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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A 2 year record of atmospheric mercury species at a background Southern Hemisphere station on Amsterdam Island" *by* H. Angot et al.

Anonymous Referee #1

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Review of Angot et al. "A 2 year record of atmospheric mercury species at a background Southern Hemisphere station on Amsterdam Island"

Recommendation

Publish with minor revisions

General comments

Angot and co-authors present a 2-year record of speciated atmospheric Hg from a remote island in the southern Indian Ocean. The authors have characterized the behavior of Hg at Amsterdam Island using a comprehensive suite of ancillary observa-

tions and HYSPLIT back trajectories. The use of 222Rn is a nice touch. This helps fill in a large data gap and is a very welcome addition to the available measurements. The manuscript is generally well-written, easy to follow, and will be of interest to the community. I recommend the manuscript for publication after minor revisions.

A general area for improvement:

The work needs to include a more critical discussion of the limitations of the RGM and PBM measurements in the context of the identified sampling biases and artifacts (e.g., [Gustin et al., 2013; Huang et al., 2013; Lyman et al., 2010; Malcolm and Keeler, 2007; Talbot et al., 2011]). At this point, the weight of evidence is clear – RGM and PBM suffer from serious biases and interferences [Jaffe et al., 2014]. The authors acknowledge that the KCl denuder has problems with collection efficiency, but do not discuss what this means for reliability and interpretation of the Amsterdam Island dataset. There is probably meaningful information embedded in the RGM and PBM that can be defensibly interpreted (e.g., elevated PBM associated with African biomass burning), but not all aspects of the RGM and PBM data are reliable and these need to be more openly acknowledged. For example, the RAMIX intercomparison conclusively demonstrated Tekran RGM concentrations are biased very low [Gustin et al., 2013; Huang et al., 2013]. And Rutter et al. [2008], Talbot et al. [2011], and Malcolm and Keeler [2007] have all published studies suggesting PBM is biased as well. All of this work suggests we cannot trust the absolute magnitudes of Tekran RGM and PBM. This then places serious limitations on how useful RGM and PBM data are for model evaluation and development, so I'd like to see the authors be more thoughtful about how exactly they suggest modelers (or policy makers) use the Amsterdam Island data.

Specific comments

Title: Should there be a hyphen in "2-year"? If so, please correct here and elsewhere in the text.

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Lines 2-3: “Scarcity of mercury species records in the Southern Hemisphere is a critical weak point for the development of appropriate modeling and regulation scenarios.” It’s debatable whether or not lack of Hg speciation measurements in the SH is actually a “critical weak point” for modelers or regulators. For example, I would contend that uncertainty in the atmospheric chemistry or anthropogenic emission inventories are more serious weak points. The second half of the sentence is also not helpful because it’s unclear what you mean by “appropriate modeling and regulation scenarios”. I strongly suggest revising or replacing this sentence.

Lines 14-15: “Lowest concentrations of GEM” is grammatically incorrect.

Line 17: I suggest deleting “for further modeling studies”. First, it’s ambiguous what sort of modeling studies the authors refer to and, second, the Amsterdam Island don’t are helpful more than just models and so why limit yourself to just supporting model studies?

Line 23: “were” should be “have been” and “exposition” should be “exposure”.

Line 26: “However, research gaps for mercury control policies at regional or global scale still remain such as our understanding of mercury sources, atmospheric chemistry or deposition processes (Selin et al., 2007).” First, the grammar of this sentence needs to be corrected. Second, what’s being said isn’t helpful because it’s so general. What would be helpful is a more specific statement about what knowledge gaps are really limiting Hg regulations? You may find Selin [2011] or Selin [2014] helpful for ideas and references.

Page 14441:

Line 5: Ocean upwelling is not a primary source of Hg. Please delete.

Lines 3-8: The 10-30-60 partitioning is an original result from Amos et al. [2013] and not UNEP [2013].

Lines 19-22: “Nevertheless, the cycling of mercury at the global scale is not fully un-

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derstood and the role of the oceans still remains unclear, mainly due to the lack of long-term records of atmospheric mercury in the Southern Hemisphere (Pirrone et al., 2013)." This sentence needs to clarify that atmospheric observations place an important constraint on the global Hg cycle, but atmospheric measurements in the SH are not the largest uncertainty in ocean cycling. Uncertainties in air-sea exchange and carbon dynamics/particle scavenging play a much larger role in the ocean.

