

# Office of Science

## Research Project Summary

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### Ambient elemental, reactive gaseous and particle-bound mercury concentrations in New Jersey, U.S.: measurements and associations with wind direction

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#### Abstract

Two and a half years of ambient concentrations of elemental mercury ( $\text{Hg}^0$ ), reactive gaseous mercury (RGM), and particle-bound mercury ( $\text{Hg}_p$ ) were collected at measurement sites at Elizabeth, NJ and New Brunswick, NJ with Tekran sampling units in order to gather information on ambient atmospheric mercury levels, to determine whether these levels could be associated with known sources, and to develop a method to analyze these data. The data were processed, summarized, and evaluated from a variety of perspectives. Data quality control and quality assurance procedures are described. Wind direction and wind speed data were also collected. Significant temporal variations in concentrations of all three species were observed. Some significant directional variations were also seen. The sporadic nature of many of the temporal variations is consistent with and could reflect highly variable emission patterns from anthropogenic mercury sources. Overall mean concentrations of all species were determined. These were, for  $\text{Hg}^0$ ,  $\text{Hg}_p$ , and RGM respectively:  $2.25 \pm 0.04$  nanograms per cubic meter ( $\text{ng}/\text{m}^3$ ),  $8.21 \pm 0.39$  picograms per cubic meter ( $\text{pg}/\text{m}^3$ ), and  $8.93 \pm 0.31$   $\text{pg}/\text{m}^3$  (arithmetic means and 95% confidence intervals) at Elizabeth, and  $2.15 \pm 0.02$   $\text{ng}/\text{m}^3$ ,  $10.73 \pm 0.45$   $\text{pg}/\text{m}^3$ , and  $6.04 \pm 0.30$   $\text{pg}/\text{m}^3$  at New Brunswick. Mean concentrations were determined for 16 different sectors representing wind directions. The impact of one known large source is suggested by these data. Reasons for some directional variations are not apparent and suggest a need for further investigation.

#### Introduction

Mercury (Hg) is persistent and toxic and can enter water bodies where it can be converted to methyl mercury (MeHg), its biologically available form. Mercury is emitted to the atmosphere through both anthropogenic and natural processes, with anthropogenic sources contributing 50-75% of the total.<sup>1,2</sup> Much of these emissions are from coal-burning power plants, waste incinerators,<sup>3</sup> and iron and steel manufacturing plants.<sup>4</sup>

Ambient concentrations of total gaseous mercury have been found to exhibit wide variability.<sup>5</sup> These inconsistencies can be due to fluctuations in emission sources from one locale to the next, variations in mercury content of feedstocks or fuels and differences in combustion conditions. Atmospheric and environmental variables, such as precipitation and wind speed/direction, can also affect mercury air concentrations.

The goals of this study were to gain information on ambient levels of atmospheric mercury species in New Jersey, to ascertain whether these observed levels could

be reliably associated with known sources of mercury and to establish a protocol to interpret these data. Two and one-half year's worth of data were collected at two sites. The two sites sampled were Elizabeth, NJ and New Brunswick, NJ.

Elizabeth is a highly industrialized area with a number of possible mercury sources in close proximity. The Elizabeth Tekran unit is located directly adjacent to the toll plaza of exit 13 of the New Jersey Turnpike, in an area that is surrounded by several major highway ramps and overpasses. The site is within a mile of the Arthur Kill, a tidal estuary on the western edge of Staten Island, New York.

New Brunswick is a suburbanized area dominated by the Rutgers University campus, several pharmaceutical companies, and residential development. There are no known significant mercury sources in close proximity to the site. There is a solid waste transfer station located more than five miles away and an active landfill within two miles of the site. The New Brunswick Tekran unit is located in a cleared area near a forested portion of the university campus within a mile of a major highway.

## Methods

### Equipment

This study used automated mercury speciation analyzer systems manufactured by Tekran, Inc. (Toronto, Canada) that are capable of measuring elemental mercury, ( $\text{Hg}^0$ ); reactive gas mercury, (RGM); and particulate bound mercury, ( $\text{Hg}_p$ ), in ambient air.

$\text{Hg}^0$  is sampled for 5 min at 1 l/min, while the reactive gas and particulate mercury are sampled at 10 l/min for 60 min. These different sample volumes are required since  $\text{Hg}^0$  is present in the ambient air at the  $\text{ng}/\text{m}^3$  level, while reactive gas and particulate mercury are present at the  $\text{pg}/\text{m}^3$  levels.

