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Atmospheric Chemistry and Physics Discussions

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

J. L. Thomas et al.

jenniet@atmos.ucla.edu

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Response to interactive comment on "Overview of the 2007 and 2008 campaigns conducted as part of the Greenland Summit Halogen-HOx Experiment (GSHOX) by J. L. Thomas et al. Anonymous Referee #2

COMMENT: This paper presents the major findings of two field campaigns conducted at Summit, Greenland. It is generally well-written and does the job it needs to do. It should be published following minor revisions.

Much of the material has been presented in published papers that have already been through a review process, so I have not commented on these conclu-

sions/interpretations.

My comments are generally minor.

- We thank the anonymous referee for these helpful comments, which have helped to improve the manuscript. We address the individual comments below.

COMMENT: P 17137 Line 17 to 20 – this sentence needs some fine tuning, for example, e.g. source of what? Also maybe emphasis directly that the scenario was controlled by chemistry rather than transport.

- The sentence has been change from: “A coupled snow-atmosphere one-dimensional model that assumed snow photochemistry as the only source successfully simulated observed NO and BrO at Summit during a three day interval when winds were weak (transport not a factor).”

To: “A coupled snow-atmosphere model simulated observed NO and BrO at Summit during a three day interval when winds were weak.”

COMMENT: P17138 Line 1 – The “history” part: The South Pole research involved ambient measurements of NO, and discovered unexpectedly high concentrations. They did not strictly “discover that sunlight shining on snow caused production of NO and NO2 that were released to the overlying air” but, interpreted their findings as such in the light of a literature survey (as explicitly stated in Davis et al section 3). In their literature survey, Davis et al cite Honrath et al 1999, Honrath et al 2000, Jones et al 2000, and Ridley et al 2000. Yes, the observations reported in Davis et al were done in December 1998 (so fit with your “in 1998...” angle), but Honrath et al’s work was done January 1999 (i.e. 1 month after Davis et al), and the Jones et al was done February 1999 (i.e. 2 months after). Given that Davis used the Honrath and Jones results to interpret their unexpected findings, and that Honrath and Jones actually carried out specific, designed experiments to quantify production and emission of NO and NO2 from snow, these references need to be included, and the “history” section amended

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to explain this all more accurately.

- The reviewer brings up a very important point, which has been discussed in detail in the review of Grannas et al. (2007). We have referred the reader to this review for a more complete perspective on the history of these measurements.

COMMENT: P17138 Line 26 – change to “and passive (multi-axis or MAX) DOAS instruments.” The point is “active” and “passive” DOAS

- This change has been made

P17139 Line 1 – “fluxing” is not a word; change to “being emitted” or something similar.

- This change has been made.

COMMENT: P17139 Line 21 – Personally, I think the place for a table summarising the full suite of measurements is in the overview paper. Given that it is already published, then okay, leave it like this. But please check the Liao et al really did include everything. For example, in the Liao et al table, BrO observations by DOAS are not described specifically as either LP or MAX, and a single uncertainty (10%) is given. Is this really for both techniques..? To answer that, I needed to go to the Stutz paper, in which there is no mention of MAX-DOAS at all.. So, what happened to the MAX-DOAS data..?? If there weren’t any good data that could be used, I’d suggest there is no need to mention MAX-DOAS in the overview paper.

- We have clarified in the text that Liao et al., 2011 contains a summary of the chemical measurements made as part of GSHOX. Adding the same table in the GSHOX overview would duplicate this information. Therefore, it has not been added to the review. In 2008, we added tether sonde profiles because the available delta temp (10 m vs 2 m) data, while suggestive of vertical mixing intensity, were an inadequate proxy for BL dynamics. Example tether sonde profiles can be found in Thomas et al., 2011. We have added wording to the manuscript to reflect this.

We have added the text: “Unfortunately, the retrieval of BrO profiles from the MAX

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DOAS proved unreliable at the low mixing ratios found at Summit." to clarify that the MAX-DOAS analysis has not provided additional information about bromine chemistry at Summit, due to the low levels of bromine found at Summit.

COMMENT: P17140 Line 4 – "there is little direct evidence" – maybe not from CIMS or LP-DOAS, but what about the MAX-DOAS observations..?

- As noted above, we have added text to clarify that the MAX-DOAS retrievals were unreliable at Summit due to the low levels of bromine.

COMMENT: P17140 Line 13 – what was the meteorological situation during this apparently highest BrO mixing ratios event..? Was there anything unusual that was not seen at other times during the two campaigns..? e.g. was there strong maritime air flow..?

- We have modified the text to clarify this: "It is likely that the highest mixing ratios of BrO at Summit during the two campaigns occurred during the MDE early in 2007 when BrO measurements were sparse. This was also the time with the lowest temperatures the two experiments were encountered at Summit and was characterized by relatively low wind speeds and air masses that were influenced by local processes."

COMMENT: P17141 Line 3 – change to "firn air"

- This change has been made.

COMMENT: P17141 Line 6 – Liao's table 1, which apparently reports measurements for both 2007 and 2008 does not mention tethersonde observations – goes back to my earlier point/question above.

- As noted above, we now refer to Thomas et al., 2011 for the tether sonde observations.

