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Factors Affecting Soil Total Mercury in Seasonal Pond Basins within a Northern Hardwood Forest in Minnesota, USA

Mikayla J. Boche
Thomas M. DeSutter*
North Dakota State Univ.
Dep. of Soil Science
Fargo, ND 58105

Randall K. Kolka
USDA Forest Service
Northern Research Station
Grand Rapids, MN 55744

Forest canopies are sites for mercury (Hg) deposition, and forests can act as Hg sources to downstream aquatic environments. This study examined soil total Hg (THg), carbon (C), and nitrogen (N) to 15 cm in 10 seasonal pond basins in a northern hardwood forest in Minnesota. Pools (mass per area) and concentrations (mass per soil mass) of THg to 15 cm were lower in uplands than in ponds, indicating downslope transport. In uplands, THg concentrations were the same at 0- to 2-cm and 2- to 5-cm depths and then decreased, whereas THg density (mass per volume) peaked at 2 to 5 cm, highlighting the importance of bulk density on mass. Carbon and N trends were similar to THg. Apart from pond centers, strong positive relationships between THg and C were observed. Upland slope length, graminoid cover, basin area, and tree height accounted for over half of THg variance at pond edge. Understanding the distribution, trends, and contributing factors of soil THg can further efforts toward immobilization and sequestration, thus minimizing the potential for bioaccumulation.

Abbreviations: SOM, soil organic matter; THg, total mercury.

Core Ideas

- **THg uniform among basins in uplands with pond centers variable by substrate.**
- **THg greatest at pond center and uniform among upland landscape positions.**
- **Positive THg relationships with C and N, except organic pond centers.**
- **Upland THg concentration peaked at 0 to 5 cm, with mass peaking at 2 to 5 cm.**
- **Fifty-six percent of THg pool was explained by slope length, graminoid cover, basin area, and tree height.**

Forest ecosystems are important recipients of dry deposition because of their large canopy surface areas that can interact with the atmosphere (Johnson et al., 2007; Miller et al., 2005; Munthe et al., 1995; St. Louis et al., 2001). Dry deposition occurs via adhesion to surfaces or absorption into leaf tissue through stomata (Laacouri et al., 2013). Coniferous stands are more efficient at scavenging mercury (Hg) from the atmosphere than deciduous stands due to a higher leaf surface area, allowing for more throughfall inputs than deciduous stands (Witt et al., 2009). Mercury adsorbed to leaf surfaces has the potential to be washed off by precipitation and to become a throughfall input to soils below (Iverfeldt, 1991; Kolka et al., 1999; Rea et al., 2000, 2001; Schroeder and Munthe, 1998). Evasion of Hg from vegetation may occur, whereby elemental Hg volatilizes back into the atmosphere (Schwesig and Krebs, 2003). Litterfall transfers Hg stored in and on leaf tissues from the canopy to the forest floor (O horizon), which is generally the largest input to forested watersheds (Rea et al., 2000). Deciduous stands produce larger inputs of litterfall to the forest floor than coniferous stands (Ericksen et al., 2003; Demers et al., 2007), where such dry deposition often accounts for a majority of inputs, as opposed to wet deposition (Demers et al., 2007; Risch et al., 2011; Sheehan et al., 2006).

Once senesced, leaves become part of the forest floor and may contribute Hg to soils (Ericksen et al., 2003). Other inputs to soils include direct dry and wet deposition from the atmosphere (Grigal, 2003). Soils contain up to 90% of Hg in forested landscapes, with the remainder within vegetation (Ericksen et al., 2006; Schroeder and Munthe, 1998). Although inputs from above result in high Hg concentrations within the forest floor (Arfstrom et al., 2000; Friedli et al., 2007; Matilainen et al., 2001), four to five times the mass of Hg occurs within underlying mineral soils

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*Corresponding author (thomas.desutter@ndsu.edu).

(Grigal, 2003). For example, concentrations and pools of total Hg (THg) in Minnesota forest floors (O horizon) range from 30 to 448 ng g^{-1} and from 0.25 to 1 mg m^{-2}, respectively, with underlying mineral soil concentrations and pools ranging from 10 to 350 ng g^{-1} and from 2.9 to 5.5 mg m^{-2}, respectively (Grigal et al., 1994, 2000; Kolka et al., 2014; Mitchell et al., 2012; Nater and Grigal, 1992; Wiener et al., 2006; Woodruff and Cannon, 2010).

Upland soils are often sinks for Hg (Grigal et al., 2000; Kolka et al., 2001), largely due to the affiliation between Hg and organic matter (Grigal et al., 1994; Hurley et al., 1995; Yin et al., 1997) because Hg can form stable bonds with sulfur (S), carbon (C), and nitrogen (N) (Meili, 1991). Immobilization of Hg in forest soils has been attributed to significant binding to the most decomposed fraction of organic matter (Schwesig et al., 1999) and the dominant form in the mineral layer (Gladkova and Malinina, 1999; Grigal et al., 1994). Correlations between soil Hg with both C and N have been reported, with some studies finding a stronger relationship between Hg and N compared with Hg and C (Gunda and Scanlon, 2013; Obrist et al., 2009). Obrist et al. (2009) attribute the stronger correlation of Hg and N to the binding of Hg with ligand-containing N groups. The correlation of the accumulation of both Hg and N in the forest floor has also been attributed to microbial immobilization that results in an increase of reduced sulfur (S) groups, to which Hg binds (Demers et al., 2007). Similarly, correlation of Hg with total C has also been linked to reduced S groups in organic matter (Selvendiran et al., 2008b). In addition to C, N, and S, the accumulation of Hg has been linked to other soil characteristics, such as texture, cation exchange capacity, and pH (Navrátil et al., 2014; Obrist et al., 2009; Schwesig et al., 1999).

Mercury that is not sequestered in the soil can be transported elsewhere. For example, some Hg in the soil is volatilized back into the atmosphere (Grigal, 2002). Volatilization can depend on sunlight (Gustin et al., 2002), soil pH, and content of clay and soil organic matter (SOM), factors that affect binding sites and how tightly Hg is bound to the soil (Schlüter, 2000). A minor amount of Hg in soil is taken up by plant roots and is either incorporated into plant tissues or released through transpiration (Bushey et al., 2008). Mercury can also be lost to the atmosphere during fire events (Amirbahman et al., 2004; Kolka et al., 2014). Finally, Hg can be transported from upland soil to locations downslope via runoff, likely subsurface runoff in forest soils (Bushey et al., 2008).

Inorganic Hg that reaches wetland sediments is susceptible to becoming transformed into more toxic organic methylmercury (MeHg) by sulfate-reducing bacteria that thrive in anoxic conditions (Reddy and DeLaune, 2008). Methylmercury readily bioaccumulates and biomagnifies through trophic levels, potentially resulting in concentrations dangerous to organisms (Scheuhammer et al., 2007). Higher concentrations of MeHg in seasonal pond basins have been tied to longer hydroperiods (Brooks et al., 2012) and factors that affect sulfate reduction rates of bacteria such as temperature, organic matter content, and sulfate supply (Reddy and DeLaune, 2008). Wetlands can act as sources of MeHg downstream (Galloway and Branfireun, 2004; Selvendiran et al., 2008a), whereby higher concentrations of wetlands in catchments have been linked to higher MeHg concentrations in fish in lakes (Driscoll et al., 1995) and streams (Chasar et al., 2009).

