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Regional precipitation mercury trends in the eastern USA, 1998–2005: Declines in the Northeast and Midwest, no trend in the Southeast

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Abstract

Mercury emissions in the USA declined between the 1990s and the beginning of this decade, largely due to the closure of municipal and medical waste incinerators. Declines in emissions have been greater in the Northeastern (NE) than the Midwestern (MW), and Southeastern (SE) regions of the eastern USA. During this time, global emissions of mercury have declined in Europe but have not declined in Asia and Africa. We have examined the patterns and trends in annual volume-weighted mean precipitation concentration for 33 Mercury Deposition Network (MDN) sites in the eastern USA, for the period 1998–2005. Individual linear regressions indicate that, 11 of 12 sites in the NE and all nine sites in the MW show declining patterns in mercury concentration, but only nine of these 21 sites show statistically significant downward trends ($p<0.10$). For the SE, seven of 12 sites show a decreasing pattern, but only two sites show significant downward trends ($p<0.10$). Random coefficient models were used to test the trends in mercury concentration for each of the three regions as a whole. These results for the NE and the MW are statistically significant ($p<0.01$), and show annual declines of $-1.70\pm0.51\%$ (S.E.) per year, and $-3.52\pm0.74\%$ per year for the NE and MW, respectively. The SE region as a whole shows no significant trend in mercury concentration during the same period. Different mercury transformation and deposition processes in the SE may account for these results. A comprehensive mercury emissions record was not available for the same 1998–2005 time period as the precipitation concentration analysis. Therefore, while the empirical relation between changing regional mercury emissions and changing precipitation mercury concentrations is suggestive, it cannot be confidently assessed at this time.

Keywords: Mercury wet deposition; Mercury emissions; Mercury precipitation concentration

1. Introduction

Mercury deposition patterns and the sources of mercury deposition have become a major environmental issue, largely because of the health effects

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associated with the ingestion of methyl mercury by humans through the consumption of fish (National Research Council, 2000; US EPA, 2001). More recently, the impact of methyl mercury on other organisms such as fish-eating birds and mammals, and insectivorous songbirds has also become an issue (Evers et al., 2005, 2007; Rimmer et al., 2005). The pathway from mercury emissions to deposition involves a number of complex processes (Cohen et al., 2007). There are various forms of mercury—elemental (Hg^0), particulate ($Hg_{(p)}$), and reactive gaseous or divalent mercury (RGM or Hg (II))—present in the atmosphere. There are also transformations of these mercury species, as well as reemission of mercury to the atmosphere, largely as Hg^0, after it has been deposited.

At present, in the USA, wet mercury deposition has been measured in a national network by the NADP/MDN (National Atmospheric Deposition Program/Mercury Deposition Network (NADP, 2006)). This network began operation in 1996 and by 1998 had 32 sites operating. As of October 2007, there were 97 sites in the USA and five in Canada (Gay, 2007), with some sites measuring not only total mercury but also methyl mercury. There is currently an initiative coordinated by NADP to form a national air monitoring network which will measure event-based mercury wet deposition, air concentrations of mercury in its gaseous and particulate forms, and meteorological and land-cover variables needed for estimating mercury dry deposition (NADP, 2007).

Direct, anthropogenic mercury emissions in the USA have declined from the early-to-mid 1990s to 2002, primarily due to decreased emissions from municipal and medical waste incinerators (Fig. 1). These reductions can be attributed to efforts to reduce mercury in the waste stream, added pollution control equipment, and the closure of many facilities. Declines in emissions have occurred in all mercury species (Fig. 2) for the eastern USA (and Southeastern Canada) with greater declines in the Northeast (NE) than in the Midwest (MW) and Southeast (SE). From 1998 to 2002, mercury emissions in the Northeastern USA have declined from 15.9 to 4.7 tons (NESCAUM, 2005). Based on an analysis of ambient concentration data, Sigler and Lee (2006) estimate that emissions in the Northeastern USA have declined by 20% between 1999–2000 and 2003–2004. A comprehensive nationwide, detailed mercury emissions record from the US EPA is only available up to 2002, and

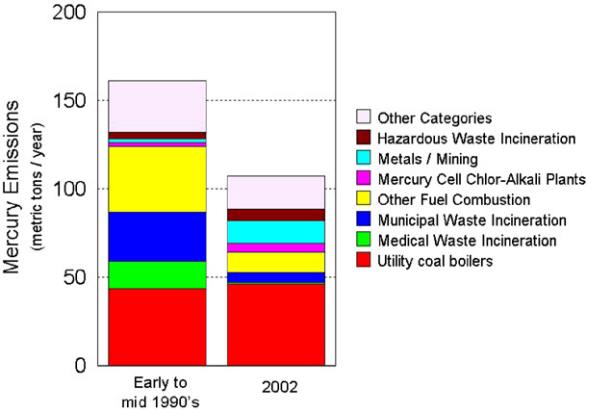

Fig. 1. Estimated US anthropogenic mercury emissions in the early-to-mid 1990s and 2002. As described in the methodology section, the estimates for the earlier period are based on US EPA data as described in Cohen et al. (2004) and the 2002 estimates are based on the US EPA 2002 National Emissions Inventory (US EPA, 2007).

