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Patterns of Mercury Deposition and Concentration in Northeastern North America (1996–2002)

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Abstract. Data from 13 National Atmospheric Deposition Program Mercury Monitor Network (NADP/MDN) monitoring stations (1996–2002) and the Underhill (VT) event-based monitoring site (1993–2002) were evaluated for spatial and temporal trends. More precipitation and mercury deposition occurred in the southern and coastal MDN sites, except for the Underhill site, which received more mercury deposition than surrounding sites. Precipitation patterns varied. Regionally, higher concentrations of mercury were recorded during the late spring and summer months. Several sub-regional clusters of MDN sites were evident, based on mercury deposition patterns. In general, more mercury was deposited during the summer months. ''Enhanced'' weekly deposition (>250 ng/m^2) and distinct seasonal deposition patterns were evident at all MDN sites. Regionally, high depositional periods contributed significantly to annual loads ($<20\%$–$\sim 60\%$). Southern and coastal sites measured more frequent periods of high deposition than inland sites. Spring and summer ''enhanced'' deposition may be important contributing factors to mercury bioaccumulation during the growing season. Recent regional reductions of mercury emissions were not reflected in the regional mercury concentration or deposition data. Few sites showed linear relations between the concentration of mercury in precipitation and acid rain co-contaminants (sulfates and nitrates).

Keywords: atmospheric deposition; mercury; regional patterns

Introduction

Atmospheric mercury deposition plays a significant role in mercury loading to fresh water ecosystems and watersheds (Scherbatskoy et al., 1994; US EPA, 1997; Scherbatskoy et al., 1998; Kamman and Engstrom, 2002). Mercury deposition to lakes and watersheds is influenced directly by the strength and proximity of mercury emission sources, the type of mercury emissions, and indirectly by local and regional weather patterns.

The rate of mercury loading to freshwater ecosystems is influenced by several factors including:

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direct mercury input from the atmosphere, mercury transport through the watershed (from contemporary and historical atmospheric inputs), inputs of acidifying compounds, and the rate of sedimentation of biological and particulate material (Bloom et al., 1991; Gilmore et al. 1992; Hurley et al., 1995; Regnell et al., 1997; Lorey and Driscoll, 1999; Shanley et al., 1999).

Mercury contamination of aquatic ecosystems in northeast North America has been extensively documented (USEPA, 1992; DiFranco et al., 1995; Newman et al., 1996; Smith and West, 1996; Schetagne et al., 1997; Beauchamp et al., 1998; Burgess et al., 1998; NESCAUM, 1998). Paleolimnological studies of mercury contamination in North American lake sediments and ombrotrophic bogs indicate elevated mercury loading for more than a century (Swain et al., 1992; Engstrom and Swain, 1996; Norton et al., 1997; Schuster et al., 2002). Loading patterns to the sediments of North American lakes suggest a steady increase during the last quarter of the 19th century and the first quarter of the 20th century. Peak loading appears to have occurred sometime during the 1960s and 1970s. A reduction in mercury loading to sediments has been observed over the last two decades (Engstrom and Swain, 1996; Lorey and Driscoll, 1999; Kamman and Engstrom, 2002; Kamman et al., 2002).

The state and provincial governments of the Northeast United States and Eastern Canadian Provinces have initiated a comprehensive mercury control strategy. The control program calls for a virtual elimination of mercury emissions from the region, with interim targets of a 50% reduction of mercury emissions by 2003 and a 75% reduction of mercury emissions by 2010. The first target reduction was met in 2002. Stationary sources emissions of mercury for 2002 (New York, New Jersey, and New England) have been estimated at 3600 kg, a reduction of approximately 75 + % from 1998 emissions (Round and Irvine, 2004). Legislation in the United States, targeted at the utility industry, promises additional significant reductions in mercury emissions, estimated as 70%–90% from 1990 base emissions for this major source sector.

One of the most important components of any regulatory effort is tracking the effectiveness of control programs, as evidenced by the acid rain program. The National Atmospheric Deposition Program/National Trends Network (NADP/NTN) provides valuable data to measure the effectiveness of acid rain control programs and subsequent changes in ecosystem acidification (Gbondo-Tugbawa and C. Driscoll, 2002; Stoddard et al., 2002). The Mercury Monitoring Network (MDN), initiated by the NADP in the mid-1990s, provides routine measurements of mercury deposition in precipitation. These data are useful in determining the effectiveness of regional and national mercury control strategies.

In Eastern Canada and the northeastern United States, widespread mercury contamination in aquatic ecosystems prompted provincial, federal and state agencies to deploy a regional atmospheric mercury monitoring network. By the late 1990s, more than a dozen routine and research monitoring sites were established in the Eastern Canadian Provinces and the northeastern United States. Since 2000, a score of additional sites have been added to fill gaps in the network.

The northeastern North America mercury monitoring network is a hybrid, composed of the NADP/MDN weekly composite mercury monitoring sites, and event-based monitoring components, such as the one in Underhill, Vermont. These sites provide a rich data set. Recent analyses of the data indicate that seasonal and spatial patterns for mercury are common (Scherbatskoy et al., 1994; Burke et al., 1995). New England data, provided by the USEPA Regional Environmental Monitoring and Assessment Program (REMAP), show higher mercury deposition and higher ambient levels of gaseous and particulate mercury in urban environments compared to rural and remote locations (Keeler and Yoo, 2003). Strong seasonal wet depositional patterns are evident, with more mercury deposited on the landscape during the late spring, summer and fall than during other times of the year (US EPA, 1999; Ryan et al., 2003). There is the suggestion of a depositional gradient from south-to-north, with the more southern monitoring sites receiving higher mercury deposition. In addition, coastal sites appear to receive more mercury deposition than inland sites (VanArsdale et al., Unpublished Manuscript; Ryan et al., 2003).

This paper provides a baseline to measure the effectiveness of current and future North American mercury emission control strategies. It explores the importance of periods of regional and

site-specific ''enhanced'' mercury deposition; relationships between mercury and acidic precipitation; and inter-site associations for deposition, precipitation and concentration. The wet deposition data provide a context for patterns of ecosystem mercury contamination, topics discussed in accompanying papers.

