Atmos. Chem. Phys. Discuss., 13, 10521–10546, 2013
www.atmos-chem-phys-discuss.net/13/10521/2013/
doi:10.5194/acpd-13-10521-2013

Atmospheric Chemistry and Physics Discussions

The Atmospheric Mercury Network: measurement and initial examination of an ongoing atmospheric mercury record across North America

D. A. Gay[1], D. Schmeltz[2], E. Prestbo[3], M. Olson[1], T. Sharac[2], and R. Tordon[4]

[1]National Atmospheric Deposition Program, Illinois State Water Survey, 2204 Griffith Drive, Champaign, IL 61820, USA
[2]US Environmental Protection Agency, Office of Atmospheric Programs, Clean Air Markets Division, 1200 Pennsylvania Avenue, NW, MC6204J, Washington, D.C. 20460, USA
[3]Tekran Research and Development, 2707 NE 125th Street, Seattle, WA 98125, USA
[4]Environment Canada, 45 Alderney Drive, Dartmouth, Nova Scotia, Canada B2Y 2N6, Canada

Received: 22 January 2013 – Accepted: 5 March 2013 – Published: 19 April 2013

Correspondence to: D. A. Gay (dgay@illinois.edu)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

The National Atmospheric Deposition Program (NADP) developed and operates a collaborative network of atmospheric mercury monitoring sites based in North America – the Atmospheric Mercury Network (AMNet). The justification for the network was growing interest and demand from many scientists and policy makers for a robust database of measurements to improve model development, assess policies and programs, and improve estimates of mercury dry deposition. Many different agencies and groups support the network, including federal, state, tribal, and international governments, academic institutions, and private companies. AMNet has added two high elevation sites outside of continental North America in Hawaii and Taiwan because of new partnerships forged within NADP. Network sites measure concentrations of atmospheric mercury fractions using automated, continuous mercury speciation systems. The procedures that NADP developed for field operations, data management, and quality assurance ensure that the network makes scientifically valid and consistent measurements.

AMNet reports concentrations of hourly gaseous elemental mercury (GEM), two-hour gaseous oxidized mercury (GOM), and two-hour particulate-bound mercury less than 2.5 microns in size ($PBM_{2.5}$). As of January 2012, over 450 000 valid observations are available from 30 stations. The AMNet also collects ancillary meteorological data and information on land-use and vegetation, when available. We present atmospheric mercury data comparisons by time (3 yr) at 22 unique site locations. Highlighted are contrasting values for site locations across the network: urban versus rural, coastal versus high-elevation and the range of maximum observations. The data presented should catalyze the formation of many scientific questions that may be answered through further in-depth analysis and modeling studies of the AMNet database. All data and methods are publically available through an online database on the NADP website (http://nadp.isws.illinois.edu/amn/). Future network directions are to foster new network partnerships and continue to collect, quality assure, and post data, including dry deposition estimates, for each fraction.

ACPD
13, 10521–10546, 2013

1 Introduction

The current Mercury Deposition Network (MDN), initiated in the mid-1990's, provides data on the concentration of total mercury (Hg) in precipitation at 110 sites across North America. MDN is one of five networks of the National Atmospheric Deposition Program (NADP), which monitors the rate of pollutant removal from the atmosphere and deposition loadings to ecosystems. MDN is a critical and valued component of a comprehensive Hg monitoring strategy (Schmeltz et al., 2011). However, a lack of scientific information on the dry deposition of Hg and limited monitoring coverage over different geographic scales provides an incomplete picture of Hg atmospheric transport and total (wet + dry) Hg deposition. Based on atmospheric model estimates and several field studies, the dry deposition of Hg (particles and gases that deposit without the assistance of precipitation) has been estimated to be wide ranging relative to wet deposition. Earlier modeling found that dry deposition could be from much lower to much higher than wet deposition at different locations (Seigneur et al., 2004), while later modeling suggested that dry deposition in the US is greater than wet deposition (Selin et al., 2008) or equal in magnitude (Zhang et al., 2012a). Several field-level studies also estimated dry deposition to be of the same magnitude as wet deposition (Lamborg et al., 2002; Caldwell et al., 2006; Lyman et al., 2007).

Three atmospheric Hg fractions (i.e. individual chemical species and groups of the same) contribute to dry deposition: (1) gaseous elemental Hg (GEM), (2) gaseous oxidized Hg (GOM), and (3) particulate-bound Hg ($PBM_{2.5}$). The impact of Hg dry deposition can be substantial, and has been attributed to local sources (Gustin et al., 2012). Even though estimated GEM dry deposition rates are small relative to GOM and $PBM_{2.5}$, GEM comprises more than 95 percent of the total Hg in the air at ground level, and can be a significant component of dry deposition. GEM can be rapidly oxidized and deposited locally or regionally (Lindberg et al., 2002; Weiss-Penzias et al., 2003; Driscoll et al., 2007), is important in forested ecosystem deposition (Grigal, 2002; Ericksen et al., 2003), and long range transport of GEM and in-situ oxidation to GOM/$PBM_{2.5}$

ACPD
13, 10521–10546, 2013

in the free troposphere has been reported at high elevations in the US (Swartzendruber et al., 2006).

While scientists have quantified Hg in precipitation, the approaches to measuring dry deposition continue to evolve. Methods to measure dry deposition are actively under development (Lyman et al., 2009; Huang et al., 2011; Lai et al., 2011; Castro et al., 2012; Gustin et al., 2012, among others). When the NADP membership began considering an atmospheric Hg network in 2004, the most promising and available approach to estimate dry deposition in select locations was high-resolution, continuously measured concentrations of atmospheric Hg fractions, combined with modeled deposition parameters. Although recognized as important for improving the scientific understanding of the fate of atmospheric Hg (for example, Fitzgerald, 1995; Mason et al., 2005; Harris et al., 2007), network-scale atmospheric Hg data have not been widely available.

Starting in 2006, NADP advocates consulted a variety of Hg scientists to determine network viability and to explore standard methods to measure air Hg fractions in a network-mode. In 2009, NADP formally launched the Atmospheric Mercury Network (AMNet) to measure atmospheric Hg concentrations and estimate the dry deposition of Hg to complement the Hg wet deposition measurements of MDN. Over 100 scientists contributed to the current instrument selection, development of equipment operating procedures, and data management methods adopted for use in AMNet. AMNet uses automated, continuous measuring systems to measure the atmospheric Hg fractions GEM, GOM, and $PBM_{2.5}$. All network sites use standard operating procedures to operate and maintain the measuring equipment, including routine documentation. A single data management system processes all of the data with three levels of review in order to report consistent, quality assured observations. We report here for the first time an accessible, standardized North American database of atmospheric Hg measurements that should provide for future trend analysis, model development, and total mercury deposition estimates. We also report on three years of observations from a number of monitoring locations and site groupings, compare and contrast the results, and identify several research questions as yet unanswered.

