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Watershed Nitrogen and Mercury Geochemical Fluxes Integrate Landscape Factors in Long-term Research Watersheds at Acadia National Park, Maine, USA

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Abstract This paper is an overview of this special issue devoted to watershed research in Acadia National Park (Acadia NP). The papers address components of an integrated research program on two upland watersheds at Acadia NP, USA (44° 20′ N latitude; 68° 15′ E longitude). These watersheds were instrumented in 1998 to provide a long-term foundation for regional ecological and watershed research. The research was initiated as part of EPA/NPS PRIMENet (Park Research and Intensive Monitoring of Ecosystems Network), a system of UV-monitoring stations and long-term watershed research sites located in US national parks. The initial goals at Acadia NP were to address research questions about mercury, acid rain, and nitrogen saturation developed from prior research. The project design was based on natural differences in forests and soils induced by an intense wildfire in one watershed in 1947. There is no evidence of fire in the reference watershed for several hundred years. We are testing hypotheses about controls on surface water chemistry, and bioavailability of contaminants in the contrasting watersheds. The unburned 47-ha Hadlock Brook watershed is 70% spruce-fir mature conifer forest. In contrast, burned 32-ha Cadillac Brook watershed, 4 km northeast of the Hadlock watershed, is 20% regenerating mixed northern hardwoods and 60% shrub/rocky balds. Differences in atmospheric deposition are controlled primarily by forest stand composition and age. The watersheds are gauged and have water chemistry stations at 122 m (Cadillac) and 137 m (Hadlock); watershed maximum elevations are 468 and 380 m, respectively. The stream water chemistry patterns reflect, in part, the legacy of the intense fire, which, in turn, controls differences in forest vegetation and soil characteristics. These factors result in higher nitrogen and mercury flux from the unburned watershed, reflecting differences in atmospheric deposition, contrasting ecosystem pools of nitrogen and mercury, and inferred differences in internal cycling and bioavailabilty.

Keywords watershed science · hydrology · mass balances · mercury · acidic deposition · nitrogen · forest health · paleoecology · forest fire · Acadia National Park

1 Introduction

Acadia National Park (Acadia NP) has been a magnet for scientists and naturalists for over a century due to its pronounced glacial features, interesting bedrock geology, scenic beauty, and diversity of plant and animal life. The park is within the southern coastal range limit for spruce-fir forests of the northeastern USA (Davis, 1966). Several species and communities are at the edge of their geographic ranges. Thin soils, steep slopes, abundant surface waters, and the highest mountains on the east coast of the United States contribute to the park's sensitivity to disturbance (Kahl, Manski, Flora & Houtman, 2000).

The steep slopes, high peaks, and exposure to coastal fog create an environment conducive to interception of polluted air masses. Therefore, an issue of particular importance at Acadia NP is long-range transport of atmospheric contaminants, including toxic trace substances such as trace metals (Kahl et al., 2000; Norton, Evans, & Kahl, 1997), persistent organic substances (Matz, 1998), mercury (Hg)(Stafford & Haines, 1997) and acidic deposition (Kahl, Andersen, Norton, 1985; 1992, 2000). The inputs of acids and Hg are well characterized at Acadia NP, based on data from the National Atmospheric Deposition Program (NADP) since 1980 (NADP, 2004), and Mercury Deposition Network (MDN) since 1995. Fog pH below 3.5 has been documented (Jagels, Cunningham, Serreze, & Tsai, 1989; Weathers et al., 1988).

Elevated deposition of contaminants, including Hg, has been inferred from sediment accumulation rates at Acadia NP (e.g., Norton et al., 1997; Kahl et al., 1985). Kahl et al. (1992, 1985) and Heath, Kahl, Norton and Fernandez (1992) documented salt- and strong acid-driven acidic episodes in streams, with pH values as low as 4.7. Nitrate concentrations in several streams are chronically elevated (Johnson et al., in press; Kahl et al., 1985), suggesting that nitrogen (N) saturation of the forest (Aber et al., 1998, 1989) is an issue to evaluate. Longcore et al. (in press a, b) documented that tree swallow eggs and chicks from areas of Acadia NP are at least as contaminated with Hg as birds living at a Hg-contaminated Superfund site in Massachusetts, USA.

Concern over ecological issues such as acidification and Hg bioaccumulation led to the recommendation for permanent long-term ecological research at the watershed scale (Kahl et al., 2000). The initial focus of this project has been atmospheric deposition of N and Hg, and their ecological consequences. A primary goal of the long-term watershed research program is to determine how watershed characteristics influence the reservoirs and fluxes of Hg and N in watersheds and surface waters.

1.1 PRIMENet watersheds at Acadia NP

PRIMENet (Park Research and Intensive Monitoring of Ecosystems Network) was a joint nationwide program between the U.S. Environmental Protection and Agency and the National Park Service, established in the late 1990s to assess the effects of environmental stressors on ecological systems. The U.S. Geological Survey (USGS) Biological Resources Division was a co-funder of the program, and the USGS Water

Resources Division was a collaborator at Acadia NP. The network of monitoring and research sites uses park units as outdoor laboratories, where environmental changes are monitored through time in relatively undisturbed and protected sites (http://www.forestry.umt.edu/research/MFCES/programs/primenet/).

At Acadia NP, small, high-elevation watersheds provide a natural experimental setting for investigating long-term influences of forest-type and disturbance history on watershed geochemistry. One possible difference between otherwise similar watersheds, is the history of disturbances such as severe fire (Goodale, Aber, & McDowell, 2000; Goodale, 2003; Magill et al., 1997; Hornbeck, Bailey, Buso, & Shanley, 1997). A major wildfire burned one-third of Acadia NP in 1947, including one of the two watersheds selected for this study (Figure 1). This paper provides a summary of the papers in this special issue dedicated to PRIMENet research at Acadia NP. We describe the use of stand-scale paleoecological tools, historic written records, and tree-ring analyses, combined with modern soil, precipitation, and stream chemistry data, to identify linkages among long-term vegetation history, disturbance, and the cycling of Hg and N within the two watersheds.

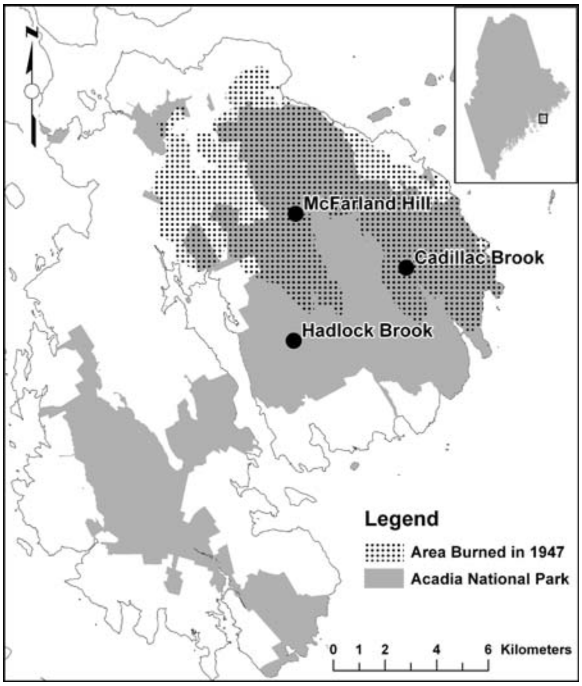


Figure 1 Location of study watersheds in Acadia National Park on Mount Desert Island, Maine, USA. The National Atmospheric Deposition Program site is shown for reference (McFarland Hill). The patterned area was burned in wildfire in 1947. Park boundary and fire extent were provided by Acadia National Park, Resource Management. Map projection is NAD83, Zone 19 North.