Methods

Precipitation, concentration and deposition data were collected from the Mercury Deposition Network (MDN) and National Trends Network (NTN) internet sites of the National Atmospheric Deposition Program (NADP) web site (http://nadp.sws.uiuc.edu/) during 2003. Historical data (1996–2002) from 13 monitoring sites (Table 1) were downloaded to spreadsheets. All data were reviewed for inconsistencies, such as missing weeks, and quality assurance flags noted. No attempt was made to synthesize ''missing'' weekly data based on annual or seasonal concentration of precipitation contaminants, for either the NTN or MDN data sets. Weekly start and end dates for each monitoring program (NTN and MDN) were reviewed and inconsistent data eliminated.

The Air Resources Lab of the University of Michigan provided mercury and precipitation data from the Proctor Maple Research Center (Underhill, VT) monitoring site. Monitoring and analysis protocols discussed in Landis and Keeler (1997) were employed at this site. The event data were not aggregated into weekly composites and therefore were not compared with MDN data. Only annual comparisons were made between the MDN data and those from the Underhill site.

Figure 1 presents the locations of the Underhill (VT) event-based site and the 13 MDN monitoring sites in the Northeast. Table 1 provides additional geographical information for these sites. Roughly half of the mercury monitoring sites are within 50 miles of the coast. The 13 MDN sites cover an area roughly a 1000 miles on the southwest to northeast axis, and 400 miles on the southeast to northwest axis. All sites are located in rural or remote areas, although the mid-coastal sites of New England (New Castle (NH05) and Freeport (ME96)) may be considered in the immediate depositional shadow of the east-coast megalopolis. The southwestern sites, Milford (PA72), Huntington (NY20), and St. Anicet (PQ04) are located downwind from the industrial heartland of the United States and the Great Lakes Basin. The most northeastern site, located in Cormak (NF09), is far removed from any local or regional sources of mercury. Six MDN sites are collocated with NADP/NTN sites: Bridgton (ME02), Freeport (ME96), Greenville (ME09), Acadia NP (ME98), Huntington (NY20), and Milford (PA72).

Table 1. Geographical and descriptive information on 14 mercury monitoring sites in northeastern North America (NADP, 2004)

Site	MDN #	Latitude	Longitude	Elevation	NTN[a]	Year[b]
Cormak, NF	NF09	49.3167	−57.3833	168 m		2001
Kejimkujik NP, NS	NS01	44.4328	−65.2056	155 m		1998
Mingan, QC	PQ05	50.2667	−64.2333	11 m		1999
St. Andrews, NB[c]	NB02	45.0833	−67.0833	11 m		1997
Acadia NP, ME	ME98	44.3739	−68.2606	129 m	yes	1996
Greenville, ME	ME09	45.4897	−69.6644	322 m	yes	1997
Freeport, ME	ME96	43.8319	−70.0628	15 m	yes	1998
Bridgton, ME	ME02	44.1075	−70.7289	222 m	yes	1998
New Castle, NH[c]	NH05	43.1667	−70.8667	10 m		1998
Laconia, NH[d]	NH00	43.5000	−71.5000	213 m		1999
St. Anicet, QC	PQ04	45.2000	−74.0333	49 m		1999
Huntington, NY	NY20	43.9731	−74.2231	500 m	yes	2000
Milford, PA	PA72	41.3275	−74.8203	212 m	yes	2001
Underhill, VT	na	44.5283	−72.8689	400 m	yes	1993

[a]National Trends Network.
[b]First year of complete data.
[c]Currently inactive.
[d]Moved to the Hubbard Brook Research Forest (NH) in 2004.

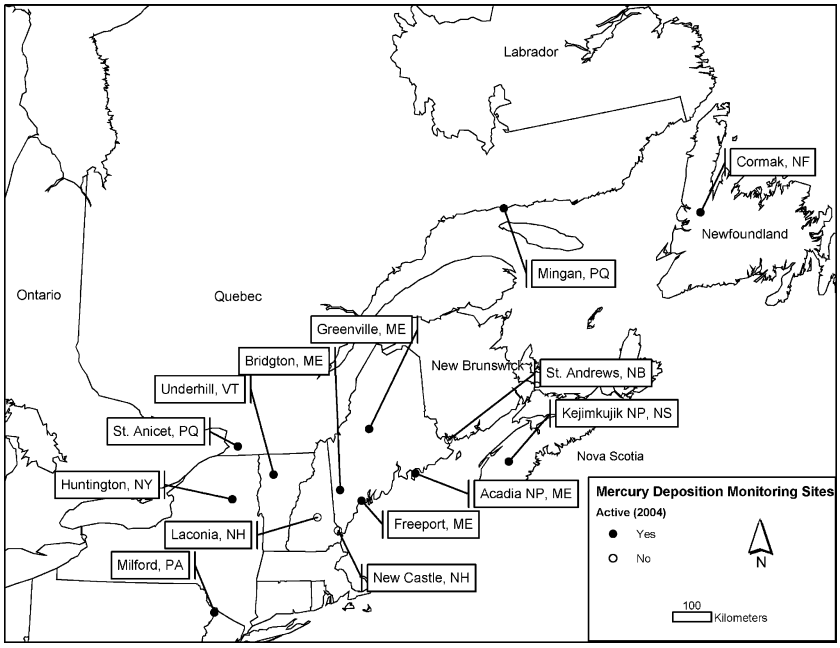

Figure 1. The location of the Underhill (VT) monitoring site and 13 Mercury Deposition Monitoring (MDN) sites in northeast North America.

All data used in this study were first imported into Microsoft Excel, and then exported to Statistica 6 (2003) (Statsoft, Inc.) for basic statistical analyses. Annual and weekly data for each site were subjected to basic statistical calculations (medians, quartiles and non-outlier ranges (the outlier coefficient is 1.5), means, standard deviations), plotted as time series, and interpreted for covariance and clustering. The annual volume-weighted concentration of mercury for each site was computed by summing weekly mercury deposition (weekly mercury concentration (ng/l)*, precipitation depth (mm)), and then dividing by the sum of corresponding weekly precipitation. Hierarchical tree plots were assembled using Euclidean distance and both single (nearest neighbor) and complete linkage methods.

Results

Regional patterns

The box plots presented in Fig. 2a–c, summarize the network-wide, within-year patterns of precipitation, mercury deposition, and volume-weighted concentration of mercury for the 13 MDN sites (all weekly data recorded from 1996–2002). Medians, upper and lower quartiles, and non-outlier ranges were plotted to help visualize within-year seasonal patterns. No obvious seasonal patterns of precipitation are evident for the region, except the months of August and September (weeks 29–37), when less precipitation (upper quartiles and medians) was recorded. Regionally, mercury deposition peaked during the late spring and summer (weeks 20–28) and was lowest during the late fall and winter (weeks 40–52 and weeks 1–13). This regional pattern was also apparent for the unweighted concentration of mercury in precipitation.