ACPD
13, 10521–10546, 2013

1.1 Network objectives

AMNet's goal is to coordinate, quality-assure, store, and share atmospheric concentration measurements of Hg fractions that contribute to dry and total Hg deposition. The network builds on NADP's 35-yr history and experience of successful, collaborative environmental monitoring by offering a database of high quality Hg measurements that complement the existing MDN program.

It is AMNet's objective to provide the database to multiple and diverse stakeholders groups to support an array of science, policy, and management objectives, including:

- assessing geographic patterns and long-term temporal trends in the concentrations of atmospheric Hg fractions and dry and total Hg deposition in selected areas;
- improving the information base for evaluating models of atmospheric Hg chemistry, transport, and deposition;
- assessing the impact of local, regional, and global Hg emitting sources as Hg emission reduction programs are implemented; and
- evaluating the status and trends in total deposition of atmospheric Hg to ecosystems with a high potential for Hg methylation and Hg bioaccumulation in fish and wildlife.

As examples, the network has catalyzed new scientific investigations involving many different collaborators and sites, including regional-scale photochemical modeling (Baker and Bash, 2012), GOM dry deposition measurement (Castro et al., 2012), seasonal and diurnal variation of Hg fractions (Lan et al., 2012), a regional modeling assessment (Zhang et al., 2012b), and an atmospheric Hg simulation over North America (Zhang et al., 2012c).

ACPD
13, 10521–10546, 2013

2 Operation of the AMNet

2.1 Monitoring locations

To help categorize Hg cycling in different ecosystems, AMNet has a variety of siting classifications, including rural, urban, coastal, and high altitude locations. Many AMNet sites were previously established, long-term air Hg research or monitoring sites. Current operating AMNet sites are described in Fig. 1 and Table 1. North American coverage is better throughout the East but major gaps remain. Several other sites operate in the West and South. Several urban sites operate in Birmingham, AL, New York City and Rochester, NY, and Salt Lake City, UT. High elevation sites operate in Hawaii (3384 m) and in a new Asian location of Taiwan (2862 m). A new site began operating in January 2012 in rural Wisconsin, but it is not considered here.

2.2 Field operation

Currently, all AMNet sites use the Tekran Continuous Mercury Vapour Analyzer Model 2537 coupled with the speciation models 1130 for GOM, and 1135 for $PBM_{2.5}$ (Tekran Instruments Corp., Toronto, Ontario Canada). This equipment is not exclusive for use in the network, but meets the AMNet requirements and is commonly available. The operation and principles of the instrument are described in Landis et al. (2002). Data captured using either personal computers or data loggers are submitted to the network monthly.

Standard Operating Procedures (SOP) for AMNet were developed and reviewed by experts as described in network documentation. Full SOPs are available on-line (NADP, 2011a,b) and include sections on field operations, data management, site selection, and field maintenance and reporting procedures. Specific site information is available to data users, since location, terrain, land use, and other characteristics may have a significant influence on atmospheric Hg observations and deposition model application.

ACPD
13, 10521–10546, 2013

2.3 Quality assurance and validation

Prior to posting, the observed Hg data are quality assured using a three-step process. Only valid data are made available six months after collection. Step one uses the NADP's automated quality assurance software to screen the raw data. This software evaluates the data utilizing 35 potential flags (NADP, 2011a; Steffen et al., 2012). Twenty of the flags are warning limits established to draw attention to the data for possible corrective actions. If only warning flags are assigned, the data are considered valid. The remaining flags are control limits, which invalidate data when exceeded. Following automated QAP review, the data are set to a quality rating of "1".

The AMNet site liaison performs step two by incorporating field observations, manually reviewing the data, and identifying any anomalies present. Data affected by maintenance are invalidated. At this point, the data are set to a quality rating of "2". The site liaison then provides site operators and investigators the opportunity for field verification by supplying them with a monthly report, including data anomalies.

The third step requires the site operator/investigator to approve the summarized data and initial data flags. Sites can clarify data records and have data reevaluated by the network. Once this process is completed, the data are set to a quality rating of "3". Data with a quality rating of 3 are fully approved for reporting to the NADP website.

As an additional quality assurance step, the AMNet site liaison performs annual site audits, following specific audit criteria and testing procedures. These testing procedures include evaluation of siting criteria, instrument operation, and any additional training.

Recently, a potential GOM measurement interferant has been noted (Lyman et al., 2010; Gustin et al., 2013). The research of Lyman et al. (2010) suggests that the oxidant ozone, leads to a chemical reduction of collected GOM compounds on KCl-coated annular denuders, releasing GEM from the denuder. The results suggest that although total mercury concentration would be unaffected, the fraction of GOM could be biased low. The additional research on this topic just reported by Gustin et al. (2013)

ACPD
13, 10521–10546, 2013

is complex and perplexing owing to their acknowledged challenges and observed biases in the different types of analyzers and especially transport of mercury through the common manifold. For example, the two standard, carefully calibrated and quality assured mercury speciation analyzers (Tekrans) had approximately a 30 % difference in GEM concentration when connected to the manifold as reported in Gustin et al. (2013). In contrast, co-located, calibrated and quality assured mercury speciation analyzers (Tekrans) in the field typically have an average difference of less than 5 % (Prestbo et al., 2011). What is conclusive from these studies is that research on the accuracy of GOM measurements continues to be studied and debated. Nevertheless, the GOM measurements reported here were made using the currently best available equipment, procedures and quality assurance as determined by the consensus of the expert atmospheric mercury community.

3 Data and availability

3.1 Measurements

The Tekran 2537 continuously measures GEM for two hours in five-minute intervals. During this two-hour period of GEM measurements, GOM and $PBM_{2.5}$ samples are being collected by denuder adsorption and filtering, respectively. The concentration of each fraction in ambient air is exceedingly low, hence the need for a high-flow, two-hour sample period. Following the two-hour sample period, the filter and denuder are heated sequentially, in zero air, to release the Hg into the Tekran 2537 in order to determine the concentration of $PBM_{2.5}$ and GOM, respectively. The $PBM_{2.5}$ and GOM analysis requires one hour. Thus, for every three-hour period, AMNet reports two GEM one-hour average values and the time-correlated two-hour $PBM_{2.5}$ and GOM values. GEM concentrations are reported in nanograms per cubic meter ($ng\,m^{-3}$), and GOM and $PBM_{2.5}$ are reported in picograms per cubic meter ($pg\,m^{-3}$). Each Hg value is reported with the AMNet site identifier, start time, end time, and additional quality assurance

ACPD
13, 10521–10546, 2013

information. All of the valid AMNet data are made available to the public through the NADP website (http://nadp.isws.illinois.edu).