We hypothesized that severe disturbance by fire may influence the long-term storage and processing of atmospheric contaminants within watersheds. In particular, burning of soils may have volatilized Hg from organic soil layers, and changed the ratio of N to other soil constituents that control N flux in stream water. We also hypothesized that the distribution of fire would be reflected in the stream chemistry draining the two watersheds.

This pair of research watersheds generally represents disturbance of New England landscapes, some of which have burned over the past several hundred years, and most of which have experienced some level of vegetation change due to different land use histories. Moreover, these watersheds, with thin soils and steep slopes, are inferred to be especially sensitive to perturbations and recovery. This sensitivity makes them ideal to serve as advance indicators of the effects of regional change.

1.2 Regional setting of paired watersheds

Pollen records from Acadia National Park and other coastal settings in New England suggest that large-scale, stand-replacing disturbances of coastal forests occurred at 300 to 500 year intervals prior to European colonization of the region, (Schauffler, 1998; Patterson & Backman, 1988). More frequent fires (both accidental and intentional) and forest clearing accompanied European settlement. Beginning in the late 18th Century and extending through most of the 19th Century, much of the land along Maine's coast was cleared for timber and sheep grazing (McLane & McLane, 1989; Tolonen, 1983). Using land-use/land-cover history as a factor, our research considers whether major *historical* disturbance influences *modern* ecological processes including soil and stream chemistry. Because fire history may be a major control on N cycling (Riggan et al., 1994; Tiedemann et al., 1979), and on Hg accumulation and speciation, research conducted in the northern hardwood or spruce-fir forest at Acadia NP has *regional* implications for both Hg and N biogeochemistry. The recovering burned zone at Acadia NP provides a setting for 'representative' research: Any specific point in the New England landscape may have burned within the past 300 years.

2 Project Design

We established one gauged watershed (Cadillac) in the zone of the intense wildfire of 1947, and one in an unburned area (Hadlock; Figure 1). Neither historical records nor paleoecological reconstructions reveal any indication of fire in the Hadlock watershed for at least 500 years. In addition to basic stream and precipitation data collection, the first year of the project included site characterization using paleoecology and historical records (Schauffler et al., in press), forest health indicators (Wiersma, Elvir, & Eckhoff, in press), and characterization of soil chemistry (Amirbahman, Ruck, Fernandez, Haines, & Kahl, 2004; Parker, Fernandez, Rustad, & Norton, 2002, 2001).

The southeastern slope of the Cadillac Mountain watershed is drained by a headwater stream unofficially called Cadillac Brook. The gauging station for this burned watershed is located at latitude 44°20′41.0″,

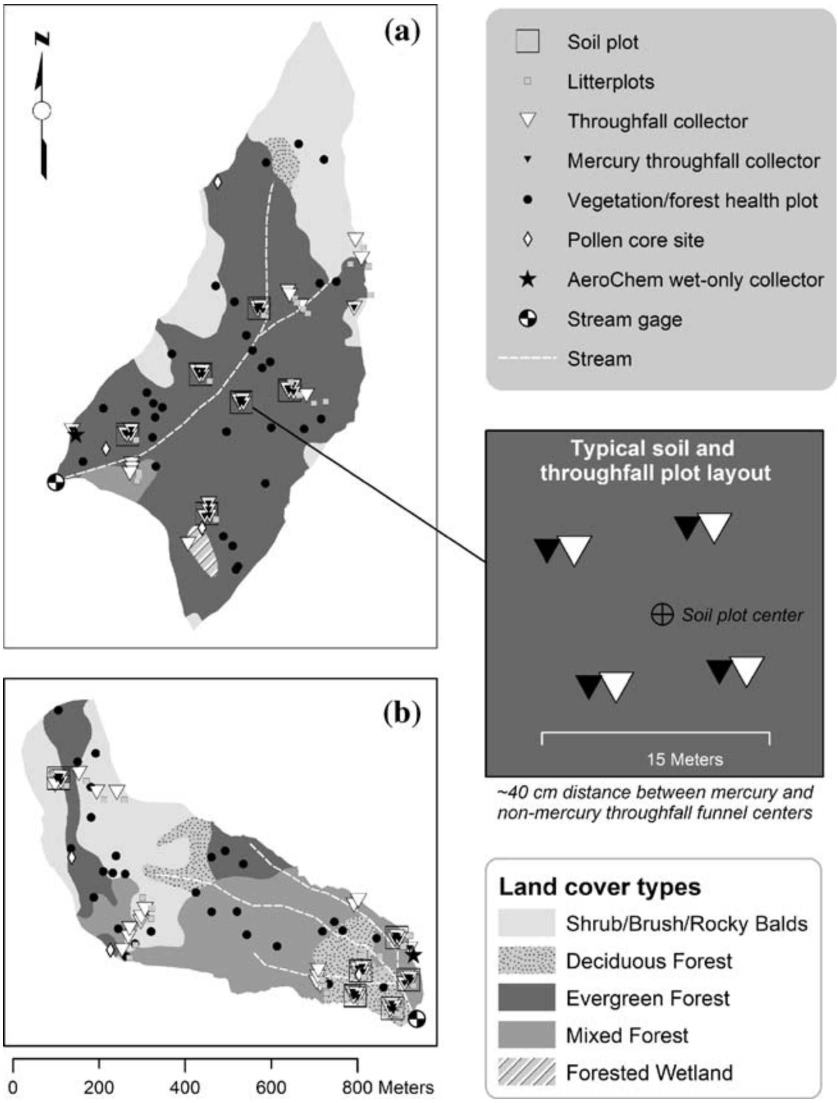


Figure 2 Study watersheds at Acadia National Park: (a) Hadlock Brook watershed, and (b) Cadillac Brook watershed with locations of throughfall and litter collectors, wet-only precipitation collectors, stream discharge gauges, soil plots, Hg soil assay sites, and paleoecological cores; National Park Service – USGS mapping project vegetation classification (Lubinski et al., 2003) for Cadillac Brook watershed and Hadlock Brook watershed is displayed in the *background*. The *inset at right* shows the typical soil and throughfall plot layout.

longitude 68°13′01.5″ (NAD 27) (Figure 2a) at an elevation of 122 m. The watershed area is 31.6 ha, extending from the summit of Cadillac Mountain (468 m) to 122 m above sea level. The average slope is 28%. The stream begins at about 440 m in a small valley, descending through relatively unvegetated exposed bedrock *via* multiple drainage channels that converge in the bottom third of the watershed.