Regional inter-annual patterns

Tables 2, 3, and 4 provide annual summary data for precipitation, volume weighted concentration of mercury in precipitation, and mercury deposition for each MDN site and the Underhill (VT) site. Precipitation amounts were generally lowest during 2001. The region-wide volume-weighted concentration of mercury in precipitation (sum weekly mercury deposition (ng/m^2)/sum weekly

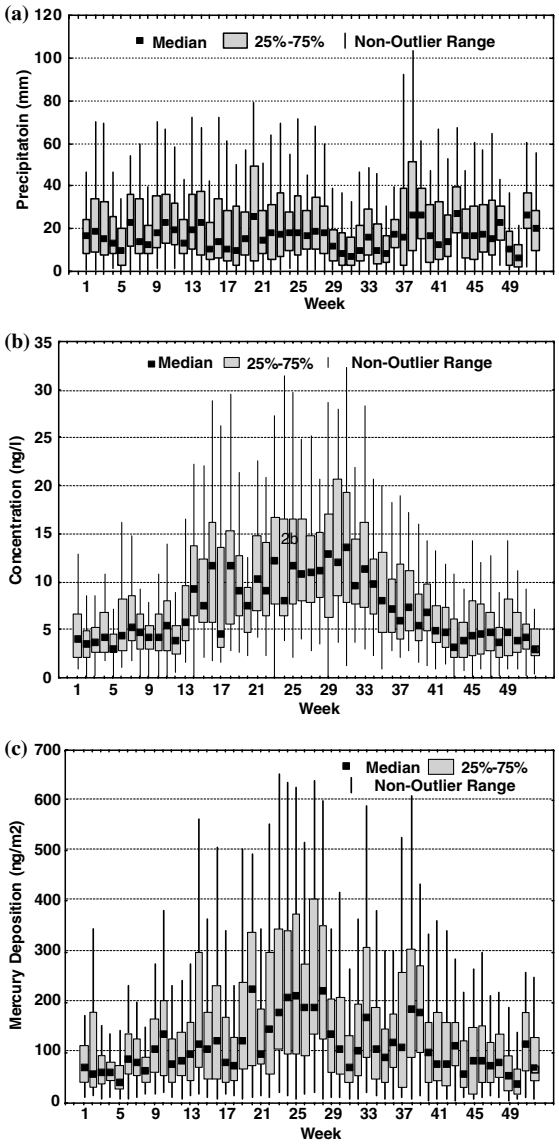

Figure 2. Median, upper and lower quartiles, and non-outlier range of weekly precipitation (mm), mercury concentration (ng/l), and mercury deposition (ng/m^2) for 13 MDN sites in northeast North America.

precipitation (mm)) declined to its lowest or near lowest level in 2002 (5.2 ng/l). The highest region-wide, volume-weighted concentration of mercury occurred in 1999 (6.7 ng/l). The highest network-wide deposition rates occurred during 1998 and 1999 (7.5 and 6.4 ng/m^2, respectively). During 2002, all sites recorded higher mercury deposition and more precipitation than the previous year, except the Mingan (PQ05) site.

Site-specific patterns

Figure 3a–c present weekly summary data for each MDN site for the period of record of each site. Median, lower and upper quartile, and non-outlier data indicate that the Kejimkujik NP (NS01), Acadia NP (ME98), Freeport (ME96) and Milford (PA72) sites receive more precipitation than neighboring sites. Over the period of record the St. Anicet (PQ04) and Mingan (PQ05) sites appear to receive less precipitation. The un-weighted mercury concentration data (Fig. 3b) indicate that the St. Anicet (PQ04), Milford (PA72), and New Castle (NH05) sites receive precipitation with higher concentrations of mercury than the other sites. The lowest median mercury concentrations were recorded at the most northern MDN sites, Mingan (PQ05) and Cormak (NF09) and at the Huntington (NY20) site. The depositional data (Fig. 3c) indicate higher weekly deposition at the Acadia NP (ME98), Freeport (ME96), Milford (PA72), and Kejimkujik NP (NS01) monitoring sites.

The Freeport (ME96) monitoring site recorded the highest two-year depositional total (1998–1999) of 18.9 ug/m^2 (Table 4) of all sites during the period of record. This contrasts with the two-year total mercury deposition of 7.9 ug/m^2 (2001–2002), recorded at the Greenville monitoring site (ME09), located less than 200 km north and inland of the Freeport MDN site. The Acadia NP (ME98), Freeport (ME96), Kejimkujik NP (NS01), and Milford (PA72) MDN sites consistently received higher mercury deposition than other sites.

Within-network associations

The MDN data were analyzed for inter-site clusters, groups of sites that showed similar precipitation, deposition and mercury concentration patterns. Hierarchical tree diagrams for the MDN sites are presented in Fig. 4a–c. The plots were constructed using complete linkage (the distances between two clusters determined by the farthest distance between neighbors) and Euclidean (geometric) distances. Distinct site associations were evident for precipitation, mercury concentration in precipitation, and deposition. Two distinct precipitation clusters included the Cormak,

Table 2. Annual precipitation (mm) measured at 13 MDN monitoring sites and the Underhill (VT) site (1996–2002)

Site	1996	1997	1998	1999	2000	2001	2002
Cormak (NF09)	–	–	–	–	–	1002	1208
Kejimkujik NP (NS01)	–	–	1203	1231	**1279**	1023	1427
Mingan (PQ05)	–	–	–	976	949	958	826
St. Andrews (NB02)	–	858	1120	1127	935	666	1201
Acadia NP (ME98)	1200	**1155**	**1445**	**1310**	1256	665	**1563**
Greenville (ME09)	–	940	1111	1266	1016	621	860
Freeport (ME96)	–	–	1251	1062	1180	706	1107
Bridgton (ME02)	–	–	1019	1082	1064	699	945
New Castle (NH05)	–	–	975	805*	–	–	–
Laconia (NH00)	–	–	–	938	–	–	–
St. Anicet (PQ04)	–	–	–	654	792	597	764
Huntington (NY20)	–	–	–	–	1100	**1026**	1073
Milford (PA72)	–	–	–	–	–	894	1187
Underhill, VT	1000	967	1416	1068	1045	820	1173

*Low capture efficiency.