Forested landscapes scavenge Hg from the atmosphere, and Hg deposited to soils in these systems has the potential to accumulate in wetlands, where it may become biologically available. Understanding the distribution of soil Hg in the landscape, and how and why that distribution changes with landscape position, is essential for efforts aiming to sequester Hg and prevent it from entering aquatic environments. The objectives of this study were (i) to characterize the distribution of THg in the soil across landscape position and profile depth, (ii) to examine upland characteristics that contribute to the downslope accumulation of THg, and (iii) to investigate the relationship of soil THg with C and N in seasonal pond (wetland) basins within a northern hardwood forest.

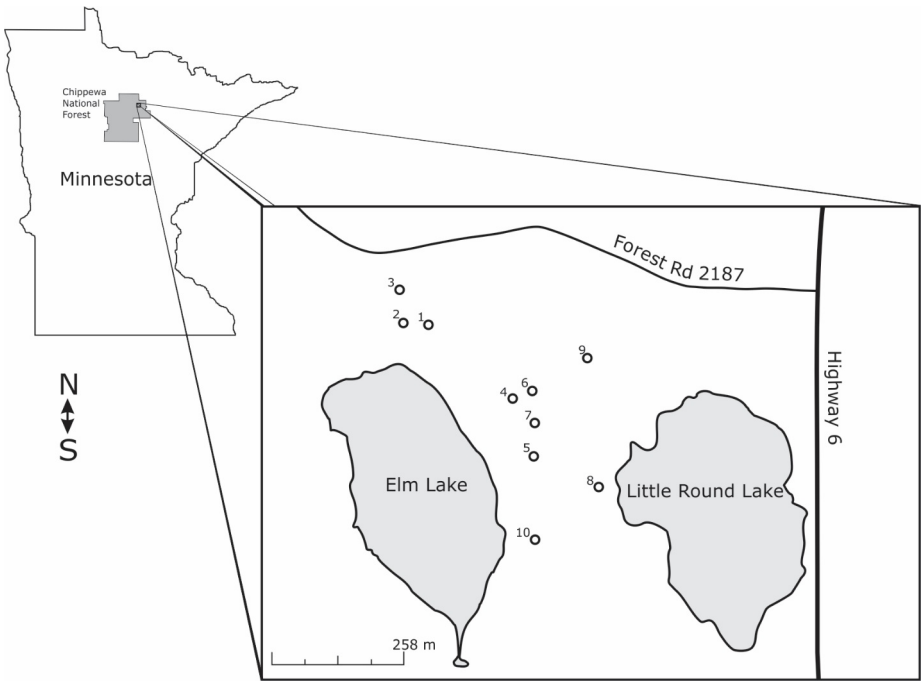

Fig. 1. Sampling locations within the Chippewa National Forest, Minnesota.

MATERIALS AND METHODS

Site Characterization

The 20-ha study area is located about 60 km northwest of Grand Rapids, MN, in the Chippewa National Forest (47.692275 N, −93.780926 W) (Fig. 1). This area is situated within the Northern Minnesota Drift and Lakes Plain Section, where the loamy till parent material was deposited throughout multiple glaciation events that left

behind outwash plains and end, stagnation, and ground moraines (Hanson and Hargrave, 1996). Upland soils within the study area are mapped as Warba (fine-loamy mixed superactive frigid Haplic Glossudalfs) (Soil Survey Staff, 2016a, 2016b) in a landscape with 0 to 33% slopes and many depressional vernal and permanent wetlands that were not separately mapped. The mean annual precipitation is 73.1 cm, and mean annual temperature for the region is 4.2°C (Midwestern Regional Climate Center, 2012). The area is classified as "birch" and "hard maple/basswood" forest dating from 1944 and 1929, respectively, with no fire or harvest treatments on record (A. Gustafson, District Silviculturist, Chippewa National Forest, personal communication, 2011). The study area was dominated by quaking and bigtooth aspen (*Populus tremuloides* Michx. and *Populus grandidentata* Michx., respectively), basswood (*Tilia Americana* L.), sugar maple (*Acer saccharum* Marshall), and paper birch (*Betula papyrifera* Marshall) in the uplands, with paper birch and black ash (*Fraxinus nigra* Marshall) dominating in the seasonal ponds.

Twenty-six ponds were identified in the study area during scoping visits in spring 2011. Of these, 10 seasonal pond basins were identified that appeared to have no surface water connection to other ponds, although groundwater connections may exist. All 10 ponds exhibited hydric soil and hydrophytic vegetation. Ponds had either mineral or organic (peat) substrates to 15 cm, as determined by visual observation in the field and laboratory analysis of bulk density and C content. Six ponds were identified as mineral (Ponds 1, 2, 7, 8, 9, and 10) and four as organic (Ponds 3, 4, 5, and 6) (Table 1). Pond 1 was somewhat intermediary between organic and mineral and was treated as mineral. The 10 basins ranged in size from 730 to 6300 m^2, with ponds ranging from 24 to 1200 m^2. Mean upland slopes per basin ranged from 10.9 to 21.3%, and mean slope lengths ranged from 6.54 to 28.2 m.

Table 1. General basin (including pond) characteristics.

Basin	Pond center		Basin area	Pond area	Mean upland slope	Mean upland slope length	Pond substrate
	Latitude	Longitude					
	decimal degree		m^2		%	m	
1	47.692797	–93.783851	2490	238	11.9	13.5	mineral
2	47.692841	–93.784476	895	47.3	10.9	9.18	mineral
3	47.693429	–93.784593	6300	951	13.4	8.38	organic
4	47.691508	–93.781641	5220	155	21.3	14.9	organic
5	47.690498	–93.781101	4380	1220	17.7	12.5	organic
6	47.691661	–93.781141	1230	218	17.7	9.66	organic
7	47.691106	–93.781081	729	23.5	11.9	6.54	mineral
8	47.689951	–93.779432	1660	45.3	17.4	17.1	mineral
9	47.692223	–93.779711	3080	223	14.1	28.2	mineral
10	47.689029	–93.781082	4970	144	16.3	23.5	mineral

Experimental Design

Within each of the 10 selected basins, sampling occurred at five landscape positions along five transects (Fig. 2). Transects were laid out from pond center to the summit of the basin enclosing the pond. A randomly generated compass bearing was determined (by 10°), and transects were spaced ~70° apart using a compass. If a transect fell in an area where the slope was <8.75% grade, the transect bearing was moved 10 degrees in the direction closest to a slope of at least 8.75% grade so that definite basin landscape positions could be identified and characterized. Landscape positions were defined as: 1 m from pond center ("pond center"), 1 m inside the high water line ("pond edge"), toe slope, midpoint of back slope ("back slope"), and at the slope shoulder ("shoulder"). The high water line was deduced by examining the extent of hydrophytic vegetation, visual observations of inundation, watermark stains on trees, drift lines, sediment deposits, and water stained leaves (Environmental Laboratory, 1987).

Pond and basin area calculations were obtained using a handheld GPS unit. The upland slope angle of each transect from pond edge to hill summit was recorded using a clinometer. Each sample location was marked on the GPS with a waypoint to obtain the elevation and coordinates. Sample locations were marked with flagging tape for identification during follow-up visits. The distance from pond edge to each sampling location was measured using a rangefinder.

