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ORIGINAL ARTICLE

Landscape Controls on Mercury in Streamwater at Acadia National Park, USA

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Abstract Fall and spring streamwater samples were analyzed for total mercury (Hg) and major ions from 47 locations on Mount Desert Island in Maine. Samples were collected in zones that were burned in a major wildfire in 1947 and in zones that were not burned. We hypothesized that Hg concentrations in streamwater would be higher from unburned sites than burned watersheds, because fire would volatilize stored Hg. The Hg concentrations, based on burn history, were not statistically distinct. However, significant statistical associations were noted between Hg and the amount of wetlands in the drainage systems and with streamwater dissolved organic carbon (DOC). An unexpected result was that wetlands mobilized more Hg by generating more DOC *in total*, but upland DOC was more efficient at transporting Hg because it transports more Hg *per unit DOC*. Mercury concentrations were higher in samples collected at lower elevations. Mercury was positively correlated with relative discharge, although this effect was not distinguished from the DOC association. In this research, sample site elevation and the presence of upstream wetlands and their associated DOC affected Hg concentrations more strongly than burn history.

Keywords Acadia National Park · Dissolved organic carbon · Maine, USA · Mercury · Surface water quality

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1 Introduction

Mercury (Hg) has a global distribution from both natural and anthropogenic sources. Because Hg bioaccumulates, human fish consumption advisories based on Hg have been posted in forty-two U.S. states (U.S. Environmental Protection Agency 2003) and all Canadian provinces (Environment Canada 2003). Mercury at levels as low as 0.15 mg/kg in fish tissue can cause reproductive and developmental damage to humans (U.S. Environmental Protection Agency 2001). Fish Hg concentrations are spatially variable, with elevated Hg concentrations found in systems as diverse as remote 'pristine' high elevation lakes and to major lakes and rivers.

Mercury is distributed worldwide via atmospheric processes (Mason et al. 1994). However, the relationship between Hg in atmospheric deposition and Hg in biota is not clear since a number of environmental factors control the bioavailability of Hg, such as dissolved organic carbon (DOC), sulfur, redox conditions, and the environment for bacteria that are thought to control bioavailability (Evers et al. 1998; Morel et al. 1998; Krabbenhoft et al. 1995; Driscoll et al. 1994). Even though Hg is deposited to landscapes primarily from the atmosphere, watershed processes modulate the amount of Hg that is detectable in surface waters (Grigal 2002; Landis et al. 2002; Babiarz et al. 1998; Hurley et al. 1995) or sediments (Norton et al. 2003). Scherbatskoy et al. (1998) and Johnston et al. (1990) investigated the effects of various landscape features on Hg in streamwater, finding that wetland proximity was important. The quantity and quality of DOC produced in watersheds has a control on Hg mobility (Amirbahman et al. 2002; Haitzer et al. 2002; Reuter and Perdue 1977).

The occurrence of Hg in surface waters is of concern in Acadia National Park (Fig. 1), as well as the State of Maine. Elevated concentrations of Hg in fish (up to 3.86 μg/g ww) were documented for samples collected on Mount Desert Island (Bank et al. in press), contributing to the statewide fish consumption advisory. Acadia National Park could reasonably be considered to be remote from industrial sources of Hg, but in reality Mount Desert Island appears to be intercepting long-range transport of polluted air masses from the Midwest and moving up the east coast of the U.S., leading to high Hg deposition (Mercury Deposition Network/National Atmospheric Deposition Program (NRSP-3) 2003).

Landscape disturbance could change how Hg is processed and may help illuminate the controls on Hg biogeochemistry. Our research took advantage of the natural experimental design provided by a major wildfire in Acadia National Park in 1947. About one-third of the Park burned in what was described as an intense fire that killed most of the vegetation and probably burned much of the organic soil horizons (Schauffler et al. in press). Johnson (2002) and Nelson et al. (in press) found that total Hg concentrations in a stream draining a research watershed located in the burned zone were lower than in an unburned, reference watershed stream.

