

# Singular and Combined Effects of Blowdown, Salvage Logging, and Wildfire on Forest Floor and Soil Mercury Pools

Carl P. J. Mitchell,<sup>\*,†</sup> Randall K. Kolka,<sup>‡</sup> and Shawn Fraver<sup>‡</sup>

<sup>†</sup>Department of Physical and Environmental Sciences, University of Toronto Scarborough, 1265 Military Trail, Toronto, Ontario, Canada M1C 1A4

<sup>‡</sup>USDA Forest Service, Northern Research Station, Grand Rapids, Minnesota 55744, United States

**S** Supporting Information

**ABSTRACT:** A number of factors influence the amount of mercury (Hg) in forest floors and soils, including deposition, volatile emission, leaching, and disturbances such as fire. Currently the impact on soil Hg pools from other widespread forest disturbances such as blowdown and management practices like salvage logging are unknown. Moreover, ecological and biogeochemical responses to disturbances are generally investigated within a single-disturbance context, with little currently known about the impact of multiple disturbances occurring in rapid succession. In this study we capitalize on a combination of blowdown, salvage logging and fire events in the sub-boreal region of northern Minnesota to assess both the singular and combined effects of these disturbances on forest floor and soil total Hg concentrations and pools. Although none of the disturbance combinations affected Hg in mineral soil, we did observe significant effects on both Hg concentrations and pools in the forest floor. Blowdown increased the mean Hg pool in the forest floor by 0.76 mg Hg m<sup>-2</sup> (223%). Salvage logging following blowdown created conditions leading to a significantly more severe forest floor burn during wildfire, which significantly enhanced Hg emission. This sequence of combined events resulted in a mean loss of approximately 0.42 mg Hg m<sup>-2</sup> (68% of pool) from the forest floor, after conservatively accounting for potential losses via enhanced soil leaching and volatile emissions between the disturbance and sampling dates. Fire alone or blowdown followed by fire did not significantly affect the total Hg concentrations or pools in the forest floor. Overall, unexpected consequences for soil Hg accumulation and by extension, atmospheric Hg emission and risk to aquatic biota, may result when combined impacts are considered in addition to singular forest floor and soil disturbances.



## INTRODUCTION

Forest soils are a globally significant sink of atmospherically deposited mercury (Hg),<sup>1</sup> but disturbances can lead to large emissions of Hg from surface litter and soils, which may contribute significantly to atmospheric pools.<sup>2</sup> Several factors influence the amount of Hg in surface soils, including levels of atmospheric deposition, soil–air emissions, soil chemistry, and both natural and anthropogenic disturbances. Increases in Hg deposition may lead to corresponding increases in soil Hg concentrations and pools, both at local scales near point sources, as well as across larger-scale regional gradients, but some variation may exist, particularly with respect to patterns of carbon accumulation.<sup>3–5</sup> Soil organic matter is a key factor influencing the accumulation of Hg in soils.<sup>1</sup> At the molecular level, reduced sulfur groups in soil organic matter are particularly important for Hg binding.<sup>6</sup> Losses of Hg in the soil pool are through both leaching of Hg, which is mostly associated with dissolved organic matter,<sup>7,8</sup> and Hg<sup>0</sup> emission

to the atmosphere. Solar radiation, moisture, microbial activity, and land cover can exert a strong control on the magnitude of Hg<sup>0</sup> emission.<sup>9–13</sup> Consequently, ecosystem processes affecting these physical, chemical and biological characteristics will alter soil Hg pools and directly affect both atmospheric and aquatic loading.

Forest harvesting and some forest management practices represent landscape-level disturbances, which may alter soil Hg pools and fluxes to both the atmosphere and surface waters. Through the removal of trees and disruption of the forest floor by machinery, forest harvesting leads to increased Hg mobilization and increased concentrations of Hg in watershed runoff,<sup>14</sup> as well as periphyton,<sup>15</sup> zooplankton,<sup>16</sup> and fish in

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watershed lakes.<sup>17</sup> Bishop et al.<sup>18</sup> estimate that one-tenth to one-quarter of the Hg in fish in high latitudes is an indirect consequence of forest harvesting.