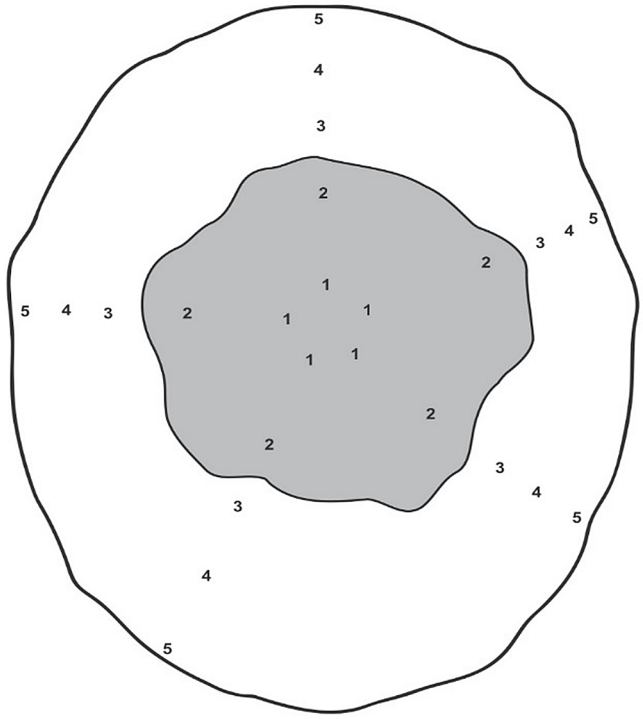

Fig. 2. Sampling layout for each basin. The shaded area represents a seasonal pond, and the white area represents the upland area of a catchment basin. Five transects were spaced approximately 70 compass degrees apart beginning at the center of the pond. Samples were obtained at five landscape positions: 1 = 1 m from pond center, 2 = 1 m inside pond edge, 3 = toe slope, 4 = back slope, and 5 = shoulder.

Soil Sampling

Soil samples were taken during June and July 2011 using a 3.12-cm-diameter stainless steel T-handled core sampler (PN012; JMC, Newton, IA). Prior to sampling, large debris (e.g., twigs and leaves) that impeded sampling was cleared from the sampling location, leaving the majority of the forest floor intact. To obtain a representative sample, a composite sampling technique was used, whereby five 0- to 5-cm and three 5- to 15-cm cores were collected from each sample location. The cores were cut with a stainless steel knife into four increments: 0 to 2, 2 to 5, 5 to 10, and 10 to 15 cm. Like-depth increment sections were aggregated into polyethylene bags. Equipment was wiped clean between cores and rinsed with 4:1 methanol/water solution between sample locations. Samples were kept on ice in coolers until they could be refrigerated at 3°C. After weighing and hand homogenization, samples were stored frozen at −15°C.

A subsample of thawed soil was weighed and oven-dried at 105°C for a minimum of 24 h to determine gravimetric water content. A second subsample was air dried for a minimum of 96 h on cellophane for use in THg, C, and N analysis. The air-dried samples were ground with a porcelain mortar and pestle, passed through a 1-mm stainless steel sieve, and stored in polyethylene bags. Grinding and sieving equipment was rinsed with deionized water before and after use with a 5% nitric acid solution and then rinsed with 4:1 methanol/water solution before being allowed to air dry between samples. A subsample of air-dried soil was oven-dried at 105°C for a minimum of 24 h to determine gravimetric water content for moisture corrections.

Total Hg analysis was done using a direct mercury analyzer (DMA-80; Milestone Inc., Shelton, CT) following USEPA Method 7473 (USEPA, 2009). Total C and N elemental analyses were done by high-temperature combustion using a LECO TruSpec CHN Analyzer (200–290; LECO Corporation, St. Joseph, MI). A mineral soil calibration was initially used for all samples during C and N analysis. Samples with C concentrations >12% were re-analyzed using an organic material calibration.

Vegetation Assessment

Vegetative characteristics, including canopy closure, basal area of trees, predominant species, and ground cover, were recorded at each soil sampling location during peak coverage between 31 July and 21 Aug. 2011 (Appendix Table A1). Canopy closure was determined by averaging concave spherical densiometer readings taken in each of the four cardinal directions. A 10-factor wedge prism was used to quantify basal area of trees >2.5 cm in diameter at 1.5 m above the ground surface. The species, diameter, and height class estimate of each of these trees was recorded. A 1-m^2 quadrat was placed on a randomly determined side (along the same contour) of each soil sampling location at a distance of 1.5 m to avoid the area disturbed by sampling. For vegetation rooted in or overhanging the quadrat, the five most prevalent species were recorded along with their Daubenmire cover class (<5, 5–25, 25–50, 50–75, 75–95, 95–100%) (Daubenmire, 1959) and primary height class estimate (<0.5, >0.5–2, >2–5, >5–15, >15–30, >30 m) (USEPA, 2011). Ground cover type within the quadrats was also recorded using Daubenmire classes for the following categories: bare ground, water presence (with depth, if any), litter/debris, nonvascular vegetation (bryopsida and sphagnum mosses), graminoids, forbs, shrubs, and trees (modified from USEPA, 2011).

Calculations and Statistical Analyses

Soil bulk density was calculated for each depth increment and the weighted averages of bulk densities per increment were summed to calculate the bulk density for the entire 15 cm sampled. The mean bulk density (0–15 cm) per landscape position across basins ranged from 0.11 to 1.55 g cm^{-3} (mean, 1.03 g cm^{-3}). Due to the wide range of bulk densities observed across landscape positions and depth increments, and because organic matter and bulk density are inversely related, one may observe high concentrations of elements such as THg, C, and N, yet calculate small pools, and vice versa (Grigal et al., 1994; Juillerat et al., 2012; Navrátil et al., 2014). Therefore, THg, C, and N pools to 15 cm (mass per area) or densities for each depth increment (mass per volume) are presented alongside concentrations (mass per soil mass) to standardize comparisons. To standardize comparisons among depth increments, THg, C, and N mass for each increment were calculated by multiplying the respective concentration by the corresponding bulk density.

Because data were collected per increment, the weighted sum of THg, C, and N concentrations per increment was calculated to obtain THg, C, and N pools to 15 cm using the equation:

$$S_{0\text{–}15\ \mathrm{cm}} = \sum_{i=1}^{n} \left(x \cdot \rho_b \cdot h \cdot f \right) \qquad [1]$$

where $S_{0–15}$ is the THg, C, or N pool to 15 cm (mg m^{-2} for THg; kg m^{-2} for C and N); n is the number of increments (4); i is the depth increment (0–2, 2–5, 5–10, and 10–15 cm); x is the concentration of THg, C, or N per depth increment (ng g^{-1} for THg; g kg^{-1} for C and N); ρ_b is the bulk density (g cm^{-3}) per increment; h is the height (cm) per increment (2, 3, 5 , and 5 cm); and f is the unit conversion factor for THg, C, and N (0.01 for TH; 0.1 for C and N) (modified from Mishra et al., 2009). Similarly, a weighted sum was used to combine THg, C, and N concentration data per increment to calculate THg, C, and N concentrations (ng g^{-1} for THg; % for C and N) to 15 cm, whereby the THg, C, or N concentration per depth increment (0–2, 2–5, 5–10, and 10–15 cm) was multiplied by the proportion of the increment's height out of the 15 cm sampled (2/15, 3/15, 5/15 and 5/15) and then summed. The C/N ratios were calculated for each increment and for the entire 0 to 15 cm by dividing C concentration by N concentration.

Statistical analyses were done using SAS software, Version 9.2 of the SAS System for Windows (SAS Institute Inc., Cary, NC). For all statistical analyses, an α level of 0.05 was considered as indicative of differences.

Simple linear regression was conducted to examine THg relationships with C and N with respect to pools (mg m^{-2} for THg; kg m^{-2} for C and N) and concentrations (ng g^{-1} for THg;

% for C and N) to 15 cm. Linear regression was repeated for all landscape positions together, pond center alone, and all landscape positions apart from pond center.

