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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Synoptic-scale meteorological control on reactive bromine production and ozone depletion in the Arctic boundary layer: 3-D simulation with the GEM-AQ model" *by* K. Toyota et al.

Anonymous Referee #2

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The paper by Toyota et al reports on a 3D model study of meteorology and chemistry related to atmospheric bromine chemistry and boundary layer ozone depletions in the Arctic. The methodology involves use of a 3D model, GEM-AQ, that has simplified gas and aerosol phase bromine chemistry, to investigate a range of open questions about this complex polar phenomenon. The methodology employed is essentially state-of-the-art, albeit with some significant simplifications, and the paper is reasonably well-written, though it does needs some editorial review. However, while the title of the paper refers to synoptic meteorological control on boundary layer ODEs, the paper

covers issues, in some detail in each case, about: activation of bromide, blowing snow and release of halogens, total column BrO, free tropospheric background BrO, stratospheric BrO and "very short lived" organobromine compounds, the relationship between column BrO and tropopause height, the temperature dependence of the surface chemistry, MY vs FY sea ice activation, direct release of bromine from the snowpack, and, to a small extent, "meteorological control". This then points out a serious problem with the paper - it does not have a focus. I appreciate that a 3D model of a process as apparently complicated as Arctic halogen chemistry involves a range of processes that must be properly parameterized, but a single paper needs to have a scientific focus, addressing a manageable number of clearly phrased scientific questions/hypotheses. This paper appears (as implied in places in the text) to have started out with an aim to investigate meteorological control, but then ended up focusing on various other issues, e.g. ozone activation of bromide in the snowpack. I think the authors should completely rewrite this paper, starting out with some reasonable hypothesis, and then use the model to test that hypothesis. It does appear that this paper could be broken up into more than one paper, with some work. But I find that after reading it carefully, I don't see that I have learned much that is new, other than that ozone activation of bromide could well be a/the important bromine activation mechanism, and that this may well occur in the snowpack. But to some extent this is already "known". The paper could also focus on the blowing snow mechanism, with more focus on supporting boundary layer meteorological evidence. With a rewrite, and appropriate change of the title, this could be a very interesting paper. More minor comments/suggested corrections are listed below in the order that the issues arose in the manuscript.

1. Much of the Introduction reads like a (5 page) review, without clearly laying out the questions the paper will address.

2. Page 26213, line 14 - why is a pH below 6.5 "hardly attainable"? I believe they have frequently been measured to be below this. Deposition of sulfate/haze aerosol can do a good job of acidifying the surface snowpack.

3. Page 26216: I think the paper should explain the impacts of ignoring chlorine chemistry. Chlorine chemistry is well known to be important, e.g. in terms of accelerating BrO conversion to Br atoms via the BrO + ClO reaction, by producing HCHO from Cl+ CH4, etc. Could you come to inappropriate conclusions by ignoring chlorine chemistry?

4. I do not understand Reaction G132 in Table 1. While the text says that HBr deposition only provides Br- to the snowpack, and that HOBr and BrONO2 deposition results in oxidation to produce Br2, Table 1 shows Reaction G132 producing Br2. That seems quite inappropriate, assuming it is not a typo. Explain. 5. Page 26218, line 7. The only source of Br- on MY ice is HBr deposition? That seems unreasonable. After all, Br- is measurable on the surface of MY ice at all times. Can you justify this?

6. Bottom paragraph, page 26218. It makes little sense to me that on MY ice, Cl- is assumed to be "inexhaustible", but on the terrestrial snowpack, a x100 decrease means it is effectively gone? How would you reconcile that with measurements of Cl- in the terrestrial snowpack (cf. Simpson et al., 2005), and with the measurements of BrCl above the terrestrial snowpack, by Foster et al. (2001)?

7. Page 26219: The discussion of O3 reaction with Br- to produce Br2 should also cite Wren et al., JGR, 2010.

8. The top paragraph on page 26221 raises the question of how you control NOx, if you are not releasing it and HCHO from the surface. If you have no source of NOx, there would quickly be none with which to form BrONO2. Is it held constant? What are the initial conditions for the model, in terms of chemical concentrations?

9. Bottom of page 26223: to take an increase in R from 0.56 to 0.68 to imply that "the model realistically represents reactive bromine release from the surface snowpack" is just too big a leap. One could easily hypothesize that met. is not perfectly represented in the base model and that adding halogen chemistry fortuitously improves the agreement. This comment should be removed.

10. Top page 26224: of the places you are studying, Barrow may well have the most tractable meteorology. Why don’t you make use of the extensive ozonesonde data from Barrow? The data are readily accessible.

11. Starting with Figure 4, the Figures are not discussed in order. They should be referred to in order, or renumbered.

12. Page 26226, starting line 7: these points might be more clearly presented as a color-coded map of the Arctic showing the distribution of BL ozone.

13. The middle paragraph on page 26227 should cite Sjostedt et al. 2007, and their discussion of possible bromine chemistry at Summit. The paper should also state clearly why comparison of model output to measurements at Summit, which is very different from the other sites, is important.

14. Figure 8 can be removed, as not much is done with it.

15. Page 26230, line 17: you should explain this observation of improved simulation at higher T, as it is somewhat counterintuitive.

16. Page 26233, line 18, should cite Salawitch et al., 2010. However, the discussion of VSL bromocarbons should also discuss how these compounds get into the stratosphere. If they are actually VSL, their presence should be sporadic and spatially heterogeneous, related to deep convection and transport.

17. Page 26235, line 10: should an increase in R from 0.64 to 0.68 be regarded as significant?

18. Page 26236, line 13 should cite Fickert et al., 1999. Line 21 should cite Cho et al., 2002.

19. Page 26236, line 27: this last statement is inappropriate; this conclusion cannot be drawn from the information presented in this paper.

20. Page 26237, title line: the answer to this question was published by Foster et al.,

in 2001.

21. Page 26238, line 7: This conclusion was made in the 1997 paper by Impey et al., and should be cited.

22. Given the last paragraph in section 3, one again wonders what the focus of the paper should be, and what the new information is.

23. Page 26240, line 2: this statement is way too strong. You simply cannot prove that with a model study such as this.

24. Table 4: the R values do say one thing clearly - that halogen chemistry has little impact on the correlations, and that other factors (mixing?) are more important. This is not reflected in the content of the paper.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 26207, 2010.