Each sample cycle is two hours in duration. The analyzer collects air continuously for the first hour, sampling is then stopped and the air collected in the first hour is analyzed, resulting in 12  $\text{Hg}^0$  readings, one  $\text{Hg}_p$  reading, and one RGM reading per sample cycle. The manufacturer reports a minimum detection limit of  $<0.1 \text{ ng}/\text{m}^3$  for  $\text{Hg}^0$ , and a minimum detection limit in the range of 1 to 5  $\text{pg}/\text{m}^3$  for RGM and  $\text{Hg}_p$ .

### Data management procedures

The data underwent a series of steps to determine validity. In order to be considered valid, the data had to conform to a specific pattern; they had to fall within a specific range based on clean cycles, generally accepted background levels, and sample volume and baseline deviation results. Once validity was established, data were associated with corresponding wind speed and direction collected at both sites.

## Results and Discussion

### Variability in data

A striking feature of the measured concentrations of all three species,  $\text{Hg}^0$ , RGM, and  $\text{Hg}_p$ , is the infrequent but persistent presence of a small number of high readings. Nothing unusual or consistent from a meteorological standpoint was observed at the times when these high readings occurred, and, because the data were subjected to the comprehensive quality control and quality assurance procedures described above, there is no reason to suspect the values are a result of errors in sampling or analysis. These data exemplify a consistent variability that is seen throughout the entire data set for all three species. Such variability has been observed elsewhere. A likely explanation, as noted above, is that short-scale temporal variations in emissions from industrial and anthropogenic sources are responsible.

### Mean Values

Over 3,000 each of  $\text{Hg}_p$  and RGM 1-h average samples, and more than 5,000  $\text{Hg}^0$  samples, each representing a 1-h composite of twelve discrete  $\text{Hg}^0$  values, were validated for each site over a 2 ½ year period. The large

number of samples permits determination of well-constrained mean values. These values are shown in Table 1.

**Table 1**  $\text{Hg}^0$ , RGM, and  $\text{Hg}_p$  mean values with 95% confidence intervals

|               | Elizabeth                             | New Brunswick                          |
|---------------|---------------------------------------|--|
| $\text{Hg}^0$ | $2.25 \pm 0.04 \text{ ng}/\text{m}^3$ | $2.15 \pm 0.02 \text{ ng}/\text{m}^3$  |
| $\text{Hg}_p$ | $8.21 \pm 0.39 \text{ pg}/\text{m}^3$ | $10.73 \pm 0.45 \text{ pg}/\text{m}^3$ |
| RGM           | $8.93 \pm 0.31 \text{ pg}/\text{m}^3$ | $6.04 \pm 0.30 \text{ pg}/\text{m}^3$  |

The  $\text{Hg}^0$  values are consistent with an annual average concentration of 2.0 to 2.6  $\text{ng}/\text{m}^3$  reported for Northeast US locations.<sup>6</sup> The mean  $\text{Hg}_p$  values are consistent with results of a recent study<sup>7</sup> performed in New Jersey that measured fine aerosol ( $\text{PM}_{2.5}$ ) mercury concentrations at five sites; mean values ranged from 4.9 to 16  $\text{pg}/\text{m}^3$ . The RGM concentrations measured in the current study are lower than the 40  $\text{pg}/\text{m}^3$  concentration measured in Solomons, MD<sup>8</sup> but are higher than the mean values in the range of 3 to 4  $\text{pg}/\text{m}^3$  recently measured in Quebec, Canada.<sup>9,10</sup>

### Estimated dry deposition and portion of total atmospheric deposition

Mean volume-weighted wet deposition of mercury in New Jersey during the period from 1997 to 2002 was measured as 13.8  $\mu\text{g}/\text{m}^2/\text{year}$ .<sup>11</sup> Additional dry deposition in the form of settling of particles containing mercury and adsorption of gaseous mercury species to terrestrial surfaces also occurs. This deposition quantity is difficult to measure directly but can be inferred from  $\text{Hg}_p$  and RGM concentrations by assuming a dry deposition velocity ( $V_d$ ) for these species. Based on  $V_d$ s determined for these species from previous studies,<sup>12,13,14,15</sup> the  $V_d$  of RGM was assumed to be approximately 2.5 cm/s, and that of  $\text{Hg}_p$  to be approximately 0.5 cm/s; it was also assumed that the mean concentrations of these two species found in this study are typical of the entire state. From this, a state-wide dry deposition of mercury in the form of  $\text{Hg}_p$  and RGM of between 6 and 9  $\mu\text{g}/\text{m}^2/\text{year}$  can be inferred.