Our objective was to determine if total Hg concentrations in stream water were affected by landscape factors such as wetland presence/absence, water chemistry variables, season, or hydrologic condition. In particular, we wanted to determine if there was a significant difference in Hg concentrations between those areas burned in 1947 and those not burned using a larger spatial scale than Nelson et al. (in press).

2 Materials and Methods

Stream water was collected from actively flowing streams at 47 locations under baseflow conditions. Areas affected by the 1947 fire were determined from National Park Service maps. Seventeen sample sites were from burned zones and 30 were from unburned zones (Fig. 1). Samples were collected under autumn baseflow conditions in November 1999, during low spring-baseflow in April 2000, and during high spring-baseflow in late April/early May 2000. Not all stream sites could be sampled at each

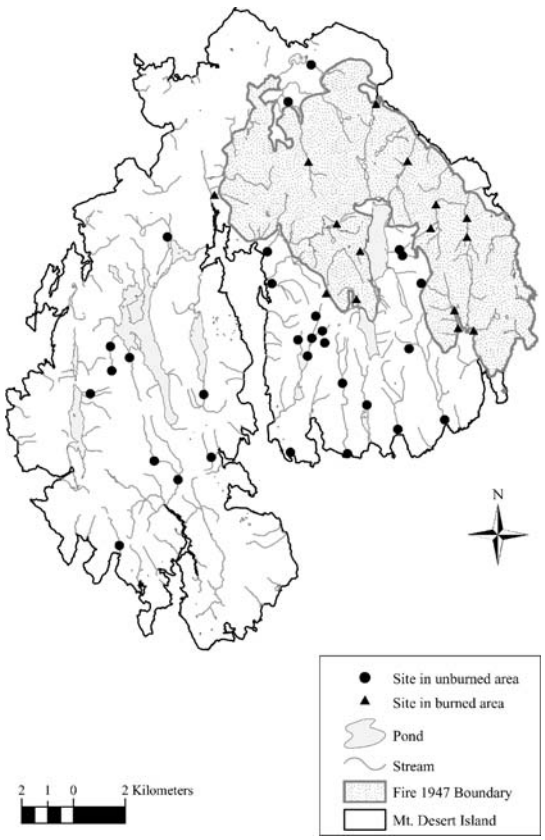

Fig. 1 Locations of surface water samples for mercury analysis in and near Acadia National Park, Mount Desert Island, Maine. Samples from within the limits of the 1947 fire (burn) are shown as *triangles* and those outside the fire zone are *circles*

round due to difficulties in access and depth of active channel (usually too shallow or dry). Samples for major ion chemistry were collected into acid-washed 500 ml HDPE bottles. Samples for total Hg analysis were collected into duplicate 500 ml Teflon bottles using the 'clean-hands dirty-hands' technique (e.g., Nelson et al. in press).

All samples were analyzed at the laboratories of the Senator George J. Mitchell Center for Environmental and Watershed Research at the University of Maine, using clean techniques and instruments involved in related research for more than a decade. Samples to be analyzed for Hg were preserved with bromine monochloride in the laboratory and analyzed on a Tekran model 2600 cold-vapor atomic fluorescence spectrometer, adhering to EPA method 1631 for the analysis of total Hg in water. Purge techniques used in this study differed from those specified in the method, namely that a phase-separator tube was used in place of the bubblers, eliminating foaming in samples with a high organic concentration. Quality control methods included duplicated analyses and matrix spiked samples. Reporting limit was 0.5 ng/l. The major ions (Ca^{2+}, Mg^{2+}, Na^{+}, Cl^{-}, NO_3^{-}, and SO_4^{2-}) and DOC were determined using standard techniques employed in EPA acid rain research in the University of Maine for more than 20 years (Kahl et al. 1993, 2004; Peck 1992; Hillman et al. 1986). These methods are described in more detail in Nelson et al. (in press).

