

Air Monitoring Plan for Establishing an
Ambient Mercury Baseline for
New York State

Local-Scale Air Toxics Ambient Monitoring Project
In Response to EPA
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Appendix A: Final Report Summary in Power Point Format: Presented at the EPA National Air Toxics Meeting April 4-7, 2011 (Dallas, Texas) (weblink)

Appendix B: Presentations of Study Findings

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1. Introduction

The New York State Department of Environmental Conservation (NYSDEC) appreciated the opportunity to pursue this monitoring and data analysis project under the US EPA's Local-Scale Air Toxics Monitoring Grant program. The field monitoring portion of the project was delayed due to the extensive developmental work necessary to make the remote field operation of the Tekran instrumentation reliable. The data and experiences gained during the project have assisted in the development of standard operating procedures (SOPs) which are now in use by all of the agencies participating in the Atmospheric Mercury Network (AMNet) program, and the collected data have highlighted unexpected Hg concentrations in large urban areas. The NYSDEC is planning to continue the measurements initiated under this program and has received indication that grant funding will be provided by the Great Lakes Commission (GLC) for this purpose.

2. Responsiveness to Grant Objectives

This project was submitted under the Community-Scale Monitoring Objective of the US EPA's grant solicitation. The c. and d. subcategories of this objective as specified in the Grant Application are:

- c. Developing a baseline reference frame of air quality concentrations that support estimates of community exposure and provide the basis for the longer term measuring of progress of a planned emissions strategy program.
- d. Characterizing pollutants that are not ubiquitous (e.g., ambient/divalent mercury emissions, or pollutants from source categories that will be considered in future residual risk determinations).

This project's results are being used to develop a baseline and to characterize non-ubiquitous pollutants including ambient elemental, divalent and particle bound mercury. The NYSDEC is planning to continue the measurements so that the baseline data collected to date can be used as a reference to determine if the expected environmental results are obtained from planned and enacted emission controls at the state, regional, and federal level.

The work involved in collecting the data also provided information that pertains to the grant categories of Method Development and Method Evaluation and Comparison. Much of the development work performed in the NYSDEC lab on the Tekran systems contributed to the development of the comprehensive SOP developed in conjunction with other AMNet Hg researchers.

3. Project Background

Mercury is a highly toxic compound present in many consumer products, and is also released into the atmosphere through combustion of coal and other incineration processes. Mercury deposition continues to be a major threat to public health in New York State. Due to the high levels of mercury in freshwater fish, the NYSDEC and the New York State Department of

Health (NYSDOH) have issued specific warnings advising that pregnant women and children should not consume any servings of certain fish species caught in more than 130 water bodies, many of which have no known direct inputs of mercury other than atmospheric deposition (NYSDOH, 2009). In addition, there is a general fish advisory alerting the public not to eat more than one meal (one-half pound) per week of fish taken from New York's fresh waters and some marine waters at the mouth of the Hudson River. This list of restricted water bodies and fish species continues to grow each year. Many of the lakes sampled are in remote mountainous areas of New York that do not have any known mercury inputs other than atmospheric deposition.

The US EPA's Mercury Study Report to Congress concluded that the Great Lakes, the Ohio River Valley, the Northeastern United States and scattered areas in the South are predicted to have the highest annual deposition rates of mercury in the United States (US EPA, 1987). A study of mercury deposition in the Adirondack Mountain region of New York showed that sources located in the United States contribute 42% of the mercury deposition in this region (NYSERDA, 2002). Regional modeling work conducted by the Electric Power Research Institute concluded that 80% of the mercury deposition that occurs in the southern New York region and surrounding states comes from sources based in the United States (Levin, 2003).

On March 15, 2005, the US EPA promulgated the Clean Air Mercury Rule (CAMR; US EPA, 2006). The final rule established a cap-and-trade program for the control of mercury from coal-fired utilities and required states to create and submit regulations to implement the cap-and-trade program and distribute mercury allocations to affected units. In 2008 the CAMR was vacated, and in March 2011 the US EPA proposed new national standards for mercury, arsenic, and other air toxics from utilities (<http://www.epa.gov/airquality/powerplanttoxics>). This project will allow New York State to establish a baseline of speciated mercury compounds in the ambient air and in wet deposition prior to implementing statewide and federal emission control strategies.

Recently the NYSDEC finalized its own regulations to address emission reductions in coal-fired utilities, 6 NYCRR Part 246. Part 246 will achieve a 90% reduction from 1999 levels from the coal-fired electricity generating units in two phases. Phase I of Part 246 imposes a facility-wide mercury emission limitation, based upon the state mercury budget distributed to New York State by the US EPA. The NYSDEC will limit applicable facilities to a specified amount of mercury emissions expressed in pounds per year in total, not to exceed New York's mercury state budget. The facility-wide emission limitations will be in effect from 2010 to 2014. The Department is withholding 40 pounds of emissions from the overall State mercury budget of 786 pounds for new sources. Phase II, starting in 2015, will establish a facility-wide emission limit. This is expected to result in annual emissions of mercury from applicable units of less than 175 pounds. In addition to coal-fired utility emissions, the US EPA has recently issued rules affecting emissions from Portland cement plants. When fully implemented in 2013, mercury emissions from cement plants are expected to be reduced by 92%.

The goal of this project is to establish a reference baseline for mercury air concentrations and wet deposition in urban areas in New York State. In ambient air, more than 90% of mercury is in the elemental form (Hg(0)). Compared to Hg(0), only trace amounts of mercury are particle-bound mercury (PBM) or reactive gaseous mercury (RGM; divalent or oxidized forms); however, these

forms are highly reactive and are readily wet- and dry-deposited to ground and water surfaces. Hence, to fully characterize the mercury burden in these areas, it is crucial to measure both ambient air concentrations and wet deposition. These baseline data will be used in conjunction with other mercury monitoring measurements within New York and the surrounding states to track the overall progress of mercury reduction strategies for the two largest source categories, municipal waste combustors and coal fired electric utilities. Establishing a baseline as well as documenting progress with regulatory actions is consistent with the federal program to quantify regulatory achievement under the Government Performance and Results Act.

4. Site Descriptions

The NYSDEC used this US EPA funding to operate a suite of instruments consisting of Tekran Model 2537B, 1130 and 1135 Mercury Speciation Units and an external calibrator at two urban locations in New York – Rochester and Bronx – to characterize ambient air concentrations of PBM, RGM, and Hg(0). The funding also allowed the NYSDEC to measure wet Hg deposition using a Mercury Deposition Network (MDN) collector and Universal Recording Rain Gauge. Table 1 lists the location information for the two monitoring sites. Figure 1 presents aerial views of the locations of these two sites, indicated by red arrows, in relation to nearby roadways and commercial and residential areas. Figure 2 displays the mercury instrumentation installed at the Bronx site.

The Rochester monitoring site is located in a metropolitan area of about 1.03 million people, along the southern shore of Lake Ontario. This site is located downwind of numerous coal-fired power plants in western New York and the Midwest. The monitoring trailer also hosts an acid deposition collector for sulfate (SO₄), nitrate (NO₃), and other ions, EPA PM_{2.5} Chemical Speciation Network site, National Air Toxics Trends Station (NATTS), and multi-pollutant NCore site. Various criteria pollutants, including sulfur dioxide (SO₂), carbon monoxide (CO), ozone (O₃), and fine particulate matter (PM_{2.5}), as well as standard meteorological parameters also monitored at this site.

The Bronx monitoring site is located in a metropolitan area of >19 million people. The region has a long manufacturing, petrochemical and industrial legacy that includes contamination from Hg and other toxic compounds. The Tekran and MDN collector are located on the roof of the Pfizer Plant Research Laboratory at the New York Botanical Gardens. The site also monitors acid deposition, various criteria pollutants (SO₂, CO, O₃, nitrogen oxides (NO_x), and PM_{2.5}), Photochemical Assessment Monitoring Station (PAMS) non-methane hydrocarbons (NMHC), various air toxics, PM_{2.5} speciation, and standard meteorological parameters.

Table 1. Mercury monitoring site information.

Site	EPA AQS ID	NYS ID	Metropolitan area population, millions	Latitude north	Longitude west
Rochester	36-055-1007	2701-22	1.03	43° 08' 36"	-77° 32' 52"
Bronx	36-005-0133	7094-10	>19	40° 52' 05"	-73° 52' 42"

Figure 1. Aerial views of the Rochester (top panel) and Bronx (bottom panel) monitoring sites, indicated with red arrows.

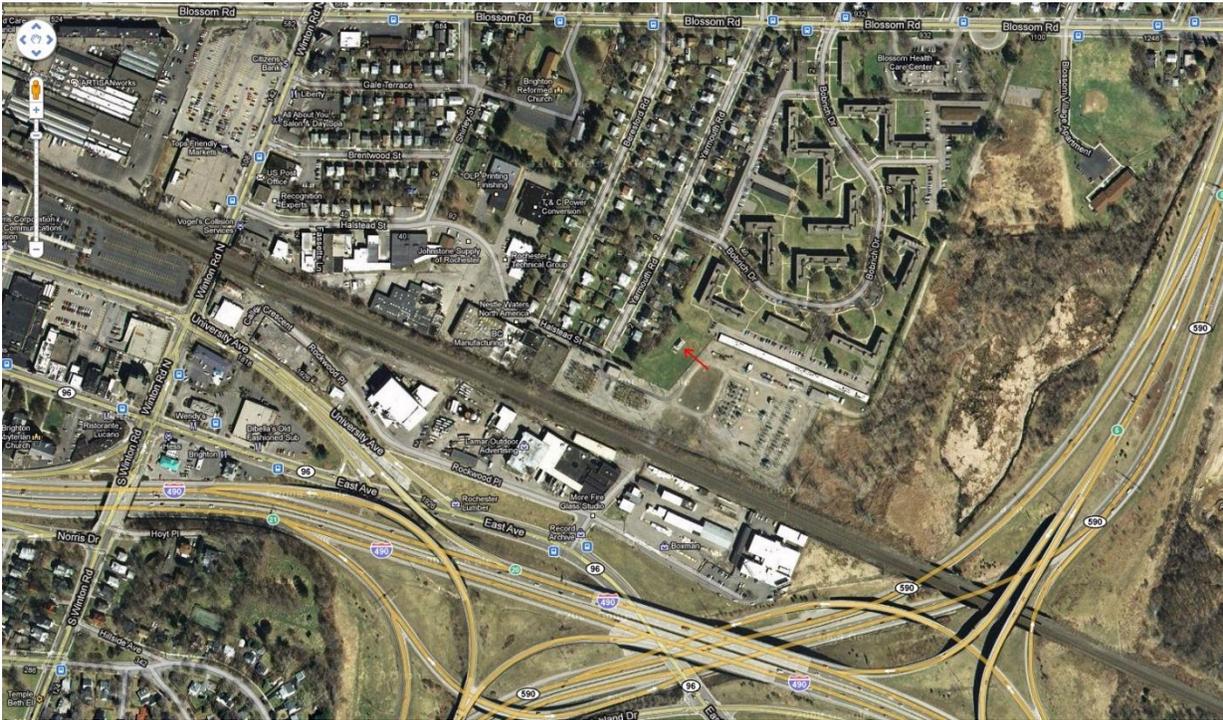


Figure 2. The mercury instrumentation at the Bronx monitoring site. The top panels show the MDN wet deposition collector (N-CON 00-125-2 automatic precipitation sampler, top left) and ETI NOAH IV total precipitation gauge (top right). The Tekran Model 2537B analyzer and Model 1130 pump box are on the bottom left, and the RGM (Model 1130) and PBM (Model 1135) units are on the bottom right.



