

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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Anonymous Referee #2

1. This manuscript is an interesting initiative in order to introduce to a broader –and probably European- audience the work being done by the North American Atmospheric Mercury Network. An impressive data set is collected since 2009 and severe quality criteria are applied to the data. This manuscript is more an attempt of promoting and establishing the merits and benefits of such a network than a solid scientific discussion based on experimental facts and data.

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I am not sure that this manuscript in his current format is appropriate to publication in a journal such as ACP. To my opinion, it would be a good introduction to a special issue dedicated to AMNET, followed by other papers that are data scientifically sound and based on the analysis of the data.

We are not sure how to respond to this comment and the stated opinions by the reviewer. We purposely chose to offer a stand-alone paper and firmly believe we meet and exceed the publication criteria set by ACP (http://www.atmospheric-chemistry-and-physics.net/review/ms_evaluation_criteria.html). We think that access and communication of long-term, quality-assured field data is a critical and important contribution to atmospheric mercury science. The title and introduction clearly states that the contribution is not an exhaustive scientific analysis, but only an initial examination of the data, highlighting particular topics for future work. We expect our contribution to catalyze many more in-depth data analysis and modeling manuscripts to help advance scientific understanding.

2. The authors may decide what should be the scope of this paper. If the paper is an attempt to present the network only, then it should be reinforced by a better discussion on data quality and procedures, and by a discussion on the scientific strategy of data processing, modeling, and priorities. If the paper wishes to present some data, it has to be seriously improved. The analysis of data is very poor and would deserve a more comprehensive discussion.

Our goal was to balance the description of the network, the rigorous data quality procedures and provide an initial examination of the data. We feel our balanced approach is appropriate, and in keeping with the scope of the ACP. Where possible, the revised manuscript adds factual information about the data quality screening results. The reviewer does not give specific examples of where the data analysis is poor in this comment, so we cannot respond. The analysis of the data and discussion was limited by design and clearly indicated, so the authors believe that no major changes are necessary.

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3. While the introduction, objectives and needs for the network are clearly presented, I found it at times repetitive. For instance, the network objectives as presented in section 1.1 are similar to what is written in section 4. The introduction can lead to a misapprehension of the content of the paper as it is orientated towards important scientific issues: estimation of dry deposition and wet deposition. These two aspects are not dealt with in the following parts of the ms, but I think they would deserve a better discussion.

The reviewer makes a good point regarding repetition in section 1.1 and section 4, so we modified the paper accordingly. We do not understand why the reviewer experienced misapprehension, when the title, abstract and introduction do not suggest that the content would be focused on dry or wet deposition results. Thus, no expanded discussion on these topics is warranted in our opinion. We think the introduction provides the appropriate scientific backdrop and motivation for the development of AMNet and the results we present. However, we took the opportunity to slightly modify the introductory text to help further clarify that the paper is not about dry deposition estimates.

4. The section on quality assurance fails to present the artifacts on speciation measurements carried out with Tekran units. Authors start a sort of controversy that looks private. I feel it not appropriate, and I would better like a comprehensive list of artifacts and how they are handled or not within the network.

The authors believe that a discussion or list of artifacts is beyond the scope of this paper and deserves a focused paper on the topic. The paragraph discussing the possible artifacts suggested by Gustin et al. was added after reviewer comments requested it. The authors believe we presented factual information about the publically available Gustin et al. (2013) paper. It was important to the Authors to highlight a few of the many acknowledged and factual uncertainties of the experimental results presented in the Gustin et al. (2013) and contrast this with other uncertainties presented in similar studies. We did not believe that the Gustin et al. (2013) results meet the criteria for concluding that there was a measurement bias and thus are inconclusive. We have

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tried to re-craft the language in this section to be cleaner and more balanced. However, we also needed felt that a clear statement must be made that to assess the accuracy of a method, the assessment method must be proven to be artifact free and have an uncertainty and system accuracy that is much better than the method being evaluated.

5. Now, as said above, the scientific discussion on data is poor and under-referenced. I acknowledge that a clear attempt was made to discuss the data, but the analysis does not reach the standard of ACP publication. I admit that this is a huge task ahead due to the size of the data base, so I suggest to the authors to stick to a pure introductive paper and additional /companion papers should be focused on in-depth data analysis.

We agree that additional companion papers should be focused on in-depth data analysis and that this paper serves as a catalyst for such future activity. As noted previously, we offered this paper as a limited analysis of the observations, since much of the paper is dedicated to the basis for measurement and data quality procedures. It is common in science for quality manuscripts in quality peer-reviewed journal to focus on the descriptive rather than the interpretive as we have done. Examples include describing new methods, new research initiatives, new models and the like.

6. Abstract: isn't it 21 sites instead of 22 It is 21 instruments. We have changed the text to read "21 unique sites and instruments" in the abstract to reflect that two instruments are collocated.

