Reviewer comments for "Air-snowpack exchange of bromine, ozone and mercury in the springtime Arctic simulated by the 1-D model PHANTAS - Part 2: Mercury and its speciation"

The fate of mercury in polar regions remains largely uncertain. Toyota et al. present a model that incorporates some of the physical and chemical processes that are thought to affect mercury fate in the arctic. This work provides some new ideas about the factors leading to mercury depletion events, and highlights several areas where further field observations or laboratory measurements could significantly reduce uncertainties. I believe this work should be published after some issues are addressed.

Major comments

1. Some details of the model are unclear. Likely these are explained in the companion paper, but this needs to made clear in this manuscript. Some questions I had were: What is the specific surface area of the snow? What solute concentration is used in the model? These parameters will determine the liquid volume of the snowpack, and its thickness at snow grain surfaces, since the method of Cho et al. is used to determine LLL liquid volume in this model.

2. I am unclear as to why the authors choose to model the LLL as being interconnected between snow grains. They cite some previous studies as suggesting this, but I am not convinced that this is supported. For example, Rosenthal et al. (one of the papers cited by the authors) say that they saw impurity networks along grain boundaries only on snow that had been stored for 8 months and sublimated under high vacuum. In fresh snow and snow stored for two months, no such networks were observed. Recently, Domine et al. (2012) also reported seeing networks of impurities at grain boundaries only after significant sublimation of snow samples.

3. The authors use Henry's law to determine partitioning of mercury compounds between the atmosphere and the snowpack. I am curious as to whether Henry's law is a good way to model this. To my knowledge, the uptake of most species to ice occurs via adsorption (and is almost exclusively a surface process). Since Henry's law deals with bulk partitioning, it may not be an appropriate way to model partitioning between the atmosphere and snow surfaces.

4. Have the authors done sensitivity runs to determine how variations in LLL thickness will affect mercury partitioning between air and snow, and on how this these variations will affect its ultimate fate? This might justify the authors' decision not to account for effects of temperature and impurity concentrations on LLL thickness.

Minor comments

1. What volume does the SIA comprise in the model? What is the surface area to volume ratio of the SIA in the model? This is important to know since the model predicts different SIA and ambient gas-phase concentrations of several species, presumably due at least in part to the high surface area to volume of the SIA.

2. This model assumes that all solutes are excluded to the ice surface to form a liquid solution that is in contact with the atmosphere due to the interconnection of the LLL assumed in this model. However, not all solutes will be expelled to the surfaces of snow grains. A large fraction will be excluded into regions within the grains such as veins and pockets. The liquid solution in these regions will not come in contact with the atmosphere. Therefore this model may significantly overestimate the amount of "liquid" (or LLL) that will participate in multiphase chemistry. Finally, Domine et al. 2013 point out that even in fairly salty snow, there will not be enough liquid formed to wet snow surfaces. I do not expect models to present entirely physical descriptions of snow surfaces, but I think this issue should be acknowledged in the manuscript.

3. The authors state the following: "In this work, the e-folding depth for the attenuation is assumed to be 7.5 cm (King and Simpson, 2001; Peterson et al., 2002; Qiu et al., 2002; Simpson et al., 2002). We thus neglect the possibility of enhanced actinic fluxes that can occur near the top of the snowpack under certain conditions (Abbatt et al., 2012)."

   I do not see an issue with ignore potential actinic flux enhancements at snowpack surfaces, but I don't think the above discussion provides a justification for it. I recommend either removing these statements and simply stating that enhanced actinic fluxes were not considered, or providing a more relevant justification.

4. page 6 line 3: "...but the share of Br-atoms against that of BrO-radicals is uncertain owing to error bars associated with the unknown surface and third-body effects." Are the authors talking about the relative concentrations of Br and BrO? This should be more clearly worded.

References

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Rosenthal, W., Saleta, J., and Dozier, J.: *Scanning electron microscopy of impurity structures in snow*, Cold Reg. Sci. Technol., 47, 80–89, 2007.

Domine F, Bock, J., Voisin, D., Donaldson, D. J.: *Can we model snow photochemistry? Problems with current approaches*, J. Phys. Chem. A., 117, 4733-4749, 2013.