JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 111, D14316, doi:10.1029/2005JD006814, 2006

Recent trends in anthropogenic mercury emission in the northeast United States

J. M. Sigler[1] and X. Lee[1]

Received 6 May 2005; revised 30 March 2006; accepted 14 April 2006; published 28 July 2006.

[1] We combine wintertime measurements of total gaseous mercury (TGM) with a combustion tracer (carbon dioxide) at a background site in Connecticut to investigate changes in regional Hg emission from 1999/2000 through 2003/2004. This method allows timelier flux estimates than inventory approaches. We find a 20% decrease in emission between 1999/2000 and 2003/2004, although the rate has increased since 2001/2002. Air trajectory analysis reveals a source region of Hg influencing the site that encompasses most of the mid-Atlantic and New England states, and Maryland and Delaware, with small interannual variation. The significant interannual changes in Hg emission therefore cannot be fully explained by climatological changes. Inventory calculations of Hg flux from the regional electric power sector show significant correlation with the emission trend derived from the atmospheric measurements, suggesting that the power sector strongly influences the regional Hg flux to the atmosphere. However, averaged over the five winters, the Hg flux from the power sector was 47–75% of the atmospheric flux, which is assumed to be >95% elemental Hg. If 50% or more of Hg emitted by power sources is speciated and deposits locally, the power sector can account for only 23–40% of the observed elemental Hg flux to the atmosphere. Possible explanations include conversion of some reactive Hg emitted by power plants to elemental Hg, underestimation of inventory emissions from sources outside the power sector, the possibility that emissions from municipal and medical waste combustion have not been curbed to the EPA targeted level, or that a combination of these factors exists.

Citation: Sigler, J. M., and X. Lee (2006), Recent trends in anthropogenic mercury emission in the northeast United States, *J. Geophys. Res.*, *111*, D14316, doi:10.1029/2005JD006814.

1. Introduction

[2] Mercury (Hg) is a potentially dangerous environmental contaminant. The degree to which anthropogenic Hg emissions impact human health is dependent on the emission rate, as well as the chemical form, of the emitted species. The northeastern United States is characterized by the highest regional anthropogenic Hg flux density in the country, due primarily to combustion of coal and municipal waste in urban areas [*U.S. Environmental Protection Agency* (*U.S. EPA*), 1997]. The emissions reported by the EPA were based on an inventory approach, which combines facility-specific emission factors, activity levels, fuel characteristics and control technology. The inventory approach is a valuable means of providing source-specific, spatially resolved emissions. However, lack of emission test data for many types of sources renders the accuracy of the EPA's emission inventory to no better than 30%. Moreover, timely assessment of temporal trends in Hg emission using the inventory approach is difficult given the sheer number of point sources, the complexity of source categories, ever-changing trends in mercury consumption, advancements in control technologies and state and federal legislation advanced since the EPA study. The need for an alternate approach is underscored by additional, recent studies that show that Hg sources in urban areas are diverse and can change rapidly across timescales. Therefore long-term measurements, preferably at multiple locations, are important [*Kim and Kim*, 2001; *Nadim et al.*, 2001; *Landis et al.*, 2002; *Chen et al.*, 2004].

[3] An alternative technique for deriving the regional Hg emission rate is the correlation of ambient Hg with an atmospheric tracer. This technique provides an estimate of emission on a fine time resolution (<1 year) and can be based on a chemical species characterized by inventory data that are more reliable and timely than Hg inventories. Furthermore, the derived emissions ratio is understood to have a footprint representative of a large area (on the order of 500 km) when established with data over timescales of weeks or longer [*Potosnak et al.*, 1999].

[4] Tracer analysis has been used extensively in atmospheric chemistry and transport studies. For example, *Wofsy et al.* [1994] used CO and hydrocarbons as tracers to estimate the contribution of chemicals released from biomass burning to the subarctic. *Wang et al.* [2002] used emission ratios of CO and SO_2 with NO_y and other industrial tracers to infer combustion fuel type and sources

---

[1] School of Forestry and Environmental Studies, Yale University, New Haven, Connecticut, USA.

0148-0227/06/2005JD006814$09.00

of elevated CO in rural eastern China. CO_2/CO correlation analyses have been used to infer combustion source types and regions [*Suntharalingam et al.*, 2004]. CO and CO_2 have also been used as tracers for determining Hg emission from biomass burning events [*Brunke et al.*, 2001; *Friedli et al.*, 2003; *Sigler et al.*, 2003].

[5] Wintertime CO_2 has been demonstrated to be a viable tracer for combustion sources in the northeast United States [*Bakwin et al.*, 1994; *Potosnak et al.*, 1999]. These sources compose more than 90% of regional Hg emissions and tend to be located near noncombustion (e.g., area and manufacturing) sources of Hg [*U.S. EPA*, 1997]. The fact that the two emission sources are colocated geographically makes CO_2 an excellent tracer for Hg. For example, *Lee et al.* [2001] used the Hg and CO_2 atmospheric concentration data for winter 1999/2000 collected at the same site to determine a regional anthropogenic elemental Hg (Hg°) flux of 41(±2) g km^{-2} yr^{-1} for the northeast United States.

[6] In this study, we use measurements of ambient TGM (total gaseous mercury) and wintertime CO_2 at a background site in northwest Connecticut to determine interannual variability in TGM emission rate in the northeast United States during the winters of 1999/2000 to 2003/2004. This study appears to be the first that applies the tracer technique to a multiyear time period. Utilizing wintertime air mass trajectory data, we create potential source contribution functions (PSCF) for each year in order to identify potential geographical source regions and dominant flow pathways that contribute the most to ambient TGM concentrations at our measurement site. We also analyze changes in the consumption of fuels by electric power producers in order to explain changes in regional Hg emissions.

2. Data and Methodology

2.1. Ambient Measurements and Tracer Analyses

[7] TGM was monitored at Great Mountain Forest in northwest Connecticut (41°58′N, 73°14′W) from the summer of 1999 through the spring of 2004. The measurement was made at 5-min intervals using a mercury vapor analyzer (Model 2537A, Tekran, Inc.) housed in a temperature-controlled shelter. The analyzer is calibrated daily against an internal permeation source to an accuracy of 5%. TGM data were carefully screened so that low-mercury episodes (TGM < 1.0 ng m^{-3}) and periods during which the analyzer malfunctioned were excluded. The instrument cannot distinguish elemental and reactive gaseous Hg (RGM), but over 95% of TGM is thought to be in elemental form [*Lindberg and Stratton*, 1998]. We assume this condition is met at Great Mountain since TGM measurements at similar, rural, background sites in southern Quebec [*Poissant et al.*, 2005] and New York State [*Han et al.*, 2004] have been found to be >97% Hg^0, and this condition is also met even in urban settings, though RGM may at times account for as much as ∼11% of TGM (e.g., Detroit [*Lynam and Keeler*, 2005]).

[8] Although measurements of ambient TGM concentrations may suggest influences from local or regional pollution sources, such measurements by themselves do not indicate actual emission rates. Rather, they must be correlated with measurements of a tracer species that is emitted by the same sources or sources that are colocated with Hg emissions. We use CO_2 as our tracer species for TGM emissions. CO_2 was measured at Great Mountain concurrently with TGM using a Licor infrared gas analyzer, calibrated weekly to an accuracy of 1%.

[9] We limit our tracer analyses to daytime conditions (1000–1600 LST) when the atmospheric boundary layer is well mixed. Where CO_2 data are concerned, we consider only the colder months as defined by the 5-cm soil temperature less than 3°C, so that biological CO_2 signals were negligible. This condition is typically met at Great Mountain from late December to the end of April. For convenience, the atmospheric measurements are compared with inventory data reported for the four winter calendar months (January to April).