Page 14443:

Line 19: Please rephrase or delete "most relevant". Tekran GEM, RGM, and PBM are operationally defined and not objectively the most relevant forms to measure, rather they are the forms we can measure current instrumentation.

Page 14444:

Line 10: I'd recommend saying just "4 h" instead of "3 to 4 h" since you later say that the sampling period is 4 hours.

Line 22: Is checking the permeation source frequent enough? How does this compare to other long-term measurement sites, such as Alert?

Page 14445: Line 1: How does the GMOS QA/QC protocol compare to the protocols of AMNet/CAMNet? I'm just curious for the sake of intercomparison.

Lines 3-13: It's confusing to state the detection limits as "better than X". If possible, please provide the actual detection limit or your best estimate.

Lines 10-11: Landis et al. [2002] reported a 15% precision for RGM and PBM. Lyman et al. [2010] and Gustin et al. [2013] have suggest that RGM interferences vary with O3 and RH, which would suggest the precision reported in one sampling environment does not apply to sampling environments with different O3 and/or RH levels. Please provide a justification for why the Landis et al. [2002] can apply to RGM and PBM at Amsterdam Island.

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Page 14446:

Line 8: “till” should be “until”.

Section 3.2: It would be appropriate to include a citation to [Sprovieri et al., 2010] for their review on Southern Hemisphere atmospheric Hg data. Also, please provide a rationale for why you directly compare TGM and GEM. Lastly, I suggest providing the months instead of seasons. It will be easier for Northern Hemisphere readers (likely the bulk of your readership) to follow the text.

Page 14447:

Line 11: “did follow” should be “followed”.

Line 28: Should “this assumption” be “this observation”?

Page 14448: Is Amsterdam Island really representative of background “tropospheric conditions”? Does the marine boundary layer where you’re sampling really represent the free troposphere as well?

Page 14450:

Section 3.2.3: You might consider adding a table with summaries of the data (e.g., monthly mean, median, std dev, max, min, n) to make the Amsterdam Island more accessible for modelers. Alternatively, you could provide the QA/QC’d data online as an Excel spreadsheet in the SI.

Based on your analysis, can you comment on the existing estimates for biomass burning Hg emissions [Friedli et al., 2009; Holmes et al., 2010]? Are they consistent with what you’re finding?

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Line 6: Is it a good assumption everywhere that RGM can only be transported <100s kilometers? What about RGM formed in situ in the dry upper troposphere/lower strato-

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sphere where wet scavenging is infrequent?

Page 14452:

Lines 3-12: Please connect this paragraph more to RGM. It's relevance is not obvious. Also, please conclude with the take home message for RGM. It's not entirely clear what the bottom line is for RGM and what are the most important controlling variables at Amsterdam Island.

Line 14: Why would GEM adsorb to particles? Does that make sense based on its vapor pressure?

Line 16: Rutter and Schauer [2007] and Amos et al. [2012] should be included as references in addition to Steffen et al. [2014].

Section 3.3.2: Are RGM and PBM decoupled at Amsterdam Island? Is there any influence from marine sea salt?

Page 14453:

Lines 22-23: Please be more specific about how your work "opens the way for new avenues in future modeling studies".

Why are future efforts at Amsterdam Island focused on lower detection limits? Huang et al. [2013] suggests RGM is biased low by 1.3-3.7x. The detection limit issue is moot if what Huang et al. [2013] found is true at Amsterdam Island. Why go after the detection limit and not aim improve oxidized Hg measurements by addressing the biases, interferences, and lack of calibration?

Figure 3: Consider adding the month to the x-axis to help orient the reader.

Figure 4: The asterisk (*) to denote statistical significance looks like an outlier.

Figure 6: Please put dGEM under Rn 222 and align the x-axis. Then the reader can track the temporal evolution of the two together.

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Figure 8: You might consider merging this with Figure 5. It would be easier to see the relationship between fire counts and PBM. Also please define “events” in the caption.

Figure 9: Please provide a bit more of a description in the figure caption, so that the figure can stand alone from the text. Right now, if I was a reader scanning the figures I wouldn’t know why Figure 9 was important without digging into the main text.

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