Generalized linear mixed models (GLIMMIX procedure) were used to generate Tukey groupings to compare least squares means of THg, C and N (pools, densities, and concentrations), and C/N ratio among basins, landscape positions, and depth increments, where the main effects were basin, landscape position, or depth increment. All models included Tukey–Kramer multiple comparison adjustments. To account for pseudoreplication (Hurlbert, 1984), concentrations, pools, and densities from like-landscape positions and like-depth increments (where applicable) within each basin were averaged where possible for comparison. The intent was to arrive at $n = 10$ when comparing basins with each other by landscape position (0–15 cm); $n = 5$ when comparing landscape positions to each other within an individual basin (0–15 cm); and $n = 10$ when making comparisons where all ponds were considered together among landscape positions (0–15 cm), depth increments at a given landscape position, and landscape positions for a given depth increment. However, to achieve sufficient degrees of freedom, all five replications (transects) in each basin were used when comparing basins with each other at each landscape position ($n = 50$) and when comparing landscape positions with each other within each basin ($n = 25$).

Distributions of data used in GLIMMIX models were assessed for normality using the Anderson–Darling goodness-of-fit test. Some datasets fit neither normal nor γ distributions, whereas others fit both. This lack of clear distribution is presumably due to the relatively small sample size. For consistency, and because there was little to no difference between results of each specified distribution, a normal distribution was specified for all GLIMMIX models.

Multiple regression was used to identify upland independent variables that predict the THg pool (0–15 cm) at the pond edge landscape position. Pond edge was selected for prediction because it can be assumed to be affected by processes from the adjacent upland. Where applicable, data from the three upland landscape positions (toe slope, back slope, and shoulder) along each transect were averaged together to produce one value for use in analysis. The predictors were assessed for collinearity so that variables contributing collinearity problems were removed.

Stepwise regression (significance levels to stay and enter = 0.25) was used to interpret factors contributing to the THg pool at the pond edge landscape position using the following upland variables: upland slope length (m), upland slope grade (%), basin area (m^2), upland means of C/N ratio (0–15 cm), THg pool (0–15 cm; mg m^{-2}), mean tree diameter (cm), mean tree basal area (m^2 ha^{-1}), mean tree height class midpoint (m), canopy openness (%), tree cover class midpoint (%), shrub cover class midpoint (%), forb cover class midpoint (%), graminoid cover class midpoint (%), nonvascular cover class midpoint (%), litter and debris cover class midpoint (%), and bare ground cover class midpoint (%). Residuals were examined for normality, randomness, and leverage. Iterations of the models were performed to obtain the greatest R^2 and acceptable residual plots. Three data points with large residuals were discarded ($n = 47$).

RESULTS AND DISCUSSION

Basin and Landscape Position Comparisons of Total Hg, C, N, and C/N Ratio

Mean soil THg concentrations (0–15 cm) per landscape position across individual basins ranged from 20.1 to 235 ng g^{-1} (mean, 54.0 ng g^{-1}), whereas THg pools ranged from 2.41 to 15.1 mg m^{-2} (mean, 5.31 mg m^{-2}) (Table 2). Total Hg concentrations and pools were generally within range of other Minnesota forest soils (Grigal et al., 1994, 2000; Kolka et al., 2014; Mitchell et al., 2012; Nater and Grigal, 1992; Wiener et al., 2006; Woodruff and Cannon, 2010), although there were some THg pools in the center of the ponds that were higher than reported previously.

In general, THg, C, and N concentrations and pools, and C/N ratios were not significantly different among basins for any of the upland landscape positions (i.e., toe slope, back slope, shoulder) (Table 2). In contrast to uplands, these parameters exhibited significant differences among basins at the pond center and pond edge landscape positions. These variations were dependent on the substrate of the pond (mineral versus organic; Table 1). For example, the concentration of C at pond centers in organic ponds was up to 14 times higher than concentrations from mineral pond centers. Nitrogen concentrations among ponds followed the same pattern across pond centers as C concentrations.

Total Hg concentrations at pond centers roughly mirrored trends for C and N concentrations; however, there were fewer statistical differences among ponds, and Pond 1 was anomalous in that it was among the highest ponds in terms of THg concentration but was among the mid-low range for C and N concentrations (Table 2). For THg pools at pond center, Ponds 3 and 5 had the lowest pools, and Ponds 1 and 10 had the greatest pools compared with other ponds. It is evident that concentration cannot accurately predict mass present because of differences in bulk density and that these pond centers are quite variable in terms of THg, C, and N storage. As such, care should be taken not to use concentration as a surrogate for distribution of mass.

For each individual basin, THg concentration was greater at pond center than upland landscape positions; however, regarding THg pools, pond centers in Basins 3, 4, 5, and 6 were either less than or not significantly different from upland landscape positions (Table 2). For C concentration, pond centers in Basins 2, 7, 8, 9, and 10 were not statistically different from some or all upland landscape positions; these pond centers all had lower C concentrations than other ponds. Similarly, where N concentrations and pools at pond center in individual basins were not statistically different from uplands, those N concentrations and pools were among the lowest among basins at the pond center landscape position.

Differences in concentrations among basins at the pond center may be related to differences in substrate (organic versus mineral). For example, many of the ponds with low C, N, and THg concentrations (Table 2; Ponds 3, 4, 5, and 6) were mineral soils, whereas many of the ponds with higher concentrations had organic or high C soils. For most parameters in Table 2, there was little difference among upland landscape positions for individual basins, and, except for C/N ratio, pond centers were

Table 2. Comparison of least squares means for soil total Hg (THg), C and N pools and concentrations, and C/N ratio to 15 cm across basins and landscape positions. Capital letters compare basins within a landscape position. Lowercase letters compare landscape positions within a basin. For all comparisons, *n* = 5.

