

Mercury distribution in two Sierran forest and one desert sagebrush steppe ecosystems and the effects of fire

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Abstract

Mercury (Hg) concentration, reservoir mass, and Hg reservoir size were determined for vegetation components, litter, and mineral soil for two Sierran forest sites and one desert sagebrush steppe site. Mercury was found to be held primarily in the mineral soil (maximum depth of 60 to 100 cm), which contained more than 90% of the total ecosystem reservoir. However, Hg in foliage, bark, and litter plays a more dominant role in Hg cycling than the mineral soil. Mercury partitioning into ecosystem components at the Sierran forest sites was similar to that observed for other US forest sites. Vegetation and litter Hg reservoirs were significantly smaller in the sagebrush steppe system because of lower biomass. Data collected from these ecosystems after wildfire and prescribed burns showed a significant decrease in the Hg pool from certain reservoirs. No loss from mineral soil was observed for the study areas but data from fire severity points suggested that Hg in the upper few millimeters of surface soil may be volatilized due to exposure to elevated temperatures. Comparison of data from burned and unburned plots suggested that the only significant source of atmospheric Hg from the prescribed burn was combustion of litter. Differences in unburned versus burned Hg reservoirs at the forest wildfire site demonstrated that drastic reduction in the litter and above ground live biomass Hg reservoirs after burning had occurred. Sagebrush and litter were absent in the burned plots after a wildfire suggesting that both reservoirs were released during the fire. Mercury emissions due to fire from the forest prescribed burn, forest wildfire, and sagebrush steppe wildfire sites were roughly estimated at 2.0 to 5.1, 2.2 to 4.9, and $0.36 \pm 0.13 \text{ g ha}^{-1}$, respectively, with litter and vegetation being the most important sources. © 2005 Elsevier B.V. All rights reserved.

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1. Introduction

Although mineral soil is the largest mercury (Hg) reservoir in most ecosystems, it is generally less reactive than smaller pools such as vegetation and litter (Krabbenhoft et al., 2005). For example, foliage exhibits a dynamic bi-directional Hg flux and the atmosphere is

considered the dominant source of Hg in leaves with uptake from soils being a minor component (Hanson et al., 1995; Rea et al., 2002; Erickson et al., 2003; Frescholtz et al., 2003; Erickson and Gustin, 2004). Tree bole (trunk) bark and branches are also possible receptors of Hg via dry deposition (Munthe et al., 1995) with some soil contribution (Erickson et al., 2003). Litterfall is an important source of Hg to forest floors (Johnson and Lindberg, 1995). The litter itself interacts directly with the atmosphere and underlying mineral soil (Schwesig and Matzner, 2001; Grigal,

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2003). These processes involving vegetation and litter are significant in the cycling of Hg in natural vegetated systems and demonstrate the importance of these smaller ecosystem reservoirs. Despite the significance of these processes and interactions involving the cycling of Hg there are few studies characterizing the relative Hg concentrations and distribution in vegetation, litter, and mineral soil at an ecosystem level (e.g., Grigal et al., 2000; Grigal, 2003).

One potential mechanism that redistributes and interrupts the cycling of Hg in ecosystems is fire. Recent studies have demonstrated that fires are an important natural source of atmospheric Hg (Brunke et al., 2001; Friedli et al., 2003a,b; Sigler et al., 2003). For example, Friedli et al. (2003b) estimated an annual release of Hg to the atmosphere from fires in temperate forests in the continental United States of 3.7×10^6 g yr⁻¹. These relatively large Hg losses result from the low volatilization temperatures of mercury species. For example pyrolytic analysis of soil samples by Biester and Scholz (1997) demonstrated that most Hg species are volatilized between 100–300 °C. Mercury released from fires is predominantly in the elemental state (Hg⁰) (Friedli et al., 2001), which can be readily transported significant distances from the original source (Jaffe et al., 2005).

Most previous studies of Hg released from fires estimated emissions using Hg and CO measurements in smoke plumes downwind of burning (Brunke et al., 2001; Friedli et al., 2003a,b; Sigler et al., 2003). This method has the advantage of determining emissions from a large area; however, the relative contribution from burned vegetation, litter, and mineral soil cannot be directly determined.

This study quantified and compared the Hg reservoirs in two different ecosystem types and examined potential loss due to prescribed burns and wildfire. Given that Hg in the mineral soil is the largest pool of Hg in most ecosystems (e.g., Krabbenhoft et al., 2005) we hypothesized that it would be the major contributor to Hg emissions during a fire despite its generally unreactive nature.

2. Methods

Study areas were established in two Sierran forest ecosystems and one sagebrush steppe ecosystem. The sites were selected to compare the effects of prescribed burns versus wildfires and to examine differences in Hg distribution based on variations in vegetation and climate. The forest prescribed burn site is at an elevation of 1767 m in the Sierra Nevada Mountains in northern

California, near the town of Truckee. This site receives an average of 940 mm of annual precipitation. Overstory vegetation is dominated by Jeffrey pine (*Pinus jeffreyi*) with a few white fir (*Abies concolor*). Understory vegetation was a negligible component of the biomass and was not considered in biomass, Hg concentration, or Hg pool estimates. Soil at this site is Kyburz series, fine-loamy, mixed frigid Ultic Haploxeralf [alfisol] derived from an underlying andesite flow (Rogers, 1974). The A horizon contains moderate clay content (10–18%) and organic matter with increasing clay (20–35%) and decreasing organic matter in deeper horizons. A prescribed fire was set in the spring of 2002. The fire was patchy in distribution burning ~50% of the forest floor and primarily burning areas with greater litter mass. Although the heat scorched some tree foliage, none of the tree branches, bole bark, or bole wood burned significantly in the fire. In the summer of 2003, roughly 1 year after the fire, five 0.08 ha plots were established and sampled in both burned and adjacent unburned areas.

