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Wet deposition of mercury in the U.S. and Canada, 1996–2005: Results and analysis of the NADP mercury deposition network (MDN)

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ABSTRACT

One of the most critical measurements needed to understand the biogeochemical cycle of mercury, and to verify atmospheric models, is the rate of mercury wet-deposition. The Mercury Deposition Network (MDN) operates sites across North America to monitor total mercury in wet-deposition. MDN's primary goal is to provide both spatial and temporal continental-scale observations of mercury wet-deposition fluxes to support researchers, modelers, policy-makers and the public interest. MDN represents the only continental-scale mercury deposition database with a >10-year record of continuous values. This study provides analysis and interpretation of MDN observations at 10 years (1996–2005) with an emphasis on investigating whether rigorous, statistically-significant temporal trends and spatial patterns were present and where they occurred. Wet deposition of mercury ranges from more than 25 $\mu g\ m^{-2}$ yr in south Florida to less than 3 $\mu g\ m^{-2}$ yr in northern California. Volume-weighted total mercury concentrations are statistically different between defined regions overall (Southeast ≈ Midwest > Ohio River > Northeast), with the highest in Florida, Minnesota, and several Southwest locations (10–16 ng L^{-1}). Total mercury wet-deposition is significantly different between defined regions (Southeast > Ohio River > Midwest > Northeast). Mercury deposition is strongly seasonal in eastern North America. The average mercury concentration is about two times higher in summer than in winter, and the average deposition is approximately more than three times greater in summer than in winter. Forty-eight sites with validated datasets of five years or more were tested for trends using the non-parametric seasonal Kendall trend test. Significant decreasing mercury wet-deposition concentration trends were found at about half of the sites, particularly across Pennsylvania and extending up through the Northeast.

1. Introduction

The atmospheric emission, transport, and deposition of mercury (Hg) to the Earth's surface is complex, much more akin to sulfur, nitrogen, and carbon cycling than to other trace metals. Many large, direct sources of mercury to lakes, rivers, wetlands, and estuaries have been eliminated or greatly reduced in North America; thus, the dominant pathway for new mercury input is typically via atmospheric emission, transformation, transport, and deposition (Swain et al., 1992; Downs et al., 1998; Munthe et al., 2001; Dvonch et al., 2005; Keeler et al., 2006; Driscoll et al., 2007). The natural ecosystem problem with mercury arises because inorganic Hg is microbially transformed to mono-methyl mercury (MMHg) and biomagnified in the aquatic food chain. MMHg is a neurotoxin and teratogen (Mergler et al., 2007). Human and wildlife exposure to Hg is primarily due to the consumption of contaminated fish and may be enhanced close to Hg sources (Evers et al., 2007). Recent research has strengthened the direct link between new atmospheric Hg deposition and MMHg production in aquatic ecosystems and recovery of Hg-contaminated fisheries (Orihel et al., 2006; Munthe et al., 2007). Currently, 48 states in North America and eight Canadian provinces have fresh- and salt-water fish consumption advisories due to toxic levels of mercury (U.S. Environmental Protection Agency [EPA], 2005).

Recent knowledge of the atmospheric cycle of Hg has been summarized in the literature (Schroeder and Munthe, 1998; Mason and Sheu, 2002; Seigneur et al., 2004; Lindberg et al., 2007). Mercury in the atmosphere is emitted by both natural and anthropogenic sources (Mason et al., 2005). Although natural Hg sources are significant, human activities such as coal combustion, waste incineration, commercial product manufacture and disposal, metals refining, cement production, and artisanal gold mining result in the majority of annual emissions to the atmosphere (Pacyna et al., 2006). Emissions of mercury to the atmosphere occur

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in three primary forms: 1) elemental mercury (Hg^0), 2) gaseous oxidized mercury (GOM)[1] and 3) particulate-bound mercury (Hg_p). However, landfills and the ocean are known sources of organic mercury compounds to the atmosphere (Pongratz and Heumann, 1999; St. Louis et al., 2005). GOM is less volatile and more water-soluble than Hg^0, and is more likely to be removed by rain, absorbed by terrestrial surfaces, and adhered to atmospheric particulate matter. GOM and Hg_p are the primary atmospheric forms responsible for dry deposition of Hg (Lyman et al., 2007). However, recent work has suggested that Hg^0 dry deposition rates may be more significant than previously understood (Lindberg et al., 2004). Mercury dry deposition is a complex topic that is beyond the scope of this work. Broadly speaking, the deposition of Hg at any one location is complexly dependent on the amount, form, atmospheric chemistry, meteorology, and distance from emission sources. Current estimates indicate that even in remote background locations, deposition is enhanced by a factor of 3 ± 1 over preindustrial levels due to continued anthropogenic mobilization of Hg to the atmosphere (Lamborg et al., 2002).

Both wet and dry depositions are important processes for the movement of mercury from the atmosphere to land and water surfaces. A number of researchers have estimated that direct wet deposition accounts for between 50 and 90% of the mercury entering surface waters (Sorensen et al., 1990; Lamborg et al., 1995; Scherbatskoy et al., 1997; Mason et al., 1997; Landis and Keeler, 2002). The average concentration of mercury in precipitation samples collected at remote sites ranges between 1 and 7 ng per liter (ng L^{-1}) (Downs et al., 1998; Guentzel et al., 2001). Increasing trends in mercury wet-deposition were reported for Wisconsin and Minnesota from 1990 to 1995 (Glass and Sorensen, 1999). While Glass and Sorensen (1999) is a valuable record, they combined all of the annual precipitation-dependent deposition values for all sites and calculated trends using linear regression. Confusing the matter, Watras et al. (2000) report a decrease in bulk precipitation concentration at a single site in Wisconsin from 1993 to 1999. Keeler et al. (2005) conclude that there was not an obvious trend in mercury deposition from 1993 to 2004, based on data from one of the highest quality mercury wet-deposition records at a site in Northern Vermont. The application of statistics or anything quantitative was not offered to support the conclusion (Keeler et al., 2005). A study focused on U.S. New England States and Canadian Maritime Provinces provides an excellent statistical spatial analysis, but no temporal trends were presented (Van Arsdale et al., 2005). Butler et al., 2007, used linear regression statistics on annual MDN values to report decreasing temporal trends and random coefficient models to examine regional trends at fewer sites. Using annual MDN values and linear regression is less statistically robust for non-parametric and seasonally influenced data such as wet-deposition concentration and deposition rate.