Kejimkujik NP, St. Andrews and Greenville group and the Freeport, Bridgton, Laconia and New Castle group. The Huntington and Milford sites also grouped well. Two multi-site clusters for weekly mercury concentration included: the Cormak, Greenville, Bridgton and Huntington sites; and the Freeport, Laconia, New Castle and St. Anicet sites. Kejimkujik NP and St. Andrews shared similar patterns of mercury concentration in precipitation. Depositional clusters included: Cormak, Greenville, St. Anicet, Mingan, Laconia, and Huntington: and Kejimkujik NP, St. Andrews, Bridgton, New Castle, Acadia NP and Freeport. Dissimilar (poor associations) sites included Mingan for weekly precipitation, Milford and Acadia NP for weekly mercury concentration, and Milford for weekly deposition.

Enhanced mercury deposition

Information presented as non-outlier range in the previous box plots (Fig. 3a–c) strongly suggests periods of high mercury deposition network-wide. In addition, several MDN sites appeared to receive more high depositional periods than others. Since high deposition periods (single weeks or multiple week clusters) influence annual and seasonal deposition totals, as well as bias within-network associations, data for high deposition weeks were distilled from the total data set. These ''enhanced''

Table 3. Annual volume-weighted mercury concentration (ng/l) measured at 13 MDN monitoring sites and the Underhill (VT) site (1996–2002)

Site	1996	1997	1998	1999	2000	2001	2002
Cormak (NF09)	–	–	–	–	–	4.7	4.1
Kejimkujik (NS01)	–	–	5.3	4.9	4.9	6.3	5.2
Mingan (PQ05)	–	–	–	5.1	4.5	6.1	3.8
St. Andrews (NB02)	–	**6.7**	5.9	6.4	6.5	6.4	4.6
Acadia NP (ME98)	6.1	6.1	6.0	6.1	6.8	7.0	5.1
Greenville (ME09)	–	5.7	5.9	5.2	4.6	6.1	4.7
Freeport (ME96)	–	–	**8.4**	7.8	6.6	6.8	4.9
Bridgton (ME02)	–	–	6.3	6.3	5.1	6.6	5.2
New Castle (NH05)	–	–	7.4	5.3*	–	–	–
Laconia (NH00)	–	–	–	6.4	–	–	–
St. Anicet (PQ04)	–	–	–	**8.8**	**8.6**	8.8	7.3
Huntington (NY20)	–	–	–	–	6.3	5.0	5.0
Milford (PA72)	–	–	–	–	–	**9.4**	**8.0**
Underhill (VT)	7.9	9.1	8.9	7.3	8.8	8.9	7.5

*Low capture efficiency.

Table 4. Annual mercury deposition (ug/m^2) measured at 13 MDN monitoring sites and the Underhill (VT) site (1996–2002)

Site	1996	1997	1998	1999	2000	2001	2002
Cormak (NF09)	–	–	–	–	–	4.7	4.9
Kejimkujik NP (NS01)	–	–	6.4	6.0	6.2	6.5	7.5
Mingan (PQ05)	–	–	–	5.0	4.3	5.8	3.1
St. Andrews (NB02)	–	5.7	6.6	7.2	6.0	4.3	5.5
Acadia NP (ME98)	7.3	**7.0**	8.6	7.9	**8.6**	4.7	7.9
Greenville (ME09)	–	5.4	6.6	6.6	4.7	3.8	4.1
Freeport (ME96)	–	–	**10.6**	**8.3**	7.8	4.8	5.5
Bridgton (ME02)	–	–	6.5	6.8	5.5	4.6	4.9
New Castle (NH05)	–	–	7.2	4.2*	–	–	–
Laconia (NH00)	–	–	–	6.0	–	–	–
St. Anicet (PQ04)	–	–	–	5.8	6.8	5.2	5.6
Huntington (NY20)	–	–	–	–	7.0	5.2	5.3
Milford (PA72)	–	–	–	–	–	**8.4**	**9.5**
Underhill, VT	7.9	**8.8**	**12.6**	7.8	**9.2**	7.3	8.8

*Low capture efficiency.

mercury deposition week data (here defined as any week with mercury deposition in excess of 250 ng/m^2) have been summarized in Table 5. The >250 ng/m^2 threshold represented depositional loading greater than 2.5× the average of all median depositional values for the network (98.1 ng/m^2) for all years, and lies outside 60% the average upper non-outlier range for all sites. This threshold was ~1.5× the mean value of all weekly mercury deposition measurements recorded during the pilot MDN program in 1993 (Vermette et al., 1995).

Summary data are presented in Table 5 for the core MDN sites (Maine, New Brunswick and Nova Scotia MDN sites) and all other sites. The highest number of "enhanced" deposition weeks were recorded in 1998. During this year the Freeport (ME96) recorded the highest average annual "enhanced" weekly mercury deposition (566 ng/m^2 averaged over the 14 weeks), accounting for roughly 75% of the total mercury deposition for the year. The second highest average annual "enhanced" deposition was recorded during 2002 at the Milford (PA72) site (545 ng/m^2, $n = 10$, accounting for ~58% of the annual mercury deposition). The 2001 and 2002 data contrasted to those of other years when fewer "enhanced" deposition weeks and lower deposition occurred at many sites, particularly those in Maine. The Greenville (ME09) site recorded the least number of "enhanced" mercury deposition weeks (average 299 ng/m^2 over 2 weeks), contributing only 16% of the total annual deposition during 2001. The Acadia NP site also recorded few "enhanced" mercury deposition weeks (3), and little contribution of these weeks to the annual total (~25%).

For the period 1998–2002, 8 out of 12 MDN sites (the NH00 was excluded because it recorded only one year of valid data) experienced one or more years when "enhanced" mercury deposition contributed at least 50% of the annual total wet deposition. The four sites that did not receive at least 50% of their annual deposition from "enhanced" deposition weeks were Mingan (PQ05), Cormak (NF09), St. Anicet (PQ04), and Huntington (NY20).The average deposition measured for "enhanced" deposition weeks recorded at these sites (13 site-years) was 345 ng/m^2.

A strong relationship between the annual number of "enhanced" mercury deposition weeks at a given site, and their contribution (as percent) to annual mercury deposition is suggested in Fig. 5. The regression line predicted that when 8 or more "enhanced" mercury events occur during a given year, at least 50% of the total annual mercury deposition during that year would be from these weeks. The scatter plot presented in Fig. 6 indicates that more the "enhanced" deposition weeks in a year the greater the average loading for those weeks.