[10] Tracer analysis assumes that the slope of a regression line between TGM and CO_2 concentrations is equivalent to the regional emission ratio of the two species [*Bakwin et al.*, 1994; *Stehr et al.*, 2000; *Lee et al.*, 2001]. Rather than ordinary least squares regression, we use orthogonal or geometric mean regression as outlined by *Draper* [1992] and *Richardson* [2000]. Combining the emission ratios with an inventory carbon flux yields the annual regional TGM flux. We use methods and data described by *Andres et al.* [1996] to derive an inventory carbon (C) emission rate of 529 t of C per km^2 $year^{-1}$ over the potential source region for the period 1994–1995. This includes most New England and mid-Atlantic states (Connecticut, Massachusetts, Rhode Island, New York, New Jersey, and Pennsylvania) as well as Maryland and Delaware. The total carbon emissions from fossil fuel burning in the northeastern states changed on average by no more than 2% during the period of 1990–2001 [*Blasing et al.*, 2004]. The state-by-state CO_2 inventory activity was terminated after 2001. However, total CO_2 emissions within the source region have been known to consistently compose the same percentage (approximately 15%) of total U.S. emissions each year [*Blasing et al.*, 2004], and U.S. emissions changed by less than 2.5% between 1999 and 2003 [*U.S. Energy Information Administration* (*U.S. EIA*), 2004c]. For these reasons, the same total CO_2 emission rate (529 t of C per km^2 $year^{-1}$) is used for the entire study period. Wintertime (January to April) monthly C flux is given as 1/12 of the annual flux, which represents the typical fraction of annual CO_2 emissions from fossil fuel burning in the United States [*Blasing et al.*, 2005].

2.2. Air Trajectory Analysis

[11] We combine the TGM and CO_2 measurements with air trajectory analysis to determine the source region impacting the measurement site and the potential influence of interannual variation in climatology and air transport patterns on the tracer correlation result. Trajectory analysis is commonly used to identify air mass source regions, transitions and atmospheric transport history [e.g., *Moody et al.*, 1998; *Cooper et al.*, 2001]. We use NOAA HYSPLIT-4, an online air trajectory model [*Draxler and Hess*, 1997], to build a trajectory database for each winter of TGM observation. Three 48-hour backward trajectories were simulated per day, at 1500, 1800 and 2100 GMT (1000, 1300 and 1600 LST) for the period considered in the tracer analyses. There were some significant gaps

in TGM data during December of 2001 and March of 2002. Trajectories were not simulated during these gaps.

[12] Choice of trajectory length is somewhat subjective. For pollutant studies in remote environments with no local sources, longer time periods (10 days) are necessary [e.g., *Cheng and Schroeder*, 2000; *Lin et al.*, 2001]. For tracking of regional pollution events or determination of source regions of, for example, TGM [*Weiss-Penzias et al.*, 2003; *Sigler et al.*, 2003], RGM [*Han et al.*, 2005; *Lynam and Keeler*, 2005], NO_3^- [*Fischer and Talbot*, 2005], SO_x [*Butler et al.*, 2001] and O_3 [*Moody et al.*, 1998; *Mao and Talbot*, 2004], 1–5 day back trajectories are more suitable. We use back trajectory length of 48 hours for three primary reasons. First, backward trajectories simulated for each Hg pollution event observed at Great Mountain during the 5-year study period most often traversed geographic areas expected to be highly sparse in pollution sources beyond a 48-hour time length (e.g., the Great Plains, northern and western Canada, the Arctic, northern Pacific Ocean). Second, because covariance in time series of pollutants with atmospheric lifetimes on the order of months or more, in this case, Hg and CO_2, is caused by sources within 500 km, or 2 days transport at average wind speeds [*Bakwin et al.*, 1997]. Third, research by *Moody et al.* [1998] indicated that 2-day back trajectories from Harvard Forest, Massachusetts (120 km northeast of Great Mountain) were sufficient to identify regional-scale transport patterns influencing chemical composition at that site. This may in part explain the results of *Han et al.* [2005], who found better correlation between RGM emissions inventory and source area impacting sites in New York State when 2-day, rather than 5-day trajectories were used.

[13] HYSPLIT and other trajectory models are typically initialized at starting heights which represent well-mixed, boundary layer air representative of surface measurements. Starting heights ranging from 100 m [e.g., *Han et al.*, 2005] to 2000 m [e.g., *Butler et al.*, 2001] have been used in previous studies. We choose 500 m [e.g., *Weiss-Penzias et al.*, 2003; *Fischer and Talbot*, 2005], as this is within wintertime boundary layer heights observed in the region (600–700 m [*Moody et al.*, 1998]) and sensitivity analysis (not shown) revealed little difference in source area when starting heights were varied below that level. HYSPLIT uses gridded wind fields from the Eta Data Assimilation System (EDAS-40 km). The output of the HYSPLIT model gives the location in latitude and longitude of each hourly endpoint (48 endpoints per trajectory data file).

2.3. Potential Source Contribution Function

[14] Backward trajectories calculated during Hg pollution events at Great Mountain may indicate source regions of polluted air masses impacting the site. To identify these source regions, trajectory data can be incorporated into a receptor model that creates a probability field for emission source potential.

[15] Using the trajectory data, we create a single-layer potential source contribution function (PSCF) for each winter. The concept of PSCF has previously been used to determine, for example, sources and transport pathways of aerosols and TGM in the Canadian high Arctic [*Cheng et al.*, 1993; *Cheng and Schroeder*, 2000; *Lin et al.*, 2001], sources of TGM in the St. Lawrence River valley [*Poissant*, 1999] and sources of RGM in New York State [*Han et al.*, 2005]. Detailed descriptions of the model can be found in these and other studies. In summary, we divide the source region into an array of 0.5° latitude by 0.5° longitude grid cells. If a trajectory endpoint lies in a particular grid cell, the trajectory is assumed to collect material emitted from that grid cell and transported to the receptor site. Mathematically, the probability that an endpoint lies in a particular cell is $P[A_{ij}] = n_{ij}/N$, where n_{ij} is the number of endpoints falling in the ijth grid cell and N is the total number of endpoints. A certain number m_{ij} of these endpoints corresponds to the observation of TGM concentrations above a criterion value at the receptor site, which is defined as the 85th percentile concentration of all TGM measurements for that winter. The probability of this event is $P[B_{ij}] = m_{ij}/N$. The PSCF value for each grid cell is therefore $PSCF_{ij} = P[B_{ij}|A_{ij}]$, or m_{ij}/n_{ij}. This value, between 0 and 1, describes probable geographic source regions based on the trajectories arriving at the source site.

[16] PSCF values may be biased in cells where n_{ij} is small. Therefore a weighting function (W) is introduced to minimize such biases, as

$$W(n_{ij}) = \begin{bmatrix} 1.0 & \dfrac{n_{ij}}{n_{mean}} & \geq 1.0 \\ 0 & \dfrac{n_{ij}}{n_{mean}} & \leq 1.0 \end{bmatrix} \tag{1}$$

where n_{mean} is the mean of the n-value for those cells with n_{ij} >= 10. Choice of a weighting function is somewhat subjective, but we find that equation (1) gives precedence to cells with higher count totals as those cells likely influence the TGM signal at the site most significantly. Other studies use the function $W(n_{ij}) = 0.1\ n_{ij}$ for $n_{ij} \leq 10$ [*Lin et al.*, 2001] or employ weight functions for $n_{ij} \leq 4$ [*Poissant*, 1999], with far fewer n-values than in our study. Application of these functions to our trajectory data results in physically unrealistic source region maps. For example, rural areas in the midwestern and western United States and Canada would be designated as highly probable source regions when only 2–3 trajectories, all of which happen to be polluted, impact them during the entire study period.

2.4. Fuel Consumption Data for the Power Sector

[17] It is known that combustion sources accounted for more than 85% of all Hg emissions in the United States in the mid-1990s. In the United States, coal-fired power plants are the most important emissions category [*U.S. EPA*, 1997]. As recently as 1997, municipal waste combustors (MWC), common along the East Coast of the United States, were the largest single Hg source category in the northeastern states. However, EPA new source performance standards of 1997 sought to reduce emissions from MWC (as well as medical waste incinerators) by >90% by 2000. There is evidence that these and other state-imposed regulations have been successful and that emissions from MWC have declined significantly in the northeast. For instance, Hg emissions from this source category were reduced by two thirds in New Jersey between 1997 and 1999 [*Themelis and Gregory*, 2002] and by 87% from medical and municipal waste combustion combined in Massachusetts from the late 1990s to 2004 [*Massachusetts Department of*

Table 1. Total Wintertime (January–April) Consumption of Coal, Petroleum and Natural Gas by the Electric Power Sector Within the Source Region

	2000	2001	2002	2003	2004
Coal, short tons	3.04×10^7	2.5×10^7	2.66×10^7	2.88×10^7	3.0×10^7
Petroleum, barrels	2.71×10^7	2.89×10^7	1.33×10^7	2.97×10^7	3.02×10^7
Natural gas, Mcf	3.6×10^8	2.06×10^8	2.28×10^8	1.8×10^8	1.98×10^8

Environmental Protection, 2006]. Total U.S. emissions from this category could be as low as approximately 1 ton per year, less than 3% of 1995 emissions (N. J. Themelis, personal communication, 2005).