Since wet deposition accounts for a large component of the Hg input to the environment, monitoring Hg in precipitation is the most direct way of assessing inputs from the atmosphere to sensitive aquatic ecosystems. The Mercury Deposition Network (MDN), coordinated through the National Atmospheric Deposition Program (NADP), is designed to study and quantify spatial and temporal trends in Hg deposition by precipitation and to provide data that support research on the environmental effects of mercury deposition. In 1995 following a year of field testing (Vermette et al., 1995), NADP began "transition phase" mercury monitoring at 17 sites (data reported by Vermette et al., 1996). Since 1996, an ever-increasing number of sites (currently more than 100 sites) have operated across the U.S. and Canada, each providing weekly integrated precipitation depth, Hg concentration, and wet-deposition data. MDN data are used for temporal trend determination, ecosystem model input, air model evaluation, regulatory accountability, and policy decisions. In the future, the MDN database will be particularly valuable to help evaluate the effectiveness of North American state, provincial, or federally mandated controls on mercury emissions to the atmosphere. Further, any significant alterations in the global mercury cycle due to shifting anthropogenic emission rates (e.g., Asia) and expected climate-change impacts should be observable over time by MDN.

The focus of this study is an analysis and interpretation of MDN observations at 10 years with an emphasis on investigating whether rigorous, statistically-significant temporal trends and spatial patterns were present and where they occurred. The complete weekly database and further information are available on the MDN web site (NADP, 2008a).

2. Sampling locations and methods

Fig. 1 shows the locations of all MDN sites that were active between 1996 and 2005, which includes urban, suburban, rural, and isolated sites. Since 2005, many new western MDN sites have been added (NADP, 2008a). All MDN sites use the same sampling equipment and follow the same strict sampling protocols, but sites are sponsored by many different individual organizations (usually federal, state, or local environmental agencies). MMHg is measured along with total mercury in precipitation samples collected at a subset of about 20 MDN sites; MMHg measurements are the subject of a future report and are not considered here. Further information can be found on the NADP web site (NADP, 2008a).

A special collector was designed for MDN based on the highly characterized IVL designed mercury wet-deposition sampler used in Sweden (Vermette et al., 1995). The MDN collector includes replacing the NADP's National Trends Network (NTN) collection bucket with a smaller insulated sampling chimney to hold a sample train (Lindberg and Vermette, 1995). The Hg sampling train consists of an average 123.6-mm diameter, borosilicate glass funnel, an acid-cleaned, wide-bore (3-mm) capillary tube, and a 2-liter (1/2 U.S. gallon) borosilicate glass bottle. Field operators receive a pre-cleaned sampling train for sample change out every Tuesday morning following NADP protocols. The weekly precipitation amount is also recorded for deposition determination. Glassware preparation and mercury analysis methods are more stringent modifications of U.S. EPA Methods 1669 and 1631 (U.S. EPA, 2007a,b; Frontier Geosciences, 2003a,b,c). In summary, sample equipment, collection, preparation, and analysis are completed using ultra-trace clean techniques.

Returned samples are treated with 0.2 normal BrCl (U.S. EPA, 2007a) to ensure complete oxidation of the mercury prior to analysis. Samples are shaken and left standing for at least 24 h at room temperature. Weeks with zero precipitation and no sampler lid openings serve as field blank samples. Weighed sample aliquots (50–100 mL) are pretreated with an aliquot of 20% hydroxylamine hydrochloride solution and tin chloride ($SnCl_2$) solution to chemically reduce free halides and the oxidized mercury to elemental mercury. The elemental mercury is then purged quantitatively from solution and analyzed by dual gold trap amalgamation and cold vapor atomic fluorescence (Tekran Inst. Corp.). Quantification is by peak area. Average blank values are determined for each analytical run and subtracted to determine sample mercury concentrations. The method detection limit for a 100 mL sample is approximately 0.1 ng L^{-1} (3 standard deviations above the reagent blanks). Low precipitation volume samples (<1.5 ml) have very high uncertainly and a miniscule impact on the volume-weighted mean

[1] The commonly used term is reactive gaseous mercury (RGM), which vaguely implies an undefined chemical behavior. The use of gaseous oxidized mercury is more accurate and used throughout this document.

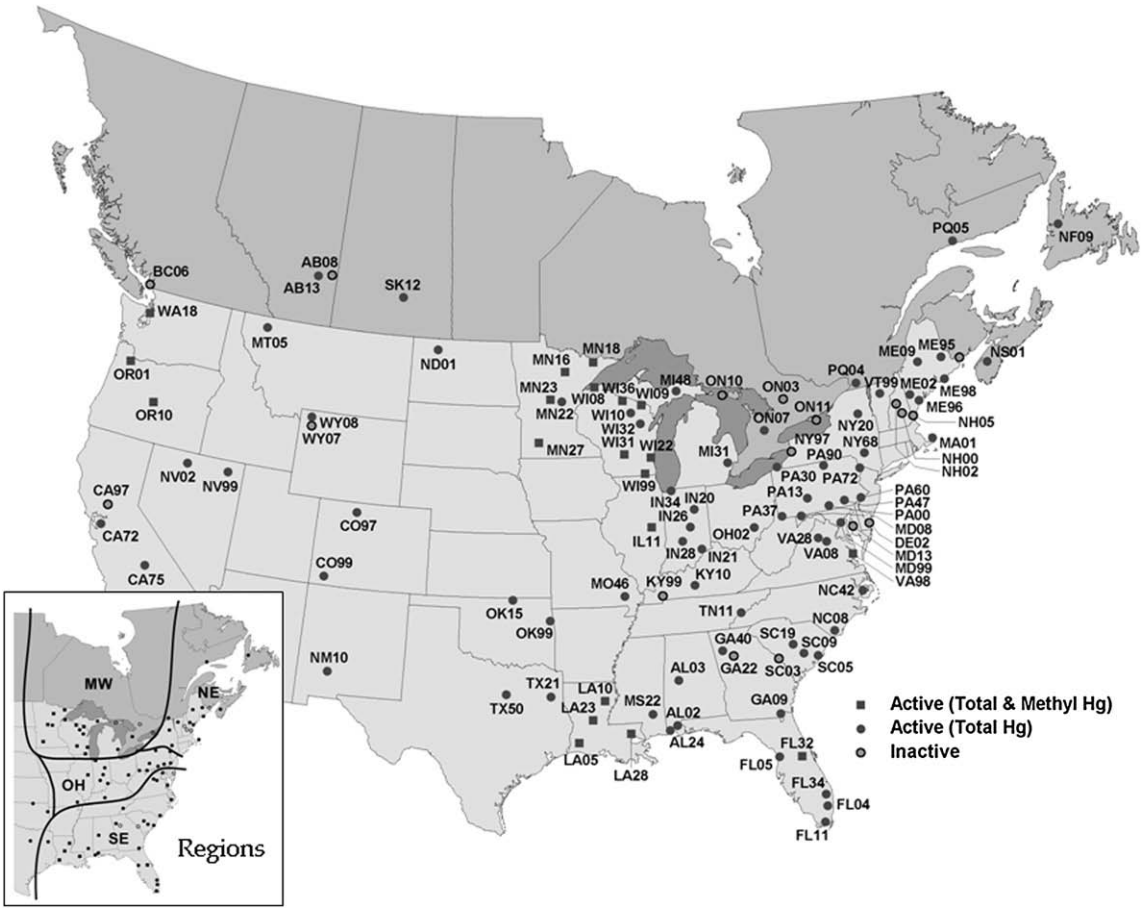

Fig. 1. Map of MDN locations for current and past sites as of December 2005, along with regional definitions used in the paper (inset).

concentration, and are therefore not reported. The weekly precipitation volume-weighted mean (VWM) concentration is determined using data only from samples considered valid (NADP, 2008a).