5. Implementation of Program

The Tekran measurements were initiated in September 2008 at Rochester, and August 2008 at the Bronx site. Supplemental funding from the GLC will allow the NYSDEC to continue collecting data through at least March 2012. The Tekran instrumentation was chosen because it has a high time resolution and suitable detection limits for ambient measurements of PBM, RGM, and Hg(0). The high frequency species data from this instrument provide insight into the conversion from one form to another during the course of a day (Poissant et al., 2005). The majority of ambient mercury is present in elemental form, but speciation data are important due to the greater deposition rates and localized impacts. Reactive mercury has much higher wet and dry deposition rates than does elemental. Speciation is of particular interest close to industrial sources such as waste incinerators and fuel-burning equipment, which may discharge the majority of their mercury emissions in ionic form, particularly mercuric chloride. The Rochester site is downwind of a majority of New York's coal-fired utilities, while the Bronx site is situated near numerous commercial and industrial facilities in the metropolitan region.

The NYSDEC has been operating the wet deposition monitoring equipment and a Universal Recording Rain Gauge at both sites since January 2008. Due to the proximity of potential localized sources of mercury emissions, it is possible that these wet deposition monitors will collect more mercury than other sites in the mostly rural MDN network. The data from this project's MDN-approved sampler will be used for comparisons with the existing rural MDN data set as well as to more completely evaluate the mercury burden in the two populated urban areas. The weekly samples from the MDN collector are picked-up by NYSDEC staff and sent to the MDN contract lab (Frontier Geosciences) for total mercury analysis.

The data from the Tekran monitor gives the grantees the ability to determine the ratio of elemental mercury to reactive mercury in the ambient air. Data procured from the Information Collection Request (ICR) showed that the majority of mercury emitted from coal fired utilities, located in the eastern half of the United States emit speciated mercury compounds in the following percentages, 31%, 66%, and 3% for Hg(0), RGM, and PBM, respectively (DeSantis, 2002). Coal-fired utilities in the states surrounding New York burn predominantly eastern bituminous coal with high chlorine content creating the ratio of in-stack gases listed above.

5.1 Tekran Method Development and Improvement

The original schedule for this project did not include the time and effort required to make the Tekran Models 2537B, 1130 and 1135 reliable and ready for operation in distant field offices. The development work was done in-house prior to field deployment. This developmental work was much more extensive and time consuming than expected and an earlier field deployment would have resulted in poor data capture and potentially significant damage to the analyzers.

Some of the developmental work included familiarization with the instrumentation and troubleshooting, collaborative work on the AMNet SOP, upgrades to the Model 2537B lamp driver board, standardization of glassware preparation techniques, development of the NYSDEC Tekran Field Data Form and work with the manufacturer on a zero air drying system. This

method development work was necessary so that the Tekran could operate reliably unattended between the bi-weekly service visits.

6. Data Analysis

6.1 Wet Deposition

All quality-assured wet deposition data through September 2010 are available on the NADP MDN website (<http://nadp.sws.uiuc.edu/MDN/mdndata.aspx>). The following sections describe the analysis performed on these wet deposition data.

6.1.1 Summary Statistics

The precipitation and Hg wet deposition measurements at Rochester and Bronx were initiated in January 2008, and these data are presented here through September 2010. Figure 3 displays the distributions of weekly (a) precipitation amount (mm), (b) Hg concentration (ng/L), and (c) Hg wet deposition (ng/m²). The asterisks denote the corresponding median values, the boxes denote the 25th and 75th percentiles, and the whiskers denote the 5th and 95th percentiles. The numbers of weekly samples are also included in Figure 3. In addition to the two urban locations, Figure 3 includes the same summary statistics at the three more rural MDN sites in New York – Huntington Wildlife (Adirondack Mountains), Biscuit Brook (Catskill Mountains), and West Point (Hudson River Valley). It should be noted that the West Point monitor was closed in October 2010.

Precipitation is typically higher at the Biscuit Brook site compared with the other MDN sites in New York, and typically lowest at Rochester. The low precipitation at Rochester may be due to the moderating effects of Lake Ontario. The median precipitation amounts at Bronx, West Point, and Huntington Wildlife are very similar. In contrast, Hg concentrations are typically higher at the two urban monitors (median values ~7-8 ng/L) than the three rural monitors (~5-6 ng/L). This is not surprising due to Rochester's relative proximity to coal-fired utilities, and the Bronx site potentially affected by numerous industrial, chemical, and legacy sources of mercury. Over this 2.5-year period, the median Hg concentration at Rochester (8.38 ng/L) was higher than that at the Bronx site (7.29 ng/L).

Wet Hg deposition, a function of both Hg concentration and precipitation, was often higher at Bronx and Biscuit Brook compared to the other three sites. The median weekly wet deposition amount was about 150 ng/m² at these two sites, compared to only about 90-100 ng/m² at the other three MDN sites in New York.

6.1.2 Seasonal Variations

Figure 4 displays quarterly composite values through the third quarter of 2010: (a) total precipitation (cm), (b) average Hg concentration (ng/L), and (c) total wet Hg deposition (µg/m²). As noted previously, of the five MDN sites in New York precipitation tends to be lowest at Rochester. There is also very little seasonal variation in precipitation at Rochester, likely due to the moderating effects of Lake Ontario.

The seasonal pattern in Hg concentration is qualitatively similar at all five MDN sites, generally higher in quarters two (April-June) and three (July-September) than during the colder seasons. In 2008, the average Hg concentrations in quarters two and three were considerably higher at Rochester than the other four sites; currently, the range in concentration from the lowest value (Huntington) to the highest value (Rochester) is not as large. Although there is some indication that Hg concentrations may be decreasing statewide, without prior deposition data at Rochester, it is not known whether 2008 was an anomalous year. The only MDN site in New York with 10 or more years of data is Huntington Wildlife, and seasonal Kendall trend analysis suggests that Hg concentrations are decreasing by about 3% per year. The seasonal patterns in wet Hg deposition are less clear. This seems to be due largely to year-to-year variations in precipitation, in addition to changes in ambient Hg concentrations.

6.1.3 Comparison with MDN Data in the Surrounding Region

Figure 5 displays the 2009 annual average Hg concentration (top panel), and accumulated wet Hg deposition (bottom panel) at sites in New York and the surrounding states and Ontario. In terms of Hg concentrations, the highest levels generally occurred to the south and west, likely due to proximity to emission sources. Across this multi-state region, the annual average concentration varied by a factor of about 1.6. The annual average Hg concentrations at the Rochester and Bronx sites in 2009 were about 6.6 ng/L.

As noted earlier, the spatial pattern in wet deposition is driven by variations in ambient concentrations and precipitation amounts. Thus, the pattern is less distinct than for concentrations. In this year, the highest wet deposition amounts ($>8 \mu\text{g}/\text{m}^2$) were recorded at the Biscuit Brook and Bronx sites, with slightly lower totals at sites in Pennsylvania.

It is also worth examining Hg concentrations and wet deposition across the nation to put the New York data in perspective. Figure 6 displays the Hg concentration (top panel) and wet deposition (bottom panel) for 2009 (NADP, 2010). In 2009, the highest Hg concentrations occurred in the more arid western states; MDN sites in Arizona, Colorado, and Nevada had average Hg concentrations that exceeded 30 ng/L. High values were also recorded in Kansas, Nebraska, and southern Florida. The highest wet Hg deposition amounts tended to occur in the Gulf Coast states, due to large emissions sources and high rainfall. The highest deposition amounts in this region were roughly 2-3 times larger than those in Rochester and Bronx. High deposition amounts also occurred in the Ohio River Valley, as well as eastern Nebraska and Oklahoma.

6.1.4 Sulfate Versus Mercury in Wet Deposition

Coal combustion is a source of both SO_4 and Hg, and both the Bronx and Rochester sites are collocated with wet deposition samplers operated by the NYSDEC. This allows for a comparison of SO_4 and Hg concentrations in precipitation on a weekly basis. Figure 7 displays the weekly average concentrations of SO_4 and Hg at the Bronx (magenta squares) and Rochester (blue squares). Although there is some scatter in the data, and it is difficult to examine the data on a single event basis, there is a general degree of correlation between these two pollutants at both sites.

Figure 3. Distributions in weekly (a) precipitation, (b) Hg concentration, and (c) wet Hg deposition at the five MDN sites in New York, January 2008 – September 2010. Asterisks denote medians, boxes denote 25th and 75th percentiles, and whiskers denote 5th and 95th percentiles.

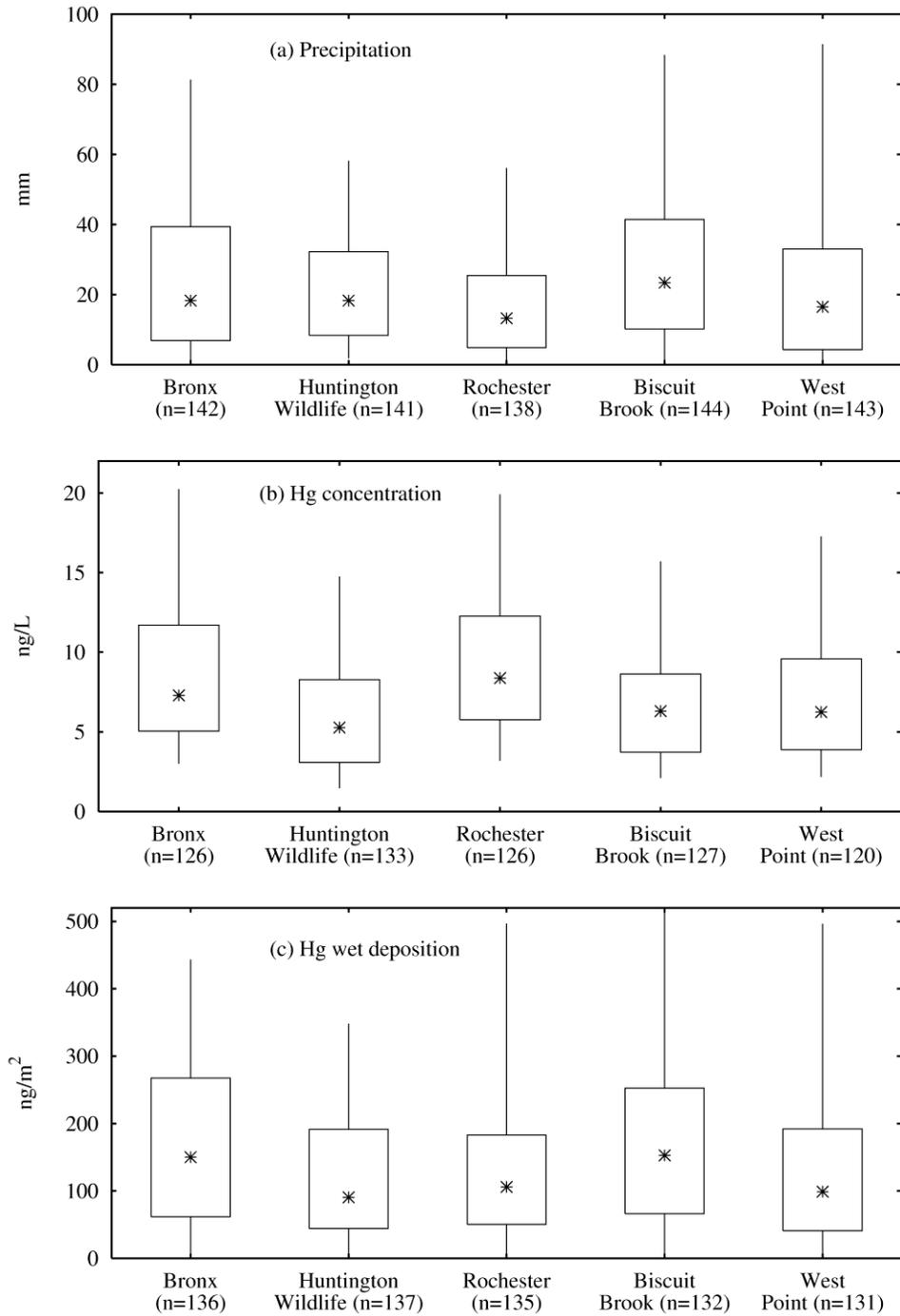


Figure 4. Quarterly (a) total precipitation, (b) average Hg concentration, and (c) total wet Hg deposition at the five MDN sites in New York, January 2008 – September 2010.

