

***Interactive comment on “Overview of the 2007 and 2008 campaigns conducted as part of the Greenland Summit Halogen- $\text{HO}_x$  Experiment (GSHOX)” by J. L. Thomas et al.***

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Response to interactive comment on “Overview of the 2007 and 2008 campaigns conducted as part of the Greenland Summit Halogen- $\text{HO}_x$  Experiment (GSHOX) by J. L. Thomas et al. Anonymous Referee #1

COMMENT: The paper by Thomas et al is an overview paper for the GSHOX project that took place at Summit Greenland in the late spring to early summers of 2007 and 2008. The paper is well written and succinct, as it should be. It does a good job of portraying what is exciting about this set of papers. Specifically, to me, the most important result is the implied presence of a significant reservoir of Br(y) in the free

C8381

troposphere, which is providing a bromide loading to the surface snowpack at Summit. This observation/conclusion should spur additional experiments (which I think really should involve focused aircraft flights from the surface in the Arctic basin, to the free troposphere, aimed at speciating Br(y) compounds, and testing the hypothesis that Br(y) is transported from the Arctic basin surface into the free troposphere, and from there, dry and wet deposition can occur to the Greenland ice sheet. I think the paper could play up the significance of this conclusion, and better cite other papers that discuss this issue, e.g. those from the satellite column BrO community, which has been agonizing over this issue for years. Other than this, I have only minor comments, which are listed below in the order they appeared in the manuscript.

- We thank the reviewer for the helpful comments, the specific responses to each specific comment below.

COMMENT: 1. Motivation section - it might be useful to summarize why we care about this chemistry, i.e. where else you find bromine in significant enough concentrations, and how in general bromine might perturb radical cycles, e.g. those that determine global background O<sub>3</sub>.

- This is a good suggestion and a set of equations that summarize bromine chemistry has been added to the Motivation section to address this point. To facilitate this discussion we have also changed order of the figures.

COMMENT: 2. Page 17139, line 1, might say "BrO efflux from the snow...", and transport, not transported. Line 2 - I don't think it should be "MBL", should it be BL? Or do you mean to say from the surface of the Arctic Ocean? This sentence is a bit confusing.

- We are discussing the flux bromine out of the snow at Summit. The suggested corrections have been made to clarify this point.

COMMENT: 3. Line 22 on page 17139 is a non-sentence.

- The original sentence: "Table 1 in Liao et al. (2011) summarizes the full suite of

C8382

measurements that were made, additional details are provided in other papers in this special issue.”

Has been changed to: “Table 1 in Liao et al. (2011) summarizes the full suite of measurements that were made during the campaign and additional measurement details are provided in the other papers in this special issue.”

COMMENT: 4. Line 27 - is it a bit misleading to say it is highly correlated with J(Br<sub>2</sub>)? I suspect this means it is correlated with radiation in general. If so, the presence of RGM is "indirect" evidence at best!

- This is a good point. This sentence has been changed from: “Mid-day peaks of RGM in excess of 200 pg m<sup>-3</sup> were highly correlated with JBr<sub>2</sub> and nearly stoichiometric decreases in GEM (Brooks et al., 2011).”

To: “Mid-day peaks of RGM in excess of 200 pg m<sup>-3</sup> were highly correlated solar radiation (including JBr<sub>2</sub>) and nearly stoichiometric decreases in GEM (Brooks et al., 2011).”

COMMENT: 5. Page 17140, line 8 - should say "exhibit", rather than "cause".

- This change has been made.

COMMENT: 6. Page 17141, line 1 - emission of what, specifically? Do you mean to suggest Br<sub>2</sub>? You certainly don't have emission of BrO.

- This sentence has been changed from: “Both trends could be reflecting boundary layer control if the primary source of BrO was emission from the snowpack, as is established for NO.”

To: “Both trends could be reflecting the influence boundary layer dynamics on gas concentrations if the primary source of bromine is release from the snowpack, as is established for NO<sub>x</sub>.”

COMMENT: 7. Page 17142, lines 14-18 - this is a run-on sentence.

C8383

- This has been changed from: “Dibb et al. (2010) reported significant variability in the Br- concentration in surface snow over the ~6-week long campaigns in both years, intervals with enhanced Br- concentration tended to coincide with elevated concentrations of the radionuclide tracers <sup>7</sup>Be and <sup>210</sup>Pb (monitored as part of Summit station baseline observations (Dibb, 2007)).”

To: “Dibb et al. (2010) reported significant variability in the Br- concentration in surface snow over the ~6-week long campaigns in both years. In addition, the intervals with enhanced Br- concentration tended to coincide with elevated concentrations of the radionuclide tracers <sup>7</sup>Be and <sup>210</sup>Pb (monitored as part of Summit station baseline observations (Dibb, 2007)).”

COMMENT: 8. Page 17142, lines 22 and 23 - again, this is by far the most important part of the overall conclusions, and should be emphasized. I believe there are recent reports in the literature about free tropospheric BrO, and these should be referenced.

- We have added the following to the conclusions: “However, the correlation of enhanced surface Br- with radionuclide tracers suggests a free tropospheric pool of Br<sub>2</sub> may be supplying bromide to surface snow during the summer.”

COMMENT: 9. Page 17142, end of line 24 - specify the actual range of Br- concentrations observed.

- We added an upper limit for the Br- mixing ratios “..(<0.4 pmol/mol).” To this sentence

COMMENT: 10. Page 17143, line 5 - provide a reference and/or explanation of what you mean, specifically, by "distillation", and by "towards Summit" line 8 - transport to Summit from where?

- This is only a possibility and has not been demonstrated to occur. However, the process is highlighted in Figure 1. Therefore, we have changed from: “Second, it is possible, but less likely, that distillation of bromine via snowpack chemistry within the boundary layer towards Summit occurs, which would commence at polar sunrise.”

C8384

To: "Second, it is possible, but less likely, that processing of bromine via snowpack chemistry within the boundary layer that results in net bromide transport towards Summit occurs (demonstrated as uptake and release of bromide in the BL in Fig. 1), which would commence at polar sunrise."

COMMENT: 11. Page 17143, line 23 - I note that active halogen chemistry in the Arctic Ocean basin occurs well into May.

- This has been changed from: "We therefore speculate that bromine activation in the Arctic basin just after sunrise provides most of the Br<sup>-</sup> that later mixes down to the surface at Summit, but recognize that the delay between sunrise in March and delivery to Summit in May is not fully understood."

To: "We therefore speculate that bromine activation in the Arctic basin just after sunrise and throughout the spring provides most of the Br<sup>-</sup> that later mixes down to the surface at Summit, but recognize that the delay between sunrise in March and delivery to Summit in May is not fully understood."

COMMENT: 12. Page 17145 - Again, you might (here?) mention specifically the need for aircraft studies of transport from the Arctic basin (or elsewhere?) to Summit, with speciated Br<sub>y</sub> measurements.

- We have added the text: "Additional studies characterizing free tropospheric and boundary layer Br<sub>y</sub> (including speciation and concentrations) concentrations in the Arctic are clearly warranted. "

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 17135, 2012.