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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 ± 13 μg kg^{-1}) while Hg amount at northern sites (39 ± 6 g ha^{-1}) was significantly greater than at western sites (11 ± 4 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 Hg^0 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
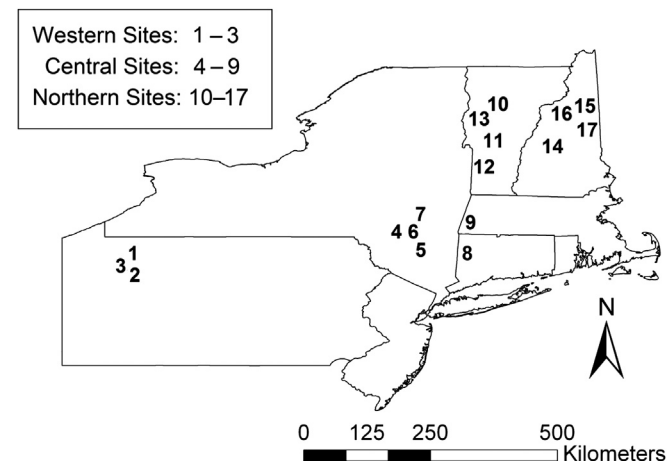

Fig. 1. Location of the upland forest, long term research sites and their sub-region across the northeastern United States.

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 (HNO_3, 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 $HgCl_2$ (3.7 nM $HgCl_2$); 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

Table 1
Site location, vegetation type, and soil taxonomy.

Sub-region[a]	Site	Site name	Elev.(m)	Mean annual precipitation[b] (mm yr^{-1})	Latitude	Longitude	Vegetation	Soil order	Soil temperature regime
W	1	Heart's Content,PA	580	1170	41.689	−79.252	Pine/Hemlock	Ultisol	Mesic
W	2	Cook's Forest, PA	430	1163	41.347	−79.212	Pine/Hemlock	Inceptisol	Mesic
W	3	Tionesta, PA	520	1141	41.477	−79.379	Oak	Ultisol	Mesic
C	4	Balsam Lake, NY	820	1268	42.067	−74.574	N. Hardwood	Inceptisol	Frigid
C	5	Mohonk, NY	366	1288	41.770	−74.158	N. Hardwood	Inceptisol	Mesic
C	6	Mt Tremper, NY	305	1365	42.071	−74.312	Pine/Hemlock	Inceptisol	Mesic
C	7	Windham, NY	580	1215	42.301	−74.170	N. Hardwood	Inceptisol	Frigid
C	8	Mohawk, CT	503	1192	41.820	−73.297	Oak	Inceptisol	Frigid
C	9	Mt. Everett, MA	790	1276	42.102	−73.431	Oak/Pitch Pine	Inceptisol	Frigid
N	10	Appalachian Gap, VT	778	1533	44.211	−72.931	Spruce/Fir	Spodosol	Frigid
N	11	Sherburne Pass, VT	671	1467	43.662	−72.833	N. Hardwood	Spodosol	Frigid
N	12	Bromley, VT	625	1465	43.214	−72.967	N. Hardwood	Spodosol	Frigid
N	13	Bristol Cliffs, VT	555	1112	44.140	−73.064	Pine/Hemlock	Spodosol	Frigid
N	14	Mt Cardigan, NH	579	1306	43.645	−71.933	Spruce/Hemlock	Spodosol	Frigid
N	15	Valley Way, NH	433	1217	44.369	−71.287	Spruce/N. Hardwood	Spodosol	Frigid
N	16	Gale River, NH	440	1181	44.232	−71.608	N. Hardwood	Inceptisol	Frigid
N	17	Wildcat Mt, NH	590	1527	44.266	−71.238	Spruce/N. Hardwood	Inceptisol	Frigid

[a] W = western region, C = central region, N = northern region.
[b] Precipitation values interpolated from *PRISM database* (PRISM Climate Group, 2012).

and Technology Gaithersburg, MD) were used as reference Hg values for forest floor and mineral soil samples, respectively. All measured Hg concentrations for SRM materials were within 7% of their certified values. Recovery rates for spiked samples were >95%. Hg concentration in the preparation blank samples was below detection limits. The digests were also analyzed for extractable Fe using an IRIS Intrepid II XSP ICP-OES (Thermo Electron Franklin, MA). Iron concentrations were within 8% of the Peach leaves SRM 1547 and Montana soil SRM 2711 reported values.

