

Review of MS No.: acp-2016-517: “Five-year records of Total Mercury Deposition flux at GMOS sites in the Northern and Southern Hemispheres”, by F. Sprovieri et al.

(2nd Review by Mark Cohen, Dec 10, 2016)

General Comments

Francesca Sprovieri and co-authors have prepared a new draft of the manuscript in which they have attempted to respond, as appropriate, to the concerns and suggestions of the three reviewers. In this 2nd review, I will primarily focus on the responses to items mentioned in my first review. In general, the authors appear to have comprehensively responded in thoughtful and constructive ways to my earlier comments, both in the point-by-point “narrative” response and in the changes made to the manuscript. This began as an important, well-written paper, representing a tremendous amount of work, and the authors’ have quite impressively improved the paper significantly. There are just a few relatively minor issues remaining that I would like to mention. None of these are “deal-breakers”, but the authors may wish to consider them in preparing the final version of the paper.

Specific Comments

- Perhaps the wet deposition flux calculation could be explained a little better (e.g., Page 6, lines 4-14). It still seems somewhat unclear to me. Given how fundamental these flux estimates are to the paper, I believe the paper could be improved by additional clarification and/or explanation.
 - First, the text makes it seem like the individual “Pⁱ” amounts are the precipitation amounts that occurred during each particular sample. However, based on the calculations I can infer from Tables S1 and S2, it seems that the annual wet dep flux for a given site is being calculated by multiplying the volume-weighted concentration estimated for days that the sampler was operated by the total precipitation continuously measured at the site over the entire year, i.e., including periods when the mercury sampler was not operating. If this is the case, then it should be made clear in the text and in the explanation of Tables S1 and S2. If this is not the case, and the Rainfall (mm) amounts in Tables S1 and S2 only represent the rainfall that was measured during the times that the Hg sampler was operating, then I don’t see how an estimate for the entire year could be made.
 - Second, the authors have added in text mentioning that the samples have been “normalized” with respect to a 15-day sampling time (in response to review questions about this issue). It’s not clear what is meant by this normalization. If it is too unwieldy to put in the manuscript text, the explanation could be added to the Supplement. I see in some later explanation that this new estimation methodology has changed some of the conclusions (e.g., in relation to GOM vs. wet deposition flux), and so all the more important to make sure it is very clear what this normalization process entailed. This will be useful for future work in the field, as this issue will probably continue to be something that has to be factored into the data analysis.

● Figure 2:

- Perhaps I have missed it in the text, but I believe it would be helpful to clarify in the caption or the text that the Rainfall (mm) is the total over the entire year, measured continuously, and not just the total for the days that the mercury sampler operated. This is really the same question that is mentioned in the earlier specific comment.
- How is frozen precipitation represented in these “Rainfall (mm)” values? An explanation of this could be added somewhere in the paper. Should the y-axis be labeled “Precipitation (mm)”? The term “Rainfall” is used throughout the document, but it seems unlikely that all of the precipitation at all of the sites would be liquid “rain”. I imagine that the frozen precipitation is melted for each sample and the totals expressed as “liquid rain equivalent”.
- In relation to these points, you wouldn’t necessarily have to change any of the Figures, Tables, or text, but perhaps you could add somewhere that “rainfall” and “precipitation” are used interchangeably throughout the paper, and the term “rainfall” includes all forms of precipitation. And you could also mention at some point – if this is indeed true -- that unless otherwise indicated, any “rainfall” or “precipitation” amount is the total measured continuously at a site, independent of whether the mercury sampler was operating at any given time at a site.

Technical Corrections and Suggestions

- Capitalization of words in the title seems inconsistent, e.g., “...Mercury wet Deposition flux...”. Could change to be consistent with whatever title capitalization conventions used are generally used in ACP.
- Page 2, lines 8-12: New sentence regarding dry deposition seems to be too long, and could potentially be reworded as follows:

“Currently, Hg dry deposition is often estimated by models, using measured ambient concentrations of Hg measurements and meteorological parameters, due to the lack of existing direct and accurate measurement methodologies (Gustin et al., 2012; Zhang et al., 2012). Therefore the investigation of Hg fluxes to terrestrial and aquatic surfaces in different parts of the world are often mainly based on performed by wet deposition measurements (Gratz et al., 2009; Feng et al., 2009).”

- Reference in new sentence about IVL-Bulk sampler unclear (page 4, line 26): “en, 2010”. Is “en” the last name of the author?
- Tables S1 and S2 – it is stated that “Measures in bold are related to the calculations based on a restricted number of sampling days, therefore statistically less representative than the others”. However, I don’t seem to see any bold vs. non-bold entries. If you do differentiate the entries as to significance, then expressing the less significant values as “bold” seems a little counterintuitive.