Wildfire is another widespread disturbance that affects many processes related to Hg, including emission of soil Hg to the atmosphere, deposition elsewhere, and bioaccumulation in the food chain. The emission of Hg from forest soils due to fire is of particular significance because without this disturbance, the strong Hg–organic matter association in soils would otherwise result in relatively long-term sequestration of Hg.<sup>19</sup> Annual global Hg emissions due to biomass burning have been estimated at  $675 \pm 240 \text{ Mg yr}^{-1}$ , with potential for emissions to increase as temperatures increase in boreal regions and the pool of Hg stored in soils is subject to more frequent and intense wildfire.<sup>20</sup> A number of studies have measured soil Hg either before and after fire<sup>21</sup> or compared burned to nearby unburned areas post fire<sup>22–26</sup> and generally found that fire leads to large releases of forest floor Hg to the atmosphere, as well as Hg emission from surface mineral soil horizons, with the effect of fire decreasing with soil depth. The release of Hg during fire is estimated to be >95% in the gaseous elemental form, with the remainder associated with particulate matter, although other studies have found greater particulate releases under certain conditions, particularly when fuel moisture is high.<sup>27–29</sup> Recent studies in northern Minnesota demonstrate that the time since fire influences O- and A-horizon soil Hg concentrations with progressively higher concentrations as the time since fire increases.<sup>30</sup> Fire also enhances local Hg deposition, thereby increasing the potential for bioaccumulation in the food chain, but results have thus far been equivocal.<sup>31</sup> Studies of aquatic biota have shown that fire may lead to elevated Hg concentrations<sup>17</sup> or little change in Hg concentrations.<sup>16,32,33</sup>

Other than forest harvesting and fire, there has been little research on the effect of other forest disturbances or management practices on Hg in soils. In particular, forest blowdown due to windthrow is a widespread forest disturbance within the Great Lakes region<sup>34–37</sup> and in Europe.<sup>38</sup> For example, windstorms in recent decades have damaged 140 000 ha of forest in Wisconsin,<sup>39</sup> 18 200 ha in Wisconsin,<sup>40</sup> and 200 000 ha in Minnesota,<sup>34</sup> and evidence from U.S. Public Land Surveys estimate that windstorm patches during the 1800s reached 34 000 ha to 344 000 ha in this same region.<sup>37,41</sup> In Europe, between 1958 and 2001, approximately  $0.11\% \text{ yr}^{-1}$  of standing timber volume was damaged by blowdown, an extent approximately equal to the area affected by wildfire.<sup>38</sup> Historical studies and inferential studies about more frequent and extensive severe weather occurrences also suggest that wind damage to forests has increased in the recent past and/or will continue to increase.<sup>36,38,42–44</sup>

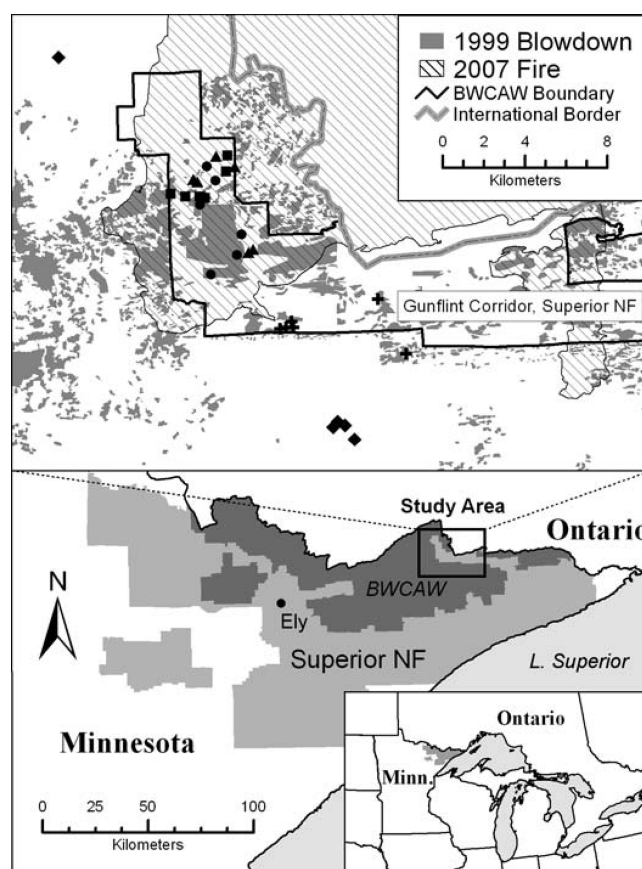
Intuitively, blowdown is likely to provide large, but short-term biomass inputs to the forest floor, which could increase Hg pools. Alterations to light and moisture regimes, however, have as yet unknown consequences for Hg accumulation. To the best of our knowledge, no field data currently exists to describe how forest blowdown affects Hg accumulation in or loss from soils, but Munthe et al.<sup>45</sup> have presented a modeling approach for estimating leaching of Hg from forest soils following a combination of blowdown and salvage logging. Following blowdown, a common practice is to salvage fallen timber with the intent of mitigating economic losses, and reducing fire severity and intensity by reducing fuel loads, although the efficacy by which this practice actually reduces fire severity or intensity is contentious.<sup>46</sup> This management practice

represents a mechanical disturbance to the forest floor through the use of machinery and alters biomass, light, and moisture, which is likely to alter soil Hg pools. This potential impact on soil Hg pools has also not been previously studied.