The Hadlock watershed is drained by a headwater stream called Hadlock Brook. The gauging station for this unburned reference watershed is located at latitude 44°19′54.0″, longitude 68°16′47.5″ (NAD27) (Figure 2b) at 137 m above sea level. The watershed area is 47.2 ha, extending from the summit of Penobscot Mountain (380 m) to the gauging station. The average slope is 21%, and the watershed faces southwest. The stream headwaters are in a 0.7-ha woodland bog/fen, descending through a mature spruce-fir forest.

3 Approach

Our approach included intensive site characterization and determination of long-term chemical and hydrological budgets to serve as baseline indicators of status and changes in watershed function (e.g. Likens, Bormann, Pierce, Eaton, & Johnson, 1977). Schauffler et al. (in press) and Wiersma et al. (in press) describe the history, landscape, and ecosystem compartments of the research watersheds. Subsequent papers define Hg wet deposition, throughfall and litterfall (Johnson et al., in press) determine processes of Hg speciation, evaluate the status of N retention, estimate N-loading to selected estuaries (Nelson, Johnson, Kahl, Haines, & Fernandez, in press; Nielsen & Kahl, in press; Peckenham, Kahl, Nelson, Johnson, & Haines, in press) and document bioaccumulation of Hg in biota (Bank, Burgess, Evers, & Loftin, in press; Longcore et al., in press a, b).

The burned watershed is dominated by hardwood forest (Wiersma et al., in press) with lower organic matter in soils (Parker et al., 2002, 2001). The unburned watershed is dominated by conifer forest, and has older and thicker, more acidic soil organic horizons. One hypothesis of this research was that the ecosystem pools of carbon (C), N, and Hg were quantitatively reduced in the 1947 fire. This hypothesis was confirmed by Parker et al. (2002, 2001) and Amirbahman et al. (2004). The two contrasting watersheds provided a contrast of ecosystem function and response in forested watersheds at Acadia NP: Regenerating hardwoods vs. mature conifers. This characterization serves as the baseline for ongoing assessments of the status of ecosystems in the park.

The methods for the research components are described in the individual papers. In addition to this special issue, several papers based on research in these watersheds have been published elsewhere (Amirbahman et al., 2004; Campbell et al., 2004; Parker et al., 2002, 2001; Peckenham, Kahl, & Mower, 2003; Heath et al., 1992; Kahl et al., 1992, 1985).

4 Site Characterization

4.1 Climate

Acadia NP is located at the temperate and boreal transition zone in North America. Its coastal location and prominent topography result in frequent cloud and fog cover. Mean annual daytime temperature for Bar Harbor, Maine is 13 °C and mean annual nighttime temperature is 2 °C (Acadia NP Official Website, *www.nps.gov/acad*). The prevalent wind direction at Bar Harbor is 220° to 240° (west–southwest; G. Zielinski, personal communication). Average annual precipitation for Acadia NP is 122 cm as rain, plus 155 cm as snow near park headquarters (Table 1), for an annual average wet total of 137 cm during the past 20 years. Seasonal precipitation is relatively evenly distributed, with minimum values occurring in summer and the highest average monthly amount in November (Table 1).

The project period encompassed weather conditions including Hurricane Floyd (September, 1999), which deposited approximately 18 cm of rain in less than one week (NADP, 2004). The project also included severe drought in 2001, the driest year ever recorded in Maine. Statewide precipitation in 2001 totaled 75 cm, 33 cm below the annual statewide average of 108 cm and 3.5 cm below the previous record drought year, 1965. Streamflow reductions were greatest in August and September of 2001; dry conditions persisted into the 2002 winter when groundwater levels across the state reached record lows (Stewart, Caldwell, & Cloutier, 2003). By late spring 2002, rains had replenished surface water levels, although groundwater levels remained below

Table I Monthly mean temperature and precipitation for Acadia National Park, Maine, September 1982–November 2003

	Annual	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Mean temperature (°C)	7.2	−5.7	−4.4	−0.3	5.8	11.9	17.2	20.1	19.8	15.4	9.3	3.6	−2.5
Mean maximum temperature (°C)	13.2	−0.2	1.1	4.9	11.4	18.2	23.5	26.2	25.8	21.0	14.5	8.2	2.6
Mean minimum temperature (°C)	1.6	−11.2	−10.0	−5.4	0.3	5.6	10.9	13.9	13.7	9.7	4.2	−1.1	−7.5
Precipitation* (cm)	122	13.3	11.0	14.0	12.1	11.6	9.3	8.6	7.0	10.8	11.9	17.1	12.7
Snow (cm)	155	48.7	35.4	40.6	11.3	0.1	0.0	0.0	0.0	0.0	0.7	9.2	29.5

Annual temperature data: U.S. Climate Normals (NCDC, 2002).
Annual precipitation data: Acadia National Park official website (www.nps.gov/acad).
Monthly data: daily surface data collected by the National Climatic Data Center as obtained from G. Zielinski, Maine State Climatologist.
* Includes melted frozen precipitation

normal until spring, 2003 (Maine Drought Task Force, 2002a, 2002b).

4.2 Hydrology

The U.S. Geological Survey, in Augusta, Maine, monitors stream stage at the outflow of each watershed with a pressure transducer linked to the World Wide Web via satellite (http://waterdata.usgs.gov/me/nwis/current/?type=flow). Streams were instrumented at natural control locations, with stage recorders that recorded stage at five-minute intervals (Rantz et al., 1982). Stage was used to calculate streamflow in each stream based on stage relationships and the stream profile (Table 2). Stream discharge is greatest in spring and fall and typically low in summer (Figure 3) and winter.

4.3 Geology

The watersheds are underlain by the Cadillac Granite of Devonian age (Gilman, Chapman, Lowel, & Borns, 1988), a spatially uniform bedrock, which minimizes bedrock lithology as a source of environmental data variability. The highest points of both watersheds are exposed and glacially scoured bedrock as a result of continental glaciation that ended about 14,000 years ago (Lowell & Borns, 1988). Downslope migration of exfoliated granite bedrock has produced local talus slopes. Till underlies much of the lower slopes of both catchments. The gauging stations of each watershed are located above the post-glacial marine limit at *ca.* 80 masl. As a result, neither watershed contains the glacio-fluvial marine silts and clays (the Presumpscot Formation) that formed during de-glaciation (Heath, Kahl, Norton, & Brutsaert 1993).

4.4 Pedology

The predominant soil is a shallow-to-bedrock, stony Schoodic-rock outcrop-Lyman complex, derived from granite and schist-bearing tills. This classification includes extensive areas of exposed bedrock, along with areas where soils exist as thin deposits of gravelly sandy loam less than 15 cm deep

Table II Streamflow statistics for two gauged streams at Acadia National Park by calendar year. Streamwater stage monitoring began in April 1999 (Cadillac) and May 1999 (Hadlock), in liters per second (l/s)

Year	Annual mean, l/s (cfs)		Peakflow, l/s (cfs)	
	Cadillac Brook	Hadlock Brook	Cadillac Brook	Hadlock Brook
2000	8.2 (0.29)	15 (0.53)	425 (15)	821 (29)
2001	6.2 (0.22)	9.1 (0.32)	481 (17)	1388 (49)
2002	9.9 (0.35)	16 (0.55)	623 (22)	2011 (71)
2003	11 (0.40)	20 (0.69)	906 (32)	2209 (78)

Discharge in cubic feet per second (cfs), as reported by the U.S. Geological Survey, is indicated in parentheses. Discharge is calculated from established relationships with stage and stream channel morphometry. More information, instantaneous data, and streamflow statistics are available on the World Wide Web at http://waterdata.usgs.gov/me/nwis/. Study streams are identified as sites 01022835 (Cadillac Brook) and 01022860 (Hadlock Brook).