2.5. Data analyses

Descriptive statistics for soil Hg and soil properties were calculated using Matlab (Matlab INC Natick, MA). Correlative relations between Hg concentration and amount with regional factors (mean annual precipitation, latitude and longitude) and soil properties (% SOM, pH, Fe concentration, % clay) were investigated using stepwise regressions and multiple regression in Matlab. Non-continuous data (e.g. soil temperature regime) were not included in the stepwise regression models. Variables from the stepwise regressions were linearly regressed with each other and the variables with R^2 values greater than 0.25 and a causal relationship were determined to covary and not included in the multiple regressions. The independent variables found to be significantly correlated with Hg concentration or Hg amount in the stepwise regression were used in multiple regression models and shown in

Table 2
Selected chemical properties of the forest soils by sub-region for the three depths sampled (±1 standard error). The value *n* is number of sites.

	n	Thickness cm	Soil pH[a]	% SOM[b] g g^{-1}	% Clay[c] g g^{-1}	Fe mg kg^{-1}	Fe kg ha^{-1}
Western Region							
Forest floor	3	3.2 ± 0.5	4.48 ± 0.35	52 ± 5	n.a.	9 ± 3	0.2 ± 0.1
Upper mineral soil	3	4.1 ± 0.6	4.43 ± 0.37	9 ± 2	11 ± 2	12 ± 6	6 ± 3
Lower mineral soil	3	5.9 ± 0.5	4.38 ± 0.32	5 ± 1	12 ± 3	15 ± 6	12 ± 6
Central Region							
Forest floor	6	6.9 ± 1.4	4.24 ± 0.10	67 ± 5	n.a.	6 ± 1	0.6 ± 0.2
Upper mineral soil	6	5.9 ± 0.4	4.10 ± 0.06	15 ± 4	12 ± 3	14 ± 4	6 ± 1
Lower mineral soil	4	9.7 ± 1.2	4.12 ± 0.19	9 ± 3	6 ± 2	16 ± 3	20 ± 1
Northern Region							
Forest floor	9	8.7 ± 1.8	3.97 ± 0.04	73 ± 3	n.a.	4 ± 1	0.7 ± 0.2
Upper mineral soil	9	6.4 ± 0.9	3.69 ± 0.03	9 ± 2	14 ± 2	12 ± 5	7 ± 3
Lower mineral soil	7	7.5 ± 0.6	4.13 ± 0.14	11 ± 1	12 ± 2	28 ± 2	25 ± 2

[a] measured in a 2:5 soil water extract.
[b] estimated from loss on ignition.
[c] determined by particle size distribution.

Table 3. The variation in Hg with soil order, and sub-region, were determined with the Kruskal–Wallis test. Post-hoc tests utilized include Student's two-sample *t*-tests and Wilcoxon rank signed tests. An alpha-level of 0.05 was used for all tests and mean values given in text and figures show ±1 standard error.

3. Results and discussion

3.1. Mercury in the forest floor

Forest floor Hg concentration ranged from 118 to 373 with a mean of 274 ± 13 µg kg^{-1} and did not vary significantly at the forested sites across the study region as shown in Fig. 2. These concentrations are similar to those reported by Evans et al. (2005), Obrist et al. (2011), and Juillerat et al. (2012) for forest floor samples from the northeastern U.S. Forest floor Hg amount ranged from 4 to 51 g ha^{-1} with a mean of 30 ± 4 g. The observed Hg amounts were similar to those reported by Evans et al. (2005) and Demers et al. (2007), with their values ranging from 26 to 46 g ha^{-1}. The total mass of Hg in the forest floor was significantly greater at northern sites (39 ± 6 g ha^{-1}) compared to western sites (11 ± 4 g ha^{-1}) ($P < 0.05$). The elevated concentration and amount of Hg in the forest floor in the northeastern U.S. has been considered to be largely anthropogenic, as human activities have significantly increased Hg deposition rates to forests (Fitzgerald et al., 1998; Grigal, 2003; Seigneur et al., 2003; Driscoll et al., 2007). Although

Table 3
Calculated multiple-regression model outputs for Hg concentration and amount with significant site characteristics and soil properties from stepwise-regressions for all 17 sites.

