



Spatial and vertical distribution of mercury in upland forest soils across the northeastern United States



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ABSTRACT

Assessing current Hg pools in forest soils of the northeastern U.S. is important for monitoring changes in Hg cycling. The forest floor, upper and lower mineral horizons were sampled at 17 long-term upland forest sites across the northeastern U.S. in 2011. Forest floor Hg concentration was similar across the study region ($274 \pm 13 \mu\text{g kg}^{-1}$) while Hg amount at northern sites ($39 \pm 6 \text{ g ha}^{-1}$) was significantly greater than at western sites ($11 \pm 4 \text{ g ha}^{-1}$). Forest floor Hg was correlated with soil organic matter, soil pH, latitude and mean annual precipitation and these variables explained approximately 70% of the variability when multiple regressed. Mercury concentration and amount in the lower mineral soil was correlated with Fe, soil organic matter and latitude, corresponding with Bs horizons of Spodosols (Podzols). Our analysis shows the importance of regional and soil properties on Hg accumulation in forest soils.

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1. Introduction

Mercury (Hg) is a global pollutant and is of particular concern to humans in the northeastern U.S. who consume local fish, as most states in the region have fish consumption advisories due to their tissue exceeding levels deemed hazardous by the U.S. Environmental Protection Agency (USEPA, 2011). Mercury is also a threat to wildlife in general; piscivorous birds, for example, have shown multiple signs of chronic Hg poisoning (Evers et al., 2004; Driscoll et al., 2007). Forest soils are a large, potentially mobile reservoir that may supply Hg to aquatic ecosystems (Aastrup et al., 1991; Lorey and Driscoll, 1999; Grigal et al., 2000; Schwesig and Matzner, 2001; Driscoll et al., 2007). This is important for the northeastern U.S., as forest soils receive comparatively elevated deposition rates of Hg and are widely forested (Rea et al., 2002; Miller et al., 2005). The transfer of Hg from forest soils to aquatic ecosystems in this region may be altered by future shifts in regional emissions and changes in climate. Mercury deposition rates to this region may be reduced as a result of new measures adopted by the

United States Environmental Protection Agency (USEPA) to reduce coal-fired power plant Hg emissions to 30% of current values by 2025 (Han et al., 2008; USEPA, 2011). The forest soil Hg pools must first be characterized in order to establish a baseline upon which to compare with future assessments.

The forest floor, the organic horizons overlying the mineral soil, is a characteristic feature of forest soils and is dominated by decomposing litter and woody debris. The forest floor is a strong accumulator of certain atmospherically-deposited metals and has been used to assess the distribution and deposition of other metals such as lead (Johnson et al., 1982; Kaste et al., 2006; Steinnes and Friedland, 2006; Kaste et al., 2011). However, the mineral soil often contains greater quantities of metals than the forest floor due to its greater volume (Grigal, 2003; Gabriel and Williamson, 2004; Stankwitz et al., 2012). Mercury distribution in the forest floor and mineral soil has been shown to be dependent on both regional abiotic factors and soil properties (McNeal and Rose, 1974; Nater and Grigal, 1992; Lorey and Driscoll, 1999; Grigal, 2003; Demers et al., 2007; Obrist et al., 2011; Stankwitz et al., 2012; Shi et al., 2013). It is unclear if the distribution of Hg in the forest floor across the northeastern U.S. is dependent on the same regional factors and soil properties as identified by previous Hg studies in

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other regions, at different scales. In addition, few studies have conducted a spatial approach to study Hg in the mineral soil.