Statistical analyses (regression, *t*-test, ANOVA) were conducted using SYSTAT® v.10 (SYSTAT 2003). Samples below detection limits were assigned a value equal to half the detection limit for statistical analysis. Wetland areas were determined from Maine Office of GIS (2003) wetland maps (1:50,000 base) and streams were assigned to one of two categories: 1) 'wetland-dominated streams', those with >5 ha total wetlands above the sample point; or 2) 'upland streams', those with <5 ha total wetlands above the sample point. A total of 38 sampling locations were identified as wetland-dominated. We did not determine the spatial configuration of wetlands upstream of the sampling site (i.e., wetland proximity, as in Johnston et al. 1990). Sample site elevations were determined using GPS sample site locations and U. S. Geological Survey (USGS) 7.5' topographic maps. Sample point elevations were assigned to the nearest 25-m interval. Stream flow conditions were determined from USGS stream gauges on Hadlock and Cadillac Brooks and assigned to two categories: high (flow > 0.5 cfs) and low (flow < 0.2 cfs) (U.S. Geological Survey 2003).

3 Results and Discussion

Mercury was detected in 90 of 118 stream water samples with a maximum of 5.1 ng/l and a median of 1.2 ng/l (Table 1). The Hg results were evaluated with respect to landscape and chemical associations in order to discern factors that affected the concentrations of Hg in stream water.

3.1 Landscape associations

3.1.1 Burn history

The Hg concentrations were not statistically different between the burned area (1.8 ng/l) and the unburned area (1.6 ng/l)(*t*-test, $p > 0.7$), even when disaggregated by season (*t*-test, $p > 0.3$). The occurrence of a major fire in 1947 appears to have had little effect on present-day Hg concentrations in these study streams.

3.1.2 Seasonality and base flow conditions

An analysis of variance (ANOVA) was performed on Hg concentrations by burn history, season, and flow state. In some individual streams the high spring-baseflow had five times the Hg concentrations of low spring-baseflow. However, in other streams Hg concentrations changed in the opposite direction. Overall, seasonal changes were not uniform in direction or magnitude between high and low flow conditions. Sample populations were not statistically different when compared by season and baseflow condition (all *p*s > 0.3).

3.1.3 Elevation

We evaluated how Hg concentrations in streams changed with elevation. Regression analyses yielded a weakly defined negative correlation between sample point elevation and Hg concentration ($r^2 < 0.6$) that suggests Hg could be accumulating lower in the landscape (Fig. 2). A χ^2 analysis of Hg *versus* elevation (25-m intervals) had a significant association between Hg and elevation ($p < 0.001$). This significant

Table 1 Summary of mercury (Hg) concentrations in stream water at 47 stream locations in Acadia National Park, November 1999 to May 2000