|	Variables	R^2	*P*
Forest floor			
Mercury concentration	% SOM, pH and latitude	0.77	<0.01
Mercury amount	Mean annual precipitation, pH and % SOM	0.65	<0.01
Upper mineral soil horizon			
Mercury concentration	Fe concentration	0.35	0.06
Mercury amount	Fe amount	0.21	0.38
Lower mineral soil horizon			
Mercury concentration	Latitude, % SOM and Fe concentration	0.80	< 0.01
Mercury amount	Latitude, % SOM and Fe amount	0.73	0.02
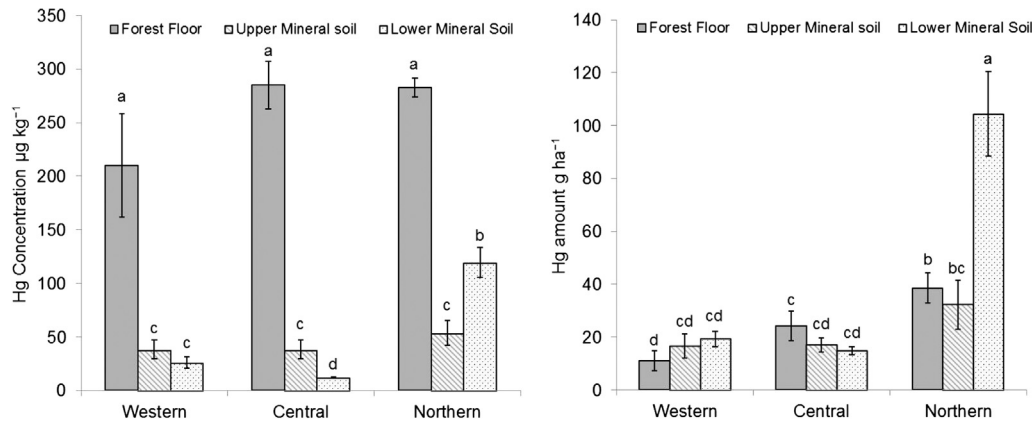
Fig. 2. The means ± 1 standard errors for Hg concentrations (μg kg^{-1}) and Hg amounts (g ha^{-1}) are shown for the three sub-regions. Letters group values by statistical significance ($P < 0.05$).

increased deposition due to human activity has been documented (e.g. Fitzgerald et al., 1998), the sources of Hg in this study were not distinguished and should be assumed to be both geogenic and anthropogenic.

The spatial distribution of Hg in the forest floor has been hypothesized to be dependent on both regional (latitude, longitude, climate, precipitation) and soil properties (SOM, pH, Fe, clay), with temperature and precipitation being the two dominant regional factors (Evans et al., 2005; Obrist et al., 2011). Soil temperature may influence Hg accumulation in the forest floor by controlling kinetics of chemical processes and SOM dynamics (Nater and Grigal, 1992; Grigal, 2003). When sites were grouped by their soil temperature regime, forest floor Hg concentration was not significantly different between mesic and frigid soils. However, frigid soils had greater forest floor Hg amount (33 ± 5 g ha^{-1}) compared to mesic forest floor (18 ± 5 g ha^{-1}) ($P < 0.05$). Further, frigid soils had a greater mean forest floor depth (10 ± 2 cm) compared to mesic soils (5 ± 1 cm) ($P < 0.05$).

The dependence of Hg on SOM dynamics has been observed in previous spatial studies (Nater and Grigal, 1992; Obrist et al., 2011; Stankwitz et al., 2012; Shi et al., 2013). The cooler frigid soil temperature regime may have promoted the accumulation of Hg through multiple mechanisms. Cooler forest floor temperatures generally lead to longer turnover times due to slower decomposition rates of SOM (Berg et al., 1993; Moore et al., 1999; Schwesig and Matzner, 2001; Obrist et al., 2011; Stankwitz et al., 2012). This legacy effect was further supported by Stankwitz et al. (2012), who observed greater Hg accumulation in soils with longer ^{210}Pb residence times. The forest floor at northern sites may have a corresponding longer turnover time than at western sites and may be exposed to longer periods of Hg deposition. Assuming that forest floor depth is roughly proportional to turnover rate regionally, then our results also may be a result of the legacy effect. Thus, it is hypothesized that the forest floor at northern sites has a longer turnover rate, was exposed to a longer duration of Hg deposition and has accumulated greater amounts of Hg.