Associations between sulfates, nitrates, and mercury in precipitation

Data collected from six collocated NADP/NTT and MDN monitoring sites and the Underhill site provide unique opportunities to investigate

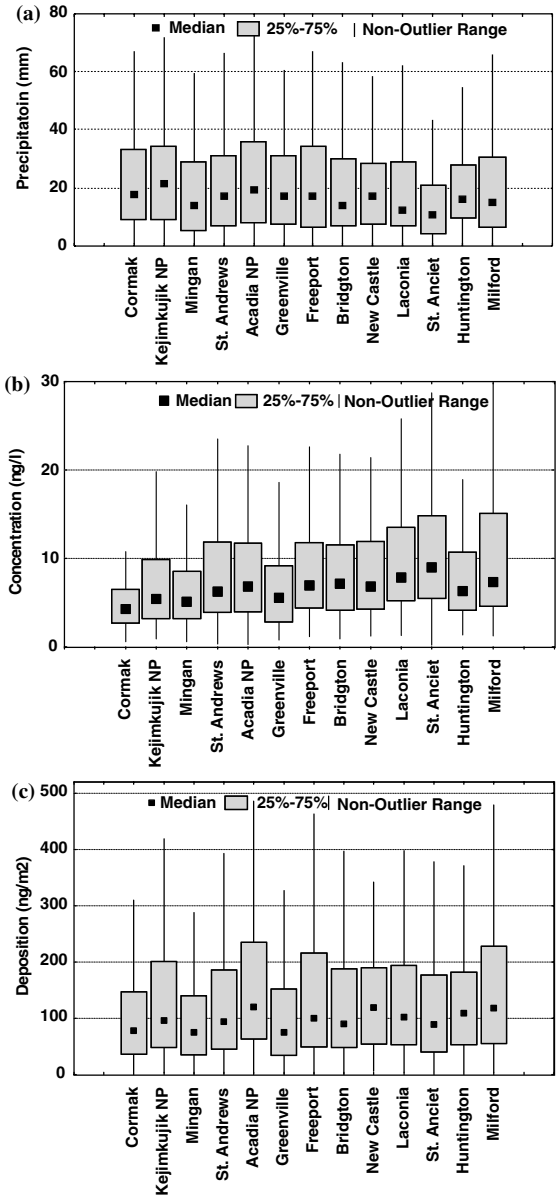

Figure 3. Median, upper and lower quartiles, and non-outlier range of weekly precipitation (mm), concentration (ng/l), and deposition (ng/m^2) for 13 MDN sites in northeast North America (1996–2002).

potential relationships between mercury, sulfate and nitrate contamination in precipitation. Regression equations for annual volume-weighted concentration data for these co-contaminants measured at the collocated monitoring sites are presented in Table 6. Bivariate relationships between sulfates and nitrates were common and paired annual volume-weighted concentrations of sulfates and nitrates for the northeast United States showed strong association (R^2 of 0.81). The annual precipitation-weighted associations for sulfate and mercury and nitrate and mercury (R^2 of 0.47 and 0.39, respectively) suggested that individual sites might exhibit stronger weekly co-contaminant relationships. Only the Milford (PA72) and Huntington (NY20) sites exhibited moderate associations between mercury, sulfate and nitrate. The Huntington (NY20) site, however, recorded the lowest weekly sulfate and nitrate associations. The four Maine sites all exhibited dissimilar co-contaminant associations, with coefficients of determination (R^2) less than 0.2 for paired weekly data of mercury, with sulfates and nitrates (data not presented in Table 6).

The relationships between co-contaminants (mercury, sulfate, and nitrate) were explored for weeks when MDN sites recorded "enhanced" mercury deposition (>250 ng/m^2). These data represented a significantly different subset of the data than in the weekly individual site comparisons. In this case, the mercury data were chosen as the independent variable. For the Maine, New York and Pennsylvania MDN sites, during weeks when mercury deposition exceeded 250 ng/m^2, the concentration of sulfates and nitrates strongly co-varied (R^2 of 0.76). Moderate anion dependence on mercury concentration during enhanced mercury deposition weeks was evident for few sites. None of the Maine sites exhibited any tendency of co-contaminant covariance (R^2 were all below 0.2), except for Bridgton (ME02). The Milford (PA72) and Huntington (NY20) monitoring sites exhibited moderate–strong co-contaminant association, with mercury–nitrate association somewhat stronger than mercury–sulfate association.

Discussion

The topic of mercury deposition and its link to mercury contamination of ecosystems is of considerable scientific interest. The data presented in this paper provide additional insights into patterns of mercury deposition that may influence regional patterns of mercury contamination in wildlife. Although spatial, inter- and intra-annual patterns