3.2 Network intercomparison

Environment Canada has a long history of network-based Hg monitoring going back to 1996 with the start of the Canadian Atmospheric Mercury Measurement Network (CAMNet, Kellerhals et al., 2003; Temme et al., 2007). The Canadian network uses their own quality assurance software, the Research Data Management and Quality Assurance System (RDMQ™), to quality assure and quality control data across CAMNet. In order to quantify the level of agreement between the two systems, raw data sets from multiple sites were processed through both software programs (Steffen et al., 2012). The final qualified data sets compared favorably between networks at the four tested sites, and the number of flags assigned by each program was generally very similar. For two of the longer-term, mid-latitude sites, results showed very good comparability. Mean differences in validated Hg fraction concentrations between the resulting datasets were small for GEM (0.3 %), and somewhat larger for GOM and $PBM_{2.5}$ (8.6 and 15 %, respectively). However, for the High Arctic site, with extremely variable Hg values and difficult monitoring conditions, the quality assured and validated data sets showed larger mean concentration differences (2.7, 27, and 33 %, respectively). The robustness and general agreement between the two quality assurance programs provides confidence that Hg data generated by AMNet are quality assured consistently with this network, although some differences were present.

3.3 Network observations and analysis

Statistics describing the currently available data and specific, by-site statistics are in Supplemental Table 1. As of January 2012, there were approximately 150 000 valid hourly and 2-h averages for each gaseous Hg fraction made by the network sites. Specific site-by-site observation number ranges from approximately 2500 to 11 000

ACPD
13, 10521–10546, 2013

observations per site. Data for sites NJ30, NJ32, NJ54, NY43, NU15 (Alert), PA13, TW01 (Taiwan), WI99, and WV99 were not used for this overview.

The Mauna Loa, Hawaii high elevation site has unique Hg observations compared to the other sites, so it is interpreted separately. The HI00 observations are similar to the other AMNet high elevation site, Mt. Lulin, Taiwan (TW01, Sheu et al., 2010), and will be available in the future on the NADP website. Both high elevation sites receive nighttime, subsiding free tropospheric air, which frequently has enhanced GOM and $PBM_{2.5}$ and depleted GEM levels (Swartzendruber et al., 2006; Obrist et al., 2008; Lyman and Jaffe, 2011; Fig. 2a–c herewith). The HI00 site receives naturally-emitted Hg from the active Kilauea Volcano (Nriagu and Becker, 2003). Even with a similar median GEM value, the interquartile range (2nd and 3rd quartiles) was the largest of any site at about 0.65 $ng\,m^{-3}$ and the 5th to 95th percentile range was 0.4 $ng\,m^{-3}$ to 2.7 $ng\,m^{-3}$ (Fig. 2a). Strikingly, the GOM and $PBM_{2.5}$ median and mean values were five to ten times greater than any other site (Fig. 2b, c). Further analysis of the HI00 and TW01 high elevation sites is highly encouraged, but is beyond the scope of this work. One thing is clear; inclusion of network-based, continuous long-term measurements of atmospheric Hg at multiple high elevation sites is essential for modeling and understanding trends, fate, and transport.

The AMNet GEM observations highlight several expected and unexpected results (Fig. 2a). As expected, for 15 of the 21 AMNet sites (excluding HI00), the median and mean GEM values were between 1.3 and 1.5 $ng\,m^{-3}$, with a typical interquartile range of about 0.25 $ng\,m^{-3}$. Three of 21 sites with means and medians well above this GEM range are urban or urban-influenced sites. Why does NY06, located in the heart of New York City, not match the other urban sites with higher GEM concentrations? With the largest, single database of multi-year urban observations, AMNet provides a new opportunity to evaluate how urban areas influence mercury deposition. There is no clear reason why three of 21 sites have means and medians below 1.3 $ng\,m^{-3}$. For example, consider NY20 and VT99; both are remote, continental forested sites in the Northeastern USA approximately 100 km apart. NY20 and VT99 had contrasting GEM

ACPD
13, 10521–10546, 2013

mean, interquartile, 95 % and min/max ranges. We postulate that site elevation and local effects may explain the differences. VT99 is located at a local high elevation and has a long fetch not influenced by surface exchange. In contrast, NY20 is located in a lake valley and has a very short fetch to dense forest.

Most sites observed GEM below 2.0 ng m^{-3} for the overwhelming number of hours during all years. Minimum observations rarely went below 0.5 ng m^{-3}. The 95th percentile GEM concentrations for the sites were highly variable, and ranged from 1.6 to 2.85 ng m^{-3}. Year to year, the median change at the typical site was small and limited to 0.1 or 0.2 ng m^{-3}. The average GEM value does not appear to predict the number of extreme values. Nearly all sites had multiple events above 3 ng m^{-3}, with some sites with a great frequency of extreme values.

Ranges of GOM and $PBM_{2.5}$ are shown in Fig. 2b and c, respectively. The median GOM concentrations were typically between 1.2 and 2.5 pg m^{-3}. The more remote sites were clearly the lowest, particularly for the marine Pacific and Nova Scotia sites. But the southern, coastal NJ05 site and the forested NY20 site were also very low. Perhaps at these sites, the combination of relative remoteness and coastal locations both led to lower values. At these sites, the GOM concentrations rarely exceeded 10 pg m^{-3}. The highest medians were at two of the urban sites (Salt Lake City and Rochester, 5–12 pg m^{-3}). These two sites also had the largest interquartile range and 95th percentiles. However, a similar median and range was also measured in western Maryland (MD08). Conversely, New York City and Washington, D.C. were somewhat suppressed, relative to the other urban sites. Clearly, there are many factors at play in the resulting GOM values that may be better understood using source-receptor analysis or similar.

$PBM_{2.5}$ medians were typically between 2.5 and 5.0 pg m^{-3}. The highest two medians were measured at NY95 and UT96 (10 pg m^{-3}, north of Salt Lake UT97), and the lowest at the coastal and continental remote sites (~2.5 pg m^{-3}). $PBM_{2.5}$ was less than 15 pg m^{-3} for the majority of sites and observations. The interquartile ranges were usually between 2.0 and 7.5 pg m^{-3}. The largest ranges were measured at NY95 and UT96, following with the highest medians. UT96 is in dry and dusty Utah, but in the

ACPD
13, 10521–10546, 2013

middle of the Great Salt Lake and downwind of significant mining sources. Rochester, NY has an industrial history, but is also on the shores of Lake Ontario. Considering the very different environmental conditions, further analysis may show why these two different sites would both observe high particulate Hg concentrations.