While plentiful research exists to describe the impacts of singular disturbances, particularly wildfire, on soil chemistry and forest ecology, little research has addressed the cumulative impact of multiple disturbances occurring in relatively rapid succession on forest structure and composition (but see refs 47 and 48) and no previous research has applied this concept to Hg cycling. Paine et al.<sup>49</sup> suggest that multiple disturbances may interact to produce “ecological surprises” not easily predicted from an understanding of single disturbances. In this study we capitalize on a sequence of recent events—blowdown, salvage logging, and fire—in the Superior National Forest of northern Minnesota that allow us to directly address the concepts presented above. Specifically, our objective was to evaluate the effect of these disturbances, singly and in combination, on forest floor and soil Hg pools and concentrations. Results from this study will advance our knowledge of soil Hg accumulation and emissions related to ecological disturbance, and they will inform forest management decisions aimed at reducing the ecological risk of Hg exposure in forest ecosystems.

## EXPERIMENTAL SECTION

**Study Site.** The study was conducted within the Superior National Forest, northeastern Minnesota (Figure 1). The area



**Figure 1.** Location of study sites in northeastern Minnesota showing the various treatment combinations (■ = blowdown-salvage logging-fire, ● = blowdown-fire, ▲ = fire, + = blowdown, ◆ = control).



has a mean annual precipitation of about 71 cm, with mean July and January temperatures of 17 °C and −8 °C.<sup>48</sup> Stony sandy loam soils were formed from ground moraine until deposited by the Rainy Lobe during the Wisconsin glacial period. The portion of the Superior National Forest where this study was conducted had been dominated by mature jack pine (*Pinus banksiana*) prior to the series of disturbances described below.

On July 4, 1999, severe straight-line winds damaged approximately 200 000 ha of sub-boreal forest, including large areas within the Superior National Forest. Between the fall of 1999 and fall 2002, the US Forest Service contracted salvage logging in the blowdown area to reduce fuel loads and fire risk.<sup>50</sup> In May 2007, after a period of drought, a forest fire (Ham Lake fire) burned approximately 15 000 ha through this same landscape. The patchiness of these three disturbances created various treatment combinations of blowdown, salvage logging, and wildfire: blowdown-salvage logging-fire (BSF), blowdown-fire (BF), fire (F), blowdown (B), and control (C). Because all salvage logged areas ultimately burned, the combination of blowdown-salvage logging-no fire did not exist.

**Experimental Design, Sample Collection and Analysis.** This study takes a modified experimental approach to previous research investigating the singular effect of fire on Hg pools<sup>22–26</sup> by investigating undisturbed and various disturbed areas in close (approximately 10 km) proximity to one another. In 2008, we established six study sites within treatment types BSF, BF, F, and B; in 2009, we added six control (C) sites, for a total of 30 sites. We selected eight of these sites because of preexisting data from another study;<sup>51</sup> we chose the remaining 22 sites by random selection from a complete set of potential sites for each treatment type. Undisturbed control sites were selected to be as near as possible to the disturbed sites, have similar soils, and support mature jack pine. By necessity, the blowdown-only and the undisturbed control sites had to be located outside of the burn perimeter. Thus, these sites are more dispersed than those clustered within the burn perimeter, but still generally within 10 km of the burn cluster (Figure 1). Importantly, our analyses assume sites were similar in structure prior to this sequence of disturbances. Archived U.S. Forest Service inventories (from 1976 to 1994), as well as predisturbance data from Johnson,<sup>52</sup> were available for a subset of the disturbed sites. Data show a narrow range of stand origin dates (1903–1915) and similar basal areas (BSF at 29.6 m<sup>2</sup> ha<sup>−1</sup>, *N* = 3 sites; BF at 27.6 m<sup>2</sup> ha<sup>−1</sup>, *N* = 5; F at 28.7 m<sup>2</sup> ha<sup>−1</sup>, *N* = 5; and B at 29.8 m<sup>2</sup> ha<sup>−1</sup>, *N* = 4). Postdisturbance (2009) data for the control (C) sites show a higher mean basal area of 34.3 m<sup>2</sup> ha<sup>−1</sup>, as would be expected given the later sampling date. Given the predisturbance similarity among sites, we do not believe their locations had any meaningful influence on our findings.