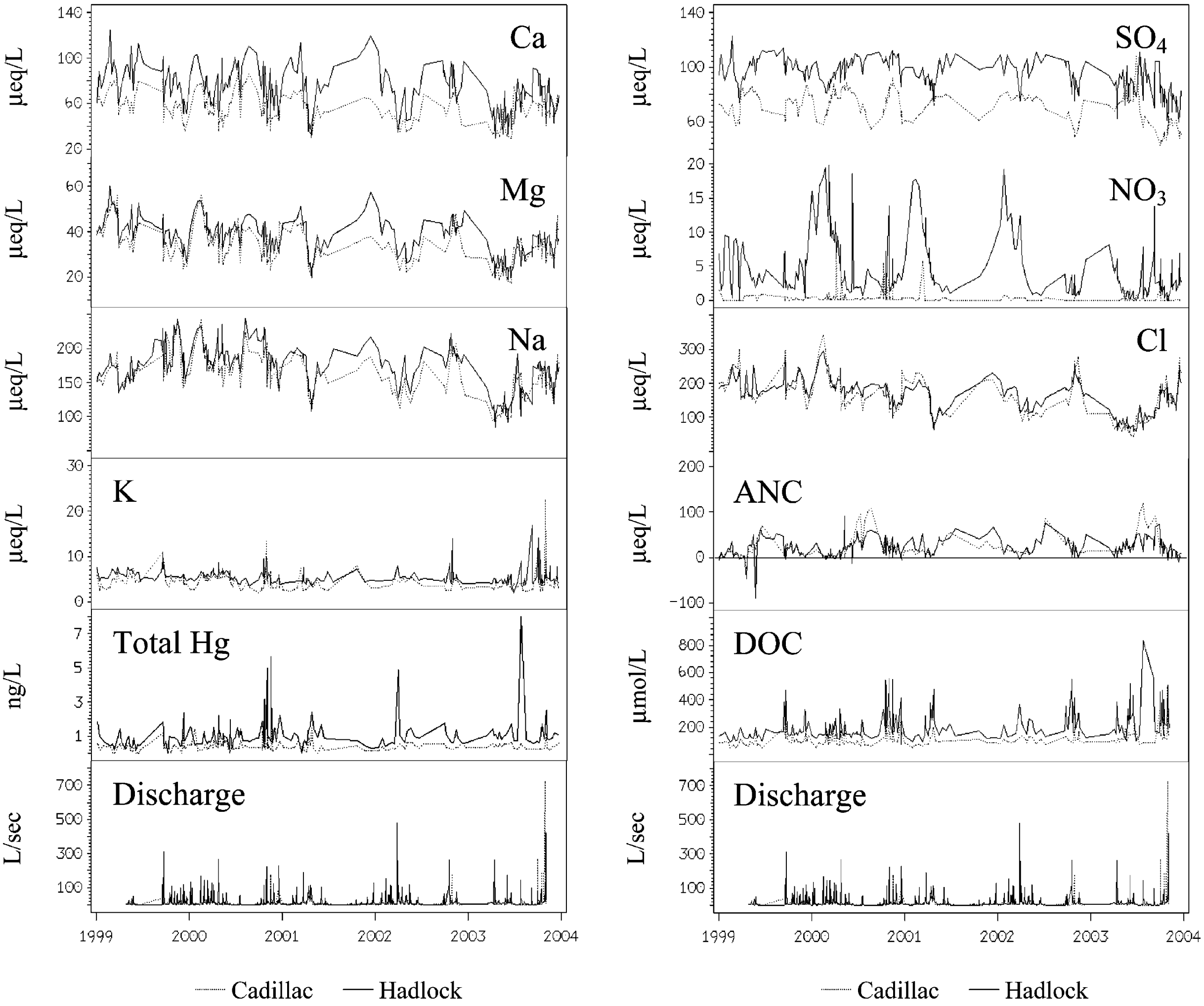


Figure 3 Concentrations of major ions, mercury (Hg), and dissolved organic carbon (DOC) in Cadillac and Hadlock brooks for 1999–2003. Stream discharge is shown at the *bottom* of each panel.

(Schoodic soils) and areas where soils form a black and reddish sandy loam less than 50 cm deep (Lyman soils). This soil complex is excessively well-drained, with slopes that range from flat to vertical for bare rock regions, and from 0% to 80% in areas with soil. On steep slopes these soils are usually droughty. Lyman and Schoodic soils are Spodosols – acidic forest soils characterized by an accumulation of iron (Fe), aluminum (Al), and organic matter in the B horizon.

Organic Lithic Borofolists are common on bedrock where mineral soil is absent. Depending on orientation and slope, Lithic Borofolists range from poorly to excessively drained. Organic soils are also associated with wetlands found in the study area. A range of poorly to well- (fibric to sapric) decomposed peats occur, with fibric peats occurring in bogs and as small blanket peats at higher elevation on bedrock. Sapric peats are associated with forest, shrub/fen and emergent shrub/fen communities (Calhoun et al., 1994). Generally, soils are shallow in depth which reduces infiltration and increases surface runoff in watersheds following storm events. Precipitation percolating through the shallow mineral or organic soils in the steeper upland watersheds becomes enriched in weak organic acids which contribute acidity to solutions. Aluminum, potentially toxic to terrestrial and aquatic organisms, is mobilized from soils and stream sediments in this acidic environment. When thicker mineral soils are present, acidic soil solutions in the upper soil profile are neutralized as they react with the constituents of the B/C horizons of mineral soils. As a result of these processes, the ephemeral upland streams described here have the lowest pH and exhibit greater episodic acidifications than perennial streams in the valley fill (Heath et al., 1993; Kahl et al., 1985).

The soils in the burned watershed had significantly higher sand percentages and lower clay and silt percentages in the upper 5 cm of the mineral soil than in the reference watershed (Parker et al., 2001). The wildfire and concomitant shift in vegetation type in the burned watershed were associated with significantly higher pH values in the upper 5 cm (pH = 4.5) compared to the reference soils (pH = 3.6). The burned watershed dominated by hardwoods had significantly lower forest-floor C and N concentrations than the reference watershed dominated by conifers (Hadlock; Parker et al., 2002, 2001). Forest-floor C and N pools were lower in the burned watershed compared with the reference (Figure 4).

