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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Seasonal and diurnal variations of atmospheric mercury across the US determined from AMNet monitoring data" *by* X. Lan et al.

Anonymous Referee #2

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This manuscript by Lan et al. provides an overview of atmospheric mercury measurements over the US collected during the first 3 years of the AMNet monitoring program. The authors describe consistencies and differences among the sites, and attempt to provide mechanistic explanations. Since the diurnal and seasonal cycles of atmospheric mercury at many individual AMNet sites have already been documented, including in papers by the coauthors, the value of this paper is in characterizing the AMNet system.

The manuscript contains major problems with the flow of its argument, and occasional jumps to conclusions that are not supported by the evidence provided. In the para-

graphs below, I list the 3-4 most egregious examples. In addition, there are errors in grammar thoughout, such as subject-verb agreement, that impede understanding. The paper certainly requires major revisions, but given the number of changes required, I recommend rejecting the manuscript in its current form.

I refer to page numbers by their last 2 digits.

Discussion and interpretation of the Utah observation sites regularly causes problems throughout the paper. For example, Lan et al. write, "UT97 was heavily impacted by nearby anthropogenic emission sources" (P51), but then a few lines later they contradict themselves, saying, "The emission sources corresponding to UT97 were similar with UT96." The authors own Table 1 states that emissions near (<10km) from UT97 are 20,000 times larger than those near UT96, so the sources definitely aren't "similar". The next sentence suggests that "UT97 could also capture long-distance transported mercury, which may include mining and/or Asian emissions." There are at least 3 major problems with this. First, there's already a better explanation (local emissions) for the mean difference between UT96 and UT97. Second, it is highly unlikely that plumes transported over hundreds or thousands of kilometers would impact site UT97, but not site UT96, 30 miles away. Third, I am not aware of any study that has conclusively identified Asian anthropogenic pollution at ground level in the North America, besides mountaintops, so this claim would require strong evidence that is not provided.

Later, the authors attribute the diurnal cycle of GEM at UT97 to loss near the Great Salt Lake: "At UT97, the largest diurnal difference reached 55 ppqv, revealing the exceptionally large daily loss of GEM near the Great Salt Lake" (P58). As stated, this is unsupported speculation. I don't know what specific aspect of the diurnal amplitude leads the authors to implicate the lake, and it conflicts with previous work by Peterson and Gustin (2008), who did not find large GEM loss in the same area. I don't understand why the authors don't consider diurnal lake or mountain breezes as the cause, especially since they think, reasonably so, in my opinion, that lake breezes are important at other sites. On the next page, Lan et al. suggest that "photochemical oxi-

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dation of GEM was likely a dominant factor controlling GOM" (P59). Again, the authors have not considered diurnal wind variations, which could be very important in the midst of large industrial and urban sources. Obvious alternatives explanations need to be discussed before concluding that photo-oxidation is "likely" or "dominant."

On P55-56, the second and third paragraphs of this section state a lot of numbers without clear interpretations or significance. These could be condensed. Many explanations here are purely speculative, such as "the summer and fall GEM exhibited large interannual differences, which probably were the consequences of different annual meteorological conditions" (L8). Unless there is some additional evidence, these statements should be cut or, at the very least, clearly marked as speculations.

Revisions to this manuscript, should also address the possible role of mercury emissions from soil, water, and vegetation as a cause for differences between sites. Since the sites range from deserts, forests, lake shores, and ocean coasts, natural mercury emissions are likely quite different across AMNet. The authors do recognize the importance of diurnal uptake and release of GEM on moistened vegetation, but should do more. The 2008 UNEP mercury assessment and Bash (2010) provide good overviews.

The following comments list localized problems.

A recent of seasonal and diurnal cycles of mercury by Nair et al. (Atmos Environ, 2012) is clearly relevant to this manuscript; it should be referenced.

P47 L11. GEM reactions with O3, OH, and NO3 are all controversial, as is stated in one of the cited references (Calvert and Lindberg 2005). This should be noted.

P47 L16. Misleading."GOM and PBM. . . are thought to be readily deposited on the order of 1–7 days near emission sources." Atmospheric oxidation of GEM is an important source of GOM and PBM, so GOM and PBM are frequently not deposited near where their constituent mercury atoms were emitted.

P56 L18. "Higher GOM mixing ratios in spring and summer may be due to the in-

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creased length of the growing season at this time." I don't know what this means.

P56 L19. Subsidence from the free troposphere is an important source of GOM at some sites (Weiss-Penzias et al. 2009). This may also contribute to seasonal cycles.

P57 L9. The following paragraph contains many repetitions, such as listing the sites with strong GEM diurnal cycles.

P60 L16. This paragraph first repeats the 2 before it. Then I don't understand why MS12 specifically is described as having very low GOM at night when several other sites have equally low or lower GOM during the same hours (e.g. NY20, VT99, OH02. . .)

Table 1. Missing elevations. Emission units are not SI. Change to kg or explain in footnote. Emissions from a belt 10-50km or 50-150km from a site are not obviously relevant. It would be more helpful to list cumulative emissions within 10, 50, and 150km.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 10845, 2012.