assessments are only made at 3-year intervals (1999, 2002, etc.). Further reductions in North American and European mercury emissions are expected due to government regulation, but global emissions may rise due to the increases in emissions from Asia and Africa (Pacyna et al., 2006). Mercury emissions from India have been growing at about 2.5% per year (EPRI, 2004). Chinese emissions have been steadily increasing at 5% per year or greater (Zhang et al., 2002) and were estimated to be around 536 (Streets et al., 2005) to 600 (Pacyna et al., 2006) tons per year in 1999–2000. These emissions may have recently stabilized to a certain extent due to the installation of coal combustion control technologies such as flue gas desulfurization and particulate removal. Asian emissions of 1179 metric tons, excluding Russia, accounted for over half of the global anthropogenic mercury emissions of 2190 metric tons in 2000 (Pacyna et al., 2006).

While there are insufficient data to quantify recent trends in mercury emissions beyond 2002, we can assess whether wet mercury concentration and deposition have been changing at MDN sites that have operated for an extended period. In this analysis, we examine changes in mercury concentration in precipitation in the eastern USA from 1998 to 2005.

2. Study area

We have focused our analysis of patterns in MDN mercury concentration in precipitation on the

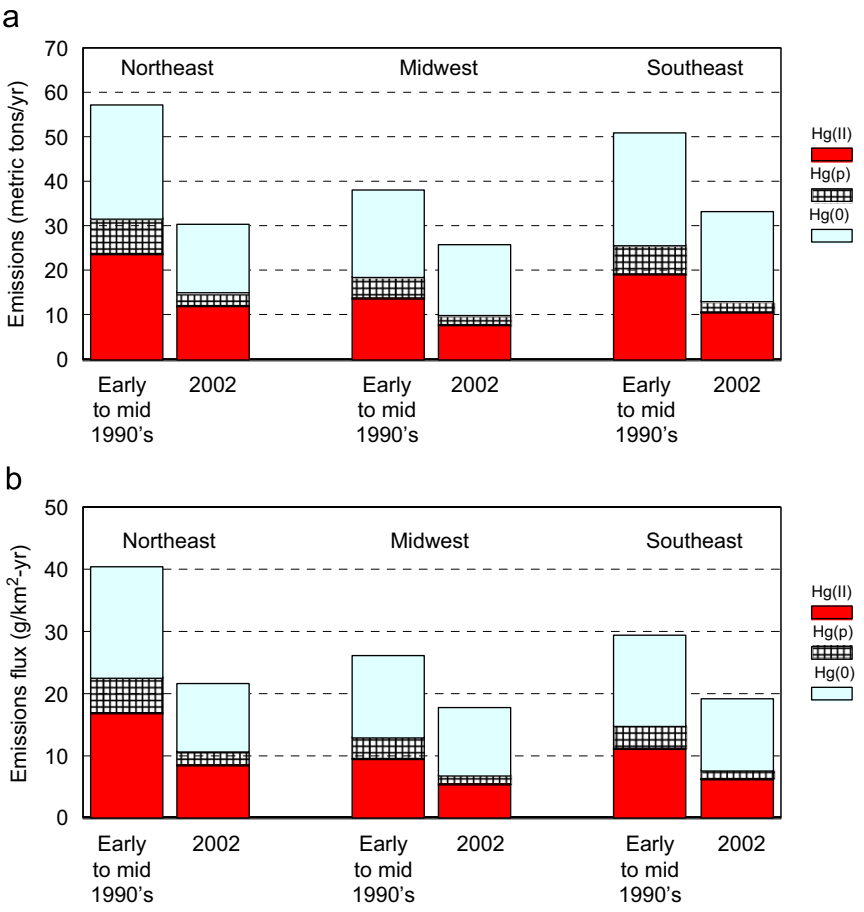

Fig. 2. Estimated changes in regional mercury emissions between the early-to-mid 1990s and 2002 in terms of (a) mass and (b) mass flux. The regions are defined as shown in Fig. 3.

eastern half of the USA where the data are more extensive. We have included 33 MDN sites, most of which have been operating since 1998 (Fig. 3), including 12 sites in the NE (including three sites in eastern Canada), 12 sites in the SE, and nine sites in the MW. Twenty-two of the 33 sites have a complete 8-year record from 1998 to 2005. Due to sites being established after 1998 or not having complete records, three sites (PQ04, LA28, IL11) are missing 1 year of data, five sites (PA60, NB02, PA37, LA05, LA10) are missing 2 years of data, and NY20 is missing 3 years of data.