Depending on site size, 6–10 circular plots, each 200 m<sup>2</sup>, were established along 40 m grid intervals originating from a randomly chosen starting point, for a total of 239 plots. These plots were established to characterize vegetation as part of a companion study. At a standard predetermined location within each plot, we collected forest floor material and mineral soils. Forest floor material is defined as the sum of litter (Oi horizon) and duff (Oe and Oa horizons), with woody debris removed, and was collected within a 25 cm diameter collar down to mineral soil. Mineral soil was collected with a 4 cm diameter core tube to a depth of 10 cm or where we met a bedrock contact. As is standard, we removed roots and stones larger than 2 mm diameter. The mean depth of mineral soil sample

retrieved was 8.6 cm, with a range of 2–10 cm. All 239 forest floor samples were analyzed, but to minimize analytical burden, 60 of the 239 mineral soil samples were analyzed (2 per site, which is 12 per treatment, randomly chosen from all samples).

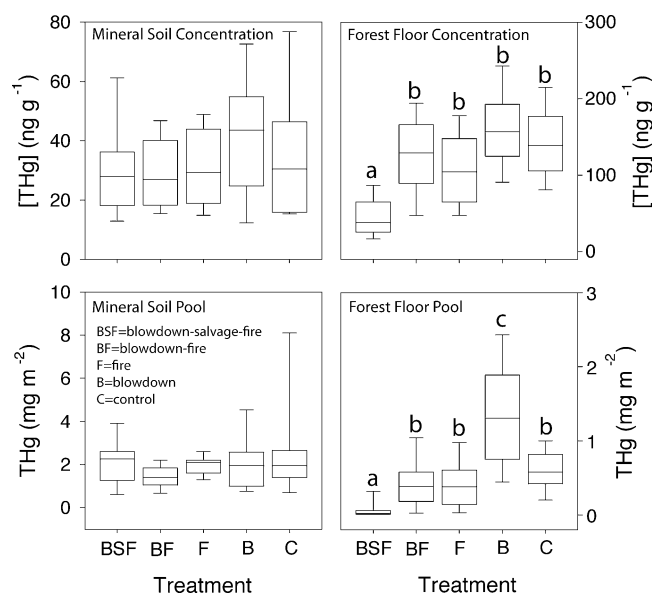
Both forest floor and mineral soil samples were air-dried and ground with a Wiley mill to homogenize. Sample concentrations were corrected for true dry mass by subsampling the air-dried sample and oven drying for 24 h at 105 °C. Samples were analyzed for total carbon (C) by combustion on a LECO TruSpec Total Elemental Analyzer. To determine total Hg (THg) concentrations, samples were microwave digested in high-purity nitric acid and analyzed by cold vapor atomic fluorescence on a Tekran 2600 automated Hg analyzer. Briefly, aliquots of digested sample were diluted with high-purity deionized water and all Hg species in the sample were oxidized by addition of BrCl and left to react for 18 h. During analysis, samples are mixed with SnCl<sub>2</sub> to reduce all Hg present in the sample to Hg<sup>0</sup> vapor, which is stripped from the liquid and then dual amalgamated and thermally desorbed from gold traps prior to determination by atomic fluorescence spectroscopy. Calibrations with certified standards were conducted daily. Quality assurance measures such as sample replicates, blanks, matrix spikes, and analysis of the certified reference material MESS-3 were included every 5–10 samples. Mean ± standard deviation sample replication was 3.9 ± 3.2%, matrix spike recovery was 92 ± 7%, and recovery of the certified reference material was 98 ± 7%. The calculated mean detection limit was 1.3 ng g<sup>−1</sup>. To determine forest floor Hg pools per unit area (μg m<sup>−2</sup>), THg concentrations (ng g<sup>−1</sup> dry weight) in forest floor samples were multiplied by the total dry mass of the sample collected within a 25 cm diameter collar. Mineral soil pools (mg m<sup>−2</sup>) were calculated by multiplying THg concentration by soil bulk density and soil depth, to a soil depth of up to 10 cm. Thus, the mineral soil pool represents the pool either to bedrock or within the upper 10 cm of mineral soil because of limits related to bedrock depth or core lengths.

