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Biological Mercury Hotspots in the Northeastern United States and Southeastern Canada

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Biological mercury (Hg) hotspots were identified in the northeastern United States and southeastern Canada using a data set of biotic Hg concentrations. Eight layers representing three major taxa and more than 7300 observations were used to locate five biological Hg hotspots and nine areas of concern. The yellow perch and common loon were chosen as indicator species for the human and ecological effects of Hg, respectively. Biological Hg hotspots receive elevated atmospheric Hg deposition, have high landscape sensitivity, and/or experience large reservoir fluctuations. In the Merrimack River watershed, local Hg emissions are linked to elevated local deposition and high Hg concentrations in biota. Time series data for this region suggest that reductions in Hg emissions from local sources can lead to rapid reductions of Hg in biota. An enhanced Hg monitoring network is needed to further document areas of high deposition, biological hotspots, and the response to emissions reductions and other mitigation strategies.

Keywords: biological mercury hotspots, mercury sources, common loon, mercury monitoring, yellow perch

Mercury (Hg) is a local, regional, and global pollutant that affects fish, wildlife, and human health. Recently, 71 scientists from New England, New York, and eastern Canada compiled and analyzed more than 30,000 observations of Hg levels in biota, including 40 fish and 44 wildlife species (Evers and Clair 2005). The resulting database is a powerful tool to quantify spatial patterns of Hg in biota across the northeastern United States and southeastern Canada (referred to here collectively as the Northeast).

We focus on biological Hg hotspots in the Northeast because the spatial heterogeneity of Hg deposition and methylmercury (MeHg) in biota is an issue of international concern. For example, fish consumption advisories concerning Hg contamination exist in each of the eastern Canadian provinces and 44 states in the United States, including all states within our study area. This pattern of advisories demonstrates that Hg contamination is widespread.

Current state and national policies to control Hg emissions from point sources include the consideration of cap-and-trade options. Trading allows the providers of coal-fired electric utilities to purchase pollution credits in order to meet a national cap, rather than requiring reduced emissions for all facilities. Thus, trading has the potential to lead to static or increased emissions in some areas of the United States, which may produce changes in Hg deposition, cycling, and biological uptake. Increased deposition near areas that are highly sensitive to Hg or already affected by Hg deposition could increase Hg contamination in fish, and may increase the risk to people and wildlife that consume fish. An understanding of the mechanisms contributing to biological Hg hotspots is important when Hg trading policies are considered.

Given the growing scientific evidence of Hg contamination (Evers et al. 2005, Kamman et al. 2005) and the public policy interest in identifying specific geographic areas that are disproportionately elevated in Hg, it is important to develop a common definition for the term "biological mercury hotspot." We define a biological Hg hotspot as a location on

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the landscape that, compared to the surrounding landscape, is characterized by elevated concentrations of Hg in biota (e.g., fish, birds, mammals) that exceed established human or wildlife health criteria as determined by a statistically adequate sample size.

There are important considerations in defining and identifying biological Hg hotspots. The sources of Hg contamination are not easily differentiated in ecosystems. Therefore, the identification of biological Hg hotspots, based on the effects of Hg pollution, should not be constrained to those areas where high Hg concentrations can be attributed to a single source or sector. Rather, multiple sources from multiple sectors can contribute to a hotspot, and as a result we do not limit the definition of a hotspot to a single source or sector.

Biological Hg hotspots can occur in diverse locations across the landscape, and are not restricted to areas of high Hg deposition. Landscapes have critical characteristics that influence Hg transport to surface waters, the methylation of ionic Hg, and the bioaccumulation of MeHg in biota, thereby modifying sensitivity to Hg inputs (Driscoll et al. 2007). These characteristics include land cover, oxidation–reduction conditions, hydrologic flow paths, and nutrient loading. Modifications of the landscape, such as changes in land disturbance, can alter the supply of Hg to downstream aquatic ecosystems.

To further define and identify biological Hg hotspots in the Northeast, we analyzed the extensive existing database developed for Hg in fish and wildlife (Evers and Clair 2005). Although these summarized data are comprehensive, some areas within the Northeast remain poorly characterized for Hg, and additional biological Hg hotspots may exist. We also hypothesize mechanisms that contribute to the formation of the biological Hg hotspots. We use a case study of the lower and middle Merrimack River watershed, located in northeastern Massachusetts and southern New Hampshire, to estimate the impact of local emissions and assess the extent to which biota may respond to changes in local Hg emissions and deposition. Finally, we describe the need for increased long-term monitoring, process-level science, and improved Hg models to fill data gaps critical to locating hotspots, tracking changes in Hg levels, following emission controls, and assessing the impact of policy decisions.

Study area and methods

Regional databases of Hg in biota were gathered during a four-year effort by the Northeastern Ecosystem Research Cooperative (NERC) and published in a series of papers describing the distribution of Hg and MeHg in northeastern North America (Evers and Clair 2005). We used a subset of 7311 observations for seven species, in three major taxonomic groups that represent eight data layers, to quantify the spatial heterogeneity in tissue Hg concentrations (table 1, figure 1). Spatial data for Hg concentrations in biota were used to identify areas where the tissue burdens of Hg exceeded levels known to result in adverse effects.

The primary data layers for Hg concentrations in fillets of yellow perch (*Perca flavescens*) and in the blood and eggs of the common loon (*Gavia immer*) were used to locate biological Hg hotspots. Secondary data layers for whole-fish analysis of yellow perch and for Hg concentrations in largemouth bass (*Micropterus salmoides*), brook trout (*Salvelinus fontinalis*), bald eagle (*Haliaeetus leucocephalus*), river otter (*Lontra canadensis*), and mink (*Mustela vison*) were used to locate areas of concern. All data are presented in terms of wet weight (ww) unless otherwise described as fresh weight (fw), which includes biotic material such as feathers and fur. All means are arithmetic. We also used data on surface water chemistry and land cover to evaluate the factors contributing to the spatial heterogeneity of Hg in biota.

Data preparation. To develop a common measure across the data set, we calculated standardized conversions of Hg concentrations for different tissue types in yellow perch and common loons. We used the Hg concentrations of standard-length (20-cm) yellow perch (Kamman et al. 2005), relying on whole-fish concentrations as an indicator of ecological risk and on fillet concentrations as an indicator of human health risk. Where only whole-fish concentrations were available, we converted these values to fillet equivalents using a regression of average-age mean Hg concentrations for fillets against mean whole-fish Hg concentrations developed from a set of statistically randomized lakes (fillet Hg = [1.63 • whole-body Hg] + 0.06; $F_{41,1} = 46.6$, $p < 0.001$, $r^2 = 0.54$; Kamman et al. 2004). This regression is similar to one performed for Hg levels in fish analyzed from lakes in the western United States (Peterson et al. 2005). Similarly, Hg values for the eggs of the common loon were converted to equivalent values for the blood of the adult female loon (female loon blood Hg = [1.55 • loon egg Hg] + 0.22; $r^2 = 0.79$; Evers et al. 2003).

Impact thresholds. The effects of MeHg exposure are difficult to measure. The US Environmental Protection Agency (USEPA) bases human health criteria on consumption models. We used the USEPA suggested advisory level of 0.30 μg Hg per g (ww) in fish muscle tissue to identify biological Hg hotspots of human health concern (USEPA 2001). This level triggers advisories of one or fewer fish meals per month for sensitive groups, such as pregnant women, women of childbearing years, and children less than 12 years of age.

To identify biological Hg hotspots that pose risks to ecological health, we used accepted thresholds for adverse effects from Hg in several wildlife species, as derived from the literature. One of the more comprehensive data sets for assessing the adverse effects of Hg on wildlife is from studies on the common loon.

Blood and egg Hg concentrations have been linked to demonstrated adverse effects in the common loon. The level of 3.0 μg Hg per g ww, which was developed *in situ,* is based on (a) physiological effects, such as higher average corticosterone levels and increased developmental instability (Evers et al. 2004); (b) behavioral effects, such as lethargy in chicks

Table 1. Summary statistics of biological data layers for mercury (Hg) concentrations in fish and wildlife (μg per g) in the northeastern United States and southeastern Canada.

