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ORIGINAL ARTICLE

Mass Balances of Mercury and Nitrogen in Burned and Unburned Forested Watersheds at Acadia National Park, Maine, USA

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Abstract Precipitation and streamwater samples were collected from 16 November 1999 to 17 November 2000 in two watersheds at Acadia National Park, Maine, and analyzed for mercury (Hg) and dissolved inorganic nitrogen (DIN, nitrate plus ammonium). Cadillac Brook watershed burned in a 1947 fire that destroyed vegetation and soil organic matter. We hypothesized that Hg deposition would be higher at Hadlock Brook (the reference watershed, 10.2 μg/m^2/year) than Cadillac (9.4 μg/m^2/year) because of the greater scavenging efficiency of the softwood vegetation in Hadlock. We also hypothesized the Hg and DIN export from Cadillac Brook would be lower than Hadlock Brook because of elemental volatilization during the fire, along with subsequently lower rates of atmospheric deposition in a watershed with abundant bare soil and bedrock, and regenerating vegetation. Consistent with these hypotheses, Hg export was lower from Cadillac Brook watershed (0.4 μg/m^2/year) than from Hadlock Brook watershed (1.3 μg/m^2/year). DIN export from Cadillac Brook (11.5 eq/ha/year) was lower than Hadlock Brook (92.5 eq/ha/year). These data show that ~50 years following a wildfire there was lower atmospheric deposition due to changes in forest species composition, lower soil pools, and greater ecosystem retention for both Hg and DIN.

Keywords mercury · nitrogen · DIN · methylmercury · forest fire effects · watershed mass balances · Acadia National Park · Maine, USA

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1 Introduction

Acadia National Park, Maine, is a designated Class I area by federal Clean Air Act regulations (40 CFR Section 52.21). However, it is well documented that the Park receives elevated inputs of atmospheric pollutants (Kahl et al., 1991; Heath, Kahl, Norton,

Brutsaert, 1993; Nelson 2002 and this volume; Johnson 2002 and in press; NADP/NTN Data, 2003). Acidification of some aquatic resources has been documented by Kahl and co-workers (Kahl, Andersen, & Norton, 1985; Kahl et al., 1995, 1999), including seasonal episodic acidification (Kahl et al., 1985, 1992; Heath, Kahl, Norton, & Fernandez, 1992). High concentrations of mercury (Hg) in fish (Stafford & Haines, 1997) are well documented. Continued deposition of nitrogen (N) may result in nitrogen saturation of forests (Aber, Naderhoffer, Steudler, & Melillo, 1989, 1998), or contribute to surface water acidification (Kahl et al., 1993; Stoddard, 1994). We established two long-term gauged watersheds in 1998, to serve as the foundation for research on these issues in Acadia National Park.

Changes in N dynamics in eastern U.S. forested watersheds have been a focus of ecological research since Aber et al. (1989) identified the issue of ecosystem N accumulation over a decade ago. Subsequently, *nitrogen saturation* has received increasing attention (Henriksen & Brakke, 1988; Hauhs, Rost-Siebet, Raben, Paces, & Vigerust, 1989; Schulze, 1989; Murdoch & Stoddard, 1992; Sullivan, 1993; Stoddard, 1994; Jeffries, 1995; Mitchell, Raynal, & Driscoll, 1996; Williams, Baron, Caine, Sommerfeld, & Sanford, 1996). In the U.S., nitrate (NO_3^-) concentrations in streams draining forests in the Northeast are higher than in any other forested region in the country, and have increased by a factor of 3 to 4 since 1970 (Helsel, 1996).

Research suggests that initial N status of the site drives differences in N dynamics between sites (Aber et al., 1998). Campbell et al. (2004) suggest that there are multiple controls on N retention in forested watersheds: hydrology, vegetation, and landuse history among them. Major forest fires are relatively infrequent in the Northeast; however, recent evidence suggests that when fires do occur, they have dramatic and lasting effects on N retention in forested watersheds (Goodale, Aber, & McDowell, 2000). In a regional survey of 159 small, forested watersheds in central New England, Cone Pond Watershed, burned ca. 1820, in New Hampshire was one of only three watersheds having no detectable NO_3^- in streamwater (Hornbeck, Bailey, Buso, & Shanley, 1997).

While watershed-scale N dynamics have been widely studied, relatively few whole-watershed studies have been completed that consider Hg in both throughfall flux (considered to be wet plus dry deposition) and intensively sampled streamwater. Lake Gardsjon and the Svartberget watershed in Sweden, the Experimental Lakes Area in Canada, and Walker Branch Watershed in Tennessee are a few notable studies (Grigal, 2002). Even fewer studies have attempted to describe MeHg cycling in forested watersheds and the data that exist indicate that MeHg dynamics are not controlled by the same mechanisms as Hg (Lee, Bishop, & Munthe, 2000; Grigal, 2002).