3. Quality assurance

The NADP internal and external quality assurance programs are rigorous and well documented (NADP, 2008b; USGS, 2007). Routine quality assurance procedures include analysis of laboratory bottle blanks, field blanks, and system blanks. The average amount of mercury in bottle blanks is 0.05 ± 0.05 ng. Field blanks average 0.06 ± 0.03 ng per bottle. System blanks, sample bottles placed out for collection but with no precipitation, average 0.06 ± 0.05 ng per bottle. Therefore, little contamination is picked up during the week while in the sampler housing. A typical (10 mm) rainfall event with a mercury concentration of 10 ng L^{-1} results in 127 mL of sample and 1.27 ng of mercury attributable to the rain. Therefore, most samples have mercury concentrations well above the detection limit.

Weekly analysis of a standard reference sample with a mercury concentration of 4.64 ng L^{-1} gave an average result of 4.38 ± 0.16 ng L^{-1}. The mean relative percent difference between weekly analyses from duplicate co-located samplers was 3.5 ± 3.6% (WA18 site). Finally, weekly laboratory spike recovery tests resulted in an average recovery of 100.2 ± 6.8%. Full laboratory quality assurance reports can be found at NADP (NADP, 2008b). Additionally, Wetherbee et al. (2006) reported the variability between collocated MDN samplers was between 3 and 14% for collector catch (rain depth) and approximately 11% for concentrations; deposition varies between 6 and 17%.

4. Statistical results and discussion

4.1. Statistical summary

The precipitation collection efficiency of the Belfort raingage compared to the National Weather Service (NWS) standard stick gage can be estimated from the NADP's Atmospheric Integrated Monitoring Network (AIRMoN) data (http://nadp.sws.uiuc.edu/AIRMoN). AIRMoN collects samples daily using the same protocols as the NTN. Precipitation is measured with both the Belfort and NWS raingages. The NWS gage requires a daily check to avoid errors due to evaporation. For more than 4000 precipitation events during all seasons through 2000, the Belfort amount averaged 94.9% of the daily NWS gage amount.

The MDN bottle precipitation catch is not significantly different from the Belfort raingage amount at warm weather sites (average collection efficiency = 98.8 ± 4.3%). At cold weather sites, the annual MDN bottle catch is significantly less than the Belfort raingage amount (efficiency = 87.1 ± 6.5%) due to the MDN sampler's lower sampling efficiency for snow. NADP completeness guidelines specify that at least 75% of the total precipitation measured by the Belfort raingage must be sampled in order to calculate valid seasonal or annual averages (NADP, 1994).

Table 1 provides a statistical summary of the annual VWM mercury concentrations and wet deposition for samples collected in the MDN network between 1996 and 2005. During this period, MDN has collected and analyzed nearly 28,000 weekly samples. These included 21,095 valid wet precipitation samples (75.5%) and 3876 valid dry samples (14%).

Table 1
The statistics for total mercury concentration in precipitation (ng L^{-1}) and wet deposition (μg m^{-2} week) for all MDN Stations are summarized.

Statistic	Number of Weekly Values		
Total Observations	27,776		
Invalid	2086 (7.5%)		
Valid Samples	25,690 (92.5%)		
Valid Samples (% of all samples)			
Wet Samples	21,095 (76.0%)		
Trace Precipitation	628 (2.2%)		
Dry Sample	3876 (14.0%)		
Other/Miscellaneous	91 (0.3%)		
Valid, Wet Samples			
N	21,095		
	Hg Concentration (Vol. weighted, ng L^{-1})	Hg Deposition (ng m^{-2})	
Mean	9.5 (vol. wt.)	479.2	
Percentiles	min	<0.1	0.2
	1%	1.3	19.5
	5%	2.2	49.4
	25%	4.7	154.8
	median	7.7	313.6
	75%	12.3	604.6
	95%	22.0	1515.4
	99%	33.7	2467.5
	max	772	4768

The VWM mercury concentration for all wet weeks, sites, and years is 9.5 ng L^{-1} (median = 7.75 ng L^{-1}) with 90% of samples falling between 2.2 and 22 ng L^{-1}. The average is not representative of North America because sites are overwhelmingly eastern locations. Less than 1% of samples are above 40 ng L^{-1}; however, concentrations above this level are almost always associated with very low volume rainfall events (<10 mm) that contribute very little to total mercury deposition. The network-wide median weekly integrated deposition rate is 313 ng m^{-2} (mean of 479 ng m^{-2}) and a 99% percentile of 2467 ng m^{-2} (Table 1). All values are highly skewed, following precipitation distributions.

The relationship between mercury concentration and precipitation amounts for all valid samples shows that observed concentrations decrease rapidly with an increasing rainfall amount of up to about 81 mm (3.2 inches), equivalent to the capacity of the 1-liter original bottle. Concentrations do not vary significantly between 81 and 162 mm (6.4 inches), equivalent to the capacity of the 2-liter bottles (later collection vessel). The data show that the mercury concentration in samples from 1-liter bottles (valid data 1996 and 1997) was not statistically different from 2-liter bottle concentrations (valid data 1999, 2000, 2001. $\alpha = 0.01$). This implies no bias due to overflow situations in the concentrations recorded. Additionally, these overflow situations are very rare, and are typically associated with hurricanes.

4.2. Geographic distribution of mercury concentration and deposition

First, it is important to note that the concentration and deposition of Hg in North America are higher than amounts reported from remote locations far from emission sources, such as open ocean sites. Furthermore, even at remote locations, deposition of mercury is estimated to be on average three times greater than historical preindustrial values (Lamborg et al., 2002). Thus the relative comparison of MDN regions and sites described below must be kept in a global perspective. Additionally, many of these same patterns have been noted by a number of other researchers (e.g., Schroeder and Munthe, 1998; Van Arsdale et al., 2005; Dvonch et al., 2005; Hall et al., 2005; Butler et al., 2007).

Mercury concentration and deposition can be distinctly different across the MDN network (Fig. 2a, b). VWM total mercury concentrations are greatest in the South and Southwest and lowest in the Northeast and along the Pacific Coast (Fig. 2a). VWM concentrations in the U.S. Gulf Coast region are consistently high, especially in Florida, and have been the subject of intensive study (Dvonch et al., 2005). Although sparse, the few MDN sites in the Southwest U.S. have consistently higher VWM mercury concentrations than regional VWM mercury concentrations in the Southeast (SE), Midwest (MW), Ohio River Valley (OH), and the Northeast (NE). For example, observations of VWM concentrations at MDN sites in New Mexico and in southwest Colorado are typically the highest of all sites on an annual basis. Several new locations within Oklahoma are also showing relatively high VWM concentrations, which are similar to the high amounts at Southwest sites. (Fig. 2a). The U.S. Midwest as a region has comparatively moderate to high concentrations year after year (9–14 ng L^{-1}). These higher concentrations span from Minnesota down to Texas. A northeastern triangle from North Carolina through Lake Ontario to Nova Scotia and beyond has lower VWM concentrations year after year, typically less than 9 and greater than 4 ng L^{-1}. At various locations in this region, however, concentrations are greater than 9 ng L^{-1} (see PA during 2001).