		Basin									
Parameter	LP†	1	2	3	4	5	6	7	8	9	10
THg, mg m^{-2}	A	15.1Aa‡	6.77CDa	2.41Ec	5.56DEb	2.56Ec	6.66CDa	9.24BCa	7.75CDa	9.15BCDa	12.6ABa
	B	6.20ABCb	4.62ABCb	5.70ABCa	7.98Aa	4.04Cb	6.93ABCa	5.02ABCb	4.14BCb	7.60ABa	6.83ABCb
	C	3.87Ab	4.09Ab	4.51Aab	4.16Ab	5.66Aa	4.66Aa	4.79Ab	4.04Ab	4.71Ab	4.96Abc
	D	3.84Ab	3.85Ab	3.45Abc	4.89Ab	4.90Aab	3.96Aa	4.79Ab	3.36Ab	4.44Ab	4.00Ac
	E	4.17Ab	3.90Ab	3.67Abc	4.45Ab	4.40Ab	4.10Aa	4.74Ab	3.54Ab	4.03Ab	4.53Abc
C, kg m^{-2}	A	9.48Ba	4.40Da	9.58Ba	12.3Aa	6.80Ca	12.6Aa	5.14CDa	5.09CDa	4.00Db	4.81Da
	B	6.45ABb	4.18BCa	7.19ABb	9.32Ab	4.71BCb	6.14BCb	3.33Cb	4.92BCab	6.59ABa	5.81BCa
	C	4.08Ac	4.03Aa	5.20Ac	4.44Ac	5.20Ab	4.89Ab	4.39Aab	4.24Aab	4.82Ab	4.82Aa
	D	3.73Ac	4.05Aa	3.52Ac	4.25Ac	4.44Ab	3.28Ab	4.02Aab	3.77Aab	4.34Ab	4.20Aa
	E	4.31Ac	4.10Aa	4.07Ac	4.33Ac	4.42Ab	3.56Ab	4.66Aa	3.71Ab	4.45Ab	4.30Aa
N, kg m^{-2}	A	0.502BCa	0.323DEa	0.441CDa	0.626ABa	0.306Ea	0.650Aa	0.392CDEa	0.391CDEa	0.383CDEab	0.423CDEa
	B	0.397ABCb	0.271Cab	0.442ABCa	0.472Ab	0.290BCa	0.387ABCb	0.266Cb	0.329ABCab	0.465ABa	0.414ABCab
	C	0.231Ac	0.222Ab	0.277Ab	0.249Ac	0.305Aa	0.251Abc	0.270Ab	0.270Abc	0.293Abc	0.310Abc
	D	0.196Ac	0.215Ab	0.208Ab	0.252Ac	0.268Aa	0.215Ac	0.256Ab	0.226Ac	0.275Ac	0.259Ac
	E	0.238Ac	0.239Aab	0.225Ab	0.260Ac	0.277Aa	0.218Ac	0.279Ab	0.198Ac	0.283Abc	0.274Ac
THg, ng g^{-1}	A	235Aa	53.1Ea	118CDa	198ABa	158BCa	209ABa	73.3DEa	60.0Ea	66.7DEa	97.6DEa
	B	60.9Bb	30.0Bb	45.8Bb	132Ab	35.2Bb	77.7ABb	25.3Bb	31.9Bb	80.6ABa	74.9ABa
	C	25.3Ab	25.3Ab	28.6Ab	27.7Ac	38.9Ab	38.0Ab	33.0Ab	25.8Ab	35.2Ab	32.6Ab
	D	26.2Ab	25.4Ab	20.4Ab	30.4Ac	32.0Ab	23.3Ab	30.7Ab	20.1Ab	28.8Ab	27.4Ab
	E	27.0Ab	24.4Ab	21.7Ab	27.9Ac	28.4Ab	24.4Ab	31.6Ab	21.4Ab	24.5Ab	30.3Ab
C, %	A	13.4Ca	3.88Da	39.0Aa	34.4Ba	30.1Ba	32.6Ba	4.22Da	4.20Da	2.80Db	3.62Db
	B	6.24Bb	3.05Ba	5.67Bb	15.7Ab	4.15Bb	6.35Bb	1.76Bc	4.11Bab	7.43Ba	6.58Ba
	C	2.75Ac	2.51Aa	3.31Ac	3.24Ac	3.65Ab	4.45Ab	3.08Aab	2.73Aab	3.72Ab	3.19Ab
	D	2.56ABc	2.71ABa	2.10ABc	2.68ABc	2.96Ab	1.98Bb	2.64ABbc	2.26ABb	2.82ABb	2.86ABb
	E	2.81Ac	2.53Aa	2.40Ac	2.76Ac	2.93Ab	2.13Ab	3.18Aab	2.29Ab	2.68Ab	2.93Ab
N, %	A	0.724Ca	0.267Da	1.79Aa	1.77Aa	1.36Ba	1.70Aa	0.304Da	0.311Da	0.260Db	0.306Dab
	B	0.374BCb	0.183BCab	0.344BCb	0.806Ab	0.244BCb	0.388BCb	0.132Cb	0.264BCab	0.498ABa	0.435BCa
	C	0.152Ac	0.137Ab	0.175Ac	0.169Ac	0.208Ab	0.203Abc	0.183Ab	0.173Abc	0.217Ab	0.201Ab
	D	0.132Ac	0.142Ab	0.121Ac	0.153Ac	0.174Ab	0.126Ac	0.161Ab	0.134Ac	0.175Ab	0.171Ab
	E	0.155Ac	0.146Ab	0.132Ac	0.158Ac	0.179Ab	0.129Ac	0.182Ab	0.121Ac	0.167Ab	0.178Ab
C/N	A	18.9Ca	13.6Dc	21.7ABa	19.8ABCa	22.3Aa	19.3BCa	13.2Dbc	13.0DEb	10.5Eb	11.4DEc
	B	16.3Ba	15.4BCbc	16.3Bb	19.7Aa	16.5Bb	15.9Ba	12.6Cc	14.9BCb	14.2BCa	13.9BCb
	C	17.7Aa	18.2Aa	19.0Aab	17.5Aa	17.1Ab	19.4Aa	16.2Aa	16.0Aab	16.6Aa	15.5Aab
	D	19.2Aa	19.0Aa	17.4Ab	16.9Aa	16.8Ab	15.3Aa	15.7Aab	16.8Aab	16.0Aa	16.3Aa
	E	19.3Aa	17.3Aab	18.2Ab	16.7Aa	16.2Ab	16.4Aa	16.7Aa	19.3Aa	15.7Aa	15.7Aab

† Landscape position, where A = pond center, B = pond edge, C = toe slope, D = back slope, and E = shoulder.

‡ Least squares means with the same capital letter within landscape positions are not significantly different at $\alpha = 0.05$. Least squares means with the same lowercase letter within basins are not significantly different at $\alpha = 0.05$.

generally greater than uplands, indicating that THg, C, and N are likely transported downslope from uplands and accumulating in ponds. Alternatively, aerobic versus anaerobic decomposition rates may vary, or there could be differential deposition in the form of throughfall and litterfall at the pond center when compared with the uplands. However, forest cover among basins was similar (Appendix Table A1), which is important in terms of homogeneity of THg deposition (Bushey et al., 2008; Juillerat et al., 2012; Perry et al., 2006) and C pools in the soil (Weishampel et al., 2009). Canopy structure and composition affects soil THg in terms of deposition via interaction of the canopy with the atmosphere and litterfall inputs to the soil (Bushey et al., 2008; Juillerat et al., 2012; Perry et al., 2006; Witt et al., 2009). For C pools, Weishampel et al. (2009) suggest that the forest floor may be more affected by canopy type than underlying mineral soil, whereas Powers et al. (2011) found similar C pools between canopy type but differing pools in mineral soils depending on canopy species.

For all basins together, pools and concentrations of THg, C, and N (0–15 cm) were greater at the pond center landscape position than at any of the upland landscape positions, whereas C/N ratio did not vary by landscape position (Table 3). Within the upland soils, no differences were found in THg, C, or N pools and concentrations across the three landscape positions (toe slope, back slope, shoulder). Others have found maximum soil THg concentrations and pools (83 ng g^{-1} and 5.4 mg m^{-2}, respectively) at the lowest sampled landscape position along forested slopes (toe slope), with summit and backslope positions exhibiting the lowest concentrations and pools (~29 ng g^{-1} and 2.9 mg m^{-2}) (Grigal et al., 1994).

Gladkova and Malinina (1999) found 24.1% of variation in THg concentration along a forested riverine catchment slope to be explained by landscape position, with 75.3% attributed to litter subhorizonation, illustrating the importance of considering sampling depth, particularly by horizon and subhorizon, when exploring soil THg distribution.

Accumulation of THg has been linked to the translocation of Hg bound to dissolved organic C (Selvendiran et al., 2008a) and dissolved organic matter (Ravichandran, 2004). Because of the affiliation of Hg with SOM, pedological concepts relating to SOM, such as differential horizon accumulation and SOM accumulation downslope in a catena, can also be used to predict the distribution of Hg in the landscape (Grigal, 2003). Total Hg and C pools in a lake-plain wetland have been attributed to depth of soil, which varied by landscape position (Nave et al., 2017). Landscape position can also affect soil moisture, aeration, texture, chemical composition, plant association, and nutrient regime of microorganisms, which can influence SOM and related processes (Gladkova and Malinina, 1999).