2 Objectives

The focus of this research was to compare watershed responses between the Cadillac Brook watershed that burned in 1947, and the Hadlock Brook watershed that has been largely undisturbed for at least 300 years (Schauffler et al., in press). In this paper, we estimate the input–output mass balances for dissolved inorganic carbon (DIN, nitrate plus ammonium), Hg, and MeHg. We hypothesized that fluxes from the unburned watershed would be higher because: 1) Deposition of Hg and N would be higher in the Hadlock Brook watershed because mature coniferous forest would enhance dry deposition; 2) elemental pools in the soil would be higher compared to the burned soils in the Cadillac Brook watershed; and, 3) uptake of nutrients would be enhanced in the regenerating forest.

The watershed Hg mass balance does not include volatilization. Research regarding volatilization rates has been conducted (see Grigal, 2002; Zhang & Lindberg, 1999; Lindberg, 1996), but there is no clear consensus on how to quantify this flux as part of a watershed mass balance. We did not quantify litterfall Hg for this study, but results from studies in these watersheds (Johnson, 2002; Sheehan, 2005) indicate that Hg in litterfall is a significant flux to the forested watersheds. Grigal (2002) reviews relevant literature and reports that leaves in litterfall had concentrations 1.5 to 2 times higher than live foliage (Grigal, Kolka, Fleck, & Nater, 2000; Lindberg, 1996; Rea, Keeler, & Scherbatskoy, 1996), similar to the results from the Acadia watersheds. Concentrations of Hg in twigs, bark and other litter components can be higher still than in leaf litter (Grigal, 2002; Grigal et al., 2000). The contribution of litterfall to MeHg input is reported to be small (Grigal et al., 2000).

3 Materials and Methods

3.1 Site description

The research watersheds were chosen for key dissimilarities (burn history and vegetation), and otherwise are similar (Table I). The watersheds are described in detail in three papers in this volume: Kahl et al. (in press), Schauffler et al. (in press), and Wiersma, Elvir, and Eckhoff (in press).

3.2 Experimental design

The natural experimental design utilizes contrasting vegetation and soil characteristics between the two watersheds that exist, in part, due to the severe wildfire that burned Cadillac Brook watershed in 1947 (Figure 1). The contrasting site histories have caused significant differences in ecosystem properties, as described in this volume by Kahl et al. (in press), Schauffler et al. (in press), and Wiersma et al. (in press). The burned watershed has regenerating hardwood forests and less organic matter in soils, in contrast to the unburned softwood forest with mature softwoods and older (thicker), more acidic soil organic horizons. An underlying premise of this research was that in the burn zone, the ecosystem pools of C, N, and Hg were depleted in 1947. By comparing the two contrasting watersheds, we characterized 'modern' accumulations and altered cycling processes that may control N-saturation and the bioavailability of Hg.

Table I Summary of physical characteristics for the study watersheds, including streamflow statistics for water year 2000, September 1, 1999–September 30, 2000

Watershed	Cadillac Brook	Hadlock Brook
Minimum elevation (m)	122	137
Maximum elevation (m)	468	380
Average slope	28%	20%
Aspect	E-SE	W-SW
Annual mean streamflow (l/s)	8.21	14.7
Peak streamflow (l/s)	425	821
Watershed area (ha)	31.6	47.2

Streamflow is measured by the U.S. Geological Survey and data are available online at http://waterdata.usgs.gov/me/nwis/.

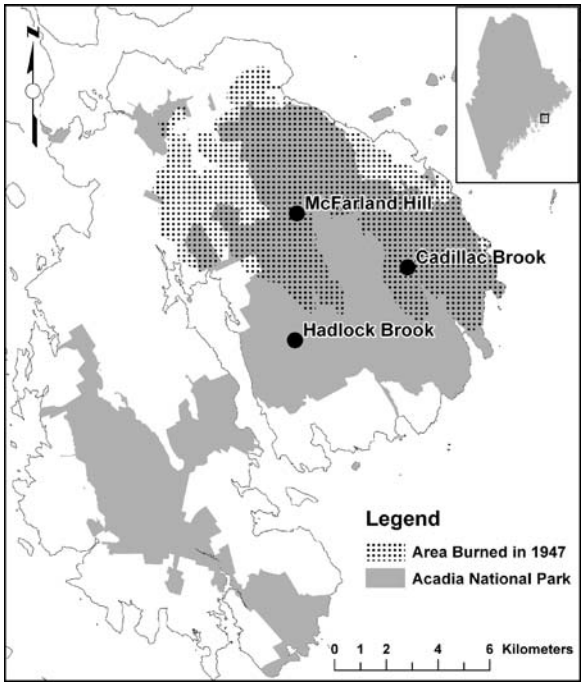

Figure 1 Locations of study sites at Acadia National Park, which is located primarily on Mount Desert Island, Maine, USA. Cadillac Brook watershed was burned in a wildfire in 1947, and Hadlock Brook watershed is undisturbed. Wet deposition of mercury (*Hg*) and nitrogen (*N*) is measured at nearby McFarland Hill, National Atmospheric Deposition Program/Mercury Deposition Network (*NADP/MDN*) site ME98. GIS layers are courtesy of the National Park Service at Acadia National Park.