VWM total Hg concentrations are statistically different between defined regions, following from a Wilcoxon test (SAS, 2001, $\alpha = 0.1$). In terms of VWM concentrations, the Southeast is approximately equal to the Midwest, and both are greater than the Ohio River Valley, which is greater than the Northeast (Fig. 3a).

Pacific site VWM concentrations seem to fall into two groups. First, concentrations in the Oregon and Northern California sites (closed in 2001) are consistently very low (between 3 and 5.6 ng L^{-1}), and along with Newfoundland sites are usually the lowest values in the network. Second, observations at urban west coast sites, Seattle, Vancouver, and the San Francisco Bay Area show that concentrations are twice as high as those in rural Oregon and Northern California. In any event, although much attention has been focused on Asian long-range transport as a source of Hg to western North America (Jaffe et al., 2005; Weiss-Penzias et al., 2007), the MDN observations show that VWM concentrations on the West Coast are relatively low in comparison to sites located in high-density source regions of the eastern half of North America.

Mercury deposition follows VWM concentration for the most part across the MDN network, except in the Southwestern U.S. desert, where precipitation is relatively low. Wet deposition of mercury ranges from more than 25 mg m^{-2} yr in South Florida to less than 3 mg m^{-2} yr in Northern California (see earlier concentrations, NADP, 2008a). The highest deposition rates in the MDN are observed in the southeastern U.S., especially along the Gulf Coast to Florida. Consistently high concentrations combined with higher annual rainfall amounts in this region result in very high deposition rates. This condition is seen in the extreme in the case of hurricanes and other large precipitation events; very high deposition rates can occur over large areas. High deposition rates can extend throughout the Mississippi Valley. Lower deposition rates are seen in the Northeast, extending through New England and into Atlantic Canada. These Canadian locations show deposition rates as low as those in remote sites in Northern California and Oregon (4–6 ng L^{-1}). Midwestern deposition levels are moderate, with deposition rates in the 8 to 13 μg m^{-2} yr range. All concentration and deposition maps are available on the NADP web site (NADP, 2008a). Total mercury wet deposition is significantly different between defined regions ($\alpha = 0.1$): the Southeast is greater than the Ohio River Valley, which is greater than the upper Midwest, which is greater than the Northeast (Fig. 3b).

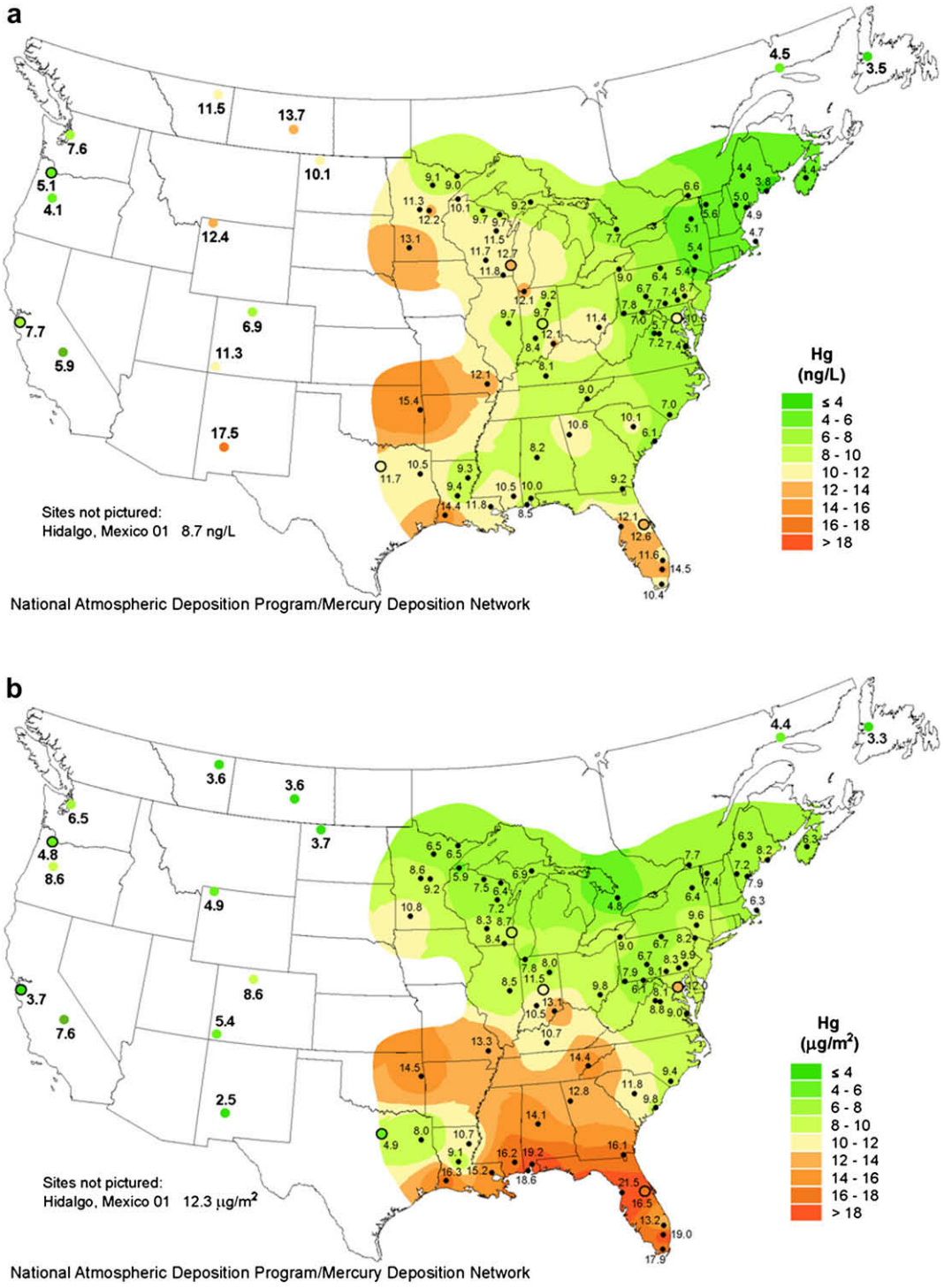

Fig. 2. a,b. Map of MDN mercury concentrations (ng L^{-1}) and deposition rates (μg m^{-2}) for the year 2005 (NADP, 2008a).