4.5 Vegetation

Acadia NP is within the southern coastal range limit for spruce-fir forests of northern and eastern Maine and maritime Canada (Davis, 1966). Cadillac Brook watershed has three relatively distinct zones that reflect burn zones during the fire of 1947 (Figure 2a). Forest communities in the bottom portion of the watershed are heterogeneous, largely early successional types, dominated by American beech (*Fagus grandifolia*), striped maple (*Acer pensylvanicum*), and some mixed spruce-fir (*Picea rubens-Abies balsamea*) and hardwood stands. Twenty percent of the watershed is deciduous forest, located almost entirely in the lowest portion of the watershed that provides near-perennial stream flow. The center section of the watershed consists of steep slopes, with more open-canopy, sub-alpine scrub or shrub type communities, such as blueberry-ash shrub (*Vaccinium angustifolium-Sorbus americana*), red spruce-grey birch (*Picea rubens-Betula populifolia*) communities, and a large section that consists primarily of patchy, mixed stands. The top section of the watershed has a relatively large area of older spruce-fir forest and some mixed- to open-shrub summit communities.

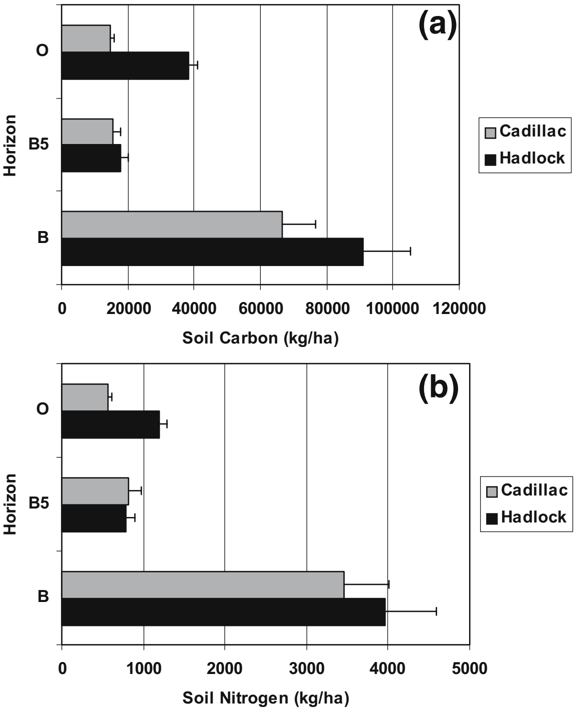


Figure 4 (a) Soil carbon content and (b) soil nitrogen content in three soil horizons at Cadillac and Hadlock Brook watersheds.

Approximately 70% of the Hadlock Brook watershed is spruce-fir forest (Figure 2b). Summit shrubland communities exist at higher elevations. There are woodland bogs at the top of the watershed and along the southern edge, which are primarily sphagnum (*Sphagnum* spp.), black spruce (*Picea mariana*), and various shrubs. A small patch of mature deciduous forest exists just south of the USGS stream gauging station. A land cover classification was provided for this study by the National Park Service, and analyzed for the two watersheds (Figure 2a and b; Lubinski, Hop, & Gawler, 2003).

4.6 Forest history and disturbance

Pollen records from Mount Desert Island and other coastal settings in Maine suggest that large-scale, stand-replacing disturbances of coastal forests were infrequent prior to European colonization of the region in the mid- and late-1700s. Fires are estimated to have burned at 300- to 500-year intervals during the two thousand years prior to European settlement (Schauffler, 1998; Patterson, personal communication). However, beginning in the late 1700s and extending through most of the 1800s, the majority of land along Maine's coast was cleared for timber and sheep grazing (McLane & McLane, 1989; Tolonen, 1983).

Reconstructions of vegetation and disturbance history were based on historical evidence, aerial photographs, and paleoecological analysis of locally derived pollen and charcoal preserved in small, partially forested bogs and forested wet hollows within the watersheds (Figure 2a and b). Pollen deposited each season in forested bogs and wet depressions is derived from sources dominantly within 50 m (Calcote, 1995; Sugita, 1995). In many settings, the stratigraphy of deposition is preserved, providing a decade-to-century scale record of changes in local dominant tree taxa. The studied wet depressions range from 1 m to several tens of meters in diameter with sediments between 20 and 50 cm deep. They are commonly covered by a mat of *Sphagnum* moss, or they may contain partially decomposed forest duff that holds standing water during wet seasons, classified as vernal pools. Although the hollows are dry at the surface in summer and fall, trapped drainage and the absorbent organic material keep the underlying sediment wet year-round, creating anaerobic conditions that preserve pollen.

5 Results and Discussion

5.1 Current status of watershed forests

USDA Forest Health Monitoring protocol indicators used in these watersheds indicated that species composition and stand structure differences are likely due to the wildfire (Wiersma et al., in press). Hadlock Brook watershed is dominated by older growth spruce (*Picea rubens*) and fir (*Abies balsamea*) and has no apparent record of fire. Cadillac Brook watershed contains a heterogeneous and patchy mix of hardwoods and conifers and is known to have burned in 1947.

Foliar N concentrations (Picea rubens) and foliar Al concentrations (Acer rubrum) were significantly higher in Hadlock Brook watershed, and foliar calcium (Ca) concentrations were lower in Hadlock Brook watershed for both species, compared to the Cadillac Brook watershed (Wiersma et al., in press). Foliar nutrient differences indicate more acidic soils in Hadlock and are suggestive of early stages of N saturation (Wiersma et al., in press). The results suggest that soil nutrient availability for plant uptake may differ between watersheds as a function of soil geochemical characteristics and microecological disturbances within each watershed.

5.2 Paleoecological reconstructions

Reconstructed forest stand histories based on pollen and charcoal analysis record major vegetation and disturbance differences in Hadlock and Cadillac watershed forests during the last several centuries (Schauffler et al., in press). The pollen data indicate that Hadlock Brook watershed has not burned or been significantly cleared for 500 years or more. Most of Cadillac Brook watershed burned in the wildfire in 1947, and likely burned several times in the 1800s. Cadillac Brook watershed has supported a heterogeneous forest for 200 years or more.

5.3 Characteristics of soils

Fifty years after wildfire, the burned watershed with hardwood regeneration (Cadillac) had significantly lower forest-floor C and N concentrations than the reference watershed dominated by conifers (Hadlock; Parker et al., 2002, 2001). Forest-floor C and N inventories (mass/area) were lower in the burned watershed compared with the reference (Figure 3), consistent with the project hypotheses related to the effects of the fire on soil chemistry. The soils in the unburned watershed contain more organic material and more Hg, also consistent with project hypotheses.