3.3 Throughfall sampling and watershed inputs

For Hg and N species, throughfall collectors were deployed from 3 May to 16 November 2000. The samples were collected 10 times at varying intervals according to techniques in Johnson et al. (in press) and Nelson (2002). Throughfall was collected under each of five vegetation classes: deciduous, coniferous, mixed conifer/deciduous, scrub/shrub, and open/no canopy. Hadlock Brook watershed was 70% conifer and 23% open/shrub (at higher elevations), while Cadillac Brook watershed vegetation was more evenly distributed between vegetation types (Wiersma et al., in press).

For each collection and location, deposition was calculated based on the depth of precipitation (determined from volume), catch area of the funnel, and chemistry data. Throughfall flux estimates were areally weighted by the proportion of each vegetation type in each watershed. For the freezing season, when throughfall was not collected, Mercury Deposition

Network (MDN; http://nadp.sws.uiuc.edu/mdn/) inputs of Hg and MeHg were used. For freezing-season DIN, National Atmospheric Deposition Program (NADP; http://nadp.sws.uiuc.edu/) inputs were used after being scaled-up by vegetation-specific factors as described in Nelson (2002).

3.4 Streamwater sampling

Streamwater grab samples were collected weekly and (bi-weekly in the winter) for full lab analysis, following standard EPA stream sampling procedures (Kahl et al., 1992; Peck, Morrison, Mader, & Chaloud, 1993), including semi-clean collection methods for total and MeHg modified from EPA Ultraclean method 1669 (Johnson, 2002). Stream samples were taken from small pools just upstream from the USGS gauging stations for each watershed. Samples for mercury analysis were collected in two Teflon™ bottles for each stream, one for Hg (500 ml), and one for MeHg (250 ml). Major ion samples were collected in HDPE bottles and syringes. Duplicate samples were collected on an alternating watershed schedule. Event sampling occurred when flow exceeded ~28 l/s, then ISCO™ auto samplers collected one sample every 4 h until stream flow decreased to below ~28 l/s. Event samples were collected in borosilicate glass for Hg samples and in HDPE bottles when Hg analysis was not performed. Events were not analyzed for MeHg because of holding time constraints.

The U.S. Geological Survey in Augusta, Maine, monitors stream stage at the outflow of each research watershed with a pressure transducer linked to the Internet via satellite. Stage is recorded at 15-min intervals, and was used to calculate streamflow in each stream based on empirical stage–discharge relations. For water year 2000, annual mean streamflow for Cadillac Brook was 8.21 l/s, and peak streamflow was 424 l/s. For Hadlock Brook in water year 2000, annual mean streamflow was 14.7 l/s and peakflow was 821 l/s.

3.5 Laboratory methods

Throughfall and streamwater samples were analyzed for major ion chemistry at the laboratories of the Senator George J. Mitchell Center at the University of Maine, using standard EPA methods (Peck et al., 1993; Norton & Fernandez, 1999) in place for more than a decade as part of several major EPA projects. Briefly, NO_3^- was measured using ion chromatography and NH_4^+ was analyzed using an ammonium autoanalyzer. Dissolved organic carbon (DOC) was quantified with persulfate oxidation and IR detection. Total Hg was measured with cold vapor atomic fluorescence spectrometry (CVAFS) using EPA Method 1631 with a dual gold trap. Methyl Hg was analyzed using distillation, aqueous ethylation, purge and trap, and CVAFS, as in EPA Method 1630. Equipment blank Hg concentrations ranged from the detection limit (0.1 ng/l) to 1.46 ng/l, averaging 0.35 ng/l, and the value of the blank was always less than 10% of the sample Hg concentration.