3. Methodology

3.1. Statistical analysis of mercury wet deposition data

Volume-weighted annual concentrations ($ng\,l^{-1}$) and annual depositions ($\mu g\,m^{-2}$) of total mercury for each site were obtained from the NADP/MDN (http://nadp.sws.uiuc.edu/mdn/). In addition, weekly MDN data for each site were aggregated to volume-weighted means for May through September, when weekly concentrations and depositions are generally higher than for the remaining months of the year. Fig. 4 shows higher concentrations and depositions for all three regions during the summer (June–August). Spring (March–May) shows the second highest seasonal mercury concentrations at the MDN sites for all three regions. The warm-season record eliminates data from the winter months when precipitation collection efficiencies, especially at Northern sites, are lower due to blowing snow and other cold-weather issues.

The MDN includes an extensive Quality Assurance Program for both laboratory and field operations. Data quality for the annual records meet the criteria that valid samples are available for at least

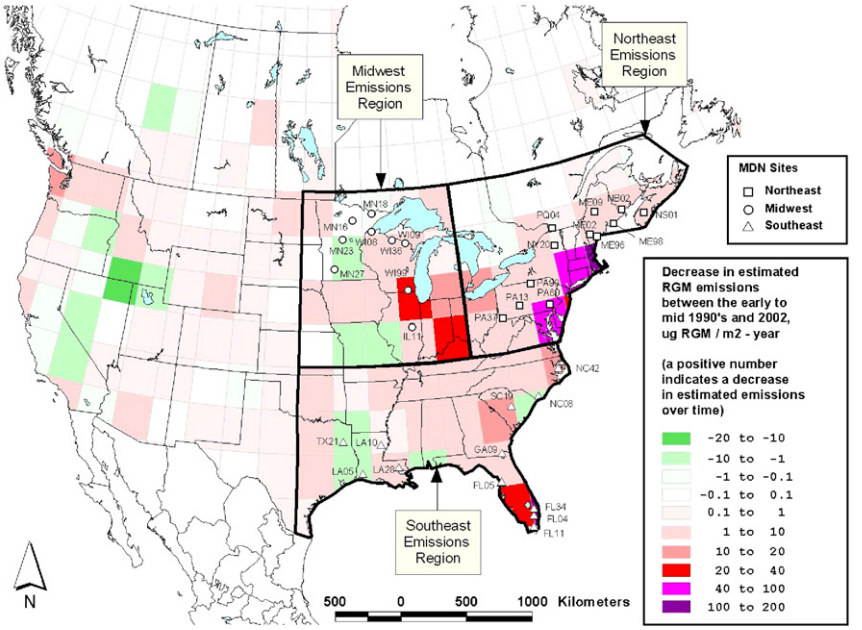

Fig. 3. MDN sites/regions analyzed and reactive gaseous mercury (RGM) emissions flux changes between the early-to-mid 1990s and 2002. The emissions inventories described herein were allocated to a $3^\circ \times 3^\circ$ grid to produce the geographical distribution of RGM changes shown.

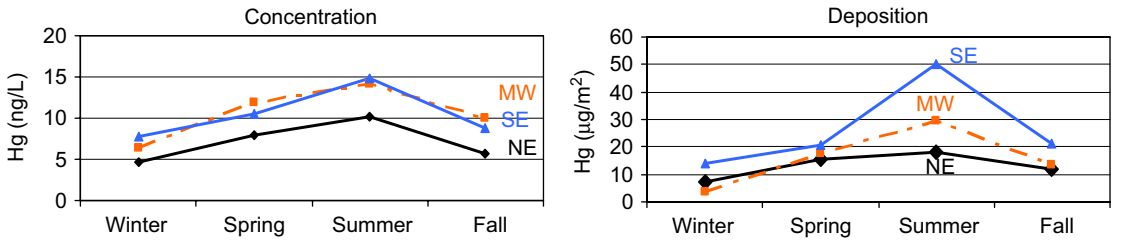

Fig. 4. Volume-weighted mean seasonal concentrations and mean depositions measured at MDN sites for the Northeastern (NE), Midwestern (MW), and Southeastern (SE) regions for 1998–2005.

90% of the year. Valid samples must have a 75% collection efficiency for precipitation volume. Field and laboratory analytical quality control, system audits and other quality assurance measures are detailed in Welker (1997), and are available at http://nadp.sws.uiuc.edu/QA/.

First, to see initial patterns we examined individual sites by performing a least-square, linear regression on the annual volume-weighted mercury concentrations vs. year, annual mercury deposition vs. year and annual precipitation vs. year. We did the analyses of concentration and deposition data without including the effect of precipitation, and also including precipitation as a main effect in the regression analyses.