Table 3. Comparison of least squares means for soil total Hg (THg), C and N pools and concentrations, and C/N ratio at 0–15 cm across landscape positions. Lowercase letters compare landscape positions. For all comparisons, n = 10.

Landscape position	THg	C	N	THg	C	N	C/N
	mg m^{-2}	kg m^{-2}		ng g^{-1}	%		
Pond center	7.78a†	7.42a	0.444a	127a	16.8a	0.879a	16.4a
Pond edge	5.91ab	5.86ab	0.373a	59.4b	6.11b	0.367b	15.6a
Toe slope	4.54b	4.61b	0.268b	31.1b	3.26b	0.182b	17.3a
Back slope	4.15b	3.96b	0.237b	26.5b	2.56b	0.149b	16.9a
Shoulder	4.15b	4.19b	0.249b	26.2b	2.66b	0.155b	17.1a

† Least squares means with the same letter within columns are not significantly different at $a = 0.05$.

Total Hg Relationships with C and N

A positive relationship was observed in 15 of 16 relationships considered between THg with C and N (concentrations and pools). In the remaining situation, a negative relationship was observed between THg and C concentration in organic ponds (Table 4). Regression fits were comparable in relationships between soil THg with C and N, although r^2 values were slightly higher in most cases for THg and N relationships than for THg and C relationships. Except for organic pond centers, the linear relationships for concentrations were stronger than for pool relationships, which indicates that concentrations of C and N are more closely related to THg concentration than C and N pools are related to THg pools. Where all landscape positions were considered together, the pond center landscape position plot appeared more variable than other landscape positions, which is consistent with the range of THg, C, and N concentrations observed among pond centers (Table 2). Removing pond center from consideration and the variation associated with it improved the fit of the models (Table 4).

Considering all basins together at the pond center landscape position, linear relationships were weak and/or insignificant. By grouping data points by pond number, it was apparent that Ponds 3, 4, 5, and 6 were distinct from Ponds 1, 2, 7, 8, 9, and 10 in THg versus C and N plots (Table 4). Because these ponds where characterized by organic or mineral substrates, respectively (Table 1), ponds were grouped together accordingly for regression (Table 4). From the grouped pond center regression lines, it is evident that C and N at pond centers in organic ponds have a different relationship with THg than mineral ponds. For relationships between THg and C concentration, mineral ponds had a strong positive slope (16.9), whereas organic ponds exhibited a slope that was both smaller in magnitude and negative (–4.69). Regarding relationships between THg and N, slopes were positive for both organic and mineral ponds; however, the slope for organic ponds (5.93) was much lower than for mineral ponds (360).

Numerous studies note a correlation between THg and organic matter (de Klerk et al., 2013; Gabriel et al., 2012; Gruba et al., 2014; Juillerat et al., 2012). A few studies have observed different relationships between THg and C in high-organic-matter scenarios compared with soils with lower organic matter content. A curvilinear distribution of THg versus C concentrations has been reported for lake sediments, where a positive relationship was observed at low C concentrations and a negative relationship at high C concentrations (Kainz and Lucotte, 2006). The difference between the relationships was attributed to the type of organic matter, where sediments originating from catchments with little to no wetland surface area had low C concentrations and catchments containing a relatively

Table 4. Linear regression relationships between soil total Hg (THg) concentration to 15 cm (ng g^{-1}) and pool (mg m^{-2}) with corresponding units of C or N with consideration of landscape position and basin characteristics. Slopes and intercepts are significant at $\alpha = 0.05$, unless noted.

Relationship of THg with C or N	Concentration or pool	Landscape position†	Basins in model‡	Prediction equation, THg =	r^2
C	concentration	A, B, C, D, E	all	4.80(C) + 23.8	0.64
C	pool	A, B, C, D, E	all	0.46(C) + 2.93	0.16
C	concentration	B, C, D, E	all	8.14(C) + 6.08	0.85
C	pool	B, C, D, E	all	0.78(C) + 1.04	0.58
C	concentration	A	mineral	16.9(C) + 6.89§	0.91
C	pool	A	mineral	1.31(C) + 2.91	0.59
C	concentration	A	organic	-4.69(C) + 330	0.23
C	pool	A	organic	0.65(C) - 2.43	0.69
N	concentration	A, B, C, D, E	all	105(N) + 17.6	0.71
N	pool	A, B, C, D, E	all	14.0(N) + 0.90	0.39
N	concentration	B, C, D, E	all	158(N) + 2.1	0.91
N	pool	B, C, D, E	all	14.2(N) + 0.70	0.70
N	concentration	A	mineral	360(N) - 32.8	0.94
N	pool	A	mineral	35.7(N) - 4.27	0.74
N	concentration	A	organic	5.93(N) + 161¶	0.00
N	pool	A	organic	11.7(N) - 1.63#	0.77

† A = pond center, B = pond edge, C = toe slope, D = back slope, and E = shoulder.

‡ All = Ponds 1–10; mineral = Ponds 1, 2, 7, 8, 9, and 10; organic = Ponds 3, 4, 5, and 6.

§ Intercept not significant ($p = 0.304$)

¶ Slope not significant ($p = 0.894$).

Intercept not significant ($p = 0.052$).

Table 5. Comparison of least squares means for soil total Hg (THg), C and N densities and concentrations, and C/N ratio across landscape positions and depth increments. Capital letters compare depth increments within a landscape position; lowercase letters compare landscape positions within a depth increment. For all comparisons, *n* = 10.

Parameter	Landscape position	Depth increment			
		0–2 cm	2–5 cm	5–10 cm	10–15 cm
THg, ng cm^{-3}	pond center	58.1Aa†	63.4Aa	54.9Aa	39.5Ba
	pond edge	39.6Bb	49.2Aab	42.2ABab	30.5Cab
	toe slope	30.7Bb	35.2Ab	33.0ABb	24.5Cb
	back slope	31.6Bb	35.3Ab	30.2Bb	19.0Cb
	shoulder	31.6Bb	34.7Ab	30.5Bb	19.0Cb
C, g cm^{-3}	pond center	0.048Aa	0.047Aab	0.042Aa	0.040Aa
	pond edge	0.046Aab	0.049Aa	0.038Bab	0.026Cab
	toe slope	0.040Abc	0.039Abc	0.031Bab	0.020Cb
	back slope	0.036Ac	0.036Ac	0.027Bb	0.015Cb
	shoulder	0.038Ac	0.037Ac	0.029Bab	0.017Cb
N, g cm^{-3}	pond center	0.003Aa	0.003Aa	0.003Aa	0.002Aa
	pond edge	0.003ABa	0.003Aa	0.002Ba	0.002Cab
	toe slope	0.002Ab	0.002Ab	0.002Bb	0.001Cbc
	back slope	0.002Ab	0.002Ab	0.002Bb	0.001Cc
	shoulder	0.002Ab	0.002Ab	0.002Bb	0.001Cbc
THg, ng g^{-1}	pond center	210Aa	168Ba	122Ca	73.7Da
	pond edge	112Ab	93.2Ab	52.7Bb	25.0Cb
	toe slope	45.0Ac	42.3Ab	31.4Bb	18.3Cb
	back slope	40.0Ac	37.5Ab	27.1Bb	13.8Cb
	shoulder	39.5Ac	36.7Ab	27.0Bb	13.7Cb
C, %	pond center	21.8Aa	18.3ABa	15.4Ba	15.4Ba
	pond edge	13.6Ab	9.36Bb	5.01Cb	2.26Cb
	toe slope	6.05Abc	4.75Bb	3.02Cb	1.50Db
	back slope	4.60Ac	3.83Bb	2.46Cb	1.07Db
	shoulder	4.78Ac	3.89Bb	2.55Cb	1.19Db
N, %	pond center	1.255Aa	1.03Ba	0.797Ca	0.723Ca
	pond edge	0.765Ab	0.560Bb	0.306Cb	0.153Cb
	toe slope	0.297Ac	0.250Bb	0.174Cb	0.102Db
	back slope	0.242Ac	0.208Bb	0.147Cb	0.078Db
	shoulder	0.250Ac	0.212Bb	0.153Cb	0.084Db
C/N	pond center	16.5Ac	16.2Ab	16.2Aa	16.1Aa
	pond edge	17.4Abc	16.4ABab	15.3BCa	13.8Ca
	toe slope	19.8Aa	18.8ABa	17.3Ba	14.6Ca
	back slope	19.1Aab	18.6Aab	16.9Ba	14.3Ca
	shoulder	19.1Aab	18.6Aab	17.0Aa	16.9Aa

