Mercury at MDN sites, 1998-2005: Declines in the Northeast, No Change in the Southeast

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1 Mercury Emissions

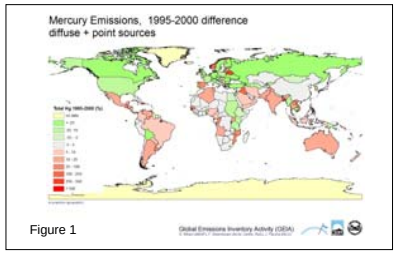


Figure 1

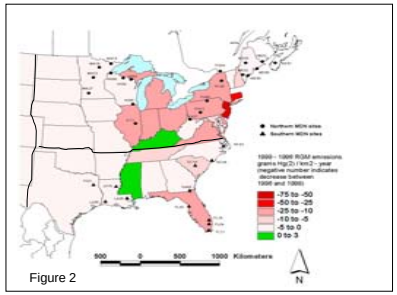


Figure 2

Evidence suggests that anthropogenic mercury emissions declined during the 1990's in Europe and North America. In most other areas it has increased (Figs. 1 and 2) . What has happened since then is unclear. However, in rapidly developing areas such as India and China mercury emissions are estimated to be growing at 2.5% and 5.0 % per year, respectively (EPRI 2004, Zhang 2002). Data from the eastern USA (Fig. 3, based on data from the EPA) show that all forms of mercury are declining in this region. The distribution of emissions in North America ~1999 is shown in Fig. 4, based on data from the EPA and Environment Canada.

There are many uncertainties in the 1996 and 1999 U.S. mercury emissions inventory, and the 2002 emissions inventory at the state level has not been released yet. In addition, details on temporal changes in emissions – even on weekly or seasonal time scales – are generally not available. Wet deposition is a highly episodic phenomena, and source-receptor relationships are comparably episodic. Since variations in emissions are unknown but believed to be significant, it is very difficult to investigate the effect of changing emission levels to changes in MDN concentrations and deposition.

Figure 3

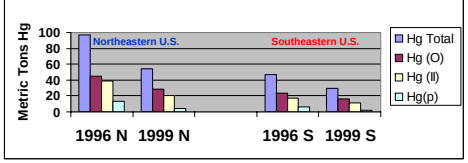
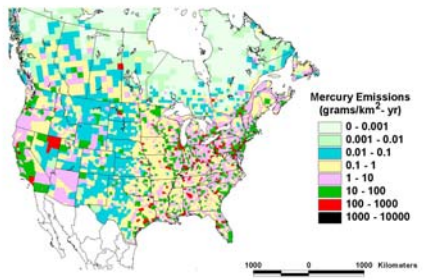


Figure 4



2 Concentration and Deposition Trends at MDN Sites

Individual MDN sites often do not show strongly significant trends in either concentration or deposition over time. However Figures 5 and 6 show the direction of the linear trend for MDN sites, and the relative strength of the relationship (given as the strength of the R^2 term on the y axis). It can be seen that Northeastern sites are more likely to have exhibited more significant decreases than Southeastern sites.

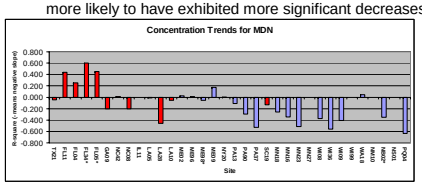


Fig. 5 R^2 values for linear regressions of individual MDN sites, annual concentration vs. year. A minus value indicates a negative trend in concentration over time. Data are from beginning of first full year of site operation (often 1997) through 2004. Red bars (SE sites); Blue bars (NE sites).

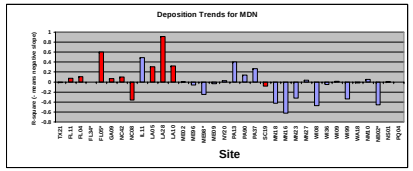


Fig. 6 R^2 values for linear regressions of individual MDN sites, annual deposition vs. year. A minus value indicates a negative trend in deposition over time. Data are from beginning of first full year of site operation (often 1997) through 2004. Red bars (SE sites); Blue bars (NE sites).