5.4 Precipitation inputs

Precipitation and throughfall volume and chemistry measurements (Tables 3a, b, and 4) and stream discharge were used to determine watershed water balance (Johnson et al., in press; Nelson, 2002). Differences in watershed and vegetation characteristics exert a significant control on the input of water and major ions to these watersheds because vegetation type influences throughfall chemical and hydrologic inputs (e.g., Houle, Oimet, Paquin, & LaFlamme, 1999; Lovett, Nolan, Driscoll, & Fahey, 1996; Cronan & Reiners, 1983). At coniferous sites, greater scavenging efficiency and year-round foliage resulted in greater sulfate (SO_4), chloride (Cl), and sodium (Na) concentrations and lower pH in throughfall at Hadlock Brook watershed (Nelson, 2002). Throughfall SO_4 was two- to three-times wet deposition, in the range reported for SO_4 for the nearby Bear Brook watershed in Maine (Rustad, Kahl, Norton, & Fernandez, 1994). Seasonal differences were impor-

Table III National Atmospheric Deposition Program (NADP) and Mercury Deposition Network (MDN) precipitation weighted mean (a) concentrations of major ions and total Hg, pH and specific conductance and (b) annual deposition of major ions and mercury, and precipitation depth for Acadia National Park – McFarland Hill (Site ME98), by calendar year

Year	Ca	Mg	K	Na µeq/l	NH_4	NO_3	Cl	SO_4	pH SU	Specific Conductance µs/cm	Total Hg ng/l
(a)											
1999	3.0	7.08	0.79	33.2	5.0	10.8	37.2	19.4	4.74	15.03	9.3
2000	4.0	8.64	0.97	40.5	7.2	15.0	42.0	25.4	4.61	19.4	9.8
2001	3.0	3.87	0.49	18.1	6.7	15.3	21.2	21.0	4.63	15.7	11.8
2002	2.5	6.75	0.74	31.3	6.7	10.5	36.7	18.9	4.8	14.52	10.2
(b)											

Year	Ca	Mg	K	Na	NH_4 kg/ha	NO_3	Inorg. N	Cl	SO_4	H+(Lab)	Precip. depth cm	Hg µg/m^2/year
1999*	0.82	1.155	0.416	10.264	1.16	8.97	2.92	17.79	12.51	0.25	134.34	7.9
2000*	1.08	1.351	0.489	11.988	1.67	12.01	4.01	19.14	15.74	0.32	128.63	8.6
2001	0.39	0.297	0.12	2.632	0.73	5.98	1.92	4.74	6.38	0.15	63.26	4.6
2002*	0.85	1.297	0.459	11.368	1.87	10.29	3.78	20.59	14.32	0.25	158.11	7.9

* NADP Data Completion Criterion 4 was <75% for these years; all other Data Completeness Criteria were acceptable.

tant, with highest dry deposition of major ions (inferred from throughfall:Wet deposition) in fall and summer. However, NADP data indicate that wet deposition inputs of Cl and Na were highest in winter, influenced by sea spray and marine storm tracks. Ongoing research is quantifying winter deposition in these study watersheds (Nelson, Weathers, Loftin, Johnson, & Kahl, 2005).

The NADP/MDN national wet deposition network is invaluable for regional spatial patterns and trends, but is not adequate for understanding total chemical deposition to specific landscapes. However, we

Table IV Descriptive chemistry for sixteen throughfall collections at Acadia National Park for the initial throughfall project period, August 1999 to November 2000

	Depth	SO_4	SO_4*	NO_3	Cl	H^+	Ca	Mg	K	Na	NH_4	EqpH
Cadillac												
N	437	441	441	441	441	440	442	442	442	442	441	440
	mm	←				µeq/L					→	
Min.	6	5	4	<0.5	8	0.1	<1.0	<0.8	<1.0	<0.9	<1.1	3.90
Max.	196	277	258	249	816	126	171	183	473	605	94	7.02
Median	46	53	42	16	58	27	16	20	22	49	2	4.58
Mean	52	60	51	24	93	34	26	29	44	74	7	4.72
St. dev.	27	42	37	30	104	27	25	30	67	76	11	0.61
Hadlock												
N	430	431	431	431	431	429	430	430	429	430	430	429
	mm	←				µeq/L					→	
Min.	3	4	3	<0.5	5	0.1	<1.0	<0.8	<1.0	4.3	<1.1	3.61
Max.	194	289	275	257	1120	245	119	200	338	848	107	6.85
Median	47	63	51	23	84	40	22	27	31	72	7	4.40
Mean	54	77	65	36	118	48	28	33	42	98	13	4.50
St. dev.	29	57	51	39	121	36	22	27	38	93	16	0.52

SO_4* is sea-salt corrected sulfate.

suggest that once the relationships are developed between nearby NADP stations and throughfall inputs to specific watersheds, NADP results can be used for total deposition estimates. However, the relationships may have changed (e.g., due to changes in legislative requirements for emissions). Thus, re-evaluation of the relationships with new data is prudent.

5.5 Hg input

Landscape aspect and vegetation type were the most influential factors affecting Hg deposition (mass/area) (Johnson et al., in press). Sites that face south to southwest received the highest Hg deposition, presumably due to the interception of continental contaminated air masses and prevailing wind direction. Sites with conifer vegetation received higher Hg deposition than other vegetation types because of the greater scavenging efficiency of the canopy. Because these sites at Acadia are largely forested and mountainous and located in the relatively polluted coastal air masses, they may be subject to greater dry deposition flux of Hg as compared to other rural areas in the Northeast (Miller et al., 2005).

Hg deposition was lower in Cadillac watershed (burned) than in Hadlock watershed (unburned) because regeneration after the fire was dominated by deciduous species rather than conifers. Litterfall contributed two- to five_times the amount of Hg to the forest floor as throughfall. (Sheehan, Fernandez, Kahl, & Amirbahman, 2006; Johnson, 2002). Our ongoing investigation of winter inputs suggests that snow throughfall has concentrations of total Hg similar to those measured in rain, but volatilization from the snowpack is a large potential loss (Nelson et al., 2005).

Methyl mercury (MeHg) deposition was not affected by aspect nor landscape factors. MeHg was typically 5% of total deposition (Johnson, 2002). We believe that this percentage may as much reflect the equilibrium of the methylation process in the collector as it does the actual deposition of MeHg. The low percentage suggests that it is neither cost-effective nor particularly instructive to measure MeHg concentrations in throughfall.

5.6 Stream chemistry and watershed mass balances

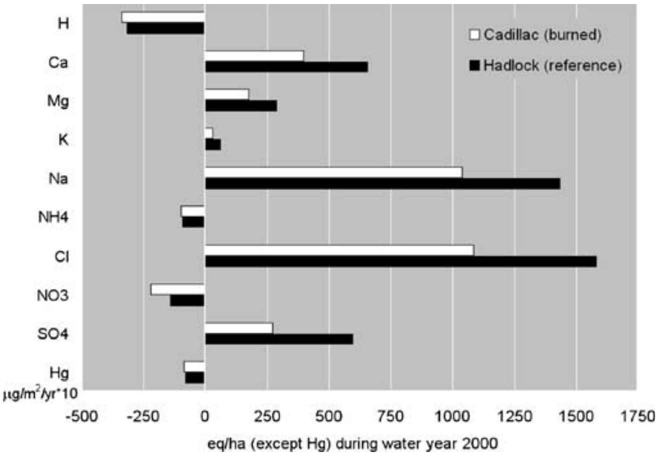
Discharge-weighted mean cation concentrations in streamwater were similar between watersheds (Table 5, Figure 3). Acid neutralizing capacity (ANC), pH, and Cl tended to be higher in Cadillac streamwater, while SO_4, nitrate (NO_3), dissolved organic carbon (DOC) and Hg were higher in Hadlock streamwater (Table 5, Figure 3). In both watersheds, H^+, ammonium (NH_4), NO_3, and Hg are retained in the soils and vegetation, although there are