[18] Given the decline in emissions from municipal and medical waste combustion after 1997, the electric power sector is now thought to be the most important Hg source category in the northeastern states. It includes electric utilities, independent power producers (nonutilities) and combined heat and power plants that primarily burn coal, natural gas, or petroleum (usually residual or distillate fuel oil). Despite uncertainty in knowledge of Hg emission factor and variation among coal types and geographic origin, coal is known to have the highest Hg emission factor of these three main fuel categories [*U.S. EPA*, 1997]. Natural gas is relatively clean, with an emission factor that is several orders of magnitude lower than petroleum or coal [*Electric Power Research Institute* (*EPRI*), 1996].

[19] Given the disparity in Hg content and emission factor among coal, natural gas and petroleum, it is possible that the regional emission rate of Hg from electricity generation may be influenced by the relative percentages of these three fuels that are combusted seasonally or annually in the northeast. Therefore we examined changes in total wintertime consumption of coal, petroleum and natural gas for electricity generation among the northeastern states.

[20] We compile data on total consumption of coal, petroleum and natural gas for electricity generation from the U.S. Department of Energy (DOE) Energy Information Administration (EIA)'s Monthly Energy Review [*U.S. EIA*, 2005]. For each winter, we combine the total consumption of each fuel type for each state in the source region (Connecticut, Massachusetts, Rhode Island, New York, Pennsylvania, New Jersey, Maryland, and Delaware; see section 3.3) for the four winter months (Table 1). We also calculate a Hg:C emission ratio by applying approximate heat contents, CO_2 and Hg emission factors to each fuel consumption total, as shown in equation (2):

$$\frac{Hg}{CO_2} = \frac{(c \times H \times F_m) + (p \times H \times F_m) + (g \times H \times F_m)}{(c \times H \times F_C) + (p \times H \times F_C) + (g \times H \times F_C)} \qquad (2)$$

where c is total coal (short tons), p is total petroleum (barrels), g is total natural gas [(thousand cubic feet (Mcf)], H is the heat content of the fuel (expressed as British thermal unit (Btu) per ton coal, barrel petroleum or Mcf natural gas) and F_m and F_C are the Hg (lb Hg/Btu) and CO_2 (lb CO_2/Btu) emission factors, respectively, for the particular fuel. We calculate Hg emission from coal using emission factors based on the 1999 EPA Information Collection Request (as reported by *EPRI* [2000]), which obtained emission test data on more than 80 coal-fired power plants. Since Hg emission factor from coal is uncertain and power plants do not employ uniform types of pollution control technology which incidentally removes Hg, we use both uncontrolled and postcontrol emission factors. Table 2 shows heat contents and emission factors used in our calculation, and information on data sources.

[21] We note that the consumption data included in our study represent the entire energy sector. In the United States, electric utilities are responsible for approximately 60% of total electric generation, down from more than 90% in the mid-1990s [*U.S. EIA*, 2004a]. For the northeastern and mid-Atlantic states, electric utilities accounted for only 15% of net generation and total coal combustion during 2004 [*U.S. EIA*, 2005]. Therefore we do not distinguish between utilities and independent power producers.

3. Results and Discussion

3.1. TGM Concentrations

[22] Figure 1 presents seasonal statistics of the TGM data. It indicates that Great Mountain is a background site with little interannual variation in TGM concentration. The mean TGM concentration during 1999–2004 was 1.6 ng m^{-3}, which is comparable to other Northern Hemisphere, mid-latitude background sites [*Slemr et al.*, 1985; *Xiao et al.*, 1991; *Kim et al.*, 1995], as well as other forest sites in Connecticut [*Sigler and Lee*, 2006]. Median TGM concentrations varied by only 0.2 ng m^{-3}, between 1.4 and 1.6 ng m^{-3}, and wintertime concentrations tended to be higher than the previous summer. However, it is improper to suggest from the seasonal mean concentration data that the regional TGM emission changed little over the observational period, as TGM data alone cannot discrim-

Table 2. Heat Content and Hg, CO_2 Emission Factors (EF) for Coal, Petroleum and Natural Gas Combustion[a]

	Coal	Petroleum	Natural Gas
Heat content	2.04×10^7 Btu/ton	6×10^6 Btu/barrel	1.02×10^6 Btu/Mcf
CO_2 emission factor, lb CO_2/Btu	2.07×10^{-4}	1.73×10^{-4}	1.17×10^{-4}
Hg emission factor, lb/10^{12} Btu	10, 5.5[b]	6.8	0.0008

[a] The CO_2 EF for coal represents a mean value of subbituminous and bituminous coal and that of petroleum is the emission factor for residual fuel oil, which represents >80% of petroleum consumption for electric power in the U.S [*U.S. EIA*, 2004a, 2004b]. The Hg EFs for coal are taken from uncontrolled power plant emissions and from power plants with minimal control devices (EPA 1999 ICR, as reported by *EPRI* [2000]). The Hg EF for petroleum is based on average values for residual oil combustion [*U.S. EPA*, 1997] and the natural gas EF is taken from *EPRI* [1996].

[b] Uncontrolled and controlled, respectively.

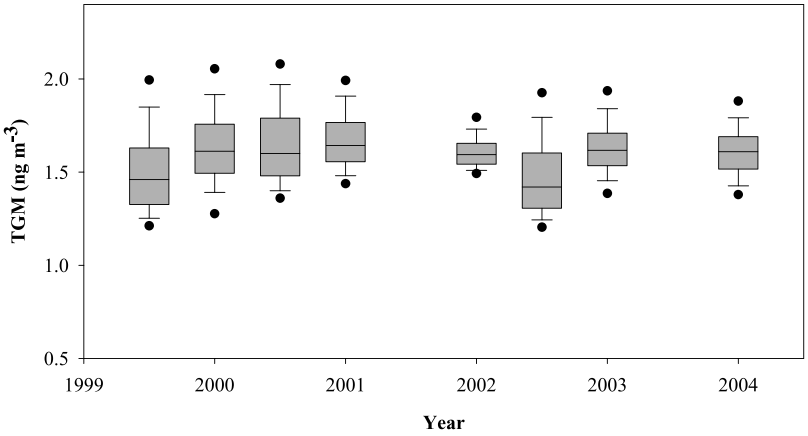

Figure 1. Box plots of interannual TGM concentration at Great Mountain Forest, Connecticut, 1999–2004. Median, 10th, 25th, 75th and 90th percentile values are shown. Midyear data points represent summertime data, and all others are wintertime.

inate contributions of regional sources from the global background.

[23] A detailed analysis of the TGM time series reveals that the site is frequently impacted by pollution events. These events, during which TGM and CO_2 exhibit significant and well-correlated temporal enhancements, typically for 1–4 hours, are common, occurring approximately once per week during the winter. Comparison of measurements taken at Great Mountain with CO_2 measurements from Harvard Forest near Petersham, Massachusetts (42°32′N, 72°11′W, 120 km to the northeast of Great Mountain) shows a high correlation between CO_2 measurements (linear correlation coefficient (r) = 0.73) at both locations (Figure 2a). Similarly, a high correlation (r = 0.81) was also found for TGM during the summer of 2002 when both sites were equipped with TGM monitors (Figure 2b). Given the distance between the sites, the elevated TGM concentrations at Great Mountain are caused by regional, rather than local, pollution sources. Therefore Great Mountain is a site of regional character and suitable for tracer analysis.

3.2. Tracer Analysis of TGM:C Emissions Ratio

[24] Wintertime correlation between CO_2 and TGM at Great Mountain showed r-values ranging from 0.62 to 0.77, with the number of observations ranging between 800 and 1650 for each winter. The strength of these correlations supports our use of CO_2 as a tracer for Hg as they are generally comparable to other tracer studies [e.g., *Bakwin et al.*, 1997; *Potosnak et al.*, 1999]. Figure 3 (open circles) shows the derived TGM:C emissions ratio for each winter. The largest incremental change was observed between the winters of 1999/2000 and 2000/2001, when the emissions ratio declined by more than 40%. However, after a further decline in the winter of 2001/2002, a gradual increase was observed since that year. The tracer analysis suggests that, unlike the trend in mean TGM concentration (Figure 1), the regional TGM emission rate in the northeast may vary significantly from year to year.