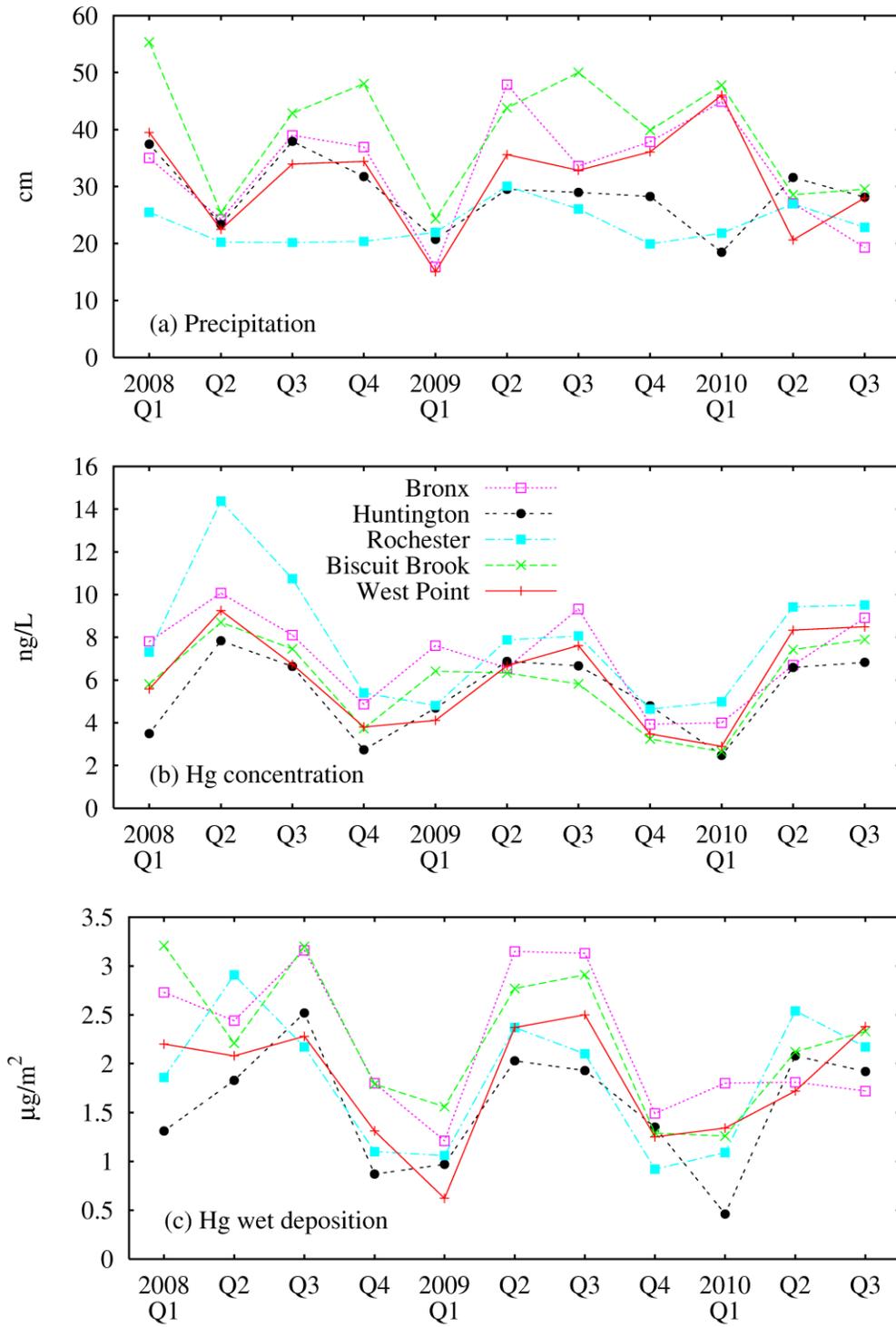


Figure 5. The 2009 average Hg concentration (ng/L; top panel) and total wet Hg deposition ($\mu\text{g}/\text{m}^2$; bottom panel) in New York and the surrounding region.

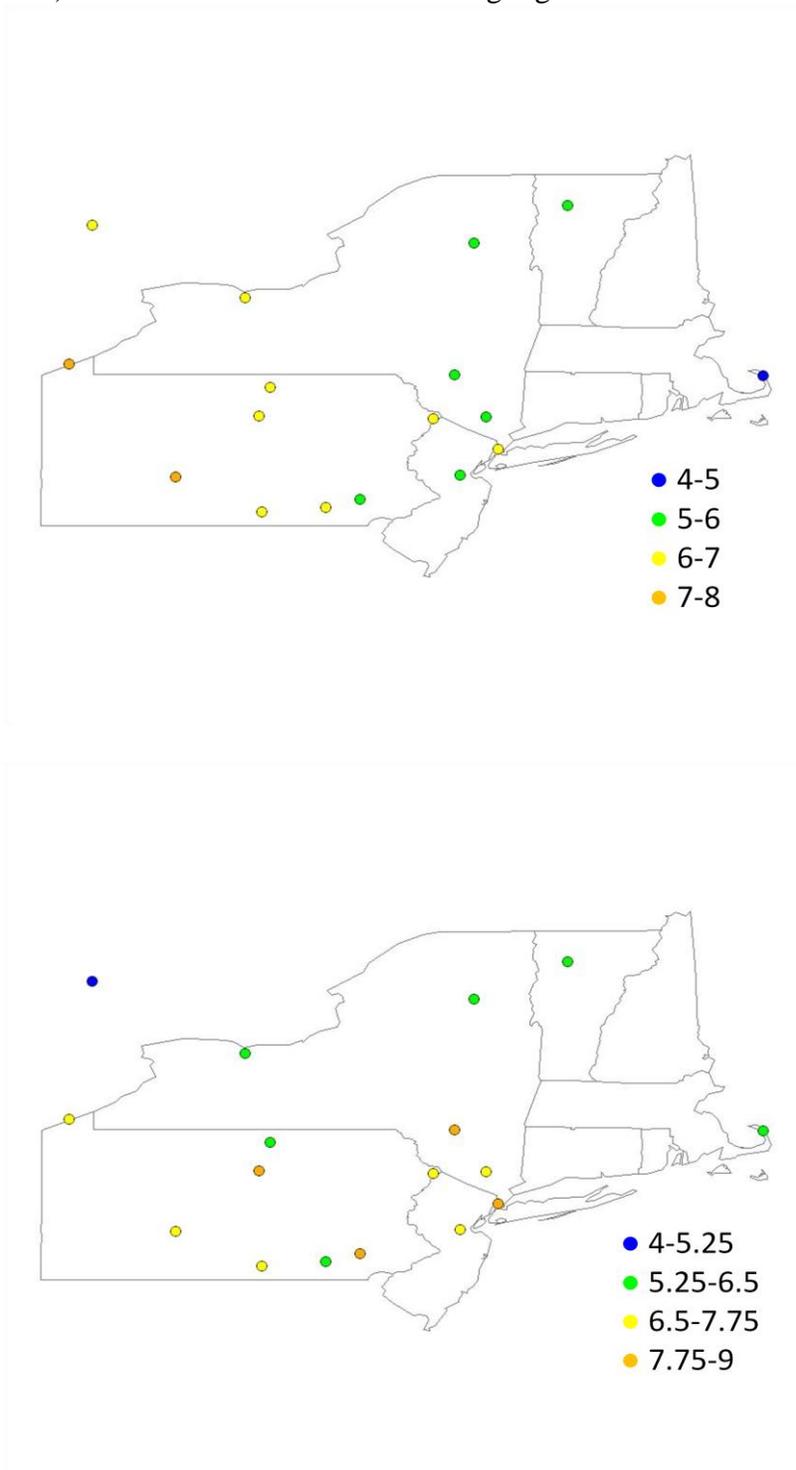
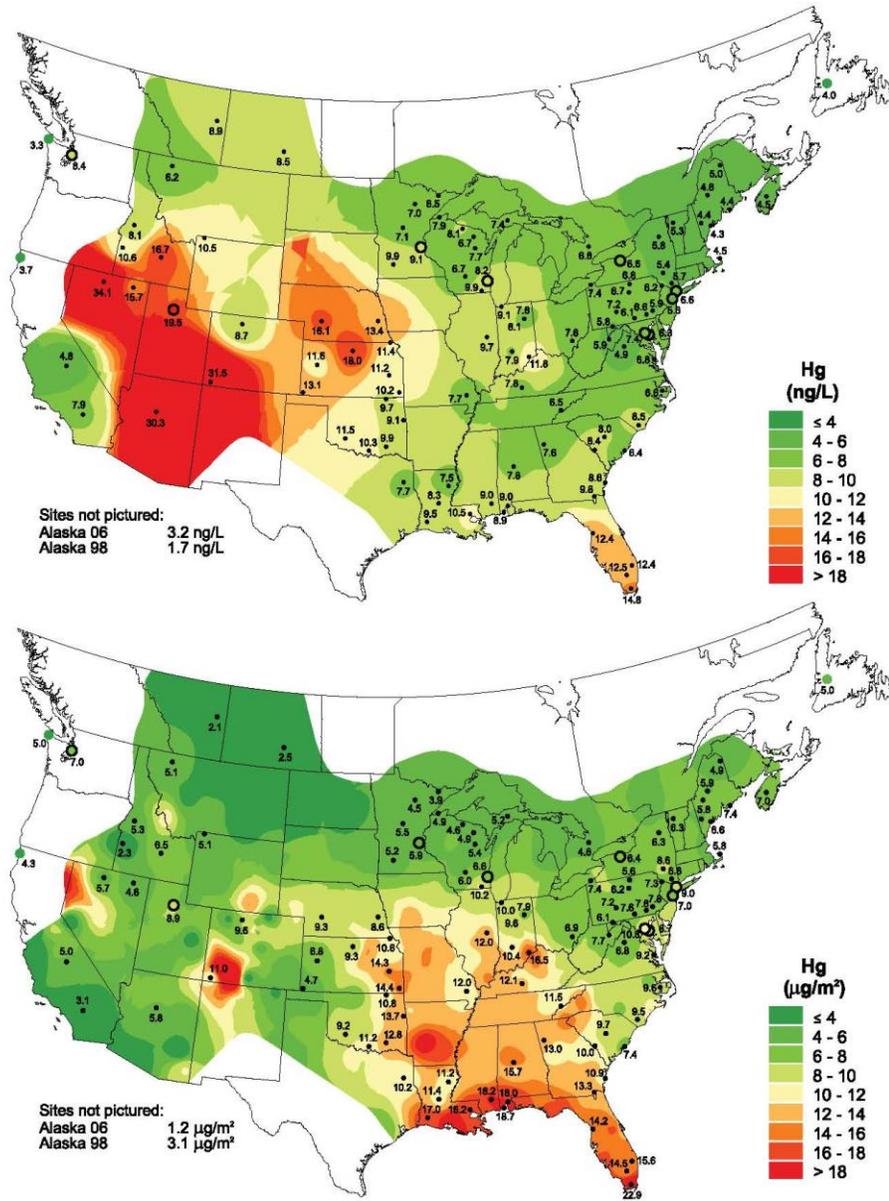
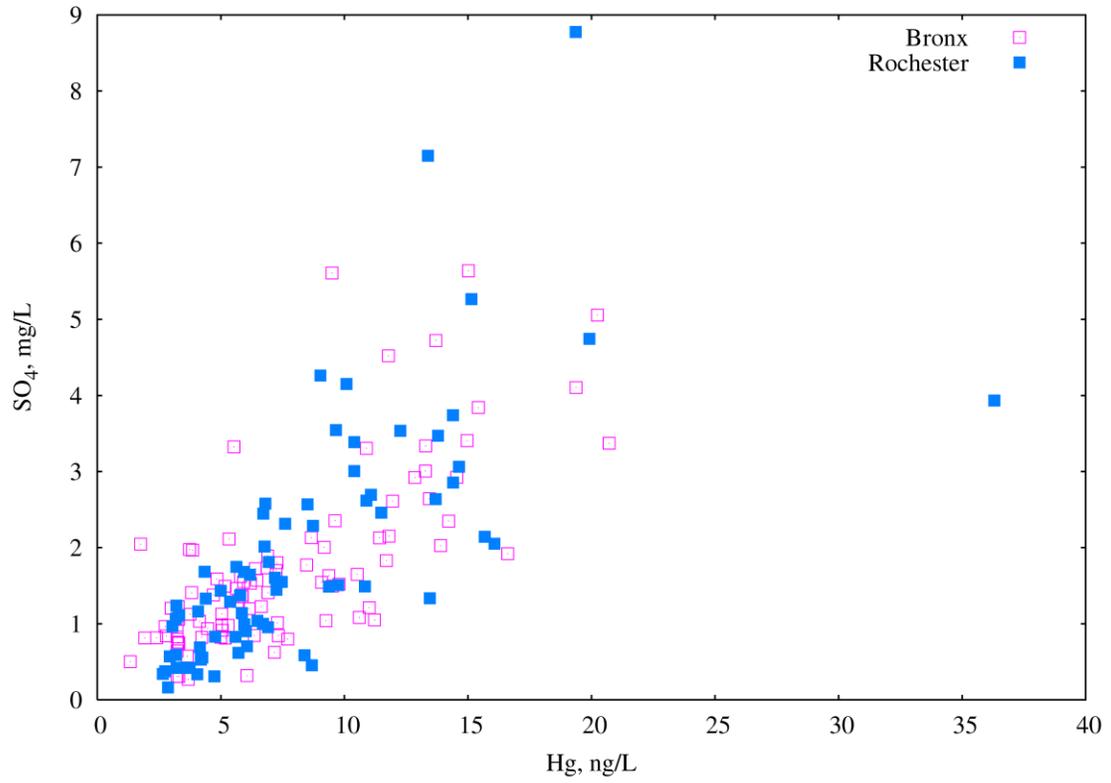