The forest wildfire site is located in the southeast portion of the Lake Tahoe Basin in western Nevada on a hill slope at elevations ranging from 1950 to 2100 m. The area receives an average of 870 mm of annual precipitation. Similar to the forest prescribed burn site, overstory vegetation is dominated by Jeffrey pine (*P. jeffreyi*), white fir (*A. concolor*), and a few scattered sugar pine (*Pinus lambertiana*) and incense cedars (*Calocedrus decurrens*). Soils at this site are the Cagwin Series, coarse, loamy sand, mixed Typic Cryosamments [entisol] derived from granodiorite (Rogers, 1974). The upper three soil horizons (A1, A2, and AC) consist of loamy coarse sand with increasing roots and gravel (up to ~10%) with depth. The deepest horizon (C) is dominated by coarse sand and grus. A primary difference between the two Sierran forest sites is the mineral soil parent material (andesite vs. granodiorite). Ten 0.08 ha plots were established in the fall of 2001 and were sampled and inventoried in the spring of 2002 (preburn data). In July of 2002, the 283 ha Gondola wildfire burned seven of the plots. At those plots, fire burned nearly all of the litter mass (~87% mass loss) and trees (~39% reduction in total tree mass). Before postburn samples could be collected, runoff from a large rainstorm eroded a significant mass of soil and ash away from the burned plots down slope. In July of 2003 (approximately one year after the fire), plots (approximately 1 m away from preburn sampling points) and material that had been transported during the erosion event were sampled and inventoried. Preburn and postburn samples were also collected from the three unburned plots, which were used as control plots.

The desert wildfire site is located in a sagebrush steppe area in the Basin and Range physiographic province in north-central Nevada, approximately 15 km northwest of the town of Battle Mountain. The semiarid site is located at an elevation of 1414 m and has an annual precipitation of 200 mm. An ~11,700 ha wildfire swept through the site in August of 1999 burning all of the sagebrush and litter. Five random 0.01 ha burned and five adjacent unburned plots were established and sampled in December of 2004, roughly 5 years after the fire. Vegetation is dominated by Wyoming big sagebrush (*Artemisia tridentata* var. *wyomingensis*) in unburned areas, and tumble mustard (*Sysimbrium altissimum*), cheatgrass (*Bromus tectorum* L), Russian thistle (*Salsola kali*), and Fescue (*Vulpia spp.*) in burned areas. Soils at the desert wildfire site are Enko fine sandy loam, coarse-loamy mixed, mesic Durixerollic [molli-sol] Camborthids derived from a stable alluvial fan. The A horizon consisted primarily of fine loamy sand with a significant platy structure with decreasing structure and organic matter and increasing concretion and gravel content down into the deeper horizons.

Mercury reservoir analysis and mass balance were determined using methods which have been successfully used for Hg and other elements (cf. Harden et al., 2004; Caldwell et al., 2002). At the two forest sites (forest prescribed burn and forest wildfire) diameter at breast height (dbh) was measured for all trees in the 0.08 ha plots. Dry tree mass for each component (foliage, branches, bole wood, and bole bark) was estimated using regressions between dbh and biomass from each plot (Johnson et al., 1997). Samples were collected from each vegetation component of trees in each plot and homogenized by plot for Hg analysis. Clean hands procedures were used for sample collection (Rasmussen, 1995). Previous studies have shown that the variability of Hg concentrations in the tree component is small ($CV < 15\%$) suggesting that bulking methods are appropriate (Rasmussen et al., 1991; Frescholtz et al., 2003). At the desert wildfire site, basal diameters were measured for all sagebrush within the unburned plots (0.01 ha); no sagebrush were present in the burned plots. Foliage and woody material samples for Hg analysis were collected using clean gloves from 3 random sagebrush in each unburned plot and placed in plastic bags. Vegetation mass for the unburned plots at the desert wildfire site were estimated based upon a site-specific regression between sagebrush basal stem diameter and dry foliage mass ($R^2 = 0.71$, $n = 11$, $p < 0.05$) and dry wood mass ($R^2 = 0.96$, $n = 11$, $p < 0.05$).

Litter (defined as the entire O-horizon) and ash were collected from five or six points in each plot (5 replicates

per plot at forest wildfire and desert wildfire sites; 6 replicates per plot at forest prescribed burn site) within a 15 cm diameter litter ring and homogenized. Samples were collected using clean gloves and placed into plastic or paper bags. A subsample of the litter was removed for Hg analysis while the remainder was dried and weighed to estimate mass per area. After litter and/or ash was removed, the soil was sampled at depths representing major soil horizons for each plot (see Table 1 for soil horizon depths). Because of the relatively thick B horizon at the desert wildfire site (~40 cm), samples for Hg concentrations were collected from both the upper and lower 20 cm thicknesses and averaged. Bulk density was estimated using the core method for one point in each plot at the top of each soil horizon. The mass of coarse (> 2 mm) and fine (< 2 mm) soil particles was measured for each horizon using a spring scale. Fine soil particle mass was calculated for each horizon and plot and were used as soil reservoir mass in this study.

After collection all samples were stored in a $-15\text{ }^{\circ}\text{C}$ freezer to prevent volatilization of Hg. Vegetation and soil samples were bulked by plot in the field or prior to sample preparation. Each litter and ash replicate sample was analyzed separately. Vegetation and litter samples were frozen with liquid nitrogen, homogenized using a mortar and pestle, and freeze dried for > 48 h using a Sentry™ Vitris Benchtop 3L (Ericksen et al., 2003). Samples were lyophilized then analyzed for total Hg using thermal decomposition, amalgamation, and atomic absorption spectrometry on a Milestone™ Direct Mercury Analyzer (EPA Method 7473). National Institute of Standards and Technology (NIST) reference materials (San Joaquin soil #2709, Peach leaves #1547, and Apple leaves #1515) were used for quality assurance at least once every 10 samples, and error on soils was less than 10% and less than 5% on plants. Triplicate analyses showed minimal sample variance ($< 5\%$ coefficient of variance for vegetation samples; $< 10\%$ coefficient of variance for litter and soil samples; $n = 52$).