Random Coefficient Models

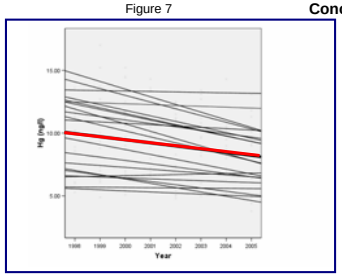
A much more robust analysis uses random coefficient models to test for temporal trends at MDN sites. The graphs presented below show the relation between mercury concentration and time (Figs. 7 and 8) and mercury deposition and time (Figs. 9 and 10) for the period 1998 to 2005 for the Northern sites (Fig. 2). The black individual lines represent individual sites. The red regression line is the overall linear regression of all Northern sites combined. The graphs on the left are annual data. The graphs on the right are for the months May – Sept. when mercury concentrations tend to be higher. Declines in concentration are 2.5% +/- 0.5% to 3.6% +/- 0.6% per year. Declines in deposition are 2.1% +/- 0.4% to 2.9% +/- 0.5% per year.

Why do the Northeastern U.S. sites show declines while the Southeastern U.S. sites do not show declines in MDN concentrations and deposition?

The mercury emissions record is not up-to-date and contains many uncertainties. However, the available data indicates that there have been greater emissions declines in the northeastern U.S. than in the southeastern U.S. Another possibility is that the Northeastern U.S. is more impacted by local/regional emissions while the southeastern USA has a proportionately greater impact from global emissions, which may be on the rise, especially in Asia. Guentzel et al. (2001) propose that high altitude long-range transport of RGM and particulate Hg are a significant source of mercury deposition in Florida. This is due to summertime large convective storms that scavenge globally derived RGM and particulate mercury from the middle and upper troposphere. This scavenging may be a widespread phenomenon in the southeast.

Northern Sites Concentration

Figure 7

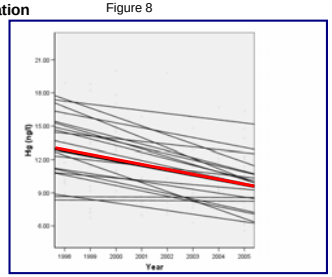


Annual

Concentration is -2.53% per year (s.e.= 0.49%)
P<0.0001 n=20

(southern sites (not shown) Conc 0.01% /yr (s.e.=0.71%) P=0.988 n=12)

Figure 8



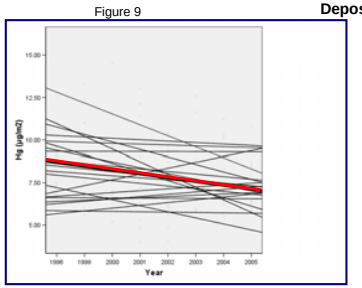
May - Sept

Concentration is -3.56% per year (s.e.= 0.58%)
P<0.0001 n=19

(southern sites (not shown) Conc 0.52% /yr (s.e.=1.16%) P=0.666 n=11)

Deposition

Figure 9

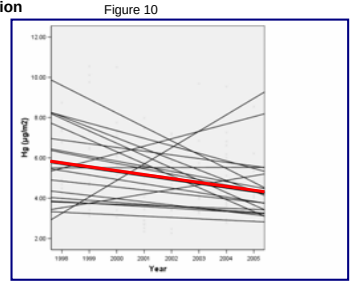


Annual

Deposition is -2.14% per year (s.e.= 0.43%)
P<0.0001 n=20

(Southern sites (not shown) Dep is 0.50% /yr (s.e.=0.91%) P=0.595 n=12)

Figure 10



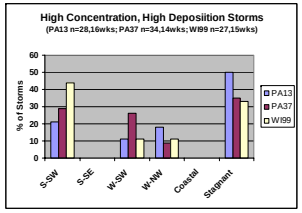
May - Sept

Deposition is -2.91% per year (s.e.= 0.53%)
P<0.0001 n=19

(Southern sites (not shown) Dep is 1.00% /yr (s.e.=2.11%) P=0.645 n=12)

3 High vs. Low Concentration and Deposition Storms at 3 Sites

In an attempt to assess if particular source regions are responsible for high levels of mercury deposition we examined back trajectories for individual storm during weeks (excluding colder months) when particular MDN sites (PA13, PA37, and WI99) showed either very high concentrations and depositions (45 weeks) of mercury, or very low concentrations and depositions (40 weeks) of mercury.



