

# Organic horizon and mineral soil mercury along three clear-cut forest chronosequences across the northeastern USA

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**Abstract** Mercury (Hg) is a globally distributed pollutant trace metal that has been increasing in terrestrial environments due to rising anthropogenic emissions. Vegetation plays an important role in Hg sequestration in forested environments, but increasing tree removal for biofuels and wood products may affect this process. The long-term effect of clear-cutting on forest soil Hg remains uncertain, since most studies are limited to measuring changes for < 10 years following a single harvest event. The chronosequence approach, which substitutes space for time using forest stands of different ages since clear-cutting, allows for investigation of processes occurring over decades to centuries. Here, we utilized three clear-cut forest soil chronosequences across the northeastern USA to understand Hg accumulation and retention over several decades. Total Hg concentrations and pools were quantified for five soil depth increments along three chronosequences. Our results showed Hg concentrations and pools decreased in the initial 20 years following clear-cutting. Mineral soil Hg pools decreased 21–53% (7–14 mg m<sup>-2</sup>) between 1–5-year-old stands and 15–25-year-old stands but mineral soil Hg pools

recovered in 55–140-year-old stands to similar values as measured in 1–5-year-old stands. Our study is one of the first to demonstrate a decrease and recovery in Hg pool size. These changes in Hg did not correspond with changes in bulk density, soil C, or pH. We utilized a simple two-box model to determine how different Hg fluxes affected organic and mineral soil horizon Hg pools. Our simple model suggests that changes in litterfall and volatilization rates could have caused the observed changes in organic horizon Hg pools. However, only increases in leaching could reproduce observed decreases to mineral soil Hg pools. Further studies are needed to determine the mechanism of Hg loss from forest soils following clear-cutting.

**Keywords** Trace metal · Pollution · Forest harvest · Soil disturbance · Mineral soil · Biogeochemistry

## Introduction

Mercury (Hg) is a globally distributed pollutant trace metal, found widely in terrestrial ecosystems due to anthropogenic emissions (Smith-Downey et al. 2010; Streets et al. 2017) as well as natural emissions (Hylander and Meili 2003). Mercury is sourced to the atmosphere from geologic and human emissions as both gaseous and particulate phases (Miller et al. 2005; Driscoll et al. 2007). After emission and transport in the atmosphere, Hg is deposited as gaseous aerosols, dry particulates, or dissolved ions in precipitation (Miller et al. 2005; Mitchell et al. 2008; Obrist et al. 2011; Yu et al. 2014). Global atmospheric deposition rates of Hg have been increasing since the Industrial Revolution due to human pollution from artisanal gold mining and coal combustion (Kamman and Engstrom 2002; Hylander and Meili 2003; Driscoll et al. 2007; Streets et al. 2017). Mercury can be lost from forest soils via two main

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processes: volatilization and leaching. Depending on temperature, atmospheric pressure, species, and moisture content, Hg can volatilize from the soil into the atmosphere, from as deep as several decimeters (Schlüter 2000; Obrist et al. 2014). Mercury may also be lost from the soil profile due to leaching, generally not as dissolved ions but rather an organic or inorganic complex (Shanley et al. 2008; Brigham et al. 2009; Mitchell et al. 2008).

In the northeastern USA, much of this deposited natural and pollutant Hg has been sequestered in forest soils due to vegetation (Seigneur et al. 2004; Driscoll et al. 2007; Obrist et al. 2011; Richardson et al. 2013; Blackwell et al. 2014; Yu et al. 2014). Plant leaves act as surfaces for interception of atmospheric Hg due to stomatal uptake or surface sorption (Weathers et al. 2006; Zhang et al. 2009; Blackwell et al. 2014). In addition, the uptake of Hg within leaf tissue and subsequent deposition to the soil can be an important flux, constituting from 20 to 70% of total Hg flux to forest soils (Rea et al. 2002; Blackwell and Driscoll 2015). Upon reaching upper soil horizons, atmospheric Hg is sorbed or complexed by soil organic matter (SOM), as well as inorganic ligands, and secondary minerals (Schlüter 2000; Gabriel and Williamson 2004; Brigham et al. 2009). Plants influence the total soil quantity of SOM, the formation of reduced sulfur compounds and other organic ligands, and the generation of acidity from organic acids, all of which impact Hg sequestration in soils (Gabriel and Williamson 2004). As a result, forest soils may retain Hg for decades to centuries in their organic and upper mineral soil horizons (Stankwitz et al. 2012; Richardson et al. 2014; Yu et al. 2014).

Timber harvesting, especially clear-cutting, disrupts vegetation's effect on Hg cycling and may cause less sequestration and greater mobilization of Hg from forest soils across the northeastern USA (Grigal 2003; Driscoll et al. 2007; Bishop et al. 2009; Demers et al. 2013; Eklof et al. 2014). Clear-cutting, where all trees > 5 cm in diameter are removed, is still a common forest harvest technique in the northeastern USA and is important for understory plants and animals that rely on disturbed forest stands (USDA 1973, 2011; Pierce et al. 1993; Palmer 2008). The removal of the trees can cause substantial impacts on the forest soil physicochemical properties that promote Hg sequestration. Log-skidding and the movement of heavy machinery during clear-cutting can destroy soil aggregation, mix soil horizons, and increase the decomposition rate of SOM (Bormann et al. 1968; Federer 1984; Fuller et al. 1988; Johnson et al. 1991; Ballard 2000; Bishop et al. 2009). Forest soil C pools may decrease significantly for decades after clear-cutting due to stimulation of SOM decomposition by soil microorganisms (Diochon et al. 2009; Zummo and Friedland 2011; Grand and Lavkulich 2012; Petrenko and Friedland 2014; Kaarakka et al. 2014). Since Hg is strongly sorbed to SOM (Weng et al. 2002; Gabriel

and Williamson 2004), changes to SOM pools affect Hg sequestration in soil (Demers et al. 2013; Eklof et al. 2014; Kiikkilä et al. 2014).

