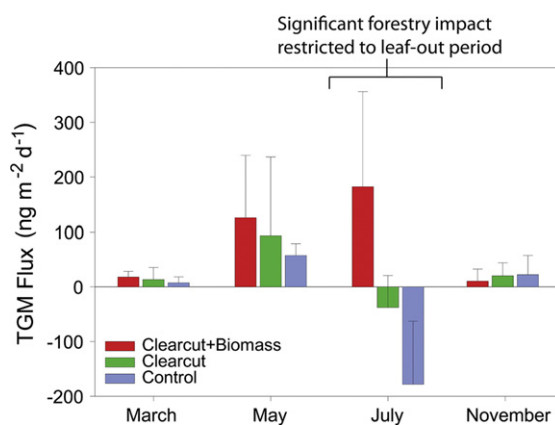
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HIGHLIGHTS

- We investigated forestry impacts on gaseous Hg flux in a field experiment.
- We measured ambient Hg fluxes and fluxes from added enriched Hg isotope tracers.
- Biomass harvesting following clearcut had the greatest impact on ambient fluxes.
- These impacts were seasonally restricted to the leaf-out growing season.
- Isotope results suggest emissions dominated by legacy Hg pools.

GRAPHICAL ABSTRACT



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ABSTRACT

Forest harvesting leads to changes in soil moisture, temperature and incident solar radiation, all strong environmental drivers of soil–air mercury (Hg) fluxes. Whether different forest harvesting practices significantly alter Hg fluxes from forest soils is unknown. We conducted a field-scale experiment in a northern Minnesota deciduous forest wherein gaseous Hg emissions from the forest floor were monitored after two forest harvesting prescriptions, a traditional clear-cut and a clearcut followed by biomass harvest, and compared to an unharvested reference plot. Gaseous Hg emissions were measured in quadruplicate at four different times between March and November 2012 using Teflon dynamic flux chambers. We also applied enriched Hg isotope tracers and separately monitored their emission in triplicate at the same times as ambient measurements. Clearcut followed by biomass harvesting increased ambient Hg emissions the most. While significant intra-site spatial variability was observed, Hg emissions from the biomass harvested plot ($180 \pm 170 \text{ ng m}^{-2} \text{ d}^{-1}$) were significantly greater than both the traditional clearcut plot ($-40 \pm 60 \text{ ng m}^{-2} \text{ d}^{-1}$) and the unharvested reference plot ($-180 \pm 115 \text{ ng m}^{-2} \text{ d}^{-1}$) during July. This difference was likely a result of enhanced Hg²⁺ photoreduction due to canopy removal and less shading from downed woody debris in the biomass harvested plot. Gaseous Hg emissions from more recently deposited Hg, as presumably representative of isotope tracer measurements, were not significantly influenced by harvesting. Most of the Hg tracer applied to the forest floor became sequestered within the ground vegetation and debris, leaf litter, and soil. We observed a dramatic lessening of tracer Hg

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emissions to near detection levels within 6 months. As post-clearcutting residues are increasingly used as a fuel or fiber resource, our observations suggest that gaseous Hg emissions from forest soils will increase, although it is not yet clear for how long such an effect will persist.

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1. Introduction

Gaseous mercury (Hg) is a problematic pollutant because its relatively long atmospheric lifetime allows for long-range transport and deposition in remote locations, where it has the potential to be methylated and subsequently accumulated in biota (Schroeder and Munthe, 1998). Mercury in soil, whether present due to geologic enrichment or atmospheric deposition, can be reduced to volatile elemental mercury (Hg^0) and emitted to the atmosphere through a number of complex processes (Lin et al., 2010). Soil Hg concentrations and mineralogy control the quantity of Hg available for emission (Miller et al., 2011). Soil emissions have been shown to vary temporally in response to numerous environmental variables, including soil moisture (Gustin and Stamenkovic, 2005; Song and Van Heyst, 2005; Xin et al., 2007), air temperature (Kim et al., 2012; Lin et al., 2010), soil temperature (Johnson et al., 2003; Magarelli and Fostier, 2005; Moore and Castro, 2012; Sigler and Lee, 2006; Zhang et al., 2001), atmospheric Hg concentrations (Kuiken et al., 2008b; Wallschlagler et al., 2002), and incident solar radiation (Carpi and Lindberg, 1998; Converse et al., 2010; Gustin et al., 2002; Moore and Carpi, 2005; Poissant and Casimir, 1998; Stamenkovic and Gustin, 2007; Xin et al., 2006). Typically, several of these variables act synergistically in their impact on Hg emission.

Several studies have found that incident solar radiation is the single strongest control on gaseous Hg emissions (Erickson et al., 2005; Gustin et al., 2002; Kuiken et al., 2008a). Where solar radiation is able to penetrate soil it facilitates photo-reduction of soil bound Hg^{2+} to gaseous Hg^0 , which is then emitted to the atmosphere from the soil surface (Carpi and Lindberg, 1997; Gustin et al., 2002; Moore and Carpi, 2005). Hg fluxes tend to be highest during periods of peak solar inputs. However even during moderate levels of solar radiation (as little as 70 W m^{-2}), Hg emission from soil can continue (Gustin et al., 2006; Kuiken et al., 2008a). As solar radiation heats soils and air, strong associations have also been identified among soil temperature, air temperature, and gaseous Hg emissions (Kim et al., 2012; Lin et al., 2010; Magarelli and Fostier, 2005; Moore and Castro, 2012; Zhang et al., 2001) because these factors effectively lower the activation energy for Hg reduction ($18.0\text{--}24.9 \text{ kcal mol}^{-1}$ – Carpi and Lindberg, 1998). Increasing soil moisture may further enhance (through upward transport of Hg via capillary action) or suppress (saturated soils reduce photo-reduction efficacy) Hg emissions depending on the degree of wetting (Gustin and Stamenkovic, 2005; Johnson et al., 2003; Lin et al., 2010; Song and Van Heyst, 2005).