3.3. Potential Source Regions for TGM

[25] There are two lines of argument for the observed interannual variation in regional TGM emission as revealed by the atmospheric measurements. First, interannual changes in the source region impacting Great Mountain may lead to changes in the measured correlation between Hg and CO_2 irrespective of any actual interannual variation in emissions. For this to occur, the emission source types and hence the resulting Hg:C emissions ratio must vary geographically. The second argument is that there are changes in the major anthropogenic sources of Hg (e.g., changes in capacity, activity levels, emission category, fuel type, or control technology) which would lead to actual interannual variation in emission.

[26] We test the first line of argument by building PSCF maps for each year. The potential source map for 1999/2000, which exhibited the highest TGM:C emissions ratio during the 5-year study period, is shown in Figure 4. For comparison, Figure 5 shows the potential source region for 2001/2002, when the lowest emissions ratio was observed.

[27] The highest-value cells for 1999/2000, which could be viewed as more likely source regions (Figure 4) lie in southeastern Pennsylvania, centered approximately over Philadelphia, with slightly lower though still high values reaching into lower Pennsylvania and central/lower New Jersey. This is an area determined by *U.S. EPA* [1997] to have a high anthropogenic Hg emission rate. There are also potential source regions in northern New York State, Delaware and eastern Maryland, as well as western Pennsylvania (Pittsburgh). Coal-burning power plants are the most important point sources of Hg in all of these areas (see section 2.4).

[28] The potential source region for 2001/2002 (Figure 5) is similar to that derived for 1999/2000, although somewhat less extensive and with generally lower probabilities throughout the source region. As in 1999/2000, most of the source region for 2001/2002 lies in the states of New York, New Jersey and Pennsylvania. There is some similarity in the location of cells with the largest probabilities in both Figures 4 and 5, as high-value cells are found in lower New York, New Jersey and eastern Pennsylvania. PSCF maps created for the winters of 2000/2001, 2002/2003 and 2003/2004 (not shown) were not much different from those shown in Figures 4 and 5. In any given year, the most likely source regions tend to lie in Connecticut, western Massa-

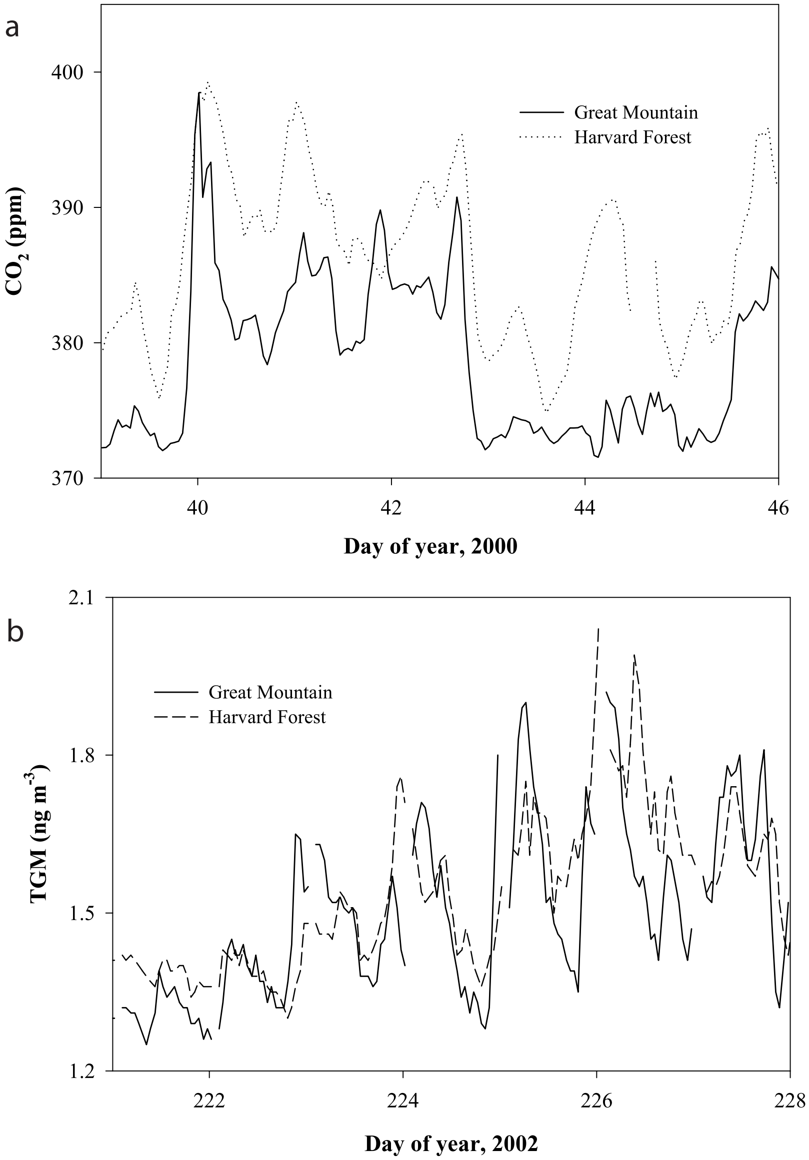

Figure 2. (a) Temporal variation in ambient CO_2 measured at Harvard Forest (Massachusetts) and Great Mountain Forest (Connecticut) during winter 2000 and (b) TGM at Great Mountain plotted concurrently with TGM at Harvard Forest, summer 2002. The small difference in CO_2 concentrations at both sites is due to different calibration standards.

chusetts, lower New York, New Jersey, and eastern and northern Pennsylvania. To further illustrate this point, all trajectory and TGM data (1999/2000–2003/2004) were combined to create a composite source region during the entire study period (Figure 6). According to Figure 6, the source region encompasses most New England and mid-Atlantic states (Connecticut, Massachuestts, Rhode Island, New York, New Jersey, and Pennsylvania), as well as Maryland and Delaware, with the most likely source regions being lower New York, New Jersey, eastern Pennsylvania, and the upper Chesapeake Bay area (Maryland, Delaware).

[29] The PSCF analysis suggests that interannual, climatological changes in geographic source region are, at the most, mitigating factors affecting the correlation analysis. Visual inspection of Figures 4 and 5 indicate some differences in probability and size of the total source region and therefore perhaps some degree of climatological influence on the estimate of TGM emission rate. However, there is significant overlap in both total source region (low and high-probability cells combined) and likeliest source regions (high-probability cells) among all winters. Furthermore, regional emissions ratio derived from fuel consumption inventory data does not reveal much sensitivity to change in the geographic size of the source region (Figure 3). Thus we conclude that changes in source region alone cannot explain the observed interannual variation in the emissions ratio, especially the difference of nearly 50% in the Hg:C emissions ratio between 1999/2000 and 2001/2002.

3.4. Hg:C Emissions Ratio Based on Fuel Consumption

[30] The fact that the interannual variation in the source region cannot explain the observed change in the emissions ratio leads us to test the line of argument that the observed

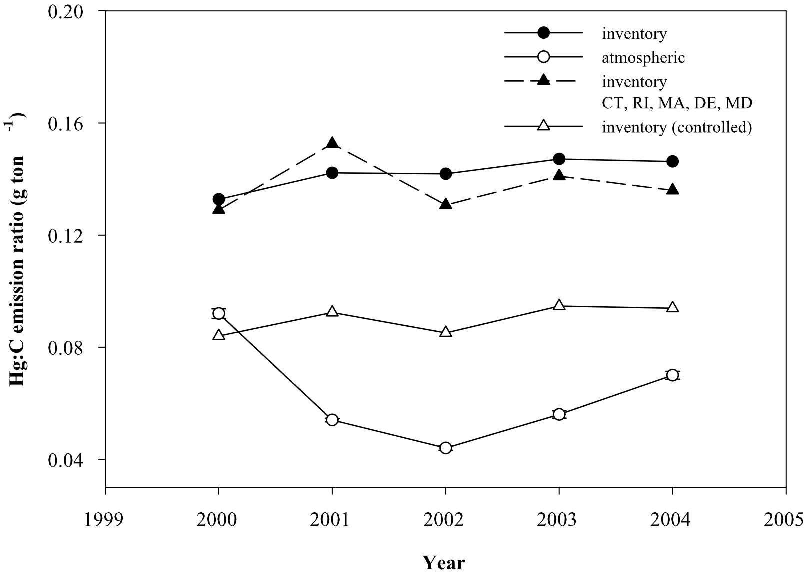

Figure 3. Comparison of wintertime Hg:C emission ratios derived from atmospheric measurement and those derived from inventory fuel consumption data for electric power sector. Error bars on atmospheric plot are 95% confidence intervals.

changes in the Hg:C emissions ratio, and therefore the regional TGM emission rate, is related to interannual changes in the anthropogenic sources themselves. For this we choose to focus on one factor, change in fuel type, as the inventory data are more or less complete. This is done by calculating Hg:C emission ratios from the electric power sector within the source region depicted in Figure 6, and as outlined in section 2.4. It should be noted that there is a high level of uncertainty in Hg emission factors and heat content among different types of coal and petroleum, as well as the influence of coal cleaning and pollution control technology, and we include Hg:C emission ratios based on both controlled and uncontrolled Hg emission factors for coal.