Mercury reservoir size was calculated by applying mean Hg concentrations to mass estimates for each component and plot. Statistical differences were determined using nonparametric Mann–Whitney U (unpaired data) and Wilcoxon sign-rank (paired data) tests and denoted as significantly different for $p < 0.05$. All statistics were run using Intercooled Stata® 8.

Raison et al. (1985) demonstrated that ratios of Ca:C in burned materials can be used as a surrogate for fire severity (a function of both temperature and duration) because C is volatilized at relatively low temperatures (200 to 400 $^{\circ}\text{C}$; Hosking, 1938) while Ca is unlikely to

Table 1
Mass estimates (dry) for vegetation, litter and soil reservoirs

	Unburned data	Burned data	Unburned– Burned
	Avg. (Mg ha ⁻¹) ±S.D.	Avg. (Mg ha ⁻¹) ±S.D.	Avg. (Mg ha ⁻¹) ±S.D.
<i>a) Forest prescribed burn site</i>			
<i>Vegetation</i>			
Foliage	6.7±2.6	5.5±1.9	–
Branches	20±8.0	17±6.7	–
Wood	64±37	9.6±3.3	–
Bark	12±6.6	50±19	–
Total tree	100±55	83±31	–
<i>Litter</i>			
Unburned litter	45±23	8.4±4.8	–
Burned litter	–	30±9.5 ^a	–
Total Litter	45±23	18±3.9	27±23
<i>Mineral soil</i>			
A Horizon (0–20 cm)	1200±460	1300±270	–
B Horizon (20–40 cm)	1200±120	1200±190	–
C Horizon (40–60 cm)	1100±130	1100±360	–
Soil total ^b	3400±620	3600±300	–
<i>b) Forest wildfire site</i>			
<i>Vegetation</i>			
Foliage	8.5±3.2	1.4±1.5	3.0±5.0
Scorched foliage	NA	4.2±2.8	NA
Branches	28±10	22±8.2	6.3±4.7
Wood	110±50	82±42	31±24
Bark	28±12	20±10	7.7±5.9
Total Tree	180±74	110±52	70±36
<i>Litter</i>	29±13	3.8±6.9 ^a	25±11
<i>Mineral soil</i>			
A1 Horizon (0–10 cm)	1100±310	–	–
A2 Horizon (10–30 cm)	1500±350	–	–
AC Horizon (30–60 cm)	2900±550	–	–
C Horizon (60–100 cm)	2 5 0 0 ±2300	–	–
Soil total ^c	8 1 0 0 ±2500	–	–
<i>c) Desert wildfire site</i>			
<i>Vegetation</i>			
Foliage	1.2±0.4	–	1.2±0.4
Wood	15±5.0	–	15±5.0
Sagebrush total	16±5.4	–	16±5.4
<i>Litter</i>	2.4±0.4	2.0±0.9 ^d	2.4±0.4
<i>Mineral soil</i>			
A Horizon (0–~4 cm)	640±640	380±130	

Table 1 (continued)

	Unburned data	Burned data	Unburned– Burned
	Avg. (Mg ha ⁻¹) ±S.D.	Avg. (Mg ha ⁻¹) ±S.D.	Avg. (Mg ha ⁻¹) ±S.D.
<i>Mineral soil</i>			
B Horizon (~4–~45 cm)	5400±1400	5700±940	
C Horizon (~45–80 cm)	4000±1000	4600±1100	
Soil total ^e	10,000±650	11,000±380	

^a Primarily ash.

^b To a depth of 60 cm.

^c To a depth of 100 cm.

^d Includes both litter and vegetation.

^e To a depth of 80 cm.

be volatilized (volatilization temperature=1484 °C; Raison et al., 1985). To assess the loss of Hg from mineral soils resulting from variations in fire severity using ratios of Ca:C as a surrogate, soil samples (0–5 cm depth) were collected at the forest prescribed burn ($n=15$) and forest wildfire ($n=11$) sites to provide a large range in apparent fire severity. The soil samples were sieved to <2 mm. Sample aliquots were analyzed for total Hg, total C (dry combustion C analyzer at the Oklahoma State University Soil, Water, and Forage Analytical Laboratory), and Ca (inductively coupled plasma atomic emissions spectrometry at A and L Western Agricultural Laboratories in Modesto, California).

3. Results and discussion

Data collected from unburned or preburn plots are herein termed unburned data, while those from burned or postburn plots are referred to as burned data. Mass estimates (all reported masses are dry) of unburned ecosystem components are provided in Table 1. Significantly higher above ground vegetation and litter masses were measured at the two forest sites (forest prescribed burn and forest wildfire) relative to the desert wildfire site. Vegetation mass was 2 to 7 times greater than litter at all three sites. Tree component masses at the forest prescribed burn and forest wildfire sites were dominated by bole wood, followed by bark and branches, which showed similar masses, and by foliage. At the desert wildfire site, the woody material in sagebrush exceeded foliage mass by roughly 10 times.

Mercury concentration data for the three sites are provided in Table 2. Unburned data show that the smaller mass reservoirs including litter from all three