3.6 Calculation of mass balances

Mass balances for the Acadia watersheds were calculated using a general watershed mass balance equation: inputs − outputs = change in pool (e.g., Kahl, Norton, Fernandez, Rustad, & Handley, 1999). We used measured and calculated inputs, stream chemistry data and interpolated values, and hourly discharge to calculate mass balances for each watershed. The granite bedrock in the two watersheds has a slow weathering rate and is assumed to be tight with respect to losses to groundwater for mass balance purposes (Gilman, Chapman, Lowell, & Borns, 1988). Volatilization of Hg to the atmosphere was not quantified, nor were changes in deep groundwater storage given the difficulties of field measurement (Grigal et al., 2000). The subtraction of output from input yields the amount of Hg, MeHg, or DIN retained in the watershed (and lost to volatilization for Hg or denitrification for DIN). For MeHg, we used one-half the method detection limit (MDL; 0.025 ng/l) to calculate fluxes for individual observations below the MDL. For NO_3 (MDL=0.1 μeq/l) and NH_4 (MDL = 0.05 mg/l) we used actual values that were reported, usually 0.0 or 'noise' that was 1/2 MDL or less. Most values of NH_4 in streamwater were below the MDL, so the fluxes should be viewed as estimates. We also assumed that organic N was a small percentage of the total N in streamwater. Analysis of 225 individual streamwater solutions collected between 1999 and 2002 indicated that most of the N was indeed NO_3^-, and DIN was more than 70% of total N for 80 of those samples. Inorganic N is typically the dominant form of N in low-DOC northeastern streamwaters (Campbell et al., 2004).

4 Results and Discussion

4.1 Watershed inputs

The mean volume of throughfall precipitation collected in individual samples was 985 ml in the Cadillac Brook watershed and 991 ml in the Hadlock watershed. These deposition inputs were not significantly different (ANOVA, $P = 0.94$). These volumes correspond to 51.0 and 51.3 mm of precipitation falling on the Cadillac and Hadlock watersheds, respectively, during the period reported here.

4.1.1 Hg inputs

The average total Hg concentration in throughfall (494 samples) over the duration of this study was lower in the Cadillac Brook watershed (14.2 ng/l) than in Hadlock Brook watershed (18.8 ng/l) (Table II). The vegetation-weighted total Hg deposition for the study period was 9.4 μg/m^2/year in Cadillac Brook watershed compared to a 10.2 μg/m^2/year for Hadlock Brook watershed. These input estimates use MDN data from the nearby station at park headquarters for the period when throughfall was not sampled, November 16, 1999–May 2, 2000 (Table II). Total Hg throughfall deposition was significantly higher at Hadlock Brook (ANOVA, $P < 0.001$). Total Hg in throughfall in Vermont was 11.6 μg/m^2/year in Lake Champlain watershed and 10.5 μg/m^2/year in the Lake Huron watershed (Rea, Lindberg, & Keeler, 2000). Both measurements were made under mixed hardwoods.

4.1.2 MeHg inputs

A total of 131 bulk and throughfall samples were analyzed for MeHg. The average concentration of MeHg in precipitation was 0.07 ng/l for Cadillac Brook watershed and 0.10 ng/l for Hadlock Brook watershed (Table II). The concentrations were consistently near the detection limit, and variability was

Table II Concentration of mercury (Hg, in ng/l) and nitrogen (NO_3^- and NH_4^+, in μeq/l) in Cadillac and Hadlock Brook watershed throughfall (May 3 to November 16, 2000), and National Atmospheric Deposition Program (NADP) and Mercury Deposition Network (MDN) wet-only concentrations for site ME98, McFarland Hill, Acadia National Park

		n	Mean (SD)	Minimum	Maximum
May 3, 2000–November 16, 2000					
Cadillac	TotHg (ng/l)	241	14.2* (1.5)	2.1	68.4
	MeHg (ng/l)	63	0.07** (0.06)	<DL	0.28
	NO_3^- (μeq/l)	279	19.2* (18.6)	<DL	116
	NH_4^+ (μeq/l)	279	5.37* (8.3)	<DL	85
Hadlock	TotHg (ng/l)	253	18.8* (1.4)	2.2	55.9
	MeHg (ng/l)	68	0.10** (0.11)	<DL	0.63
	NO_3^- (μeq/l)	273	24.3* (22.6)	<DL	145
	NH_4^+ (μeq/l)	273	10.1* (17.4)	<DL	188
May 3, 2000–November 14, 2000					
MDN (ME98)	TotHg (ng/l)	24	11.8 (11.4)	1.7	52.4
NADP (ME98)	NO_3^- (μeq/l)	18	21.8 (18.5)	2.26	68.4
	NH_4^+ (μeq/l)	18	12.2 (10.5)	<DL	31.6
Nov. 16, 1999–May 2, 2000					
MDN (ME98)	TotHg (ng/l)	18	6.87 (4.66)	0.3	20.2
NADP (ME98)	NO_3^- (μeq/l)	22	24.7 (27.6)	3.71	124
	NH_4^+ (μeq/l)	22	10.0 (15.0)	<DL	65.4

*$P < 0.01$

**$P < 0.05$

One-half the detection limit was used for samples below the detection limit for computing means for N species and MeHg. All means were significantly different between watersheds (ANOVA). NADP and MDN data were not statistically evaluated.

large. Using MDN estimates for winter deposition, the annual vegetation-weighted MeHg deposition for Cadillac Brook watershed was 0.05 and 0.1 μg/m^2/year for Hadlock Brook watershed (Table IV).