Category/species	Sample size	Data layer designation	Hg concentrations: Mean ± standard deviation	Hg concentrations: Range	Hg level of concern (tissue type)	Percentage of samples with concentrations > level of concern
Human health						
Yellow perch[a]	4089	Primary	0.39 ± 0.49	< 0.05–5.24	0.30 (fillet)	50
Largemouth bass[b]	934	Secondary	0.54 ± 0.35	< 0.05–2.66	0.30 (fillet)	75
Ecological health						
Brook trout	319	Secondary	0.31 ± 0.28	< 0.05–2.07	0.16 (whole fish)	75
Yellow perch[c]	(841)[d]	Secondary	0.23 ± 0.35	< 0.05–3.18	0.16 (whole fish)	48
Common loon[e]	1546	Primary	1.74 ± 1.20	0.11–14.20	3.0 (blood)	11
Bald eagle	217	Secondary	0.52 ± 0.20	0.08–1.27	1.0 (blood)	6
Mink	126	Secondary	19.50 ± 12.1	2.80–68.50	30.0 (fur)	11
River otter	80	Secondary	20.20 ± 9.30	1.14–37.80	30.0 (fur)	15

Note: All data are in wet weight except for fur, which is on a fresh-weight basis.

a. Fillet Hg in yellow perch is based on individuals with a standardized length of 20 cm.

b. Fillet Hg in largemouth bass is based on individuals with a standardized length of 36 cm.

c. Whole-fish Hg in yellow perch is based on individuals with a standardized length of 13 cm. Whole-fish Hg for yellow perch was converted to fillet Hg.

d. The sample population of 841 yellow perch examined for whole-fish Hg is included with the 4089 fillets (i.e., the total number of all biotic data layers does not double-count yellow perch).

e. Egg Hg for the common loon was converted to the adult blood equivalent.

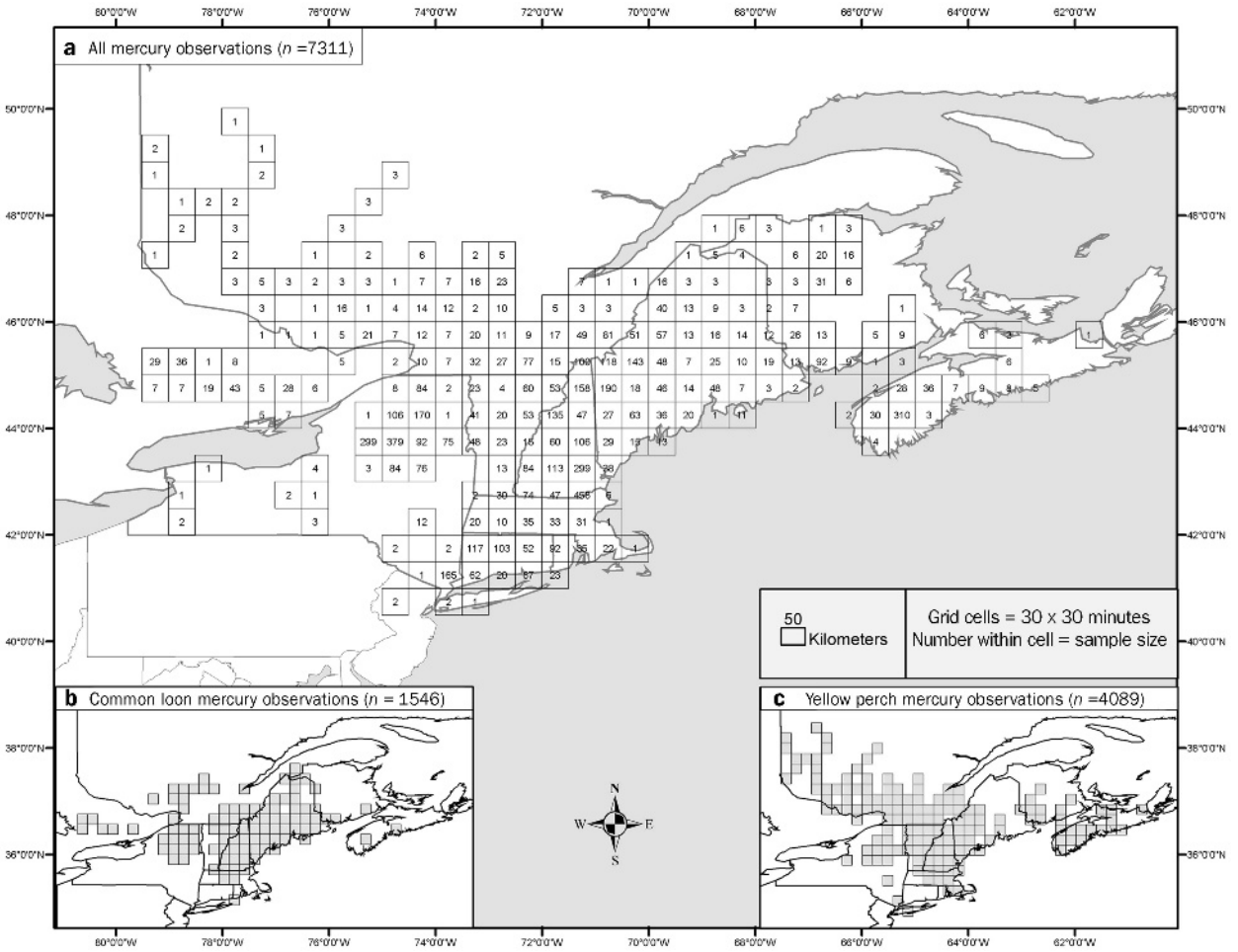

Figure 1. (a) Distribution of biotic mercury (Hg) observations across the northeastern United States and southeastern Canada, and specific distribution of Hg observations for (b) the common loon and (c) yellow perch.

(Nocera and Taylor 1998) and abnormal incubation patterns (Evers et al. 2004); and (c) reproductive effects, such as fewer fledged young from a territorial pair (Barr 1986, Burgess et al. 1998, Evers et al. 2004). Based on this level of concern and on estimates of nesting success, productivity levels can be modeled to determine population sinks and sources for loons (Evers et al. 2004, Nacci et al. 2005). Stage-based models indicate that when more than 25% of a loon population produces 40% fewer fledged young, a population sink occurs.

A second group of human health and ecological indicators was selected to identify areas of concern in the region. These secondary indicators are largemouth bass, brook trout, yellow perch (whole-fish concentrations), bald eagle, river otter, and mink. In this analysis, a whole-fish Hg concentration of 0.16 μg per g (ww) for yellow perch and brook trout was used as an adverse-effect level for piscivores, reflecting the documented risk to loons foraging on fish with whole-body concentrations above this level (Evers et al. 2004, Seiler et al. 2004). A blood Hg concentration of 1.0 μg per g (ww) in bald eaglets was selected as the adverse-effect level that is related to significant negative effects on reproductive success in Maine (DeSorbo and Evers 2006). Because of uncertainties in the accepted level of adverse effects for furbearers, a value of 30 μg per g (fw) in fur was used for river otter and mink, rather than the 20 μg per g (fw) used in some studies (Thompson 1996).

Spatial analysis. The biotic Hg data layers were plotted using a 30' × 30' polygon grid interval (or 0.5° × 0.5° grid) to summarize the data and provide a relevant geographic coverage using GIS (geographic information system) techniques. The grid size was selected on the basis of our understanding of the NERC data, reflecting the trade-offs between spatial detail and the number of sites with biotic Hg data within a cell. Grid interval size varied according to latitudinal and longitudinal position but averaged approximately 2200 to 2300 km^2. We employed power analyses to determine the minimum acceptable number of yellow perch and loon samples needed within any given grid cell to maintain a likelihood of detecting biological threshold limits ($p \pm 0.01$ and $\beta = 0.80$ for yellow perch; $p \pm 0.001$ and $\beta = 0.95$ for common loons). These analyses indicate that a minimum sample size of 10 independent sites per grid cell for yellow perch, and 14 for common loons, is needed to characterize Hg concentrations accurately.

The perch data were queried to display standardized Hg concentrations of at least 0.30 μg per g (ww), with each data point representing an independent sampling site. These data were joined to a 30' × 30' polygon grid, and the resulting grid was queried for a sample size of at least 10. We verified this analysis by converting the entire NERC fish Hg data set of more than 15,000 observations (Kamman et al. 2005) to a data set for standard-length yellow perch using the model created by Wente (2004). These data showed agreement with the spatial analysis, demonstrating that the yellow perch database was a robust indicator for biological Hg hotspots. The loon data were joined to a 30' × 30' polygon grid. These data were then queried to display (a) cells with a sample size of at least 14 and (b) cells with at least 25% of the data showing 3.0 or more μg Hg per g (ww).