COMMENT: P17141 Line 13 – what do you mean by "successfully evaluated"?

- Due to the uncertainties in modeling snow photochemistry, the model needs to be

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evaluated against measurements. We have changed the sentence from: “This model assumed that snow photochemistry was the only source of NO and BrO and was successfully evaluated during a case study when the airmasses arriving at Summit had been over the Greenland Ice Sheet at low altitude for the previous 3 days.”

To: “The model assumed that snow photochemistry was the source of NO and BrO and was evaluated using a case study when the airmasses arriving at Summit had been over the Greenland Ice Sheet at low altitude for the previous 3 days.”

COMMENT: P17141 Line 21 – “by” 6 – 20% (could be “to”...)

- This change has been made.

COMMENT: P17141 Line 23 – “by” 12 and 10%

- This change has been made.

COMMENT: P17141 Line 26 – “time periods” rather than “intervals”

- This change has been made.

COMMENT: P17142 Line 1 – was there a consistent bias between LP-DOAS and CIMS? If so, it would be helpful for the reader to know at this point.

- Unfortunately the overlap between the two instruments was too sparse to allow a clear answer to this question.

COMMENT: P17142 Line 23 – if the pool was Br-, then it would be aerosol; is this what you mean? If you have in mind a gas-phase pool, you should not write Br-

- We have changed this to say a pool of bromine instead of Br-.

COMMENT: P17142 Line 28 – same point as above re Br-

- We have changed this to say bromine instead of Br-.

COMMENT: P17142 Line 29 – doesn’t it also seem likely that the source of Br- in snow

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at Summit is aerosol..?

- This is certainly a possibility. We haven't distinguished between gas and aerosol bromine, we have updated the capture of Figure 1 to reflect this by adding: "We use Bry to refer to both gas and aerosol bromine in the boundary layer and troposphere."

COMMENT: P17142 Line 29/P17143 Line 1 – I don't think you can say that the bromine loading at Summit likely originates from bromine explosion events. The BE would generate gas-phase bromine, and as suggested above, the source of Br- at Summit could easily be via aerosol. The BE could coincide with high aerosol, if the aerosol was the BE bromine source.

- We agree that aerosol may be a source of bromide in surface snow at Summit. However, this bromide in aerosol particles may also originate from bromine explosion events. As noted now noted in the Figure 1 caption.

COMMENT: P17143 Line 1 – do you mean gas phase or aerosol phase, or are you not distinguishing? Plus, and more substantively for your argument – to what height does convection operate over open water leads? Could this really inject something into the free troposphere? I guess what I'd like to know is, to what height could an open water lead convection plume extend?

- Gas phase bromine released during bromine explosion events in the coastal Arctic is at least partially taken up on aerosol and further processed (e.g. as cloud condensation nuclei) in the troposphere. Given the uncertainties in all of these processes is large, we have not separated gas and aerosol bromine in the troposphere. Regarding the second question, wide Arctic leads are known to amplify turbulent exchange (up to 300 meters above the ocean surface) (e.g. Esau, 2007). Therefore, this may increase the rate of exchange of bromine compounds released at the surface with the free troposphere.

We have realized we overstated the uncertainty in processes involving Bry in the boundary layer and troposphere. Therefore, additional process that are known to occur

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(even if the uncertainty is large) are now in red in this figure.

COMMENT: P17143 Line 3 – you mean open “water” leads

- We have added the text (narrow openings in the sea ice cover) after the word lead to clarify.

COMMENT: P17143 Line 16 – no evidence for active halogen chemistry... do you mean “bromine” chemistry..? This is puzzling, given that conclusions from GSHOX are precisely that there _is_ active bromine chemistry at Summit

- Given that the clarifying statement “via perturbed NMHC ratios” follows this phrase, we have left it as is.

COMMENT: P17144 Line 28 – Arctic needs capital A

- This has been corrected.

COMMENT: P17144 Line 29 – entrainment over the ocean – I guess this refers back to the discussion about convection over open water leads. If so, keep the two consistent – ocean is not the same as open water leads, for example (even though I wonder about open water leads for such a mechanism, as reflected in my earlier comment).

- This has been reworded from: “Transport via entrainment of bromine into the free troposphere over the ocean, followed by transport in the free troposphere and dry or wet deposition to the snow at Summit.”

To: “Transport via exchange of bromine from the boundary layer to the free troposphere from bromine explosion events, followed by transport in the free troposphere and either dry or wet deposition to the snow at Summit.”

COMMENT: Fig 1 – “lead” is spelt incorrectly (spelt “leeds”)

- This has been corrected.

COMMENT: Final thought – is it surprising that BrO is not measured by satellites over

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Summit..? Do you know how close they are to being able to measure these low mixing ratios?

- The levels of BrO are likely too low to be detected by satellites over Summit. The issue is not only the low mixing ratios, but also the shallow boundary layers where bromine enhancements are likely be to found at Summit. Satellites either need very large bromine enhancements or small enhancements that extend over hundreds to thousands of meters.

References: Esau, I. N.: Amplification of turbulent exchange over wide Arctic Leeds: Large-eddy simulation study, J. Geophys. Res., 112, D08109, doi: 10.1029/2006JD007225, 2007.

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