† Least squares means with the same capital letter within rows are not significantly different at $\alpha = 0.05$. Least squares means with the same lowercase letter within columns are not significantly different at $\alpha = 0.05$.

high wetland surface area had high C concentrations. Although not investigated here, these studies explore the implications of aerobic versus anaerobic conditions on THg adsorption. The authors of these studies attributed the curvilinear distribution to the nature of Hg being less adsorbed to C in wetland sediments compared with upland soils, indicating that the type of organic matter is important in addition to the amount. Other researchers have obtained similar results, indicating negative relationships between THg and C in forest litter with high C concentrations versus positive relationships in underlying mineral soil with lower C concentrations (Navrátil et al., 2014; Obrist et al., 2009).

The type of organic matter and level of decomposition of organic matter may explain the negative relationship between THg and C concentration in organic ponds (Table 4). For ponds with organic rather than mineral substrates (Ponds 3, 4, 5, and 6) (Table 1), it may be that, although C concentration continues to increase, the bulk of the organic matter is less decomposed and thus is not as conducive to Hg binding than if it were more decomposed.

For THg versus C concentration relationships considering plots with all landscape positions together, where pond center was excluded, and for mineral ponds, slopes ranged from 4.80 to 16.9 (Table 4). These slopes are comparable to those reported elsewhere for mineral soils, ranging from ~5 to 20 (Juillerat et al., 2012; Mitchell et al., 2012; Navrátil et al., 2014; Obrist et al., 2009). For pond centers with organic substrates, a slope of –4.69 was observed for THg versus C concentration (Table 4), whereas other researchers have reported slopes of approximately –11 to –10 for organic forest floor horizons (Navrátil et al., 2014; Obrist et al., 2009).

The better general fit of regression lines for relationships between THg and N versus fits for THg and C has been attributed to Hg binding to ligands containing N groups (Gunda and Scanlon, 2013; Obrist et al., 2009). Unlike C, slopes for THg concentration versus N among organic ponds had a positive slope, which supports the hypothesis that Hg may bind to N groups. For THg versus N concentrations considering plots with all landscape positions together, where pond center was excluded, and for mineral ponds, slopes ranged from 105 to 360 (Table 4). These slopes are comparable to a slope of 151.8 reported for forest floor horizons in Sierra Nevada ecosystems (Obrist et al., 2009). For pond centers with organic substrates, an insignificant slope of 5.93 was observed for THg versus N concentration, whereas Obrist et al. (2009) reported a slope of 110, and Navrátil et al. (2014) found an insignificant negative correlation between THg and N concentrations in Czech Republic forest floors. Again, the differences in N trends may be attributed to differences in Hg binding to ligands containing N groups and highlights that the type of organic matter affects Hg accumulation (Navrátil et al., 2014).

Relationships between THg with C and N pools all had positive slopes, thus increasing mass of THg can be predicted by increasing masses of C or N (Table 4). Slopes for relationships between THg and C pools ranged from 0.46 to 1.31, and slopes for THg versus N pools ranged from 11.7 to 35.7 (Table 4). These slopes are similar to slopes of 0.38 and 13 for THg versus C and N pools, respectively, reported in a different forested landscape (Obrist et al., 2009). At pond center, slopes for THg versus C and N pools were larger for mineral ponds compared with organic ponds, whereby peak THg pools were found in mineral ponds. This may be a function of differing bulk densities, where bulk density in organic ponds ranged from 0.10 to 0.23 g cm^{-3} (0–15 cm), and mineral ponds ranged from 0.39 to 1.46 g cm^{-3}. Because pools correct for bulk density, organic ponds with low bulk densities have lower THg mass than mineral ponds with higher bulk densities.

Depth Increment Comparisons of Total Hg, C, N, and C/N Ratio

For most THg, C, and N concentrations and densities considered, depth increments at the pond center landscape position were not significantly different compared with upland landscape positions where more significant differences were observed (Table 5). Uniformity among depth increments at pond center may be a result of several possibilities, including that these increments are saturated from inputs from upslope, that infiltration or fluctuating groundwater serve to distribute elements evenly throughout the profile, or that deposition rates are consistent over time. At pond center, there was no statistical difference among increments for C or N densities despite decreases in concentrations down the profile, demonstrating that consideration of concentrations alone does not provide an accurate representation of how masses are distributed.

Apart from pond center, THg densities in the 0- to 2-cm increments were lower than the 2- to 5-cm increments, although concentrations in those increments were not statistically different (Table 5). Several studies have found THg pools to be smaller in the forest floor compared with underlying mineral soil, despite higher concentrations in the former (Fleck et al., 1999; Grigal et al., 1994, 2000; Juillerat et al., 2012; Mitchell et al., 2012; Nater and Grigal, 1992). This has been attributed to the inverse relationship between organic matter and bulk density, which can result in high concentrations with small pools and vice versa (Grigal et al., 1994; Juillerat et al., 2012; Navrátil et al., 2014). Although the forest floor was not explicitly considered here, bulk densities in the 0- to 2-cm increments at all landscape positions (range, 0.31–0.83 g cm^{-3}) were the lowest among increments, indicating higher organic matter (and C as discussed below). However, although bulk densities increased with depth for all landscape positions (up to 0.94–1.51 g cm^{-3}), THg densities did not increase accordingly (Table 5). The decrease of THg densities with depth beyond the 2- to 5-cm increment is an indication that deeper soils are less affected by postindustrial deposition from above than those closer to the surface and that the THg is not geogenic.

Despite the correlations between THg and C (Table 4), THg concentration does not decrease down the profile in the same manner as C. For THg concentrations apart from pond center, the 0- to 2-cm and 2- to 5-cm increments are not statistically different, with the 5- to 10-cm and 10- to 15-cm increments being less than the increment before (Table 5). A similar trend has been observed across horizons in a lake-plain wetland, where C concentration decreased with each horizon, whereas THg concentration peaked in the middle horizon (Nave et al., 2017). Other studies have explored the implications of SOM decomposition on Hg binding. The authors of these studies attributed Hg binding with more decomposed SOM, the dominant form in the mineral layer (Gladkova and Malinina, 1999; Grigal et al., 1994). Although organic matter binding was not investigated here, it may be that, despite a lower concentration of C in mineral soils, this C is associated with more decomposed organic matter, allowing for more stable binding with THg compared with C in less decomposed organic matter found in the upper soils. This may allow the THg concentration to remain constant in the 0- to 2-cm to 2- to 5-cm increments despite a decrease in C concentration with depth.