For those grid cells that did not meet the sample size requirements for yellow perch and common loons, we examined Hg concentrations in the six secondary biotic data layers (table 1). Independent of sample size, those grid cells that had two or more biotic data layers with mean Hg concentrations that exceeded associated adverse-effect levels were identified as areas of concern. Locations of major historic and current Hg discharges at industrial sites (e.g., mercury-cell based chlor-alkali facilities, textile plants) were also identified (figure 2).

To help ascertain possible mechanisms responsible for biological Hg hotspots, we examined land-use and water-chemistry attributes of water bodies within each grid cell based on standardized data sets, such as those available through the USEPA Environmental Monitoring and Assessment Programs (both national and regional versions). Land-use percentages for forested, wetland, and agricultural areas were extracted from the US Geological Survey's National Land Cover Dataset, while total phosphorus (TP), dissolved organic carbon (DOC), pH, and acid neutralizing capacity (ANC) in surface waters were summarized in relation to sensitivity thresholds established by Driscoll and colleagues (2007) using NERC data (TP < 30 μg per L, DOC < 4 mg carbon [C] per L, pH < 6, and ANC < 100 microequivalents [μeq] per L).

Spatial analysis based on multiple data layers of mercury

Mercury concentrations within the two primary and six secondary data layers were available for 234 grid cells covering an area of 513,471 km^2. Five biological Hg hotspots were identified in the study region, based on the two primary data layers (yellow perch and common loon). A total of 663 sites, with 4089 measurements of yellow perch Hg concentrations, were analyzed for 147 grid cells representing an area of 336,723 km^2. A total of 101 grid cells (approximately 70% of the study region) had mean Hg concentrations for yellow perch that exceeded the USEPA human health criterion at one or more sites. Nine grid cells had mean Hg concentrations for yellow perch at 10 or more independent sites that exceeded the criterion, resulting in five biological Hg hotspots with a total area of 20,616 km^2 (figure 2).

In general, where standard-length yellow perch exhibited Hg concentrations in excess of 0.30 μg per g, other larger, more predatory, and more sought-after game fish, such as largemouth bass, also had elevated Hg concentrations. Mean perch Hg concentrations were highest in the western Adirondack Mountains of New York (H1a) and the middle part of the Merrimack River watershed in New Hampshire (H3a), followed by the lower part of the Merrimack River watershed

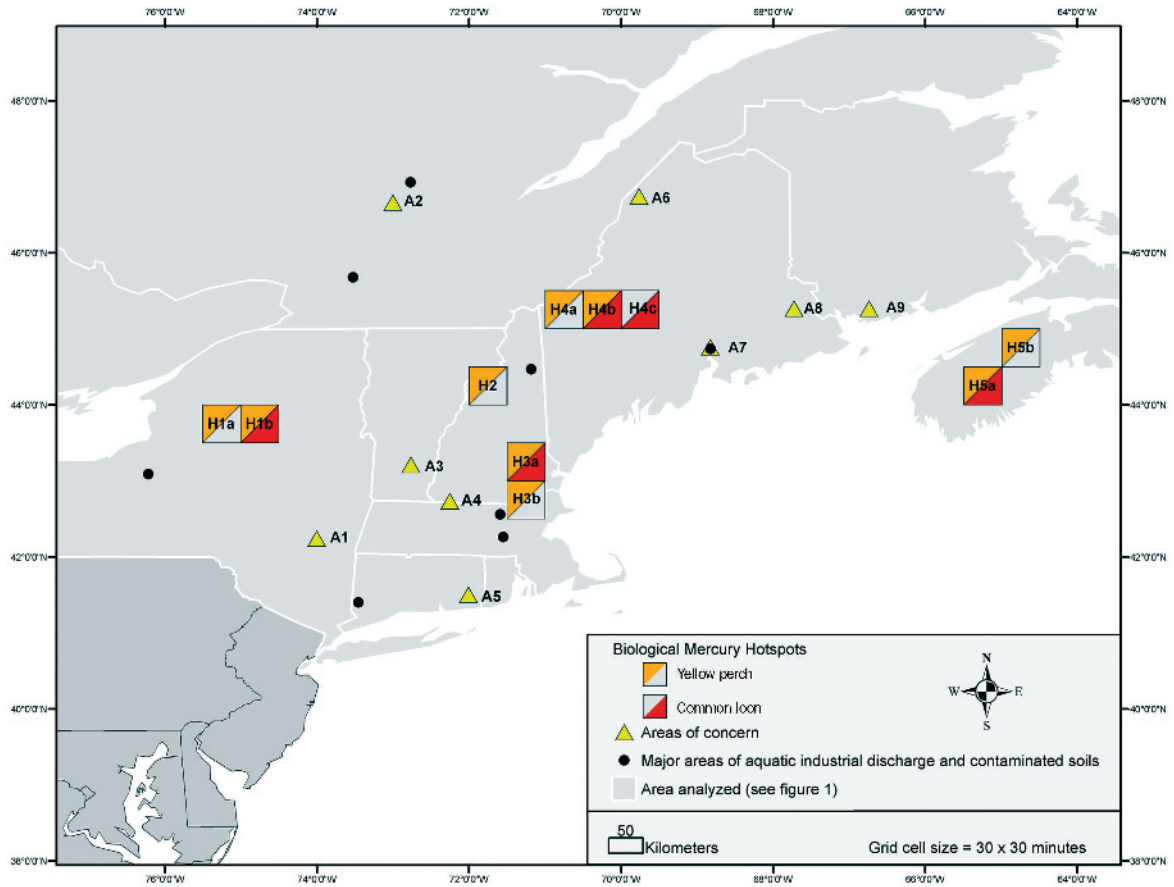

Figure 2. Distribution of biological mercury hotspots (H1a–H5b) and areas of concern (A1–A9). Areas of concern: A1, Catskill Mountains, New York; A2, LaMauricie region, Quebec, Canada; A3, Deerfield River, Vermont; A4, north-central Massachusetts; A5, lower Thames River, Connecticut; A6, upper St. John River, Maine; A7, lower Penobscot River, Maine; A8, Downeast region, Maine; A9, Lepreau region, New Brunswick, Canada. Hotspots: H1a, western Adirondack Mountains, New York; H1b, central Adirondack Mountains, New York; H2, upper Connecticut River, New Hampshire and Vermont; H3a, middle Merrimack River, New Hampshire; H3b, lower Merrimack River, Massachusetts and New Hampshire; H4a, upper Androscoggin River, Maine and New Hampshire; H4b, western upper Kennebec River, Maine; H4c, eastern upper Kennebec River, Maine; H5a, Kejimkujik National Park, Nova Scotia, Canada; H5b, central Nova Scotia.

in Massachusetts (H3b), the central Adirondack Mountains (H1b), and Nova Scotia, Canada (H5a and H5b).

Of the 1546 loons sampled in 102 grids, representing an area of 226,503 km^2, 33 grid cells met the minimum sample size requirement. Biological Hg hotspots associated with loons occur in five grid cells within four of the biological hotspots, covering an area of 11,027 km^2 (table 2, figure 2). In these grid cells, 25% to 93% of the sampled loon population had Hg concentrations above adverse-effect levels. In these biological Hg hotspots, common loons therefore are most likely to experience significant adverse effects at the population level. Mean loon blood Hg concentrations were highest in the upper Kennebec River region of Maine (H4b and H4c) and in Kejimkujik National Park in Nova Scotia (H5a).

Nine areas of concern were identified based on the six secondary data layers. These areas include the Catskill Mountains, New York (A1); the LaMauricie region, Quebec, Canada (A2); Deerfield River, Vermont (A3); north-central Massachusetts (A4); the lower Thames River, Connecticut (A5); the upper St. John River, Maine (A6); the lower Penobscot River, Maine (A7); the Downeast region, Maine (A8); and the Lepreau region, New Brunswick, Canada (A9; figure 2).

Identification and interpretation of biological mercury hotspots

To understand the mechanisms that may contribute to these biological Hg hotspots, it is necessary to consider Hg sources, atmospheric processes, landscape characteristics, and human disturbance to the landscape (figure 3). We hypothesize that three factors amplify the effects of regional and global atmospheric Hg emissions and deposition and are the likely major mechanisms contributing to the biological Hg hotspots identified here: (1) elevated atmospheric Hg deposition from local sources, (2) high landscape sensitivity, and (3) large

Table 2. Summary of data layers for mercury (Hg) concentrations (µg per g, wet weight) in yellow perch and common loons for each biological Hg hotspot in the Northeast.