Table 2

Average mercury concentration and standard deviation of environmental components

	Unburned data		Burned data		Burned?
	Hg conc. (ng g ⁻¹)±S.D.	<i>n</i>	Hg conc. (ng g ⁻¹)±S.D.	<i>n</i>	
<i>a) Forest prescribed burn site</i>					
<i>Vegetation</i>					
Foliage	19.7±10.5	5	32.2±23.1	5	Yes
Branches	18.5±7.8	5	—	5	No
Bole wood	2.9±0.4	5	—	5	No
Bole bark	12.5±3.6	5	—	5	No
Litter	81.3±20.3	5	24.5±4.7	4	No
Litter (burned)	NA		24.7±4.4 ^a	4	Yes
<i>Mineral soil</i>					
A Horizon (0–20 cm)	22.1±1.7	4	24.6±2.5	4	Yes ?
B Horizon (20–40 cm)	18.5±1.6	4	19.7±1.1	4	No
C Horizon (40–60 cm)	20.0±3.2	4	17.4±4.4	4	No
<i>b) Forest wildfire site</i>					
<i>Vegetation</i>					
Foliage	63.6±18.9	7	30.9±5.5	7	No
Scorched foliage	NA	7	34.0±4.5	7	Yes
Branches	19.4±2.2	7	17.1±8.0	7	Yes
Bole wood	2.3±0.2	7	3.6±0.7	7	No
Bole bark	15.2±2.7	7	10.0±2.8	7	Yes
Litter	91.4±32.2	7	39.2±8.3 ^a	7	Yes
<i>Mineral soil</i>					
A1 Horizon (0–10 cm)	8.8±3.1	7	10.1±8.4	7	Yes ?
A2 Horizon (10–30 cm)	7.6±4.9	7	5.5±5.4	7	No
AC Horizon (30–60 cm)	4.9±2.7	7	4.2±1.0	7	No
C Horizon (60–100 cm)	3.4±1.1	7	3.7±1.7	7	No
<i>c) Desert wildfire site</i>					
<i>Vegetation</i>					
Foliage	54.5±6.7	5.0	—	5	Yes
Wood	21.9±6.1	5.0	—	5	Yes
Litter	95.8±10.9	5.0	49.2±7.3 ^b	5	No
<i>Mineral soil</i>					
A Horizon (0~4 cm)	47.4±26.8	5	49.4±11.3	5	Yes?
B Upper (4~25 cm)	16.7±7.5	5	18.0±9.4	5	No
B Lower (~25–45 cm)	11.2±9.5	5	11.2±5.8	5	No
C Horizo (~45~80 cm)	7.1±2.3	5	8.4±3.8	5	No

Burned columns indicate whether or not samples collected in the burned plots were burned. *n*=number of plots.

^a Primarily ash.

^b Includes both litter and vegetation.

sites (81.3 ± 20.3 to 95.8 ± 10.9 ng g⁻¹) and foliage at the forest wildfire and desert wildfire sites (54.5 ± 6.7 to 63.6 ± 18.9 ng g⁻¹) had the highest Hg concentrations. Similar distributions have been observed by previous workers (Rasmussen et al., 1991; Frescholtz et al., 2003; Grigal, 2003). Vegetation from the unburned samples showed a general decrease in Hg concentrations from foliage to branches, to bark, to wood (Table 2). Mercury in foliage has been demonstrated to result primarily from the accumulation of atmospheric Hg with soils exerting a minor influence (Hanson et al., 1995; Rea et al., 2002; Ericksen et al., 2003; Frescholtz et al., 2003; Ericksen and Gustin, 2004). A substantial difference in Hg concentrations in foliage between the two forests sites (forest prescribed burn = 19.7 ± 10.5 ng g⁻¹, forest wildfire = 63.6 ± 18.9 ng g⁻¹) was observed. These differences may be a result of variations in air Hg concentration and/or age of the foliage (Hanson et al., 1995; Rea et al., 2002). Litter Hg concentrations in unburned samples were similar between all sites (81.3 ± 20.3 to 95.8 ± 10.9 ng g⁻¹). This is surprising given the different vegetation concentrations, litter masses, and vegetation types between the desert wildfire site and the two forest sites (Tables 1 and 2).

Table 3 and Fig. 1 show the Hg reservoirs at the three sites. In general, the unburned litter Hg reservoir was larger than the vegetation Hg reservoir at all three sites, although the differences were not statistically significantly different (Table 2). Results from the two forest sites (Table 3a,b) compare well with calculations from Grigal (2003) who estimated the average Hg reservoirs in temperate forests of the contiguous United States (vegetation = 0.8 g ha⁻¹, litter = 5.5 g ha⁻¹, mineral soil to 100 cm depth = 117 g ha⁻¹). The Hg vegetation pool at the forest wildfire site was roughly double that at the forest prescribed burn area. This is primarily a result of higher tree biomass at the forest wildfire site (Table 1). Ash and charcoal were observed in litter at unburned plots at the forest prescribed burn site suggesting previous fires at the site may have limited tree growth. At the forest prescribed burn site the average distribution of Hg in the unburned vegetation was roughly 44% in the branches, 20% in the bole wood, 19% in the bole bark, and 18% in the foliage (Table 3a). The average distribution of Hg in samples of unburned vegetation from the forest wildfire site was similar (30% branches, 26% bole bark, 24% bole wood, and 20% foliage; Table 3b). In samples from the unburned plots at the desert wildfire site, vegetation (sagebrush) and litter reservoirs were a relatively small portion of the total ecosystem relative to the forest sites (0.12% and 0.15%, respectively; Table 3c). In these small above ground reservoirs,

the majority of the Hg was held in the foliage (~80% on average) relative to the wood.

Mineral soil was by far the largest reservoir of Hg at all three sites, accounting for >90% of the total ecosystem Hg reservoir (Fig. 1). This dominance of the

Table 3
Average mercury content and standard deviation of environmental components

	Unburned plots		Burned plots		Significantly different?
	Mean (g Hg ha ⁻¹) ±S. D.	n	Mean (g Hg ha ⁻¹) ±S. D.	n	
<i>a) Forest prescribed burn site</i>					
<i>Vegetation</i>					
Foliage	0.15±0.07	5	0.17±0.09	5	No
Branches	0.37±0.17	5	0.32±0.12	5	No
Bole wood	0.17±0.10	5	0.13±0.05	5	No
Bole bark	0.16±0.08	5	0.12±0.04	5	No
Tree total	0.84±0.39	5	0.74±0.24	5	No
Litter	4.0±2.6	5	0.4±0.1	4	Yes
<i>Mineral soil</i>					
A Horizon (0–20 cm)	26.5±12.1	4	32.0±8.6	4	No
B Horizon (20–40 cm)	21.2±1.9	4	22.7±2.5	4	No
C Horizon (40–60 cm)	21.2±2.7	4	15.5±5.5	2	No
Min. soil total	68.9±10.4	4	70.3±7.2		No
Ecosystem total ^a	73.8±10.2		71.5±13.7		No
	Preburn		Postburn		Significantly different?
	Mean (g Hg ha ⁻¹) ±S. D.	n	Mean (g Hg ha ⁻¹) ±S. D.	n	
<i>b) Forest wildfire site</i>					
<i>Vegetation</i>					
Foliage	0.33±0.18	7	0.19±0.11	7	No
Branches	0.49±0.18	7	0.39±0.19	7	No
Bole wood	0.39±0.16	7	0.27±0.10	7	Yes
Bole bark	0.42±0.19	7	0.20±0.12	7	Yes
Tree total	1.63±0.48	7	1.05±0.43	7	Yes
Litter	2.77±1.75	7	0.16±0.23	7	Yes
<i>Mineral soil</i>					
A1 Horizon (0–10 cm)	9.8±4.3	7	10.5±8.0	7	No
A2 Horizon (10–30 cm)	11.3±5.7	7	9.6±12.3	7	No
AC Horizon (30–60 cm)	13.9±7.4	7	12.2±3.3	7	No
C Horizon (60–100 cm)	7.7±7.3	7	9.2±11.2	7	No
Min. soil total	42.7±12.9	7	41.5±21.4	7	No
Ecosystem total ^b	47.1±12.3		42.7±21.8		No