Regarding concentrations and pools of THg, C, and N for individual depth increments, the pond center landscape position was greater than upland landscape positions for most increments (Table 5). The relative consistency of each depth increment among upland landscape positions coupled with peaks at pond center for each increment further suggests that upland runoff downslope plays an important role in THg accumulation in the ponds. In addition, each increment is enriched at pond center and not in uplands, indicating that transport down the profile occurs to a greater degree at pond center compared with upland landscape positions.

Despite the near-identical trends of C and N concentrations, there were some differences in C/N ratios among depth increments and landscape positions (Table 5). For the 0- to 2-cm increment, the C/N ratio at pond center was lower than at upland landscape positions, which indicates the relative enrichment of N at the soil surface of pond centers. Among depth increments, the C/N ratio declines with depth at the pond edge, toe slope, and back slope landscape positions. In uplands, Kolka et al. (2014) also reported a larger C/N ratio higher in the profile (forest floor) compared with the underlying mineral soil (32.2 and 23.3, respectively) despite C and N both decreasing from forest floor to the underlying mineral soil. Depth increments in this study were not statistically different at both the pond center and shoulder landscape positions (Table 5), which indicates that C and N are similar throughout the 15-cm depth sampled.

Upland Influences on Total Hg at Pond Edge

Approximately 56% of the variance of THg pool (0–15 cm) at the pond edge landscape position can be accounted for by the linear combination of upland slope length (18.5%), mean upland graminoid cover class (15.5%), basin area (14.7%), and mean upland tree height class (7.04%) with the regression equation

$$S_{THg} = 2.32 + 0.0460a + 0.000355b - 0.0833c + 0.122d \quad [2]$$

where S_{THg} is THg pool (0–15 cm) at pond edge, a is slope length (m), b is basin area (m^2), c is mean upland graminoid cover class (%), and d is mean upland mean tree height class (m).

Catchment factors that influence water flow and leaching are important to THg accumulation in lakes (Matilainen et al., 2001), and slope grade (Betemariam et al., 2013; Burns et al., 2012; Kainz and Lucotte, 2006; Lorey and Driscoll, 1999) and basin area in relation to lake area (Betemariam et al., 2013; Matilainen et al., 2001) have been implicated as such drivers. Here, basin area was predictive, although only slope length, not slope grade, was significant in the model (Eq. [2]). Upland graminoid cover was found to have a negative influence on THg at pond edge, which follows from grass buffers slowing runoff and intercepting pollutants (Veum et al., 2009).

Canopy features have also been cited as predictive for soil Hg due to the implications that coverage and species have on deposition, litterfall production, and throughfall inputs (Bushey et al., 2008; Juillerat et al., 2012; Perry et al., 2006; Witt et al., 2009). Although

other researchers have found that the degree of decomposition of SOM, as evidenced by C and N contents whereby lower C/N ratios correspond to a higher degree of decomposition, predicts elevated THg concentration in soils (Gunda and Scanlon, 2013; Juillerat et al., 2012; Obrist et al., 2011), C/N ratio was not a good predictor of THg concentration in this study (Eq. [2]). Mean upland tree height class was included in the model, which may result in increased surface area to collect dry deposition; however, several other upland vegetative factors were not found to be predictive of THg at pond edge, such as tree diameter, tree basal area, canopy openness, tree cover, shrub cover, and litter/debris cover. Certain types of vegetation and vegetative structure receive differential THg deposition (Bushey et al., 2008; Johnson et al., 2007; Laacouri et al., 2013); thus, exploration of THg pools and concentrations of vegetative components themselves may be useful.

Given the relatively low landscape position that pond edge occupies and landscape trends observed here where THg appears to be accumulating downslope at pond center (Table 3), it might be expected that the upland THg pool would be a driver of pools lower in the landscape. However, upland THg pools were not found to be a driver of pools at the pond edge landscape position (Eq. [2]). The THg concentrations and pools at pond edge were not statistically different from upland landscapes positions (Table 3).

CONCLUSIONS

Few studies have investigated THg in small, closed-basin vernal ponds and demonstrated how the interaction of bulk density with concentration can affect the total mass present in a given volume. Given that these basins do not have a surface water outlet, these ponds may have the potential to accumulate large quantities of Hg over time. Without an outlet, such accumulation may be sequestered in place, which decreases the likelihood of its transport to lacustrine habitats; however, risks to organisms that use these ponds remain. The accumulation of THg, C, and N observed at the pond center landscape position and the relatively uniform saturation of these elements throughout the profile likely indicate that these elements are being transported downslope, possibly in association with dissolved organic matter. The consistent pattern of greater THg, C, and N concentrations and densities at the pond center landscape position for each depth increment also suggests that downward transport throughout the profile is more pronounced at pond center and/or is indicative of accumulation from upslope in that location. Higher concentrations of THg, C, and N in the upper soil depths at upland landscape positions indicate that deposition from above is a more likely source than geogenic processes. Forested ecosystems play important roles in cycling of Hg, C, and N, making an understanding of the distribution and drivers of Hg in these environments key to their management objectives and strategies.

APPENDIX

Table A1. Mean basin (including pond) vegetation characteristics.

Basin	Dominant tree species	Canopy openness	Basal area tree coverage	Tree height class midpoint	Tree diameter	Cover class midpoint							
						Bare ground	Water	Litter/ debris	Nonvascular	Graminoid	Forb	Shrub	Tree
		%	$m^2\ ha^{-1}$	m	cm	%							
1	*Acer saccharum*, *Populus tremuloides*, *Fraxinus nigra*	6.30	32.3	15.5	36.0	1.40	19.5	94.0	5.20	10.7	21.9	7.60	85.8
2	*Acer saccharum*, *Betula papyrifera*, *Fraxinus nigra*	4.58	23.2	12.7	24.0	4.70	0.00	82.2	5.90	2.80	34.8	9.40	89.6
3	*Populus* spp., *Acer* spp., *Fraxinus nigra*	7.26	22.1	12.1	27.9	3.20	3.30	61.7	16.4	8.60	41.2	20.7	84.9
4	*Fraxinus nigra*, *Acer saccharum*, *Populus grandidentata*	7.63	31.6	20.5	25.4	7.60	12.7	79.4	4.10	3.30	11.9	19.1	88.1
5	*Acer saccharum*, *Tilia americana*, *Populus grandidentata*	10.9	23.5	17.8	28.7	4.40	29.5	73.5	5.50	13.4	33.6	23.7	80.3
6	*Fraxinus nigra*, *Acer saccharum*, *Populus grandidentata*	9.70	34.2	21.7	31.5	12.3	27.1	70.4	0.70	6.60	9.10	11.0	84.5
7	*Acer saccharum*, *Tilia americana*, *Fraxinus nigra*	7.92	35.5	19.9	29.8	8.00	24.7	77.8	0.30	7.80	20.6	37.7	84.0
8	*Acer saccharum*, *Tilia americana*, *Fraxinus nigra*	4.63	30.0	13.6	30.7	4.40	0.00	90.2	2.60	7.00	13.6	2.80	88.0
9	*Populus tremuloides*, *Fraxinus nigra*, *Betula papyrifera*	8.15	26.0	20.7	33.9	5.50	19.5	83.3	1.50	6.80	27.7	14.3	78.6
10	*Fraxinus nigra*, *Tilia americana*, *Acer saccharum*	3.22	31.3	17.4	29.6	12.7	0.00	77.3	0.20	19.4	16.5	10.3	88.8

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