Soil temperature may also influence Hg accumulation in the forest floor by controlling the kinetics of Hg^0 re-emission or volatilization (Schlüter, 2000; Schwesig and Matzner, 2001; Demers et al., 2007; Tipping et al., 2008). Volatilization has been recognized as an unconstrained pathway of Hg loss from soils (Johnson and Lindberg, 1995; Grigal et al., 2000; Grigal, 2003; Obrist et al., 2011; Demers et al., 2013). Estimates for Hg volatilization using mass balances have varied widely and ranged from 0 to 300 mg ha^{-1} yr^{-1} (Carpi and Lindberg, 1998; Grigal et al., 2000; Demers et al., 2007). Volatilization rates for Hg in northeastern

U.S. soils are poorly understood with few empirical studies in North America and volatilization may control local accumulation of Hg in soil (Demers et al., 2013). Demers et al. (2007) reported 95% of atmospherically deposited Hg was retained in forest soils of New York. The METALLICUS project in the Experimental Lakes Area of Ontario, Canada reported losses of 8% of Hg amended to soil (Hintelmann et al., 2002). Lindqvist et al. (1991) and Driscoll et al. (1994) both estimated that approximately 5% of the annual atmospherically deposited Hg to Spodosols in southern Sweden would be lost via volatilization. From these estimates, the influence of re-emission and volatilization of Hg may be considered limited but requires further attention to determine the effect of soil temperature on Hg accumulation in the forest soils.

Wet deposition is a major pathway in which Hg reaches forest soils and may explain the spatial variation of Hg in the forest floor (Tipping et al., 2008). Miller et al. (2005) has concluded that rural, upland forests of the northeastern U.S. receive elevated rates of Hg in precipitation, with estimates >120 mg ha^{-1} yr^{-1}. Further, total Hg deposition projected for these sites by data from Miller et al. (2005) suggest values from 200 to 350 mg ha^{-1} yr^{-1}. It must be noted that this study and Miller et al. (2005) have considered only Hg deposition estimates for forested ecosystems; land-use and vegetation cover significantly affects Hg deposition to soils and was not considered extensively in this study (Smith-Downey et al., 2010). For a rough estimate of wet deposition of Hg, the interpolated mean annual precipitation from the *PRISM database* was assumed to be proportional to the wet deposition of Hg (*Prism database*; PRISM Climate Group, 2012). The *PRISM database* does not account for many site-specific Hg deposition fluxes for total deposition such as litterfall (Hall and St. Louis, 2004), but may be used as a general estimate for wet Hg deposition (Tipping et al., 2008). The *PRISM database* is used by the National Atmospheric Deposition Program and Mercury Deposition Network for this purpose. When regressed, forest floor Hg concentration was found to be poorly correlated with mean annual precipitation. However, forest floor Hg amount was moderately correlated with interpolated mean annual precipitation amount with an R^2 value of 0.46 ($P < 0.05$) (Fig. 3). There are advantages and disadvantages with using interpolated mean annual precipitation amounts; 30 year averaged data is less susceptible to the problems associated with annual variations and error from sample collection. The primary disadvantage of using the interpolated values to estimate the mean annual precipitation at each site is the lack of comprehensive measurements for validation (Miller et al., 2005). Despite this drawback, a general association can be observed between forest floor Hg amount and mean annual precipitation, suggesting sites
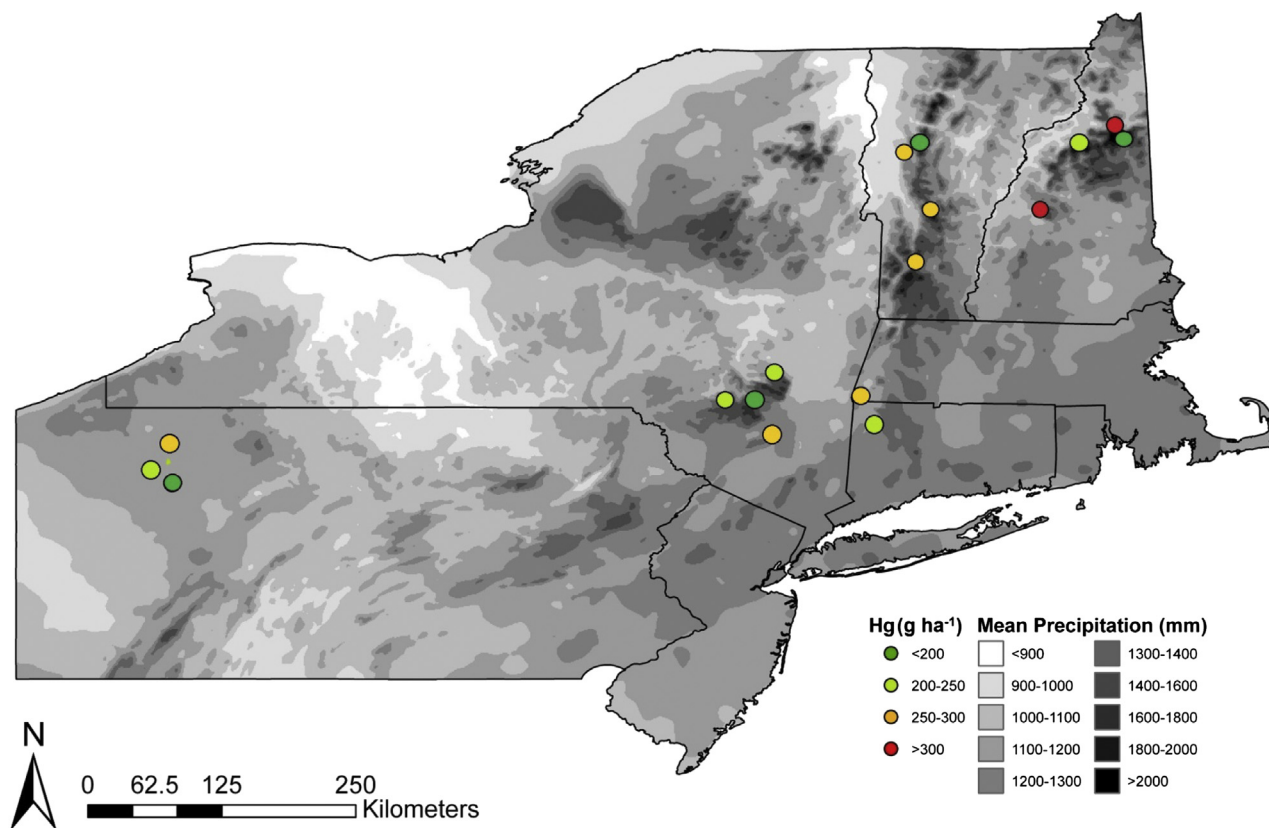
Fig. 3. Forest floor Hg amount at 17 sites (in circles) displayed with mean annual precipitation (1981–2011) map graphics from the PRISM Climate Group. Copyright © 2011, PRISM Climate Group, Oregon State University, http://prism.oregonstate.edu, Map created 10 21 2012.