Table 3 (continued)

	Unburned plots		Burned plots		Significantly different?
	Mean (g Hg ha ⁻¹) ±S. D.	n	Mean (g Hg ha ⁻¹) ±S. D.	n	
<i>c) Desert wildfire site</i>					
<i>Vegetation</i>					
Foliage	0.13±0.05	5	–	5	Yes
Wood	0.03±0.01	5	–	5	Yes
Veg. total	0.16±0.06	5	–	5	Yes
Litter	0.20±0.09	5	0.11±0.01 ^c	5	Yes
<i>Mineral soil</i>					
A Horizon (0–4 cm)	24.6±15.1	5	19.6±9.7	5	No
B Horizon (4–45 cm)	77.5±35.7	5	84.2±42.0	5	No
C Horizon (45–80 cm)	29.1±14.3	5	36.5±12.2	5	No
Soil total	131.2±19.1	5	140.3±39.2	5	No
Ecosystem total ^d	131.5±19.1	5	140.3±39.2		No

Each plot consisted of 5 or 6 replicates. Statistical significance considered at $p < 0.05$. n = number of plots.

^a To a depth of 60 cm.

^b To a depth of 100 cm.

^c Includes both vegetation and soil.

^d To a depth of 80 cm.

mineral soil has been previously documented (e.g., Grigal, 2003; Krabbenhoft et al., 2005). Mineral soil Hg concentrations exhibited some variation with depth (Table 2, Fig. 2). The desert wildfire site soil showed a statistically significant decrease in mean Hg concentrations with soil depth (Fig. 2c). There is a similar trend at the forest wildfire site but it is not statistically significantly different. Changes in soil Hg concentrations are typically attributed to variable concentrations of organic matter (Grigal, 2003); Hg has a strong affinity for organic matter (Schuster, 1991). The highest soil Hg concentrations were observed in unburned samples of the A-horizon (Fig. 2c) at the desert wildfire site. Fig. 2b demonstrates that Hg concentrations in unburned mineral soils from the forest wildfire site are significantly lower than for other sites, especially at depth, but the mineral soil was still the largest reservoir for Hg at the site (~91% for unburned data). The lower concentrations in the deeper soil horizons are likely a result of low Hg concentrations in the source rock (granodiorite); deeper soil horizons generally contain element concentrations similar to the source rock (Kauranne, 1992). All soil Hg concentrations were below the threshold for un-enriched soils (<100 ng g⁻¹; Connor and Shacklette, 1975).

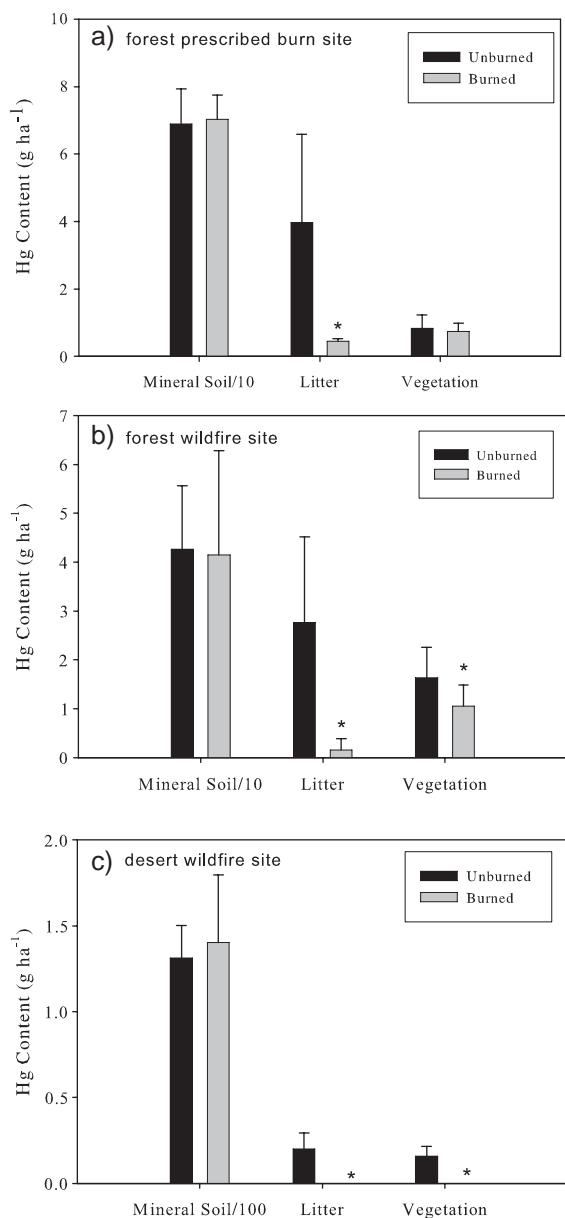


Fig. 1. Average and standard deviation of Hg content for vegetation, litter, and mineral soil at the three study sites. * symbol indicates the burned data Hg content was significantly smaller ($p < 0.05$) than unburned content.