For high-concentration storms, the most common trait at all three sites were back trajectories that showed slow moving, relatively "stagnant" air mass flows (Fig. 11). Figs 12 and 13 are representative of such systems. Such storms would be expected to be high in other atmospheric pollutants ($SO_4^{=}$, NO_3^- and H^+) as well as mercury. High deposition storms also commonly arrive from the South-Southwest direction, especially for WI99 (Fig 11), where over 40% of the high-deposition storms examined were classified in this category. At WI99, southerly storm tracks can be influenced by the mercury emission sources in the Chicago/ Gary area, a region with high emissions (see Figure 4) that also contributes significantly to mercury deposition to Lake Michigan (Landis et al. 2002).

Figure 11 Percent of high-deposition storm back-trajectories by category

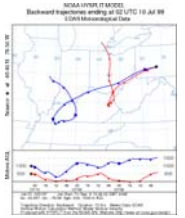


Fig. 12 PA13, 72-hour back trajectory for 1000m and 500m above ground level for a high-deposition precipitation event during the week of 7/6/99 to 7/13/99. Precipitation was 25 mm, mercury concentration was 21.9 ng/l and wet deposition was 558 ng/m2. The 1000m back trajectory indicates an air mass passing directly over the industrialized Ohio River Valley.

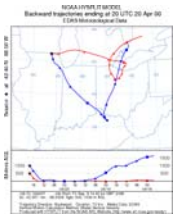


Fig. 13 WI99, 72-hour back trajectory for high-deposition precipitation event during the week of 4/18/00 to 4/25/00. Precipitation was 84 mm, mercury concentration was 17.3 ng/l and wet deposition was 1453 ng/m2. 1000-m back trajectory--- shows air mass near ground passing over Chicago/Gary metropolitan area.

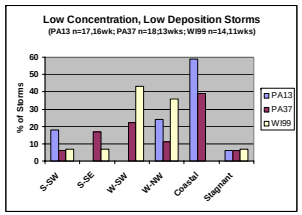


Figure 14 Percent of low-deposition storm back-trajectories by category

Significant anthropogenic sources of mercury occur along the eastern seaboard (Fig. 4). However, for sites within 400 – 500 km of the Atlantic coast, storms derived from oceanic air and moisture provide relatively low wet deposition rates (i.e. PA13 and PA37), even when the volume of precipitation is high. A similar result was found for other pollutant species, NO3- and SO4= concentration and deposition, at the Hubbard Brook Experimental Forest in New Hampshire (Likens et al. 1990). PA13 low-deposition storms can also occur when air mass trajectories are originating from a northerly direction. For the storms examined, 24% were classified in this category (W-NW). Areas north of PA13 are generally rural in nature and represent an area of relatively low mercury emissions. PA37 has only 11% of its low-deposition storm tracts originating from the north. In contrast to PA13, the area north of PA37 has significant mercury emission sources (e.g. Pittsburg area). For the WI99 site, air masses from the W-SW and W-NW are the dominant sources of air masses resulting in low-deposition storms. These directions represent non-industrial areas with low mercury emissions in the region (see Fig. 4). Higher mercury emission source regions are located to the east and south of the site.

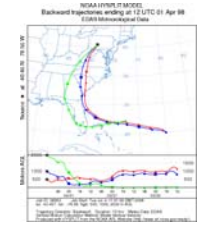


Fig. 15 PA13, 72-hour back trajectories (500m, 1000m and 2000m agl) for low deposition precipitation event during the week of 3/31/98 to 4/7/98. Precipitation was 30 mm, mercury concentration was 3.6 ng/l and wet deposition was 109 ng/m2.

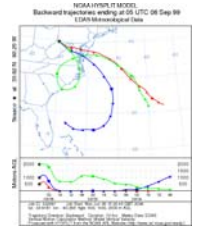


Fig. 16 PA37, 72-hour back trajectories (500m, 1000m and 2000m agl) for low deposition precipitation event during the week of 8/31/99 to 9/7/99. Precipitation was 20 mm, mercury concentration was 7.9 ng/l and wet deposition was 160 ng/m2.

References

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