Biological Hg hotspot	State/province	Hg concentrations: mean ± standard deviation (*n*, range)		Percentage of loons with Hg concentrations > level of concern
		Yellow perch	Common loon	
H1a: Adirondack Mountains (west)	New York	0.73 ± 0.15 (10, 0.57–0.96)	1.5 ± 0.3 (6, 1.1–2.1)	0
H1b: Adirondack Mountains (central)	New York	0.54 ± 0.15 (12, 0.39–0.80)	2.0 ± 1.2 (44, 0.3–4.1)	25
H2: Upper Connecticut River	New Hampshire, Vermont	0.35 ± 0.13 (17, 0.14–0.58)	1.1 ± 0.7 (45, 0.1–2.9)	0
H3a: Merrimack River (middle)	New Hampshire	0.78 ± 0.99 (38, 0.05–5.03)	2.6 ± 1.8 (39, 0.7–7.1)	28
H3b: Merrimack River (lower)[a]	Massachusetts, New Hampshire	0.65 ± 0.78 (17, 0.23–3.81)	NA (no loons sampled)	NA
H4a: Upper Androscoggin River	Maine, New Hampshire	0.44 ± 0.27 (12, 0.21–1.25)	1.9 ± 1.0 (92, 0.15–5.47)	14
H4b: Upper Kennebec River (west)	Maine	0.40 ± 0.09 (11, 0.24–0.52)	3.1 ± 2.1 (77, 0.6–14.2)	43
H4c: Upper Kennebec River (east)	Maine	0.38 ± 0.30 (3, 0.14–0.72)	2.2 ± 1.0 (31, 0.6–4.1)	26
H5a: Kejimkujik National Park	Nova Scotia	0.50 ± 0.18 (27, 0.14–0.85)	5.5 ± 1.4 (14, 2.9–7.8)	93
H5b: Central Nova Scotia	Nova Scotia	0.58 ± 0.86 (16, 0.14–3.79)	NA (no loons sampled)	NA

NA, not applicable.
a. *Source:* Hutcheson et al. 2003.

water-level manipulations (table 3). Atmospheric deposition is the major Hg input to the region (Fitzgerald et al. 1998), and both local sources and long-range transport of Hg are likely to be important in the formation of biological Hg hotspots. Although biological Hg hotspots may also originate from local sources of Hg-contaminated soils and waters, the impacts from these sources are less pervasive, and we therefore focus here on biological Hg hotspots originating from atmospheric deposition.

Mercury is emitted to the atmosphere from a variety of sources. The largest single source in the United States is coal-fired electric utilities. Mercury can be deposited locally or travel great distances, depending mostly on its oxidation state (i.e., 0, +2). Mercury is present in the atmosphere in several forms: elemental Hg, or Hg^0; gaseous divalent Hg, or Hg(II); and particulate Hg, or Hg(p). Elemental Hg has an approximately 0.5- to 2-year residence time in the atmosphere, so it constitutes the majority of airborne Hg. Gaseous divalent Hg and Hg(p) are generally deposited much more rapidly than

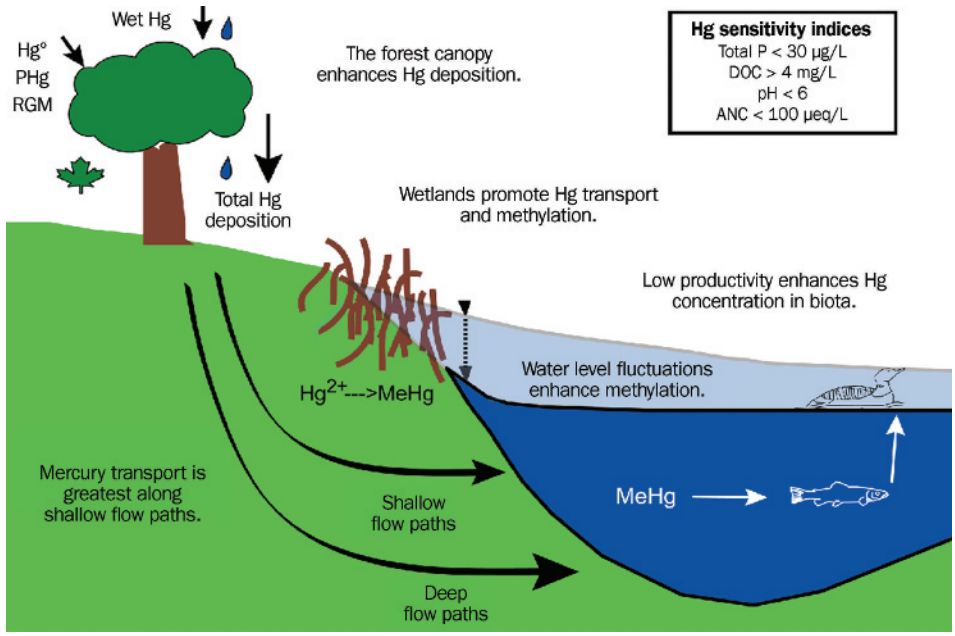

Figure 3. Conceptual figure illustrating important processes controlling the sensitivity of forest and linked aquatic ecosystems to atmospheric mercury (Hg) deposition and artificial water level regulation. The forest canopy enhances dry Hg deposition. Water transported along shallow flow paths supplies greater quantities of Hg than water in deep flow paths. Wetlands are important in the supply of dissolved organic carbon (DOC), which enhances the transport of ionic Hg and methylmercury (MeHg), and are important sites for the production of MeHg. The nutrient status and productivity of surface waters also control concentrations of MeHg in aquatic biota. Indicators of lakes sensitive to Hg inputs are shown in the insert (after Driscoll et al. 2007). Reservoir creation and water-level fluctuation will stimulate MeHg production in the littoral region. Abbreviations: ANC, acid neutralizing capacity; Hg^0, elemental Hg; P, phosphorus; PHg (i.e., Hg[p]), particulate Hg; RGM (i.e., Hg[II]), reactive gaseous Hg.

Table 3. Hypothesized mechanisms for presence of biological mercury (Hg) hotspots in the Northeast.

		Hypothesized mechanisms of Hg contamination				
Biological Hg hotspot	State/ province	Regional and global atmospheric deposition	Water-level management	Landscape sensitivity	Local air emissions	Local soil contamination
H1a: Adirondack Mountains (west)	New York	x	–	x	–	–
H1b: Adirondack Mountains (central)	New York	x	–	x	–	–
H2: Upper Connecticut River	New Hampshire, Vermont	x	x	–	–	–
H3a: Merrimack River (middle)	New Hampshire	x	–	–	x	–
H3b: Merrimack River (lower)	Massachusetts, New Hampshire	x	–	–	x	–
H4a: Upper Androscoggin River	Maine, New Hampshire	x	x	–	–	–
H4b: Upper Kennebec River (west)	Maine	x	x	–	–	x
H4c: Upper Kennebec River (east)	Maine	x	x	–	–	–
H5a: Kejimkujik National Park	Nova Scotia	x	–	x	–	–
H5b: Central Nova Scotia	Nova Scotia	x	–	x	–	–

Hg^0 and therefore have much shorter residence times. These oxidized species make up a small fraction of the total atmospheric Hg (less than 5% at remote sites) but can be responsible for a significant fraction of the total deposition. Gaseous divalent Hg and Hg(p) make up 50% to 90% of the Hg emitted from coal-fired electric utilities in the northeastern United States (NESCAUM 2005, NHDES 2005).

Although Hg^0 generally has a low deposition velocity, under some conditions Hg^0 can be rapidly converted to gaseous Hg(II) and deposited locally and regionally (Wang and Pehkonen 2004). Elemental Hg can also interact with the forest canopy, enhancing deposition rates (discussed below). Gaseous Hg(II) and Hg(p) have high deposition velocities; therefore, proximity to sources and the form of Hg emitted from sources play key roles in determining the amount of Hg deposited to a given area.

We hypothesize that once Hg has been emitted to the atmosphere and deposited to the landscape, the potential for biological Hg hotspots to develop depends on several factors, including the rate of deposition as well as site-specific characteristics such as landscape sensitivity, water-level management in reservoirs, and direct Hg input from water discharges and contaminated soils. Examples of how these factors affect organisms at higher trophic levels are provided below.