that receive greater precipitation have a greater forest floor Hg mass.

Although only wet deposition was evaluated in this study, dry deposition may be an equally important but complex flux of Hg to forest soils (Miller et al., 2005; Demers et al., 2013). The dry deposition of Hg in forests of the northeastern U.S. has been estimated to be equal to or greater than Hg wet deposition (Miller et al., 2005). Rea et al. (2002) estimated dry deposition of Hg was nearly 33% of the total Hg entering the Lake Champlain watershed in northern Vermont. Dry deposition of Hg at our study sites was not estimated because of its complex variance with parameters at the landscape scale to leaf stomata scale (Ericksen et al., 2003; Miller et al., 2005; Smith-Downey et al., 2010). Furthermore, dry deposited Hg is readily washed off during precipitation making it difficult to quantify and generally it is integrated into throughfall (Rea et al., 2002; Miller et al., 2005). Dry deposition may explain variation in Hg concentration and amount in the forest floor and should be considered in future studies.

Winter fluxes of Hg are a less-understood mechanism that may influence Hg accumulation in the forest floor. Mean annual precipitation includes all forms of precipitation. However, Hg in snow may not permeate into the forest floor and mineral soil during winter months as effectively as rainfall (Schwesig and Matzner, 2001). In addition, Hg has been observed to transfer from the soil into overlying snow (Susong et al., 2003). The precipitation type, snowpack and snowmelt across the northeastern U.S. are likely different in total amount and temporally. This may influence the amount of Hg reaching the forest floor and entering the mineral soil (Schwesig and Matzner, 2001; Demers et al., 2007).

Many investigators have observed a strong correlation between Hg with % SOM and soil pH in the forest floor (Schuster, 1991;

Gabriel and Williamson, 2004; Szopka et al., 2011; Obrist et al., 2011; Shi et al., 2013). Mercury accumulation is influenced by SOM and pH as they control sorption capacity of soil (Schuster, 1991; Gabriel and Williamson, 2004). Northern sites had significantly higher % SOM and lower pH compared with western sites ($P < 0.05$) (Table 2). The higher % SOM may have increased the sorption capacity for Hg (Gabriel and Williamson, 2004). Further, our results suggest lower pH promoted higher Hg concentration and amount, which does not agree with other studies (e.g. Gabriel and Williamson, 2004). The mechanism responsible for the negative correlation between forest floor Hg and pH is unclear but may be due to a soil properties not investigated in this study such as biological activity of fungi or annelid. Greater % SOM did not correlate with lower pH ($R^2 = 0.19$) and is presumed not to be the masking mechanism for greater pH correlating with greater Hg.

