

Interactive comment on “Numerical Analysis of the Role of Snowpack in the Ozone Depletion Events during the Arctic Spring” by Le Cao et al.

Anonymous Referee #2

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This paper presents results from a model that contains a mixed boundary layer, and a multi-layer model of the snowpack, to pursue questions about the role of the snowpack in boundary layer ozone depletion in polar regions. The model presents some interesting results, in particular that the model is quite sensitive to the size of the snow grains, and the thickness of the liquid-like layer (LLL) on the snow grains, and that boundary layer ozone depletion is most sensitive to chemistry occurring on the near-surface layer of the snowpack. The model itself could, in principle with amendments, be a useful new tool for these members of the community, and perhaps others. However, the paper is not publishable in its present form, and major revisions would be necessary to make it so. At issue is the fact that the work is motivated by other models, informed almost entirely solely by those models, the conclusions are about the reality simulated by the model, and the conclusions refer to what work needs to be done to

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produce an improved model. The Introduction to the paper betrays a philosophy that mechanisms can be verified from models alone, even if those models are highly parameterized, and limited in scope. What is almost completely missing in this paper is a discussion of the fact that the model is nothing more than a mathematical representation of our understanding of some reality, and reflects that understanding, however poor that understanding may be. A model should be used to test that understanding, against observations. But the authors make very little use of any observational data, and because of this complete focus on the model and its reality, they fail to connect the model outcomes to any future observational needs, that might enable us to test hypotheses about the real atmospheric and cryospheric physics and dynamics that control the phenomenon at hand. The conclusions of the paper contain an eloquent set of examples of the issue I have with the paper, i.e. "we found that the diameter of snow grains, volume fraction of the LLL, and the uptake rate of HOBr by snow particles are the deterministic factors which critically control the occurrence of ODEs in the polar boundary layer." Well, no, these are the factors that are important in the representation of the PBL and snowpack processes in the model. They are not necessarily those that are important in the real world. There is a huge (known) difference between this model and reality, and the authors do not discuss these differences. Their conclusions are only valid if their model faithfully represents the relevant important processes in the environments under study, and does so well. But they really don't discuss much about what is known, other than that ozone decays at some observed rate in sunlit stable boundary layers (in which mass transfer is really poorly understood!). Let's take this sentence quoted above as an example. First of all, when they discuss the snow grain diameter used in the model, they only refer to other models, when in fact there is a large amount of observational information in the literature which they could have analyzed to assess the actual distribution of snow grain sizes. For example, the Domine group has published several papers (Domine et al., 2002, and Cabanes et al., 2002 for Alert, and Domine et al. 2012 for Barrow) on specific surface area of snowpacks (as a function of depth), along with the density of the snow, from which snow grain

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size could be estimated. But this paper does not refer to or draw on those very useful papers which are aimed at observations and understanding of reality. The next part of the sentence is about the LLL. However, it is important for the authors to read the Domine et al., J. Phys. Chem 2013 paper that discusses the idea that a uniform LLL around snow grains is not a physical possibility, and that snowpack photochemistry must be a very complex multi-phase process, and that the community is currently only able to simulate such processes through highly parameterized models that are under-constrained by observation of all the processes and by true understanding of even what phase in which the chemical processes are occurring. That paper suggests that much of the chemistry may be occurring through reactions of adsorbed species on a solid surface, the kinetics of which are not understood at all, or in aerosols embedded in the snow crystals. Unfortunately, the authors of this paper fail to recognize the existence of these concerns, and on this issue, again only refer to other models as sources of information. On the last part, it is discussed that the rate of Br₂ production is limited by the rate of HOBr uptake by snow surfaces. The paper fails to discuss that in the context of actual observations of HOBr (e.g. the Liao et al. paper which they did cite), nor does it discuss the state of knowledge of whether or not this chemistry is in fact the most important source of Br₂. For example, it has been discussed in the literature (e.g. Abbatt et al., 2012) that condensed phase OH radicals may be important oxidants for the production of Br₂. If the model simulated that, would the results and conclusions be different? A conclusion from this paper is that Br₂ cannot be produced at pH above 7. However, while the authors only refer to another modeling study on this conclusion, this fact has been discussed from observations in the Arctic (Pratt et al., 2013). The authors lack of awareness of the literature affects their ability to know what is a new piece of information worthy of their focus and reporting as a new result. The importance of chlorine chemistry has been discussed in another Liao et al. paper (2014), but chlorine chemistry is not discussed in this paper. It is known that chlorine chemistry produces HO₂, an important precursor to HOBr. I feel that this study, and the model construction, should be redeveloped after thorough familiarization with and

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study of the relevant observational literature, so that the model can be discussed in the context of an up-to-date discussion of what we actually know, and don't know, about this phenomenon.

Other more minor issues are:

1. The paper has not been carefully proofread.
2. Line 22 - I am not aware of any evidence of any kind that ODEs have "a great influence on the human beings". Line 29 - I am not aware of any proof that this process is a significant source of marine Hg.
3. Bottom page 2 - ODEs happen in spring with low solar elevation angles, in the thin layer of the atmosphere (surface to 400m). I am not aware of any radiative transfer calculations that show that ODEs are important to the energy balance in the Arctic.
4. Top page 4 - that the snowpack is important is a result of observations of reality, discussed in many papers in the literature, not just simulations of that reality.
5. Page 7 - the paper should discuss that it is using the resistance model for mass transfer between the atmosphere and the snowpack, and how well that has been shown to apply to stable polar boundary layers.
6. Line 4 on page 8 - the decrease of the Cl-/Br- ratio as described is not possible.
7. Page 13 line 11 - explain why aerosols in the Arctic would be assumed to be halogen-free? What impact does that have on the results?
8. Bottom page 17 - this discrepancy could be from 100 different reasons, like the absence of chlorine chemistry, the dilution of the snowpack air during sampling, less Br- in that snow, different pH in that snow, that the simulated chemistry is occurring in a liquid phase, and in reality it is a solid phase process, etc. etc.
9. Regarding the ozone gradient at the surface, the authors refer to Helmig et al., 2007, but that is about Summit, Greenland. What about Helmig et al. 2012?

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10. The sensitivity values (e.g. line 10, page 23) need a quantitative definition.

11. Middle page 23 - what do observations, e.g. Boylan et al., 2014, indicate?

12. The Conclusions are mostly a restatement of what is in the paper. What do the authors think we need a better fundamental understanding of, which could then be tested with observations, using an improved model? What observations would be helpful?

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