The regional spatial distribution pattern for mercury contrasts sharply with the other NADP/NTN spatial distribution of sulfate ion where the highest concentration and deposition is clearly the Ohio River Valley. Given the high-density of sulfate and mercury emissions in both the Midwest and Ohio River, a regional spatial pattern for mercury deposition should match that of sulfate. The contrasting results suggest that there is a much more complicated source–receptor chemistry and transport fate for mercury than for sulfate.

4.3. Annual, regional, and seasonal distribution of mercury deposition

In the eastern half of North America, concentrations of total mercury in precipitation and mercury wet deposition amounts show a strong seasonal pattern. The data in Fig. 3a and b shows that average summer (June–August) concentrations of mercury in rain are generally double the average winter concentrations (December–February), and that average summer wet deposition values are

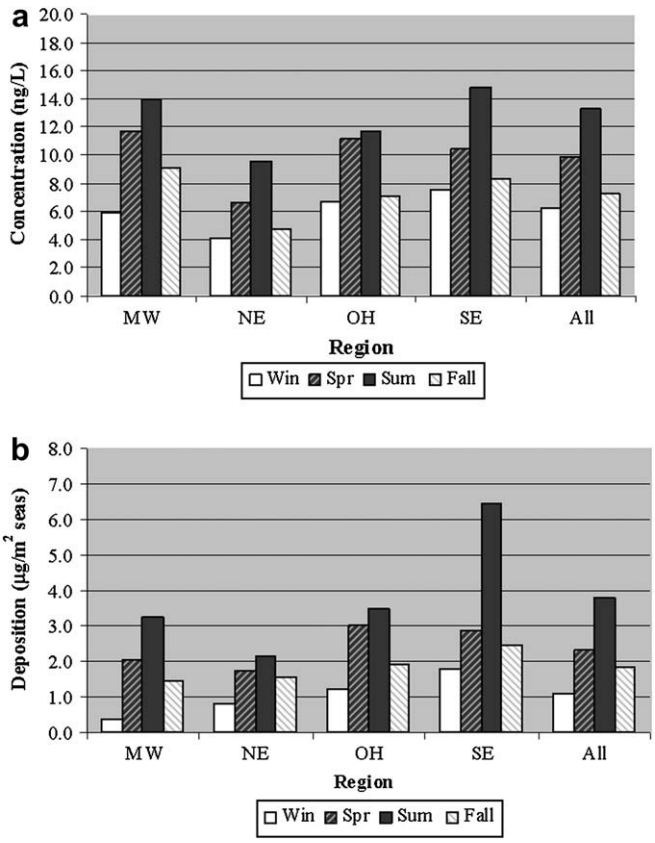

Fig. 3. a,b. Seasonal variation of weekly VWM mercury concentrations and the sum of deposition, 1996–2005. Summer is June–August, etc; with an increasing number of sites for the individual years. Following a Wilcoxon test, regions have significantly different concentrations and deposition rates for all seasons ($\alpha = 0.1$) and by season ($\alpha = 0.1$). See Fig. 1 for the delineation of the regions.

typically three times higher than deposition in the comparable winter period. Spring and fall averages for both concentration and deposition are in between the summer and winter values. This pattern was similar every year and for all eastern regions between 1996 and 2005. Higher mercury deposition in the summer months is a function of both the higher average Hg concentration in rain and higher summer precipitation amounts at most sites. Western sites were excluded because of limited data and the fact that seasonal precipitation patterns can be very different than those typical of eastern North America. These observations were confirmed by a non-parametric Wilcoxon test, which indicated that concentrations and deposition rates are significantly different (SAS, 2001, $\alpha = 0.1$) between summer and winter seasons for all sites as one group and for each region individually.

All eastern regions follow the same general annual concentration pattern (Fig. 4a), in which concentration is generally decreasing from year to year (see "trends" section below). The upper Midwest generally has higher concentrations year after year, but concentrations for both the SE and OH are higher during certain years. Consistently, the NE has the lowest averages. The regional deposition rates show a different pattern (Fig. 4b). No general trends are evident. The SE clearly has the highest deposition values. Comparatively, the NE has low deposition, given lower concentrations and moderate precipitation. The upper Midwest is very similar to the NE with somewhat higher concentrations and less precipitation. Deposition in the OH and SE appears higher during the later half of the record. Wilcoxon tests show significant differences ($\alpha = 0.1$) between annual depositions for all eastern sites as a group and for each region individually. Therefore, we can conclude that the depositions are not static from year to year.

One feature of the weekly mercury deposition record at many MDN sites is the occurrence of individual weeks with unusually high mercury deposition amounts, shown in both a timeline (Fig. 5), and in summary (Fig. 6a, b). A high deposition week is defined here as having rates greater than 1.5 µg m^{-2}, and typically features an average or above average mercury concentration along with much higher than average rainfall amounts. This can result in much of the mercury wet deposition for the entire year falling in a single summer week. Weeks with high mercury wet deposition usually occur during the warmer months (April through October).

Normalized per the number of total observations, the SE dominates the list of sites with a high occurrence of heavy deposition. For example, FL11 (tip of Florida, Fig. 1) has about 7% of its weeks with heavy deposition. Site FL04 is very close, followed by the other Gulf sites, then the Atlantic coastal sites. Notable exceptions to this coastal relationship include IN21, which is located within the Ohio River Valley. Also, OK99 and OK15 (2.5%) have high percentages, but both of these sites have relatively short records. Oaxaca, Mexico (OA02) is also of interest given its southern location and monsoonal precipitation; but only a limited number of observations are available.

4.4. Temporal trends in mercury concentration

Precipitation amount, Hg concentration, and wet deposition data were investigated for seasonal and annual trends. Sites with five or more years of data and greater than 75% valid data within the period of 1996–2005 (49 sites) were used in the Seasonal Kendall Trend Test (Gilbert, 1987). The seasonal Kendall is a generalized Mann-Kendall test, run for data containing seasonal cycles. The test is run for each season over all years independently. Trend magnitudes are median changes between all observations, with the analysis subdivided by season; i.e., summer observations are compared only against other summer observations, with different seasons combined for annual summaries. It is important to recognize that different sites may not have congruent time periods of record used for trend analysis since each site often has a unique starting date.

At the 90% confidence level, many stations show trends in concentration and deposition. Since deposition is the product of concentration and precipitation, deposition trend analysis and interpretation can be difficult to discern clearly. Because several MDN sites have significant tends in precipitation depth, the primary focus of this work is on trends in mercury concentration. However, the statistically-significant precipitation and deposition trends are included in Fig. 7.

Results of this weekly concentration trend analyses show a spatially-consistent decrease over most sites (Fig. 7). This is particularly true from Nova Scotia through New York and Pennsylvania. In addition to this consistent and clear decrease, the magnitudes of the decreases are also consistent; decreases range from 1 to 2% per year (see Table 2). This consistent trend continues down the eastern seaboard (through Georgia), and several sites in Indiana. Of note is the largest trend and the only trend greater than 2.5%: a 4.4% annual decrease at the NF09.