A stepwise regression was used to rank the relative importance of each regional factor (mean annual precipitation, latitude and longitude) and soil property (pH, SOM, and Fe concentration and amount) on forest floor Hg concentration and amount. Forest floor Hg concentration was significantly correlated with % SOM, pH, and latitude while forest floor Hg amount was correlated with % SOM, pH and precipitation (Table 2). These regressions correspond with the variables used in spatial Hg models by Smith-Downey et al. (2010) and Obrist et al. (2011). In addition, the stepwise regressions also suggest the importance of soil pH in the northeastern US, a variable not used in models by Obrist et al. (2011) or Smith-Downey et al. (2010).

A multiple regression of Hg concentration in the forest floor was calculated using % SOM, pH, and latitude, with an R^2 value of 0.77 ($P < 0.01$) (Table 3). Similarly, a multiple regression to describe forest floor Hg amount was determined using mean annual

precipitation, % SOM and pH with an R^2 value of 0.65 ($P < 0.01$) (Table 3). The dependence of forest floor Hg on latitude, precipitation, pH and % SOM demonstrate that regional and soil properties are both integral to Hg accumulation in forest soils. Regional factors (latitude and precipitation) likely control Hg deposition to the forest floor while soil properties (SOM and pH) may dictate the retention of Hg in the mineral soil.

3.2. Mercury in the mineral soil

The mean Hg concentrations in the upper and lower mineral horizons were 45 ± 6 and 71 ± 13 µg kg^{-1}, respectively. The Hg concentrations are similar to those reported by previous studies in the northeastern U.S. (McNeal and Rose, 1974; Rea et al., 2002; Obrist et al., 2011; Juillerat et al., 2012). Mercury concentration in the upper mineral horizon was similar across the study region (Fig. 2). In the lower mineral horizon, Hg concentration was significantly higher at the northern sites (Fig. 2). Mercury concentration in the upper and lower mineral horizons were significantly lower compared with the forest floor ($P < 0.01$) (Fig. 2). The mean Hg amount in the upper and lower mineral horizons were 23 ± 5 and 64 ± 12 g ha^{-1}, respectively. These values are similar with Evans et al. (2005) but are much lower than those reported by Demers et al. (2007), likely due to site-specific characteristics of the soil profiles (i.e. profile depth and rock content). Mercury amount in the lower mineral horizon at northern sites was significantly higher than all other soil horizons at all sub-regions (Fig. 2).

Many of the soil properties of the upper and lower mineral soil also varied significantly across the study region. Soil horizons were thinner at western sites compared to central and northern sites (Table 2). Soil pH was highest in the upper mineral soil of the western sites and lowest in the upper mineral soil of the northern sites (Table 2). % SOM in the upper mineral was similar across the study sites with an average of 12% and a median of 10%. However, the lower mineral horizons at western sites were significantly lower in % SOM than central and northern sites ($P < 0.05$) (Table 2). The clay content of the soil was generally low and ranged from 6 to 23% with a mean of 13 ± 1.4%. Iron concentrations in the upper mineral soil were similar across sites with a mean of 13 ± 3 mg kg^{-1}. In the lower mineral horizons, northern sites contained greater Fe concentrations than western and central sites ($P < 0.01$) (Table 2). Similarly, Fe amounts in the upper mineral horizons were comparable, with a mean of 7 ± 2 but, northern sites were significantly greater than western and central sites ($P < 0.05$) (Table 2).