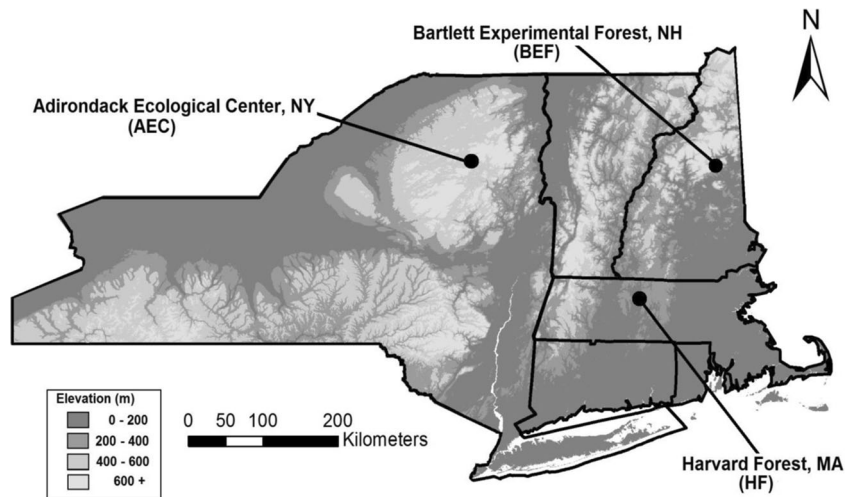
Previous studies on the effects of clear-cutting on Hg in forest soils have focused on changes < 10 years following a clear-cutting event (e.g., Porvari et al. 2003; Eklof et al. 2014). These studies have observed significant Hg increases in soil leachate and river water, suggesting Hg exports from soils after forest harvest (Porvari et al. 2003; Bishop et al. 2009; Eklof et al. 2014). However, we are not aware of studies that address the effect of clear-cutting on Hg for longer time periods. The objective of this study was to determine long-term effects of clear-cutting (> 20 years) on Hg concentrations and pools in forest soils and if effects were driven by changes in soil properties. Understanding Hg biogeochemistry following clear-cutting is essential for quantifying impacts from forest practices and the role of forest soils in providing the ecosystem service of mediating the global Hg cycle.

## Methods

### Study areas and stand descriptions

Our study utilized clear-cut forest stand-age chronosequences at three study areas on United States Forest Service and privately owned research lands. The study areas were the Adirondack Ecological Center (AEC) in Newcomb, NY, Bartlett Experimental Forest (BEF) in Bartlett, NH, and Harvard Forest (HF) in Petersham, MA (Fig. 1, Table 1). At each study area, we focused on a set of forest stands ranging in age from 1 year up to at least 140 years, to substitute space for time. All three study areas are temperate biomes (Table 1), with mean summer temperatures ranging from 16 to 22 °C and mean winter temperatures ranging from − 8 to 0 °C (PRISM Climate Group, <http://prism.oregonstate.edu>). Mean annual precipitation (MAP) ranged from 1074 to 1326 mm year<sup>−1</sup> (Table 1). Four forest stands at AEC and HF and five forest stands at BEF were selected with assistance from local land managers and ages were determined using harvest records for the area (Leak and Smith 1996; Zummo and Friedland 2011; Vario et al. 2014; Petrenko and Friedland 2014). The forest stands ranged in size from less than 1 ha to several hectares and were of varying ages and clear-cutting histories. Forests were northern hardwoods consisting of *Betula* spp., *Populus* spp., *Fagus grandifolia* Ehrh., *Quercus* spp., and *Acer* spp., with limited, interspersed eastern hemlock (*Tsuga canadensis*). Soils were Spodosols at AEC and BEF, while the most southern study area, HF, was dominated

**Fig. 1** Location of study areas are displayed. State borders are outlined



by Inceptisols (Table 1). All soils were moderately to well-drained.

**Soil sampling methods**

Sampling took place during the summers of 2012 and 2013. Three plots were sampled at each forest stand. Plots were at least 10-m apart from each other, at < 10° slope, away from pits and drainage areas, and > 1 m from the nearest tree (Vario et al. 2014). The organic horizon (Oi + Oe + Oa), commonly referred to as the forest floor, was sampled by collecting three 15 × 15 cm blocks of all organic material overlying the mineral soil (Johnson et al. 1982). At each plot, three soil cores were extracted using a gas-powered auger (Earthquake™ 9800B) with a 9.5-cm diameter diamond-tipped, stainless steel drill bit following methods of Rau et al. (2011). The mineral soil was sampled incrementally in four depth increments: 0 to 10 cm; 10 to 20 cm; 20 to 30 cm; and 30 to 45 cm (Zummo and Friedland 2011). Soil horizons for the 0–10-cm depth increment were A, E, and Bs horizons at AEC and BEF but were A and Bw horizons at HF (Table 2). After collection, the three cores were bulked together by depth increment for each plot in order to create a larger, more representative

sample. In total, this study includes 108 individual soil cores, with physical and chemical soil properties measured for 36 bulked soil cores. Soils were placed into polyethylene bags, stored in a cool, dry area, and transported to the laboratory following sampling. The addition of Hg from the use of a diamond-tipped stainless steel drill bit is very unlikely given the traditional composition of steel.

**Soil and mercury analyses**

Bulked soil samples were air-dried in paper bags at 25 °C to a constant weight. Air-drying can lead to a systematic loss of Hg from samples during drying, which necessitated our use of a closed porous medium to decrease photoreduction and increase removal of water. Oven drying was not performed in order to reduce Hg volatilization. Subsamples were ground to < 0.5 mm using a ball mill. Percent C per gram of soil was determined by combustion of 5-mg oven-dried soil in a Carlo-Erba NA 1500 Element Analyzer (Thermo Scientific, Waltham, MA). For every 12 samples, we included a duplicate, a quartz blank, and the standard reference materials (SRMs) atropine and acetanilide. Soil pH was determined using a 1:2 soil–water slurry for mineral soil samples and