The contribution of individual soil horizons to the total soil reservoir was roughly equal for the two forest sites (Table 3a,b). A large variation in contribution between the A (19%), B (60%), and C (21%) horizons relative to the entire mineral soil pool was observed in samples from the desert wildfire site (Table 3c) and is a result of large variations in Hg concentrations (7.1 ± 2.3 to 47.4 ± 26.8 ng g⁻¹; Table 2c) and horizon thicknesses (~4 to ~40 cm).

Comparison of mass, Hg concentration, and Hg reservoir sizes in unburned and burned data provides insight on the effect of fire on Hg distribution and cycling in these systems. Fire has several direct and indirect mechanisms for mobilizing elements found in ecosystems. Direct mechanisms include conversion to gaseous forms and subsequent emission, vaporization of volatile compounds, and release as/on ash particles (Binkley and Christensen, 1991). Indirect mechanisms include conversion to more soluble forms that can be leached into the soil, removal of stabilizing vegetation, and development of a hydrophobic layer allowing for transport of element-bound particles (Gimeno-García et al., 2001).

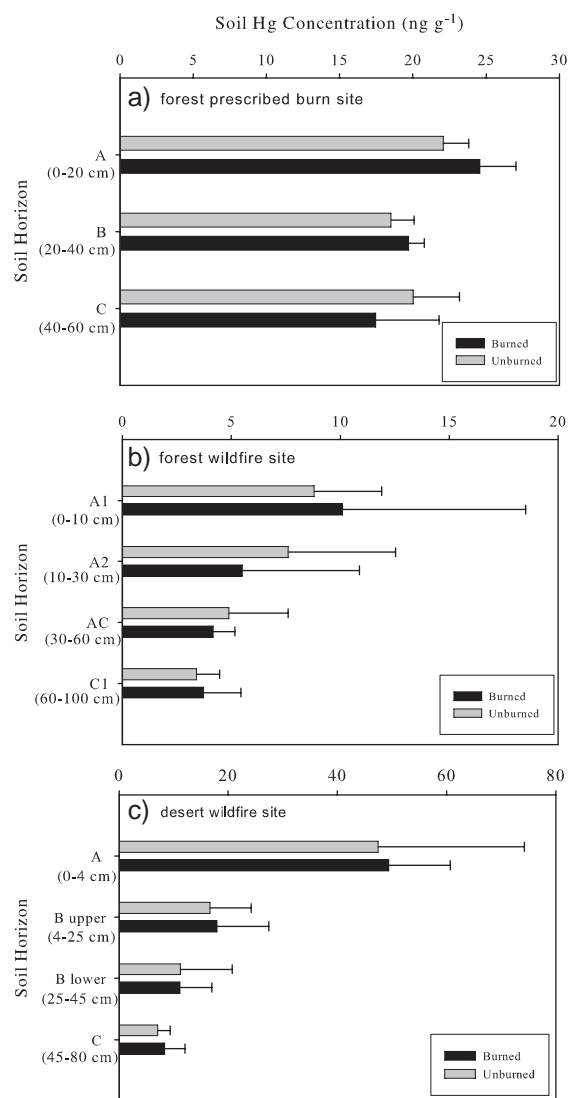


Fig. 2. Average and standard deviation of Hg concentration by soil horizon.

Results from Mann–Whitney *U*-tests comparing Hg reservoir size between unburned and burned plots at the forest prescribed burn site indicated a statistically significant decrease in the litter Hg reservoir even though burned samples were collected one year after the fire (Table 3a). No significant differences were noted in the vegetation at this site, which was not notably burned in the fire, or for the mineral soil reservoir. Comparison of litter mass at the forest prescribed burn showed 60% less mass at the burned plots on average than the unburned plots (Table 1a) and 70% lower Hg concentrations in burned samples (Table 2a). The difference in the litter Hg reservoir size between the burned and unburned plots is estimated at $3.5 \pm 2.6 \text{ g ha}^{-1}$ (88% lower) which represents a $\sim 5\%$ ecosystem Hg reservoir loss.

Unburned versus burned mass estimates at the forest wildfire site showed a large decrease in total vegetation mass ($\sim 40\%$) and litter mass ($\sim 87\%$) as a result of the fire and post-fire erosion of the soil and litter (Table 1). Burned litter was dominated by ash. Statistical comparison of Hg reservoir size at the three control sites for unburned versus burned data at the forest wildfire site (data not shown) showed no significant difference. For the plots that burned (Table 3b) statistically significant differences between unburned and burned Hg reservoirs were noted for the litter. Mercury concentrations (Table 2b) in scorched post-fire foliage ($34.0 \pm 4.5 \mu\text{g g}^{-1}$) and unscorched post-fire foliage ($30.9 \pm 5.5 \mu\text{g g}^{-1}$) were markedly smaller than for litter in unburned plots ($63.6 \pm 18.9 \mu\text{g g}^{-1}$). This may be due to unburned foliage having differences in concentration due to leaf age (Rasmussen, 1995) and/or the elevated temperatures produced by fire, which caused a loss of Hg in both scorched and unscorched post-fire foliage. The total Hg vegetation reservoir exhibited a statistically significant decrease ($0.6 \pm 0.5 \text{ g Hg ha}^{-1}$, 36% decrease) after burning even for data collected approximately one year later. A $2.6 \pm 1.7 \text{ g Hg ha}^{-1}$ difference between unburned and burned litter Hg reservoir size at the forest wildfire site was also calculated (94% decrease). This estimate includes both direct and indirect fire effects because some of the ash (litter) and soil was transported from the plots during the post-fire rainstorm.

Mercury concentrations in ash samples (i.e., burned litter) from the forest prescribed burn and forest wildfire sites were much higher ($> 20 \text{ ng g}^{-1}$; Table 2) than in ash collected immediately after a burn reported by Friedli et al. (2001) of $< 5 \text{ ng g}^{-1}$. Given the strong affinity of Hg for carbon in ash (Olson et al., 2000; Gustin and Ladwig, 2004) it is likely that these elevated ash Hg concentrations are a result of sorption of atmospheric Hg

in the ~ 1 year period between the fire and the collection of the samples during burned conditions.

