

Interactive comment on “The Atmospheric Mercury Network: measurement and initial examination of an ongoing atmospheric mercury record across North America” by D. A. Gay et al.

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1. ...However, I consider the section on initial inter-comparison of data between stations as weak, poorly referenced, and not ready for publication as is. For example, the authors highlight differences between various groups of stations (such as coastal versus continental, rural versus urban/pollution impacted), but they do not extensively discuss the reasons for such differences and don't provide references that show the various emission, deposition, and redox pathways that may lead to these observed patterns. I fully understand the providing a detailed discussion of the patterns at all stations is not within the scope of this publication and that such discussions have been

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and will continue to be done by individual site investigators; still, the currently presented highlights on differences between groups of stations lacks detail and discussions of the possible reasons for differences, and is very poorly referenced.

This work, in our collective opinion, had a two-fold purpose as noted in the title; both the 1) years-long development of a functioning network of mercury instruments and a consistently qualified and verified database of consistently quality assurance of mercury observations in the atmosphere, and 2) initial review of the measurements that only a network view of mercury fractions can provide, along with several initial observations of what we see within the data. We provide many specifics of how sites operate, how our quality assurance system was developed and operates (with documented references of the specifics), and many specifics from the observations, comparison graphics between the sites (showing mean, median, distributions, outliers, etc.). But we also attempted to keep the article a reasonable length, all the while keeping the article appealing to a more general audience following the ACP scope. We have agreed to several of the comments received, to increase the specificity of the observations (contained within multiple comments above and below). And specific to this comment, we have added a number of additional references to guide the reader to the potential causes of the observations presented here.

However, we still maintain that a very important point of this article is to present to the mercury community, a community that publishes within ACP quite often, the availability of a wealth of new data, all of which is very useful and accessible to ACP's wide audience. We have always considered this article primarily a catalyst for further research by others, as it was originally written. We agree that the article is somewhat different from a typical discovery article. However, we argue that it is appropriate and follows the ACP scope as written, by providing the overview of many field observations and summarizes the general conditions observed, and specifically points to areas where further research is needed.

2. I also miss an analysis of spatial patterns across the country in association with

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some of the model predictions published in the literature, such as vertical mixing of oxidized Hg in the western U.S. and other processes that are well known to control atmospheric Hg patterns.

3. I suggest to completely revise the sections on GEM, GOM, and PBM measurements and inter-comparisons between sites, including discussion of potential reasons for differences and much better referencing of previous work associated with this discussion. The main highlights of such a station inter-comparison and spatial patterns also should be provided in the abstract and in the summary.

Comments 3 & 4 answered together. We have added in several references, per this and other comments. We agree that this will guide the reader to some of the specific topics raised in the paper and by the observations. However, as we have discussed in previous responses, we do not feel that a complete rearrangement of this section is necessary.

4. Abstract: Page 10522, lines 20-30: We present atmospheric mercury data comparison by time at 22 unique site locations. What is unique about these 22 stations? Need to highlight and discuss in detail the inherent differences between various groups of stations in regards to GEM, GOM, and PBM patterns. Also should provide range, median/means, minimum and maximum values of observations.

The word unique was used to suggest that we had individual sites and instruments. We have changed the wording to "individual sites and instruments".

We have added summary GEM, GOM, and PBM2.5 statistical tables into the supplemental information for the paper. There are now five tables, each giving mean, st. dev., median, min/max, 5th, 25th, 75th, and 95th percentiles for: 1) all data all years and all sites; 2) all data all years and all sites, but without Hawaii given its unique situation; 3) all data and all sites without HI00 by year; 4) all years by site group; and 5) all years by site. We had assumed that the box and whisker plots would have provided graphics indications of these data, but we understand the reviewer's wish for more numerical

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observations.

5. Network Objectives: Page 10525, lines 19-24: This section is quite weak, I seem to recall much more examples of publications associated with individual stations, comparisons with model data, etc. Maybe a separate section should be added that provides current published and data products (publications, novel investigations, model constraints) that summarize the use of the AMNet program network and data for current scientific discussion of atmospheric Hg patterns.

The Network Objectives are as were stated to our initial scientists in the network, and as proposed to NADP for the development of the network. And perhaps we are misunderstanding the reviewer, but we do provide a listing of peer reviewed papers that have used our data (including references), and provided an idea of what was done with the network data (Page 10525, lines 20-25).