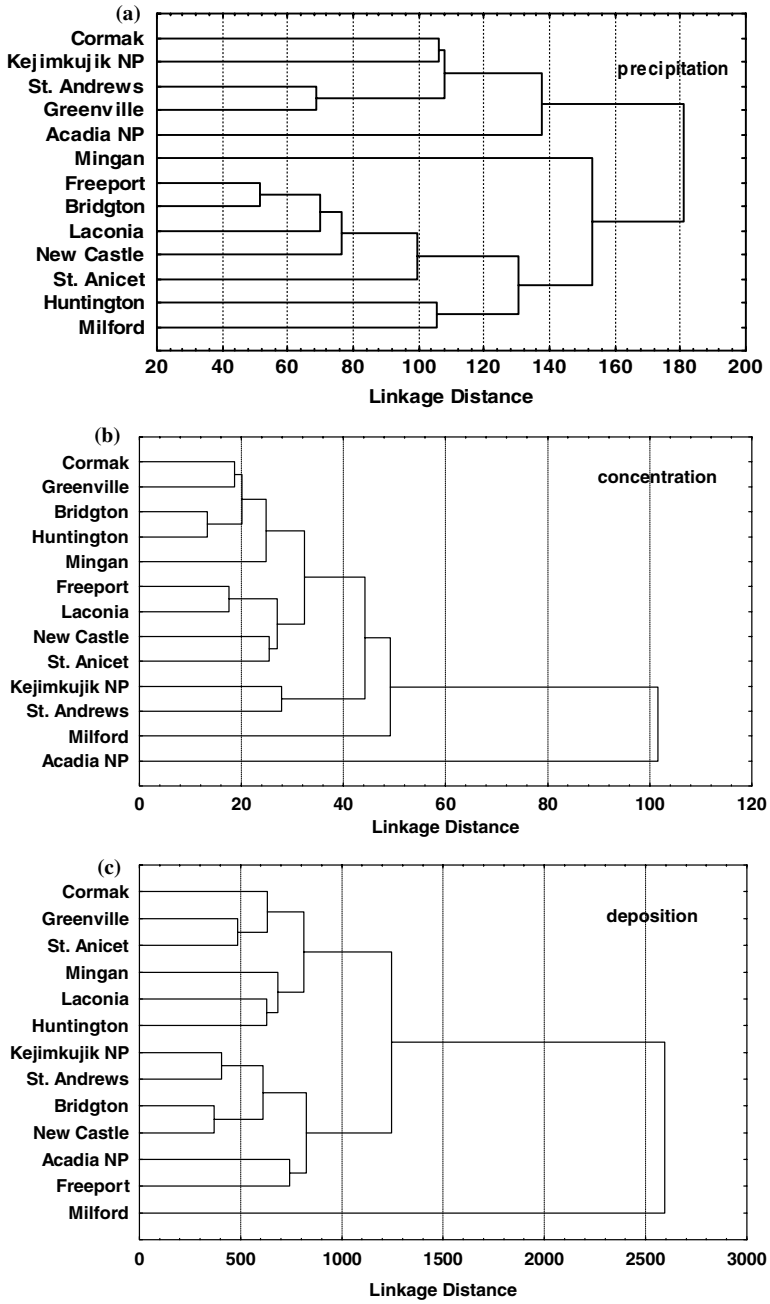

Figure 4. Hierarchical tree diagrams for 13 MDN sites. Complete linkage, Euclidean distance derived from weekly precipitation, mercury concentration and mercury deposition (1996–2002 data).

of mercury contamination in precipitation complicate those insights, our analyses provide evidence that the region can be viewed as two and possibly three distinct sub-regions defined by patterns of mercury loading (wet deposition).

Individual site data, presented as box plots Fig. 3a and b, support the north-to-south gradient in wet mercury deposition and contamination in precipitation. In addition, inland sites and sites located away from the coast generally show a

Table 5. Summary data of "enhanced" mercury deposition (weekly mercury deposition >250 ng/m^2) for network-wide and core MDN sites (1998–2002)

	Core sites*		All sites	
	Range	Average	Range	Average
1998				
# weeks	(6–14)	9	(6–14)	9.3
Average deposition (ng/m^2)	360–566	378	360–566	447
% Total deposition	40–75%	54%	40–75%	55%
1999				
# weeks	(5–11)	8.2	(3–11)	6.3
Average deposition (ng/m^2)	338–476	405	338–476	396
% Total deposition	28–57%	48%	27–57%	44%
2000				
# weeks	(4–11)	6.8	(4–11)	5.8
Average deposition (ng/m^2)	289–471	389	289–471	392
% Total deposition	23–50%	40%	23%–50%	41%
2001				
# weeks	(2–8)	4.5	(2–8)	5.8
Average deposition (ng/m^2)	299–417	361	293–488	369
% Total deposition	16–52%	34%	16–57%	39%
2002				
# weeks	(4–9)	6	(2–9)	5.7
Average deposition (ng/m^2)	325–432	387	325–545	395
% Total deposition	30–52%	39%	24–58%	37%

*Core sites include New Brunswick, Nova Scotia and Maine MDN sites..

gradient of increasing wet deposition and concentration from east-to-west. Taken together, these patterns show a $\sim$2$\times$ difference in annual wet mercury loading among sites within the region. Western and southern mercury monitoring sites receive higher mercury loading than the other sites. The coastal maritime sites of New England and New Brunswick, and the Nova Scotia and Newfoundland sites, also show a strong north-to-south depositional mercury and concentration gradient. These sites are influenced by coastal storms (as indicated by the chloride signal commonly found in precipitation), and in the case of the southernmost sites, lie in the immediate air pollution shadow of the east coast megalopolis. The cluster analysis for mercury deposition supports grouping the coastal New England and New Brunswick sites, with the Kejimkujik NP and Bridgton sites.

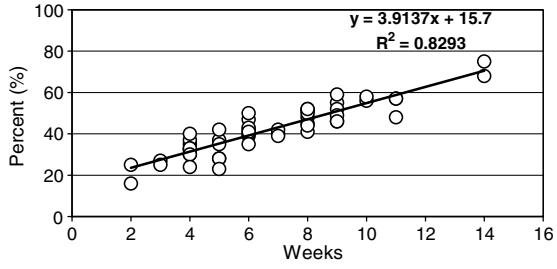

Figure 5. Precent contribution of "enhanced" mercury deposition weeks (>250 ng/m^2) to total mercury deposition as a function of the annual number of "enhanced" mercury deposition weeks for 13 MDN sites located in northeastern NA (1996–2002 data).

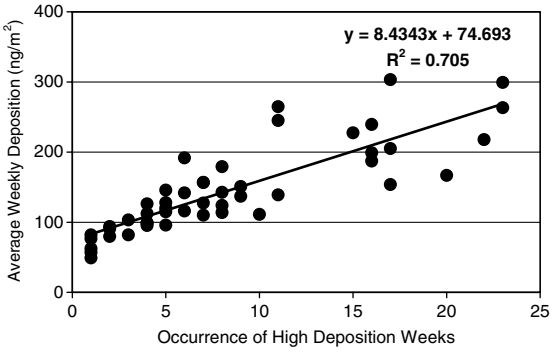

Figure 6. The relationship between the annual occurence (sum) of high mercury deposition weeks (>250 ng/m^2) and the average annual deposition for those weeks for 13 MDN monitoring sites located in northeast North America.