**Statistical Analyses.** The effect of each treatment on forest floor and mineral soil Hg (both concentrations and pools) was examined using mixed-model analyses of variance (ANOVA). Prior to ANOVA, data were tested for normality using the Shapiro-Wilk *W* test. Data were square-root transformed where necessary so that data were normally distributed and the homogeneity of variance was improved. In addition, we used the minimum variance quadratic unbiased method of model estimation to account for deviations from normality that persisted following transformation. In the mixed-model ANOVA, treatment was considered the fixed effect and site the random effect. For analyses with statistically significant treatment effects (*p* < 0.05), significant differences were further investigated by pairwise treatment comparisons, using Bonferroni adjustments for multiple comparisons. All analyses were conducted using SAS version 9.2.<sup>53</sup>

## RESULTS AND DISCUSSION

**Forest Disturbance Impacts on Forest Floor and Soil THg Concentrations and Pools.** In previous work, researchers have found that fire generally results in significantly reduced forest floor Hg pools, but may have little to no effect on underlying mineral soils depending on the intensity and/or severity of the burn.<sup>23–25</sup> The intent of this study was to determine whether different or multiple forest impacts occurring in sequence (blowdown, salvage logging, and/or wildfire) differed from singular impacts or from one another in

terms of THg concentrations and pools remaining in the forest floor and mineral soil, postdisturbance. We observed significant impacts of forest disturbances on forest floor THg, but none of the treatments investigated had a significant effect on THg concentrations or pools in the mineral soil. Neither THg concentrations nor pools in the upper 10 cm of mineral soil varied by treatment ( $F = 1.50$ ,  $p = 0.268$  for THg concentrations and  $F = 1.05$ ,  $p = 0.423$  for THg pools; Figure 2). Total Hg concentrations in the forest floor did vary



**Figure 2.** Total Hg concentrations and pools in surface mineral soils and the forest floor among treatments. Treatments include blowdown-salvage logging-fire (BSF), blowdown-fire (BF), fire (F), blowdown (B), and control (C). Lower case letters correspond to statistically similar groups for forest floor samples only ( $\alpha \geq 0.05$ ). No statistically significant differences were observed in mineral soils.

significantly by treatment ( $F = 14.27$ ,  $p < 0.0002$ ) with the BSF treatment having significantly lower concentrations than all other treatments (Figure 2). No other significant differences in forest floor Hg concentrations were observed among treatments. Forest floor THg pools also varied by treatment ( $F = 19.46$ ,  $p < 0.0001$ ) with the BSF treatment having significantly smaller pools than all other treatments, and the B treatment having significantly larger pools than all other treatments (Figure 2).

In comparing our B treatment sites to controls, blowdown resulted in a mean net gain of  $0.76 \text{ mg m}^{-2}$ , or an approximate 223% increase in the forest floor THg pool. In a recent study, D'Amato et al.<sup>47</sup> investigated the restructuring of the vegetation in these same treatment plots and found in the B treatment that outside of a significant increase in downed woody debris, the mixture of tree species remained relatively unchanged, with shade-tolerant conifer species continuing to be dominant. Thus, the change in the pool was not related to restructuring of the tree community, but more likely simply the result of a large accumulation of litter resulting from the 1999 blowdown, as windthrown trees gradually shed leaves or needles over time. Since concentrations in above-ground vegetation are generally lower than in litter or soil, the newly accumulated Oi surface litter is likely to have lower THg concentrations than that deeper in the forest floor.<sup>1,5</sup> However, we did not observe a decrease in forest floor THg concentration as a result of the

blowdown. The THg concentration of these inputs either must have increased to the level currently observed in the forest floor due to decomposition<sup>54</sup> in the time between the blowdown event and our sampling 9 years later, have accumulated Hg as a result of postdepositional sorption of Hg into the litter,<sup>5</sup> or the concentrations in vegetation may already have been very similar to that in the existing forest floor.