**Table 1** Study area descriptions

Study area	Code	Latitude	Longitude	Elevation	MAT <sup>a</sup>	MAP <sup>a</sup>	Soil taxonomy <sup>b</sup>	Stand ages
		d.d.	d.d.	m	°C	mm year <sup>-1</sup>		years
Adirondack Ecological Center	AEC	43.9694	− 74.1647	550	5.2	1074	Spodosols/Podzols	1, 19, 55, 140 <sup>c</sup>
Bartlett Experimental Forest	BEF	44.0778	− 71.2828	304	6.2	1326	Spodosols/Podzols	5, 25, 120, 140 <sup>c</sup>
Harvard Forest	HF	42.4875	− 72.1875	360	7.8	1197	Inceptisols/Cambisol	5, 25, 64, 84

<sup>a</sup> Thirty-year averages (1981–2010) for mean annual temperature (MAT) and mean annual precipitation (MAP) were retrieved from PRISM Climate Group, Oregon State University, <http://prism.oregonstate.edu>

<sup>b</sup> USDA Soil Taxonomy/FAO Soil Classification

<sup>c</sup> One hundred forty years reflect written records and actual age of stand may be older

**Table 2** Soil morphology and select soil properties by study area

Study area	Depth increment	Typical horizons	Carbon concentrations	pH	% clay
Adirondack	cm		mg g <sup>-1</sup>	log units	g g <sup>-1</sup>
Ecological Center (AEC)	Organic horizons	Oi, Oe, Oa	355 ± 37	4.3 ± 0.5	–
	0–10	A, E, Bs	104 ± 16	4.5 ± 0.3	11 ± 2
	10–20	B, Bs	36 ± 5	4.4 ± 0.2	11 ± 2
	20–30	B, Bs	66 ± 8	4.8 ± 0.4	10 ± 1
	30–45	BC	42 ± 8	4.9 ± 0.4	7 ± 2
Bartlett Experimental Forest (BEF)	Organic horizons	Oi, Oe, Oa	318 ± 34	4.5 ± 0.6	–
	0–10	A, E, Bs	50 ± 4	4.6 ± 0.7	10 ± 3
	10–20	B, Bs	31 ± 16	4.9 ± 0.7	7 ± 3
	20–30	B, Bs	28 ± 3	5.1 ± 0.4	5 ± 3
	30–45	BC	24 ± 3	5.2 ± 0.2	4 ± 2
Harvard Forest (HF)	Organic horizons	Oi, Oe, Oa	333 ± 29	4.4 ± 0.5	–
	0–10	A, Bw1	62 ± 2	4.4 ± 0.3	2 ± 1
	10–20	Bw1	73 ± 15	4.9 ± 0.2	9 ± 3
	20–30	Bw1, Bw2	18 ± 3	5.0 ± 0.2	8 ± 3
	30–45	Bw2	15 ± 2	5.1 ± 0.2	8 ± 3

1:4 soil–water slurry for organic horizon samples. The pH of the slurries was measured with a VWR 8015 electroprobe pH meter (VWR, Radnor, PA).

Mercury concentrations were quantified using a Milestone Direct Mercury Analyzer–80 (Milestone Inc., Shelton, CT) in which 100 ± 10 mg subsamples were weighed into nickel boats and combusted. To ensure instrument precision and accuracy, every 10 samples included a replicate, a preparation blank, SRM 1547, and SRM 2711a. Preparation blanks were below detection limits of 0.25 ng Hg g<sup>-1</sup> sample and duplicate variations were within 4% RSD. SRM Hg measurements were within 5% of their certified values.

### Data analyses

Concentration data were compared for each depth increment at each study area. Mercury pools were calculated using Hg concentration data and bulk density measurements for each depth increment from the soil cores, excluding rock fragments. Organic horizon pools were calculated as the Hg concentration multiplied by the organic horizon mass per square meter. Mineral horizon pools were determined by multiplying the Hg concentration with the soil bulk density for each depth interval and summing the four mineral soil increments from 0 to 45 cm. Differences in soil properties, Hg concentrations, and pools were quantified using the nonparametric Kruskal-Wallis test and post hoc Mann-Whitney *U* test for each study area. With linear regressions, we explored correlative relationships among metal concentrations, C concentrations, and pH in organic and mineral soil horizons for each study areas.

### Simple soil Hg box model

A simple two-box model was utilized to estimate how clear-cutting impacts Hg pools in the organic and mineral horizons. We modeled estimates of fluxes in and out of the organic horizon (Eq. 1) and mineral soil (Eq. 2):

$$\Delta Organic\ horizon\ pool = F_{litterfall} + F_{atm\_dep} - F_{uptake} - F_{volatilization} - F_{OH\_leaching} \tag{1}$$

$$\Delta Mineral\ soil\ pool = F_{weathering} + F_{OH\_leaching} - F_{uptake} - F_{MS\_leaching} \tag{2}$$

We assumed the annual weathering input ( $F_{weathering}$ ) and root uptake ( $F_{uptake}$ ) of Hg in the organic and mineral soil were negligible (Grigal 2003; Frescholtz et al. 2003). Annual leachings from the organic horizon to the mineral soil ( $F_{OH\_leaching}$ ) and from the mineral soil out of the soil profile ( $F_{MS\_leaching}$ ) were calculated as a set percentage of the total Hg pool for each respective horizon based upon Schlüter and Gäth (1997), Demers et al. (2007), and Yu et al. (2014).

We investigated the effect of clear-cutting on Hg cycling by varying the annual rate of fluxes in the model that are dependent on vegetation in order to quantify the influence of each flux on the system. Annual Hg volatilization (also known as evasion and evaporation) rates ( $F_{volatilization}$ ) from organic horizons and mineral soil horizons at the eight sites were adapted from Yu et al. (2014), which were calculated from a synthesis of Hg evasion measurements from across the northeastern USA by Denkenberger et al. (2012). Given that Hg volatilization rates are higher in open soils compared to forest soils