At the regional scale, Hg distribution has been reported to be dependent on soil temperature and precipitation (Nater and Grigal, 1992; Smith-Downey et al., 2010; Obrist et al., 2011; Tipping et al., 2008). Soil temperature may affect Hg accumulation in soils by controlling soil organic matter (SOM) dynamics among other processes. The dependence of Hg accumulation on SOM in forest soils has been described by Obrist et al. (2011) as the 'legacy effect'. In the legacy effect, SOM at colder climates has a slower decomposition rate and is exposed to a longer duration of Hg deposition, and accumulates more Hg. Another temperature dependent process is Hg⁰ volatilization, which is greater at higher soil temperatures (Schlüter, 2000). Furthermore, precipitation may also influence Hg accumulation because wet deposition is a major pathway by which Hg reaches the forest soil (Rea et al., 2002; Demers et al., 2007; Obrist et al., 2011; Juillerat et al., 2012; Stankwitz et al., 2012). In addition, physical and chemical soil properties have also been shown to influence Hg accumulation in soils (Grigal, 2003; Gabriel and Williamson, 2004; Demers et al., 2007). Soil organic matter, soil pH, clay-sized particle abundance and metal oxides can influence the surface sorption of Hg in soils (Schuster, 1991; Yin et al., 1996; Roulet et al., 1998; Han et al., 2003; Grigal, 2003; Gabriel and Williamson, 2004; Liao et al., 2009; Obrist et al., 2011; Shi et al., 2013).

It is unclear which regional factors (latitude, longitude, climate, mean annual precipitation) or soil properties (SOM, pH, Fe, clay) are important for Hg accumulation in the forest soils of the northeastern U.S. The objectives of this study were to determine the spatial and vertical distribution of Hg in the forest floor and mineral soil at seventeen long-term forest research sites across the northeastern U.S. and identify the regional and soil properties that explain the pattern of Hg accumulation.

2. Materials and methods

2.1. Site descriptions

Twenty-five long-term, upland forest research sites were established in 1980 as part of a larger study on metals in the forest floor (Johnson et al., 1982). Sites were situated in undisturbed forests, mostly in higher elevation regions, with many located within National and State forests. Seventeen of the twenty-five original sites were relocated using GPS and resampled in 2011 and are shown in Fig. 1. Sites were grouped into three sub-regions: western, central and northern (Fig. 1). The range of elevations at each sub-region was not significantly different for the three sub-regions. Site elevation ranged from 305 to 820 m (Table 1) with a mean of

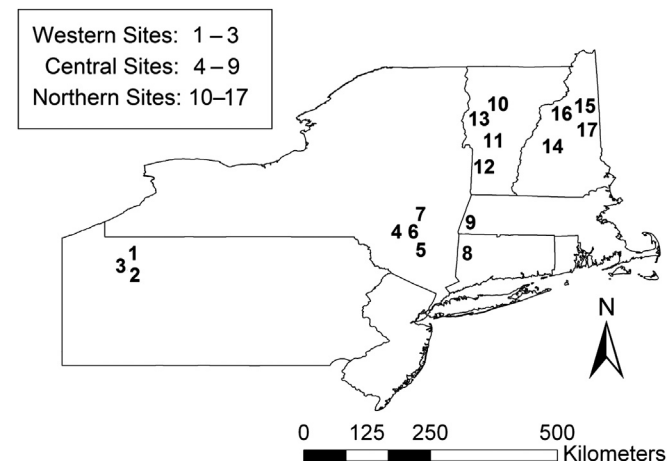


Fig. 1. Location of the upland forest, long term research sites and their sub-region across the northeastern United States.

562 ± 35 m. The soil temperature regime for each site was determined from mapped soils on Web Soil Survey (Soil Survey Staff). Soils were from the frigid (mean annual soil temperature < 8 °C) and mesic (mean annual soil temperature > 8 °C) temperature regime. Northern sites were primarily frigid while western sites were all mesic (Table 1). Precipitation for each site was interpolated from the mean annual precipitation (1981–2010) spatial model from the PRISM climate group (Prism database; PRISM Climate Group, 2012).

Vegetation at each site was mixed; sites ranged from primarily deciduous vegetation such as oak (*Quercus* spp.), beech (*Fagus* spp.), maple (*Acer* spp.), and birch (*Betula* spp.), to coniferous such as pine (*Pinus* spp.), spruce (*Picea* spp.), and hemlock (*Tsuga* spp.) (Table 1). Northern sites were generally more conifer-dominated while western and central sites were deciduous-dominated. The soils were developed from glacial till, outwash deposits, or outcrops of weathered bedrock (c.f. Siccama, 1974; Kaste et al., 2006). The soils were well-drained and on level to shallow slopes (<12%). Soils were primarily classified as either Spodosols (Podzols in FAO/UNESCO) or Inceptisols (Cambisols in FAO/UNESCO) (Table 1), except Site #1 and 3, which were classified as Ultisols (Acrisol in FAO/UNESCO). In general, western sites were Ultisols, central sites were Inceptisols and northern sites were Spodosols (Table 1).