[31] Figure 3 presents the results along with the observed emissions ratio. In general, the emissions ratio derived from the fuel inventory data is higher than the measured value at Great Mountain, by an average of 30% (assuming controlled emissions) to 55% (uncontrolled emissions) over the five year period. This is not surprising since power plant emissions should have a higher bulk Hg:C emission ratio than what was observed at Great Mountain, which includes area, combustion and other, cleaner point sources which are relatively dilute with Hg. Exclusion of the mid-Atlantic states (New York, New Jersey, Pennsylvania) from the source region results in a slight decrease in the emissions ratio by an average of 0.001 g ton^{-1} (controlled; not shown in Figure 3) to 0.007 g ton^{-1} (uncontrolled) over the five year period. The change however is much smaller than the observed interannual variability, supporting our interpretation that interannual variability in source region cannot explain the observed change in the emissions ratio. Another important point is that the observed interannual variability is absent in the inventory data.

3.5. Regional Mercury Flux

[32] Figure 7 presents three estimates of monthly Hg emission flux over the source region for the winter season (January to April). Two inventory flux estimates are shown, one employing a coal emission factor without control devices, and the other with control (i.e., electrostatic precipitators or baghouses for particulates, and wet scrubbers for sulfur oxides).The inventory flux estimates are computed as the product of the C emission flux from the electric power sector and the inventory Hg:C emissions ratio (Figure 3). The atmospheric flux estimate is computed as the product of the emissions ratio derived from the tracer correlation analysis and the total C emission flux over the source region.

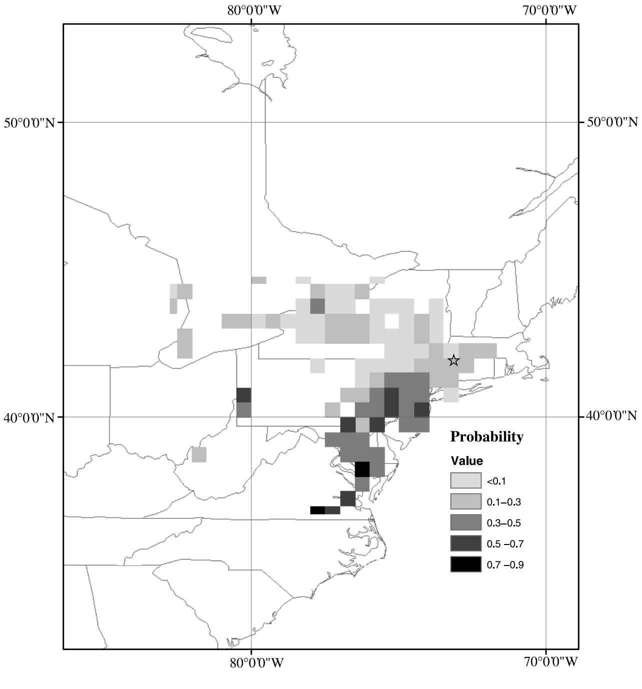

Figure 4. Map of the northeast United States showing PSCF grid values during winter 1999/2000. The star marks the location of Great Mountain.

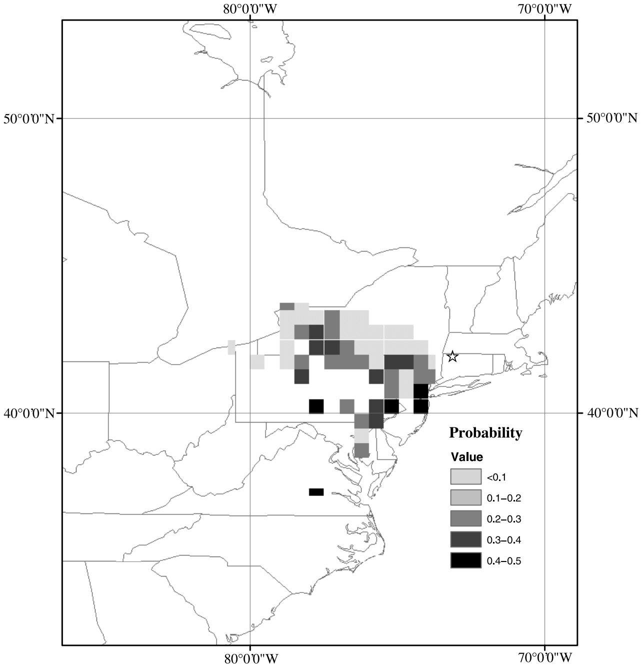

Figure 5. As in Figure 2 except for winter 2001/2002.

As stated above, the total C flux is held at a constant of 529 t C km^{-2} yr^{-1} over the study period, whereas the power sector C flux is computed annually from the inventory data. The total C flux is not sensitive to the interannual variation of the power sector C flux as the latter contributes 26–28% to the total annual C flux in the United States [*U.S. EIA*, 2004c]. For example, the power sector C flux decreased by ~20% from 2000 to 2001, while the state-by-state total C flux over the source region decreased by only ~3% over the same period [*Blasing et al.*, 2004].

[33] The two inventory flux estimates strongly correlate with the atmospheric flux (linear correlation coefficients of 0.75 and 0.81), with a clear declining trend from 1999 to 2002 and an increasing trend from 2002 to 2004. The atmospheric flux estimate appears to be highly sensitive to interannual changes among regional power plants, particularly in the amount of coal consumed. Therefore electricity generation plays a discernable role in the interannual variation of Hg flux to the atmosphere, and makes a significant contribution to the regional atmospheric Hg budget.

[34] Averaged over the five winters, the Hg flux from the power sector was 1.3 to 2.1 g km^{-2} $month^{-1}$ for controlled and uncontrolled coal combustion, respectively, and was at most only 75% of the flux from the atmospheric tracer analysis (2.8 g km^{-2} $month^{-1}$). However, since all U.S. power plants have, at minimum, particulate control technologies (EPRI, personal communication, 2005), the uncontrolled inventory flux can be interpreted as an upper bound. Assuming that the controlled inventory flux (1.3 g km^{-2} $month^{-1}$) is more representative of coal-fired power plants in the northeast, inventory Hg emissions from electric power were equal to approximately 47% of the flux from the atmospheric tracer analysis (2.8 g km^{-2} $month^{-1}$).

[35] This result is striking for several reasons. First, there has been a perceived reduction in emissions from municipal and medical waste combustion since the mid-1990s. According to *U.S. EPA* [1997], in the mid 1990s, emissions from municipal and medical waste combustion were approximately 43% of the total anthropogenic emission to the atmosphere in the northeastern United States, while electric utilities accounted for approximately 25%, commercial and industrial boilers produced approximately 23%, with the remaining 9% from minor sources such as cement manufacturing and sewage sludge incineration. The ratio of inventory (controlled) electric power flux to total atmospheric Hg flux (Figure 7) therefore represents approximately the same percentage of electric power emissions to the total inventory emissions reported by *U.S. EPA* [1997]. However, the EPA new source performance standards of 1997 sought to reduce emissions from municipal waste combustion as well as medical waste incinerators by >90% by 2000, and there is evidence that significant reduction has occurred in some northeastern states (see section 2.4). If this goal was achieved and emissions from other sources are assumed to have remained unchanged since 1995, the power sector emission would become by far the dominant source category over the study period, contributing no less than ~75% to the total emission over the source region.