Table 6. Regression equations and associated coefficients of determination (R^2) for annual (precipitation-weighted averages) and weekly mercury, sulfate, and nitrate concentrations in precipitation (highlighted data)

Annual data					"Enhanced" deposition weeks				
Sites	Comparison	Regression equation	R^2	Paired N	Sites	Comparison	Regression equation	R^2	Paired N
7 sites[a]	SO_4:NO_3	$Y = 0.78X + 0.11$	0.81	38	6 sites[b]	SO_4:NO_3	$Y = 0.68X + 0.11$	0.76	185
	SO_4:Hg	$Y = 3.00X + 3.18$	0.47	38		Hg:SO_4	$Y = 0.08X + 0.58$	0.25	185
	NO_3:Hg	$Y = 3.15X + 3.5$	0.39	38		Hg:NO_3	$Y = 0.07X + 0.37$	0.3	185
					Huntington (NY20)	Hg:SO_4	$Y = 0.14X + 0.45$	0.53	17
						Hg:NO_3	$Y = 0.14X + 0.16$	0.61	17
					Milford (PA72)	Hg:SO_4	$Y = 0.12X + 0.69$	0.42	22
						Hg:NO_3	$Y + 0.07X + 0.67$	0.52	22
Weekly data									
Huntington (NY20)	SO_4:NO_3	$Y = 0.47X + 0.98$	0.24	132	NY20 and PA72	Hg:SO_4	$Y = 0.9X + 0.54$	0.5	39
	SO_4:Hg	$Y = 3.04X + 3.21$	0.32	132		Hg:NO_3	$Y = 0.12X + 0.60$	0.46	39
	NO_3:Hg	$Y = 2.02X + 4.40$	0.12	132					
Milford (PA72)	SO_4:NO_3	$Y = 0.59X + 0.74$	0.5	92	Bridgton (ME02)	Hg:SO_4	$Y = 0.11X + 0.25$	0.39	37
	SO_4:Hg	$Y = 2.86X + 3.29$	0.51	92		Hg:NO_3	$Y = 0.07X + 0.29$	0.29	37
	NO_3:Hg	$Y = 2.61X + 4.23$	0.28	92					

[a]Precipitation-weighted data from Maine,Vermont, NewYork and PA collocated monitoring sites.
[b]MDN sites only, Underhill (VT) excluded.

On the other hand, the cluster analyses for mercury deposition and mercury concentration in precipitation isolate the Milford (PA72) site from virtually all other sites. This site receives higher deposition (2001-2002) than the other sites (Table 4), and significant ''enhanced'' loading events (21 weeks – both years, contributing ~57% of the annual deposition for each year). Annual precipitation-weighted mercury concentration is higher at this site than all other sites for these years. Although, the annual regional data and those for many individual sites show moderate or strong covariance between co-contaminants sulfate and nitrate in precipitation, only the Milford (PA72) and Huntington (NY20) data show relationships between mercury and co-contaminant acidic compounds. These sites lie downwind from the industrialized Midwest, the Great Lakes basin and the southern region of the east coast megalopolis; areas dominated by ozone and acid rain precursor and mercury emissions from utilities.

The data presented here cannot provide definitive evidence that recent emissions reductions have resulted in less mercury contamination of precipitation. It is not possible to pick out depositional or concentration signals, even though recent significant reductions in mercury emissions have occurred. The deposition and concentrations data for the period 2000–2002 (Tables 3 and 4) show substantial differences (in the order of 25–40 + %), over short time frames and distances, possibly attributable to the severe drought that occurred in areas of Maine and New Brunswick and to a lesser extent at most other sites. Although the volume-weighted annual concentrations of mercury varied during 2000–2002, the highest annual volume weighted concentrations occurred during the periods of least precipitation (2001). During this year, regional mercury emissions did not significantly increase in the northeast North America region, as indicated by the mercury emission data reported by Round and Irvine (2004). The 2000–2001 data for the six core sites (Maine, New Brunswick and Nova Scotia sites) suggest a reduction in mercury loading and a reduction in mercury contamination in precipitation, based on the number of ''enhanced'' deposition periods and the average deposition during these periods. Although the data clearly show a marked decrease in the number of ''enhanced'' mercury loading weeks

in 2001 and 2002, there is no compelling evidence to suggest that this recent pattern indicates anything other than year-to-year changes in precipitation patterns.

The Underhill data (1993–2002) show no significant changes in the concentration of mercury in precipitation or wet mercury deposition in Northern Vermont, however. This may be due to its location, lying outside the east coast megalopolis, and therefore not be influenced by emission reduction efforts in the megalopolis corridor. The Underhill site, and possible the Huntington site should be subject to orographically enhanced precipitation (respectively located 400 m and 500 m above sea level), and may receive more precipitation and higher levels of mercury contamination due to elevation, as described in an accompanying paper by Miller et al. (2005). Orographic effects also influence the amount of contamination in precipitation at these higher elevation sites. Based on prevailing winds and our current knowledge of pollution transport corridors in eastern North America, the western-most sites (Huntington (NY20), Underhill (VT), and St. Anicet (PQ04)) appear to be suitable as sentinels for changes in mercury deposition and concentration as a result of changes in continental-scale emissions. Viewed differently, data from these sites would probably not be useful for verifying the effectiveness of regional mercury emissions controls in the Northeast States and Maritime Provinces.

Periods of high mercury deposition should be important from a biological perspective. Atmospheric mercury loading can strongly influence direct mercury input to surface water, watershed release of mercury, water column mercury concentrations, methylmercury production, and bioaccumulation within the aquatic food web (Driscoll et al., 1994; 1995; Krabbenhoft and Goodrich-Mahoney, 2003). ''Enhanced'' mercury deposition events are common throughout the region and deliver significant mercury loads to receiving waters. More ''enhanced'' deposition events occur at coastal sites and sites located immediately downwind of major emission source regions than at northern or inland remote sites. During 1998, eight ''enhanced'' mercury deposition events were recorded regionally (at least one half of the sites recorded mercury deposition >250 ng/m^2). During June of that year, two regional events contributed 1062 ng/m^2 of mercury (average for all sites). The regional depositional events of May and June 1998 averaged 336 ng/m^2 and 423 ng/m^2, respectively. Mercury concentrations for these periods were not remarkable, indicating mercury loading may have been more a function of precipitation amount than mercury contamination in precipitation. The data clearly show that most ''enhanced'' deposition events occur during the late spring and summer. This is true for the five regional ''enhanced'' deposition events that occurred during 1999. Only one regional event occurred annually, during 2000 and 2001, and both were spring events. Only three ''enhanced'' mercury deposition events were recorded in 2002. None of these regional ''enhanced'' deposition events rivaled those of 1998. Based on the time when ''enhanced'' mercury deposition occurs, we would expect that the potential for direct mercury deposition to influence mercury methylation and bioaccumulation of methylmercury in receiving waters to be greater during the 1998–1999, than during the 2000–2001 period.