2.2. Soil collection

Soils were sampled in a 30 m by 30 m plot at each site between July and September 2011 (Fig. 1). Sites were relocated using GPS and instructions from previous investigators. Five forest floor (Oi + Oe + Oa) and upper mineral soil (A or E horizons) were collected from all sites. Lower mineral soil (Bw or Bs horizons) was sampled at most sites except sites 4, 6, 7, 10, and 17 due to hardpan layer, extreme rock content, or lithic contact. The forest floor sampling technique was the same as those described by Johnson et al. (1982). In brief, five 15 × 15 cm square sections of forest floor were separated from the underlying mineral horizons and collected. In an adjacent location, a pit was excavated to allow access to the mineral soil. To avoid mixing or contaminating samples, the lower mineral horizon was sampled from the soil pit face using hand trowels first, followed by the upper mineral soil. Upper mineral soils were classified as either an A or E horizon and lower mineral soils were classified as either Bw or Bs horizon following the USDA Soil Taxonomy guidelines (Soil Survey Staff, 2010). High-density polypropylene tubes were used to collect intact soil cores samples for bulk density measurements.

2.3. Soil processing

Forest floor and mineral soil samples were air-dried to a constant weight and roots >5 mm in diameter were removed. The forest floor total mass was calculated using oven-dried sub-samples and the volume was calculated using the area of the template and measured depths. Soil mass was calculated using the bulk density and the mass of the sieved, oven-dried 100 °C sample. Forest floor and mineral soil samples were milled and sieved, respectively, to <2 mm. Mineral soil mass was corrected for rock and coarse fragments using bulk cores and visual soil pit estimates. It should be noted that both methods may underestimate the rock fraction because soil pits and soil cores could not include cobbles and boulders, leading to an overestimation of total soil mass (Huntington et al., 1988). To determine the soil pH, 4 g of soil was added to 10 g of water for a 2:5 soil–water gravimetric ratio and shaken for 1 h using a wrist-action shaker and allowed to settle for 10 min. The soil pH of the supernatant was measured using a pH meter (8015 VWR) (Table 2). Loss on ignition was used to estimate the % SOM present in the samples (Table 2). For loss on ignition, 4 g of soil was held at 475 °C for 8 h and % SOM was determined from change in mass from thermal oxidation. Soil particle size distribution was determined using a modified Bouyoucos hydrometer method (Gee and Bauder, 1986). For the process, 30 g of soil were treated with 30% w/w hydrogen peroxide to oxidize SOM aggregates and dispersed with 100 mL of 0.08 M sodium hexametaphosphate overnight. Soil pH, % SOM, and % Clay are given for each sub region and soil horizon in Table 2.

2.4. Metal analyses

The U.S. EPA method 3051A was used to quantify Hg through direct digestion. Homogenized 2 g sub-samples of organic and mineral soil samples were dried to a constant weight at 45 °C for 5 days. 250 mg (±1 mg) sub-samples were digested with 5 mL of a 1:9 ratio of trace metal grade hydrochloric acid:nitric acid (HNO₃, 70%; HCl, 70%). The digest solutions were allowed to de-gas overnight in lightly sealed 50 mL polypropylene centrifuge tubes. The solutions were then heated using a CEM MARS digestion system at 90 °C for 45 min (CEM Mathews, NC). After cooling, the digested samples were brought to a 50 mL final volume with DI water. Digested O horizon samples were filtered using 0.45 μm polypropylene Whatman syringe filters. The digests were diluted a further 10× with deionized water and then analyzed by an Agilent 7500 series ICP-MS (Agilent Technologies Santa Clara, CA). With every 20 digested samples we included: one randomly spiked sample with 50 μL of 1 ppm HgCl₂ (3.7 nM HgCl₂); one replicate; one preparation blank; and one standard reference material (SRM). Peach leaves SRM 1547 and Montana soil SRM 2711 from the National Institute of Standards and Technology (National Institute of Standards

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