In comparing the GOM values to $PBM_{2.5}$ values (Sup. Fig. 1), we observed that all of the sites, save three, had median GOM concentrations less than $PBM_{2.5}$. Therefore, most sites observed $PBM_{2.5}$ levels greater than GOM, and more variable $PBM_{2.5}$ values. The three sites that showed the opposite relationship were the western Maryland MD08 site, HI00, and UT97 within Salt Lake City. UT97 is an urban site in the western US, that would be expected to have higher particulate levels. But this is not the case at nearby UT96, nor at any of the other urban sites. Gold production is important in the Great Basin Area, with gold purification emission sources in the UT97 area that could be one of the sources of high particulate mercury. MD08 is in a different environment altogether from UT97, so it is predicted that the formation, source and dry deposition rate of $PBM_{2.5}$ will be much different for these two locations.

It appears that site location relative to both natural and anthropogenic sources, elevation, and local conditions is influencing Hg concentrations. As an additional summary, we combined sites into loosely defined groups: the Pacific coast (CA48 only), coastal remote, continental remote, coastal near-sources, continental near-sources, urban and high elevation (HI00 only). Definitive conclusions based on the analysis of the group results are not recommended, due to the limited number of sites per group and years of data. In effect, the group analysis should be used to direct deeper and more refined analysis.

The group with the highest GEM median, variability and maximum concentrations was the urban group (Fig. 3a). Interestingly, the continental remote group had the lowest median. Three groups, coastal remote, coastal near-sources and continental near-sources, had surprisingly similar medians and variability for GEM. The Pacific coast group is closer to urban GEM than it was to the coastal remote group, but with a narrow interquartile range. Some small point sources near CA48 could explain the

ACPD
13, 10521–10546, 2013

higher GEM median; however, point sources usually result in more variability and some extreme values, which are not observed for CA48. A more likely explanation is GEM natural source emissions coming from the upwelling waters in Monterrey Bay (Weiss-Penzias et al., 2003).

For GOM, the Pacific and coastal remote groups had the lowest median and variability (Fig. 3b). This is in contrast to previous studies and models suggesting mid-day GOM production chemistry in the marine boundary layer. The median GOM for the continental near-sources group is high, and very similar to the urban group. Why does the continental near-sources group have a GEM median similar to other groups and the highest GOM median value? The coastal near-sources group has a lower median and range than the continental near-sources group. A coastal depletion in GOM relative to other groups is evident, at least from these observations. One would suspect this is due to precipitation removal and the lack of industrial sources in a seaward direction. This is distinct from GEM, where a coastal depletion relative to other groups is not present.

Similar to GOM, the highest $PBM_{2.5}$ medians and interquartile ranges were observed for the continental near-sources and urban groups (Fig. 3b). The lowest median values and ranges were observed for the Pacific and coastal remote groups. The overall higher $PBM_{2.5}$ to lower GOM relationship is clearly shown in all site groupings.

What may be more important, but beyond the scope of this paper is the magnitude and frequency of the GOM and $PBM_{2.5}$ values above the 75th and 95th percentile (Fig. 3b). Significant events are routinely reported for GOM and $PBM_{2.5}$ at all the site groups, with the fewest for the Pacific coast and the most for the source, urban and high elevation (HI00) groups. It will be informative to determine through future analysis how much the high GOM and $PBM_{2.5}$ values impact the overall dry deposition rate at various sites.

ACPD
13, 10521–10546, 2013

4 Importance, implications, and future directions

The importance of the AMNet is data, its availability, and its consistency over time and space. This consistency is a product of the standard operating procedures and quality assurance steps the AMNet puts in place. With long-term scientifically defensible data, the data can be used for different activities with significant implications for science and policy decisions. These data will allow for:

- the evaluation of, and improvements to, Hg air quality models, model intercomparison and development, and allow for model result comparisons;
- a baseline of atmospheric Hg concentrations to assess the magnitude of change due to regulation and expected emission reduction;
- determining the importance of atmospheric transport of local, regional and global sources to regional Hg concentrations and deposition; and
- stimulus and support of additional research and development into the biogeochemistry and cycling of mercury within ecosystems.

Several future directions are planned. The network will continue to encourage new partners to join AMNet and improve the spatial coverage of the network over North America and other countries. In the near future, the NADP, in collaboration with researchers at Environment Canada, will produce weekly dry deposition estimates from the Hg fractions measured at each operating site. Sites collocated with NADP/MDN will offer an estimate of total Hg deposition (wet + dry) to ecosystems.

This same focus on total Hg deposition has led to a trial network of litterfall deposition, used to estimate the litterfall input of Hg (NADP, 2012). Tied with AMNet and MDN, a more fully developed estimate of total deposition will be available at these select locations. Finally, the AMNet methods will be used to strengthen and facilitate consistency with other networks that measure Hg across the globe, including the European-led

ACPD
13, 10521–10546, 2013

Global Mercury Observation System (GMOS, http://www.gmos.eu/), and potential future Hg monitoring networks in Asia. The AMNet quality assurance program, including standard procedures for field operations, data review, and site audits, and the network intercomparison studies, are envisioned as a resource to the global monitoring community and an opportunity for further collaborations.

5 Summary

The NADP has successfully developed and is operating an international-scale and long-term network of atmospheric Hg monitoring sites in North America, Hawaii, and Taiwan. Many different agencies and groups support the network, including federal, state, tribal, and international governments, academic institutions, and private companies. The collaborative efforts of many Hg experts and site personnel are critical for making consistent measurements using the same equipment and operation methods and a network-wide systematic review of data.

The AMNet reports quality-assured measurements of hourly and two-hour gaseous Hg fractions, including future development of hourly dry deposition estimates for total Hg. Currently, over 450 000 individual observations are available. A short review of data available shows consistency over time at individual sites and interesting patterns between sites.

Supplementary material related to this article is available online at: http://www.atmos-chem-phys-discuss.net/13/10521/2013/acpd-13-10521-2013-supplement.pdf.