We then combined the concentration data across all sites within each region and used random coefficient models, to examine overall patterns specifically for each region or group of sites (Littell et al., 1996; Singer, 1998). We applied the random coefficient model with site as a random effect to both the annual record and the warm-month record (May–September) for concentration. Random coefficient models were also run on annual and warm-month deposition data, but we will focus on the concentration results which eliminate the complications introduced by precipitation variability. The models controlled for precipitation by including precipitation as a fixed effect. The mixed procedure in SAS was used to fit these models (Littell et al., 1996).

In a linear regression model, the residuals are assumed to be independent and normally distributed with constant variance. In a random coefficient model, there are residuals at more than one "level" (between and within sites) which require similar assumptions. These assumptions have been met for all the models presented here. In regular linear regression analysis, the coefficients are assumed to be fixed parameters. In a random coefficient model, it is assumed that one or more of these parameters are a random sample from the population of possible coefficients. To be more specific, since each site has annual data for up to 8 years, a separate regression can be run for each site. The regression model for each site then represents a random deviation from the model for the population of all sites. Site is considered a random effect because we are not addressing specific sites; instead we consider that these sites are a random sample of all the possible locations in a particular region.

We also ran a third set of models combining all sites and regions together in order to compare regions with one another. Random coefficient models combined data ($n = 238$) from the three regions (NE, $n = 85$; SE, $n = 85$; MW, $n = 67$) which were fit using the mixed procedure in SAS (Littell et al., 1996). We entered "region" as a fixed effect in the model since the regions are not regarded as a random sample of all the possible regions. We used interaction terms (region × year) to test for significant differences in trends between the regions. A significant interaction term means that the observed trends are different between regions. We also included precipitation as a fixed effect since precipitation amount can influence concentration.

As mentioned earlier, not all sites have a complete 8-year record. The random coefficient model takes the varying number of years for each site into consideration by giving more weight or influence to sites with more information (Snijders and Bosker, 1999). Further discussions of the random coefficient model and its use in assessing trends in atmospheric pollutants are available (Singer, 1998; Snijders and Bosker, 1999; Raudenbush and Bryk, 2002; Butler et al., 2005).

In assessing whether a regression is statistically significant, we use $p < 0.10$ for the linear regressions for individual sites, and $p < 0.05$ for the random coefficient models. The relatively low number of observations for the individual site regressions ($n = 8$ or less) have relatively little statistical power, compared with the random coefficient models with much higher sample numbers, and much higher statistical power.

3.2. Mercury air emissions inventories

We assembled spatially resolved anthropogenic mercury air emissions inventory data for the US and Canada for two periods: the early-to-mid 1990s and 2002. The US and Canadian emissions data for the earlier period were that used in the Cohen et al. (2004) atmospheric modeling analysis. Details regarding this inventory, based on US EPA and Environment Canada data, are provided there. For the 2002 period in the USA, the US EPA 2002 National Emissions Inventory (NEI) was utilized (US EPA, 2007). A summary of the USA emissions estimates for the two periods is shown in Fig. 1. It is believed that the reductions shown in this figure for municipal and medical waste incinerators are relatively accurate. However, emissions changes shown in two other source categories—"other fuel combustion" and "metal/mining"—may be artifacts of changing emissions inventory methodology. The "other fuel combustion" category includes emissions from commercial and industrial boilers burning coal and other fuels, and the emissions shown for this category for the earlier period are consistent with the inventory presented in the US EPA Mercury Study Report to Congress (1997). The emissions in this category in the 2002 NEI are significantly less, and it is not clear if this apparent decrease is real or resulted from a different categorization of sources or different emissions estimating assumptions (e.g., different emissions factors). For the "metals/mining" category, it is believed that the inventory for the earlier period significantly underestimated emissions from gold mining, electric arc furnaces and other metallurgical operations.

For the 2002 period in Canada, emissions estimates for major point sources in a 2000 inventory (Environment Canada, 2004) were updated using data from the Canadian National Pollutant Release Inventory (Environment Canada, 2007). The latest available area-source emissions estimates that could be obtained for Canada are for the year 2000, aggregated at the provincial level. This spatial resolution is too coarse to be useful in this analysis, and so these area-source emissions estimates were geographically distributed following the spatial pattern of area-source emissions in the

1995 Environment Canada inventory used for the earlier period.

RGM is more susceptible to wet deposition than other forms of mercury due to its much higher water solubility. The spatial distribution of RGM emissions changes between the two periods is shown in Fig. 3. Here, the emissions have been aggregated to a $3^\circ \times 3^\circ$ grid, and the changes have been expressed as fluxes by normalizing by the area of each grid square (excluding lake and ocean regions). We note that in addition to the inventory issues discussed above, the accuracy of this spatial distribution is limited by uncertainties and potential inconsistencies in the "speciation" of emissions, i.e., the apportioning of emissions from a given source into elemental, reactive gaseous, and particulate mercury forms.