In addition, dry deposition of  $\text{Hg}^0$  occurs. A range of  $V_d$ s of  $\text{Hg}^0$  has been modeled in previous studies.<sup>16</sup> Assuming a  $V_d$  for  $\text{Hg}^0$  in New Jersey of 0.06 cm/s during a 5-month growing season suggests that dry deposition of  $\text{Hg}^0$  to terrestrial surfaces with deciduous tree cover adds additional depositions in the range of 17  $\mu\text{g}/\text{m}^2/\text{year}$ . Deposition of all species to forested surfaces could be higher where conifers predominate, such as the Pinelands, because foliage is present year-round. Some of the mercury that deposits to foliage surfaces or is adsorbed to the interior of leaves and other plant tissues can be expected to re-volatize at some point during the life cycle of the foliage, and thus would not be perma-

nently deposited in a watershed. Nevertheless, it appears that dry deposition could represent a significant portion of the total deposition of mercury to land surfaces in New Jersey, and it is likely that the total deposition is considerably higher than the wet deposition quantity. Augmentation of wet deposition with a significant component of dry deposition is consistent with findings of some recent research based on concentrations of mercury in lake sediments that estimated deposition rates of mercury to watersheds in New Jersey within the last several decades to be in the range of 20  $\mu\text{g}/\text{m}^2/\text{year}$  to over 200  $\mu\text{g}/\text{m}^2/\text{year}$ .<sup>17</sup>

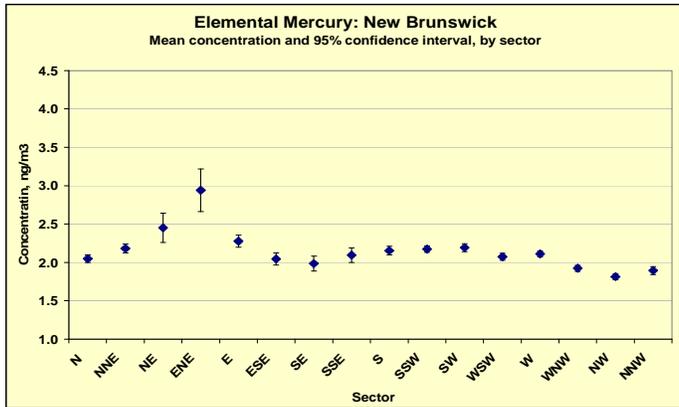


Fig. 1 Mean Hg<sup>0</sup> concentrations and 95% confidence interval, by sector: New Brunswick

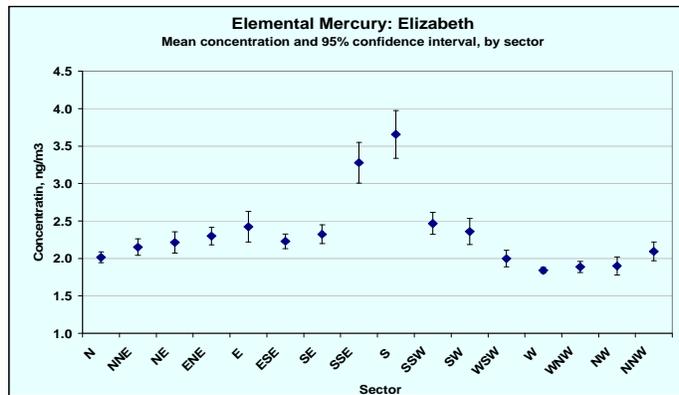


Fig. 2 Mean Hg<sup>0</sup> concentrations and 95% confidence interval, by sector: Elizabeth

### Directional Signals

There are significant differences in mean concentrations of the measured species associated with differences in wind direction. These concentration differences are apparent when each wind direction is grouped into one of 16 sectors, with each sector representing 22.5° of the compass. A Kruskal-Wallis (nonparametric ANOVA) test was performed with the data. For the Elizabeth site, it showed that variation among the sector medians is significantly greater than expected by chance,  $P < 0.0001$ . A Dunn's multiple comparisons test indicated that the difference between some sectors was more pronounced than between others. For example, at the Elizabeth site, a comparison of the

S (due south) sector showed it to be different from every sector with a  $P$  value  $< 0.001$  except for the SSE sector, with which it was different with  $P < 0.05$ . Mean values and 95% confidence intervals of each of the three measured species were determined for each sector at each location. These values are shown for New Brunswick and Elizabeth in Figures 1 and 2.

