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Interactive comment on "Snow-sourced bromine and its implications for polar tropospheric ozone" *by* Xin Yang et al.

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Response to Referee #1:

We thank you for the very constructive comments on the manuscript. Our responses to each of the questions are shown below.

Page 8142, line 19: Am I right that the recycling reactions only occur on background aerosol and not on the additional aerosol generated from blowing snow? If so, the authors might like to add a comment somewhere that the additional aerosol from blowing snow will further enhance the recycling.

Yes, you are right. In the current study, the recycling reactions only occur on background aerosol. We have add one sentence in the revised version to address 'Note

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that in the polar boundary layer the additional sea salt aerosol from blowing snow will further enhance the recycling (R1-R3) and may cause much higher BrO fraction in total inorganic bromine species; this is not included in this study.'.

Page 8145/8146 and Figure 4: I can see why it's tempting to make comparisons at specific locations, especially ones where ODEs and BEs are regularly observed. But I wonder if it's asking too much of the experiment to do regressions on these? The comparison at Barrow works fairly well and r2 is (surprisingly) good. But there are clearly issues with comparisons at many of the locations shown in Figure 4, where neither the agreement in timing nor magnitude of BrO are particularly well captured. This isn't really surprising given that a model grid box is being compared with a satellite pixel, and that the system is highly dynamic and sensitive to a wind speed threshold that might be smoothed out over an averaged grid box. The most important point I take from these diagrams is that the Ocean run doesn't get close to giving bromine explosions, whereas the blowing snow run really does. I'd take that as the conclusion, and not push into regression analyses for these other locations, unless more discussion is given as to why differences in detail (and hence reduced r2) can be expected.

We have added one sentence in the conclusion section to address: 'Our simulations show that the open ocean and organic bromine source are not sufficient to explain the bromine explosions (BEs), whereas the bromine sourced from blowing snow is.'

In the revised paper, we keep figure 4 instead of removing it. According to your suggestion, we explored the possible reasons why the correlation is poor at some sites such as Arrival Height and Neumayer. In the revised paper, we say: 'Unlike in the NH all multiyear sea ice has been ruled out as sea salt source. However, the large Ross ice shelf near Arrival Height is a sea salt and Br source, explaining why the model overestimates BrO at Arrival Height during spring. The poor correlation at Neumayer could be due to the coarse model resolution. Noteworthy are several short-term BE episodes that are consistently captured by both measurement and model results. It is of course unrealistic to expect excellent correlations at all sites, given the coarse model resolution. More importantly, it is important to realize that, in all cases, the elevated BrO in polar spring cannot be simulated without including the blowing-snow source of bromine.'

Page 8146, line 3: "during Julian days 80-90, and 120-130" – or is there a reason why the latter is not mentioned?

No. In the revised version we say: 'Comparison with GOME data shows that the model successfully reproduces the big bromine explosion events during Julian days 57-60, 82-91 and 122-127, but not for Julian days 73-75.'

Page 8146, around line 15: Neumayer and Arrival Heights (4e and f) are not inland stations, they are coastal sites, Summit, on the other hand (4b) is in central Greenland, and Harestua (4d) is some considerable distance from the coast.

Agree.

Page 8147, line 20: Figure 6 shows that emissions of Br associated with blowing snow contribute significantly to BrO at mid-latitudes. This is an important finding and should be included in the Conclusions section.

We have added it into the abstract and conclusion section.

Page 8159, Figure 4: It's noticeable that in the Arctic, the model consistently underestimates GOME trop column throughout the non-spring periods of the year. Do you have any suggestions as to why this might be?

It could be due to the following reasons: 1) there are more open oceanic sea salt contribution in the SH than in the Arctic (as the Antarctic is 100% surrounded by open ocean); 2) the mean surface wind speeds in the Arctic could be much smaller than that over the Southern Ocean in the SH; 3) the bromine depletion factor (DF) used for sea salt aerosol could be much different between the two hemispheres (DF is likely acidity related). However, in this study, we did not consider this effect; 4) GOME trop column has its own uncertainty. An overestimation of the tropo BrO column by the GOME retrieval technique can not be totally ruled out.