Figure 6. Average Hg concentration (top panel) and wet deposition (bottom panel) at MDN sites in 2009, from NADP (2010).



Total mercury concentration (top) and wet deposition (bottom), 2009.

Figure 7. A comparison of weekly average SO_4 and Hg at the Bronx (magenta squares) and Rochester (blue squares) sites.



6.2 Ambient Air Concentrations

All quality-assured Tekran data through 2010 are available on the password-protected NADP AMNet data site (<http://nadp.sws.uiuc.edu/amn/data.aspx>). The following sections describe the analysis performed on these ambient Hg data.

6.2.1 Tekran Data Capture

Table 2 lists the approximate percentages of valid data through September 2010. Data collection began at the Bronx site on August 28, 2008, and at the Rochester site on September 27, 2008. The data capture was higher at the Rochester site than the Bronx site. In addition to short down periods due to bi-weekly maintenance and calibrations, there were numerous other down periods over the past two years at each site. Bad pumps, system leaks, site problems such as electrical surges, computer/data acquisition failures, and the Tekrans not cycling properly were experienced at both sites. Both systems required substantial “shakedown” during the fall of 2008. The Rochester Tekran was also down for maintenance from late August through September 2010, while the Bronx monitor was down during significant stretches between February and May 2010. Because the Bronx monitor was not particularly stable during the early part of the monitoring campaign, it was decided in early 2009 to set the Tekran to zero and span every day, as opposed to once every three days at Rochester.

Table 2. Approximate Tekran valid data capture at the monitoring sites.

Site	PBM	RGM	Hg(0)
Rochester	76.7%	76.7%	81.4%
Bronx	64.5%	64.2%	69.7%

6.2.2 Summary Statistics

Over this two-year period, the median and range in ambient Hg concentrations at the Rochester and Bronx sites were similar. The median (and 5th and 95th percentiles) concentrations of Hg(0), RGM, and PBM at the Bronx site were 1.42 ng/m³ (1.09-2.14 ng/m³), 6.2 pg/m³ (0.8-42.9 pg/m³), and 10.3 pg/m³ (1.5-42.9 pg/m³), respectively. The corresponding values at the Rochester site were 1.41 ng/m³ (1.15-1.74 ng/m³), 6.6 pg/m³ (0.8-38.5 pg/m³), and 13.7 pg/m³ (3.3-36.2 pg/m³) for Hg(0), RGM, and PBM. While the median values of Hg(0), RGM, and PBM are similar at these two sites, the variability in the concentrations is larger at the Bronx site. This may due to more diverse sources in the vicinity of the Bronx site, or possibly a more stable PBL around Rochester as a result of proximity to Lake Ontario.

The concentrations of speciated Hg at Rochester were similar to those reported by Huang et al. (2010) with a collocated Tekran system in 2008-2009. The New Jersey Department of Environmental Protection (NJDEP) reported 2.5 years of Tekran data from two urban sites, Elizabeth and New Brunswick. Although the average Hg(0) concentrations in NJ were higher (2.25 ng/m³ at Elizabeth, 2.15 ng/m³ at New Brunswick) than the Bronx, the average RGM and PBM concentrations were very similar to those from the Bronx site, in the 6-10 pg/m³ range (Aucott et al., 2009). Choi et al. (2008) reported Tekran data from the Huntington Wildlife site

from June 2006 through May 2007. While the average Hg(0) concentration at the Huntington site were very similar to those at the two urban sites (1.4 ng/m^3), the average RGM and PBM concentrations were approximately a factor of three lower at the rural site compared with the Bronx and Rochester sites.

While Hg(0) generally varies between about $1\text{-}2 \text{ ng/m}^3$, RGM and PBM can vary from near the instrument detection limit to $\sim 50\text{-}100 \text{ pg/m}^3$ or more. There is also considerable seasonal variation in the RGM and PBM data. For this reason, Figure 8 displays the distributions of each Hg species, by season, at the two urban sites. Seasons are defined as **December-February (winter)**, **March-May (spring)**, **June-August (summer)**, and September-November (fall). The median Hg(0) concentrations are similar at the two sites, although the spread in the data is larger at the Bronx site. The median Hg(0) concentrations at the Bronx are slightly higher during the spring and summer, whereas the median Hg(0) concentrations at Rochester are slightly elevated during the winter and spring.

There is a distinct seasonal variation in RGM at the two sites. The highest RGM concentrations and highest degree of variability tend to occur in the spring months, especially April. The highest PBM concentrations tend to occur in the winter months at these sites, likely a result of a low planetary boundary layer and wood combustion for space heating (e.g. Choi et al., 2008). Both RGM and PBM tend to exhibit occasional episodes, and this will be discussed briefly in Section 6.2.7.

6.2.3 Monthly Variations

Figure 9 displays the monthly average and standard deviations in each Hg species. The corresponding averages and standard deviations from the rural Huntington Wildlife AMNet site are also included for comparison. In terms of Hg(0) all three sites generally tracked each other over these two years on an average basis, although the variability in concentrations was generally higher at the urban sites. During the winter, the monthly average Hg(0) concentrations at the three sites are very close to each other; the concentrations at the two urban sites tend to be approximately 20-50% higher than those at Huntington Wildlife during the summer months.

In contrast to Hg(0), the concentrations of RGM and PBM at Huntington Wildlife are much lower than the two urban sites, often being near the detection limit of the Tekran system. While there is distinct seasonal variation in RGM (spring peak) and PBM (winter peak) at the urban sites, the seasonal variation in the more reactive Hg species is far more muted at the rural site.

6.2.4 Diurnal Variations

Figure 10 displays the average diurnal variations in each Hg species at the two sites, on a seasonal basis with the same color-coding as Figure 8. The left side panels (10a, 10c, 10e) show the diurnal variations at the Bronx site, and the right side panels (10b, 10d, 10f) show the Rochester diurnals. The top, middle, and bottom rows denote Hg(0), RGM, and PBM, respectively.

In terms of Hg(0), there is a substantial seasonal variation in the diurnal patterns at the Bronx site. During the summer months, Hg(0) has a maximum at nighttime and a broad mid-day minimum. High nighttime values are associated with the shallow nocturnal boundary layer, and lower daytime values are associated with atmospheric mixing and oxidation reactions. At Rochester, the diurnal pattern is qualitatively similar during the spring, summer, and fall. At both sites, there is little diurnal variation during the winter months.

The diurnal pattern for RGM suggests a photochemical source. At the Bronx site, this is clearly evident during the spring, summer, and fall months, and consistent with the Hg(0) variation; during the winter, the RGM values are generally low over the entire day. At Rochester, the amplitude of the diurnal variation is less than at the Bronx, and is most distinct during the spring and fall months. In contrast to the Bronx site, at Rochester the RGM levels on average are lowest during the summer months, even though the diurnal pattern shows slightly higher levels during the afternoon hours.

There is relatively little variation in the diurnal patterns for PBM. At Rochester, there is almost no temporal variation in a given season; average concentrations are simply higher during the winter and spring months. At the Bronx site, there is some indication of photochemical production during the warmer months, but the relative diurnal amplitudes of PBM are smaller than for RGM.

6.2.5 Wind and Pollution Rose Analysis

Surface wind analysis can be used to investigate potential regional sources of Hg. Figure 11 displays the wind and pollution roses at the Bronx monitoring site, by season. The wind rose (Figure 11a) shows the percent of hours the wind blows from a particular sector, and the average concentrations of Hg(0), RGM, and PBM as a function of surface wind direction are displayed in Figures 11b-d, respectively. Because the wind data at the monitoring site were only sporadically available over the two years, hourly surface winds were obtained from LaGuardia International Airport (~4 miles to the south). In this region, surface winds are complex because of the land-sea circulations, urban heat island effects, and regional climatology. While the surface winds are complex and vary seasonally, there is a clear pattern in the ambient Hg concentrations at this site. Each Hg species exhibits a maximum concentration when surface winds are from the southwest. This is not surprising, owing to the large number of current and legacy sources of Hg in this region, including steel manufacturing, coal-fired utilities, crematoria, and incinerators (e.g. Aucott et al., 2009; Gratz et al., 2009).

6.2.6 Winter Stagnation Event

The February 4-10, 2009 period illustrates the use of speciated mercury and ancillary air quality and meteorological data to characterize a wintertime stagnation event at the Bronx site. Figure 12 consists of four panels: (a) air temperature, hourly PM_{2.5}, and estimated PBL height; (b) wind speed and direction; (c) NO_x, total non-methane hydrocarbons (NMHC), and SO₂; and (d) Hg(0), RGM, and PBM. All of these air quality parameters were collected at the same location, while the continuous meteorological data were collected at a tower ~300 m away. The PBL heights were based on estimates from archived meteorological fields in New York City from the NOAA

ARL READY website (<http://ready.arl.noaa.gov/READYamet.us.php>). On February 4 and 5, pollutant concentrations and daytime temperatures were generally low, wind speeds were moderate with winds out of the west/southwest, and maximum daytime PBL heights were high.