[36] Second, at least half of the power sector emission is thought to be in reactive and particulate form [*Galbreath and Zygarlicke*, 1996; *Prestbo and Bloom*, 1995; *Carpi*, 1997; *U.S. EPA*, 1997], and the percentage of reactive Hg emitted by MWC is thought to be at least 60% [*U.S. EPA*, 1997]. The vast majority of TGM measured at Great Mountain is assumed to be Hg^0 (see section 2.1). This means that under the assumption of 50% speciation from electricity generators [*U.S. EPA*, 1997], the electric power sector in the source region can account for, on average, approximately 23–40% (based on range of controlled and

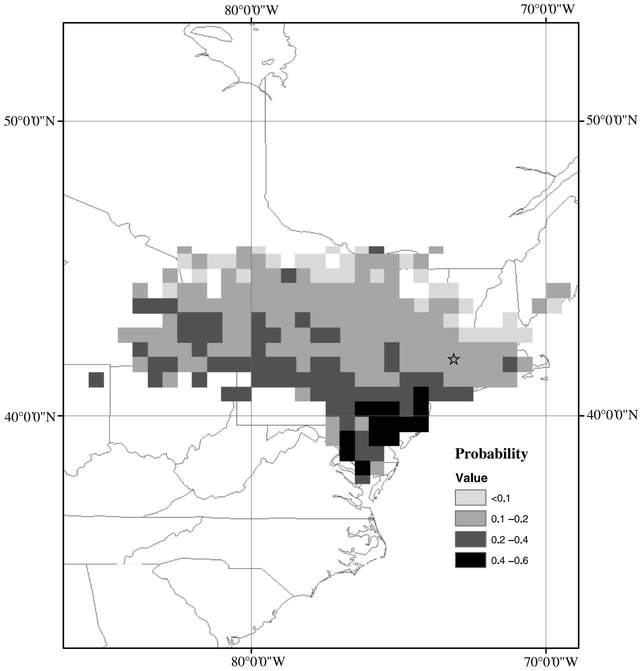

Figure 6. As in Figure 2 except for the entire study period, 1999/2000–2003/2004.

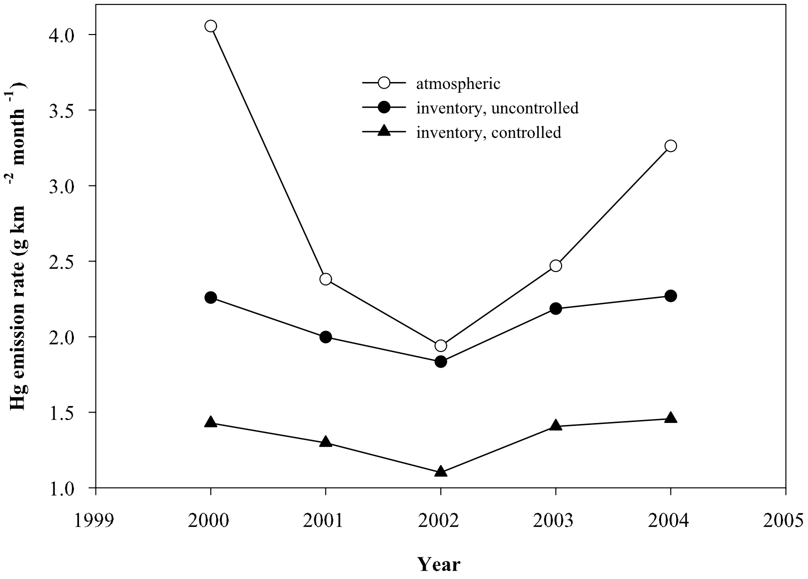

Figure 7. Three estimates of wintertime Hg emission flux over the whole source region. The linear correlation coefficients of uncontrolled and controlled electric power sector emissions with the atmospheric estimate are 0.81 and 0.75, respectively.

uncontrolled coal combustion) of the observed elemental Hg flux to the atmosphere. The disparity is even larger under the consideration that some studies have shown that reactive Hg may compose significantly more than 50% of total Hg emitted from electricity generators (60–80% [*Prestbo and Bloom*, 1995; *Carpi*, 1997]).

[37] Several possible explanations exist for the large disparity between the atmospheric and electric power inventory Hg flux. One possibility is that although the percentage of reactive Hg emitted into the atmosphere by coal combustion is believed to be high ($\geq$50%), it is possible that there is some reduction of reactive Hg to Hg^0 after emission to the atmosphere. Although observations are limited and the mechanism not understood, recent measurements by *Edgerton et al.* [2004] showed percentages of reactive Hg of only 10–15% at distances of 25-km downwind of power plants. Measurements presented by *Prestbo et al.* [2004] also suggest some conversion of reactive Hg to Hg^0 in power plant plumes. Significant conversion of reactive Hg to Hg^0 could theoretically explain much of the discrepancy between the atmospheric and electric power inventory fluxes if the uncontrolled Hg emission factor for coal (Table 2) is more representative of power plants in the region.

[38] Another possibility is that emissions from municipal and medical waste combustion have not been reduced to the level targeted by EPA in 1997 and still contribute significantly to the regional Hg emission rate. Recent work by NESCAUM (Northeast States for Coordinated Air Use Management) indicates that despite an overall decline in MWC emissions since 1998, MWC may have contributed approximately 21% of total Hg inventory emissions in the northeast (not including Pennsylvania) as recently as 2002. This may be due to lack of Hg control among many facilities, as well as an underestimate of Hg content in certain types of waste (e.g., fluorescent lamps) in the EPA inventory [*Northeast States for Coordinated Air Use Management* (*NESCAUM*), 2005].

[39] A fourth possible explanation is that other sources outside the waste combustion and electric power sectors make previously underestimated contributions to regional emissions. For example, steel foundries may be a significant emission source, emitting as much as 400 kg of Hg in New York and New Jersey in 2002 [*NESCAUM*, 2005]. Also, the source region (Figure 6) encompasses urban areas and highways with high traffic volume. Inventories [e.g., *U.S. EPA*, 1997] typically do not include vehicular emissions estimates since measurements are scarce, but recent studies have suggested emissions from vehicles may be a strong Hg source in urban areas [*Edgerton and Jansen*, 2004]. In any case, this study underscores the incomplete state of knowledge of Hg emission sources, and suggests that policy strategies for abatement of regional Hg pollution should not focus solely on the electric power sector.

[40] An uncertainty in our analysis is the Hg emission factor for coal combustion. *U.S. EPA* [1997] used an Hg emission factor of 16 lb/trillion Btu for uncontrolled, uncleaned coal. This is unrealistically high, as all coal-burning power plants are outfitted with some form of control technology that removes approximately 40% of Hg released from coal combustion in the United States [*U.S. EPA*, 1997; EPRI, personal communication, 2005]. The Hg emission factors used for coal-fired power plants in this study are based on more recent measurements from more than 80 plants with particulate and sulfur oxide control (5.5 lb/trillion Btu postcontrol; EPA 1999 ICR, reported by *EPRI* [2000]). However, *Chu and Porcella* [1995] and *U.S. EPA* [1989] reported median Hg emission factors for coal-burning power plants with minimum air pollution control that were closer to the uncontrolled value from the 1999 ICR measurements (10 lb/trillion Btu). The coal emission factor is therefore uncertain, as the data which exist are highly variable. For example, the measurements of *Chu and*

Porcella [1995] ranged from 4 to 22 lb/trillion Btu, and the data obtained by the 1999 ICR show similar variation. However, even if an emission factor as high as 16 lb Hg/trillion Btu (uncontrolled, uncleaned coal, from *U.S. EPA* [1997]) is assumed, the inventory flux for the power sector can only account for less than 60% of the atmospheric (primarily elemental) Hg flux.

[41] In addition, the emission factor used for oil combustion (6.8 lb/trillion Btu) is the same one used by *U.S. EPA* [1997]. Some studies suggest that this emission factor may be an order of magnitude too high [*Chu and Porcella*, 1995; *U.S. EPA*, 2005]. Substituting a lower emission factor for oil combustion (e.g., 0.46 lb/trillion Btu as reported by *Chu and Porcella* [1995]) would actually lower the electric power sector emission estimate even further (by, on average, 15% for controlled emissions).