Acknowledgements. This network is able to function only with the work of the site operators, and Principal Investigators (M. Allen, St. of WI; S. Brooks, NOAA; R. Callison, Cherokee Nation; M. Castro, Un. of MD; K. Crist, Ohio Un.; E. Edgerton, ARA Inc.; D. Felton, St. of NY; T. Holsen,

ACPD
13, 10521–10546, 2013

Clarkson U.; W. Luke, NOAA; E. Miller, Ecosystems Res.; N. Olson, St. of UT; K. Perry, Un. UT; G.-R. Sheu, North Central Un.; S. Steffen, Env. Canada; R. Talbot, Un. of NH; J. Dalziel, Env. Canada; P. Weiss, Un. CA Santa Cruz; and R. Zsolway, St. of NJ). Their effort and advice are very much appreciated.

This paper has not been subjected to US Environmental Protection Agency (EPA) peer and administrative review; therefore, the conclusions and opinions contained herein are solely those of the individual authors, and should not be construed to reflect the views of the EPA. A mention of trade names, products, or services does not convey, and should not be interpreted as conveying, official EPA approval, endorsement, or recommendation.

The NADP is National Research Support Project-3: A Long-Term Monitoring Program in Support of Research on the Effects of Atmospheric Chemical Deposition. More than 240 sponsors support the NADP, including State Agricultural Experiment Stations; universities; private companies and other nongovernmental organizations; Canadian government agencies; state, local, and tribal government organizations; and federal agencies, including the US Department of Agriculture-National Institute of Food and Agriculture (under agreement no. 2012-39138-20273). Any findings or conclusions in this article do not necessarily reflect the views of the Illinois State Water Survey, US Department of Agriculture, or other sponsors.

References

Baker, K. R. and Bash, J. O.: Regional scale photochemical model evaluation of total mercury wet deposition and speciated ambient mercury, Atmos. Environ., 49, 151–162, 2012.

Caldwell, C. A., Swartzendruber, P., and Prestbo, E.: Concentration and dry deposition of mercury species in arid south central New Mexico (2001–2002), Environ. Sci. Technol., 40, 7535–7540, 2006.

Castro, M. S., Moore, C., Sherwell, J., and Brooks, S. B.: Dry deposition of gaseous oxidized mercury in Western Maryland, Sci. Total. Environ., 417–418, 232–240, 2012.

Driscoll, C. T., Han, Y. J., Chen, C. Y., Evers, D. C., Lambert, K. F., Holsen, T. M., Kamman, N. C., and Munson, R. K.: Mercury contamination in forest and freshwater ecosystems in the Northeastern United States, BioScience, 57, 17–28, 2007.

ACPD
13, 10521–10546, 2013

Ericksen, J. A., Gustin, M. S., Schorran, D. E., Johnson, D. W., Lindberg, S. E., and Coleman, J. S.: Accumulation of atmospheric mercury in forest foliage, Atmos. Environ., 37, 1613–1622, 2003.

Fitzgerald, W. F.: Is mercury increasing in the atmosphere? Need for an Atmospheric Mercury Network (AMNet), Water Air Soil Poll., 80, 245–254, 1995.

Grigal, D. F.: Inputs and outputs of mercury from terrestrial watersheds: a review, Environ. Rev., 10, 1–39, 2002.

Gustin, M. S., Weiss-Penzias, P. S., and Peterson, C.: Investigating sources of gaseous oxidized mercury in dry deposition at three sites across Florida, USA, Atmos. Chem. Phys., 12, 9201–9219, doi:10.5194/acp-12-9201-2012, 2012.

Gustin, M. S., Huang, J., Miller, M. B., Peterson, C., Jaffe, D. A., Ambrose, J., Finley, B. D., Lyman, S. N., Call, K., Talbot, R., Feddersen, D., Mao, H., and Lindberg, S. E.: Do we understand what the mercury speciation instruments are actually measuring? Results of RAMI X, Environ. Sci. Technol., doi:10.1021/es3039104, 2013.

Harris, R. C., Rudd, J. W. M., Amyot, M., Babiarz, C. L., Beaty, K. G., Blanchfield, P. J., Bodaly, R. A., Branfireun, B. A., Gilmour, C. C., Graydon, J. A., Heyes, A., Hintelmann, H., Hurley, J. P., Kelly, C. A., Krabbenhoft, D. P., Lindberg, S. E., Mason, R. P., Paterson, M. J., Podemski, C. L., Robinson, A., Sandilands, K. A., Southworth, G. R., St. Louis, V. L., and Tate, M. T.: Whole-ecosystem study shows rapid fish-mercury response to changes in mercury deposition, P. Natl. Acad. Sci. USA, 104, 16586–16591, 2007.

Huang, J., Liu, Y., and Holsen, T. M.: Comparison between knife-edge and frisbee-shaped surrogate surfaces for making dry deposition measurements: wind tunnel experiments and computational fluid dynamics (CFD) modeling, Atmos. Environ., 45, 4213–4219, 2011.

Kellerhals, M., Beauchamp, S., Belzer, W., Blanchard, P., Froude, F., Harvey, B., McDonald, K., Pilote, M., Poissant, L., Puckett, K., Schroeder, B., Steffen, A., and Tordon, R.: Temporal and spatial variability of total gaseous mercury in Canada: results from the Canadian Atmospheric Mercury Measurement Network (CAMNet), Atmos. Environ., 37, 1002–1011, 2003.

Lai, S. O., Huang, J., Hopke, P. K., and Holsen, T. M.: An evaluation of direct measurement techniques for mercury dry deposition, Sci. Total. Environ., 409, 1320–1327, 2011.

Lamborg, C. H., Fitzgerald, W. F., Damman, A. W. H., Benoit, J. M., Balcom, P. H., and Engstrom, D. R.: Modern and historic atmospheric mercury fluxes in both hemispheres: global and regional mercury cycling implications, Global Biogeochem. Cy., 16, 1104, 2002.

ACPD
13, 10521–10546, 2013

Lan, X., Talbot, R., Castro, M., Perry, K., and Luke, W.: Seasonal and diurnal variations of atmospheric mercury across the US determined from AMNet monitoring data, Atmos. Chem. Phys., 12, 10569–10582, doi:10.5194/acp-12-10569-2012, 2012.

Landis, M. S., Stevens, R. K., Schaedlich, F., and Prestbo, E. M.: Development and characterization of an annular denuder methodology for the measurement of divalent inorganic reactive gaseous mercury in ambient air, Environ. Sci. Technol., 36, 3000–3009, 2002.

Lindberg, S. E., Brooks, S. B., Lin, C- J., Scott, K. J., Landis, M. S., Stevens, R. K., Goodsite, M., and Richter, A.: The dynamic oxidation of gaseous mercury in the Arctic atmosphere at polar sunrise, Environ. Sci. Technol. 36, 1245–1256, 2002.

