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Interactive Comment

Interactive comment on "Successes and challenges of measuring and modeling atmospheric mercury at the part per quadrillion level: a critical review" *by* M. Sexauer Gustin et al.

Anonymous Referee #2

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This paper is a review of the state of atmospheric mercury measurements and the associated modeling, and the current challenges associated with both. The paper provides an initial review of the methods that have been used for measurements and then discusses some results that cast doubt on the validity of the results from the automated Tekran instrument which is the most widely used instrument for measuring Hg speciation in air. The review bases much of the discussion on recent papers by Gustin, Huang et al and I feel that the review pushes the point of view of the authors and maybe there could be more general discussion of the literature that is available.

More specific comments: 1) I concur with many of the comments posted by Slemr in

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his review.

2) One area I think could be better presented is a review of the historical literature on the measurements of Hg speciation in air, and perhaps some additional references could be included. There are a number of papers that could be referenced or talked about especially for surrogate surfaces. For example, there is no mention of the use of a water surrogate surface sampler - papers by Sakata and others - and other papers on surrogate surfaces are not mentioned - Caldwell et al 2006, some earlier papers by the Holsen and Keeler groups, the Marsik et al paper from the Everglades etc. This review paper has a very North American flavor and I think it would be good to focus as well on studies done in Asia and elsewhere.

3) The writing could be improved and the paper shortened as a result. There are numerous places where the writing is cumbersome, or there are minor errors. Some examples include: pg 3783, ln 15 "..been used to as a means to remove..."; equation R1 - kJ not kj; pg 3791, 7 "...provided a start for better understanding of..."; ln 22 "...different compounds containing GOM in air"; pg 3792, ln 2 "..biased low through spikes of GOM.."; pg 3800, ln 17 "Gom and PBm due to their solubility.." not all PBM is soluble.

4) A more detailed explanation early in the paper of the typical speciation in air would help the reader understand the paper better. There could also be some discussion, based on either measurements or modeling about the likely forms of GOM in the air. This is relevant to the results of the suitability of the various surfaces proposed for GOM collection. Also I do not like the use of RM for designating both GOM and PBM. The inherent assumption is that the PBM is reactive and soluble in deposition and we know this is not the case. Another term is needed besides RM.

5) pg 3781, ln 26. A bit more explanation of the statement of the controversy about "whether the 2537 measured TGM or GEM" would be useful here. Furthermore, more detail on the Univ of Houston system should be included in the body of the paper as

this is not well explained. This is also so for the discussion about the DOHGS - line 25, pg 3783.

6) Section 3.1. there is a lack of references to the statements in this section. What does "uncovered lines" mean? There is no references associated with the discussion of Maryland data and other numbers included in this section. Other papers besides Steffen could be referred to concerning depletion events. There are also specaition papers related to the Arctic that I do see mentioned in this paper. The sentence at the end about the requirement of a pyrolyzer is not explained or any reason given.

7) Section 3.2 would be improved with a better discussion of historical literature not just the recent papers. The comment that "evidence is coalescing.... vary seasonally and spatially" is not true. We have known about this variability for a long time. This is overstating the recent advances.

8) pg 3789, ln 23 "denuders loaded with HgCl2..." is this correct? More explanation is needed.

9) pg 3790, lines 20 onwards. There is no details of how the experiments were done, what the air concentrations were and were they realistic, and how the collection efficiency was determined. More details are required here.

10) It would be good to include some discussion and contrast the results of the lab studies and those of RAMIX (pg 3792). Also some explanation of the thermal desorption approach would help.

11) As noted above, a better discussion of surrogate surfaces that have been used is needed (pg 3793).

12) The potential explanation for the Cheeka Peak data (Section 4) is that the ocean boundary layer has more Hg0 because of ocean evasion. Additionally, I think that the detailed discussion, with lots of assumption and speculation in this section is totally unwarranted and not supported. I suggest removing this section. There is no validity for

deriving an equation from one location and thinking this could be applicable elsewhere. I also think that the idea of trying to come up with a way to convert Tekran data into what may be obtained by another methods is foolhardy based on current understanding. I do not agree this is possible given all the variability and differences in sources. I think the recommendations that refer to this are not sufficiently supported to be included in the manuscript. They should be removed.

13) The section on the MBL doesnt even reference any of the papers that have made measurements that the models were based on and this is not right. Include references to the data that Holmes et al and Hedgecock et al etc used for their modeling.

14) The last paragraph on pg 3800 and the discussion on the next page should be shortened - it is repetitive and somewhat obvious.

15) I do not agree with recommendations 2 and 5 (pg 3803) and think they should not be included in the paper. 1 6) More minor comments: i) pg 3779, ln 4 - generally I thought it was now agreed that the residence time was <1 yr; ii) ln 23 - I think there are good potential explanations, such as that put forward in Soerensen et al 2012 about the importance of evasion from the ocean - this could also explain the Cheeko Peak data as noted above; iii) remove comment about "a small city" (pg 3784, ln 12.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 3777, 2015.