On the morning of February 6, high pressure settled into the region which slowly moved off the eastern seaboard on February 7. A strong low-level inversion formed overnight from February 6 to 7, and persisted throughout the day; the PBL heights were extremely low on February 7 and part of February 8. Temperatures increased over this period, and the winds became calmer and originated from the east/southeast. These meteorological conditions were conducive to trapping local and regional emissions, and the highest concentrations of PM_{2.5}, NO_x, NMHC, and SO₂ occurred on February 7.

Interestingly, the highest PBM concentrations (>150 pg/m³) occurred at this site on February 6, a day earlier than the other pollutants, although values exceeding 120 pg/m³ were also observed on February 7. In contrast, PBM concentrations were generally about 20-40 pg/m³ during the rest of this time period. The RGM levels during this time of year are generally low, but were somewhat elevated compared to the concentrations on prior and subsequent days. The time series of Hg(0) very closely tracked PM_{2.5}, NO_x, NMHC, and SO₂, with peak Hg(0) concentrations on February 7 approximately twice as high as those on previous days.

6.2.7 Hg Variations With Co-Pollutants: Two Examples

The diurnal variation in RGM, with a clear peak during the afternoon hours especially during the warmer months, suggests that photochemistry plays a key role in the formation of RGM. Figure 13 displays the time series of RGM (green lines) with hourly O₃ (orange lines) concentrations during a ten-day period, August 12-21, 2009, at the Bronx site. Both RGM and O₃ were low on August 12 and 13, and both pollutants began to rise on subsequent days. During the middle of this period, peak hourly O₃ concentrations were about 80-90 ppb, and on multiple days the diurnal patterns of O₃ and RGM concentrations were well-correlated.

The seasonal variation in PBM shows a maximum during the colder months, when PBL heights are generally low, photochemistry is low, and space heating and other sources are prevalent. The same is true for other pollutants such as SO₂. Figure 14 displays the time series of PBM (magenta line) and hourly SO₂ (blue line) concentrations during a two-week period, January 7-20, 2010, at the Bronx site. PBM and SO₂ both exhibit noticeable peak concentrations on January 14-15, and are generally correlated on subsequent days as well. Both of these relationships – RGM vs. O₃ and PBM vs. SO₂ – indicate that co-pollutants can provide insight into sources and fates of speciated Hg in the region, and that further analysis is warranted.

Figure 8. Distributions of three-hour average (a) Hg(0) (ng/m^3), (b) RGM (pg/m^3), and (c) PBM (pg/m^3). Distributions are shown by season (DJF, MAM, JJA, and SON); the left “box” denotes the Bronx site, the right “box” denotes the Rochester site. Dots denote medians, boxes denote 25th and 75th percentiles, and whiskers denote 5th and 95th percentiles.

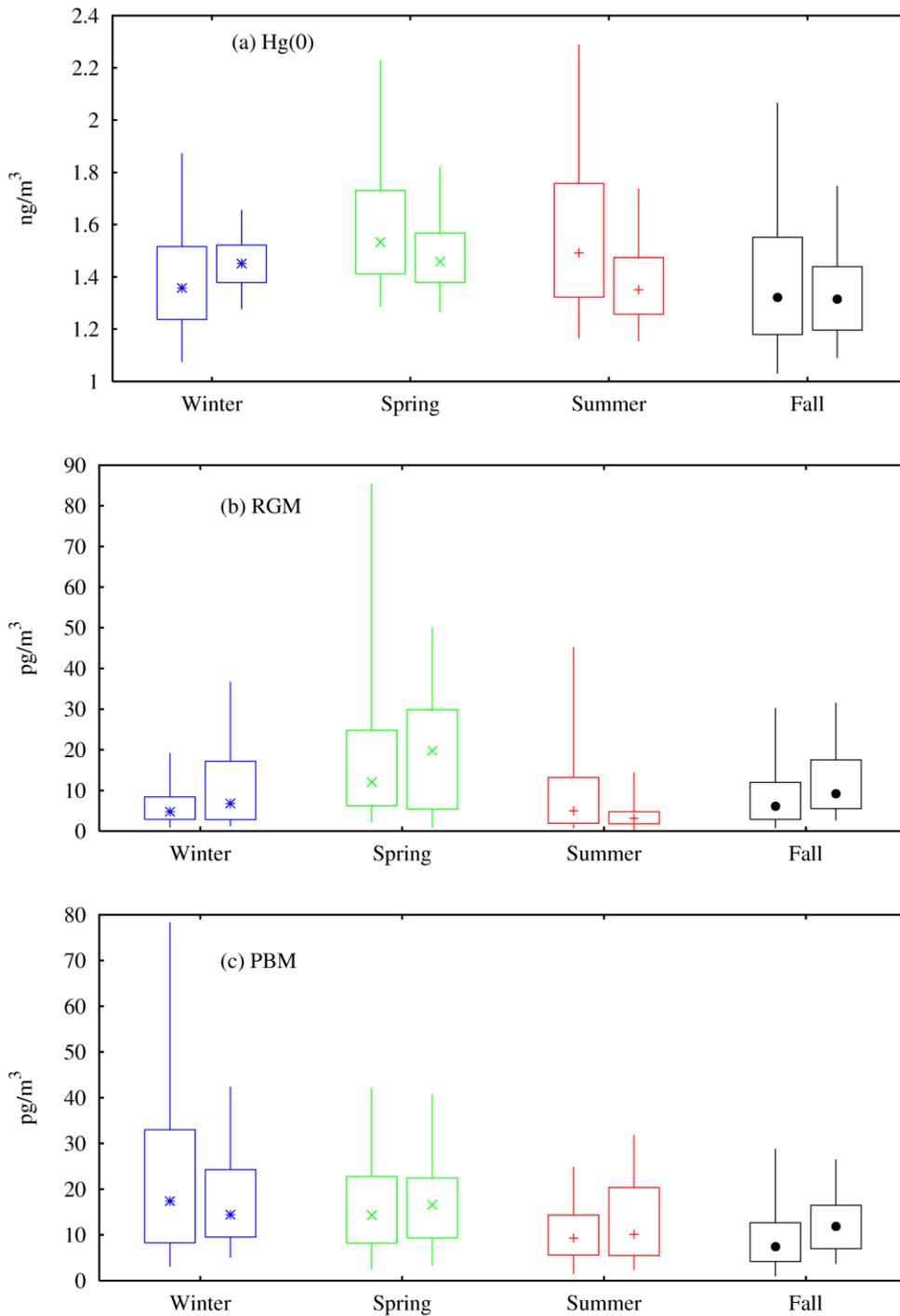


Figure 9. Monthly average and standard deviations in (a) Hg(0) (ng/m^3), (b) RGM (pg/m^3), and (c) PBM (pg/m^3) at the two urban sites and the rural Huntington Wildlife site, August 2008 – September 2010.

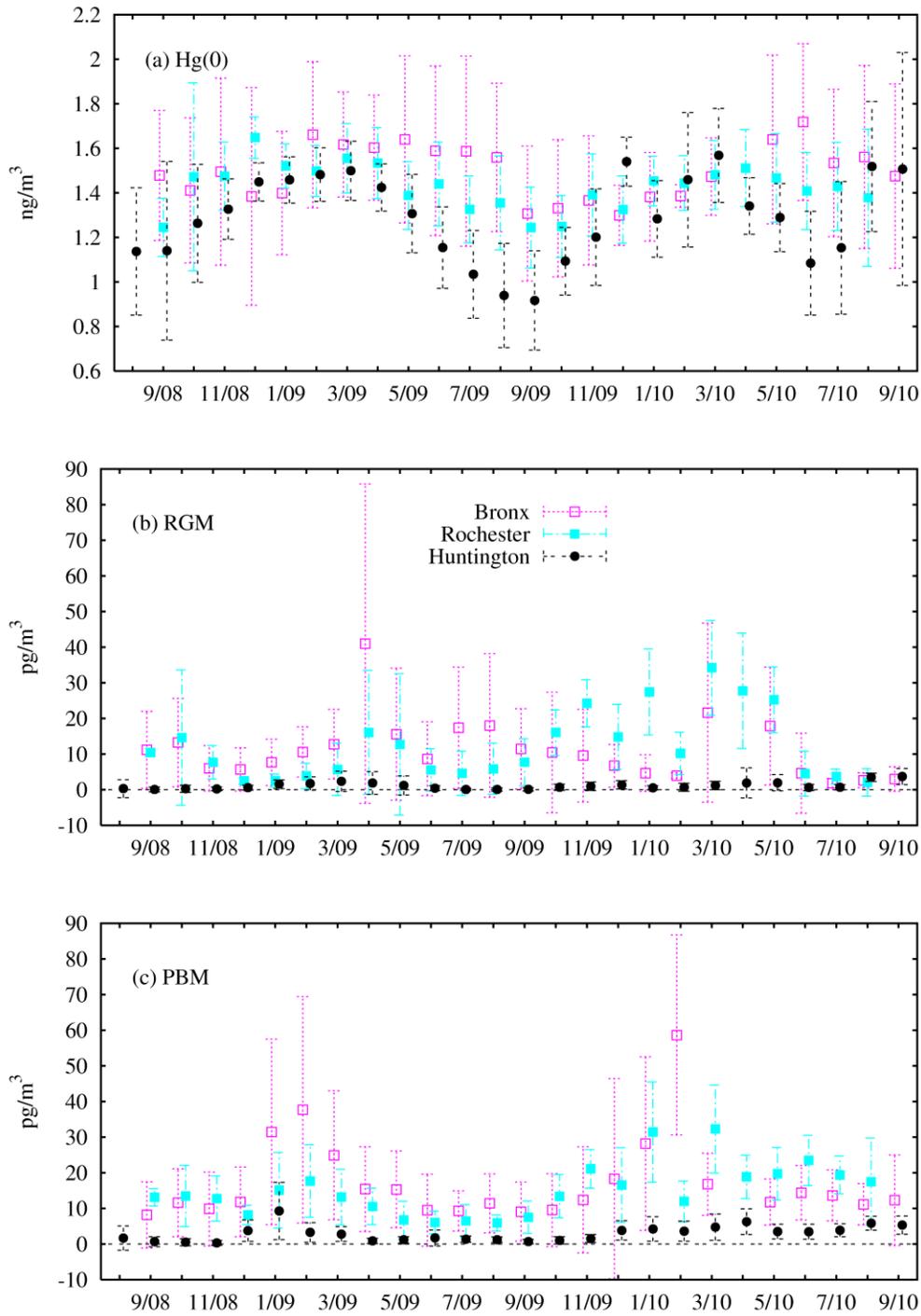


Figure 10. Seasonal (DJF, MAM, JJA, and SON) diurnal variations in Hg(0), RGM, and PBM at the Rochester and Bronx sites.