4.1.3 N input

Mean concentrations of both NO_3^- and NH_4^+ in deposition were significantly higher for the Hadlock Brook watershed than Cadillac ($P < 0.01$, ANOVA) (Table II). NADP DIN concentrations were remarkably similar to throughfall DIN concentrations for the two watersheds, despite the fact that NADP samples were collected weekly while throughfall solutions were integrated over periods of two to three weeks. We expect that any throughfall N-enrichment due to dry deposition or landscape effects was reduced by foliar uptake of N. This is especially true for Cadillac Brook watershed, where DIN in throughfall was lower than in NADP wet-only precipitation, presumably because of the more active hardwood canopy. The deposition data from each collector were weighted by the proportion of each vegetation type in the watershed to estimate a whole watershed input. (Nelson, 2002).

4.2 Streamwater

4.2.1 Hg and MeHg in streamwater

Arithmetic mean Hg concentration in Cadillac Brook (0.6 ng/l) was less than half the mean Hg concentration in Hadlock Brook (1.5 ng/l). Total Hg concentrations in Hadlock Brook during events were up to five times the arithmetic mean for Cadillac Brook, peaking at 3.14 ng/l (Figure 2). The maximum Hg concentration during an event in Hadlock Brook was 5.70 ng/l, nearly four times the mean (Table III). These concentration values are reflected in total export (Table IV).

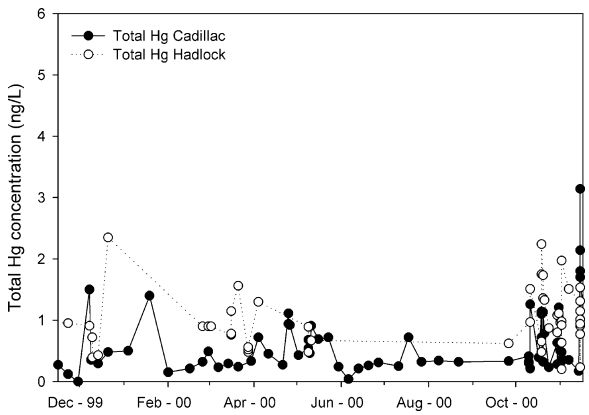

Figure 2 Streamwater mercury concentrations (ng/l) in Cadillac and Hadlock Brooks for the study period, November 17, 1999–November 16, 2000.

Table III Stream water concentrations for mercury (Hg, in ng/l), methylmercury (MeHg), dissolved organic carbon (DOC, in mg/l) and nitrogen (NO_3^- and NH_4^+, in μeq/l) in Cadillac and Hadlock Brooks, for event and regular grab samples taken between November 17, 1999 and November 16, 2000

		n	Mean	Minimum	Maximum
Cadillac	TotHg (ng/l)	104	0.6	<DL	3.14
	MeHg (ng/l)	12	0.05	<DL	0.12
	NO_3^- (μeq/l)	93	0.7	<DL	7.4
	NH_4^+ (μeq/l)	93	<DL	<DL	<DL
	DOC (mg/l)	96	1.6	0.6	3.8
Hadlock	TotHg (ng/l)	139	1.5	<DL	5.7
	MeHg (ng/l)	17	0.07	<DL	0.15
	NO_3^- (μeq/l)	126	7.4	0.3	19.8
	NH_4^+ (μeq/l)	125	<DL	<DL	3.3
	DOC (mg/l)	130	2.7	1.3	6.7

One-half the detection limit was used for samples below detect for computing means for N species.

Higher Hg export from Hadlock Brook watershed (0.4 μg/m^2/year) as compared to Cadillac Brook watershed (1.3 μg/m^2/year) may be a result of factors that include greater Hg pools in watershed soils (Amirbahman, Ruck, Fernandez, Haines, & Kahl, 2004) and greater throughfall deposition or less retention by soils. The annual export of total Hg from Hadlock Brook watershed in this study was in the same range as reported for other similar studies. For example, Lee et al. (1998) reported a value of 1.3 μg/m^2/year for a coniferous watershed, and Grigal et al. (2000) found 2.2 μg/m^2/year for a watershed dominated by mature aspen (*Populus tremuloides Michx.*) in Minnesota. Scherbatskoy, Shanley, and Keeler (1998) measured streamwater export of Hg as 3.2 and 2.2 μg/m^2/year for years one and two, respectively, for a two-year project in Vermont. However, the annual export of Hg from Cadillac Brook watershed was much lower, only about one-third of that for the other watersheds, supporting the retention hypothesis due to factors relating to the fire.