The Midwest and south Florida sites do not follow this general trend. Of these eight MW sites, no sites show a significant VWM concentration decrease over time. There is no statistical evidence for any trend in the MW region, except for one significant precipitation decrease. In other work, Glass and Sorensen (1999) have reported that mercury wet deposition increased at six Minnesota sites between 1990 and 1995. Annual total mercury wet deposition averaged 7.4 µg m^{-2}, and they calculated a statistically significant

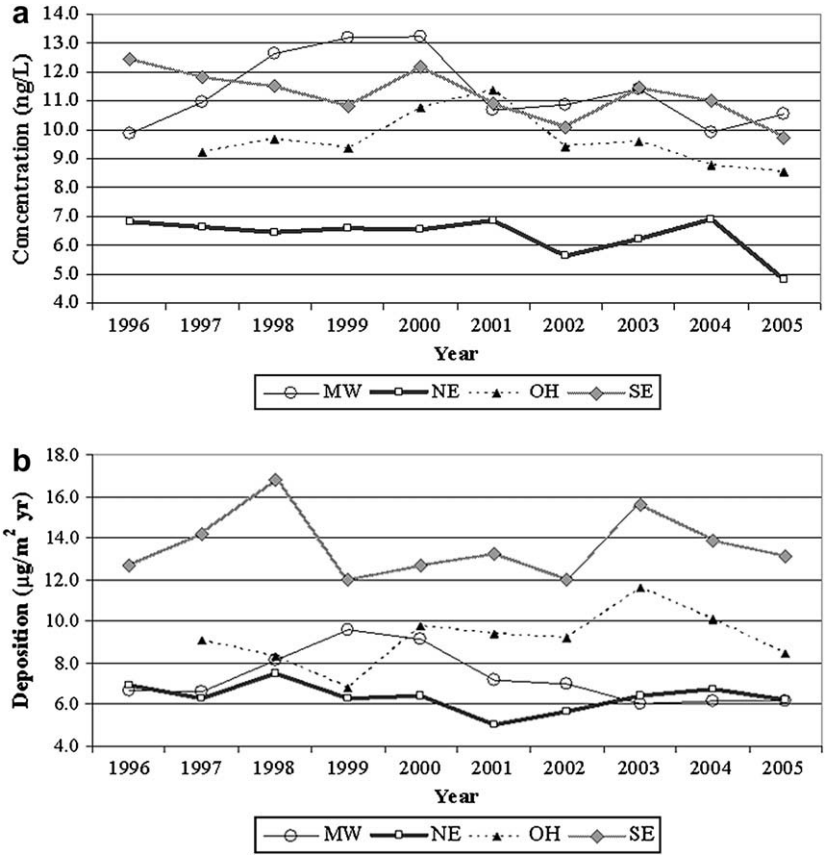

Fig. 4. a,b. Annual variation of mercury concentration and deposition sums in precipitation, 1996–2005, with an increasing number of sites for the individual years. Concentrations are volume-weighted. For each region, annual concentrations and deposition sums are significantly different over time (Wilcoxon test, $\alpha = 0.1$). See Fig. 1 for a delineation of the regions.

increase of 0.60 ± 0.06 µg m^{-2} yr (8.1% yr). This work was limited to a specific number of years (six) prior to the current study, focused on precipitation-dependent deposition rather than concentration, and calculated regional trends by grouping sites rather than individual site trends. The same areas (MN, WI) show no trends in concentration or deposition for the 1996 to 2005 period based on this study. Watras et al. (2000) showed a Midwest decrease in bulk precipitation concentration of 1.2 ng L^{-1} yr between 1993 and 1999. This significant trend is not seen in our analysis, but the two tests were for different time periods. Short-term trends could be obscured by longer-term data records, particularly with four initial years not represented in both datasets.

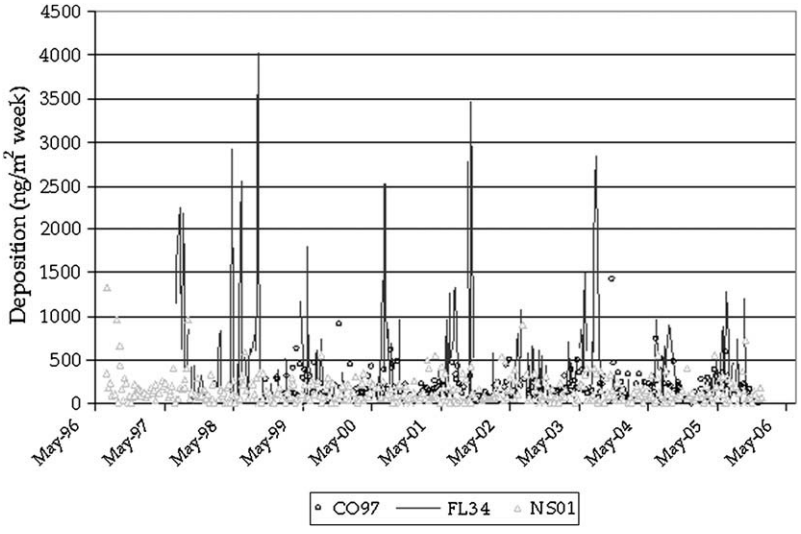

Fig. 5. Example timelines for weekly mercury deposition for several network sites, in ng m^{-2} per week.

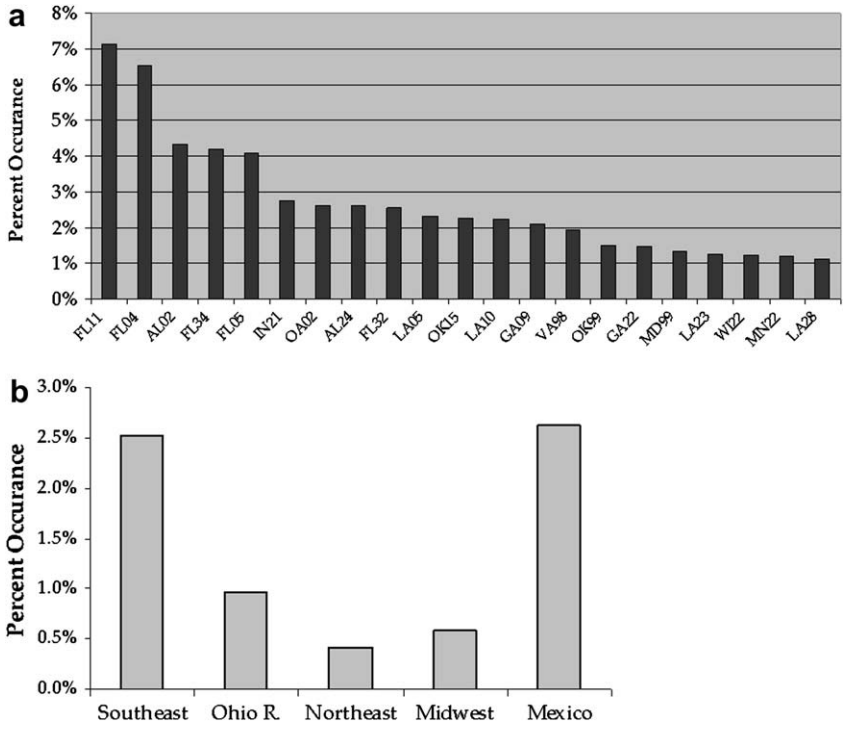

Fig. 6. a,b. Normalized occurrence of high deposition weeks across the network. High deposition events are wet deposition fluxes of greater than 1500 ng m^{-2} per week. Mexico is represented by only one site (Oachaca).

