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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.

K. Toyota et al.

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Received and published: 12 January 2014

Reply to Referee #2

We thank all the referees for providing useful comments and strong support to our manuscript. Here we reply to Referee #2 by answering each specific comment.

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Reply to major comments

Major Comment #1

Pg. 20345: It would seem appropriate to also mention recent 1-D modeling by Piot and von Glasow (2008, ACP), who showed ozone depletion within 1 day when recycling on the snow surface was included. The study by Piot and von Glasow represents an intermediate between the Lehrer et al and Thomas et al (and this study).

Reply:

Piot and von Glasow (2008) dealt with the fluxes (or recycling) of reactive bromine from the snow surface in a highly ad-hoc manner by not asking at all the type of chemical processes occurring in the snowpack, which we feel does not represent “an intermediate between the Lehrer et al. and Thomas et al. (and this study)” in describing what might be occurring inside the snowpack. However, quite a short timescale indicated from the Piot and von Glasow study is worthy of being mentioned as the referee suggests. In the introduction of our revised manuscript, we will refer to Piot and von Glasow as one of our motivation to study the timescale of simulated bromine release from the snowpack and subsequent ozone depletion.

Major Comment #2

Sec. 2.2: It is assumed that mercury reactions are included in this model, but that mercury is simply not discussed in this manuscript. Is that correct? Please clarify the text. Also, since several components of the chemical mechanism have changed, it would be useful to include a table in the supplemental information that shows all revisions to the Toyota et al 2004 mechanism. Otherwise, the vague descriptions in Sec. 2.2 do not provide sufficient information for comparison with the chemical mechanisms in other models.

Reply:

Yes, mercury chemistry is included in the model runs presented in this paper. In fact, the same model runs are re-used for the Part 2 paper. We will clarify this point in the

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introduction (as suggested by the referee Jacobi) and here in the section 2.2. In the supplement to a revised manuscript, we will include tables listing all the reactions in the present chemical mechanism.

Major Comment #3

Pg. 20349, Lines 21-23: Please clarify whether HCHO, CH₃CHO, and C₂H₂ were the only hydrocarbons included in the current model.

Reply:

In that sentence, we did not correctly state what we included in the present model, as we also accounted for the reactions of CH₄, C₂H₆, CH₃OOH, C₂H₅OOH, CH₃OH, etc. with OH-radical and Cl-atom. On the other hand, HCHO and CH₃CHO and C₂H₂ were indeed only VOCs considered as major Br-atom scavengers in this study, whereas the effect of C₂H₄ and C₃H₆ discussed in Toyota et al. (2004) was neglected. This point will be clarified by a full list of reactions to be included in the supplement to the revised manuscript, but we will also correct the sentence noted by the referee.

Major Comment #4

Pg. 20353, Line 22: Douglas et al 2012 discusses the chemistry of frost flowers. Perhaps a more appropriate reference would be Voisin et al. 2012 (JGR, "Carbonaceous species and humic like substances (HULIS) in Arctic snowpack during OASIS field campaign in Barrow").

Reply:

We will cite Voisin et al. in the revised manuscript.

Major Comment #5

Pg. 20353, Sec. 2.6: Please clarify this underlying assumption for this section. Is this saying that soluble species within the LLL on snow grains are physically transferred between snow grains in the snowpack?

Reply:

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Yes, soluble species in the LLL are assumed to be transferred physically between snow grains. Since its viability is debatable, we call this process “hypothetical”. In the revised manuscript, we will describe underlying assumption about this section – please refer to our reply to comment #3 from the referee Jacobi. Also, as mentioned in our reply to him, we have decided to revise and scale down this vertical diffusivity in the snowpack LLL network by a factor of 10 from that used for model runs in the ACPD version of our Part 1 & 2 papers, because the prescribed thickness of the LLL in our model scenario is as shallow as 0.556nm. The impact of this change is not fundamental for model results discussed in the Part 1 paper, but stated values will be adjusted accordingly.

Major Comment #6

Sec. 2.7: This section is extremely long (especially in comparison with the detail in other sections), and therefore, it may improve readability to perhaps move some of this to the supplementary information. As one example, the discussion of testing various stability functions and the reasoning behind using Cheng and Brutsaert (i.e. much of pg. 20355) could be moved to the supplemental information. Sec. 2.9 is also very long and could be moved partially to the supplemental material.

Reply:

In the revised manuscript, we will move these redundant items in Sections 2.7 and 2.9 to the supplement as suggested.

