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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Natural and anthropogenic atmospheric mercury in the European Arctic: a speciation study" *by* A. O. Steen et al.

Anonymous Referee #1

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Review of "Natural and anthropogenic atmospheric mercury in the European Arctic: a speciation study"

Steen, Berg, Dastoor, Durnford, Hole, and Pfaffhuber

General Comments

This is a valuable and an interesting contribution, and I hope that the editor will chose to accept this work for publication. There are some issues that I think need to be pursued further by the authors, but I think that the authors will be able to deal with them fairly easily.

My main comments are 1) The abstract of the paper communicates well the trends in the GEM, PHg and RGM species observed, but does not communicate well the authors' discussion of what factors control the relationship between RGM and PHg, or the seasonal variation in the relative importances of the BrO and ozone oxidation reactions.

2) Recent work which investigates the importance of the BrO reaction chemistry in marine environments vs the ozone oxidation reaction (Jacob, Harvard), and laboratory and field based gas-particle partitioning work (Schauer, U. Wisc-Madison) are not utilized.

3) The study does not adequately evaluate caveats associated with the Tekran. For example, the impact of ozone concentrations on RGM collection efficiency by the KCl coated denuder is not adequately addressed (Gustin, U. Nevada-Reno). Furthermore, the study does not discuss the consequences of keeping the particulate matter/PHg sample heated to 50oC during the collection period. This may be an important as some of the suggested species of PHg are semi-volatile, and RGM may become PHg by partitioning to airborne snow in the sample location being discussed.

Specific Comments

P27256 L9: Give RGM and PHg concentration summaries as ranges followed by a mean or median; as 8+/- 13 pg m-3 taken literally means that negative concentrations were observed, and is therefore distracting. L11: Seasonality of these species has been observed at other latitudes. Extend sentence to restrict this claim to the arctic. L11: Put the "For the complete. . ..<1%" sentence after the concentration summaries, and before the discussion of seasonality in concentrations". L14: "RGM was suggested as the precursor of PHg ..." The measurements in Fig 2 suggest that RGM may not always lead to the same concentrations of PHg from year to year which may cause some readers to questions this claim. This statement should be modified to acknowledge the uncertainties in our knowledge of how RGM and PHg are related in the environment. L16 Remove "surprisingly" L17-21 Suggest reversing discussion of BrO and ozone, or

at least state why you are discounting ozone as an oxidative source. It may be that both sources are important at different times of the year. It should be recoqnized that the Jacob (Harvard) modeling work which looks at marine RGM formation uses the Hall et al 1995 ozone rate coefficient, which is an order of magnitude slower than more recent measurements. This is not to say that the bromine reaction is not important and certainly the Jacob results remain a very important contribution, but the ozone oxidation reaction may be more important than the Jacob study might suggest. L21-23. Mention the evidence/analysis that brought you to this conclusion to give the conclusion more weight.

P27257 L9-10. Modify sentence by changing "to occur" to something like "to be an important influence on RGM and PHg concentrations". At present the sentence may suggest to some readers that partitioning will not occur if aerosol concentrations fall below a certain concentration, which is not scientifically accurate. L10-15. Since you compare the ozone and halogen oxidation pathways in the abstract, the ozone reaction studies should be added in this paragraph. I also suggest adding the aforementioned Holmes modeling study to give an idea of the relative importances of the halogen and ozone reactions. L15. Halogens may also be released by nitric acid replacement in sea salt aerosol.

P27258 L5-10. More recent work has shown that other factors also impact PHg and RGM distributions. See general comments for more details. L25. Ny-Alesund and Zeppelin names are used inconsistently between here and Fig 1b, as Ny-Alesund does not appear in Fig 1b itself as indicated by the text. To avoid confusion, perhaps present the mercury monitoring site as "Zeppelin" in the text and figure caption and then explain its geographic and functional relationship to Ny-Alesund.

P27260 L4 Explicitly state SCIAMACHY is a satellite for readers who are unfamiliar with BrO measurement methods. L9. Met data from Zeppelin should be used in parallel analyses as they may be different enough to Ny-Alesund to cause errors in interpretations. If the authors know that the differences between the met data at both sites

are negligible this should be explicitly stated, to assure the readers that this practice is acceptable. L15 Were ozone concentrations ever high enough to see reduced RGM collection efficiencies in the denuders as seen by Gustin (U. Nevada-Reno)?

P27261 L23. See comment about concentration summary ranges made in Abstract.

P27262 L26. Also prolonged darkness means that known GEM oxidants would be very low in concentration by the time spring insolation starts. You state this later in the paragraph, but I would bring first mention of it forward to here. What were ozone concentrations during the dark period? Are model predictions of BrO concentrations available during the polar night? If these are available you might consider mentioning them here.

P27263 Importance of radiation to RGM formation and the explanation of why large increases in RGM were observed in the absence of significant concentrations of PHg are valuable information which I think are under-represented in the abstract. Please also see comments made in General section about relationship between RGM and PHg, and limitations of using Tekran for understanding this relationship.

P27264 L12. Has this been demonstrated with models? What are feasible possibilities for the other 90% of the depleted GEM. It seems that there could be other possibilities beyond those the authors have presented. Having read a bit further on P27265 it seems as though you observe AMDEs which have occurred elsewhere, and much of the produced RGM and PHg has deposited before it reached the sampling site. Please explain this more clearly on P27264.

P27267 L15. As PHg concentrations were shown to be anti-correlated with temperature, were RGM concentrations positively correlated with temperature? Why is this analysis not shown in Fig 6?

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 27255, 2010.