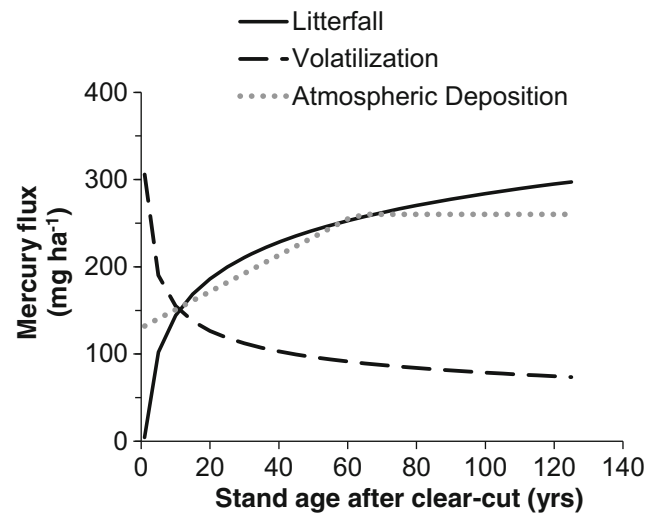
(Schlüter 2000; Choi and Holsen 2009), annual volatilization rates in the model were assumed to be highest immediately after the clear-cutting event and decreased logarithmically to a minimum equivalent to the regional mean volatilization rate from Yu et al. (2014). Annual litterfall fluxes of Hg ( $F_{\text{litterfall}}$ ) from vegetation were taken as the geometric mean of fluxes reported by Demers et al. (2007), Blackwell and Driscoll (2015), Blackwell et al. (2014), and Richardson and Friedland (2015). We assumed  $F_{\text{litterfall}}$  from vegetation to be 0 immediately after clear-cutting and treated logging slash as immediately incorporated into the forest floor. We increased  $F_{\text{litterfall}}$  with stand age logarithmically in proportion to the total aboveground biomass using allometric and measured values from Hughes and Fahey (1994), Keeton et al. (2011), and Ter-Mikaelian and Korzukhin (1997) with an upper boundary matching estimates from Demers et al. (2007) and Richardson and Friedland (2015). Similar to  $F_{\text{litterfall}}$ , we modeled annual wet and dry atmospheric deposition of Hg ( $F_{\text{atm\_dep}}$ ) with the assumption that deposition was the lowest immediately after clear-cutting due to decreased leaf area for gaseous and particulate interception in which the upper boundary values match data interpolated from Miller et al. (2005), Chalmers et al. (2014), and Yu et al. (2014).

Equations 1 and 2 were solved under five scenarios, three of which (scenarios 1–3) exclusively focused on atmospheric deposition, volatilization, and litterfall fluxes of Hg to compare their individual effects on organic horizon Hg pools. Under scenario 1,  $F_{\text{volatilization}}$  and  $F_{\text{litterfall}}$  were assumed to be constant while  $F_{\text{atm\_dep}}$  increased with time as described above and shown in Fig. 2. In scenario 2,  $F_{\text{atm\_dep}}$  and  $F_{\text{litterfall}}$  were held constant while  $F_{\text{volatilization}}$  decreased with time. In scenario 3,  $F_{\text{volatilization}}$  and  $F_{\text{atm\_dep}}$  were held constant while  $F_{\text{litterfall}}$  increased with time using estimates of Hg in litterfall with stand age from as mentioned above (Fig. 2). The last three scenarios (scenarios 4–6) estimated changes to the input and output Hg fluxes to the mineral soil. In scenario 4, all fluxes to the organic horizon ( $F_{\text{atm\_dep}}$ ,  $F_{\text{volatilization}}$ , and  $F_{\text{litterfall}}$ ) were combined and we focused on changes in the mineral soil Hg pool. In scenario 5,  $F_{\text{MS\_leaching}}$  was increased by an order of magnitude to determine its influence on losses from the mineral soil Hg pool. In scenario 6,  $F_{\text{OH\_leaching}}$  was also increased by an order of magnitude and  $F_{\text{MS\_leaching}}$  was also kept as the elevated rate from scenario 4.

## Results

### Mercury concentrations and pools

We compared Hg concentrations for each horizon among forest stands in each study area to determine if there were statistically significant linear trends. HF exhibited a significant positive trend in organic horizon Hg concentrations with stand



**Fig. 2** Litterfall ( $F_{\text{litterfall}}$ ), volatilization ( $F_{\text{volatilization}}$ ), and atmospheric deposition ( $F_{\text{atm\_dep}}$ ) inputs for our simple box model. In brief, annual Hg volatilization rates ( $F_{\text{volatilization}}$ ) from organic horizons and mineral soil horizons at the eight sites were adapted from Yu et al. (2014). Given that Hg volatilization rates are higher in open soils compared to that in forest soils (Schlüter 2000; Choi and Holsen 2009), annual volatilization rates in the model were assumed to be the highest immediately after the clear-cutting event and decreased logarithmically to a minimum equivalent to the regional mean volatilization rate from Yu et al. (2014). Annual litterfall fluxes of Hg ( $F_{\text{litterfall}}$ ) from vegetation were taken as the geometric mean of fluxes reported by Demers et al. (2007), Blackwell et al. (2014), and Richardson and Friedland (2015). We assumed  $F_{\text{litterfall}}$  from vegetation to be zero immediately after clear-cutting, treating logging slash as immediately incorporated as the forest floor, and increased  $F_{\text{litterfall}}$  with stand age logarithmically proportional to the total aboveground biomass using allometric and measured values. Similar with  $F_{\text{litterfall}}$ , we modeled annual wet and dry atmospheric depositions of Hg ( $F_{\text{atm\_dep}}$ ) with the assumption that deposition was the lowest immediately after clear-cutting due to decreased leaf area for gaseous and particulate interception using upper boundary values that match data interpolated from Miller et al. (2005), Chalmers et al. (2014), and Yu et al. (2014)

age (Table 3). However, organic horizon Hg concentrations were negatively correlated with stand age for AEC (Table 3). Mercury concentrations in the 0–10-cm mineral depth increment were positively correlated with stand age for BEF and HF (Table 3). Fewer depth intervals from 10 to 45 cm exhibited significant trends; however, 20–30-cm Hg concentrations at HF and 30–45-cm Hg concentrations at BEF were positively correlated with stand age (Table 3). Organic horizon Hg pools were positively correlated with stand age at AEC and BEF and only mineral soil Hg pools were positively correlated with stand age at HF (Table 3).