Lyman, S. N. and Jaffe, D. A.: Formation and fate of oxidized mercury in the upper troposphere and lower stratosphere, Nature Geosci., 5, 114–117, 2011.

Lyman, S. N., Gustin, M. S., Prestbo, E. M., and Marsik, F. J.: Estimation of dry deposition of atmospheric mercury in Nevada by direct and indirect methods, Environ. Sci. Technol., 41, 1970–1976, 2007.

Lyman, S. N., Gustin, M. S., Prestbo, E. M., Kilner, P. I., Edgerton, E., and Hartsell, B.: Testing and application of surrogate surfaces for understanding potential gaseous oxidized mercury dry deposition, Environ. Sci. Technol., 43, 6235–6241, 2009.

Lyman, S. N., Jaffe, D. A., and Gustin, M. S.: Release of mercury halides from KCl denuders in the presence of ozone, Atmos. Chem. Phys., 10, 8197–8204, doi:10.5194/acp-10-8197-2010, 2010.

Mason, R. P., Abbott, M. L., Bodaly, R. A., Bullock Jr., O. R., Driscoll, C. T., Evers, D., Lindberg, S. E., Murray, M., and Swain, E. B.: Monitoring the response to changing mercury deposition, Environ. Sci. Technol., 39, 14A–22A, 2005.

National Atmospheric Deposition Program (NADP): Atmospheric Mercury Network Data Management Manual (2011–12) Version 1.8., NADP Program Office, 2204 Griffith Dr., Champaign, IL 61820, available at: http://nadp.isws.illinois.edu/amn/docs/AMNet_Data_Management_Manual.pdf, 2011a.

National Atmospheric Deposition Program (NADP): Atmospheric Mercury Network Operations Manual (2011–05) Version 1.0., NADP Program Office, 2204 Griffith Dr., Champaign, IL 61820, available at: http://nadp.isws.illinois.edu/amn/docs/AMNet_Operations_Manual.pdf, 2011b.

National Atmospheric Deposition Program (NADP): NADP's Litterfall Mercury Monitoring Initiative, NADP Program Office, Illinois State Water Survey, 2204 Griffith Dr., Champaign, IL

ACPD
13, 10521–10546, 2013

61820, available at: http://nadp.isws.illinois.edu/newIssues/litterfall/, last access: November 2012.
Nriagu, J. and Becker, C.: Volcanic emissions of mercury to the atmosphere: global and regional inventories, Sci. Total. Environ, 304, 3–12, 2003.
Obrist, D., Hallar, A., McCubbin, I., Stephens, B. B., and Rahn, T.: Atmospheric mercury concentrations at Storm Peak Laboratory in the Rocky Mountains: evidence for long-range transport from Asia, boundary layer contributions, and plant mercury uptake, Atmos. Environ., 42, 7579–7589, 2008.
Prestbo, E. M., Gay, D., Olson, M., Luke, W., Kelley, P., Felton, D., Holsen, T., Huang, J., and Choi, H.-D.: An analysis of co-located atmospheric mercury speciation data from AMNet (2009–2010). International Conference of Mercury as a Global Pollutant, Nova Scotia, Canada, available at: http://www.mercury2011.org, 2011.
Schmeltz, D., Evers, D. C., Driscoll, C. T., Artz, R., Cohen, M., Gay, D., Haeuber, R., Krabbenhoft, D. P, Mason, R., Morris, K., and Wiener, J. G.: MercNet: a national monitoring network to assess responses to changing mercury emissions in the United States, Ecotoxicology, 20, 1713–1725, 2011.
Seigneur, C., Vijayaraghavan, K., Lohman, K., Karamchandani, P., and Scott, C.: Global source attribution for mercury deposition in the United States, Environ. Sci. Technol., 38, 555–569, 2004.
Selin, N. E., Jacob, D. J., Yantosca, R. M., Strode, S., Jaeglé, L., and Sunderland, E. M.: Global 3-D land-ocean-atmosphere model for mercury: present-day versus preindustrial cycles and anthropogenic enrichment factors for deposition, Global Biogeochem. Cy. 22, GB2011, doi:10.1029/2007GB003040, 2008.
Sheu, G. R., Lin, N. H., Wang, J. L., Lee, C. T., Ou Yang, C. F., and Wang, S. H.: Temporal distribution and potential sources of atmospheric mercury measured at a high-elevation background station in Taiwan, Atmos. Environ., 44, 2393–2400, 2010.
Steffen, A., Scherz, T., Olson, M., and Gay, D. A.: Atmospheric mercury measurement data: quality control and assurance protocols for data standardization, J. Environ. Monitor., 14, 752–765, 2012.
Swartzendruber, P. C., Jaffe, D. A., Prestbo, E. M., Weiss-Penzias, P., Selin, N. E., Park, R., Jacob, D. J., Strode, S., and Jaeglé, L.: Observations of reactive gaseous mercury in the free troposphere at the Mount Bachelor Observatory, J. Geophys. Res., 111, D24301, doi:10.1029/2006JD007415, 2006.

ACPD
13, 10521–10546, 2013

Temme, C., Planchard, P., Steffen, A., Banic, C., Beauchamp, S., Poissant, L., Tordon, R., and Wiens, B.: Trend, seasonal and multivariate analysis study of total gaseous mercury data from the Canadian Atmospheric Mercury Measurement Network (CAMNet), Atmos. Environ., 41, 5423–5441, 2007.

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

Zhang, L., Blanchard, P., Gay, D. A., Prestbo, E. M., Risch, M. R., Johnson, D., Narayan, J., Zsolway, R., Holsen, T. M., Miller, E. K., Castro, M. S., Graydon, J. A., Louis, V. L. St., and Dalziel, J.: Estimation of speciated and total mercury dry deposition at monitoring locations in eastern and central North America, Atmos. Chem. Phys., 12, 4327–4340, doi:10.5194/acp-12-4327-2012, 2012a.

Zhang, L., Blanchard, P., Johnson, D., Dastoor, A., Ryzhkov, A., Lin, C. J., and Zhang, K. M.: Assessment of modeled mercury dry deposition over the Great Lakes region, Environ. Pollut., 161, 272–283, 2012b.

Zhang, Y., Jaeglé, L., van Donkelaar, A., Martin, R. V., Holmes, C. D., Amos, H. M., Wang, Q., Talbot, R., Artz, R., Brooks, S., Luke, W., Holsen, T. M., Felton, D., Miller, E. K., Perry, K. D., Schmeltz, D., Steffen, A., Tordon, R., Weiss-Penzias, P., and Zsolway, R.: Nested-grid simulation of mercury over North America, Atmos. Chem. Phys., 12, 6095–6111, doi:10.5194/acp-12-6095-2012, 2012c.