4. Results

4.1. Linear regression for individual sites

Linear regressions for the MDN annual concentration data for 33 sites show a downward pattern with time (1998–2005) in most cases. This pattern holds whether the regressions include precipitation as a main effect or not. Fig. 5(a) shows the patterns (normalized by presenting concentrations as percents of the long-term means) for the linear regressions of concentration vs. year, not corrected for precipitation effects, for the 33 sites in this study. The downward concentration patterns for the NE and MW sites are clear, but only four of the 12 sites in the NE, and five of the nine sites in the MW have significant downward trends ($p<0.10$). The pattern is less clear for the SE with a mix of downward (seven sites) and upward patterns (five sites). Only two sites have a statistically significant trend, and they are downward trends in concentration over time.

Deposition patterns from the linear regressions are complicated by the effect of precipitation. If precipitation is not included as a main effect in the linear regression models (Fig. 5(b)), the NE sites exhibit only small changes (both positive and negative), and none are statistically significant at $p<0.10$. This can be explained by a pattern of increasing precipitation (Fig. 5(c)), during the period of study when most sites are experiencing a decline in concentration. Seven of nine sites in the MW show a declining pattern in deposition (Fig. 4(b)), with four of nine sites showing significant declines ($p<0.10$). Most sites in the MW show little change in the precipitation pattern (Fig. 4(c)). SE deposition shows an overall pattern of increasing deposition. However, one site shows a significant downward trend and one site a significant upward trend in deposition. The remaining sites show no significant trend ($p<0.10$). The precipitation pattern for the SE includes six of 12 sites showing increasing precipitation but only two of these sites are significant (GA09 and FL05). The six remaining sites show a negative, but not significant, pattern of precipitation. If precipitation is added as a main effect the linear regression slopes for deposition (not shown) are similar to the concentration patterns in Fig. 5(a) for the three regions.

4.2. Random coefficient models by region

An overall regression that incorporates all sites within a region was obtained using the random coefficient model as discussed above. An overall estimate of the linear relation between mercury concentration and year was obtained for each region controlling for precipitation as a fixed effect. Site-to-site variability was also incorporated into the model as a random effect. These linear relations represent an "average" regression based on all the sites in a region. The "average" is weighted by the number of years in the record for each site. The deposition results are quite similar to the concentration results because precipitation effects are included in the random coefficient models. We will focus our analyses on the concentration models where precipitation effects are less important. We also tested for quadratic-year effects (i.e., a nonlinear or variable slope model), but these were not significant and thus removed from the model.

For two of the three regions examined, the trends are highly significant ($p<0.01$) for both the annual concentration models and the models based on concentrations during the warm months of the year (Table 1). The NE and MW regions show declines in Hg annual precipitation concentrations of -1.70% (0.51% S.E.) per year and -3.52% (0.74% S.E.) per year, respectively. The concentration pattern is not significant (the regression slope is not significantly different from 0.00) for the SE region. Graphic displays of the concentration trends for the NE, MW, and SE regions are shown in Fig. 6. The individual site regressions and the overall regional trend are displayed for each region.