4. Conclusions

[42] The combination of trajectory modeling and long-term tracking of the regional TGM emission rate by tracer analysis presented in this study provides a generally applicable framework for timely assessment of trends in regional-scale emission of Hg and other pollutants. This framework may enhance our ability to detect annual or seasonal changes in emission, determine source region of pollutants and evaluate the effectiveness of legislative strategies for reducing pollutant emissions.

[43] This study reveals that the regional Hg emission rate in the northeast United States can change significantly on an annual basis. After a 40% decrease from 1999/2000 to 2001/2002, the Hg emission rate increased steadily between the winters of 2001/2002 and 2003/2004. Trajectory analysis revealed little interannual variation in source region, suggesting that the significant variation in emissions cannot be explained by climatological impacts. The strong correlation between the trend in Hg fluxes calculated from inventory consumption data from the electric power sector and the atmospheric tracer analysis indicates that regional-scale emissions of Hg are highly sensitive to interannual variation in coal and petroleum consumption by the electric power sector in the source region.

[44] The inventory Hg flux for the electric power sector was much lower than anticipated when compared to the regional, atmospheric flux estimate. Coal-burning power plants are a focal point of pollution abatement strategies because they are thought to be by far the dominant anthropogenic source of Hg in the northeast, especially given the perceived reduction in emissions from municipal and medical waste combustion during the past 10 years. While the ultimate explanation for the discrepancy between the atmospheric measurement and the power sector Hg flux is not clear, possible explanations are that some percentage of reactive Hg emitted from power plants is quickly converted to elemental Hg, Hg emissions from municipal and medical waste combustion have not been curtailed to the targeted level in the northeast, that there are other anthropogenic source that have been grossly underestimated, or that a combination of all of these hypotheses results in the discrepancy.

[45] The significant interannual variation in the observed Hg emission highlights the importance of long-term observations and the tracer analysis. While inventory approaches, despite large uncertainty, provide valuable source-specific emissions estimates with high spatial resolution, the "top-down" approach of the atmospheric tracer correlation method provides near real-time estimates of emission of toxic pollutants with less uncertainty. This approach may therefore be a valuable means of verifying existing inventories and expanding overall knowledge of Hg emissions, and for evaluating the progress of policy strategies for pollution abatement.

[46] **Acknowledgments.** We thank J. William Munger of Harvard University for access to Harvard Forest CO_2 data as well as the use of the Harvard Forest tower to measure TGM during the summer of 2002. We also thank N. J. Themelis of Columbia University for information regarding combustion sources of Hg in the northeast. Funding for CO_2 measurements was supported by the Biological Environmental Research Program (BER), U.S. Department of Energy, through the northeast regional center of the National Institute for Global Environmental Change (NIGEC) under Cooperative Agreement DE-FC03-90ER61010, and by in-kind contribution from the Great Mountain Forest Corporation, Connecticut.

References

Andres, R. J., G. Marland, I. Fung, and E. A. Matthews (1996), 1° × 1° distribution of carbon dioxide emissions from fossil fuel and cement manufacture, 1950–1990, *Global Biogeochem. Cycles*, *10*, 419–429.

Bakwin, P. S., P. P. Tans, and P. C. Novelli (1994), Carbon monoxide budget in the Northern Hemisphere, *Geophys. Res. Lett.*, *21*, 433–436.

Bakwin, P. S., D. F. Hurst, P. P. Tans, and J. W. Elkins (1997), Anthropogenic sources of halocarbons, sulfur hexafluoride, carbon monoxide and methane in the southeastern United States, *J. Geophys. Res.*, *102*, 15,915–15,925.

Blasing, T. J., C. T. Broniak, and G. Marland (2004), Estimates of annual fossil-fuel CO_2 emitted for each state in the U.S.A. and the District of Columbia for each year from 1960 through 2001, in *Trends: A Compendium of Data on Global Change*, Carbon Dioxide Inf. Anal. Cent., Oak Ridge Natl. Lab., U.S. Dep. of Energy, Oak Ridge, Tenn. (Available at http://cdiac.esd.ornl.gov/trends/trends.htm)

Blasing, T. J., C. T. Broniak, and G. Marland (2005), The annual cycle of fossil-fuel carbon dioxide emissions in the United States, *Tellus, Ser. B*, *57*, 107–115.

Brunke, E.-G., C. Labuschagne, and F. Slemr (2001), Gaseous mercury emissions from a fire in the Cape Peninsula, South Africa, during January 2000, *Geophys. Res. Lett.*, *28*, 1483–1486.

Butler, T. J., G. E. Likens, and B. J. B. Stunder (2001), Regional scale impacts of Phase I of the Clean Air Act Amendments in the USA: The relation between emissions and concentrations, both wet and dry, *Atmos. Environ.*, *35*, 1015–1028.

Carpi, A. (1997), Mercury from combustion sources: A review of the chemical species emitted and their transport in the atmosphere, *Water Air Soil Pollut.*, *56*, 745–767.

Chen, H., X. Yang, and C. Perkins (2004), Trend and variability of total gaseous mercury (TGM) in the state of Connecticut during 1997–1999, *Water Air Soil Pollut.*, *151*, 103–116.

Cheng, M.-D., and W. H. Schroeder (2000), Potential atmospheric transport pathways for mercury measured in the Canadian high Arctic, *J. Atmos. Chem.*, *35*, 101–107.

Cheng, M.-D., P. K. Hopke, L. Barrie, A. Rippe, M. Olson, and S. Landsberger (1993), Qualitative determination of source regions of aerosol in Canadian high Arctic, *Environ. Sci. Technol.*, *27*, 2063–2071.

Chu, P., and D. B. Porcella (1995), Mercury stack emissions from U.S. electric utility power plants, *Water Air Soil Pollut.*, *80*, 135–144.

Cooper, O. R., J. L. Moody, T. D. Thornberry, M. S. Town, and M. A. Carroll (2001), PROPHET 1998 meteorological overview and air-mass classification, *J. Geophys. Res.*, *106*, 24,289–24,299.

Draper, N. R. (1992), Straight line regression when both variables are subject to error, *Tech. Rep. 890*, 18 pp., Dep. of Stat., Univ. of Wis., Madison.

Draxler, R. R., and G. D. Hess (1997), Description of the HYSPLIT_4 modeling system, *NOAA Tech. Memo. ERL ARL-224*, 24 pp.

Edgerton, E. S., and J. J. Jansen (2004), Elemental Hg measurements in Atlanta, GA, USA: Evidence for mobile sources?, paper presented at 7th International Conference on Mercury as a Global Pollutant, Oak Ridge Natl. Lab., Ljubjana, Slovenia, 28 June to 2 July.

Edgerton, E. S., B. E. Hartsell, and J. J. Jansen (2004), Speciated Hg measurements at a rural site near Atlanta, GA, USA, paper presented at

7th International Conference on Mercury as a Global Pollutant, Oak Ridge Natl. Lab., Ljubjana, Slovenia, 28 June to 2 July.

Electric Power Research Institute (1996), Mercury in the environment—A research update, final report. Palo Alto, Calif.

Electric Power Research Institute (2000), An assessment of mercury emissions from U.S. coal-fired power plants, *Rep. 1000608*, Palo Alto, Calif.

Fischer, E., and R. Talbot (2005), Regional NO_3^- events in the northeastern United States related to seasonal climate anomalies, *Geophys. Res. Lett.*, *32*, L16804, doi:10.1029/2005GL023490.

Friedli, H. R., L. F. Radke, J. Y. Lu, C. M. Banic, W. R. Leaitch, and J. I. MacPherson (2003), Mercury emissions from burning of biomass from temperate North American forests: Laboratory and airborne measurements, *Atmos. Environ.*, *37*, 253–267.

Galbreath, K. C., and C. J. Zygarlicke (1996), Mercury speciation in coal combustion and gasification flue gases, *Environ. Sci. Technol.*, *30*, 2421–2426.

Han, Y.-J., T. M. Holsen, S.-O. Lai, P. K. Hopke, S.-M. Yi, J. Pagano, L. Falanga, M. Milligan, and C. Andolina (2004), Atmospheric gaseous mercury concentrations in New York State: Relationships with meteorological data and other pollutants, *Atmos. Environ.*, *38*, 6431–6446.

Han, Y.-J., T. M. Holsen, P. K. Hopke, and S.-M. Yi (2005), Comparison between back-trajectory based modeling and Lagrangian backward dispersion modeling for locating sources of reactive gaseous mercury, *Environ. Sci. Technol.*, *39*, 1715–1723.