Table IV Mass balances for the gauged watersheds at Acadia National Park for November 17, 1999–November 16, 2000 and Bear Brook Watershed, Maine for November 1, 1999–October 31, 2000

	Inputs	Outputs	Net flux	Retention
Cadillac				Percent
NH_4^+	46	6.3	40	86
NO_3^-	183	5.2	178	97
NO_3^-+NH_4^{+-}	229	11.5	218	95
Hg	9.4	0.4	9.0	96
MeHg	0.05	0.04	nm	nm
Water (mm)	1190*	792**	398	Yield – 67%
Hadlock				
NH_4^+	60	10.1	50	83
NO_3^-	168	82.4	86	51
NO_3^-+NH_4^+	228	92.5	136	59
Hg	10.2	1.3	8.9	87
MeHg	0.10	0.06	nm	nm
Water (mm)	1190*	992**	198	Yield – 83%
East Bear				
NH_4^+	137	12.9	124	91
NO_3^-	227	15	212	93
NO_3^-+NH_4^+	364	27.9	336	92
Water (mm)	1239	976**	263	Yield – 79%

*Inputs from NADP Site ME98.

**Calculated from U.S. Geological Survey daily discharge values.

Inputs were calculated from throughfall data from May 3–November 16, 2000, and NADP/MDN data and ratios for November 17, 1999–May 2, 2000. Streamwater outfluxes were calculated using the methods of Kahl et al. (1999). Mass balance was calculated as throughfall input–stream outflux. Retention was calculated as 100×(input − output)/input. Units for nitrogen species are equivalent per hectare per year and for mercury species are microgram per square meter per year. The mass balance for MeHg is not meaningful (nm), as described in the text.

MeHg export was estimated as 0.04 μg/m^2/year from Cadillac Brook watershed and 0.06 μg/m^2/year from Hadlock Brook watershed (Table III), based on fewer samples analyzed for MeHg than for Hg.

4.2.2 The DOC connection

The literature on Hg in freshwater systems shows a strong correlation between Hg and DOC (Pettersson, Bishop, Allard, & Lee, 1995; Kolka, 1996; Vaithiyanathan, Richardson, Kavanaugh, Craft, & Barkay, 1996; Lee et al., 1998; Lee et al., 2000; Shanley et al., 2002; Bishop, Lee, Pettersson, & Allard, 1995). We analyzed streamwater samples for DOC to investigate the relationship between Hg and DOC. The mean DOC concentration was 1.6 mg/l in Cadillac Brook and 2.7 mg/l in Hadlock. Both DOC and total Hg concentrations in Cadillac Brook were consistently around one-half the value measured in Hadlock Brook.

DOC and Hg were positively and linearly correlated in streamwater samples in Cadillac and Hadlock Brooks (r^2 = 0.71, P < 0.01; Figure 4). The same mechanism of DOC-Hg transport appeared to exist in both streams. There was no relationship between MeHg and DOC concentrations in either Cadillac (r^2 = 0.03, P = 0.57) or Hadlock brooks (r^2 = 0.12, P = 0.18).

High Hg concentration observed in streamwater during high discharge episodes likely results from DOC mobilization of Hg by complexation (Kolka, 1996; Lee et al., 2000) due to increased shallow and overland flow (Shanley et al., 2002; Scherbatskoy et al., 1998). The dependency of Hg transport on the DOC complexation mechanism also helps explain lower Hg export by Cadillac Brook. Cadillac Brook exports less DOC, which is attributed to the depletion of soil organic C by the fire, erosion following the fire, and faster rates of decomposition in soils due to higher quality litter from the pioneer hardwood forest types in Cadillac.

4.2.3 DIN in streamwater

Concentrations of NH_4^+ are typically below the laboratory detection limit (0.05 mg/l) in aerobic streams of the northeastern U.S. (Stoddard et al., 2003). However, concentrations of NO_3^- were dramatically different between streams (Figure 3). Mean NO_3^- concentration was 0.7 μeq/l in Cadillac Brook and 7.4 μeq/l in Hadlock Brook. Typically, Cadillac Brook streamwater NO_3^- was above detection during high discharge events such as spring snowmelt and fall high flow (Figure 3).

DIN export from Cadillac (11.5 eq/ha/year) was significantly lower than the DIN export for Hadlock Brook (92.5 eq/ha/year) (Table IV). Nielsen and Kahl (in press) reported no significant difference between burned and unburned sites for either NO_3^- or total N export from 13 small watersheds on Mount Desert Island, where Acadia National Park is located. The lack of a fire effect was attributed to masking effects of other landscape factors, such as vegetation type or near-stream soil processes (Nielsen & Kahl, in press).