ACPD
13, 10521–10546, 2013

Table 1. Atmospheric Mercury Network site locations and general descriptions, as of January 2012.

NADP site ID	Site name	Latitude	Longitude	Elev. (m)	Inlet ht.	Operating agency start	Data availability end	Data availability condition	General site environment	Notes on surrounding
AL19	Birmingham	33.5530	−86.8148	177	4.0	ARA Inc.	1 Jan 2009	ongoing	urban	urban
CA48	Elkhorn Slough	36.8100	−121.7800	10	3.1	UC Santa Cruz	1 Jan 2010	31 Dec 2011	suburban	grass, lake
FL96	Pensacola	30.5500	−87.3753	44	5.0	ARA Inc.	1 Jan 2009	ongoing	rural	grass, open
GA40	Yorkville	33.9283	−85.0456	394	4.5	ARA Inc.	1 Jan 2009	ongoing	rural	grass, open
HI00	Mauna Loa	19.5362	−155.5761	3384	5.0	NOAA	1 Jan 2009	ongoing	rural	high elevation, open
MD08	Piney Reservoir	39.7054	−79.0126	761	3.1	Un. of Maryland	1 Jan 2009	ongoing	rural	Grass, mixed forest
MD96	Beltsville_B	39.0283	−76.8171	47	10.0	NOAA	1 Jan 2009	ongoing	urban/suburban	Forest
MD97	Beltsville	39.0283	−76.8171	47	10.0	NOAA	1 Jan 2009	ongoing	urban/suburban	Forest
MS12	Grand Bay NERR	30.4124	−88.4038	1	10.0	NOAA	1 Jan 2009	ongoing	rural	woody, wetland, shrub, forest
MS99	Grand Bay NERR_B	30.4124	−88.4038	1	10.0	NOAA	1 Jan 2009	ongoing	rural	woody, wetland, shrub, forest
NH06	Thompson Farm	43.1088	−70.9485	25	4.3	U New Hampshire	1 Jan 2009	29 Nov 2011	rural	mixed forest, crops
NJ05	Brigantine[a]	39.4649	−74.4488	8	4.0	St. of New Jersey	1 Jun 2009	ongoing	suburban	wetland, lake, forest
NJ30	New Brunswick[a]	40.4728	−74.4224	21	3.0	St. of New Jersey	1 Jan 2009	ongoing	urban	crop, forest
NJ32	Chester[a]	40.7876	−74.6764	276	1.0	St. of New Jersey	1 Jan 2009	ongoing	urban/suburban	forest, wetland
NJ54	Elizabeth Lab[a]	40.6415	−74.2085	5	3.0	St. of New Jersey	1 Jan 2009	ongoing	urban	urban
NS01	Kejimkujik	44.4321	−65.2031	158	5.0	Environment Canada	26 Jan 2009	ongoing	rural	forest
NU15	Alert[a]	82.4509	−62.5084	57	3.5	Environment Canada	affiliated site	affiliated site	rural	artic
NY06	New York City	40.8679	−73.8782	26	9.1	St. of New York	1 Jan 2009	ongoing	urban	urban
NY20	Huntington Wildlife Forest	43.9736	−74.2232	502	4.9	Clarkson U.	1 Jan 2009	ongoing	rural	forest, lake, wetland
NY43	Rochester	43.1544	−77.6160	154	4.3	Clarkson U.	1 Jan 2009	13 Nov 2009	suburban	urban
NY95	Rochester_B	43.1463	−77.5483	154	3.3	St. of New York	1 Jan 2009	ongoing	suburban	urban
OH02	Athens	39.3080	−82.1182	274	2.5	Ohio Un.	1 Jan 2009	1 Aug 2012	rural	forest, shrubs
OK99	Stillwell	35.7508	−94.6696	300	4.0	Cherokee Nation	1 Jan 2009	ongoing	rural	grass, forest
PA13	Allegheny Portage	40.4571	−78.5603	739	3.6	NOAA	1 Jan 2009	ongoing	rural	grass, forest
TW01	Mt. Lulin, Taiwan	23.5100	120.9200	2862	11.5	Taiwan EPA	1 Jan 2010	ongoing	rural	high elevation, open
UT96	Antelope Island	41.0885	−112.1187	1285	3.3	Un. of Utah	18 Jun 2009	30 Jun 2011	suburban	grass, crops
UT97	Salt Lake City	40.7118	111.9612	1099	8.2	St. of Utah	23 Nov 2008	ongoing	urban	urban
VT99	Underhill	44.5285	−72.8682	397	5.9	Ecosystems Res. Gr.	1 Jan 2009	ongoing	rural	grass, lake
WI99	Horicon	43.4557	−88.6169	272	4.0	St. of Wisconsin	1 Jan 2011	ongoing	rural	grass, lake
WV99	Canaan Valley Institute	39.1189	−79.4522	985	3.2	NOAA	1 Jan 2009	ongoing	rural	forest