Major Comment #7

Table 1: This table may not be critical to the main text and could be moved to the supplementary material. The same could be true for Table 5 (particularly since fluxes are not given for the model-derived species, although this addition would be quite useful). Figures 2b, 3, and 4 could also be moved to the supplemental.

Reply:

In the revised manuscript, we will move Table 5 and Figures 3 and 4 to the supplement. We do not feel inclined to move Table 1 to the supplement, because the use of different

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diffusivity coefficients between gas and aqueous (aerosol or snowpack LLL) phases and between different model domains (FT, ABL and snowpack) was quite confusing to ourselves in the beginning and we believe the same for many readers. Also, we would like to keep Figure 2b on aerodynamic resistance in the main text, because it is crucial for discussion in Section 3.1 and 3.3.

Major Comment #8

Sec. 2.10: The lengthy discussion of the role of temperature could be condensed significantly, particularly since the main point is that role of temperature is not probed by this model.

Reply:

We will condense the discussion of temperature in the first and (very lengthy) fourth paragraphs in Sections 2.10 as suggested.

Major Comment #9

Table 3: The Cho et al. and Millero et al. manuscripts do not describe actual measured bulk snow chemistry, and this could be easily confused by a reader. Further, Krnavek et al. 2012 (Atmos. Environ.) provide chemistry data corresponding to nearly 1000 Arctic snow samples, and the median values do not agree with those shown in Table 3, as suggested in the third paragraph on page 20365. Further, the recent work by Pratt et al. (2013, Nat. Geosc.) suggests that the ratio of Br-/Cl- plays a role in Br2 activation; this is also supported by laboratory studies of HOBr uptake and Br2 release (Huff & Abbatt 2002 (J. Phys. Chem. A), Adams et al. 2002 (ACP)). If the snow chemistry values shown in Table 3 are to be used and presented in this model study, then the discussion in Sec. 2.10 should be revised to state that these are not necessarily typical values.

Reply:

We appreciate the referee for pointing out issues with our choice of snowpack composition. Unfortunately, we could not perform new model runs on the basis of suggested references within the given time frame of manuscript revision. Hence we will simply

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state that halide concentrations taken from Cho et al. and Millero et al. do not necessarily represent median values from actual measurements of bulk snow chemistry such as Krnavek et al.

Major Comment #10

Sections 3.1 and 3.2: These sections are well-written with excellent discussion and represent important contributions to our understanding of bromine chemistry. With that said, the last paragraph in Section 3.2 is not very informative to the main points of the manuscript and is suggested to be moved to the supplemental information since the reason for the chemical solver crash is unknown. This discussion detracts from the important scientific discussion and results of Section 3.2. The last sentence of the paragraph could simply be moved up and integrated into the first paragraph on page 20375.

Reply:
We will revise as suggested.

Major Comment #11

Section 3.4: This section contains a significant amount of introductory material that could be moved to the introduction. In fact, the introduction could be revised slightly to provide adequate introduction to the main themes of the results and discussion section; this would also provide the reader with the appropriate context to understand the significance of the model results.

Reply:
The introductory part in this section will be shortened and moved to Section 1 (Introduction).

Major Comment #12

Page 20380, Lines 15-17: It could be confusing to the reader that “deliquesced seasalt aerosols” are mentioned here (and elsewhere), given that only sulfate aerosols are actually considered in this particular model exercise.

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Reply:

In addition to the sentence pointed out here, similar sentences in Abstract and Section 2.2 may indeed sound confusing. In the revised manuscript, we will stop using the phrase “deliquesced sea-salt aerosols” in the sentences in the abstract and the conclusion and simply stress that a common set of multiphase chemical mechanism was employed in the atmosphere and in the snowpack. In Section 2.2 (chemical mechanism description), we will remind readers more explicitly of the fact that sea-salt aerosols (as a source of halogens) were not simulated in our model.

Reply to minor suggestions

Minor Suggestion #1

Pg. 20342, Lines 13-17: Long, awkwardly worded sentence.

Reply:

We will split and modify the sentence to make it sound more sophisticated.

Minor suggestion #2

Page 20347, Line 8: As a comment (but not something that needs to be changed in this manuscript), recent work by Kwok et al. (2011, JGR) provides data from recent IceBridge snow depth studies.

Reply:

We appreciate the referee for this information. Although not cited at this time in our paper, we will take it into consideration for possible follow-up studies.

Minor suggestion #3

Page 20366: There is discussion of results included here in the methods section, which does not seem appropriate.

Reply:

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In the revised manuscript, discussion of results in regard to mirabilite will be moved completely to the supplement. For the results on pH in the LLL, we will move and expand the discussion by creating a new subsection in Section 3 (Results and discussion).

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

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