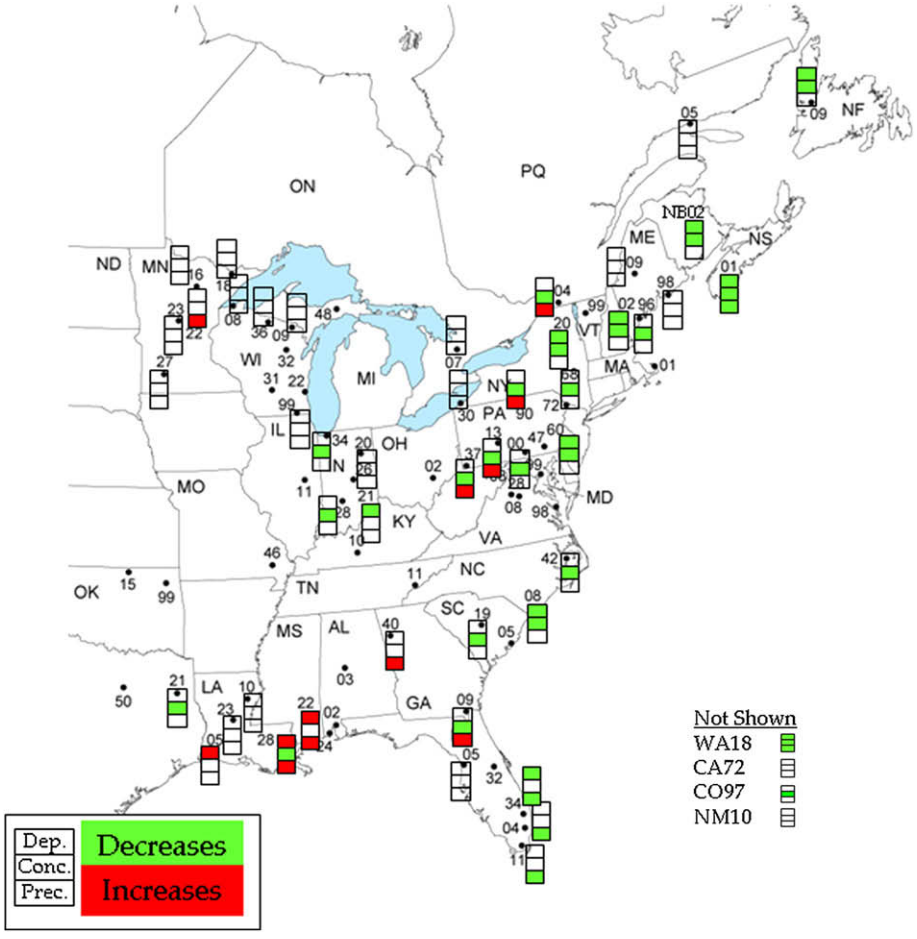

Fig. 7. Trends in mercury concentration, deposition, and precipitation between 1996 and 2005.

Table 2
Total mercury concentration in precipitation (ng L^{-1}) and concentration trends (%/year) at MDN stations. Values in italics represent datasets with between 50 and 75% completeness, normal-text values have 75% or greater data completeness with valid precipitation amounts for a minimum of 90% of sample period. Concentration and deposition averages are for 75% or greater values only (NADP, 1994). Trends not significant at a 1/4 0.1 are labeled as 'ns'. WA18 is an urban site.

Site	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	Avg	Trend (%)
CA72					9.7	9.1	6.6	13.4	9.3	7.7	9.3	*ns*
CO97				*11.2*	*8.9*	*8.0*	*7.7*	*8.1*	*7.7*	*6.9*		−1.7
FL04			13.8	12.2	15.7	13.1	14.2	16.4	14.7	14.5	14.3	*ns*
FL05			12.4	11.2	12.9	10.1	12.8	15.4	14.8	12.1	12.7	*ns*
FL11	14.1	14.7	12.9	11.5	14.6	14.1	12.4	16.4	14.7	10.4	13.6	*ns*
FL34			11.5	10.6	14.8	14.4	12.4	16.0	15.6	11.6	13.4	*ns*
GA09			11.7	11.9	10.6	13.3	9.9	11.7	10.2	9.2	11.1	−1.1
GA40					*11.7*	9.6	10.0	13.9	7.5	10.6	10.3	*ns*
IN20						11.7	11.2	11.1	11.1	9.2	10.9	*ns*
IN21						12.8	11.6	13.2	13.0	12.1	12.5	*ns*
IN28						10.2	*11.2*	9.7	9.3	8.4	9.4	−2.0
IN34						12.1	12.7	14.7	10.7	12.1	12.4	−1.6
LA05				12.8	9.9	9.6	*7.6*	12.2	11.1	14.4	11.7	*ns*
LA10				11.2	13.9	10.9	9.3	10.5	11.5	9.3	11.0	*ns*
LA23						10.0	8.6	11.9	10.3	9.4	10.1	*ns*
LA28				12.7	11.5	10.5	10.5	11.0	11.0	11.8	11.3	−1.2
ME02		*8.8*	6.4	6.4	7.5	7.0	5.6	8.5	6.2	5.0	6.6	−1.8
ME09		6.0	6.0	5.5	5.8	6.1	4.9	6.0	7.2	4.4	5.8	*ns*
ME96			8.4	7.3	6.6	7.3	5.1	6.7	10.2	4.9	7.1	−1.7
ME98	6.3	6.8	6.0	6.0	6.9	8.0	5.1	5.6	6.8	3.8	6.1	*ns*
MN16	10.3	10.8	11.3	12.4	12.5	9.1	11.4	11.2	9.0	9.1	10.7	*ns*
MN18	14.3	11.1	13.3	12.2	14.8	11.7	10.6	12.5	10.8	9.0	12.0	*ns*
MN23		11.7	14.0	17.0	11.5	13.6	11.2	11.8	10.5	11.3	12.5	*ns*
MN27		13.1	12.6	16.6	15.5	9.5	*17.1*	13.1	14.3	13.1	13.9	*ns*
MS22					*9.4*	8.9	8.6	12.4	11.6	10.5	10.4	*ns*
NB02		6.7	6.4	6.7	7.6	7.2	9.1	5.6			7.0	−1.9
NC08	11.8	10.4	11.2	7.9	9.6	10.4	8.9	8.7	8.8	7.0	9.5	−1.1
NC42	*9.4*	9.0	7.1	6.5	8.1	7.9	6.6	6.5	7.9	5.7	7.3	−1.5
NF09					*5.1*	5.0	4.1	4.0	4.0	3.5	4.1	−4.4
NM10		*17.1*	23.0	21.3	19.3	28.4	26.6	26.0	17.5	17.5	22.4	*ns*
NS01		6.3	5.3	5.0	5.2	6.6	5.5	5.0	5.2	4.4	5.4	−2.0
NY20					6.6	7.2	5.7	7.3	6.7	5.1	6.4	−2.5
ON07					*11.1*	7.3	8.7	9.6	7.7	7.7	8.2	*ns*
PA00						11.6	7.7	8.7	7.8	7.7	8.7	−2.2
PA13		9.2	10.2	9.3	9.2	14.8	9.3	8.5	8.0	6.7	9.5	−1.1
PA30					*12.5*	9.1	8.7	10.3	8.7	9.0	9.2	*ns*
PA37				*10.5*	11.3	9.9	8.3	10.1	8.2	7.8	9.3	−1.7
PA60					10.5	10.9	8.4	8.8	8.7	8.7	9.3	−1.8
PA72						9.4	8.3	7.6	9.6	5.4	8.1	−2.4
PA90		9.5	9.0	8.6	9.7	6.6	7.4	7.1	7.1	6.4	7.9	−1.4
PQ04			*9.9*	8.8	8.6	8.9	7.4	8.2	6.9	6.6	7.9	−1.5
PQ05			*5.6*	5.6	4.7	6.3	4.4	4.3	5.6	4.5	5.1	*ns*
SC19	*12.0*	10.8	11.6	10.5	12.0	10.1	9.7	10.5	10.0	10.1	10.6	−1.0
TX21	11.0	9.3	10.0	11.4	11.6	9.3	10.0	11.6	9.4	10.5	10.4	−1.2
WA18	*19.8*	17.5	5.9	8.5	9.7	6.3	9.2	6.8	8.6	7.6	8.9	−1.1
WI08	10.1	12.0	11.8	13.2	14.6	9.2	10.2	9.6	10.3	10.1	11.1	*ns*
WI09	9.8	10.0	11.7	13.6	10.9	11.0	12.4	9.7	8.7	9.7	10.8	*ns*
WI36	9.4	11.0	11.8	11.0	11.7	10.0	11.1	10.6	9.5	9.7	10.6	*ns*
WI99		10.6	13.5	10.6	18.0	13.0	12.2	13.3	10.5	11.8	12.6	*ns*