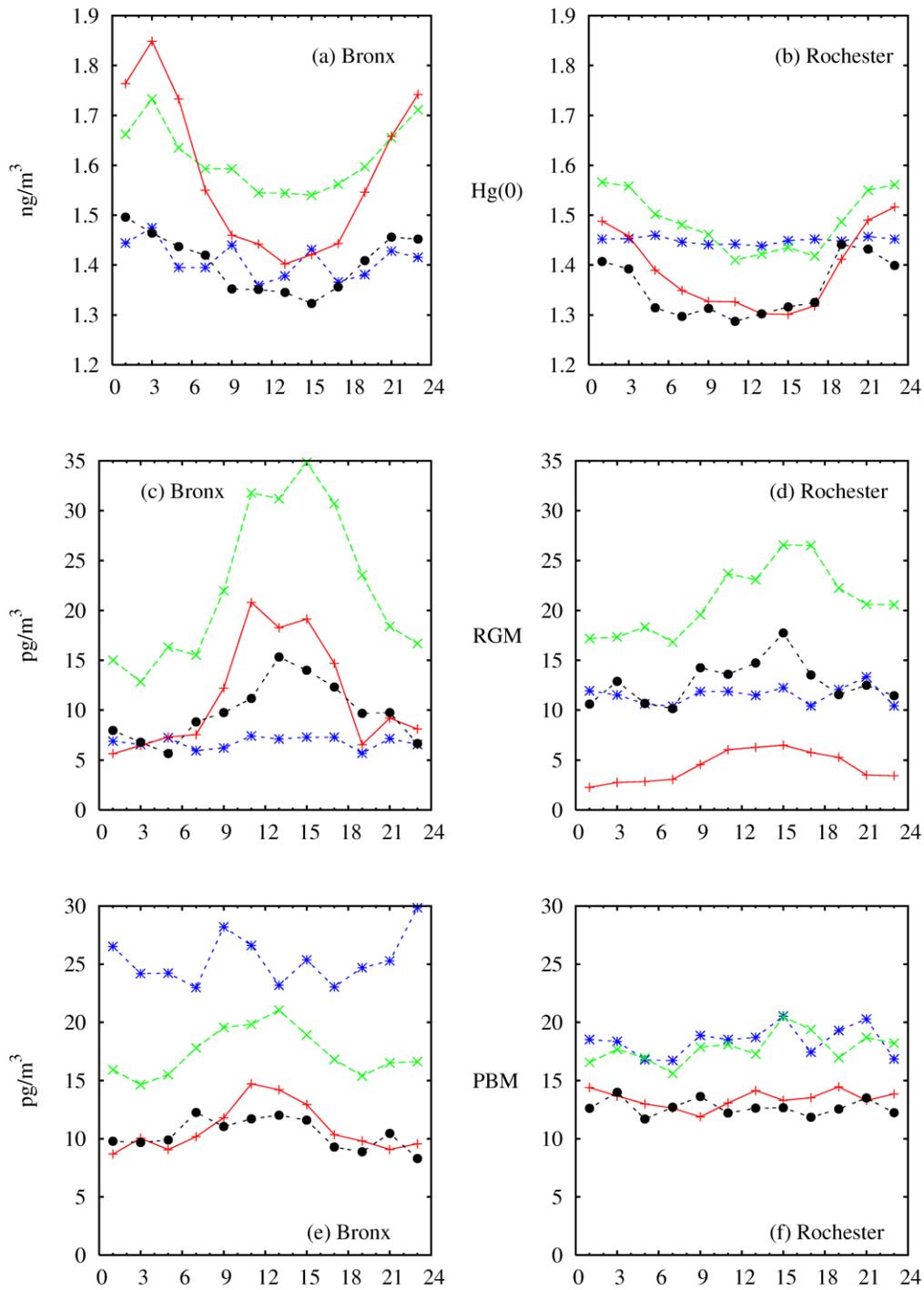


Figure 11. Wind and Hg pollution roses at the Bronx site, by season (DJF, MAM, JJA, and SON): (a) wind rose, (b) Hg(0), (c) RGM, and (d) PBM.

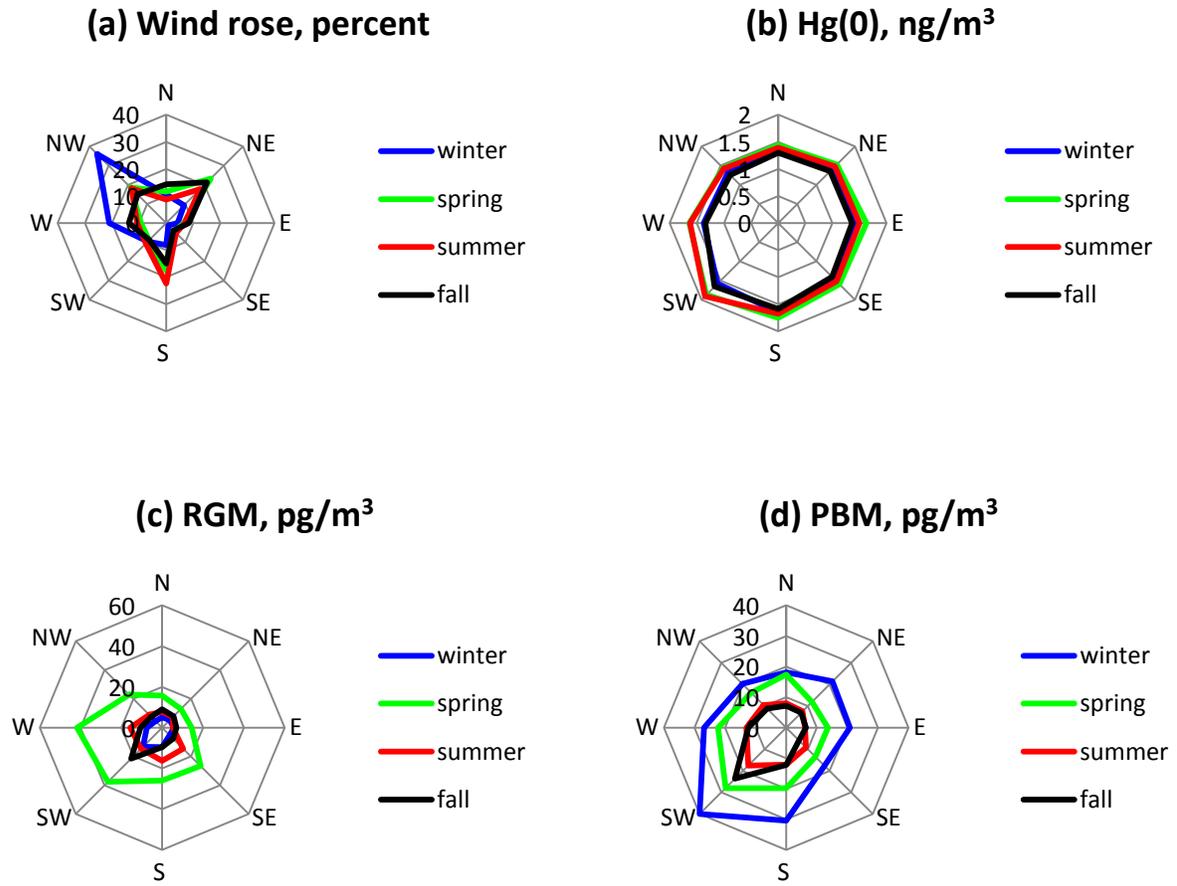


Figure 12. (a) Air temperature, hourly $PM_{2.5}$, and estimated PBL height; (b) wind speed and direction; (c) NO_x , total non-methane hydrocarbons (NMHC), and SO_2 ; and (d) $Hg(0)$, RGM, and PBM.

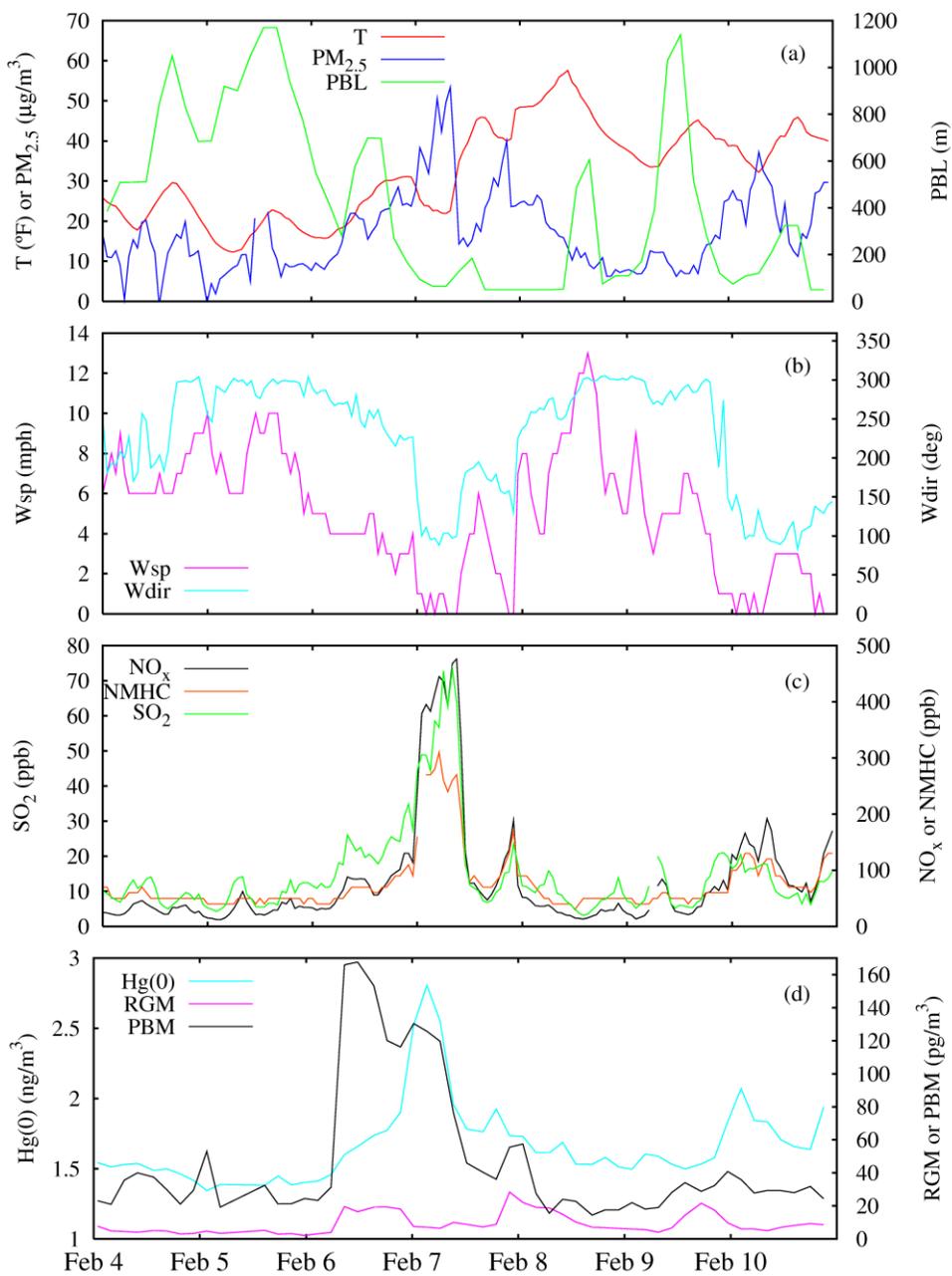


Figure 13. Time series of RGM (green line) and O₃ (orange line) at the Bronx site, August 12-21, 2009.

