

Interactive comment on “A synthesis of atmospheric mercury depletion event chemistry linking atmosphere, snow and water” by A. Steffen et al.

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This manuscript predominantly reviews the advances made in atmospheric mercury chemistry in polar regions over the past decade or so. It is well written and gives the reader a clear sense of the academic excitement following the discovery of AMDEs by Schroeder et al. in the mid 1990s. Indeed, many of the authors included in this review have played a large role in unravelling polar sunrise photochemical processes. The chemistry of atmospheric reactions is well described as is the potential significance of mercury deposition into fragile polar ecosystems. Clearly, the research in this topic has been a growth industry and Steffen et al. point out in a number of places impor-

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tant research remaining to be done before we understand the entire process linking atmospheric depletion to uptake in ecosystem. Having said that, I think this paper does have an Achilles heel - in particular, the link to water. The weakness of our understanding of how mercury enters water and what it does once there is not the fault of the authors, but if this paper is to claim in its title that it will link atmosphere, snow and water, there needs to be a better presentation of the difficulties with the water side including the problem of making aquatic measurements for trace Hg species. I say this especially because the research on AMDEs has been justified by reference to their potential ecological significance and, yet, we still seem not very far advanced in determining that significance despite the brilliant atmospheric chemistry that has been done since 1996. In a review written prior to the discovery of AMDEs, we proposed that there were two priorities in the context of Arctic contamination, one of which was Hg (Macdonald and Bewers, 1996). At that time, mercury was already established as a global contaminant with j or more of the Hg cycling in atmosphere/upper ocean due to human activities (e.g., see Mason et al., 1994). But the question for polar research was whether or not mercury deposition somehow favours polar locations thereby making the poles global mercury sinks. It seems clear that the unique deposition of Hg at polar sunrise is significant (50 to 300 tonnes in the Arctic is proposed), but is the net deposition into the water anywhere near that large (see Kirk et al., 2006), and how might that deposition be altered by change in ice climate? These questions are crucial and depend almost completely on water-column work. The authors present in Table 3 a fairly long list of mercury measurements in polar waters; the problem with this table is that 1) it presents no data (only detection limits); 2) it gives no evaluation of confidence in the data given the difficulties with mercury measurements and 3) most of the ocean data are for the North Atlantic, giving a false impression that much has been done in the Arctic Ocean. Actually, we are in a very poor position to say much about mercury geochemical cycling in the Arctic Ocean and I think that needs to be more clearly expressed. The second issue, allied with problem of Hg speciation and trends in polar oceans, is whether or not AMDEs lead to biological uptake. Scott (2001) has shown

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that some of the deposited Hg is bioavailable, but what is the efficiency of transfer from air to snow to water to biota? Without these answers, the AMDE remains an interesting academic phenomenon with unknown relevance to Hg trends found in wildlife (e.g., Lockhart et al., 2005; Stern and Macdonald, 2005) or humans (Johansen et al. 2004).

Specific comments P10841 L 25; the rich history of Hg measurements in snow, ice and water seems misleading to me. Certainly, where is the rich history of Hg measurements in polar waters? P10844-45L25-2. The arctic is actually quite well populated in some regions, and heavily industrialized in parts of the Siberian sector. P10845 L10-14. Although upper layer Hg enrichment in Arctic marine sediments is often taken to indicate industrial input, it may not always be so (Gobeil et al., 1999). P10846 L15-17. Hg and POPs deposition have a similar component - phase partitioning. Cold condensation is, accordingly, a bit of a misnomer as the POPs do not 'condense out' any more than Hg 'condenses out'; they both partition onto particles which then settle out in some manner. Some (perhaps most) of the Hg deposition is forced by the photochemistry, but Hg deposits in other parts of the world where there are no AMDEs. I would recommend rewriting this sentence a bit more carefully. P10866 L20-25. The importance of more long-term measurements of atmospheric Hg in the Arctic. No doubt more measurements would help to validate model and seek geographical variation, but is this the most important thing to do if one had money enough to establish more stations? It would help the reader if the various proposed research tasks could be ranked in order of importance with some rationale placing in context the understanding of risks to ecosystems posed by mercury. P10866 L25-26. I'm not sure what is meant by 'this information'. Assessments of fluxes and trends have been made already, so I presume they can be made. The question then is of confidence in those fluxes. P10869 L11-18. So what do these observations mean? P10869 L26. The use of the word solubility, which is usually defined as "A measure of how much of a given substance will dissolve in a liquid". Normally, one thinks of solubility at saturation, but I think what is meant here is not solubility, per se, but rather Henry's Law partitioning behaviour (or *K_{ow}*). This crops up elsewhere as well. P10872 L15-28. Are these 'net' depositions in an