The cumulative effect of blowdown-salvage logging-fire (BSF) was the only treatment that led to significant losses in the forest floor Hg pool (Figure 2). The more severe fire conditions observed following this succession of disturbances led to much greater consumption of forest floor matter (see Supporting Information, Figure S1), which likely released more Hg to the atmosphere. Other possible loss pathways exist, but they were likely minor. The loss of Hg through evasion between the initial blowdown event and sampling (9 years) can be constrained by using representative flux measurements from the region. From the Hg<sup>0</sup> net flux measurements compiled by Denkenberger et al.,<sup>55</sup> a representative value for forest cover in the Great Lakes region is approximately  $6.9 \mu\text{g m}^{-2} \text{ yr}^{-1}$ , which over a 9 year period would account for  $62 \mu\text{g m}^{-2}$  Hg, or 12% of the observed difference between the THg pools in the BSF and control treatments. It is also possible that Hg may have been lost from the forest floor due to increased leaching and runoff as a result of salvage logging or due to increased Hg mobility following the fire. It is unlikely that the removal of material during salvage logging would have contributed significantly to this loss, as slash would have been left behind, but it is possible that the disturbance of machinery could contribute to increased mobility of Hg in runoff. We did not measure the potential leaching and runoff fluxes, but a comparison with disturbance-related increases in Hg runoff in other boreal ecosystems suggests that the potential loss through runoff is also likely comparably low. Previous research estimated runoff losses due to clearcutting and soil mounding of  $1.2\text{--}4.3 \mu\text{g m}^{-2} \text{ yr}^{-1}$  in a small Finnish watershed.<sup>14</sup> Given the upper  $4.3 \mu\text{g m}^{-2} \text{ yr}^{-1}$  estimate from Porvari et al.<sup>14</sup> and a maximum 8 years between salvage logging operations and sampling of the forest floor in our study, this would account for  $0.034 \text{ mg m}^{-2}$ . Together, net Hg evasion between disturbance and sampling and increased mobility in runoff may have accounted for approximately  $0.096 \text{ mg m}^{-2}$  of the observed  $0.52 \text{ mg m}^{-2}$  difference between BSF and control treatments. Assuming that additional losses in the BSF treatment occurred only as a direct consequence of the subsequent fire, approximately  $0.42 \text{ mg m}^{-2}$  was likely emitted, representing a net loss of 68% of the forest floor THg pool.

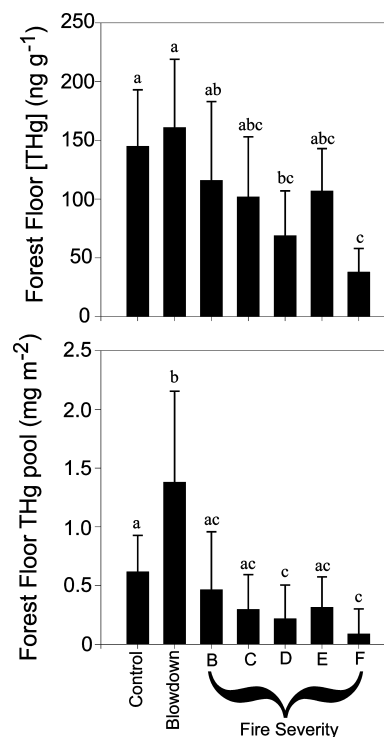
**Multiple Disturbance Impacts on Fire Severity and Hg Loss.** In a recent study by Fraver et al.<sup>48</sup> on these same treatment plots, fire intensity (heat released) was assessed by scorch-height, which was measured as the highest point of charring on tree boles, and fire severity (impact to the ecosystem) was assessed by both aboveground vegetation and forest floor assessments as per Jain and Graham.<sup>56</sup> Since our study focused on forest floor and soil Hg pools, we have focused primarily on the forest floor assessment as a measure of fire severity (see Table S1 in Supporting Information for explanation of severity indices). Fraver et al.<sup>48</sup> found that the BSF treatment had significantly lower scorch-height than either the BF or F treatments, indicating a less intense fire, but the BSF treatment experienced significantly greater fire severity as assessed through the forest floor characteristics. We note that none of the sites in this study were entirely devoid of litter,

even in the most severely burned plots. Mineral soils were black char or gray, as opposed to orange-red as would likely result from severe fire impacts.<sup>57</sup> Therefore, even in the most severely impacted sites, it does not appear that the fires following any of these series of disturbances in this study was intense enough to lead to large releases of Hg from the mineral soils. Our sampling of mineral soils as composites of the upper 10 cm would have been too coarse to observe possible effects in the surface soils, but presumably the lack of difference in THg concentrations and pools was mostly due to insulation from litter and the overall moderate severity of the wildfire. We note that the majority of wildfires are characterized by low severity burns wherein litter is only partially consumed,<sup>58,59</sup> thus while the severity of the studied fire impacts does not cover the extreme, it is likely quite representative of many wildfire impacts.