The expectation is supported by results from the METAALICUS experiments (METAALICUS – Mercury Experiment To Assess Atmospheric Loading in Canada and the US) which show that direct ''fresh'' (new) mercury input to a freshwater lake at the onset of the growing season results in rapid methylation and bioaccumulation (Krabbenhoft and Goodrich-Mahoney, 2003). Fresh mercury deposited on the watershed, on the other hand, is not exported in stream flow. Watershed throughput and methylation of mercury is relatively slow (on the order of several months) compared to direct input to the lake (roughly a month). Most of the mercury measured in stream flow is ''old'' mercury, deposited during past years. In northeast North America, most of the ''enhanced'' mercury deposition occurs between the 19th and 28th week (May through mid-July) in a year (Fig. 7). For all the sites, for all the years, roughly 31% of the annual deposition, or 2.3 ug/m^2 of mercury, falls during this period. Deposition of mercury for the mid-May through September period is on the order of 3.6 ug/m^2 ($\sim$49 + % of the annual for all years and all sites). Since the intensity and frequency of regional and site-specific ''enhanced'' deposition events vary from year-to-year and seasonally, and the amount of mercury directly delivered to surface waters varies accordingly, there

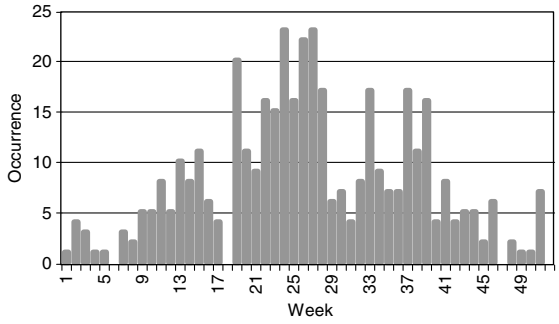

Figure 7. The weekly occurence of "enhanced" mercury deposition events (weekly deposition >250 ng/m^2) for 13 MDN monitoring sites located in northeastern North America (1996–2002 data).

should be significant year-to-year differences in within-lake reservoirs of methylmercury and mercury bioaccumulation. However, the importance of year-to-year and seasonal deposition patterns on mercury concentration in receiving waters and subsequent bioaccumulation in biota has not been fully explored.

Mercury deposition that occurs during the fall and early winter (October–December) may not be immediately important biologically, and early winter run-off and direct deposition of mercury contributes less to the near-term methylmercury burden of aquatic organisms than during the summer or spring. Although the amount of mercury deposited during the winter is far less than the spring and summer, roughly 5–20 + % during the winter versus 40–50 + % during the summer, mercury deposition during the mid- or late winter is important (Burgess et al., 1998; NESCAUM, 2003; Ryan et al., 2003). As noted by other authors (Burke et al., 1995; Shanley et al., 1999) elevated deposition events and snowmelt provide significant quantities of mercury to receiving waters, especially in high elevation snowbound terrains where snowmelt can carry enormous amounts of mercury to receiving waters.

A clear understanding of mercury deposition and reductions in mercury emissions, and the response of aquatic resources to potential reductions in mercury inputs cannot be provided without additional knowledge. There are no simple or general relationships between atmospheric mercury deposition and methylmercury contamination of aquatic resources, although it is clear that mercury deposition plays an important role in wildlife contamination. Mason et al. (2005) argue for a comprehensive monitoring framework to assess linkages between atmospheric inputs, atmospheric and depositional processes, watershed biogeochemical processes, and biological uptake of methylmercury. We agree with this approach and present data providing evidence that seasonal "enhanced" mercury deposition may significantly contribute to seasonal mercury loading patterns and that high loading periods influence mercury bioavailablity in aquatic ecosystems during periods of high biological activity. We note that all of the MDN sites in the region frequently receive "enhanced" mercury deposition and that some MDN sites also receive elevated concentrations of acid rain pollutants (nitrates and sulfates) with "enhanced" mercury deposition. Hrabik and Watras (2002) contend that direct input of mercury and sulfates significantly alter ("co-limit") bioaccumulation of mercury. Investigations along the lines of sulfate and mercury additions to freshwater microcosms provide evidence of enhanced methylation due to sulfate input (Gilmore et al., 1992). The presence of dissolved organic carbon (DOC) appears to further enhance mercury methylation (Driscoll et al., 1995; Wallshlager et al., 1996). These studies suggest that significant precipitation events during the late spring and early summer, such as the "enhanced" mercury deposition periods identified in this region, when accompanied by sulfates from acid rain and DOC released from watershed soils can increase bioaccumulation of mercury in biota of receiving waters. Other investigations show that changes in watershed acidification can result in unexpected changes in regional watershed biogeochemistry, such as increased DOC and acid neutralizing capacity (ANC) output (Stoddard et al., 2002). Arguably, these watershed changes can influence mercury release from watersheds, and that increased DOC output could lead to a greater efficiency in the transport of mercury and methyl mercury (Mierle and Ingram, 1991; Driscoll et al., 1994; Rencz et al., 2003). The model results presented by Loux (1998) suggests that organo-sulfur, as sulfhydryl groups, account for much of the mercury binding, indicating that DOC alone cannot explain ionic mercury binding and the partitioning of mercury released from watersheds. The

model results presented by Gbondo-Tugbawa and Driscoll (2002) further strengthen the importance of long-term chemical changes due to watershed acidification, organic S pools in soils, and long-term release of organic S–mercury complexes. We speculate that fundamental changes in watershed geochemistry caused by changes in watershed acidification will strongly influence processes controlling mercury transport through watersheds. We believe that watershed acidification and mercury transport processes must be studied together.

Conclusions

- Mercury deposition and the concentration of mercury in the region are not uniform. Coastal, western and southern sites receive more mercury in precipitation than other sites.
- Distinct depositional sub-regions can be identified: areas immediately downwind of the east coast megalopolis, areas immediately downwind of the Great Lakes Basin and the Midwest United States, and northern maritime and inland areas.
- "Enhanced" mercury deposition weeks (>250 ng/m^2), especially multiple weeks occurring during the late spring and summer, contribute significant mercury loading on a local (site-specific) and regional scale.
- Insufficient data are available from the atmospheric mercury monitoring network to determine if recent regional mercury emissions reductions have resulted in less mercury contamination in precipitation.

Disclaimer

The opinions of the authors and the results presented here do not necessarily reflect the views of the U.S. EPA.

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