Landscape-driven biological mercury hotspots. Ecosystems vary in their sensitivity to Hg inputs; models predicting ecosystem sensitivity can be developed using environmental indicators (Roué-Legall et al. 2005). Mercury that is deposited from the atmosphere may be reemitted to the atmosphere, sequestered in soil or sediments, or transported with drainage waters to aquatic ecosystems, where it can potentially be methylated and bioaccumulate in aquatic organisms. Generally only a small fraction of atmospheric Hg deposition is transported to aquatic ecosystems (Grigal 2002). Nevertheless, the extent to which Hg is transmitted to surface waters varies greatly, and is controlled by multiple processes in the watersheds that connect atmospheric deposition to Hg fate in surface waters (figure 3). Ecosystems with enhanced Hg deposition, transport to surface waters, methylation, and bioaccumulation are considered Hg sensitive (Driscoll et al. 2007).

Forests enhance landscape sensitivity to atmospheric Hg deposition. Canopy trees scavenge atmospheric Hg (Rea et al. 1996). Atmospheric Hg(p), gaseous Hg(II), and oxidized Hg^0 may be adsorbed by foliage and subsequently leached in throughfall (Lindberg et al. 1995). Elemental Hg also enters foliage by the stomata and can ultimately be deposited to the forest floor via leaf litter. In northeastern North America, dry deposition associated with the canopy may provide 60% to 75% of total Hg inputs to forest ecosystems (Miller et al. 2005).

Landscape characteristics including shallow hydrologic flowpaths (Grigal 2002, Galloway and Branfireun 2004), the presence of wetlands (St. Louis et al. 1994), and unproductive surface waters (Chen et al. 2005) facilitate the transport, methylation, and bioconcentration of Hg in surface waters, thereby increasing an ecosystem's sensitivity to atmospheric Hg deposition (Driscoll et al. 2007). Moreover, acidic deposition has affected forested watersheds across eastern North America (Driscoll et al. 2001). It exacerbates ecosystem sensitivity to Hg because the addition of sulfate stimulates production of MeHg (Jeremiason et al. 2006) and the acidification of surface waters enhances concentrations of Hg in fish tissue (Hrabik and Watras 2002).

Two of the biological Hg hotspots in the Northeast, located within the Adirondack Mountains (H1a and H1b) and Nova Scotia (H5a and H5b), appear to be associated with watersheds that are highly sensitive to atmospheric Hg deposition (table 3); the H5a grid cell is of especially high concern because of demonstrated negative Hg impacts on common loon reproductive success (Burgess et al. 1998, 2005). The grid cells in these biological Hg hotspots have forested and wetland cover above the 80th percentile of all grid cells, and are in the lowest 10th percentile for agricultural land uses. These same

grid cells were characterized by water chemistry within the sensitive ranges for attributes associated with high fish Hg in the Northeast (Driscoll et al. 2007). The mean values for 28 water bodies contained in these grid cells are as follows: TP = 9.5 μg per L; DOC = 4.7 mg C per L; ANC = 75 μeq per L; pH = 6.1.

Biological mercury hotspots associated with water-level management. Mercury concentrations in biota are elevated in reservoirs of the Northeast relative to other aquatic environments (Evers et al. 2004, Kamman et al. 2005). We identified two biological Hg hotspots representing four grid cells that appear to be associated with water-level manipulations in reservoirs: the upper Connecticut River in New Hampshire and Vermont (H2) and the upper Androscoggin River watershed (H4a) and upper Kennebec River watershed of Maine (H4b, H4c).

Generally, elevated Hg levels can be attributed either to reservoir creation or to water-level manipulations within existing reservoirs. The initial saturation of soils resulting from the creation of a reservoir yields a large flux of Hg and other detrital material to overlying waters (Bodaly et al. 2004). The resultant decompositional environment of the soil–water interface favors bacterial methylation of recently deposited or legacy Hg adsorbed on soil and vegetative particles. The MeHg forms complexes with various DOC compounds, and several factors, including the composition of the DOC itself, mediate subsequent bioaccumulation (Bodaly et al. 2004, Driscoll et al. 2007). Methyl Hg concentrations have been shown to increase up to 30% above initial values within the first 13 years after reservoir creation (Schetagne and Verdon 1999).

Increases in fish Hg concentrations of 1.5 to 4 times natural lake background levels have been observed in new reservoirs, with concentrations peaking approximately 10 to 15 years postconstruction and declining thereafter (Schetagne and Verdon 1999). Where reservoirs are not further manipulated or managed, fish Hg concentrations typically decline to natural lake background levels 20 to 40 years after initial flooding (Anderson et al. 1995, Schetagne and Verdon 1999).

In addition to reservoir creation, water-level fluctuation influences fish Hg concentrations. Water-level fluctuation has been identified as a key variable in explaining elevated Hg concentrations in fish tissue (Verta et al. 1986). Shallow depth and variable hydroperiods are strongly associated with increased fish Hg concentrations in southeastern US ponds (Snodgrass et al. 2000). The sediments of dewatered and reinundated littoral zones are prime environments for methylation because of their transitioning reduction–oxidation conditions, which promote bacterial sulfate reduction. Methylmercury formed in the littoral zone can be transported to the remaining open-water portion of the reservoir either during rain events or when the reservoir is refilled. The availability of MeHg to reservoir biota is likely to vary in relation to the ratio of dewatered area to reservoir size. Steep-sided reservoirs with organic-poor substrates can be expected to display less efficient MeHg production, lower ambient MeHg concentrations, and less bioaccumulation than reservoirs with wide basins and large littoral areas with more organic matter.

Several reservoir systems in the Northeast illustrate the effects of water-level manipulations (figure 4). In one study in north-central Maine, the ratio of MeHg to Hg in samples from sediment cores was shown to increase considerably, and then remain elevated, after the onset of reservoir fluctuation (Haines and Smith 1998). In another Maine study of five interconnected reservoirs, Hg concentrations in loon tissue increased with greater reservoir fluctuation. In reservoirs that had large summertime (June through September) drawdowns (> 3 m), Hg concentrations in adult loon blood were significantly higher than in reservoirs with small drawdowns (< 1 m) (figure 4). Similar patterns in fish Hg concentrations were documented in an interconnected system of three Connecticut River reservoirs for smallmouth bass (*Micropterus dolomieu*) and yellow perch (figure 4). In Minnesota, dampening water-level fluctuations resulted in significantly improved fish Hg concentrations (Sorensen et al. 2005).

Biological mercury hotspots associated with direct water discharges and contaminated soils. In contrast to sources of Hg from air emissions, direct Hg discharges (e.g., industrial wastes, wastewater, stormwater overflow) and land-based contamination (e.g., landfills, former mining and industrial facilities) tend to affect discrete drainage areas. Eight well-known sites of Hg discharges into lakes and rivers were identified, though they are not considered biological Hg hotspots under our definition, since the data for Hg in biota are currently insufficient to make such determinations (figure 2). The influence of these sources on streams is well studied; generally, streams can rapidly transport and diffuse Hg from a site (Whyte and Kirchner 2000). However, some land-based Hg sources, such as those on rivers with extensive emergent, shrub, and forested floodplains, can have significant downstream biological impacts that may reach 30 km (Wiener and Shields 2000) to 130 km or more (Hildebrand et al. 1980) from the source, decades after termination of active Hg discharges. Mercury-cell chlor-alkali plants are well-known sources of Hg contamination (Hildebrand et al. 1980), and in some cases they may influence biotic Hg levels in lakes that are downwind (A7; figure 2). Other less-described sources include landfills with Hg-containing leachate (Niebla et al. 1976), historical mining activities (Seiler et al. 2004), and municipal wastewater treatment plants (Gilmour and Bloom 1995). Storm water discharges, particularly from areas associated with impervious cover in urban and suburban footprints, also can enhance Hg supply to surface waters (Rule et al. 2006). Estuaries and other wetlands are common end points of urban watersheds, and the potential exists for negative impacts to avian reproductive success from Hg runoff (Schwarzbach et al. 2006). To further assess potential ecological impacts, monitoring and remediation efforts need to be continued long after Hg discharges to surface water from

point sources or contaminated soils are terminated.