7. Page 10523: Line 19-21: I would rather write "three measurable atmospheric Hg fractions", because other fractions that are not measured by speciation units contribute to Hg dry deposition as well. These fractions include particulate Hg with particles exceeding 2.5 μm , organic mercury and GOM that is not collected/or not proven to be collected by speciation units.

We agree and the change has been made.

8. Line 21: The impact of Hg dry deposition on ? Please clarify if it does impact

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the budget of global hg deposition or something else. Gustin et al also investigate source of GOM in the dry deposition. I am not sure that dry deposition is only of local origin since particles (biomass burning for instances) can travel on thousands of kilometers. I suggest reviewing more deeply the sources of Hg that could be involved in dry deposition. 9. Line 25: "GEM can be (...)" These sentence is odd –but I am not a native English speaker.

Both comments answered together. We agree with the first comment. We have added "to the total deposition budget" to the sentence. On the second point of comment 1 and the second comment, we have revised the last part of this paragraph as such, for clarity: "The impact of Hg dry deposition on the total Hg deposition budget can be substantial. Even though estimated GEM dry deposition rates are small relative to GOM and PBM2.5, 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; Gustin et al., 2012), is important in forested ecosystem deposition (Grigal, 2002; Ericksen et al., 2003), and can be transported over long distances before deposition occurs. In-situ oxidation of GEM to GOM/ PBM2.5 in the free troposphere has also been reported at high elevations in the U.S. (Swartzendruber et al., 2006)."

10. Table 1: Inlet height is given in meters probably. Please add this information in the table.

We added the inlet height unit '(m)' for meters to Table 1.

11. Is the site of Chester appropriate regarding inlet height? Does it comply with sitting criteria of the NADP SOPs?

The inlet height of 1 m is correct. The recommend inlet height range is 2 to 6 meters, but conformity is up to the site sponsor. Several AMNet sites were operating prior to the development of the network and siting criteria. Therefore AMNet reports all inlet

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heights allowing any data user to include or exclude individual sites and observations based on their own criteria.

12. Figure 1: The map is not clear enough. I wish to be able to locate the sites: you may include the name of the site or at least give a map with latitude and longitude. Also, you might precise by a color code, what are the sites used in the data analysis (21 sites)

We have added latitude and longitude indications to the basic figure and subfigure, changed the state and international borders and added the site IDs to the map.

13. Page 10527-28 About potential interferences with ozone. This paragraph looks polemic. Speciation units are the best fully automated instruments that are today available. There are artifacts that could be discussed in this section such as denuder efficiencies (what are the GOM species collected?), the quasi impossibility to compare parallel PBM measurements, the use of versatile sodalime (and what does it release and retain?), and even more importantly the calculation of detection and quantification limits for GOM and PBM. These artifacts could be discussed and if available should be fully referenced. Then, ozone interference might be a problem too, but I don't see the point to frontally criticize Gustin et al studies. It should be done in a critical letter. I agree (my own experience) that GEM should intercom pare very well between two co-located instruments (and that I do not understand why this paper was published with such a strong analytical problem), but this is not the place to refute this paper. Moreover, citing Prestbo et al 2011 as a reference is not appropriate since this is not a peer-reviewed article.

Refer to the response to the #4 review comment above.

14. P10528 Line 20: zero air is not a universal term. Either define it, or prefer a more meaningful term such Hg-free air or equivalent. While I acknowledge the authors to make this explanation short, I think it should more precise. You should precise that heating (pyrolysis) convert oxidized fraction of GOM and PBM into GEM that is

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subsequently released into the 2537.

We agree. We changed “zero air” to “Hg-free air”, and rewrote the sentence to reflect the precise pyrolysis action. The sentence now reads: “Following the two-hour sample period, the filter and denuder are heated, which converts the oxidized Hg in GOM and PBM2.5 to GEM. The GEM is released sequentially, into Hg-free air flowing to the Tekran 2537 in order to determine the Hg concentration of PBM2.5 and GOM, respectively.”

15. Line 25: precise that there are reported as standard (1 atm, 0°C) cubic meters. This is very important due to the differences in altitude in the network.

We agree, and added in the standard operation conditions for the measurements into the section 3.1 Measurements.

16. Page 10529 line 25 :are PBM considered here? If so, don't call it gaseous hg fraction

We have struck the word gaseous.

17. Page 10530 line 12: “Strikingly, the GOM and PBM2.5 median and mean values were five to ten times greater than any other site (Fig. 2b, c).” Is it really striking? H100 is closed to a volcano that emits oxidized Hg and a bunch of oxidant that may convert GEM. As said by the authors, it also received (probably) enhanced GOM and PBM from the high troposphere.

It is true, there are reasonable explanations for the H100 site results, however, in comparison to the other sites, we believe that the adjective “striking” is appropriate.

18. Line 15: this sentence does not contribute to a real scientific statement and discussion. It should be move to a more general discussion and a perspective section.