Table V Discharge-weighted chemistry for Cadillac Brook (CB) and Hadlock Brook (HB) for calendar years 1999–2003

	1999		2000		2001		2002		2003	
	Cadillac	Hadlock	Cadillac	Hadlock	Cadillac	Hadlock	Cadillac	Hadlock	Cadillac	Hadlock
Equilibrated pH	5.86	5.48	5.89	5.45	5.98	5.51	6.11	5.18	5.63	5.54
ANC (μeq/l)	11.6	5.0	9.1	4.7	11.2	6.3	10.8	−0.4	7.0	6.7
Ca (μeq/l)	56.8 (50.4)	62.9 (55.9)	49.4 (43.3)	60.7 (53.9)	44.9 (40.0)	46.5 (41.8)	53.9 (47.2)	45.8 (40.4)	49.3 (43.1)	47.3 (43.0)
Mg (μeq/l)	33 (−6.5)	32.7 (−3.7)	32 (0.5)	36.7 (1.4)	28.2 (2.9)	29.6 (5.4)	35.1 (0.3)	31.9 (4.3)	30.3 (−1.6)	26.7 (4.6)
K (μeq/l)	6.4 (2.7)	7 (3.5)	5.7 (2.7)	6.7 (3.2)	3.4 (1.0)	4.7 (2.4)	4.7 (1.3)	7.5 (4.8)	10.4 (7.3)	5.7 (3.5)
Na (μeq/l)	179 (5.6)	185 (24.1)	166 (26.4)	183 (26.7)	142 (30.1)	145 (38.0)	165 (11.2)	148 (25.9)	144 (3.8)	128 (29.8)
Al (μmol/l)	2.8	6.9	4.3	10	3.8	9.2	3.7	11	7.0	11
NH_4 (μeq/l)	0.6	0.7	3.1	2.8	1.0	1.3	0.5	0.8	1.7	1.7
Si (μmol/l)	73	88	64	78	57	62	76	65	63	83
DOC (μmol/l)	136	256	194	323	131	302	151	372	315	415
Cl (μeq/l)	206	187	162	182	130	124	179	142	162	127
NO_3 (μeq/l)	0.3	5.1	0.8	7.5	0.1	5.9	0.2	9.3	0.1	2.1
SO_4 (μeq/l)	73.2 (51.9)	97.2 (77.9)	77.5 (60.7)	95.0 (76.2)	72 (58.5)	84.7 (71.9)	66.2 (47.7)	78.9 (64.3)	56.7 (40.0)	73.1 (60.0)
Total Hg (ng/l)	0.59	1.29	1.17	2.14	0.70	1.48	0.44	4.05	0.93	3.36
MeHg (ng/l)	–	–	0.04	0.04	0.04	0.04	–	–	–	–

Numbers in parentheses are sea-salt corrected by subtracting marine constituents in proportion to chloride.

Figure 5 Mass balance (streamwater export minus wet precipitation input) of major ions and mercury (Hg) in the study watersheds for water year 2000. Note that Hg has different units (μg/m^2/yr) and is multiplied by 10 for graphical display purposes. Positive values indicate net loss from the watershed and negative values indicate net retention. Wet precipitation data from the National Atmospheric Deposition Program (NADP) and Mercury Deposition Network (MDN) for site ME98.



relative differences in export related both to the fire and to the subsequent differences in loading in throughfall relating to the different canopies (Figure 6); Na, Cl, Ca, magnesium (Mg), potassium (K), and SO_4 were lost from the system for water year 2000 (Nelson et al., in press; Figure 5).

For some ions, differences related to vegetation and/or soils control the relative patterns of retention and release. For other ions, dry deposition is a major component of watershed inputs. For instance, Cl and SO_4 are considered relatively conservative with respect to canopy processes. Throughfall deposition of SO_4 was 2–3 times and Cl 2–10 times wet deposition in these watersheds, dependent on season (Nelson, 2002), consistent with other regional estimates (Rustad et al., 1994). Throughfall deposition of these conservative ions at conifer sites – such as Hadlock Brook watershed – tends to be greater than at deciduous sites – such as Cadillac Brook watershed (Nelson, 2002). The increased scavenging efficiency at Hadlock Brook watershed helps to explain differences in the mass balances between watersheds when only wet precipitation is considered (Figure 5). Estimates of dry deposition *via* throughfall are important for mass balances estimates. High deposition of marine aerosols impacts stream chemistry for well more than one hydrologic year.

Of the measured deposition of total Hg, 95% was retained or lost to volatilization by the Cadillac watershed, and 87% by the Hadlock watershed. These values are similar to those reported in other watershed studies. Grigal (2002) reviewed the Hg literature and found an average throughfall flux of 17 μg/m^2/yr, and an average streamwater flux of 1.7 μg/m^2/yr, yielding an average 90% retention and/or loss *via* volatilization. We did not attempt to quantify re-volatization of Hg from surfaces beneath the canopy (re-volatization of Hg from the canopy is irrelevant to our mass balances because it is not measured as either an input nor an output term).

5.7 Hg in soils

The 1947 fire caused significant impacts on the evolution of the Cadillac watershed, raising the soil pH, altering the vegetation, and reducing C and Hg soil pools (Amirbahman et al., 2004). Total Hg content was higher in Hadlock soils compared to Cadillac soils (Figure 7a). Soil pH was significantly higher in all soil horizons at Cadillac by about 0.4 pH units, compared to Hadlock soils. The results of Hg adsorption experiments indicated that the dissolved Hg concentration was controlled by the dissolved organic carbon concentrations. Adsorption isotherms suggest that Hg is more mobile in the unburned Hadlock watershed because of higher solubility of organic carbon in that watershed (Amirbahman et al., 2004). MeHg concentrations (the form of Hg that is concentrated in biota) were consistently higher in the burned Cadillac soils than in the unburned Hadlock soils (Figure 7b). The higher MeHg concentrations in Cadillac soils may reflect generally faster rates of microbial metabolism due to more rapid nutrient cycling in the deciduous forest.

