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# ***Interactive comment on “Air-snowpack exchange of bromine, ozone and mercury in the springtime Arctic simulated by the 1-D model PHANTAS – Part 1: In-snow bromine activation and its impact on ozone” by K. Toyota et al.***

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The authors describe the development and application of the 1D chemistry model PHANTAS to simulate chemical processes in the atmosphere-snow system. Simulations focus on conditions in the springtime in the Arctic aiming to represent regularly observed activation of reactive bromine species and the depletion of ozone and mercury in the arctic atmospheric boundary layer. The manuscript deals with the model set-up, the reactive halogen species and ozone, while the results for mercury are pre-

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sented in a second manuscript. For a full consideration of possible heterogeneous processes for the generation of bromine species, the model takes into account meteorological exchange processes in the atmosphere including aerosols and processes in the snowpack. In fact, the conceptual representation of the snowpack and aerosols is very similar in the model, however, with strongly different densities. State-of-the-art parameterizations for the vertical transport in the atmosphere and in the snow and for the chemical reaction mechanisms were implemented. Like in all previously published snow chemistry models, chemical reactions in the snowpack were limited to a defined volume with assumed properties similar to a liquid phase and called here Liquid-like layer (LLL) by the authors. This concept of LLL is based on a simplified parameterization of its volume as well as the hypothesis that all impurities in the snow are located in the LLL, thus, enabling the calculation of concentrations and reaction rates. Furthermore, the same chemical mechanism based on aqueous phase reactions is used for the aerosols as well as the LLL. The composition of the snowpack was assumed to be constant without any variation compared to the layered structure of a real snowpack. With the model, the authors performed a range of simulations demonstrating among others the importance of atmospheric conditions and the emissions of aldehydes like formaldehyde and acetaldehyde for the built-up of bromine species and the destruction of ozone. They further compared concentrations of reactive bromine species inside the interstitial air of the snowpack with atmospheric concentrations indicating strong enhancements inside the snowpack. Despite the limitations of the model it represents the current knowledge of chemical processes in the atmosphere-snow system. In contrast to the study of Thomas et al. which was focused on specific conditions observed at Summit, Greenland, the authors tried to represent average arctic conditions in their model. In their model the authors derived some important and interesting limitations for the activation of reactive bromine compounds using typical Arctic conditions that can be studied in new field studies. In my opinion, these results warrant the publication of the manuscript in ACP. I only suggest some minor changes and editorial corrections.

Minor comments:

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Page 20345, line 22ff: The destruction of ozone can occur during one day as observed by Jacobi et al. (2010).

Introduction, last paragraph: The authors decided to publish their study in two parts, which is probably reasonable taking into the account the length of a merged manuscript and the somewhat different audiences regarding ODEs and MDEs. Nevertheless, I believe that the authors can better describe the two parts. I think they should explain that they used the same model set-up with the same equations and mechanisms. These mechanisms included halogen, ozone, and mercury chemistry. The manuscript here forms part I and describes the model set-up in detail and the results regarding halogen and ozone chemistry. Part II then presents the results for mercury. I recommend that this information should be given here. I also would not call part II a “companion paper”. This sounds to me like an additional study and not like part II of the same study.

Page 20348, line 15ff and chapter 2.6: What is the basis for the assumption of an interconnected liquid phase that enables vertical transport in the snow? In chapter 2.6 the authors refer to Huthwelker et al., 2006; Domine et al., 2008; Gladich et al., 2011. However in Domine et al., 2008 such a liquid network is not mentioned. I am not convinced that the other references can justify such an assumption.

Chapter 2.10, 3. Paragraph: More recent observations of the chemical composition of the seasonal snowpack in the Arctic can be found in Jacobi et al. (J. Geophys. Res., 117, D00R13, doi:10.1029/2011JD016654, 2013)

Chapter 2.10, 4. Paragraph: The calculation of the pH of the LLL in the model is certainly a very crucial point and needs to be discussed. However, I am surprised to see that the authors begin the discussion with the precipitation of mirabilite as a major source of uncertainty, while later stating that some model runs showed no large impact. I actually would not expect no impact from the formation of mirabilite. Other factors (some are also mentioned by the authors) are probably more important like the aerosol deposition, the behavior of HCl and other volatile acids, the volume of the

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LLL fraction and the inclusion of all impurities in the LLL, the parameterization of the LLL as ideal solution and so on. Unfortunately, no field observations of the pH of the surface layer of the snow grains exist and the simulated values cannot be compared. Nevertheless, I recommend re-writing this paragraph to discuss the major uncertainties regarding the simulation of the pH.

Figures: In the current size, the colored contour plots in the printed version are almost useless because it is impossible to distinguish the details described in the text (which seems in fact to be a general feature of ACP articles). These details are only visible in the electronic version after enlarging the figure by a factor of 3 or 4. I recommend using larger contour plots in the printed version.

Editorial comments:

Page 20342, line 11: A common set of aqueous-phase reactions describes chemistry...

Page 20342, line 18: Delete: , in a conventional definition,

Page 20342, line 24: ...release to the atmosphere, ...

Page 20343, line 5: “during daytime” instead of “under sunlight”

Page 20347, line 22: ...gas and condensed phases...

Page 20348, line 1: ...gas and aqueous phases...

Page 20348, line 21: The snowpack is assumed...

Page 20348, line 25: Better  $0.508\text{cm}^3(\text{liquid}+\text{solid})\text{cm}^{-3}(\text{air})$

Page 20363, line 5 (and throughout the manuscript): What are “in-snow emissions”?

Page 20368, line 12: ...under calm weather condition...

Page 20378, line 9: ... on the ice surface, high values of  $v_d$  ...

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Page 20379, line 26: ... simulated BrO columns reach ...

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Interactive comment on Atmos. Chem. Phys. Discuss., 13, 20341, 2013.

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13, C5927–C5931, 2013

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