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annual context? P10879 L24-27. I would agree with the first part of the sentence, but there is not much in the section detailing 'subsequent transfer to other compartments of the ecosystem.' P10881 L6 -10. And how did these models perform compared to DEHM? P10881 L12. Insoluble or unfavourable air to water partitioning? P10881 L11-20. What about deposition to water? Although deposition to water might involve RGM and PHg, what about the reverse flux; does that depend on Hg(0) and MHg? If so, is that flux controlled by the water side or the air side (e.g., Nightingale, 2003). It might help the discussion here to give a diagram showing a schematic of the fluxes. P10882 L5-10. It would seem appropriate to point out here that future efforts in modeling should focus on incorporating re-emission of Hg from snow and ice. P10883 L22. From the above comment, it seems to me that a current challenge for these models is to get the correct net deposition by properly incorporating the re-emission fluxes? The issue of transfer to ecosystems needs to be addressed by measurements at this point in time, more than by modeling, in my opinion. P10886 L20. Figure 6 seems to me to be un-necessary as the same information is presented, or could be presented, in Figure 3. Furthermore, the text describes some knowledge about methyl mercury which is represented in Figure 3 but not in Figure 6. P10893 L1-2. The question of Hg and MHg concentrations scaling with water flow is interesting and important for climate change; this deserves more text in this document (see also Stanley et al., 2002; Bishop et al, 1995) P10894 L9. Understanding the springtime pulse of MeHg is important from the perspective of marine mammal uptake, but the authors need to recommend the lines of research required to make that connection. Furthermore, climate change will likely have its greatest impact on MHg production through the organic carbon cycle, perhaps mediated through change in hydrological conditions - especially where such change causes submergence of soil. P10894 Section 6.4. This section is appallingly thin. For the Arctic, we can say that 1/3 of the area is covered by ocean; the margins of that ocean are where the AMDEs exert their influence because they need seawater halogens (influence extends only 25-75 km inland; P10877); the other 2/3 drainage basin in the Arctic feeds its material, including Hg, into the same margins of that ocean. And so,

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with respect to Hg deposition and ecosystem consequences, I would say that the ocean is quite a bit more than a 'considerable' portion of Polar Regions. The authors should also provide more specific guidance on the sorts of research required for oceans and their priorities in line with what they have provided elsewhere for atmospheric-snow components. I admit a fondness for oceans, but my point is this: the Arctic Ocean contains a large, vulnerable ecosystem living under the threat of very poorly understood risks from Hg. Those risks stem from 1) a potentially high net deposition of Hg via AMDEs, 2) Concentrating and methylating processes within the water column, sediments and ice and 3) climate change. Presently, AMDEs could deposit as much as 300 tonnes/yr of Hg to the surface; a lot of it might get into the water - or almost none of it (Kirk et al., 2006)). The science community needs better guidance than "more research be focussed on the cycling of Hg in and around these oceans." P10895 line 23. If a great leap has taken place, it is in atmospheric research on AMDEs; if there has been a great leap in understanding the cycle in the ocean or the passage of deposited Hg into ecosystems, it is not evident in this review. P10895 Section 7 Conclusions and future directions. Again, I would recommend prioritizing this research. What are the key research activities required to understand risks of Hg to polar ecosystems and humans?

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H. Elderfield); Chapter 6.03, Gases in Seawater, pp 49-81. Stanley, J.B., P.F. Schuster, M.M. Reddy, D.A. Roth, H.E. Taylor, and G.R. Aiken (2002) Mercury on the move during snowmelt in Vermont. *Eos, Transactions, American Geophysical Union*, 83 (5), 45, 47-48.2. Stern, G.A., and R.W. Macdonald (2005) Biogeographic provinces of total and methyl mercury in zooplankton and fish from the Beaufort and Chukchi Seas: Results from the SHEBA drift. *Environmental Science and Technology*, 39, 4707-4713.

Typos etc. P 10840 L 6 Much work has occurred but these processes are not yet P10844 L 17; typos/missing words P10853 L12; remove in the Page 10858 L7 remove and P10866 L3; they P10870 L26. How do you repeat a hypothesis? P10871 L8-11; rewrite for clarity P10871 L18-20; rewrite for clarity P10883 L4. Remove 'that' P 10886 L27 Rewrite for clarity. P10889 L12 loose should be lose. P10891 L 14. In its oxidized form, Hg is P 10895 L18. Insert and after oceans.

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