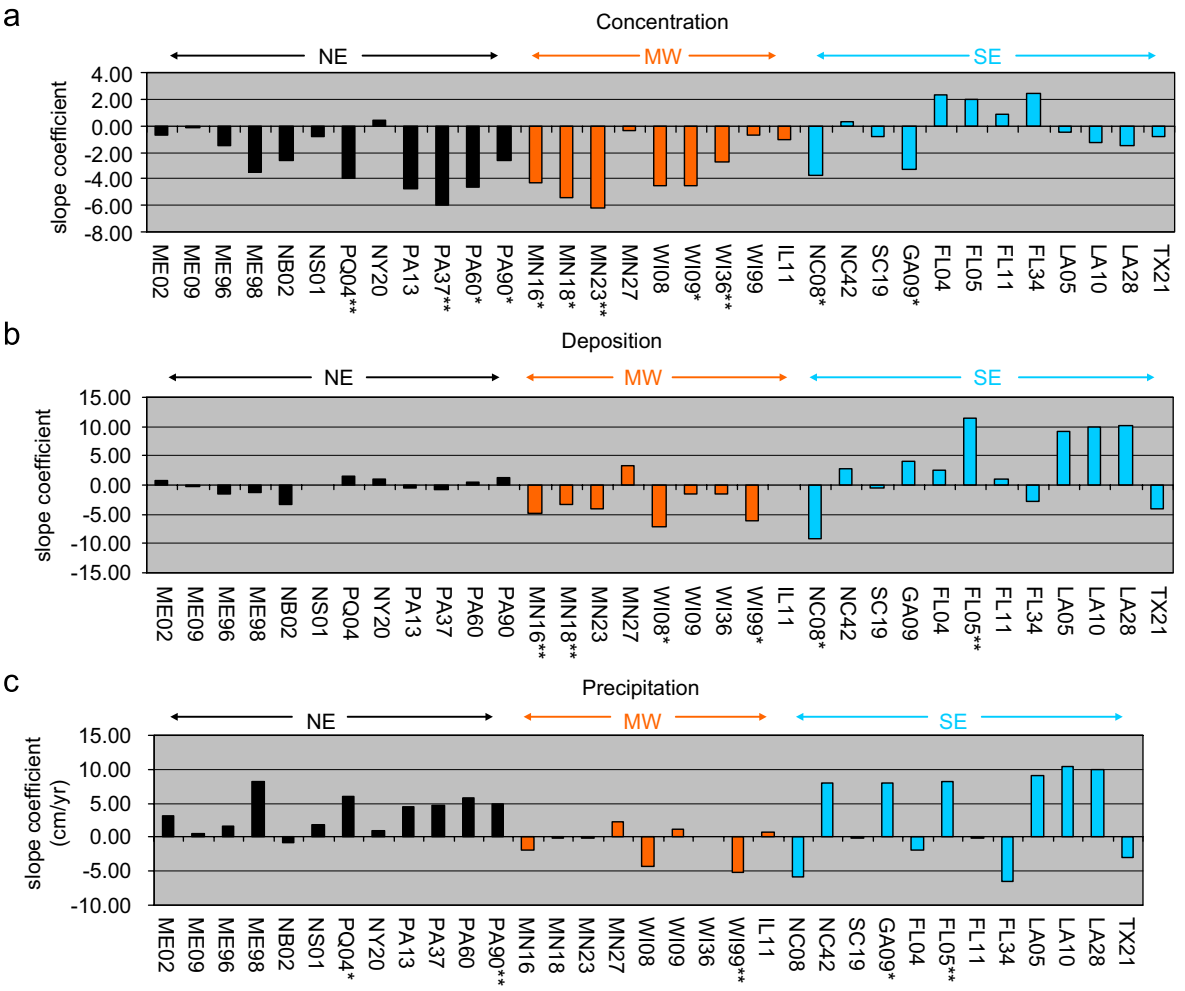

Fig. 5. The linear regression slope coefficients, for (a) concentration, (b) deposition, and (c) precipitation for individual sites in this study for 1998–2005. The slope coefficients for (a) and (b) are expressed in units of % change of the long-term mean per year. Sites labeled *significant slope at $p = 0.10$, **significant slope at $p = 0.05$. NE, MW, and SE are Northeastern, Midwestern, and Southeastern regions, respectively.

Table 1
Random coefficient model results (expressed as percent change per year) with standard errors (S.E.) for annual and May–September precipitation mercury concentration for the Northeastern (NE), Midwestern (MW), and Southeastern (SE) USA regions

	Northeastern			Midwestern			Southeastern		
	Concentration % change/year	S.E.	*p*-Value	Concentration % change/year	S.E.	*p*-Value	Concentration % change/year	S.E.	*p*-Value
Annual	−1.70	0.51	0.0014	−3.52	0.74	<0.0001	0.01	0.71	0.9881
May–September	−1.62	0.62	0.0112	−5.33	0.84	<0.0001	0.52	1.16	0.6664

The May–September data, also shown in Table 1, show enhanced yearly declines in mercury concentration that is highly significant ($p < 0.001$) for the MW region. Concentration declines are −5.33% (0.84% S.E.) per year. The NE region shows a similar decline in the warm months as in the annual record. The mercury concentration decline is −1.62% (0.62% S.E.) per year ($p < 0.01$).