6. Monitoring locations: Page 10526 line 6: "but major gaps remain" clarify where major gaps exist, such as in the Western U.S. and high-elevation sites.

We have added into the text examples of where the gaps remain: "(central plains, Southwest, Pacific Coast, much of the West, interior Canada, Rocky Mountains and high elevations, etc.)".

7. Quality assurance and validation: lines 5 to 10: the authors discuss the utilization of 35 potential flags, and warning limits, but all of this is not very specific. I understand that the details of the QAP process has been published elsewhere (NADP 2011 and Steffen et al, 2012, but the authors should be more specific what the quality control involves.

We have added into the text a brief description of the flags used. "Flags include zero and span deviations, bias between the dual GEM responses, low voltage response and drift, calibration intervals and significant changes between calibrations, presence of multiple response peaks, and sample volume variation over time, etc."

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8. What really would be helpful is to provide a statistical and quantitative overview of the main reasons for data rejection - this only can be done using a large network with many stations, and we could learn about the causes for major quality issues that would benefit the many operators of this technique;

We thought this was a good idea, and added this paragraph to the manuscript at Page 10530 Line 3: "With the abundance of data and sites available, we can make some basic observations of the quality assurance system results in network operation of the Tekran speciating system. For 5-minute GEM observations, 80% of observations are valid with 10% excluded, leaving about 10% invalid observations for all years. For GOM and PBM2.5, 86% of all observations for each are valid, with only 14% invalid. The most frequent reason for invalidation for all fractions is low instrument response (5.2%, 6.5%, and 6.3% respectively). For GEM, the second most frequent invalidation is for calibration bias (at 1.3%). For GOM and PBM2.5 the second most important invalidation is for measurement cycles longer or shorter than normal (1.1% for each). These are typically power outages or maintenance where the instrument is stopped for some reason.

9. Line 11: "identifying anomalies": how is this done?

We added this specific list to the sentence: "identifying anomalies present "through manual review of timeline graphics and statistical distributions." "

10. Line 18: "performs annual site audits" is this correct, are there annual site visits to all sites? Or what do annual site audits include?.

Yes, we do provide annual site audits. The specifics are listed in lines 20-22 and in the SOP.

11. Line 23 to end: I hope the authors or Tekran system developers will follow up with a detailed response paper to Gustin et al., 2013 showing that many successful testing, inter-comparison, and field studies have been performed with the current Tekran

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speciation system.

The NADP AMNet is an open forum for atmospheric mercury experts to improve methods, data quality assurance and data analysis that is informed by new research studies. We agree that a detailed response to Gustin et al., 2013 would be helpful and hope that the reviewer participates in this effort.

12. Network observations and analysis: this is the weakest part of the paper, this section should be completely revised and should discuss in detail observations at the stations and within groups of stations, supported by the many publications that discuss sources, sinks, and transformation of atmospheric mercury.

This topic was addressed in the responses above. Again, the paper scope was purposely a high-level overview, with the expectation that this would catalyze further in-depth analysis and future contributions.

13. Line 26: "ranges from approximately 2500 to 11000 observations per site" they should provide a quantitative measure of data coverage per site, days of the year, or % coverage;

We have put into supplemental information a table detailing the distribution of all observations, by year, and by site. However, we would suggest that it remain in the supplemental, given the number of observations and therefore the length of the table.

14. Page 10530, line8: Faïn et al., 2009 (ACP) should also be mentioned here as well; This reference was added.

15. Lines 22-24: discuss reasons for urban GEM enhancements and reference studies showing urban emission r sources;

This would be a great topic for a stand-alone manuscript. All NADP-AMNet data is available to scientists for in-depth study, analysis and for model development.

16. Page 10531, lines 1 to 4: "we postulate that site elevation and local effects may

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explain the differences” how, explain what local effects and how elevation would play into this?

We tried to clarify the text to better explain the differences in surface exchange effects between the two contrasting sites VT99 and NY20.

17. Line 6: Minimum observations rarely went below 0.5 ng m⁻³” explain which stations showed lowest values, quantify the percent of observations below a certain threshold (e.g., 1 ng m⁻³), and discuss in detail reasons for low observations below global background with supporting references.