The accumulation of SOM and Fe in the lower mineral horizon at northern sites is indicative of Spodosols and their presence may explain the higher Hg concentration and amount at northern sites. Central and northern sites were grouped by soil order, to determine if Hg concentration and amount in the soil profile varied substantially between the two dominant soil orders: Inceptisols and Spodosols (Fig. 4). For the Inceptisols, Hg concentration followed the SOM distribution and decreased with depth (Fig. 4). Mercury amount however, was nearly uniform through the profile of Inceptisols. Mercury concentration and amount in the Bs horizons of the Spodosols were significantly higher compared to all other mineral horizons ($P < 0.01$, Fig. 4). Maximum accumulation of Hg in the Bs horizon is consistent with other studies of Hg in Spodosols in
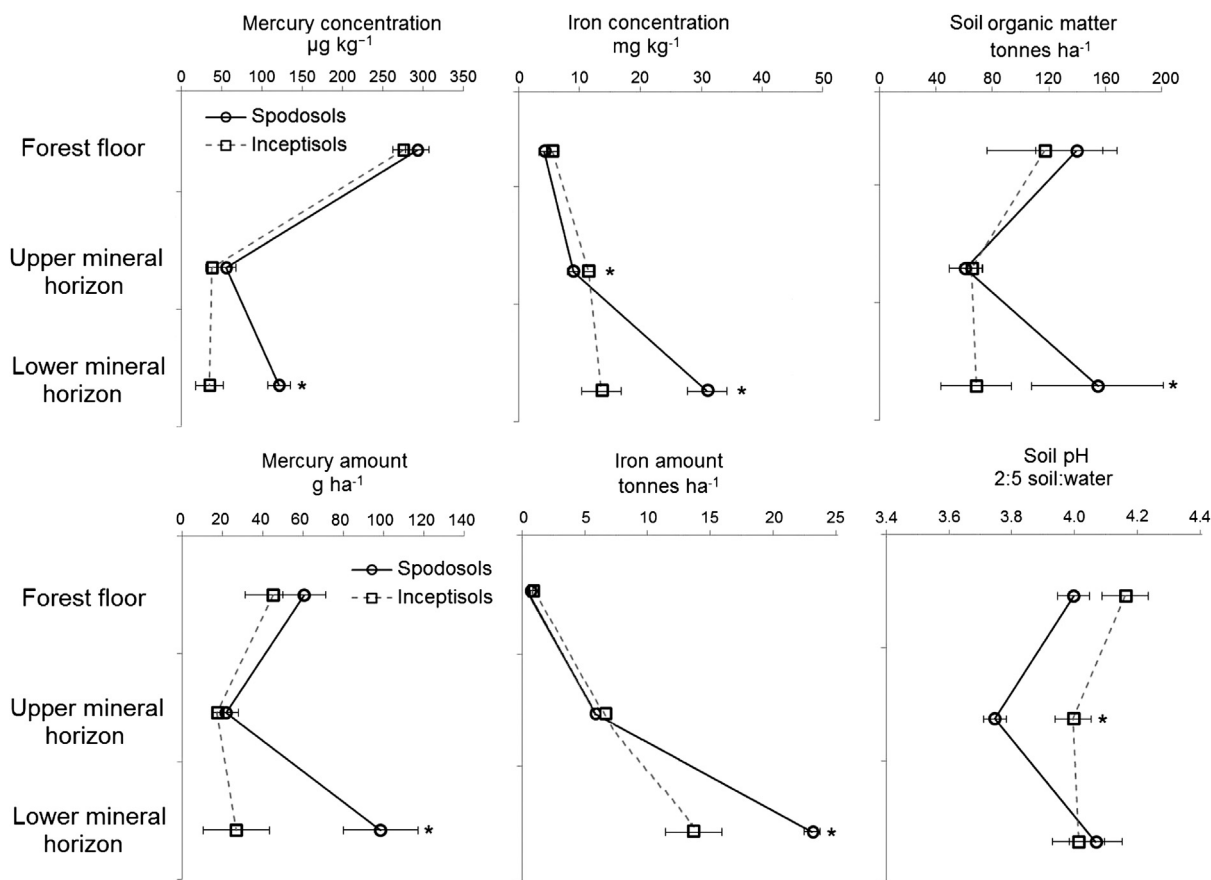

Fig. 4. Mercury and soil properties displayed by depth for three horizons for Spodosols and Inceptisols at central and northern sites. (*) designates a significant difference ($P < 0.05$) using the Wilcoxon rank signed test.

other regions (e.g. Ma et al., 1997; do Valle et al., 2005; Grimaldi et al., 2008) and explains the significantly higher Hg in the lower mineral horizons at the northern sites. The importance of spodic horizons on lead accumulation in the mineral soil has been observed in previous studies in the northeastern U.S. (e.g. Johnson and Petras, 1998; Kaste et al., 2011) and in warmer climates (e.g. Ma et al., 1997). This is a finding that has not been reported for Hg in other regional studies in the northeastern U.S.