We used the Kruskal-Wallis test and post hoc Mann-Whitney  $U$  test to compare Hg concentrations and pools among forest stands at each study area to test for age-specific differences as opposed to general linear trends. At AEC and BEF, Hg concentrations in the 10–20- and 20–30-cm depth intervals had significant decreases between 1–5-year-old stands and 20–25-year-old stands ( $p < 0.05$ ), which was followed by a significant increase in mineral soil Hg in

**Table 3** Pearson *R* correlation coefficients and *p* values for linear regressions of Hg concentrations and pools with stand age are displayed. Only information for significant linear regressions are present. At AEC and HF (*n* = 4) and BEF (*n* = 5)

	AEC	BEF	HF
<b>Hg concentrations</b>			
Organic horizon	- 0.94**	-	+ 0.96**
0–10 cm	-	+ 0.57*	+ 0.99**
10–20 cm	-	-	-
20–30 cm	-	-	+ 0.58*
30–45 cm	-	+ 0.77*	-
<b>Hg pools</b>			
Organic horizon	+ 0.73*	+ 0.90**	-
Mineral soil	-	-	+ 0.78

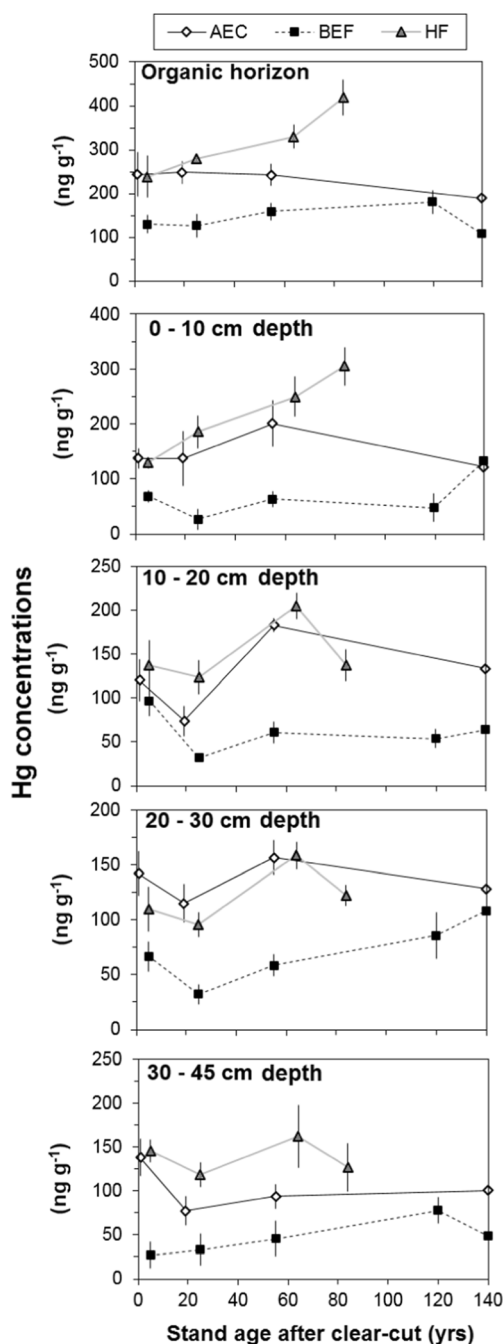
\**p* values < 0.05; \*\**p* values < 0.001

55–65-year old stands (*p* < 0.05) (Fig. 3). At all study areas, mineral soil Hg pools decreased significantly from 1–5-year-old stands to 20–25-year-old stands, which was followed by a significant increase in mineral soil Hg in 55–65-year-old stands (*p* < 0.05) (Fig. 4). Mercury pools were similar between stands that were 50–65 years old and those that were 80–140 years old (Fig. 4). The change in Hg pools from 1–5-year-old stands to 20–25-year-old stands was approximately 14 mg m<sup>-2</sup> (- 42%) at AEC, 7 mg m<sup>-2</sup> (- 53%) at BEF, and 9 mg m<sup>-2</sup> (- 23%) at HF.

Organic and mineral horizon Hg concentrations at AEC and BEF (30–265 ng g<sup>-1</sup>) were within the range of Hg concentrations measured in previous studies in the northeastern USA (Evans et al. 2005; Demers et al. 2007; Drevnick et al. 2012; Obrist et al. 2011; Juillerat et al. 2012; Richardson et al. 2013; Blackwell et al. 2014). The Harvard Forest had higher Hg concentrations in comparison to the other study areas presented here and other studies published for this region. Forest soils at HF may have had higher Hg concentrations due to its closer proximity to metropolitan and industrial areas in Connecticut, Massachusetts, and New York (Yu et al. 2014). Additional information about the spatial variation in Hg concentrations and pools in forest soils can be found in Richardson et al. (2013) or Yu et al. (2014).

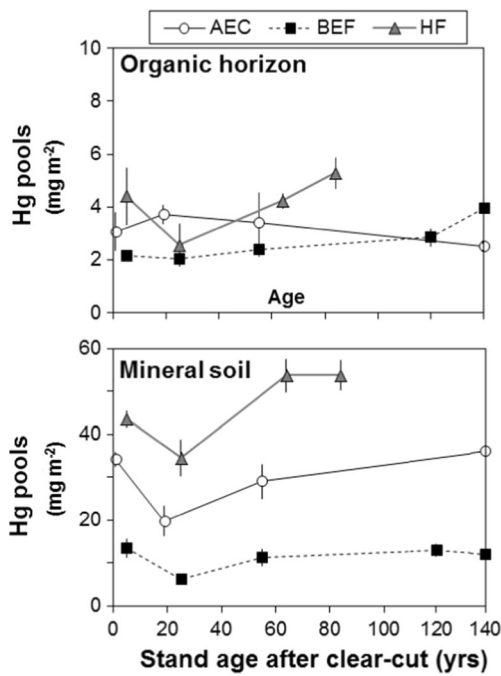
**Soil physicochemical properties**

Soil pH for each depth increment was not significantly different among chronosequence forest stands at each study area (*p* > 0.10, Kruskal-Wallis test; Supplemental Fig. 1). AEC soil C concentrations increased from the 1-year-old stand to the 19-year-old stand, which was followed by decreases at the 55- and 140-year-old stands (*p* > 0.10, Kruskal-Wallis test; Supplemental Fig. 2). BEF soil C concentrations did not show