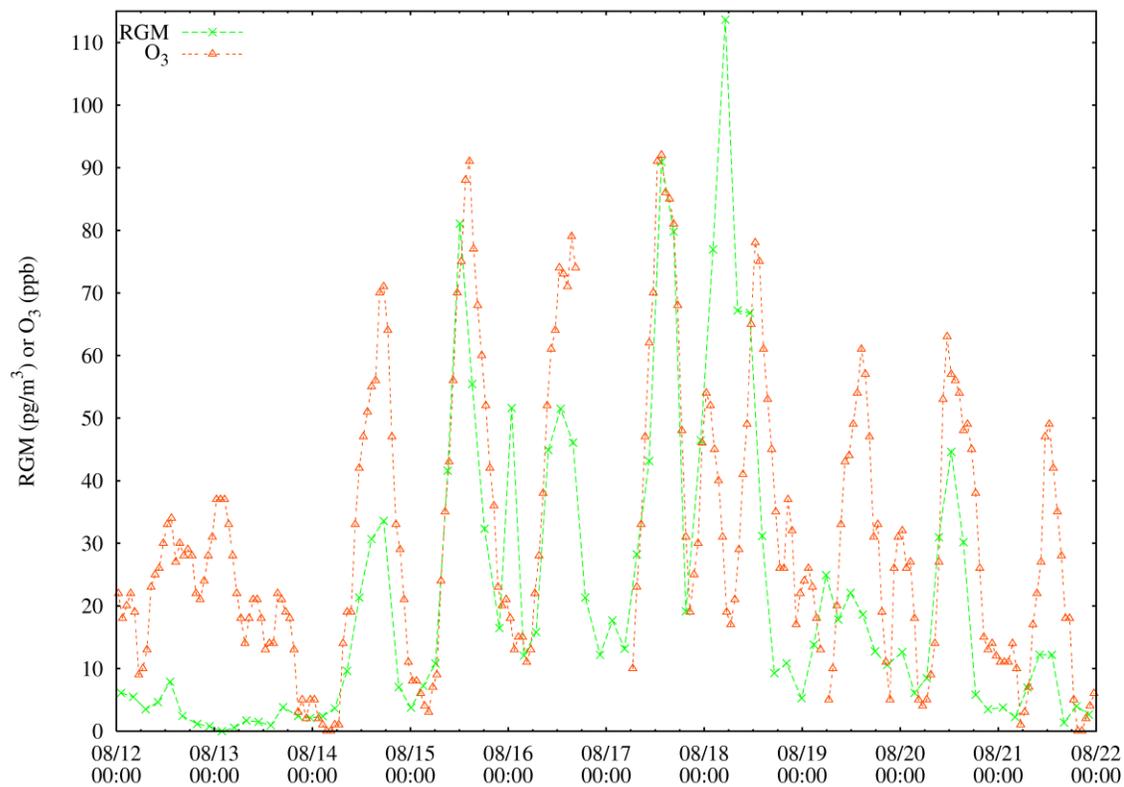
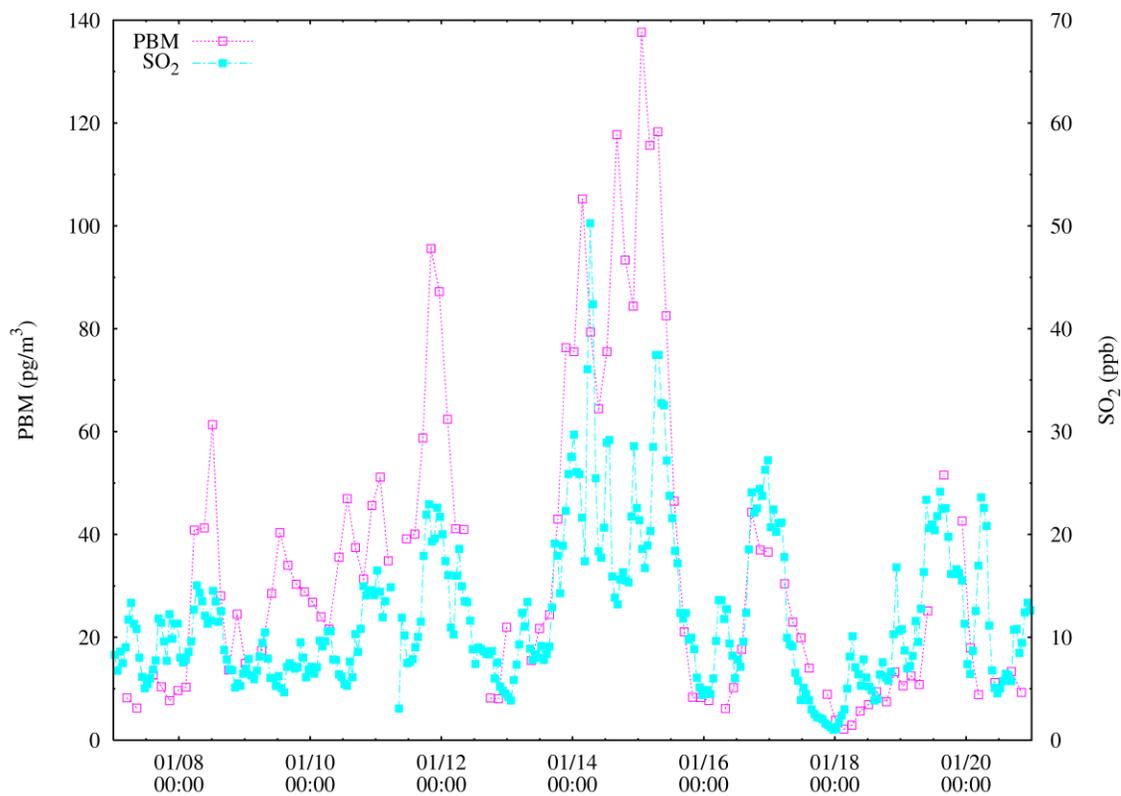


Figure 14. Time series of PBM (magenta line) and SO₂ (blue line) at the Bronx site, January 7-20, 2010.



7. Quality Assurance

The quality of the speciated ambient Hg data is based on the performance of the Tekran Model 2537B analyzer. The analyzer utilizes a gold trap to collect Hg over a five-minute collection period while a second gold trap is simultaneously thermally desorbed to measure the amount of Hg collected during the previous five-minute cycle. The analyzer uses cold vapor atomic fluorescence spectroscopy (CVAFS) to determine the amount of Hg(0) that was adsorbed onto the gold trap during each sample cycle. These five-minute Hg(0) data are produced by the analyzer during the first two hours of each three-hour interval. During the third hour, the analyzer determines the amount of RGM and PBM collected during the first two-hour interval. The RGM is collected on a quartz annular denuder coated with KCl and housed in the Model 1130 unit, and the PBM is collected on a quartz filter in the Model 1135 unit. Both the denuder and quartz filter must use a larger volume of ambient air to collect enough RGM and PBM for analysis by the Model 2537B. The flow of 10 L/min over two hours provides sufficient sensitivity for this study.

The accuracy and precision of the Model 2537B are checked over time to insure that the final dataset is accurate and consistent. The Model 2537B has a built-in permeation source that is used every 24 (Bronx) or 72 (Rochester) hours to calibrate the response of the detector to Hg. The permeation source is actually a small amount of pure Hg contained within a piece of Teflon tubing which is maintained at a specific temperature. The Hg diffuses through the Teflon at a constant rate and provides a stable and consistent absolute standard. The use of pure Hg in the internal permeation source is the preferred way to calibrate the instrument and is the way the two 2537Bs were calibrated throughout this study. There are, of course, ways in which the permeation source can fail and for this reason an external permeation source should be employed once per year in order to confirm that the internal source is functioning properly. The external permeation source is a stand-alone temperature controlled volume of air above 2 ml of pure Hg. The air which contains a known amount ($\sim 28 \text{ ng/m}^3$) of Hg vapor is injected into the 2537B and the result is compared to the result obtained from the internal permeation. The results obtained during this study from NYSDEC staff and an external audit from the AMNet site liaison are summarized in Table 3.

Table 3. External permeation injection results.

Site	Date	Result	Auditor
Bronx 2537B	Aug 19, 2008	-3.25%	NYSDEC
Bronx 2537B	Oct 7, 2009	-4.04%	NYSDEC
Bronx 2537B	Nov 16, 2010	-3.01%	NYSDEC
Rochester 2537B	Aug 21, 2008	-2.84%	NYSDEC
Rochester 2537B	Sept 29, 2009	-0.27%	NYSDEC
Rochester 2537B	Nov 19, 2010	-4.77%	NYSDEC
Rochester 2537B	Nov 11, 2008	0.0025%	AMNet

In addition to accuracy, the precision of the Model 2537B is important. The precision of the instrument response is determined by comparing the response of the instrument from one internal permeation calibration to the next providing that nothing has been adjusted. The magnitude of the instrument response changes whenever an adjustment is made to one of the operating parameters such as lamp voltage or sensitivity. To calculate precision for this study, the calibration responses were compared for periods when the instrument was unattended and therefore no adjustments had been made. The precision for this study is then calculated as the average of the percent difference of the instrument response from one calibration to the next. Table 4 shows the standard deviation and the average of the change in the instrument response from one calibration to the next.

Table 4. Average percent difference and percent standard deviation of instrument response factors from one internal calibration to the next.

Site	Trap A Ave	Trap B Ave	Trap A Std Dev	Trap B Std Dev
Bronx	0.13%	0.41%	1.9%	1.8%
Rochester	-0.14%	-0.04%	1.3%	1.3%

Other factors affecting the accuracy of the Tekran measurements include the Model 2537B analyzer and Model 1130 pump flow rates, and the capture efficiency of the denuder and quartz filter. The Model 2537B's flow rate is nominally 1 L/min and is controlled by a flow controller. The flow rate is verified during the bi-weekly service visits and the flow rates for both instruments have always been within $\pm 2\%$ of the external standard. The Model 1130 inlet flow is also verified every two weeks. This flow varies due to the nature of the flow path through the impactor, denuder, filter, pyrolyzer, heated line and the analyzer and is within $\pm 10\%$ of the nominal 10 L/min design flow. The audit flow device used in this study is a BIOS Definer 220-H, which is a primary flow standard.

The capture efficiencies of the RGM denuder and the PBM quartz filter have been determined by researchers during the development of the Tekran system. Reproducing this work is outside the scope of this study but a summary of the results as shown below demonstrate that the method is suitable for this ambient work:

- Lindberg et al. (2002) using an automated external mercury calibration source observed spike recoveries of 85 to 114% with an average of $102 \pm 3\%$ and zero checks of $0.09 \pm 0.01 \text{ ng/m}^3$ on the Model 2537A. A comparison of the Manual 1130 with the manual denuder method was found to be within 7%.
- Poissant et al. (2005) observed zero air blanks for the speciation units Models 1130 and 1135 of $0.66 \pm 1.25 \text{ pg/m}^3$. They reported a precision for the Model 2537A of approximately 2% and an average detection limit of 0.06 ng/m^3 .
- Han et al. (2004) reported a precision of $7.6 \pm 5.4\%$ for total gaseous mercury (TGM = RGM+Hg(0)) as measured by the Model 2537A using standard injection of mercury

vapor from an external calibrator. Field blanks averaged from 0.19 to 0.29 ng/m³ and the MDL ranged from 0.19 to 0.55 ng/m³.

Several researchers have also attempted to estimate the overall uncertainty of each of the components measured by the Tekran Models 2537B, 1130 and 1135. The results shown in Table 5 were estimated from older datasets from primarily rural areas. These uncertainties should be considered to be an upper bound for the data from this study.

Table 5. Estimated overall uncertainty as reported by Jaffe et al. (2005) and Landis et al. (2002).

Fraction	Measurement Uncertainty
Hg(0)	± 12%
RGM	± 15%
PBM	± 15%

7.1 Data Validation Criteria (AMNet Screening Program)

The AMNet program and its membership worked together to develop an instrument SOP and data screening criteria. The goal of this effort was to have all of the data of comparable quality from each provider so that the resulting national dataset could be used for site by site comparisons as well as for wide scale trends analysis. The five-minute data files from the Model 2537B, the electronic data from the DataComm option and the field data sheets were submitted to the AMNet data manager on a quarterly schedule. The AMNet data processor reviews the field data form, processes the data through an automated screening program, discusses potentially suspect data with NYSDEC staff, and manually inspects the data if necessary to assign specific quality assurance flags. Table 6 lists the AMNet data QA levels; as of April 11, 2011, the data from both sites have been assigned level 4 validation through all of 2010.