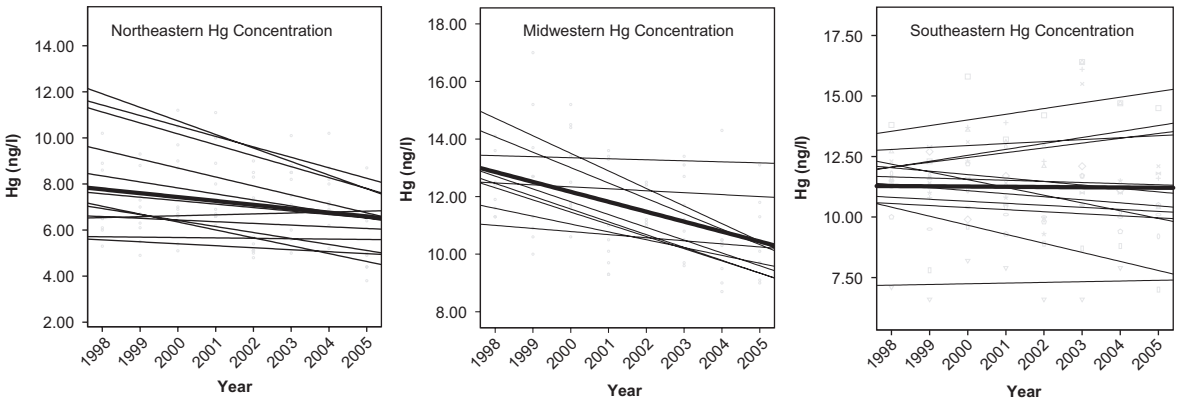

Fig. 6. Random coefficient model results for the relation between time and annual volume-weighted concentration measured at MDN sites for the Northeastern (NE), Midwestern (MW), and Southeastern (SE) USA for 1998–2005. The bold line in each graph represents the overall regression line for the region. The other lines are regressions for each site in the region.

Again, as with the annual data, the SE region does not show any significant trend in concentration (at $p<0.05$) during the May–September period.

4.3. Random coefficient models for all regions together

Tests for differences between regions using random coefficient models were also performed. In other words, are the changes in mercury concentration over time similar for these three regions, or are their slopes different? In this case, region is a "fixed effect" and each region is compared to another region. The models show that for concentration, the regression slopes are significantly different between the SE region and the MW region ($p = 0.008$), and borderline significantly different ($p = 0.063$) between the SE and the NE region. The MW and NE are not significantly different ($p = 0.090$) from each other.

5. Discussion

The comprehensive regional results from the random coefficient models demonstrate that there are significant declines in mercury precipitation concentrations from 1998 to 2005 for both the NE and the MW regions. The SE region does not show any positive or negative significant trend during this period. These annual declines in concentration are 1.70% per year for the NE region and 3.52% per year for the MW from 1998 to 2005. For the entire 8-year period, these represent declines of $14\pm4\%$ and $28\pm6\%$ for the NE and MW, respectively.

Recent declines in precipitation mercury concentration and deposition have been reported in other locations also. Wangberg et al. (2007) found declines of 10–30% in deposition, and greater declines in precipitation concentration of total mercury, at several stations near the North Sea, when comparing 1995–1998 to 1999–2002 data. These declines are attributed to emission controls in Europe. A study of Canadian sites (Temme et al., 2007) which measured total gaseous mercury (TGM) found statistically significant ($p<0.01$) downward trends at five of six eastern sites from 1995 to 2005 with the largest declines of 13% and 17% at rural sites near the urban areas of Montreal and Toronto, respectively. MDN sites in nearby areas also showed decreasing patterns in mercury concentrations, although they were often not statistically significant. The authors hypothesize that the good agreement between declines in both TGM and mercury in precipitation at these eastern Canadian sites is the result of local and regional changes in mercury emissions.

Definitive relations between mercury emissions and concentrations in precipitation cannot be assessed with certainty in our study because the emissions record contains significant uncertainties and extends from the early-to-mid 1990s to 2002, while the concentration record analyzed here is from 1998 to 2005. Despite this discrepancy, the emissions record is worth discussion. We will primarily consider here the regional changes in emissions, understanding that individual sources

within a region will have varying degrees of influence over the deposition at any given MDN site in the region (Butler et al., 2007).

Emission estimates show declines in all mercury species (Table 2) from the early-to-mid 1990s to 2002. Total mercury emissions have declined 47% in the NE, 32% in the MW, and 35% in the SE over this period, while RGM emissions have declined 50%, 43%, and 45%, respectively. Another way to view emission changes is from the standpoint of an emission flux per unit area ($\mu g\,m^{-2}\,year^{-1}$). Total mercury flux decreases from the early-to-mid 1990s to 2002 by 12.4, 7.5, and 6.9 $\mu g\,m^{-2}\,year^{-1}$ for the NE, MW, and SE, respectively. For RGM (Hg (II) in Table 2), the decreases in emissions flux are 5.6, 3.7, and 3.4 $\mu g\,m^{-2}\,year^{-1}$, respectively. A more detailed depiction of the spatial distribution of RGM flux changes is shown in Fig. 3.

During this same period, it is likely that global emissions of mercury may have increased or remained constant (Pacyna et al., 2006). We hypothesize that *if the regional temporal patterns in North American emissions shown here were similar to the patterns from 1998 to 2005* (*the period of analysis for the MDN data*) this would suggest that the effect of global sources on wet mercury concentrations in precipitation may have less of an impact than local/regional declines in mercury emissions for the NE and MW. Thus, we see significant declines in wet mercury concentrations for these regions. Others have also demonstrated that local and regional sources can have major impacts on both wet and/or dry mercury deposition (Mason et al., 2000; Landis et al., 2002; Keeler et al., 2006; Evers et al., 2007). This finding is also consistent with the modeling analysis of Cohen et al. (2004) for the Great Lakes Region, which includes both the MW and NE.