**Fig. 3** Mercury concentrations for the five soil depth intervals at each forest stand in each clear-cut chronosequence are shown. Error bars are ± 1 standard error

any significant change overall. However, soil C concentrations in the 55-year-old-stand were significantly higher than those in the other stands (*p* > 0.10, Kruskal-Wallis test; Supplemental Fig. 2). Moreover, soil C concentrations for the 30–45-cm depth interval at BEF was highest at the 5- and 55-year-old stands and significantly lower for the 120- and 140-year-old stands (*p* > 0.10, Kruskal-Wallis test; Supplemental Fig. 2). HF soil C concentrations in the organic horizon, 20–30- and 30–45-cm depth intervals increased with



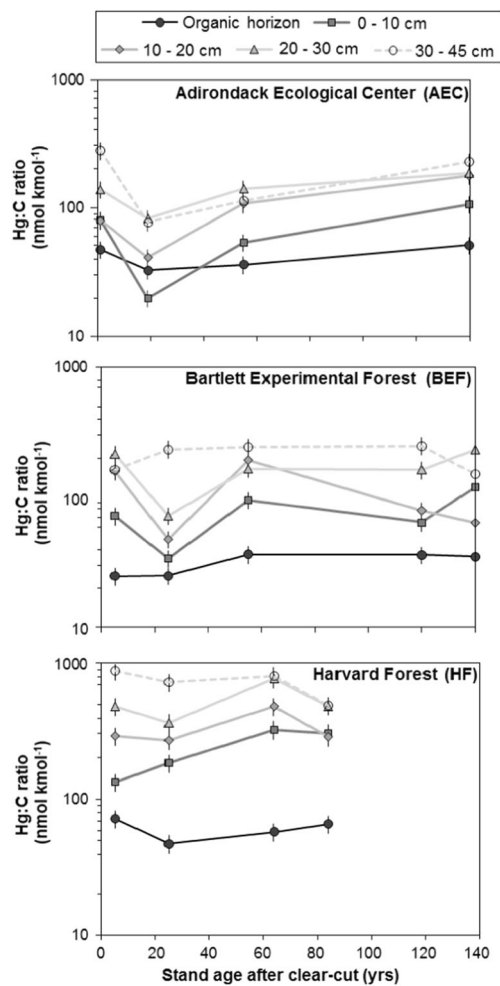
**Fig. 4** Mercury pools in the organic horizons and mineral soil horizons at each forest stand in each clear-cut chronosequence are displayed. Error bars are  $\pm 1$  standard error

increasing stand age but decreased significantly in the 10–20-cm depth interval with stand age (Supplemental Fig. 2). Soil bulk density did not vary significantly along the three chronosequences (Petrenko and Friedland 2014; Richardson et al. 2017).

We used linear regressions and Hg:C ratios to explore potential links of Hg with soil C and pH. Soil C concentrations were strongly positively correlated with Hg at each study area ( $R^2 > 0.65$ ,  $p < 0.05$ ) (Supplemental Fig. 3). Soil pH was poorly correlated with Hg concentrations for each study area ( $R^2 < 0.25$ ,  $p > 0.10$ ) (data not shown). We explored the Hg:C ratio to investigate changes in Hg with respect to C molar stoichiometry. Organic horizon Hg:C ratio ( $\text{nmol kmol}^{-1}$ ) decreased for AEC and HF between 1–5-year-old stands and 15–30-year-old stands and subsequently increased with stand age (Fig. 5). Mineral soil Hg:C ratio decreased for AEC and BEF. At AEC, all soil horizons exhibited a decrease between sites 1–5-year-old stands and 15–30-year-old stands with a subsequent increase in mineral soil Hg:C ratio at older stands (Fig. 5). At BEF, all soil horizons depths except the 30–45-cm depth interval had a similar trend as AEC in Hg:C ratio with stand age (Fig. 5). At HF, all soil horizon depths except the 0–10-cm depth interval had a similar trend as AEC and BEF in Hg:C ratio with stand age (Fig. 5).

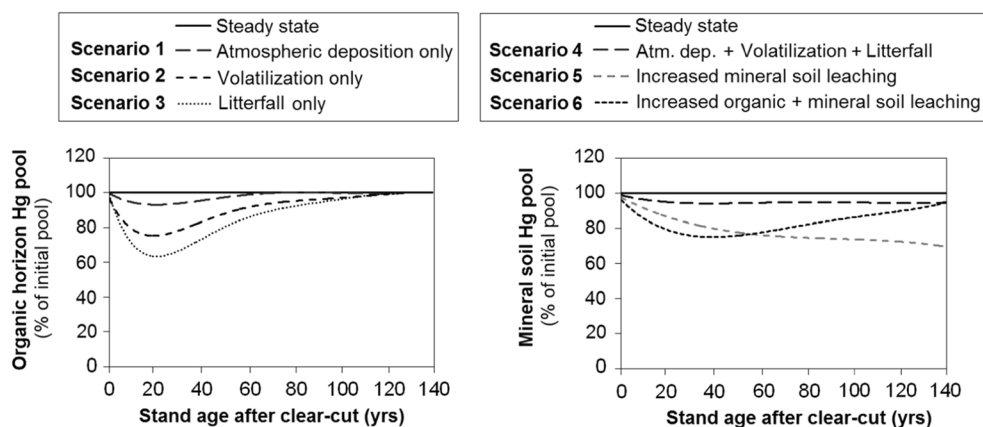
**Simple soil Hg box model**

A simple two-box model was implemented to estimate how changes in litterfall ( $F_{\text{litterfall}}$ ), atmospheric deposition



**Fig. 5** The Hg:C ratios for the five depth intervals at each stand in each clear-cut chronosequence are shown. Error bars are  $\pm 1$  standard error