Table 6. AMNet data flags.

QA Level	Description
1	Raw data that have been submitted to NADP.
2	Processed by the NADP automated QA system.
3	Site Liaison review completed.
4	Site Operator review complete and data ready for distribution.
5	Embargo; data has been placed in holding until problems are resolved.
0	No observations made or available.

The specifics of how the data are processed by AMNet and a more complete description of the data management procedures can be found in the Data Management SOP. The AMNet Tekran SOPs, including the Data Management SOP which is currently still being revised, will be made available on the NADP AMNet website (<http://nadp.sws.uiuc.edu/amn/docs.aspx>).

8. Work Products and Outputs

The NYSDEC has presented some of the findings from this report at the upcoming National Air Toxics Monitoring and Data Analysis Workshop (April 4-7, 2011) in Dallas, TX. Although travel restrictions precluded NYSDEC to appear in person, an EPA Region 2 staff member agreed to make the presentation on our behalf. Appendix A includes a link to the Workshop as well as other EPA Air Toxics websites. The NYSDEC has also submitted an abstract for a poster presentation to the upcoming 10th International Conference on Mercury as a Global Pollutant (July 24-29, 2011) in Halifax, NS. By April 15, 2011 we will be notified whether or not our abstract has been accepted. The NYSDEC has recently obtained nearly one year of speciated Tekran data from Elizabeth, NJ, operated by NJDEP. It is anticipated that the two agencies will collaborate on investigating Hg sources in the New York City metropolitan area.

The NYSDEC has made several presentations using these data over the course of this project. The NYSDEC has also provided information for several other presentations as well. Appendix B lists the eight presentations that have included ambient and wet deposition data collected during this project. In addition the NADP Program Office has received numerous requests in recent years to use AMNet data for proposed analyses, including model evaluation, estimation of seasonal and spatial patterns, and investigation of chemical and physical processes that affect mercury. Appendix C lists seven such requests, and the NYSDEC has granted use of these data in each instance. We anticipate that there will be additional opportunities to present and share these valuable data in coming years.

9. Conclusions

The NYSDEC applied for this Community Assessment Grant to fund the equipment and method development required to establish a baseline of mercury species in two urban areas in New York. The study has taken much longer than anticipated but the results have been worth the extra time and effort. The data are available on the NADP MDN and AMNet websites, and much of the method developmental work has gone into the AMNet program SOPs and the quality of some of the manufacturer's products. The NYSDEC sees value in the wet deposition and speciated mercury measurements, and staff has pursued additional grant opportunities to continue the measurements further into the future.

The MDN measurements have been going on for more than three years. Currently there are only four wet deposition monitors across New York, the Bronx and Rochester urban sites operated by the NYSDEC, and two rural monitors in the Adirondack (Huntington Wildlife) and Catskill (Biscuit Brook) Mountains. While Hg concentrations at these two urban sites are generally higher than the rural sites in the state, wet depositional loadings are generally highest at the Bronx and Biscuit Brook sites, driven largely by precipitation volume. The MDN data collected to date are valuable data for examining seasonal and annual wet Hg deposition data in New York, and will continue to provide baseline data as state and federal emissions reductions programs are promulgated in coming years.

The Tekran data have provided insight into ambient Hg levels at two contrasting urban sites. The average Hg(0) concentrations at these two urban sites are similar to that at the rural Huntington Wildlife site, whereas the average concentrations of RGM and PBM are considerably higher at the urban sites. Although speciated Hg concentrations are generally similar at the two

sites, the variability in the Bronx data is higher. This may be due to PBL dynamics and more diverse sources affecting the Bronx site. At both sites there is a springtime maximum in RGM, and a wintertime maximum in PBM. The photochemical nature of RGM is evident in the high values in the afternoon during the warmer months, while PBM is dominated by combustion sources and PBL dynamics during the colder months. Finally, pollution roses and analysis of co-pollutants provide some insights into sources and fates of Hg, especially in the New York City region, but also suggest that more work can be done to characterize the ambient Hg cycle.

10. Acknowledgments

The NYSDEC greatly appreciates the support of the US EPA (Grant # XA-97265006). Additional support was provided by the Great Lakes Commission and the New York State Energy Research and Development Authority. The NYSDEC also wishes to thank David Gay of the NADP Program Office, and AMNet site liaison Mark Olson for their assistance throughout the study period, and for making MDN and AMNet data from other sites available for use in this report. This work could not have been completed without the efforts of the following NYSDEC staff responsible for sample pickups, and site visits and maintenance: Matthew Hirsch, Thomas Everts, and Sergio Fleishaker.

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Appendix A: Final Report Summary in Power Point Format: Presented at the EPA National Air Toxics Monitoring and Data Analysis Workshop April 4-7, 2011 (Dallas, Texas)

The summary presentation will be available on the 2011 National Air Toxics Monitoring and Data Analysis Workshop webpage.

<http://www.epa.gov/ttn/amtic/toxmeet.html>

The entire report will be made available in electronic format from the EPA Community Scale Air Toxics website.

<http://www.epa.gov/ttn/amtic/local.html>

http://www.epa.gov/ttn/amtic/20052006_CSATAM.html#reports

Appendix B: Presentations of Project Data

NYSDEC staff have authored, or contributed mercury concentration and wet deposition data to support, a number of presentations as of April 11, 2011.

1. Civerolo, K., D. Felton, S. Fleishaker, and T. Everts, Wet mercury deposition in New York and the surrounding region: current status, NADP Annual Meeting and Scientific Symposium, Saratoga Springs, NY, October, 2009.
2. Felton, D., NACAA Members Meeting, NY's Experience with Tekran Speciated Hg Monitoring, the MDN program and the proposed Mercury Initiative (MI), Louisville, Kentucky, April 29 – May 2, 2007.
3. Felton, D., K. Civerolo, and M. Hirsch, A Data Review and Operational Notes: A Year Spent with the Tekran Speciated Ambient Mercury Analyzer at Two Urban Locations in New York State, National Air Monitoring Conference, Nashville, TN, November 2 - 5, 2009.
4. Felton, D., K. Civerolo, and M. Hirsch, Urban Mercury Monitoring: Data Review and Operational Notes: A Year Spent with the Tekran Speciated Ambient Mercury Analyzer at Two Urban Locations in New York State, MARAMA Monitoring Conference, Philadelphia, PA, February 23, 2010.
5. Felton, D., K. Civerolo, M. Hirsch, S. Fleishaker, and T. Everts, Baseline measurements of ambient concentrations of elemental, reactive gaseous and particle-bound mercury at two urban locations, NADP Annual Meeting and Scientific Symposium, Saratoga Springs, NY, October, 2009a.
6. Felton, D., K. Civerolo, M. Hirsch, S. Fleishaker, and T. Everts, Baseline measurements of ambient concentrations of elemental, reactive gaseous and particle-bound mercury at two urban locations, NYSERDA Environmental Monitoring, Evaluation, and Protection meeting, Albany, NY, October, 2009b.
7. Kilner, P. I., E. M. Prestbo, D. Felton, T. Holsen, S. Lyman, and Mark L. Olson, Field techniques to assure the quality of atmospheric mercury speciation measurements, 9th International Conference on Mercury as a Global Pollutant, Guiyang, China, June, 2009.
8. Prestbo, E., D. Gay, D. Schmelz, T. Sharac, and M. Olson, The North American Atmospheric Mercury Speciation Network, Air Quality VII Conference, Arlington, VA, November, 2009.

Appendix C: Specific Requests for Project Data

The NADP Program Office has received a number of AMNet data requests as of April 11, 2011. The NYSDEC has allowed their Hg data to be used by the following investigators. Please note that Hg(0) is sometimes referred to as “GEM,” RGM is sometimes referred to as divalent mercury (“Hg²⁺” or “Hg(II)”) or gaseous oxidized mercury (“GOM”), and PBM is sometimes referred to as “PHg” or “PB25.”

1. Dr. Jesse Bash, EPA. Dr. Bash would like to use data from all sites but Alert for all three fractions. His goal: I would like to use this monitoring data for model evaluation of an annual CMAQ run to be done later this year. We plan on running the model for the year of 2005 for the continental US. We have evaluated the model results with wet deposition in the past but I would like to use ambient measurements in the evaluation as well. The wet deposition evaluation can be used to inform us if the oxidation reactions are balanced against the aqueous phase reduction but we do not have enough observations to constrain the problem well. If both the oxidation and aqueous phase reduction rates are too high or low, we can still match the wet deposition but we will not capture the ambient concentrations or the transport well. If we have an idea of what the Hg(0), Hg(II), and PHg concentrations are then we should be able to evaluate the oxidation and reduction mechanisms in the models chemical mechanism. The spatial patterns of Hg(II) and PHg should give use an idea of what the dry deposition of those species should be because we do not expect them to have a very long atmospheric lifetime. His plan is to determine seasonal values from our observations to compare to his model results, specifically:“ I would like to compare the model to mean/median seasonal values (assuming that there is some sort of seasonal pattern) to get a more complete spatial coverage and an idea of what the seasonal and annual variability and because we are not ready to run 2008/2009 simulations.”
2. Prof. Kevin Crist, 159 Stocker, Ohio University, Athens, OH 45701. Dr. Crist would like to use data through 2009. His goal is to provide an improved understanding of mercury fate transport, and depositional patterns within the Great Lakes region as well as to quantify relative contribution of major mercury sources (i.e., global, Chinese, regional). To accomplish this task the Ohio University Air Quality Center is conducting regional and global chemical transport model simulations (MM5-SMOKE-CMAQ) to provide an improved understanding of the fate, transport, and deposition of mercury within the Great Lakes region. To evaluate the performance of the modeling system the output will be evaluated against the AMNET data.
3. Dr. Eric K. Miller, Ecosystems Research Group, Ltd., PO Box 1227, Norwich, VT 05055. The purpose of accessing these data is to produce an updated estimate of mercury deposition to northeastern North America following the methods of Miller et al. A major weakness of Miller et al. 2005 was the limited observations of GEM, GOM, and PBM available at the time. AMNet has changed all that and provides the opportunity for a much improved estimate of regional air concentration fields and dry deposition.

4. Dr. Lyatt Jaegle and Yanxu Zhang/University of Washington. The data will be used to provide very unique constraint on our regional mercury model, especially on the gas/particulate partition of divalent mercury and the oxidation mechanisms of elementary mercury. We will use this data in the following aspects: constraining the gas/particle phase partition scheme developing an in-plume degradation scheme of Hg²⁺ checking the assumption that Br is the main oxidation of elementary mercury constrain the diurnal profile of mercury over land constrain the chemistry and transport budget of mercury over North America.”
5. Dr. Leiming Zhang, Environment Canada. Dr. Zhang would like to use AMnet data in monthly average GEM, GOM, and PB25 values at several sites and years to compare to his model output and annual dry deposition calculations. In a second request, he asked for the data for use in the direct calculation of mercury dry deposition (using inferential method). The data may also be used to explore the relation between dry and wet deposition.
6. Dr. Robert Talbot, Professor of Atmospheric Chemistry, Director, Climate Change Research Center and NOAA AIRMAP Cooperative Institute, Institute for the Study of Earth, Oceans, and Space Morse Hall, 8 College Road, University of New Hampshire, Durham, NH 03824. Lan Xin is planning to conduct data and statistical analyses of the atmospheric mercury data collected across the U.S. by AMnet. They are interested in analyzing the spatial and seasonal trends across the U.S.
7. Dr. Christian Temme, Stellvertretender Laborleiter, Eurofins GfA GmbH, Neuländer Kamp1, D-21079 Hamburg. He is using MDN data in a statistical table of global mercury observation as a means of comparison for sites in Germany and Europe.