Mercury emissions from forest soils vary seasonally in response to changes in canopy cover, which affects solar radiation, temperature, and soil moisture (Choi and Holsen, 2008; Kuiken et al., 2008a). Estimates of the solar radiation that penetrates mature deciduous canopies are between 20 and 58%, thus Hg fluxes from forest soils are typically lower in the growing season than in leaf-free seasons (Denkenberger et al., 2012; Gustin et al., 2004; Gustin and Stamenkovic, 2005; Kuiken et al., 2008a; Schroeder et al., 2005; Sigler and Lee, 2006). Higher magnitude Hg emissions in the spring and autumn typically occur due to the lack of foliage and therefore higher proportion of incident radiation able to penetrate to the forest floor (Choi and Holsen, 2008; Kuiken et al., 2008a, 2008b). Fluxes from soils in most northern deciduous forests are considered negligible in the winter due to persistent snow cover throughout most of the season (Denkenberger et al., 2012). Currently there are few published observations of Hg fluxes from forests through seasons (Choi and Holsen, 2008; Kuiken et al., 2008a). As such the regulation of Hg fluxes by forest vegetation is relatively poorly documented (Schroeder et al., 2005).

North American deciduous forests are an important source of timber for pulp and paper production and disturbance of forest vegetation via harvesting is common in many regions. Clearcutting involves the complete removal of stems suitable for the production of wood-based products from a forested area, resulting in a large open area. Woody debris (material not economically suitable for the production of wood based products, including branches, underbrush, and small trees) is typically left on-site. Recently however, with increased incentive for sustainable harvesting and the development of new uses for woody biomass (e.g. for renewable fuels as has been legislated in the 2007 U.S. Energy Independence and Security Act or as a fiber resource), removal of woody debris after harvesting has increased (Janowiak and Webster, 2010). Though such techniques make more efficient use of woody biomass, the removal of more woody material from clearcut sites may ultimately increase the intensity of disturbance to these sites, removing shading and potentially increasing Hg emissions. Studies examining the direct impacts of forestry operations on soil Hg fluxes are very limited (Magarelli and Fostier, 2005). Currently, the only work performed on this topic has been from tropical rainforests in South America where Hg emissions have been shown to increase in response to deforestation (Almeida et al., 2009; Magarelli and Fostier, 2005). However these results may not be directly transferable to the forestry sector in higher latitude climates.

Enriched stable Hg isotope tracers (herein Hg tracers) are useful in the determination of Hg fate within environmental systems (Hintelmann and Evans, 1997). Several studies have utilized Hg tracers to examine the fate of contemporary deposition on soil surfaces (Erickson et al., 2005; Xin et al., 2007); however, few measurements of emitted Hg tracers have been made in forested environments and none have looked at the impacts of forest harvesting (Hintelmann et al., 2002; Munthe et al., 2001). These studies have shown that added Hg tracers (presumably representing recently deposited Hg) quickly become sequestered into the soil (Erickson et al., 2005; Hintelmann et al., 2002; Munthe et al., 2001). Once the Hg has infiltrated into the soil column with rainwater, its ability to be emitted is greatly reduced due to the lack of solar radiation, which only penetrates a very shallow depth below the soil surface (Xin et al., 2007; Carpi and Lindberg, 1997). Of the Hg tracer emitted back to the atmosphere, some (0.1 to 3%) is emitted within 24 h of deposition (Erickson et al., 2005; Xin et al., 2007). Emissions quickly decline in weeks to months following deposition, to barely detectable levels and on an annual basis account for only ~6–8% of the original amount deposited (Erickson et al., 2005; Hintelmann et al., 2002). Studies that have scaled surface Hg emissions annually have shown that forests (and other natural background surfaces) are a large net sink for atmospheric Hg, with inputs from wet and dry deposition exceeding levels of Hg emissions (Hartman et al., 2009; Kuiken et al., 2008a; Denkenberger et al., 2012). Given the net accumulation of Hg in forest ecosystems, it is critical to understand the potential impact of clearcutting and biomass harvesting on surface–air Hg flux dynamics.

This study aims to determine the effects of two common forms of forest harvesting, clearcutting and clearcutting with additional biomass (woody debris) removal, on Hg fluxes from forest soils. This study also expands our current understanding of seasonal Hg emission variability and the fate of contemporary Hg deposition to soils in northern forested environments. We analyzed ambient gaseous Hg emissions across seasons to elucidate some of the processes controlling observed Hg emissions. Additionally we used a field-scale enriched stable Hg isotope tracer application to represent the fate of newly deposited Hg in forest soils. We expected that both ambient and tracer Hg emissions would

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