Biological mercury hotspots associated with local atmospheric emissions and deposition: A case study. Several studies have shown that the high ambient concentrations of gaseous Hg(II) typically observed in the vicinity of high-emission areas increase dry and wet Hg deposition (USEPA 1997, Bullock and Brehme 2002) and Hg concentrations in soils and sediments (Biester et al. 2002). Here we estimate emissions and deposition in southern New Hampshire and parts of northeastern Massachusetts in order to assess the linkages among local Hg emissions, deposition, and concentrations in biota.

The industrial source complex short-term air dispersion model, or ISCST3 (USEPA 1995), was used to examine the hypothesis that the biological Hg hotspot in the middle and lower Merrimack River watershed (H3a and H3b; figure 2) is associated with high deposition from local emissions sources.

The ISCST3 model is a steady-state Gaussian plume model, which is used to assess pollutant concentrations from sources at the local scale (within 50 km). It assumes that deposition of Hg^0 from anthropogenic emissions is balanced by the reemission of previously deposited Hg^0, because of its large vapor pressure and low solubility (Bullock and Brehme 2002, Cohen et al. 2004), so only deposition of Hg(II) and Hg(p) was simulated in this analysis (table 4). The Henry's law constant and molecular diffusivity used in the USEPA *Mercury Study Report to Congress* (USEPA 1997) were adopted for Hg(II). Following Landis and colleagues (2002), it was assumed that the fine fraction (0.68 μm) accounted for 70% and the coarse fraction (3.5 μm) 30% of the Hg mass.

The model was run using a 5-km grid based on the 1996 National Emissions Inventory (USEPA 1996) for Hg and the 2002 revised emissions inventory for the Northeast states (NESCAUM 2005). The input-modeling domain was defined as New Hampshire and several counties within the adjacent states of Maine, Massachusetts, and Vermont. The output-modeling domain was limited to New Hampshire and northeastern Massachusetts. Meteorological data from Concord, New Hampshire, and Portland, Maine, were used as the surface and upper air data for 2002, respectively.

The ISCST3 results indicate that a biological hotspot (H3a and H3b) exists within an area of elevated deposition that receives considerable Hg input from local and regional sources (figure 5). Model estimates show total Hg deposition associated with local and regional sources of 17 to 804 μg per m^2 per year in 1996 and 7 to 76 μg per m^2 per year in 2002. There are two possible reasons for this area of high Hg deposition: (1) The predominant wind direction has a westerly compo-

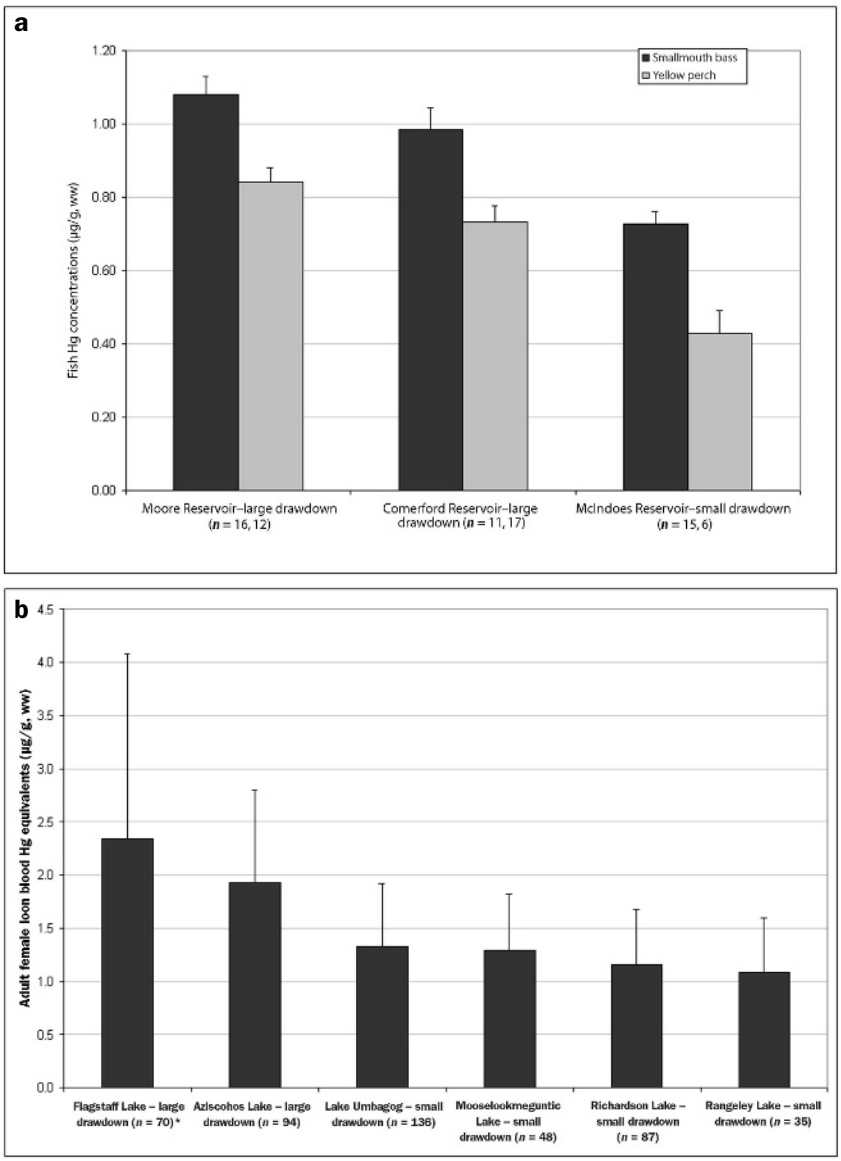

Figure 4. (a) Fillet mercury (Hg) concentrations for smallmouth bass and yellow perch (mean ± standard deviation [sd]) at three interconnected Connecticut River reservoirs in Vermont and New Hampshire and (b) blood Hg concentrations for the common loon (mean ± sd) at five interconnected Androscoggin River reservoirs in Maine and New Hampshire and one reservoir (Flagstaff Lake) in the upper Kennebec River watershed, Maine. (Although it is not hydrologically connected to the grid in the upper Androscoggin River watershed, Flagstaff Lake is illustrative of headwater reservoirs in that region that have large drawdowns.) Reservoir drawdowns from June through September that are less than 1 m are considered small, and those greater than 3 m are considered large. Kruskal-Wallis tests indicate significant differences between reservoirs with large and small drawdowns.

nent, and (2) major Hg sources are located in southern New Hampshire and Massachusetts. Of the total modeled deposition in 2002, Hg(II) deposition contributes the dominant fraction (90%) compared with Hg(p) (10%), primarily because the dry and wet deposition velocities for Hg(II) are higher than for Hg(p). In addition, the emissions of gaseous Hg(II) and Hg(p) from point sources contribute approximately 76% and 58% of the totals in the Hg(II) and Hg(p) categories, respectively (table 5). The ISCST3 results also show that dry deposition contributed more than wet deposition for Hg(II), while the opposite was true for Hg(p).

The USEPA estimated Hg deposition in the United States for 2001 using the community multiscale air quality (CMAQ) model. For the study area in northeastern Massachusetts and southern New Hampshire, they report a range in total deposition of 15 to 20 μg per m^2 per year (USEPA 2005). Miller and colleagues (2005) estimated regional Hg deposition for the study area using a "big-leaf" model and reported a range in total Hg deposition of 19 to 21 μg per m^2 per yr, with wet deposition of 5 to 6 μg per m^2 per year and dry deposition of 14 to 15 μg per m^2 per year. The values from the CMAQ model include local sources, but the emissions are averaged over a large grid cell, and therefore the model appears to underpredict total Hg deposition in the immediate vicinity of large emission sources. The big-leaf model represents regional and global deposition sources; the impact of large local emission sources was not directly accounted for. The local deposition estimates from the ISCST3 model represent an additional Hg input above the deposition estimated by the big-leaf model and therefore suggest that approximately 25% to 65% of total Hg deposition from all sources in the southern New Hampshire region is attributable to local emission sources.

Table 4. Deposition parameters of mercury (Hg) used for this study.