Kim, K., and M. Kim (2001), The temporal distribution characteristics of total gaseous mercury at an urban monitoring site in Seoul during 1999–2000, *Atmos. Environ.*, *35*, 4253–4263.

Kim, K.-H., S. E. Lindberg, and T. P. Meyers (1995), Micrometeorological measurements of mercury vapor fluxes over background forest soils in eastern Tennessee, *Atmos. Environ.*, *29*, 267–282.

Landis, M. S., A. F. Vette, and G. J. Keeler (2002), Atmospheric mercury in the Lake Michigan Basin: Influence of the Chicago/Gary urban area, *Environ. Sci. Technol.*, *36*, 4508–4517.

Lee, X., O. R. Bullock, and R. J. Andres (2001), Anthropogenic emission of mercury to the atmosphere in the northeast United States, *Geophys. Res. Lett.*, *28*, 1231–1234.

Lin, C.-J., M.-D. Cheng, and W. H. Schroeder (2001), Transport patterns and potential sources of total gaseous mercury measured in Canadian high Arctic in 1995, *Atmos. Environ.*, *35*, 1141–1154.

Lindberg, S. E., and W. J. Stratton (1998), Atmospheric mercury speciation: Concentrations and behavior of reactive gaseous mercury in ambient air, *Environ. Sci. Technol.*, *32*, 49–57.

Lynam, M. M., and G. L. Keeler (2005), Automated speciated mercury measurements in Michigan, *Environ. Sci. Technol.*, *39*, 9252–9262.

Mao, H., and R. Talbot (2004), Relationship of surface O_3 to large-scale circulation patterns during two recent winters, *Geophys. Res. Lett.*, *31*, L06108, doi:10.1029/2003GL018860.

Massachusetts Department of Environmental Protection (2006), Massachusetts Fish Tissue Mercury Studies: Long-term monitoring results, 1999–2004, Boston, Mass. (Available at http://www.mass.gov/dep/toxics/stypes/hgres.htm)

Moody, J. L., J. W. Munger, A. H. Goldstein, D. J. Jacob, and S. C. Wofsy (1998), Harvard Forest regional-scale air mass composition by Patterns in Atmospheric Transport History (PATH), *J. Geophys. Res.*, *103*(D11), 13,181–13,194.

Nadim, F., C. Perkins, S. Liu, R. J. Carley, and G. E. Hoag (2001), Long-term investigation of atmospheric mercury contamination in Connecticut, *Chemosphere*, *45*, 1033–1043.

Northeast States for Coordinated Air Use Management (2005), Inventory of anthropogenic mercury emissions in the northeast, Boston, Mass. (Available at http://www.nescaum.org)

Poissant, L. (1999), Potential sources of atmospheric total gaseous mercury in the St. Lawrence River valley, *Atmos. Environ.*, *33*, 2537–2547.

Poissant, L., M. Pilote, C. Beauvais, P. Constant, and H. H. Zhang (2005), A year of continuous measurements of three atmospheric mercury species (GEM, RGM and Hg_p) in southern Quebec, Canada, *Atmos. Environ.*, *39*, 1275–1287.

Potosnak, M. J., S. C. Wofsy, A. S. Denning, T. J. Conway, J. W. Munger, and D. H. Barnes (1999), Influence of biotic exchange and combustion sources on atmospheric CO_2 concentrations in New England from observations on a forest flux tower, *J. Geophys. Res.*, *103*, 9561–9569.

Prestbo, E. M., and N. S. Bloom (1995), Mercury speciation adsorption (MESA) method for combustion flue gas methodology, artifacts, intercomparison and atmospheric implications, *Water Air Soil Pollut.*, *80*, 145–158.

Prestbo, E. M., et al. (2004), Interconversion of emitted atmospheric mercury species in coal-fired power plant plumes, paper presented at 7th International Conference on Mercury as a Global Pollutant, Oak Ridge Natl. Lab., Ljubjana, Slovenia, 28 June to 2 July.

Richardson, A. D. (2000), Coarse root elongation rate estimates for interior Douglas-fir, *Tree Physiol.*, *20*, 825–829.

Sigler, J. M., and X. Lee (2006), Gaseous mercury in background forest soil in the northeastern United States, *J. Geophys. Res.*, *111*, G02007, doi:10.1029/2005JG000106.

Sigler, J. M., X. Lee, and W. Munger (2003), Emission and long-range transport of gaseous mercury from a large-scale Canadian boreal forest fire, *Environ. Sci. Technol.*, *37*, 4343–4347.

Slemr, F., G. Schuster, and W. Seiler (1985), Distribution, speciation and budget of atmospheric mercury, *J. Atmos. Chem.*, *3*, 407–434.

Stehr, J. W., R. R. Dickerson, K. A. Hallock-Waters, B. G. Doddridge, and D. Kirk (2000), Observations of NO_y, CO and SO_2 and the origin of reactive nitrogen in the eastern United States, *J. Geophys. Res.*, *105*, 3553–3563.

Suntharalingam, P., D. J. Jacob, P. I. Palmer, J. A. Logan, R. M. Yantosca, Y. Xiao, M. J. Evans, D. G. Streets, S. L. Vay, and G. W. Sachse (2004), Improved quantification of Chinese carbon fluxes using CO_2/CO correlations in Asian outflow, *J. Geophys. Res.*, *109*, D18S18, doi:10.1029/2003JD004362.

Themelis, N. J., and A. F. Gregory (2002), Mercury emissions from high-temperature sources in the NY/NJ Hudson-Raritan basin, *Am. Soc. Mech. Eng. Proc. NAWTEC*, *10*, 205–215.

U.S. Energy Information Administration (2004a), Electric power annual 2003, Washington, D. C. (Available at http://www.eia.doe.gov/cneaf/electricity/epa/epa_sum.html)

U.S. Energy Information Administration (2004b), Annual energy review 2003, Washington, D. C. (Available at http://www.eia.doe.gov/aer/)

U.S. Energy Information Administration (2004c), Emissions of greenhouse gases in the United States, Washington, D. C. (Available at http://www.eia.doe.gov/oiaf/1605/ggrpt/index.html)

U.S. Energy Information Administration (2005), Monthly energy review (archives), Washington, D. C. (Available at http://www.eia.doe.gov/emeu/mer/contents.html)

U.S. Environmental Protection Agency (1989), Estimating air toxics emissions from coal and oil combustion sources, *EPA-450/2-89-001*, Research Triangle Park, N. C.

U.S. Environmental Protection Agency (1997), Mercury study report to Congress, Washington, D. C. (Available at http://www.epa.gov/mercury/report.htm)

U.S. Environmental Protection Agency (2005), AP-42, Compilation of air pollutant emission factors, Research Triangle Park, N. C. (Available at http://www.epa.gov/ttn/chief/ap42/index.html)

Wang, T., T. F. Cheung, Y. S. Li, X. M. Yu, and D. R. Blake (2002), Emission characteristics of CO, NO_x, SO_2 and indications of biomass burning observed at a rural site in eastern China, *J. Geophys. Res.*, *107*(D12), 4157, doi:10.1029/2001JD000724.

Weiss-Penzias, P., D. A. Jaffe, A. McClintick, E. M. Prestbo, and M. S. Landis (2003), Gaseous elemental mercury in the marine boundary layer: Evidence for rapid removal in anthropogenic pollution, *Environ. Sci. Technol.*, *37*, 3755–3763.

Wofsy, S. C., S.-M. Fan, D. R. Blake, J. D. Bradshaw, S. T. Sandholm, H. B. Singh, G. W. Sachse, and R. C. Harris (1994), Factors influencing atmospheric composition over subarctic North America during summer, *J. Geophys. Res.*, *99*, 1887–1897.

Xiao, Z. F., J. Munthe, W. H. Schroeder, and O. Lindqvist (1991), Vertical fluxes of volatile mercury over forest soil and lake surface in Sweden, *Tellus*, *43*, 276–279.

X. Lee, School of Forestry and Environmental Studies, Yale University, 205 Prospect Street, New Haven, CT 06511, USA.

J. M. Sigler, School of Forestry and Environmental Studies, Yale University, 210 Prospect Street, New Haven, CT 06511, USA. (jeffrey.sigler@yale.edu)