There is more variation in the Hg<sub>p</sub> and RGM values than in the Hg<sup>0</sup> values, and there are also significant differences in the mean values when the wind is from certain directions. Most notable are the higher mean Hg<sup>0</sup> values when wind is from the direction of northeast and east-northeast at New Brunswick (Fig. 1), and higher mean Hg<sup>0</sup> values when wind is from the direction of south-southeast and south at Elizabeth (Fig. 2). Although there are relatively large uncertainties, higher mean Hg<sub>p</sub> values also appear to exist when the wind is from these directions.

A comparison of values associated with different wind directions and the locations of known or suspected sources of mercury in New Jersey, shown in Fig. 3, reveals at least one likely correlation. The apparently higher reading of Hg<sup>0</sup> at New Brunswick when the wind is from the direction of east-northeast is consistent with the presence of an electric-arc furnace steel manufacturer in Sayreville. This source, the more western of the two iron and steel plants shown in Fig. 3, is estimated to release several hundred pounds of mercury per year. However, the strong signal of Hg<sup>0</sup> at Elizabeth when the wind is from the S and SSE sectors remains a puzzle. There are some mercury-contaminated hazardous waste sites in this direction, but two on-site investigations found no significantly elevated readings that would be consistent with a flux high enough to cause elevated readings at Elizabeth. It is possible that a site or sites in Staten Island or, perhaps, sediments or ship traffic in the Arthur Kill could be involved. However, no likely

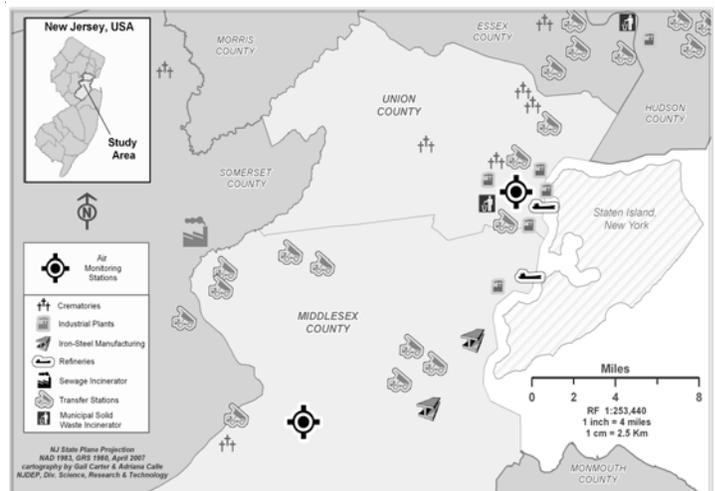


Fig. 3 Study area: known and potential sources

candidates have been identified. Adding to the puzzle is the fact that the higher than average mean concentrations appear to be caused by unusually high values that occur sporadically, with no apparent relation to temperature or tidal cycles, which would be expected if the readings were associated with contaminated environmental reservoirs or waste disposal sites.

### **Conclusions**

Measured ambient atmospheric concentrations of elemental, reactive gaseous, and particle-bound mercury showed wide temporal variations that were consistent with wide variations from anthropogenic sources, many of which are known to be sporadic in their emission patterns. Overall mean concentrations were well-constrained, however, and were relatively consistent with measurements that have been reported elsewhere.

Consistent differences in mean concentrations were found when measurements were grouped into sectors based on wind direction. Particularly notable were the findings that the mean concentration of elemental mercury at the New Brunswick facility was about 50% higher when wind was from the east-northeast than when wind was from other sectors, and that at the Elizabeth facility the mean concentration of elemental mercury was nearly twice as high when the wind was from the south and south-southeast than when the wind was from other sectors.

The first finding is consistent with the presence of a known large mercury emissions source, an electric-arc steel manufacturing plant in Sayreville, which is east-northeast of the New Brunswick site. The second finding, however, cannot be explained by locations of known sources, and may be the result of poorly characterized or unknown source or meteorological or other variables that are not readily apparent. Further research is necessary to identify the cause of this anomaly and to shed further light on directional influences on other observed variations in concentration.

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<sup>4</sup> Randy England, deceased June 10, 2007. Randy made major contributions to this research; his wisdom and kindness will be missed by all who had the pleasure and privilege of working with him, and this work is dedicated to his memory.

**RESEARCH PROJECT SUMMARY**

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