Form	Properties	Values used in this study
Divalent Hg	Molecular diffusivity[a]	0.045 cm^2 per s
	Solubility enhancement factor[a]	10^9
	Pollutant reactivity[a]	800
	Mesophyll resistance[a]	0
	Henry's law constant[a]	2.7×10^{-7}
	Liquid scavenging ratio[b]	2.5×10^{-4} (s-mm per hr) $^{-1}$
	Frozen scavenging ratio[b]	5.0×10^{-5} (s-mm per hr) $^{-1}$
Particulate Hg	Liquid scavenging coefficient (0.68 μm)[b]	7.0×10^{-5} (s-mm per hr) $^{-1}$
	Frozen scavenging coefficient (3.5 μm)[b]	2.8×10^{-4} (s-mm per hr) $^{-1}$

a. Adopted from USEPA 1997.
b. Adopted from Sullivan et al. 2004.

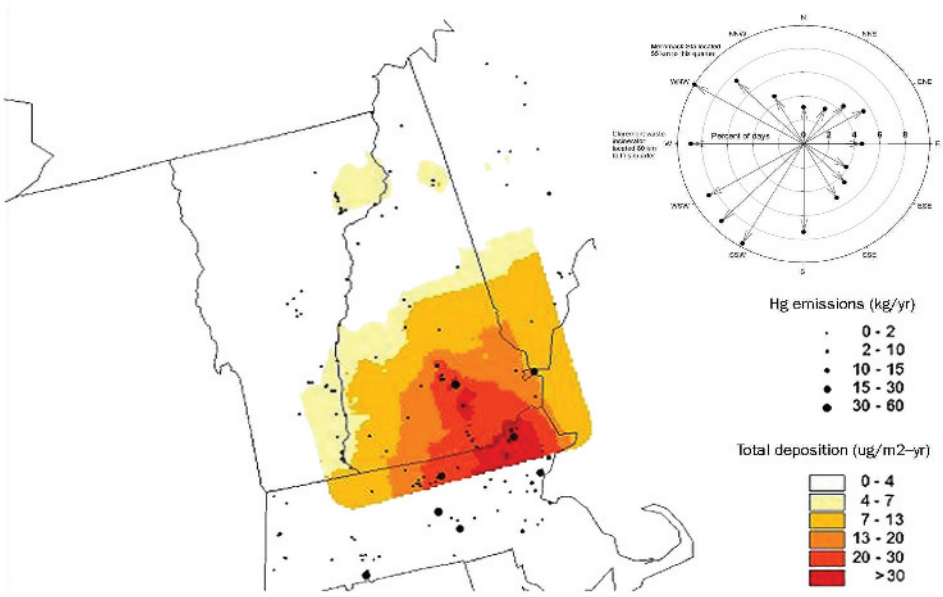

Figure 5. Left, map showing total mercury (Hg) deposition for 2002, estimated using the industrial source complex short-term model, or ISCST3; right, wind rose showing the direction of air flow for May through August 1999 to 2002 in southern New Hampshire, based on weekly wind roses from the NOAA (National Oceanic and Atmospheric Administration) Air Resources Laboratory's READY (Real-time Environmental Applications and Display System) analyses (NOAA 2006).

Table 5. Emission rates used in model domain in 2002.

Emission sources	Emission rates (kg per yr) Particulate mercury	Divalent mercury	Elemental mercury
Point sources	82.4	264.8	135.8
Area sources	60.4	90.6	245.0
Total emissions	142.8	355.4	380.8

Temporal patterns in biotic mercury. Historical data from the Merrimack River watershed biological hotspot (H3a and H3b) suggest that biotic Hg can change rapidly in response to changes in atmospheric emissions and deposition from local and regional sources. From 1997 to 2002, Hg emissions in southern New Hampshire declined 45 percent, largely as a result of restrictions on incinerators (table 6). Meteorological data from Concord were used to determine the dominant wind direction in the area of the Merrimack River watershed biological hotspot and to identify a group of study lakes downwind from major Hg sources. The average wind direction was calculated in grid cell H3a (latitude –43.08 N, longitude –71.16 W) for 1999 to 2002 using the months of May through August (a period of loon blood Hg measurements). The results show that airflow to grid cell

H3a had a westerly component during approximately two-thirds of this period (figure 5).

Based on the meteorological analysis, we selected 10 study lakes within grid cell H3a that were downwind of major Hg emission sources and, when pooled together, provided time series data for Hg in common loons. The study lakes are: Ayers, Canobie, Jenness, Massabesic, Mendums, Onway, Northwood, Pawtuckaway, Swains, and Tower Hill. Mean loon Hg concentrations in these lakes declined 64% from 1999 to 2002 (figure 6a), commensurate with the reduction in Hg emissions of 45% from upwind sources in southern New Hampshire (table 6). Recent data show no appreciable change in mean loon Hg concentrations from 2003 to 2005 (figure 6a). The grid cell immediately north of grid cell H3a, outside the area of highest Hg deposition within the middle Merrimack River watershed, provides a reference area for comparing the magnitude and temporal trends of loon Hg concentrations. This area has similar watershed cover and water chemistry to grid cell H3a. Here, mean loon Hg concentrations were 1.3 to 2.7 times lower than in grid cell H3a during the 1999 to 2002 time period, but still declined 30%. From 1999 to 2002, mean loon Hg concentrations in grid cell H3a exhibited a significant negative trend (using the Mann-Kendall test for normalized approximations; $s = -6$, $n = 4$, $z = -1.70$), and the grid cell immediately north of grid cell H3a did not exhibit a significant negative trend ($s = -4$, $n = 4$, $z = -1.02$).

Negative mercury trends in other taxa were observed within the lower Merrimack River watershed biological hotspot and demonstrated other lines of evidence during the same time period. In yellow perch, there was a significant decrease in fillet Hg concentrations between 1999 and 2004, based on individuals normalized to 24.3 cm in length within northeastern Massachusetts, which overlaps with grid cell H3b; comparatively, throughout the rest of Massachusetts, perch exhibited decreases approximately half as large as those in the Merrimack River watershed (C. Mark Smith and Michael Hutcheson, Massachusetts Department of Environmental Protection, Boston, personal communication, 7 July 2006). Mercury concentrations in zooplankton samples taken from three lakes in grid cell H3a declined between 1996 and 2002, compared with three study lakes outside grid cell H3a, in which the trend in total Hg in zooplankton did not decline (Chen et al. 2000; Carol Folt, Department of Biological Sciences, Dartmouth College, Hanover, New Hampshire, personal communication, 20 June 2006).

The consistency between the timing and magnitude of Hg emissions reductions and the declines in Hg concentrations in common loons, fish, and zooplankton could be related to several factors. A substantial amount of gaseous Hg(II) was removed from the

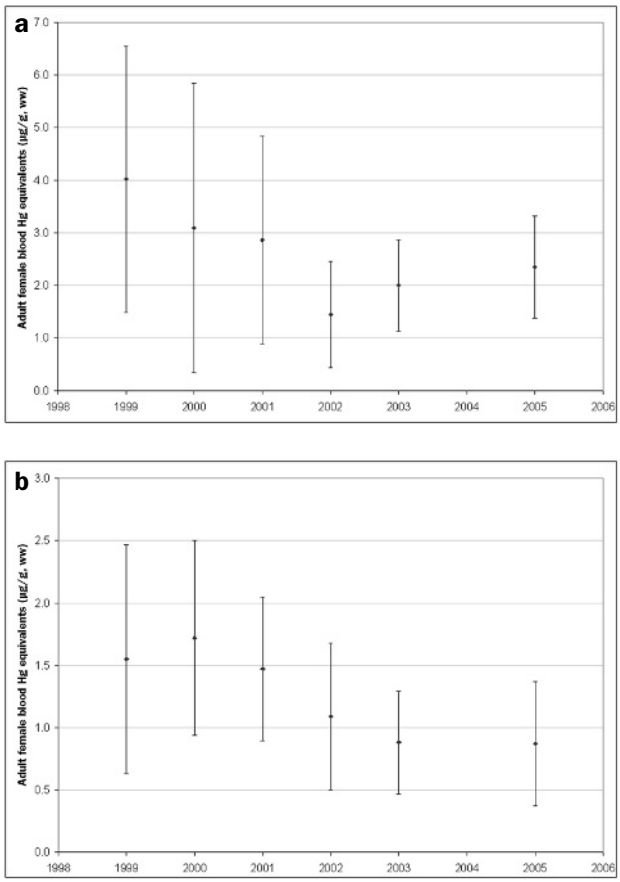

Figure 6. Temporal patterns for adult loon blood mercury (Hg) equivalents (µg per g, wet weight; mean ± standard deviation) in (a) the middle Merrimack River watershed (n = 53) and (b) the upper Merrimack River watershed (n = 43), New Hampshire. Note: The magnitude of the y axis, adult female blood Hg equivalents, differs between figure 6a and 6b.