Not burned Mercury in Streams, Mount, Desert, Island	Wetlands >5 ha	Elevation (m.a.s.l.)	Nov-99 Hg (ng/l)	Apr-00 Hg (ng/l)	May-00 Hg (ng/l)
Canada Hollow	Y	50	1.2	1.2	NS
Canon Brook/Cadillac	N	425	1.3	NS	2.7
Duck Cove	Y	19	3.8	4.1	NS
Duck Pond Brook	N	29	1.1	1.8	NS
Duck Pond Brook-Dup.	N	29	0.9	1.8	NS
Duck Pond Inlet	N	69	0.6	1.8	NS
Eagle Lake East/North	N	158	0.6	0.6	1.1
Eagle Lake East/South	N	158	1.3	0.9	1.7
Eagle Lake West	N	89	1.5	1.8	2.6
Gilmore Brook	N	151	0.3*	NS	1.4
Gilmore Brook-Dup.	N	151	0.3*	NS	NS
Great Brook	N	20	0.3*	0.5	NS
Great Brook-Dup.	N	20	NS	0.4*	NS
Hadlock Brook	N	80	1.1	0.5*	1.4
Hadlock HB-1	N	144	0.3*	0.5*	1.3
Hadlock HBB	N	152	2.8	NS	3.2
Hadlock HBC	N	263	0.7	NS	1.5
Hadlock HBE	N	263	2.2	NS	2.2
Hogdon Brook	Y	11	2.6	4.2	NS
Hunters Brook South	N	15	1.1	0.6	1.0
Little Harbor Brook	N	12	0.8	NS	1.0
Little Harbor Brook- Dup.	N	12	NS	NS	1.0
Little Hunters Brook	N	23	NS	2.6	4.2
Little Hunters Brook-Dup.	N	23	NS	2.7	4.2
Lower Old Mill	Y	3	3.6	NS	2.1
Marshall Brook	Y	13	NS	2.5	NS
Norwood Cove	N	10	2.5	2.1	NS
Norwood Cove-Dup.	N	10	2.5	NS	NS
Oak Hill Stream	Y	9	NS	4.9	4.2
Parkman Brook	N	67	ND	NS	0.6
Pemetic Brook	N	128	0.9	0.6	2.8
Richardson	Y	15	NS	2.2	4.4
Sargent Brook	N	23	0.3*	0.6	2.4
Squid Cove Brook	Y	1	4.0	NS	NS
Stanley Brook	Y	73	1.1	1.3	2.1
Steward Brook	Y	12	NS	0.8	NS
Stony Brook	Y	9	3.9	4.7	3.5
Stony Brook-Dup.	Y	9	NS	4.6	NS
The Reservoir	N	60	0.4*	NS	NS
Upper Little Harbor	N	42	NS	0.3*	0.8
Burned					
Aunt Betty Pond Inlet	Y	70	2.2	NS	2.5
Aunt Betty Pond Inlet-Dup.	Y	70	NS	NS	2.4
Breakneck Brook	N	7	ND	1.0	1.6
Canon Brook West	N	73	0.3*	NS	1.3
Canon Brook West-Dup.	N	73	0.3*	NS	NS
Canon Brook/Rte. 3	N	25	1.9	1.8	1.5
Canon CB-1	N	125	0.1	0.3*	0.9
Canon CB-1-Dup.	N	125	NS	0.3*	NS

Table 1 (*continued*)

Not burned Mercury in Streams, Mount Desert, Island	Wetlands >5 ha	Elevation (m.a.s.l.)	Nov-99 Hg (ng/l)	Apr-00 Hg (ng/l)	May-00 Hg (ng/l)
Chasm Brook A	N	169	0.1	0.4*	0.6
Cromwell Brook	Y	15	4.2	NS	1.6
Cromwell Brook-Dup.	Y	15	4.1	NS	NS
Duck Brook	Y	36	1.4	1.7	1.1
Duck Brook-Dup.	Y	36	1.3	1.7	1.2
North Deer Brook	N	98	1.1	1.1	1.0
North Deer Brook-Dup.	N	98	NS	1.0	1.0
South Kebo Brook	N	59	ND	0.2*	0.4*
South Kebo Brook-Dup.	N	59	ND	NS	NS
Upper Old Mill	N	28	1.1	1.4	1.8
Upper Old Mill-Dup.	N	28	1.2	NS	NS
West Kebo Brook	N	74	1.2	0.2*	0.4*
Whalesback	Y	4	NS	5.1	4.2
Whalesback-Dup.	Y	4	NS	NS	4.2

* Estimated, *ND* none detected @ 0.5 ng/l, *NS* not sampled, *Dup*. duplicate.

relationship was evident when data were disaggregated into burned watersheds ($p < 0.05$) and unburned watersheds ($p < 0.1$). There were insufficient data to test changes within the same stream at different elevations.

3.1.4 Wetland presence

We tested the association between the sampling locations and the presence of upstream wetlands. The data were disaggregated by the size of upstream wetlands to distinguish between active but narrow riparian zones that are more or less ubiquitous (i.e., saturated stream banks) and wetland-dominated streams as defined above. The mean concentration of Hg in wetland-dominated streams was 2.8 ng/l, compared to 1.0 ng/l for upland streams. This difference was statistically significant ($p < 0.001$). The significant differences were maintained when analyzed by season (all ps < 0.05). Also, the burn history had no effect of the differences.