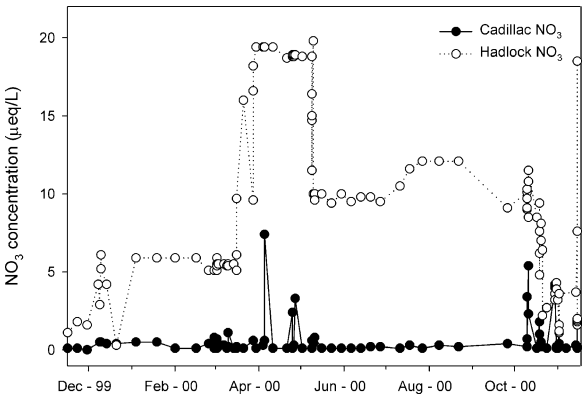

Figure 3 Streamwater nitrate concentrations (μeq/l) in Cadillac and Hadlock Brooks for the study period, November 17, 1999–November 16, 2000.

We are unable at present to attribute the differences in N flux in the upland watersheds to a fire effect, or other factors. Factors such as stand composition are affected by fire and it is likely that contrasts such as younger and more pioneer hardwood species in Cadillac create a higher demand for available N. Preliminary sampling at the outlet of an unburned sub-catchment near the top of Cadillac Brook watershed showed NO_3^- concentrations in streamwater similar to those at undisturbed Hadlock Brook (Nelson, unpublished data).

4.3 Watershed mass balances

Likens, Bormann, Pierce, Eaton and Johnson (1977) pioneered the use of watershed mass balances at HBEF, NH. The theory is simple:

$$storage = input - output \quad (1)$$

We define input as throughfall deposition as described in this paper. Output is stream output of measured solute, calculated from concentration multiplied by USGS water flux. A discrete value of concentration was interpolated for each discharge value, using the methods of Kahl et al. (1999). A negative value for storage in Equation (1) is a net outflux and a positive value is retention within the watershed.

4.3.1 Mercury mass balance

The Hg mass balance for Cadillac Brook watershed was an accumulation of 9.0 μg/m^2/year, or 95% of the total Hg deposited on Cadillac Brook was retained by the watershed (Table IV). These calculations do not include potential volatilization losses. The Hg mass balance for Hadlock Brook watershed was accumulation of 8.9 μg/m^2/year, or 87% of the total Hg deposited on Hadlock Brook was retained by the watershed (Table IV). Retention of Hg was estimated as 92% of total deposited Hg in northern Wisconsin's lake district (Krabbenhoft, Benoit, Babiarz, Hurley, & Andren, 1995). A mass balance for MeHg is not meaningful, because of chemical transformations undergone by Hg after deposition from the atmosphere.

Hg throughfall input was similar for both watersheds. The lower flux of Hg from Cadillac Brook watershed was probably a result of increased Hg retention and a smaller soil pool of Hg (Amirbahman et al., 2004). This probably reflects both the depletion of Hg due to the fire as both gaseous losses and through erosion, as well as changes in organic matter quality due to vegetation changes leading to higher rates of Hg complexation per mass of soil organic C. Recently, researchers from the METAALICUS project in Canada reported that Hg in runoff was primarily 'old' mercury, not isotopically labeled, newly applied ^{202}Hg (Hintelmann et al., 2002). The results from this study at Acadia support the conclusion that newly deposited Hg is quickly stored in the watershed, and is not necessarily reflected in short-term runoff.

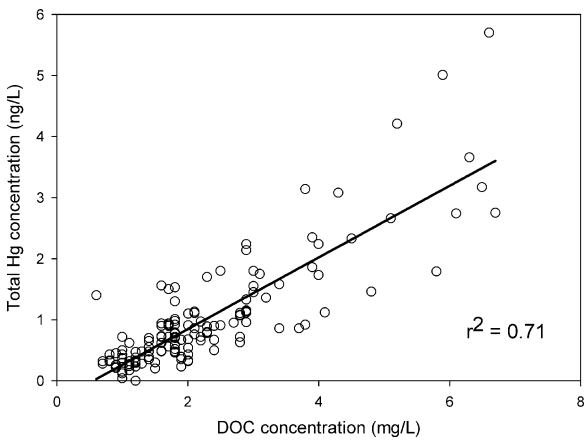

Figure 4 Mercury (*Hg*) vs. dissolved organic carbon (*DOC*) concentration in Cadillac and Hadlock Brooks for the period during 17 November 1999 to 16 November 2000. Equation for regression line for Cadillac: Hg Conc = 0.55 (DOC Conc) – 0.23, r^2=0.53. Equation for regression line for Hadlock: Hg Conc = 0.60 (DOC Conc) – 0.40, r^2=0.71.

Higher MeHg export in streams fed by bogs or fens also suggests that the presence of organic substrates may influence MeHg production (Kolka,1996; Grigal et al., 2000). Grigal et al. (2000) found that bog areas contribute twice the MeHg to the stream compared to upland areas. Hadlock Brook, which has ~33% greater export of MeHg than Cadillac Brook, is fed by two upland wooded wetlands that may have favorable redox conditions for MeHg production.