We did not find significant reductions in concentration for the SE region as a whole. It is possible that wet deposition in the SE region is more influenced by global transcontinental mercury sources. Different atmospheric processes may be a factor that differentiates the NE and MW regions from the SE with respect to mercury wet deposition. Guentzel et al. (2001) proposed that high altitude, long-range transport of RGM and particulate Hg are a significant source of mercury deposition in Florida. This is due to large convective storms in summer that scavenge abundant globally derived RGM and particulate mercury from the middle and upper troposphere. It is noted that a portion of this upper atmosphere RGM and particulate mercury will have been transported from sources and a portion will have been formed in the atmosphere via oxidation of elemental mercury. These types of storms also occur in other Southeastern areas where intense summer heating leads to major convective storm activity. Almost 1/2 of the total mercury

Table 2
Estimated changes in regional mercury emissions between the early-to-mid 1990s and 2002

	Hg (total)	Hg^0	Hg (II)	$Hg_{(p)}$
Northeastern				
Emissions in early-to-mid 1990s (metric tons)	57.2	25.2	24.0	8.0
Emissions in 2002 (metric tons)	30.5	15.4	12.0	3.1
Fractional change in emissions between the two periods (%)	−47	−39	−50	−61
Change in emissions between the two periods (metric tons)	−26.6	−9.8	−12.0	−4.9
Change in emissions flux between the two periods ($\mu g\,m^{-2}\,year^{-1}$)	−12.4	−4.6	−5.6	−2.3
Midwestern				
Emissions in early-to-mid 1990s (metric tons)	38.0	19.2	13.8	5.0
Emissions in 2002 (metric tons)	25.9	15.9	8.0	2.1
Fractional change in emissions between the two periods (%)	−32	−17	−43	−58
Change in emissions between the two periods (metric tons)	−12.1	−3.3	−5.9	−2.9
Change in emissions flux between the two periods ($\mu g\,m^{-2}\,year^{-1}$)	−7.5	−2.0	−3.7	−1.8
Southeastern				
Emissions in early-to-mid 1990s (metric tons)	50.9	25.1	19.3	6.5
Emissions in 2002 (metric tons)	33.2	19.9	10.7	2.7
Fractional change in emissions between the two periods (%)	−35	−21	−45	−59
Change in emissions between the two periods (metric tons)	−17.6	−5.2	−8.6	−3.8
Change in emissions flux between the two periods ($\mu g\,m^{-2}\,year^{-1}$)	−6.9	−2.1	−3.4	−1.5

The emissions regions are shown in Fig. 3.

deposition occurs in the summer months (June–August) in the SE (Fig. 4). However, these exceptionally large convective storms are not as likely in the cooler Northeastern and Midwestern USA, where convective activity is not as intense. Therefore, this upper level source of mercury is less available to these areas.

Additionally, many of the MDN sites in the SE are located relatively near the coast. The increased presence of halogens in the marine atmosphere, most likely leads to an increased rate of conversion from Hg^0 to RGM, and thus lead to more wet Hg deposition (Hedgecock and Pirrone, 2001; Laurier et al., 2003). This conversion may occur by different mechanisms. First, the enhanced RGM in the marine boundary layer can lead to enhanced mercury concentrations in precipitation that falls through this layer. Second, convective thunderstorms may transport reactive halogen compounds from the marine boundary layer into the precipitating clouds, and these reactive halogen compounds may significantly increase the rate of conversion of Hg^0 to RGM in cloud droplets. In either case, the incorporation of long-range transported Hg^0 into precipitation would be enhanced. The consequence of the above processes is that increasing or static emissions from global sources may offset the impact of regional emission declines in the SE.

6. Conclusions

Concentration of mercury in precipitation as measured by the MDN has declined significantly in the Northeastern and Midwestern USA regions from the period 1998 to 2005. Random coefficient models show declines of $14\pm4\%$ for the NE and $28\pm6\%$ for the MW during this 8-year period. The SE region shows no significant overall pattern in concentration for the same period.

A pattern of regional mercury emission decline exists from the early-to-mid 1990s to 2002 in all three regions, although estimating the total emission decline is difficult after 2002 because of limited data on mercury emissions. Emission declines have been proportionately greater in the NE than the MW and SE regions in terms of emissions and emissions flux.

The lack of a trend in precipitation mercury concentration in the SE may be the result of different atmospheric processes in this region. Global emissions of mercury are static or possibly increasing, and these global sources may have a greater influence on precipitation chemistry in this region. A greater marine influence that enhances the conversion of Hg^0 to RGM may also be a factor.

Better data regarding current and historical emission levels, in terms of frequency of reporting, speciation, and consistency between inventories, would allow a more thorough comparison between spatio-temporal changes in MDN concentration (and deposition) data and mercury emissions.

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