( $F_{\text{atm\_dep}}$ ), volatilization ( $F_{\text{volatilization}}$ ), and leaching ( $F_{\text{OH\_leaching}}$ ;  $F_{\text{MS\_leaching}}$ ) impact Hg pools in the organic and mineral horizons (Fig. 6). Under scenario 1, only  $F_{\text{atm\_dep}}$  was not held constant (Fig. 2) and resulted in a decrease of  $\sim 9\%$  of the original Hg pool. In scenario 2, only  $F_{\text{volatilization}}$  was not held constant (Fig. 2). By increasing the volatilization rate, organic horizon Hg pools decreased to a minimum of  $\sim 76\%$  of the original organic horizon Hg pool. Under scenario 3,  $F_{\text{litterfall}}$  was not held constant (Fig. 2) and decreased to a lower minimum of 64% of the initial size of the organic horizon Hg pool. From scenarios 1–3, it is clear that changes in atmospheric deposition rates had the least effect on the organic horizon Hg pool while volatilization and litterfall were more dominant based upon and derived from many previous studies on Hg in forests (e.g., Rea et al. 2002; Demers et al. 2007; Yu et al. 2014; Richardson and Friedland 2015). As shown in Fig. 6, the organic horizon Hg pool decreased to a greater extent for approximately 50 years following clear-cutting. These results match the observation for changes in organic horizon Hg pool at HF.



**Fig. 6** Model outputs are shown for our simple box models. Eqs. 1 and 2 were run under six scenarios: scenarios 1–3 investigated individual variable effects on organic horizon Hg pools while scenarios 4–6 investigated cumulative effects on mineral soil Hg pools. Descriptions of the six scenarios can be found in “Methods” section, “Simple soil Hg box model.” In brief, under scenario 1,  $F_{\text{volatilization}}$  and  $F_{\text{litterfall}}$  were assumed to be constant while  $F_{\text{atm\_dep}}$  increased with stand age (Fig. 5). In scenario 2,  $F_{\text{atm\_dep}}$  and  $F_{\text{litterfall}}$  were held constant while

$F_{\text{volatilization}}$  decreased with time (Fig. 5). In scenario 3,  $F_{\text{volatilization}}$  and  $F_{\text{atm\_dep}}$  were held constant while  $F_{\text{litterfall}}$  changed with stand age (Fig. 5). Scenarios 4–6 show cumulative effects of fluxes on mineral soil pools. In scenario 4, the cumulative effect of organic horizon Hg fluxes on mineral soil Hg pools is shown. In scenario 5, conditions were the same as scenario 4 except  $F_{\text{MS\_leaching}}$  was increased by an order of magnitude. In scenario 6, conditions were the same as scenario 5, except  $F_{\text{OH\_leaching}}$  was also increased by an order of magnitude

Under the conditions of scenario 4, changes in Hg fluxes to the organic horizon had minimal impacts on mineral soil Hg pools, decreasing to 92% of the initial pool with no recovery after 50 years (Fig. 6). To test if observed changes in the mineral soil Hg pool were due to increased leaching, the same conditions for scenario 4 were applied in scenario 5 but with the addition of increasing  $F_{\text{MS\_leaching}}$  by an order of magnitude. By increasing  $F_{\text{MS\_leaching}}$ , the modeled mineral soil Hg pool decreased substantially more and at a nearly constant rate but did not return to its starting size after the first 50 years following clear-cutting as found in our field observations (Fig. 4). Thus, in scenario 6, the leaching of Hg from the organic horizon to the mineral soil ( $F_{\text{OH\_leaching}}$ ) was also increased by an order of magnitude to facilitate recovery of the Hg pool. The results from scenario 6 showed that mineral soil Hg pools decreased to 71% of the initial pool size and returned to their starting sizes as observed at our three study areas (Fig. 6).

## Discussion

### The chronosequence approach

For a chronosequence to be effective, two assumptions must be met. First, environmental factors such as climate and Hg deposition rates over the chronosequence period must be relatively constant and forest soils utilized in all ages of the chronosequence must be comparable to detect changes due to clear-cutting. Mercury emissions and deposition rates have not been constant throughout the chronosequence time span. Atmospheric deposition of Hg in the northeastern USA has

increased by 2–7 times the pre-1850s rate (Kamman and Engstrom 2002). Although deposition is increasing, we argue that deposition during the chronosequence time span is still of comparable magnitude (Streets et al. 2017). Moreover, increased deposition would result in lower Hg concentrations and pools at the most recently clear-cut stands due to decreased leaf interception and litterfall and we did not observe this effect (Figs. 3 and 4).

In the second chronosequence assumption, forest stands along the chronosequence must have similar ecosystem and soil physicochemical properties to detect an effect from clear-cutting as opposed to spatial variations. Based upon our sampling procedures and comparisons of the physicochemical soil properties among forest stands at each study area, we argue that the forest stands in this study meet the second assumption. Discussions in Vario et al. (2014) and Petrenko and Friedland (2014) provide details regarding the similarities among stands in the study areas. In brief, we have held soil sampling to areas of similar slope, drainage, physical disturbances, and microtopography. Moreover, bulk density, clay percent, and parent material were similar among forest stands for each study area (Petrenko and Friedland 2014; Richardson et al. 2017). By holding all climatic and edaphic properties similar across forest stands, differences in Hg concentrations and pools may be attributed to changes in Hg biogeochemistry since the clear-cutting event.

Lastly, logging techniques and characteristics have changed through time. The oldest forest stands were likely harvested with hand tools and logs dragged across the soil surface by large animals, while more recent harvests utilized crawler tractors with rubber-tired skidders during the late 1960s (Yanai et al. 2003). Another variation in the technique



is the amount of slash left behind, which was not quantified and may have an unknown effect on Hg dynamics. However, it is important to note that our results support our assumption of similar soil properties (e.g., pH and bulk density) for stands at each study area.