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Page 8147, line 13: do the differences in BrO distribution reflect different meteorology in the two polar regions?

Yes. Frequent storms occurred over the Southern Ocean mean much stronger vertical transport of boundary layer tracers than in the Arctic. So we say in the revised version: 'reflecting dynamical process in controlling vertical transport of BrO.'

Page 8147, line 18: do the different percentages arise because of the different salinities between the Arctic and Antarctic used in the model?

Yes, you are right. In the ArcticX1 run, the snow-sourced contribution over the Arctic is significantly lower than that in the base run (40-70% in the ArcticX1 run vs. 70-80% in the base run). So in section 4, we add one sentence to say'..., which is mainly due to the higher snow salinity used in the Arctic'. In section 5, we say: 'Compared with the OCEAN run, the snow contribution (from ArcticX1 run) to the total column BrO over Arctic is still significant (40-70%).'

Page 8149, line 10: Blowing snow events are never dry. Within blowing snow, relative humidity will be 100%. What matters, in terms of climatic influence, is the windiness.

Within the blowing-snow layer (\sim 10m), the RH will reach 100% soon after the start of the blowing snow. Here what we mean is the environmental relative humidity (nor RH within blowing snow, but the background RH). In the revised version, we say 'A relatively dry and windy environmental condition favors a large sublimation rate during a blowing event.'

Please clarify this in the text.

Page 8158, Fig 3: given the discussion in the text about wind speed at Barrow, it might be interesting to see wind speed data for Barrow compared with that in the model for the spring period.

We thank reviewer #1 for raising this interesting question. One supplementary figure is added (shown as figure 1 in this response) to address the relationship between surface

wind and BrO at Barrow. The updated conclusion is:

'Compared to the observed 10m wind speeds at Barrow airport (data from National Climatic Data Center), model daily mean surface wind speeds (with horizontal resolution of 5.6°) are on average 1 m/s smaller (supplementary figure 1). Through a detailed comparison with tropospheric column BrO data as shown in the supplementary figure 1, we find that, during most periods, strong surface winds correspond to elevated BrO at Barrow, but not all elevated BrO is accompanied by windy condition. For example, during Julian days 58-60, 89-91 and 122-127, big BEs are captured by both satellite and simulation, but surface wind speeds are very low indicating non-local sources and long-range transport of lifted bromine in the free troposphere'

Page 8158, Fig 3: As the largest BE occurs between days 50-60, it would be nice to see GOME trop column data for this period as well – are there any?

The GOME trop column data has now been extended to cover the big BE during Julian days50-60 (shown as figure 2 in this response). The modelled BrO well matches the GOME data for this event. Also note is that the TOTAL GOME BrO (black line) in the figure has been updated by using the updated data (the old one was calculated based on air mass factors calculated based on a stratospheric BrO profile rather than a profile with having contribution in both troposphere and stratosphere).

Page 8160, Figure 4: Could you define what you mean by "over Weddell Sea" and "over Ross Sea"? Are the data averages across these regions..?

The data shown in figure 4g and h are not representing the average across the Weddell Sea and Ross sea. The GOME tropospheric BrO data at the given centre location as shown in figure 4g and 4h are representing averaged satellite BrO within a range of 200km. The model data are just for the corresponding gridbox BrO.

Technical comments

Page 8138, line 13: Amend title to "Main features of the blowing snow scheme" Page

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8142, line 15 and 16: can you provide a reference for the uptake coefficients used? The title of section 2.2 has been changed.

The uptake coefficients used in this study are actually based on our own estimations, thus no references are cited. In the text, we say: 'The uptake coefficients used in this study are estimated to be \ldots '

Page 8143, line 12: "Earth" should be upper case Page 8144, line 21: "boreal" and "austral" should be lower case (adjectives)

Changed

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Fig. 1. supplementary figure 1: Observed 10 m daily mean wind speeds at Barrow airport (data from National Climatic Data Center) during Julian days 40-150, 1998, and model daily mean surface wind speeds.

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Fig. 2. updated figure 1 in the manuscript