The BSF treatment likely had the least intense fire, but the severity of the fire impact on the forest floor in this study was significantly greater than all other treatments ( $p = 0.003$ ).<sup>48</sup> This was likely due to a number of impacts which may have exposed mineral soils, making them slightly, but not significantly more susceptible to the effects of fire.<sup>48</sup> The most significant of these alterations to the forest floor was likely due to harvesting equipment, which would have exposed some surface mineral soils to the fire and compacted the duff, leading to increased smoldering combustion<sup>60</sup> and hence resulted in greater Hg emission to the atmosphere. The more open salvaged sites were also likely drier, which would have led to greater forest floor consumption at a given fire intensity.<sup>61</sup> Salvage logged sites were more open than any other treatment leading to greater solar radiation reaching the forest floor. Given the positive relationship between solar radiation and Hg volatilization,<sup>9,11</sup> this condition may have further reduced Hg concentrations during the 5–8 years between salvage logging and the fire.

We further grouped our measurements by fire severity, as assessed through forest floor characteristics, and compared groups with respect to THg concentrations and pools in the forest floor. In general, we observed that forest floor THg concentrations and pools both decreased with increasing fire severity (Figure 3), but we observed significantly smaller concentrations and pools compared to the control only at severity indices D and F ( $p < 0.05$ ). There was a small increase in both THg concentration and pool size at the E fire severity index, but we attribute this to a very small and likely unrepresentative sample size ( $N = 3$  whereas  $N = 10$ – $66$  for all other indices). Given that approximately 70% of the forest floor severity assessments in the BSF treatment were at levels of D or greater and that >80% of the assessments in the BF and F treatments were at levels of C or less,<sup>48</sup> salvage logging contributes to a more severe burn at the forest floor level, leading to greater losses of forest floor mass, including Hg to the atmosphere. This is further supported by the tight relationship observed between forest floor Hg and C pools (Supporting Information, Figure S1), however we caution that this relationship is more strongly driven by forest floor mass than Hg or C concentration.

In general, we did not observe statistically significant differences among the Hg–C concentration relationships across treatments in either the mineral soils or the forest floor (Figure 4). However, large differences were observed in the Hg–C relationships within mineral soils versus the forest floor. The Hg–C relationship in mineral soils was strong ( $r^2 =$



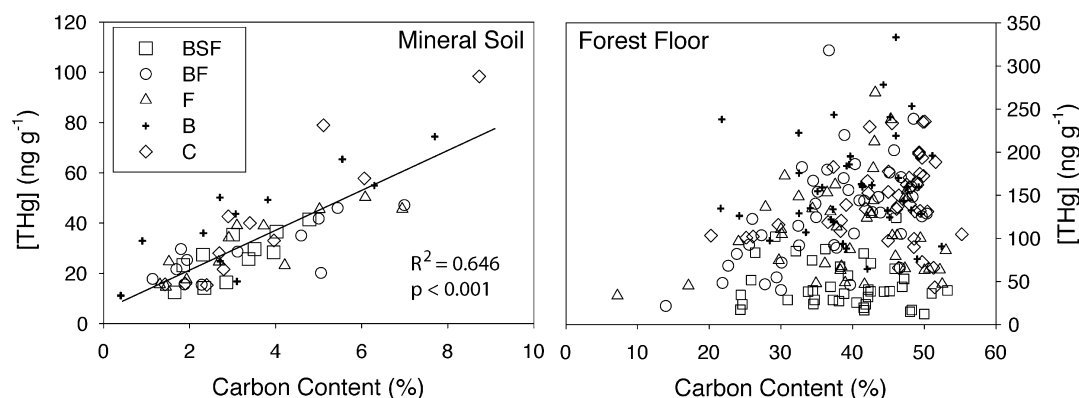
**Figure 3.** Total Hg concentrations and pools in the forest floor among nonfire treatments and increasing fire severity. Fire severity scale is explained in Table S1 of the Supporting Information. Lower-case letters correspond to statistically similar groups ( $\alpha \geq 0.05$ ).