We have struck the sentence.

19. Line 23: The authors might try to give an answer. One straightforward thing is to
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check the proximity of strong sources such as combustible-using plants (power plant, incinerators, crematorium, and so on).

As stated above, the design of the paper is to be an introduction, without in-depth site-by-site analysis, yet still highlight areas for serious future inquiry. Thus, no changes were made.

20. Why do the authors expect that 15 of the 21 sites have mean GEMem between 1.3 and 1.5? I don't get it.

We removed the “expected” and added this sentence and 4 references that compare very favorably with our observations and support our statement: “These values are similar to the range observed in contemporary measurements of total gaseous Hg and GEM measurements at long-term sites of Mace Head, Ireland (Ebinghaus et al., 2011; Slemr et al., 2011), multiple sites in the Canadian Atmospheric Mercury Measurement Network (CAMNet; Temme et al., 2007), and a relatively new rural Germany site (Temme et al., 2013).”

With this change, we added these four references.

Ebinghaus, R., Jennings, S. G., Kock, H. H., Derwent, R. G., Manning, A. J., & Spain, T. G. (2011). Decreasing trends in total gaseous mercury observations in baseline air at Mace Head, Ireland from 1996 to 2009. *Atmospheric Environment*, 45(20), 3475-3480.

Slemr, F., Brunke, E. G., Ebinghaus, R., & Kuss, J. (2011). Worldwide trend of atmospheric mercury since 1995. *Atmospheric Chemistry and Physics*, 11(10): 4779-4787.

Temme, C., Blanchard, P., Steffen, A., Banic, C., Beauchamp, S., Poissant, L., Tordon, R. and Wiens, B. (2007). Trend, seasonal and multivariate analysis study of total gaseous mercury data from the Canadian atmospheric mercury measurement network (CAMNet). *Atmospheric Environment*, 41(26): 5423-5441.

Weigelt, A., Temme, C., Bieber, E., Schwerin, A., Schuetze, M., Ebinghaus, R., & Kock, H. H. (2013). Measurements of atmospheric mercury species at a German rural
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background site from 2009 to 2011—methods and results. *Environmental Chemistry* 10(2): 102-110 <http://dx.doi.org/10.1071/EN12107>.

21. Line 29: why not discussing GA 40 which is also rural with a mean below 1.3? NY20 has an odd 10.5 ng m³ in 2010 for a rural site. I don't see how the authors can state that NY20 and VT99 have contrasting GEM mean? They look pretty the same (according to the table, and given the analytical precision). The right discussion would be to discuss why these sites are lower than the rest. I don't understand what local high elevation causes onto NY20 and VT99 data set. The authors should elaborate more on this.

We compared NY20 and VT99 which have distinctive median differences (1.3 to 1.5) because they were so close to one another (100 km). We felt that this was a better comparison particularly due to their very close proximity.

22. Page 10531: "Year to year, the median change at the typical site was small and limited to 0.1 or 0.2 ngm⁻³." I don't understand this sentence. What is a typical site? Are you trying to talk about trend by pooling together the 3 years of data?

What we meant was "a typical site in this network". We have replaced "typical site" to "at these AMNet sites".

23. "The average GEM value does not appear to predict the number of extreme values." Yes, of course not! How could a mean predict something? I do not understand. What we meant was that sites with a higher GEM values did not have significantly higher numbers of extreme values, and vice versa. We replaced this sentence with: "Large numbers of extreme values are not limited to the sites with high GEM averages."

24. Page 10533: "Perhaps at these sites, the combination of relative remoteness and coastal locations both led to lower values." This does not sound like an in-depth analysis. Why does a coastal location bring lower GOM?

The network size data set generates many important questions as the reviewer notes.

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As stated, one purpose of the paper is to generate new questions and provide the quality assured data for future in depth analysis.

25. Page 10533 line 6: please provide a ref at least. There are ship-based studies that could not be compared to your data set. Moreover, short term campaign shouldn't be compared with a 3 year average. The 3 year mean may hide production of GOM that occurs during day hours (and maximize at noon) .

We added two references: Laurier, F.J.G., Mason, R.P., Whalin, L., and Kato, S.: Reactive gaseous mercury formation in the North Pacific Ocean's marine boundary layer: A potential role of halogen chemistry. *J. of Geophys. Res.*, 108 (D17), 4529, 2003; and Holmes, C. D., Jacob, D. J., Mason, R. P., and Jaffe, D. A.: Sources and deposition of reactive gaseous mercury in the marine atmosphere. *Atmos Environ* 43(14): 2278-2285, 2009.

26. Line 13: is there a significant difference in precipitation amounts between coastal sites and urban sites?

This comment is somewhat unclear. We didn't suggest that precipitation differences between urban and coastal groups are the only reason there are differences. Perhaps we were not clear enough in our explanation. We will add the word "enhanced" to the sentence.

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

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