Podzolization, the process of SOM complexing and eluviating Fe and other metals from the E horizon to the Bs horizon (Lundstöm et al., 2000), is likely responsible for the elevated Hg accumulation in the Bs horizon (do Valle et al., 2005; Kaste et al., 2011; Bushey et al., 2013). The accumulation of SOM and Fe oxides in the Bs horizon promote greater Hg accumulation due to increased surface binding sites (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). The mean SOM amount in the Bs horizons was significantly greater compared to the all other mineral soil horizons ($P < 0.05$) (Fig. 4). Similarly, the mean extractable Fe concentration in the Bs horizons (31 ± 2 kg ha^{-1}) was higher compared to the E horizons (9 ± 3 kg ha^{-1}) ($P < 0.05$) (Fig. 4). The Bs horizons have greater Fe oxides than the other soil horizons, assuming that the extracted Fe concentration is roughly representative of Fe oxide abundance.

Regional factors (mean annual precipitation, latitude and longitude) and soil properties (pH, % SOM, Fe content, and clay content) were stepwise regressed with Hg concentration and amount in the mineral soil. Mercury concentration and amount in the upper mineral horizon were only correlated with Fe concentration and amount, respectively. The correlation between Fe concentration and amount in the upper mineral horizons with Hg are arbitrary because they only explained 35% and 21% of the variation in Hg in the upper mineral horizon, respectively (Table 3). For the lower mineral horizon, Hg was correlated with latitude, % SOM, and Fe. Soil pH and % SOM may not have been correlated with mineral soil Hg concentration and amount due to the small range in values. Iron was correlated with Hg concentration and amount in the soil for two possible reasons: 1) factors that promote the sorption of Fe and Hg in the lower mineral horizons are similar (e.g. similar affinity for SOM); or 2) Fe, present in the lower mineral horizon as an oxide or hydroxide, may promote Hg sorption, with the assumption that Fe concentration is roughly representative of Fe oxide abundance. The % clay may not have been correlated with Hg in the soil due to the mineralogy of the clay-sized particles. Soils of the northeastern US, especially at northern sites, generally lack clay minerals known to strongly sorb metals (Coen and Arnold, 1972; April and Newton, 1983). Mean annual precipitation was not correlated with Hg concentration or amount in the upper and lower mineral soil horizons, suggesting that deposition processes may be less important to the spatial distribution of Hg in the mineral soil. This finding agrees with the conclusion by Obrist et al. (2011) that Hg distribution in the mineral soil does not coincide with variables associated with Hg deposition.

The significantly correlated variables from the stepwise regressions were used in multiple regressions to estimate Hg concentration and amount in the mineral soil. As described earlier, Hg concentration and amount in the upper mineral horizon was inadequately explained by Fe concentration and amount, respectively (Table 3). For the lower mineral horizons, Hg concentration and amount were well explained using latitude, % SOM and Fe concentration and amount with R^2 values of 0.80 and 0.73, respectively (Table 3). The significant correlation of SOM, Fe, and latitude in the lower mineral horizon is suggestive of the importance of Spodosols, which only occurred in northern sites and had greater % SOM and Fe concentrations in the lower mineral horizons.

The strong correlation among Hg concentration, Fe concentration, and % SOM in the Bs horizon, coupled with the greater accumulation of Hg in the Bs horizon compared with Bw horizons in Inceptisols, suggests that podzolization promoted Hg accumulation. We hypothesize that podzolization has transferred Hg bound to SOM in the forest floor through the E horizon and into the Bs horizon. Based upon the strong contrast in the vertical distribution of Hg, soil orders were more indicative of the spatial pattern of Hg in the mineral soil than a multiple regression of soil properties (e.g. % SOM). This is a finding that has not been elaborated on by previous studies modeling Hg in soils (e.g. Obrist et al., 2011). Future studies on Hg in forest soils should consider incorporating soil orders into spatial estimates of Hg as a contrast to regressions with only regional factors and soil properties.

4. Conclusions

The findings of this study indicated that Hg accumulation in the forest floor varied with regional factors: soil temperature and precipitation; and physicochemical properties: pH, SOM, and depth. Soil temperature influenced Hg accumulation in the forest floor through multiple possible mechanisms: forest floor turnover rate, Hg volatilization, or winter Hg flux. The variables that correlated with Hg in other studies were also found to be important for multiple regressions in this region, with the addition of soil pH. The variation in Hg distribution in the mineral soil horizons was best explained by comparing the two dominant soil orders rather than by multiple regressions of regional factors and soil properties. The Spodosols had greater Hg in their Bs horizons compared to all other mineral horizons, as they occurred at higher latitudes with significant accumulations of SOM and Fe oxides. This result suggests the importance of soil order on Hg accumulation in the mineral soil and may provide an alternative approach to spatial modeling of Hg in forest soils. With this assessment of current Hg levels in forest soils of the northeastern U.S., the effect of future changes in Hg deposition or cycling at these sites may be monitored.

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