Table 6. Values of mercury (Hg) emissions, deposition, and biotic concentrations in the middle Merrimack River watershed, New Hampshire, for 1996–1997, 1999, and 2002.

Measure	Year	
	1996–1997 and 1999	2002
Emissions in model domain	1515.3 kg	879.0 kg
Maximum annual deposition[a]	810 µg per m^2 per yr	76 µg per m^2 per yr
Area of elevated deposition	50 km^2	20 km^2
Average adult common loon blood equivalent[b]	4.02 µg per g	1.45 µg per g
Average zooplankton (45–202 µm)	5.14 ng per g	0.59 ng per g
Average zooplankton (> 202 µm)	1.72 ng per g	0.17 ng per g

a. Deposition estimates are based on monitoring data from the Mercury Deposition Network and ISCST3 (industrial source complex short-term) model analysis.

b. Common loon tissue Hg equivalents were determined from 10 lakes in southeastern New Hampshire from 1999 to 2005. The decline from 1999 to 2002 represents a statistically significant change ($t = 2.1$, df = 16, $p = 0.008$). Loon blood and egg Hg concentrations were collected starting in 1999.

Table 7. Emission reduction scenarios considered in this analysis.

Location of coal-fired electric utilities	Emissions (kg per yr)		
	Current	50% reduced	90% reduced
Merrimack Station	62.4	31.20	6.24
Schiller Station	5.00	2.50	0.50
Salem Harbor Station	8.80	4.40	0.88
Mount Tom Station	1.93	0.97	0.19

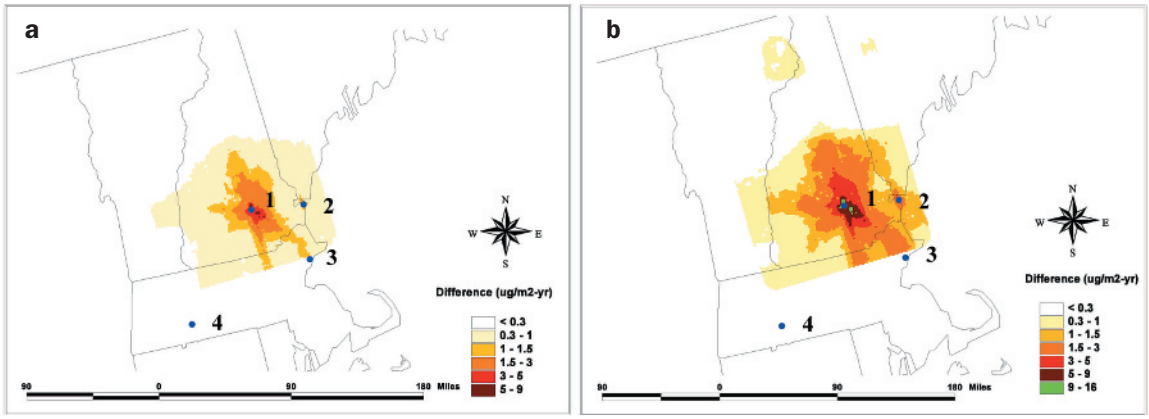

Figure 7. Total differences in mercury (Hg) deposition (μg per m² per year) statewide in New Hampshire (a) with 50% emission reduction and (b) with 90% emission reduction from four coal-fired utilities in New England. Power plant Hg emission sources: (1) Merrimack Station, (2) Schiller Station, (3) Salem Harbor Station, (4) Mount Tom Station.

local atmosphere and most likely reduced local Hg deposition, and this "new" Hg is generally thought to be more readily bioavailable than Hg that has been in the ecosystem for some time (Gilmour et al. 2003). Moreover, most of the study lakes have characteristics that are considered conducive to rapid response: They exist in close proximity to the emission sources, have small watershed-to-lake-area ratios (Grigal 2002), and have limited shoreline wetlands. Wetland areas less than 150 m from lake shoreline are predictive of loon blood Hg concentrations (Kramar et al. 2005), and therefore their extent influences the production of MeHg in the food web.

Links between local emission sources and birds have been measured elsewhere. In Britain, downward trends in piscivorous bird Hg levels were associated with reductions in local industrial air emissions (Newton et al. 1993). In the United States, recent downward trends in the Hg concentrations of Florida's wading birds were linked to reductions in Hg emissions and deposition from local sources (Frederick et al. 2004). Varying sulfate loads may also be a factor in the extent of MeHg production and availability in the Everglades (Bates et al. 2002).

Predicted future changes related to power plant emissions. The ISCST3 model was also used to evaluate two scenarios: a 50% and a 90% reduction in emissions from the four active coal-fired utilities located in the input modeling domain (table 7). The difference in deposition between the current and reduced emissions scenarios is evident in grid cells H3a and H3b (figure 7a, 7b). The average difference in deposition across all cells was 5% for the 50% reduction scenario and 9% for the 90% reduction scenario. However, the reduction in deposition was much greater in the areas of highest deposition; the model cells with the greatest percent decrease between current and projected deposition (23% for the 50% reduction and 41% for the 90% reduction) are located within 20 km of the Merrimack Station in New Hampshire, which is the largest coal utility in the modeling domain.

The scenario results indicate that a large portion of Hg(II) and Hg(p) is deposited within a short distance of these large sources, causing elevated deposition. Similarly, the results show that emissions from four coal-fired utilities in the area contribute approximately 40% of total Hg deposition attributed to local sources, and that decreased Hg emissions will result in substantial decreases in Hg deposition. The magnitude of the decreases in Hg deposition from local sources illustrated in these calculations (figure 7) should be viewed in the context of the additional Hg deposition from regional and global sources (19 to 21 μg per m² per year; Miller et al. 2005).

These results are based on the NESCAUM (Northeast States for Coordinated Air Use Management) inventory, which assumed that coal-fired utilities emit 70% of Hg as gaseous Hg(II) and Hg(p), on average. Recent stack-testing data for the Merrimack Station in New Hampshire suggest that gaseous Hg(II) emissions may constitute up to 92% of total Hg emissions at this facility (NHDES 2005). Under these conditions, we would expect baseline deposition to be higher than estimated here, and the decline in deposition associated with these emission reduction scenarios to be much greater.

Conclusions

Current levels of Hg deposition in the Northeast are 4 to 6 times higher than the levels recorded in 1900 (Perry et al. 2005). We identified five biological Hg hotspots in the region and hypothesized that these hotspots occur where the impacts of atmospheric Hg deposition are amplified by large reservoir fluctuations, highly sensitive landscapes, or elevated Hg deposition associated with large local emission sources.

Model estimates suggest that emissions from coal-fired power plants in the study region account for a large fraction of the total Hg deposited in the Merrimack River watershed hotspot, and that decreased emissions from these sources will result in decreased deposition. Significant and rapid improvements in Hg concentrations in common loons and other biota within this deposition-associated biological Hg hotspot (H3a, H3b) were documented for 1997–2002. Our analysis of the importance of local emission sources also emphasizes that emission trading rules must take local deposition and ecological conditions into account. Other management activities linked to potential reductions in biotic Hg concentrations include minimizing summertime water-level fluctuations on some reservoirs and creating suitable catchments for storm water runoff.

While existing data provide a strong basis for identifying biological Hg hotspots, large gaps in data and understanding continue to hamper our ability to quantitatively analyze sources and fully characterize the spatial and temporal patterns of deposition and biological availability across the United States and Canada. We suggest the development of comparable and linkable data sets for the primary and secondary data layers used here across North America; such data sets will further facilitate the identification of biological Hg hotspots. Developing novel indicator species, such as songbirds and bats, will enhance the ability to identify potential terrestrial biological Hg hotspots for invertivores that may or may not be directly associated with aquatic food webs.

At present, only 92 Hg wet deposition sites operate in the United States and Canada, and no coordinated national system exists to systematically collect and analyze Hg samples for dry deposition and biota in either country. A comprehensive Hg monitoring network has been developed (Mason et al. 2005) and, if employed, can be used to (a) better quantify wet and dry Hg deposition, particularly near high-emission sources; (b) detect additional deposition or biological Hg hotspots; (c) quantify the ecological and human health risks associated with existing biological Hg hotspots; and (d) track the resulting changes in management and policy actions. Ongoing process research and model development can be used to guide this monitoring network.

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