Statistical analyses of data from the desert wildfire site indicated a significant difference in Hg reservoir size between the burned and unburned plots for only the vegetation and litter reservoirs which had entirely burned during the fire. Biomass losses during fires in other sagebrush systems have also been shown to be $> 90\%$ (Sapsis and Kauffman, 1991; Kauffman and Sapsis, 1989). Assuming a total loss of vegetation and litter during the fire, $0.36 \pm 0.13 \text{ g Hg ha}^{-1}$ was released ($< 1\%$ of the total Hg in the system). This value is roughly ten times smaller than differences observed for the two forest sites and is a result of the relatively small litter and vegetation mass in semiarid environments.

Data from the fire severity points for soils at forest prescribed burn site and the forest wildfire site are shown in Fig. 3. Ratios of Ca to C are plotted on the ordinate and are used as an indicator of fire severity (Raison et al., 1985); points where significant C was lost during the fire plot further to the right and those with less loss of carbon are plotted on the left. Data from both

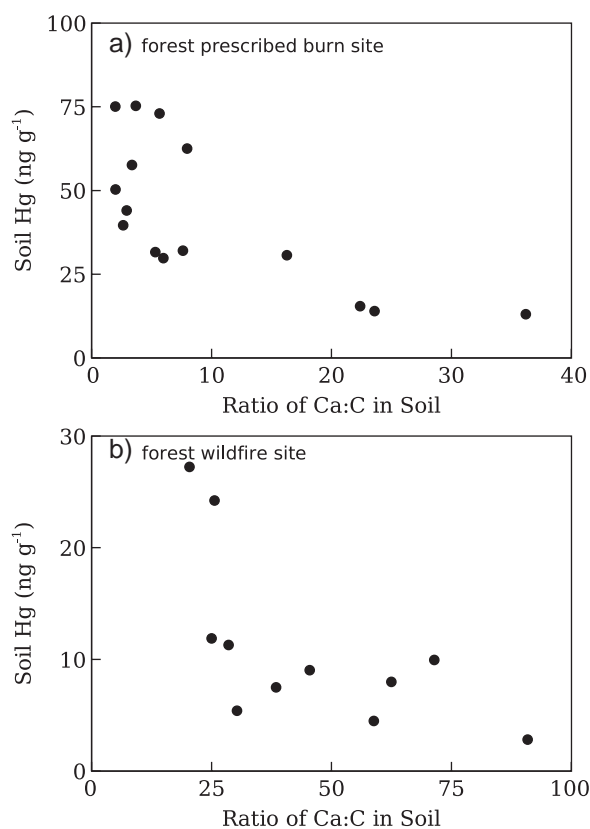


Fig. 3. Soil Hg concentration and ratios of Ca : C data from fire severity plots at the forest prescribed burn and forest wildfire sites. Ca and C are in percent units.

plots show a relatively large range in soil Hg concentrations (13 to 75 ng g⁻¹ at the forest prescribed burn site and 3 to 27 ng g⁻¹ at the forest wildfire site) from points where little or no burn occurred. With increasing burn severity (i.e., larger Ca:C ratios), Hg concentrations in the soil decreased to <30 ng g⁻¹ at the forest prescribed burn site. The data are less convincing for the forest wildfire site where there are only two points with concentrations >20 ng g⁻¹, while the rest are all below <15 ng g⁻¹, although there is a decreasing trend in the Hg concentration data at higher Ca:C ratios. Larger maximum ratios of Ca:C were observed in data from the forest wildfire site compared to the forest prescribed burn site and are likely a result of greater fire severity at the former site. These results suggest that some Hg was volatilized from soils during exposure to elevated burn intensities.

Mass balance calculations indicated that there were no significant differences in Hg reservoir sizes for mineral soils between unburned data and burned data suggesting that a negligible amount of Hg was lost from the mineral soil column despite exposure to elevated temperatures during the fire. A similar lack of emission of Hg from soils was observed from a forest fire in Minnesota (L. Woodruff, pers. comm.). This is in contrast to results from the fire severity points, which demonstrated a decrease in soil Hg concentration in the upper 5 cm at greater fire severity. Two potential reasons for this discrepancy are: 1) that the fire severity points were specifically selected to represent certain post-fire conditions and may not be similar to the randomly picked mass balance sampling points and/or 2) that the fire severity points only included the top 5 cm of the soil profile while the mass balance samples were collected through the entire A horizon (0–20 cm at the forest prescribed burn site and 0–10 cm at the forest wildfire site).

In another Sierran forest, Oakley et al. (2003) demonstrated that surface mineral soil surface temperatures can exceed 100 to 200 °C for several hours, while temperatures at a depth of 10 cm generally did not exceed 50 °C. In a *Eucalyptus pauciflora* forest, Raison et al. (1986) observed surface mineral soil temperatures exceeding 450 °C, but had decreased to ~50 °C within 2 cm depth. These findings suggest that surface temperatures at the forest prescribed burn and forest wildfire sites during fire may have exceeded temperatures necessary for emission of soil-bound Hg (>100 °C, Biester and Scholz, 1997) but temperatures at depth were likely too low to allow for emission. Soil moisture has also been shown to buffer heating of soils (Trollope, 1984) but all three fires occurred during dry periods

when soil moisture was low. Loss of Hg from the upper 1 cm of mineral soil is likely too small to be detected in bulked samples representing 0 to 10–20 cm depth, such as those used in the mass balance samples. Soil surface temperatures from a savanna fire with a similar biomass as the desert wildfire site were low (<40 °C) suggesting that Hg may not have been emitted from soils during the fire assuming similar fire severity (Jensen et al., 2001).