Interestingly, for Florida, where Hg concentration and deposition are relatively high, there are no significant concentration trends observed at any of the four stations. Unfortunately MDN measurements in Florida started after the closure or mercury emission control was installed on municipal and medical waste incinerators in the early 1990s, missing a potentially very significant decrease in deposition rates and concentration. However, at WA18 in Seattle, an abrupt decrease in Hg concentration and deposition after 1997 was observed and is linked to the closure of medical waste incinerators (MWI, Prestbo et al., 2006). Over the ten-year period of record, WA18 had significantly decreasing concentration (Table 2) and deposition. The abrupt decreasing Hg deposition change observed at WA18 was likely the case at urban and rural sites all across the U.S. when medical and municipal waste incinerators were shut down or controls were added.

In order to see if there is any statistically observable impact on deposition of mercury in western North America due to the relatively rapid increase in mercury emissions in East Asia (Pacyna et al., 2006), the authors looked at concentration and deposition trends in Seattle during 1998–2005 after local MWI sources were closed. While not ideal, Seattle is far enough west, distant from large Hg sources, and within the region known to be impacted by long-range Asian transport events (Jaffe et al., 2005; Weiss-Penzias et al., 2007). There was no trend in concentration, no trend in deposition, and no trend in precipitation. We can thus conclude that although very limited, there is no overwhelmingly rapid or significant impact due to offshore inflow of mercury.

Louisiana and east Texas MDN sites show only a few decreasing trends. The one mountain station in the analysis (CO97) showed a decreasing trend, which is based primarily on summer data, given the difficulty of winter sample collection at the 10,600-foot elevation.

Our work here compares very favorably with Butler et al., 2007. They observed decreasing concentration trends generally

consistent in rate with this work. The comparison is particularly good in the Northeast. However, we found no significant trends in the upper Midwest where they found decreasing trends in annual averages for their larger Midwest region. One explanation is the difference in regions; the Indiana sites compare quite favorably but are not in our Upper Midwest region.

Any increases in average precipitation could lead to a general decrease in concentration due to dilution of the washout loading. There are a few significant precipitation increases at some locations in the NE and SE (NY, PA, GA, and LA, Fig. 7), but this is not the case for most of the decreasing concentration sites.

Deposition trends are much more complicated than trends in Hg concentrations, because increasing precipitation over time with constant concentration would lead to increases in deposition, and vise-versa. Given this qualification, a significant number of eastern sites do have decreasing deposition trends. Among the tested sites, six Northeastern sites do have significantly decreasing concentration and deposition trends without observed precipitation trends. The spatial consistency of these complementing observations at multiple NE sites further suggests a significant finding. One possible explanation could be mercury emission reductions in the upwind high-density Midwest source region, but this is difficult to test without accurate trends in emissions, or a more extensive monitoring network.

5. Conclusions

The MDN is a continental-scale network of more than 88 active sites with 10 years of continuous operation to provide status and trends information on mercury precipitation chemistry. Sampling and analysis methods and the concomitant quality assurance oversight are based on well-established scientific principles that have been programmatically documented. The VWM mercury concentration for the network is 9.5 ng L^{-1} (median = 7.7 ng L^{-1}) and mean weekly deposition is 479 ng m^{-2} $week^{-1}$ (median = 314 ng m^{-2} $week^{-1}$) calculated for the 21,095 valid non-trace weekly total mercury wet-deposition values over 10 years. The spatial distribution of observed mercury concentration and deposition is revealing. Based on the delineated regional definitions, mercury deposition rates are greatest in the U.S. Southeast, followed by the Ohio River valley, then the U.S. and Canadian sites in the Midwest and Northeast (OH > MW for VWM concentrations). This contrasts with the spatial distribution of sulfate ion deposition, which is maximized in the Ohio River valley, suggesting a much more complicated source–receptor relationship for mercury. A prominent seasonal pattern is seen for the eastern sites and all regions with VWM concentration and deposition both higher in the summer versus the winter. Non-parametric concentration trends were significant at about half of the sites with approximately the same magnitudes (downward 1–2% per year), and over cohesive regions of the NE and the Ohio River valley. Five NE sites show clear decreases in both concentration and deposition at the same time. Therefore, with standardized and quality-assured long-term measurements, it is evident that certain regions have defined trends of small magnitudes. Also, for many regions there is now adequate baseline monitoring data to determine shifts in deposition with any new regulations on sources. At this point, however, we still must conclude that during these years, mercury concentration has shown modest change in North America.

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