3.2 Chemical associations

Mercury exhibited associations with DOC but had no other significant correlations with the major chemical variables measured. There were no statistical associations between Hg and SO_4^{2-}, or between Hg and Cl^-.

3.2.1 Dissolved organic carbon

Overall, DOC concentrations ranged from 0.5 to 18.7 mg/l with a median of 2.5 mg/l. Wetland-dominated streams exported more DOC to drainage waters than upland streams. The overall mean concentration of DOC in wetland-dominated streams was 7.3 mg/l compared to 2.5 mg/l for upland streams, regardless of burn history or season sampled. Using linear regression analysis, there was a positive correlation between DOC and Hg (Fig. 3). The Hg to DOC correlation coefficient for the burned area ($r^2 = 0.84$) was greater than for the unburned area ($r^2 = 0.69$), but this difference was not statistically significant. The concentration of Hg was strongly positively correlated with DOC ($p < 0.001$) and the presence of wetlands ($p < 0.001$). This relationship is consistent with the hypothesis that wetlands produce more DOC than uplands. Seasonal variations (spring *versus* fall) in DOC and Hg concentrations were not significantly different.

The slope of the Hg-DOC regressions ranged from 0.36 ± 0.04 ng Hg/mg DOC for upland streams to 0.27 ± 0.03 ng Hg/mg DOC for wetland-dominated streams for all data (Figs. 3 and 4). The lower Hg/DOC ratio in wetland-dominated streams suggests

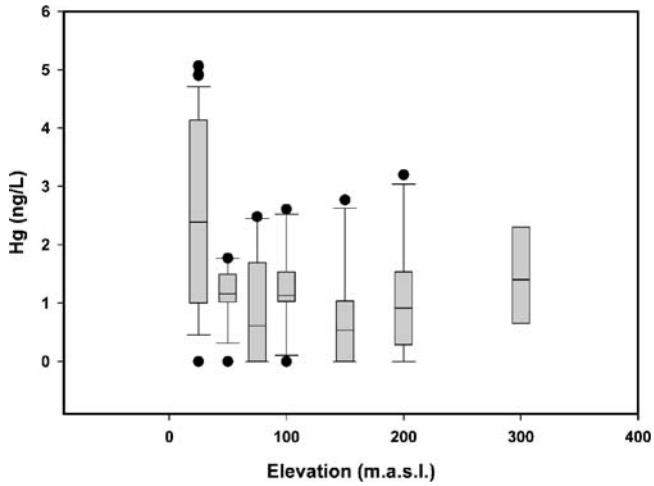

Fig. 2 The distribution of mercury in Acadia National Park streams by collection point elevation. *Box plots* are for the interval elevation ranges (steps: 25, 50, 75, 100, 150, 200, and 300 m)

that they contain more DOC, but less Hg *per unit DOC*, compared to DOC in upland streams that are more efficient, transporting more Hg *per unit DOC*. This same relationship holds when data were regressed by season, with a consistently lower Hg *per unit DOC* in wetland-dominated streams. The regression slope was 5 to 35% steeper for upland streams. The lowest slopes were for the fall wetland-dominated streams (0.25 ± 0.04 ng Hg/mg DOC) and the greatest were for the spring upland streams (0.56 ± 0.06 ng Hg/mg DOC).

4 Conclusions

Many authors have shown that Hg in natural waters is complexed and transported by organic carbon, variably reported as total (TOC) or dissolved (DOC) (Grigal 2002; Shanley et al. 2002; Amirbahman et al. 2002; Lu and Jaffe 2001; Clair et al. 1998; Clair and Ehrman 1998; Hintelmann et al. 1997; Driscoll et al. 1995). Organic carbon in natural waters is a product of many catchment processes, but is especially controlled by the presence of wetlands near water courses, or by the presence of active riparian zones that we consider to be mini-wetlands (Back et al. 2002). Most researchers suggest that the more TOC (or DOC) there is, the more Hg is transported from soils and wetlands (Haitzer et al. 2002). However, our data suggest that the quality (i.e., composition) of the DOC varies in its ability to complex and transport Hg. This means that the Hg-DOC relationship should vary by source of DOC, as we demonstrate. Indeed, it is a surprising observation that on