The mass balance data for the three study areas can be used to examine potential emissions from various fire regimes. However, Hg concentrations in litter (ash and charcoal) samples in the burned data were high (>20 ng g⁻¹) relative to ash collected immediately after burning of vegetation (<5 ng g⁻¹; Friedli et al., 2001). The samples were collected at least one year after the fire and over a year most likely accumulated Hg from the atmosphere via dry and wet deposition. To produce the entire mean postburn litter contents of 0.4 (forest prescribed burn site) and 0.16 (forest wildfire site) g Hg ha⁻¹ (see Table 3), would only require an average dry deposition rate of ~4.5 and 2 ng m⁻² h⁻¹, respectively. These values are well within the dry elemental Hg deposition rates measured for coal fly ash (Gustin and Ladwig, 2004) and measured dry deposition to soil (Engle et al., 2001). Some of the Hg could also be accounted for by inputs from precipitation although they would likely be small (<0.1 g Hg ha⁻¹ based on data from the Mercury Deposition Network; National Atmospheric Deposition Program, 2005). Therefore, the results must be used with caution due to potential enrichment of ash in Hg due to wet and dry atmospheric deposition, and the potential for soil and ash erosion such as that which occurred after the fire at the forest wildfire site. Assuming a 50% to 100% loss of the litter Hg reservoir, a 0% to 50% loss of the vegetation reservoir, and a 0% to 100% loss of Hg in the upper 5 mm of mineral soil during the forest prescribed burn fire, average emissions are estimated at 2.0 to 5.1 g ha⁻¹. At the forest wildfire site, assuming that 50% to 100% of Hg litter reservoir, 50% to 100% of the Hg vegetation reservoir, and 0% to 100% of the upper 5 mm of the mineral soil were released during fire, emissions are roughly estimated at 2.2 to 4.9 g ha⁻¹. These emission estimates for the forested ecosystems are dominated by release from litter; and at both sites mineral soil was a relatively small input (<15%). Assuming a complete removal of sagebrush and litter with no loss of Hg from the mineral soil for the desert wildfire site resulted in an emission estimate of 0.36 ± 0.13 g ha⁻¹.

A comparison of estimated Hg losses from this study and other studies is provided in Table 4. These data

Table 4
Summary of mercury emission estimates from fires

Location	Fire type	Emission estimate (g Hg ha ⁻¹)	Data source
Forest prescribed fire site in California, USA	Prescribed	2.0–5.1	This study
Forest wildfire site in Nevada, USA	Wildfire	2.2–4.9	This study
Desert sagebrush steppe in Nevada, USA	Wildfire	0.36±0.13	This study
Boreal forest in Alaska, USA	Prescribed	2±3	Harden et al., 2004
Boreal forest in Quebec, Canada	Wildfire	1.5	Sigler et al., 2003
Temperate Forest in Washington State, USA	Wildfire	2.8±1.5	Friedli et al., 2003b
Forest in Minnesota, USA	Prescribed	2	Woodruff et al., 2001

show a relatively small range in Hg emissions from different fires from 1.5 to 5.3 g ha⁻¹. Differences in these estimates between sites are likely a result of variations in fuel amount, moisture content, fire severity, and Hg concentration in the fuel. Friedli et al. (2003b) estimated an annual Hg emission of 3.6±1.9 t yr⁻¹ from temperate forests in the contiguous United States (1997–2001 average) based on data from a fire in Washington State which had losses similar to fires in other states (Table 4).

Annual emission estimates may change with time and land use. In the United States pre-settlement fires were frequent and generally low severity (McKelvey et al., 1996). With increased fire suppression and cessation of Native American fire ignitions, fuel loads have built up significantly producing much higher severity burns in forest environments with much lower frequency (Tilman et al., 2000). Because of this, emissions from many wild fires presented in Table 4 may be larger than those in pre-settlement times. Prescribed fires are currently applied by regulatory agencies to reduce fuel loads in areas of potential risk for high severity wild fires. As the application of prescribed fires begins to increase, fire frequency will likely increase and the severity will decrease. It is currently unclear how these changes will affect Hg emissions from fires and the global Hg budget.

Estimated emissions from the desert wildfire site are considerably lower than for the forest sites, reflecting the low amount of fuel at these sites. Of particular interest at the desert wildfire site is the change in vegetation from a sagebrush-dominated system before

the burn, to cheatgrass and other small bushes and shrubs after the fire. Cheatgrass-dominated systems are considerably more likely to burn than sagebrush systems and fire recurrence intervals can change from ~60–110 years to in some cases <5 years with this change in vegetation (Knapp, 1996). Because cheatgrass and other invasive species in these systems can inhibit native perennial species from re-establishing themselves after fires, these changes in fire regime can become permanent (Young et al., 1969).

4. Summary

Results from this study showed that mercury was distributed primarily in the mineral soil ecosystem reservoir (>90%) relative to the smaller litter and vegetation reservoirs at all three sites. However, these smaller reservoirs play a significantly more active role in the cycling of Hg in these systems (Krabbenhoft et al., 2005). Mercury distributions at the Sierran forest sites (forest prescribed burn and forest wildfire sites) were similar to those observed for other US forest sites (Grigal, 2003). The semiarid desert wildfire site exhibited significantly smaller vegetation and litter reservoirs due to lower biomass.

Comparison of unburned data to burned data demonstrated that litter and possibly vegetation (with high enough fire severity) are important sources for Hg emitted during fires. Our hypothesis that mineral soils were the most important source of Hg in the fire was not fully addressed by this study, although fire severity point data and results from other workers suggest that the Hg in upper few millimeters of the mineral soil may be volatilized. Our calculations suggested that mineral soils have a relatively small input (<15%) to the total Hg emission while litter appears to be the dominant source. Mercury emission estimates for fires at the two forest sites were 2.0 to 5.1 g Hg ha⁻¹ (forest prescribed burn site) and 2.2 to 4.9 g Hg ha⁻¹ (forest wildfire site). The emission estimates are surprisingly similar but similar vegetation and litter between the sites may be a factor. These estimates compare well with those from other forested sites using mass balance (Woodruff et al., 2001; Harden et al., 2004) and Hg:CO methods (Sigler et al., 2003; Friedli et al., 2003b). Emission estimates as a result of fire from the desert wildfire site were considerably lower than for the forest sites (0.36±0.13 g Hg ha⁻¹).

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