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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Horizontal and vertical structure of reactive bromine events probed by bromine monoxide MAX-DOAS spectroscopy" *by* William R. Simpson et al.

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We thank Reviewer 1 for helpful comments. In this response, the reviewer comments are included in plain text and our responses are in bold text. Line numbers refer to the ACPD version of the manuscript.

This paper presents data from 3 MAX-DOAS instruments deployed in 2012 as part of the BROMEX experiment. The paper comprises an important contribution to our knowledge of halogen activation and ozone depletion in the Arctic and should be accepted after some minor comments are addressed.

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Discussion paper

We appreciate the positive words on the manuscript.

Minor comments:

Abstract, L32 – Please define more clearly sharp edge. Figure 1 caption – Please define "cloud streets"

Wording was clarified as "sharp" meaning "smaller than 30km horizontal length scale". Cloud streets are horizontal convective rolls associated with airmass motion from over ice to over open water.

Figure 2 – This is the only figure that I cannot fully understand. In the 3rd panel, what are the flat red and blue lines? Were wind directions so consistent or is this an instrument problem? Were their wind direction measurements on the IL1 and IL2 buoys? This is not clear for me.

These red and blue lines represent the direction between the IL1 and IL2 buoys from the BARC site, which was not made clear in the text, so we modified the caption to be more informative.

L312, Section 5.2 – This section is titled "Snowpack-Based BrO events. . ." Does this refer to snow on sea-ice based events?

Both sea ice and land at this time of year are covered with snowpack, and past literature has demonstrated that "shallow" (large fraction of BrO in the lowest 200m) events are associated with recycling of reactive halogens on snowpack surfaces. For the most part, these events come from sea ice regions, but we do not explicitly separate snow on sea ice from snow on land.

L337 – It would be nice to reference some additional work on bromine activation, including studies on aerosols at warmer temperatures. There is some evidence that there are interfacial/dark reactions that are also important. Two examples include:

- Hunt, S. W., et al. "Formation of molecular bromine from the reaction of ozone with

deliquesced NaBr aerosol: evidence for interface chemistry." The Journal of Physical Chemistry A 108.52 (2004): 11559-11572.

- Oum, K. W., M. J. Lakin, and B. J. Finlayson-Pitts. "Bromine activation in the troposphere by the dark reaction of O3 with seawater ice." Geophysical Research Letters 25.21 (1998): 3923-3926.

These citations are useful and have been added, but the section at line 337 is specifically about the pH dependence of reaction 3, so we didn't add them at line 337, but instead at line 324 . Note that these processes are also heterogeneous processes, requiring either ice (snowpack) or aerosol surfaces, so they are similar to reaction 3 listed.

L376 – There is a relevant study on HOBr uptake that should be mentioned here: Roberts, Tjarda J., et al. "Re-evaluating the reactive uptake of HOBr in the troposphere with implications for the marine boundary layer and volcanic plumes." Atmospheric Chemistry and Physics 14.20 (2014): 11185-11199.

The reference was added.

L393 – Sentence that starts with "As discussed by. . ." Please consider rephrasing as this sentence reads a bit strange.

We reworded this sentence.

L423 – I have two comments on this paragraph.

1. Can't other aerosols besides sea-salt also be formed/released from open leads? Is sea-salt the only potentially important aerosol surface that can be contributing here?

2. Aerosol extinction is lower at IL2 than the other sites. Can't this just be a limitation of the ability of the MAX-DOAS to measure aerosols aloft (higher than 1.5 km or so)? Given that the lead results in increased mixing, is it really there are less aerosols or are aerosols just diluted and mixed out of the region where they can be measured reliably?

Yes, potentially other particles could be released from an open lead. For example, particles derived from the sea surface microlayer could be produced. Thus, the wording was broadened. The reduced aerosol extinction at the downwind site is not likely due to technical problems with MAX-DOAS in this case. Specifically, the aerosol extinction profiles (Fig. 7) show that the aerosol is not highly lofted. Additionally, as AOD becomes lower (e.g. the better the visibility), MAX-DOAS becomes more able to see higher in the atmosphere.

L452 – What does it mean to "deepen the BrO"?

We clarified "BrO layer thickness"

L458 – Please rephrase the sentence that begins "Thus, in-situ. . ." to be more specific

Clarified to mean surface observations (e.g. ground-based CIMS or MAX-DOAS)

L461 – The paper from Yang et al. (2010) and Theys et al. (2011) show good agreement with satellite observations in the Antarctic, but the model/satellite measurement comparison is less good in the Arctic. In addition, the study from Jones et al. (2009) primarily focuses on the Antarctic. Therefore, I think it's important to point out that the conclusion that "high winds" increase tropospheric BrO may be somewhat Antarctic specific. There may be different mechanisms that dominate in the Arctic because it's in general less stormy and more stable. I would add a reference to Theys et al. (2011) here as well:

Theys, N., et al. "Global observations of tropospheric BrO columns using GOME-2 satellite data." Atmospheric Chemistry and Physics 11.4 (2011): 1791.

This bi-polar difference in wind speed regime was added to the section.

Conclusions – I find it necessary to add a paragraph on what this means for future model studies/developments. Some examples of past work to mention include:

- Toyota, K., et al. "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." Atmospheric Chemistry and Physics 14.8 (2014): 4101-4133.

- Toyota, K., et al. "Analysis of reactive bromine production and ozone depletion in the Arctic boundary layer using 3-D simulations with GEM-AQ: inference from synoptic-scale patterns." Atmospheric Chemistry and Physics 11.8 (2011): 3949.

- Holmes, Christopher D., Daniel J. Jacob, and Xin Yang. "Global lifetime of elemental mercury against oxidation by atomic bromine in the free troposphere." Geophysical Research Letters 33.20 (2006).

Certainly, there is a need for modeling of these data; these citations and other modeling efforts were added.

L506 – Missing period at the end of the paragraph.

Fixed.

Figures 4-6 – Consider using a different color than green so that it's easier to distinguish the green and blue curves. The dots are a bit big to see small differences in the measurements.

The green was chosen as a progression from red-green-blue, so we didn't change that color. Instead, we changed the symbol for the green lines to have an open symbol for better differentiation from the blue line. We reduced the size of the dots to allow small differences to be seen.

Figures 7 8 – Consider combining into one large paneled figure so that the BrO and aerosol profiles can be viewed together.

We combined these figures to make a single very large one (new Fig. 7).

Figure 7 – Please comment in the text on the lower panel Mar 23 – What do the measurements mean above the black region? Are these real aerosol measurements

or is everything above the black portion unreliable?

This is a good point. The aerosol measurements above an optically thick layer are not reliable (you cannot see through the thick layer). The text was modified.

Figure 9 – Please mark on figure the upwind and downwind measurements for ease of understanding.

Upwind and downwind are marked.

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