[a] Affiliated sites

ACPD
13, 10521–10546, 2013

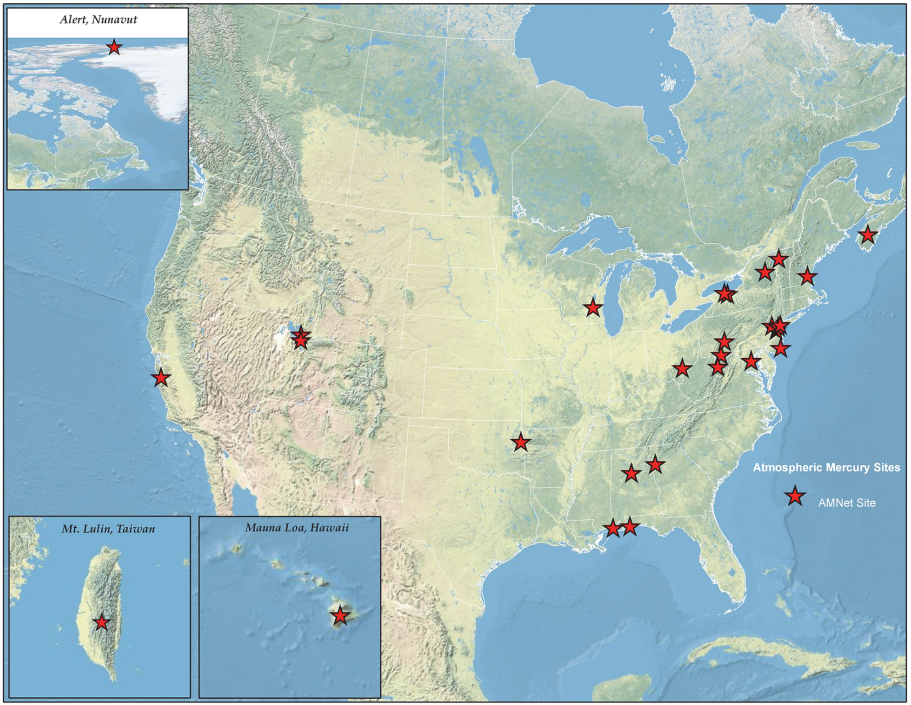
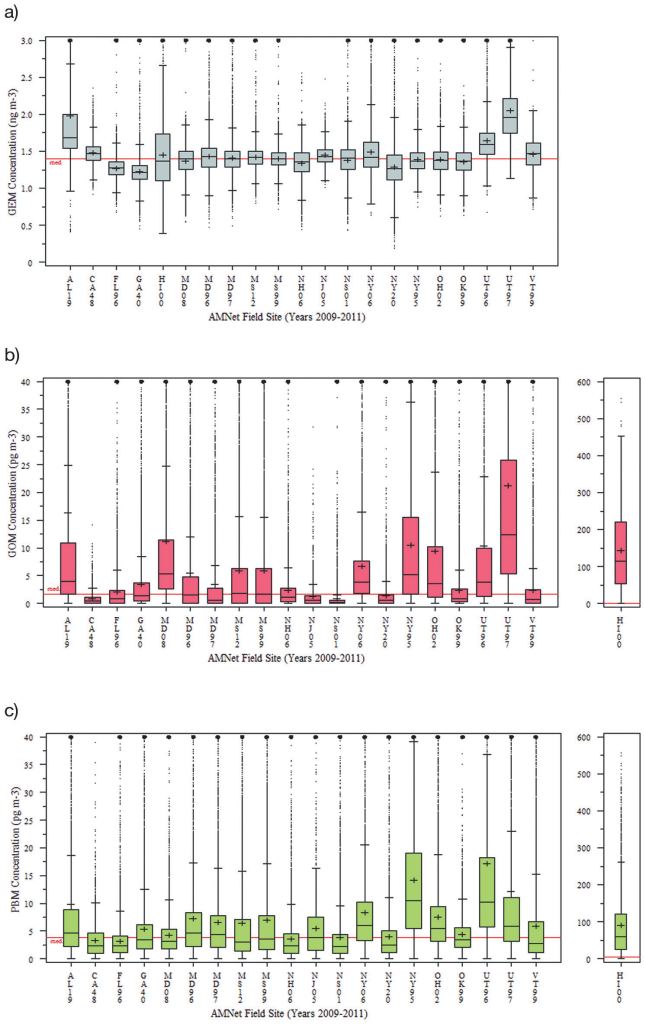


Fig. 1. Atmospheric Mercury Network sites, as of January 2012 (stars).

ACPD
13, 10521–10546, 2013



ACPD
13, 10521–10546, 2013

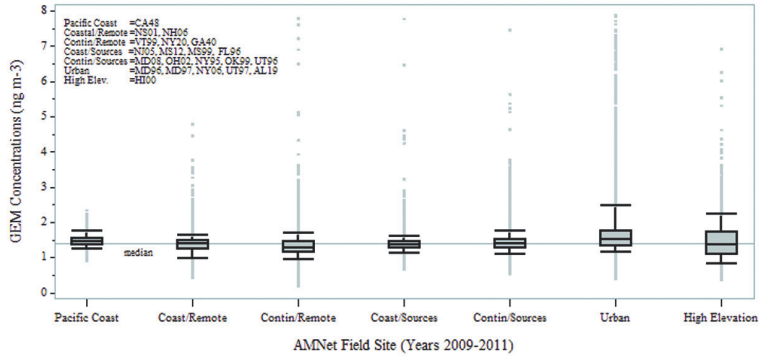
The Atmospheric Mercury Network

D. A. Gay et al.

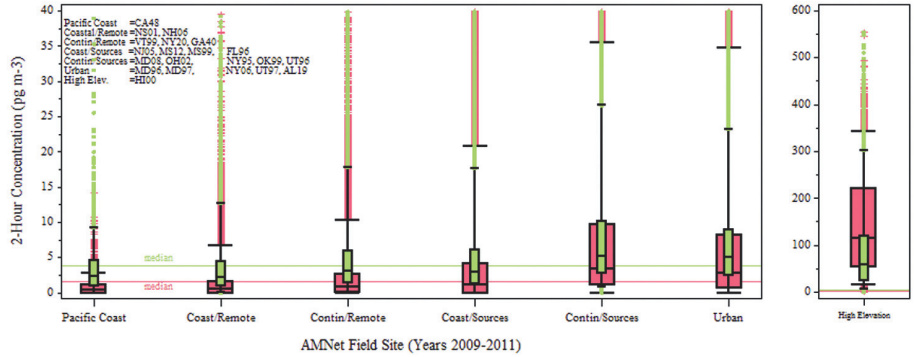
Fig. 2. Box and whisker plots of gaseous elemental mercury (GEM, $ng\,m^{-3}$) **(a)**, and for gaseous oxidized and particulate-bound mercury (GOM and $PBM_{2.5}$, $pg\,m^{-3}$) **(b)** and **(c)** observations for each site for years 2009 to 2011. Each box includes the median (midline), mean (+), 25th and 75th percentiles (box edges), 5th and 95th percentiles (whiskers), and individual values outside these limits (dots). Values above $3.0\,ng\,m^{-3}$ and $40\,pg\,m^{-3}$, respectively, are not shown (large dots). The respective medians of all observations are shown for reference (red lines, without HI00 for GOM and $PBM_{2.5}$).

ACPD
13, 10521–10546, 2013

a)



b)



ACPD
13, 10521–10546, 2013

The Atmospheric Mercury Network

D. A. Gay et al.

Fig. 3. Box and whisker plots of gaseous elemental mercury (GEM, ng m^{-3}) **(a)**, and for gaseous oxidized and particulate bound mercury (GOM in red, $PBM_{2.5}$ in green, pg m^{-3}) **(b)** observations for site groupings for years 2009 to 2011. Each box includes the median (midline), 25th and 75th percentiles (box edges), 5th and 95th percentiles (whiskers), and individual values outside these limits (dots/pluses). Values above 8.0 ng m^{-3} and 40 pg m^{-3}, respectively, are not shown. The respective medians of all observations are shown for reference (silver, red and green lines, without HI00 for GOM and $PBM_{2.5}$).

ACPD
13, 10521–10546, 2013