0.646;  $p < 0.001$ ), as has been observed in several previous studies<sup>1,5,62</sup> and likely stems from the strong association between reduced sulfur and other functional groups on organic matter with Hg.<sup>6</sup> The slopes of the different treatments did not differ significantly (ANCOVA;  $p = 0.17$ ). Across all treatments, the slope of the Hg–C relationship was approximately  $0.8 \mu\text{g Hg g}^{-1} \text{C}$ . Given a C to soil organic matter mass ratio of 0.5, this is approximately equal to the ratio observed by Nater and Grigal<sup>4</sup> across the north-central United States. The treatments including fire, particularly the BSF treatment, reduced the Hg concentration range in the mineral soil C–Hg relationship compared to the B treatment and control, but did not have a significant effect on the slope of the linear relationship (Figure 4). Our results thus suggest that, at least for wildfires of moderate intensity or less, the composition of mineral soils, including the sorption of Hg onto organic molecules, is not significantly altered, regardless of the disturbance history of the forest.

In contrast to the strong Hg–C relationships observed in our mineral soil samples, no such relationship existed within the forest floor (Figure 4). Very weak, but significant relationships were observed within the individual BF ( $r^2 = 0.19$ ;  $p = 0.002$ ) and F ( $r^2 = 0.11$ ;  $p = 0.02$ ) treatments. Otherwise, no significant relationship existed between C and Hg within the forest floor, suggesting as have others, that localized heterogeneity resulting from variations in the relative stage of decomposition, degree of burn, and Hg sorption capability of remaining charred surface materials varies substantially within the forest floor.<sup>62,63</sup>

**Implications.** We see important implications from this research, especially considering that blowdown is among the major disturbance agents in US Lake States forests,<sup>34–36</sup> possibly becoming even more prevalent under a changing





**Figure 4.** Relationship between THg concentrations and carbon content in mineral soil and forest floor samples. Individual symbols relate to treatment (BSF = Blowdown-Salvage logged-Fire; BF = Blowdown-Fire; F = Fire; B = Blowdown; C = Control).

climate.<sup>42,64</sup> Further, postdisturbance salvage logging is currently among the more contentious forest-management issues in the U.S. and worldwide.<sup>46,65–67</sup> Understanding how these individual forest disturbances affect the size of Hg pools in forest soils, which tend to be the largest Hg pools in forest ecosystems,<sup>1</sup> is important; however, of greater interest is the extent to which these disturbances interact to affect Hg pools in ways that may not be predicted based on knowledge of individual disturbances alone. In this regard, our study contributes to the nascent body of literature addressing the interaction of multiple disturbances.<sup>49</sup>

Overall, this study demonstrates the importance of considering both singular and multiple forest disturbances occurring in succession, on Hg emission from, or accumulation in, the forest floor. The singular effect of blowdown increased forest floor THg pools and may lead to Hg bioaccumulation in aquatic biota of affected watersheds, as this pool appears to be a strong contributor of THg (and possibly methylmercury) in runoff to surface waters.<sup>68</sup> Only in watersheds where multiple disturbances occurred prior to fire did we observe substantial Hg losses. Release of forest floor and soil Hg to the atmosphere during wildfire may therefore not always be significant given fire alone. Fire intensity and severity are important to this release and should be considered in relation to combined disturbances. For example, in this study, disturbances from salvage logging operations were more important from a Hg management perspective than fire alone and enhanced both forest floor fire impacts and the likelihood that wildfire following salvage logging would release significantly greater amounts of Hg from the forest floor in particular. Comparison to other studies suggests that the vast majority of this loss is to the atmosphere, leading to both long-range transport and localized increases in atmospheric mercury and deposition in other watersheds.<sup>31</sup> Although it is likely that these disturbances increase the hydrologic mobility of Hg from soils, this flux is most likely small (10–20%) compared to fluxes to the atmosphere. Because the combination of blowdown, salvage logging, and wildfire dramatically decreased both THg concentrations and pools, it may take decades, perhaps centuries, for Hg to accumulate to predisturbance levels,<sup>30</sup> which could decrease Hg levels in nearby aquatic biota over this time period. This possibility would also depend on short-term disturbance-related impacts on local Hg mobility from soils. Ultimately, the fate of the Hg released to the atmosphere is difficult to track; it is eventually deposited back to the surface, possibly leading to increased bioaccumulation elsewhere.

## ■ ASSOCIATED CONTENT

### § Supporting Information

Results of linear regression analysis of Hg pools to C pools and overall forest floor dry mass per treatment (Figure S1), and key to forest floor fire severity index (Table S1). This material is available free of charge via the Internet at <http://pubs.acs.org>.

## ■ AUTHOR INFORMATION

### Corresponding Author

\*Phone: 416-208-2744; fax: 416-287-7279; e-mail: [carl.mitchell@utoronto.ca](mailto:carl.mitchell@utoronto.ca).

### Notes

The authors declare no competing financial interest.

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