We have added a supplemental table detailing the distribution of all observations, by year, and by site. However, we would suggest that it remain in the supplemental, given the number of observations and therefore the length of the table. The frequency is also shown in the box and whisker plots.

18. Line 10 to 12: same for values above 3 ng m⁻³; which sites showed highest values, what are likely causes for these, and discuss in detail the pollution sources that have been reported for GEM in the literature.

We have added a supplemental table detailing the distribution of all observations, by year, and by site. However, we would suggest that it remain in the supplemental, given the number of observations and therefore the length of the table. The frequency is also shown in the box and whisker plots.

19. Lines 12 and following: the same needs to be for GOM and PBM, not a single references is given in this entire discussion part, discuss reasons, sources, sinks for GOM and PBM and how they relate to the groups of stations and/or spatial patterns of observations.

As mentioned above to similar reviewer comments, the paper’s scope was purposely a high-level overview, with the expectation that our limited analysis would help catalyze further in-depth analysis and future contributions.

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20. Page 10532, line 1 and lines 10-15: “mining sources” there are several studies that reported observations from the mining sites in eastern Nevada and observations in southern Idaho, these should be discussed in respect to the observations around Salt Lake city. Similar, there are studies from the Dead Sea which should be mentioned in lieu of GOM and PBM patterns observed at the Great Salt Lake;

We rewrote the paragraph, to include the comments by the reviewer, and added the reference: Obrist, D., Tas, E., Peleg, M., Matveev, V., Faïn, X., Asaf, D., & Luria, M.: Bromine-induced oxidation of mercury in the mid-latitude atmosphere. *Nature Geoscience* 4(1): 22-26, 2010.

21. Page 10532, line 8: “: : expected to have higher particulate levels”: there are good references out that discuss gas-particulate partitioning of Hg (e.g., Amos et al., 2012, and others). Generally, the ratios of GOM and PBM should be discussed, not only for this one station but for a multitude of stations/groups available in this network.

As we have suggested previously, the paper scope was purposely a high-level overview, with the expectation that this would catalyze further in-depth analysis and future contributions.

22. Page 10532 Line 20 to 24: “Definite conclusion based on the analysis of the group results are not recommended: : :.due to limited number of sites: : :. “ I don’t understand this, this study provides the best spatial, multi-station measurement record I have ever seen, the authors should highlight the unique opportunities of grouping data into different locations and spatial areas and discuss the major patterns. Clearly, there will always be uncertainties and some outliers that determine some different patterns within a certain group.

We agree that the network provides a wealth of data, and will allow for much further research. However, we believe that the network still lacks in upper air stations, Pacific Coast sites, Midwestern sites, etc., and therefore, the conclusions we are making are preliminary and that definite conclusions can only be made with further analysis,

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modeling studies and more monitoring.

23. Page 10533, lines 5 to 15: “: : in contrast to previous studies and model suggesting mid-day GOM production in the marine boundary layer: : :” I don't think the observations reported by the network are in contract to mid-day GOM production. It should be mentioned that GOM in the marine boundary layer is quickly taken up (and deposited) by sea salt particles (e.g., work by Holmes et al.). In addition, GEM shows some of the lowest values at coastal stations, in support of GEM oxidation in the marine boundary layer. These observations should be discussed in detail by other studies also reporting coastal Hg patterns (including Engle et al., 2010; Engle et al., 2008, Sigler et al., 2009, Mao et al.,2008, and others).

We agree. We have changed “previous” to “some” and added an additional reference and sentence to the paragraph.

24. Summary, page 10535, lines 6 to 20. Currently, the summary is very non-specific and weak. It does not give a summary of the manuscript, and it should be improved to summarize the network goals, network set-up, and the data provided in this initial overview of measurements.

We revised the summary and ending to be more consistent with the scope of the paper, and the comments here.

25. Figure 3. it is very difficult to see both patterns of GOM and PBM in one figure panel. I suggest to show these data in separate panels as has been done in Figure 2.

Although we agree that individual figures may be somewhat easier to see, we purposely superimposed the GOM and PBM figures onto one another to 1) reduce the figure number due to publishing limitations, and 2) so that it would also provide a figure to see the ratios between GOM and PBM without a new figure. We have left the figures as they are.

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Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/13/C6600/2013/acpd-13-C6600-2013-supplement.pdf>

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 10521, 2013.

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