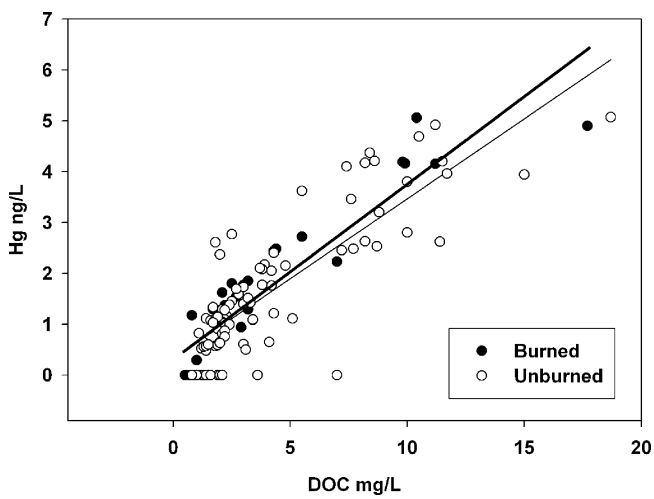

Fig. 3 Relationship between mercury (Hg) and dissolved organic carbon (*DOC*) in streamwater for burned and unburned areas in Acadia National Park, Maine. *Regression line* for burned-zone samples is emphasized

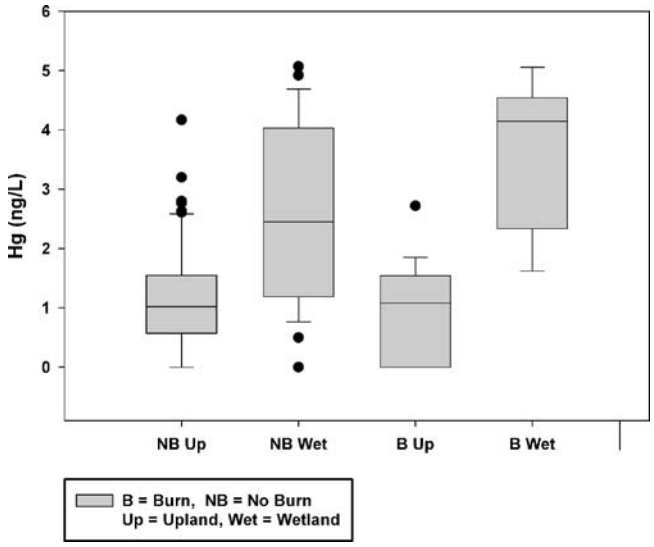

Fig. 4 *Box plots* of streamwater mercury concentrations grouped by burn history and presence of >5 ha wetlands in watershed, Acadia National Park, Maine

a per DOC-unit basis, headwater forest watersheds have the capacity to transport more Hg than wetlands. The alternative explanation, that the DOC transport of Hg from wetlands is limited by the supply of Hg, is not supported by Amirbahman et al. (2002), who found no limitation to the supply of Hg in wetland soils of Acadia National Park. This relationship warrants a more detailed field assessment.

Mercury concentrations increased with decreasing elevation. We interpret this to be a cumulative impact of wetlands and riparian zones. The highest concentrations of Hg occurred during high spring-baseflow. We expect that the same is true during high fall-baseflow, because DOC increases with discharge in the fall. We did not sample during high fall-baseflow periods because they did not occur during our study.

Landscape disturbance, such as fire, appears to be too general a variable on Mount Desert Island to explain the variations in Hg concentrations in streams. The presence of wetlands in the drainage systems had the strongest influence on the amount of Hg in the streams. The wetland effect was more pronounced than any other association, although our analysis was unable to differentiate this effect from higher Hg correlated with greater discharge, because DOC increased with discharge. Burn history may set a baseline for Hg export that is strongly modulated by other watershed processes and may eventually be obscured by wetlands.

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