MeHg concentrations were inversely proportional to total Hg in soils, suggesting that landscape factors such as soil pH, vegetation type, or land use history (e.g., fire) may be the determining factors for bioavailabilty

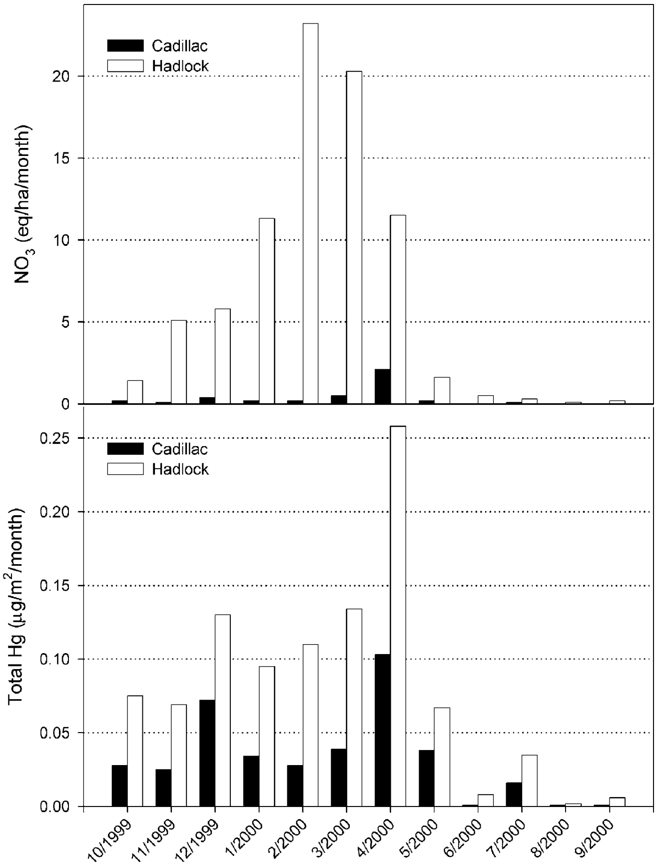


Figure 6 Monthly streamwater flux of nitrate (NO_3) and mercury (Hg) in Cadillac and Hadlock Brooks, water year 2000.

(Amirbahman et al., 2004). This lack of direct relationship between Hg and MeHg suggests that ‘recovery’ from Hg bioaccumulation is not limited by the total Hg reservoir because the labile pool of Hg is not defined by the total reservoir. Because the natural landscape is a mosaic of various land covers, the information from these watersheds may eventually provide the basis for determining the factors that control Hg bioavailability and the timing of a decline in Hg bioaccumulation and health consumption advisories.

5.7.1 Hg contamination in amphibians

Hg contamination is an important environmental stressor for amphibian populations in Acadia NP, The levels reported for northern two-lined salamander (*Eurycea bislineata*) larvae are considered to be elevated (Bank, Loftin, & Jung, 2005). The full extent of the influence of Hg contamination on *Eurycea bislineata* larvae is unknown, although it is likely that these stream salamanders are at considerable risk of reduced population performance from the effects of multiple stressors (Sparling, Linder, & Bishop, 2000; Semlitsch, 2003), including Hg (Lowe & Bolger, 2002; Barr & Babbitt, 2002; Bank, 2005; Bank et al., in press). Hg concentrations in *Eurycea bislineata* (a carnivore) were, on average, three times higher than green frog (*Rana clamitans*) (a grazer) and bullfrog (*Rana catesbeiana*) (a grazer) tadpoles (~1 year of age). These Hg differences were likely attributed to different diets (Bank, 2005; Bank et al., in press).

Hg risk assessment is affected by watershed location (Bank, 2005). For example, *Eurycea bislineata* larvae collected from Breakneck Brook (a low elevation stream at Acadia NP) contained low to moderate levels of Hg; although the stream’s receiving wetland had the highest observed methylation efficiency rate (Bank, 2005).

Although Hg bioaccumulation in amphibians from Acadia NP has been quantified, the mechanism between population performance and Hg exposure is unknown (Bank, 2005). Other contaminants, or other abiotic

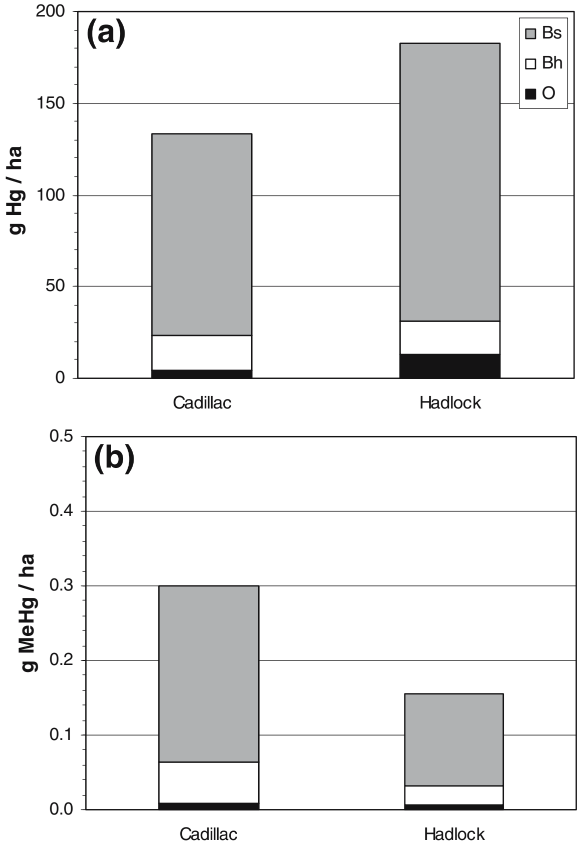


Figure 7 (a) Total Hg and (b) MeHg contents in the soils of Hadlock and Cadillac watersheds. (Amirbahman et al., 2004; used with permission).

conditions (i.e., drought, warm water temperatures) potentially decrease overall fitness in amphibians (Bank, 2005). Next steps in the evaluation of bioaccumulation might include toxicological studies that use MeHg exposure regimes that reflect natural surface water conditions, and examine the long-term and sublethal effects of this contaminant with the synergistic or cumulative effects of stress from the presence of predators and/or predator cues (Relyea & Mills, 2001).

6 Conclusions and Major Implications

The paired-gauged watershed project design at Acadia NP is based on a natural experiment caused by the major wildfire in 1947 in one of the watersheds. The two similar watersheds, but with differing fire histories, have been extensively studied from 1998 to 2004 with previous supporting data collected during 1982–1984 and 1989–1991. The results suggest the following conclusions about control of stream water chemistry by landscape factors:

1) Quantification of dry deposition in different terrain and in different vegetation is necessary to accurately quantify total deposition of water and solutes to the landscape. The NADP national wet deposition network is invaluable for regional spatial patterns and trends, but is not adequate for understanding total deposition to specific landscapes.
2) Differences in deposition of solutes and contaminants are influenced largely by vegetation species and stand age, and secondarily by aspect. The burned watershed has significantly lower deposition and accumulation of Hg, as an inferred result of the fire, and a result of lacking forest cover for deposition interception for many years after the fire. Moreover, the regenerating forest cover was deciduous, which is less effective at scavenging solutes from the atmosphere than conifers.
3) Identifying past disturbances is essential for evaluating modern patterns of forest cover differences in soil chemistry, and differences in runoff chemistry. For example, the 1947 fire is inferred to have reduced soil C, N, and Hg, compared to the reference site. The fire also promoted the regeneration of hardwood forest cover. These differences, in turn, exert control over runoff chemistry, especially for Hg and N.
4) The effects of some disturbances apparently extend for decades or centuries, at least influencing N and Hg dynamics at the watershed scale. At Cone Pond in NH, a severe fire in 1820 disrupted N dynamics to the extent that present conditions are still unfavorable for nitrification (Hornbeck & Lawrence, 1997). The results at Acadia NP support these conclusions for the more recent fire.

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