4.3.2 DIN mass balances

Throughfall deposition of DIN was the same for the two watersheds: 229 eq/ha/year for Cadillac Brook watershed and 228 eq/ha/year for Hadlock Brook watershed (Table IV). Deposition of NH_4^+ was slightly higher at Hadlock Brook watershed and deposition of NO_3^- was slightly higher at Cadillac Brook watershed. We hypothesized that there would be higher deposition at Hadlock because of greater scavenging efficiency of the primarily softwood canopy (Kahl et al., in press). The hypothesis was supported for sulfate and chloride, generally considered to be conservative with respect to the forest canopy (Nelson, 2002). However, for DIN, we propose that lower deposition at Hadlock Brook watershed may have resulted from the large proportion of scrub/shrub vegetation, 30%–40% of the watershed area. For SO_4^- and Cl^-, estimates calculated from throughfall were over 50% higher if the entire watershed were assumed to be softwood as opposed to weighting the inputs by vegetation type, which included the large areas of scrub/shrub vegetation (Nelson, 2002).

Nitrogen is strongly retained by both Cadillac and Hadlock Brook watersheds (Table IV), despite the significantly higher flux from Hadlock Brook. These results are consistent with other regional studies (Kahl et al., 1999), as well as a previous study in the Hadlock Brook watershed. In 1989, data were collected at a station 50 m downstream of the Hadlock site (Heath et al., 1993). The export of NO_3^- from in 1989–1990 was 60 eq/ha/year for the water year 1989, one-third less than for 1999–2000.

The relatively high retention of DIN in Cadillac Brook watershed is attributed to regenerating vegetation (Vitousek & Reiners, 1975; Campbell et al., 2004) and depleted soil pools of N (Parker, Fernandez, Rustad, & Norton, 2001). Campbell et al. (2004) calculated retention of DIN at 96% for Cadillac and 69% for Hadlock, based on monthly flux estimates from 1999 to 2000, using a 1 June water year. The DIN values for this study are 95% (Cadillac) and 59% (Hadlock), in close agreement.

A substantial portion of NO_3^- at Hadlock Brook watershed is exported, and reflects incipient N saturation in this undisturbed spruce-fir forest. Campbell et al. (2004) reported that watersheds with thin soils and high infiltration rates typically had less capacity to retain N due to hydrologic flushing. At Cadillac Brook watershed, soils are thin and discontinuous, yet the watershed had a high retention of N due to soil C:N characteristics and depleted N (Parker et al., 2001). High soil C:N ratios generally lead to N immobilization and less export; but conifers, which have high C:N ratios, typically have a lower demand for N (McNulty, Aber, & Boone, 1991; Campbell et al., 2004). The combination of mature conifer stands and relatively high accumulated soil N pools are likely key factors contributing to the net N loss at Hadlock Brook watershed (Stoddard, 1994).

5 Summary and Implications

The results from our mass balance analysis support the hypotheses that 1) deposition is higher in mature conifer stands of the Hadlock Brook watershed, and 2) there is higher DIN and total Hg retention in the regenerating hardwood forest in the Cadillac Brook watershed despite thinner soils and more rock outcrop compared to the Hadlock Brook watershed. The fire had significant impacts on the Cadillac watershed, including raising the soil pH, altering the forest species mix, and reducing dry deposition for decades in the regenerating forest (Kahl et al., in press, Schauffler et al., in press; Wiersma et al., in press; Amirbahman et al., 2004; Parker et al., 2001). Carbon and Hg soil pools were reduced in Cadillac compared to Hadlock. These differences in watershed characteristics produce the differences in solute fluxes and mass balances observed in this paper.

Solute flux and mass balance data are also available for the Bear Brook Watershed in Maine, determined by the same research group using the same methods as the Acadia watershed research (Table IV).We compared DIN results with those from the Bear Brook Watershed in Maine (BBWM), which

is less than 50 km from the Acadia watersheds (Norton & Fernandez, 1999). Research at BBWM is investigating mechanisms of N saturation and acidification, as well as recovery, in a long-term, whole-watershed N enrichment experiment. Cadillac and Hadlock Brook watersheds bracket the outflux of DIN for East Bear, with Cadillac exporting the least and Hadlock the most (Table IV).

After 14 years of experimental treatment, BBWM and the Acadia watersheds together offered a response continuum for N-saturation that encompasses the range of N enrichment found in the northeastern U. S. (Norton et al., 1994; Uddameri, Norton, Kahl, & Scofield, 1995). Acadia's fire history offered a natural treatment regime that is representative of the mosaic of fire history throughout New England. The continuum of nitrogen enrichment includes: 1) Augmented N (BBWM experimental treatment); 2) high N (reference watershed at BBWM and unburned watershed at Acadia); and 3) low N (burned watershed at Acadia). The BBWM experiment offers the unique opportunity to compare mechanisms of response at BBWM to evidence of similar mechanisms in the Acadia watersheds.

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