### Mercury concentrations and pools with clear-cutting frequency

The objective of this study was to determine long-term effects of clear-cutting (> 20 years) on Hg concentrations and pools in forest soils and if they were driven by changes in soil properties. We expected that Hg pools in the organic and mineral horizons would decrease following forest clear-cutting due to decreased inputs and increased mobilization for at least a decade and return to their original pool size (see Covington 1981). Our observation of lower Hg pools, and to a limited extent lower Hg mineral soil concentrations, supports our expectation that clear-cutting decreases Hg accumulation and retention in forest soils (Figs. 3, 4). However, this impact was primarily observed for only 15–25-year-old stands. When comparing 1–5-year-old stands to 15–25-year-old stands, we calculated that mineral soil Hg pools in the top 45 cm of the soil profile decreased by 23–53%, respectively. This finding agrees with that of previous studies in other regions, which observed that forestry increased Hg export from forest soils following timber harvesting (e.g., Porvari et al. 2003; Skyllberg et al. 2009; de Wit et al. 2014; Eklof et al. 2014).

Our study is one of the first to demonstrate that Hg biogeochemistry in forest soils can be impacted for time scales longer than 10 years. Our results show that Hg pools in mineral soil Hg pools recovered to a similar size in 55–65-year-old stands. This is an important finding since most studies on Hg following clear-cutting have only focused on changes occurring within 10 years following harvest. Another notable result was that only organic horizon Hg pools at HF changed over time while AEC and BEF did not exhibit a temporal trend. We expected that organic horizon Hg pools would be substantially affected by clear-cutting. For example, Mitchell et al. (2012) found that organic horizon Hg pools were the most sensitive to disruptions such as wildfire, logging, and blowdown events, while mineral soil Hg pools were commonly not found to be significantly different. However, the organic horizon may be too dynamic to identify long-term changes in Hg retention in forest soils. For example, the residence time of other trace metal pollutants, such as Pb, in the organic horizon has been estimated to be 21–143 years at upland forest stands in Massachusetts, Vermont, and New Hampshire (Richardson et al. 2014). Thus, changes occurring over > 100 years may not be recorded in the organic horizon. Mineral soil retention of natural and pollutant Hg has been estimated on the order of

centuries to millennia (Demers et al. 2007; Stankwitz et al. 2012; Yu et al. 2014; Richardson and Friedland 2015).

We investigated the Hg:C ratio to determine if changes in C were driving the observed changes in mineral soil Hg pools. We expected the addition of C compounds, particularly labile compounds, from clear-cutting coupled with increased soil temperatures from greater solar radiation may destabilize Hg:C in SOM compounds (Ballard 2000; Diochon et al. 2009; Skyllberg et al. 2009). Petrenko and Friedland (2014) found decreasing soil C pools in the mineral soil horizons across forest chronosequences in the northeastern region, which included AEC, BEF, HF, and additional study areas. However, Hg:C ratios in mineral soil horizons displayed the same trend in the mineral soil as the Hg pools. Thus, we conclude that the variations in the Hg:C ratios in the mineral soil along the chronosequences were dominated by shifts in Hg concentrations, not changes in C concentrations. We speculate differences in the Hg:C ratios may not have manifested because Hg and C systems are not entirely coupled. For example, Hg is strongly sorbed to sulfur groups in SOM (Skyllberg et al. 2009), and atmospheric deposition and volatilization are inorganic pathways that control Hg in soil. In addition, the source and type of C in the soil can affect how tightly coupled Hg is to C. Smith-Downey et al. (2010) suggested that the majority of Hg stored in soils is bound to fast-cycling C compounds, while the primary, more stable forms of C stored in mineral soils hold less Hg. In other words, the loss of more labile C may have resulted in a disproportionate decrease in the Hg pool.

### Simple two-box Hg model

We expected that changes to Hg in the organic and mineral horizons could correspond to decreased Hg inputs (atmospheric deposition or litterfall) or higher Hg losses (volatilization or leaching). We conclude that combinations of volatilization and litterfall are most likely the source of decreased organic horizon Hg pools at HF. Atmospheric deposition was not a large enough flux to substantially decrease the organic horizon Hg pool. In spite of the cumulative combination of fluxes (increased  $F_{\text{volatilization}}$  and decreased  $F_{\text{atm\_dep}}$  and  $F_{\text{litterfall}}$ ) in scenario 4, mineral soil Hg pools were not substantially affected by changes in fluxes to the organic horizon (Fig. 6). This was likely due to the fact that the magnitude of fluxes in and out of the organic horizon were small ( $0.01\text{--}0.03\text{ mg m}^{-2}\text{ year}^{-1}$ ) compared to the size of the mineral soil Hg pool ( $7\text{--}63\text{ mg m}^{-2}$ ). Thus, fluxes in and out of the mineral soil must be larger than the few empirical measurements suggest (see [Supplemental Information](#)), especially since most measurements were of short duration and not in clear-cut forests. The results from the model suggest that Hg losses from the mineral soil were likely not driven by changes in litterfall, atmospheric deposition, or volatilization but were primarily

dependent on Hg leaching from the mineral soil. Leaching rates had to be substantially increased in scenario 4, to reproduce the observed changes in Hg pools in Fig. 4. This conclusion is based upon many assumptions, such as Hg volatilization from the mineral soil being limited (see Obrist et al. 2014), negligible weathering inputs, negligible plant uptake, and the inclusion of the most pertinent parameters and variables used in equations used to describe changes in  $F_{\text{volatilization}}$ ,  $F_{\text{atm\_dep}}$ , and  $F_{\text{litterfall}}$ . In spite of these limitations, this simple two-box model approach offered insights to the overall functioning of Hg biogeochemistry in the organic and mineral soil horizons following clear-cutting.

## Conclusions

We expected that clear-cutting would decrease Hg concentrations and pools in organic and mineral soil horizons for decades to centuries after the event. Our results suggest Hg concentrations and organic and mineral soil Hg pools were impacted (21–53% decrease in Hg pools), but only for the first 20 years following clear-cutting. Afterwards, Hg concentrations and pools recovered to a pool size similar to those in 1–5-year-old stands. Our simple two-box model showed that litterfall and volatilization rates were most likely responsible for changes to organic horizon Hg pools but only increased leaching could substantially impact mineral soil horizons to match observations. The Hg:C ratio data suggest that changes to Hg were decoupled from overall C concentrations. Our chronosequence method has been able to provide an initial investigation of these changes and further studies should consider investigating which fraction of Hg (i.e., exchangeable, SOM-complexed, or mineral-bound) is most affected by clear-cutting.

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