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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Longpath DOAS observations of surface BrO at Summit, Greenland" *by* J. Stutz et al.

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Response to Interactive comment on "Longpath DOAS observations of surface BrO at Summit, Greenland" by Anonymous Referee #1

(Responses are in italics)

To summarize further: The authors conclude that while the photochemistry in snow is the major source for the BrO measured at Summit (meaning that reactive bromine is formed locally), it is not yet known how bromine is originally transported to the center of the Greenland ice sheet. The closest (and only?) source of bromide is the ocean

around Greenland and the authors speculate that air rapidly transported from the marine boundary layer could deliver bromide to the surface at Summit – but as quoted above they also say (Page 6708, line 28): "However, marine transport events are rare and most likely not the main source of bromide in surface snow at Summit." A bit of a contradiction, right?

Since their analysis shows that air masses with a clear marine origin were rare during the two campaigns, this raises the rather important question if during other times of the year the transport from the marine environment could be more frequent and could be responsible for replenishing the bromine content in the snow. It would help the discussion if the authors could provide more information on the topic if transport of marine air to Summit is more frequent during most of the year or if May-July display a typical behaviour in this regard.

The bromine needs to come from somewhere and it certainly is vital to exclude the possibility (although rather unlikely) that it is introduced by anthropogenic activities around the Summit station (see comment SC C619).

The reviewer is correct - we were referring only to the periods of our observations and rapid transport events that bring gaseous bromine to Summit. We performed snow samples to investigate if Summit station or the aircraft operations at Summit could be a source of bromine. The results of these tests showed no evidence for an impact of both possible sources (please see also a more detailed response to comment SC C619)

We have revised the first paragraph of the conclusions to address both of the reviewer's comments and added two new references that have investigated marine transport events to Summit:

"Possible sources include bromide in falling snow, or transport and deposition of inorganic or organic bromine species from the free troposphere or marine regions around

Greenland. Summit station and aircraft operation at Summit have been excluded as possible sources of bromine based on a comparison of snow samples collected close to these possible sources, with samples collected at 0.5 – 5km distance from the station.

Rapid transport from the north Atlantic was observed during at least one rapid marine transport event in 2008, showing clearly that gaseous bromine species can, at times, be transported directly from the Greenland east-coast to Summit in late Spring and Summer. A trajectory study by (Kahl et al., 1997) shows that this type of trajectory is rare (2-4% of annual trajectories). It thus appears that the observed type of rapid transport event is infrequent and that these events are unlikely to contribute significantly to the bromide budget at Summit. It is unclear how important marine influence is during other times of the year. Transport of air with low ozone associated with bromine catalyzed arctic ozone depletion events has been found to influence Summit in spring (Hirdman et al. 2010). While it is likely that these transport events bring gaseous bromine or aerosol bromide to Summit, we have no clear evidence that this is the main source of snow bromide observed at Summit. Our observations also show that boundary layer chemistry is significantly different during these marine transport events from times when air resides on the Greenland ice sheet for extended periods of time. "

Hirdman, D., Burkhart, J. F., Sodemann, H., Eckhardt, S., Jefferson, A., Quinn, P. K., Sharma, S., Strom, J. , Stohl, A.: Long-term trends of black carbon and sulphate aerosol in the Arctic: changes in atmospheric transport and source region emissions, Atmos. Chem. Phys., 10, 9351-9368, doi:10.5194/acp-10-9351-2010, 2010.

Kahl, J.D.W.,Martinez, D.A,. Kuhns, H., Davidson, C.I., Jaffrezo, J.L., Harris, J.M., Air mass trajectories to Summit, Greenland: A 44-year climatology and some episodic events, J. Geophys. Res., 102, 26861-26875, 1997.

We have also changed the language in the abstract to clarify this point: "However, rapid transport of marine air from the Greenland east coast is rare and most likely not the main source of bromide in surface snow at Summit."

Specific comments:

Page 6711, lines 3 & 10-11: Explain what GSHOx stands for when first mentioned

done

Page 6714, line 15: typo: . . . Dispersion Model . . ..

done

Page 6719, lines 19-20: Could you add some refs for the CH3Br and CH3I measurements or a very brief explanation how they are measured?

We added the following two references detailing the analytical methods of the CH_3Br and CH_3I measurements to the manuscript. These references are now also cited in the text as "(see Colman et al, 2001; Swanson et al, 2002 for details on the measurement principle)".

Colman, J.J., Swanson, A.L., Meinardi, S., Sive, B.C., Blake, D.R., Rowland, F.S.: Description of the analysis of a wide range of volatile organic compounds in whole air samples collected during PEM-Tropics A and B, J. Anal. Chem., 73, 3723-3731, 2001.

Swanson, A.L., Blake, N.J., Dibb, J.E., Albert, M.R., Blake, D.R., Rowland, F.S.: Photochemically induced production of CH3Br, CH3I, C2H5I, ethane, and propene within surface snow at Summit, Greenland, Amos. Environ., 36, 2671-2682, 2002.

Page 6720, line 4: Delete space after 'decrease'

done

Page 6720, line 11: Change 'Fig/' to 'Fig.'

done

Figure 2: This looks very convincing for around 3 ppt but how does the absorption structure look for say 1 ppt? Where is the measurement threshold so you can still comfortably recognize the measured BrO absorption?

The purpose of the figure was primarily to show the clear spectroscopic identification of BrO at Summit. The quality of the spectral retrieval is given by the error in BrO, which for the spectrum shown in Figure 2 is $\pm$0.3 ppt. The error of BrO, which is reported throughout the manuscript, is determined by the least squares fit that underlies the spectral retrieval. This is described in detail in Platt and Stutz, (2008), which is included as a reference in the manuscript. The error changes from spectrum to spectrum, as the environmental conditions, such as visibility and turbulence, change.

We quote the detection limit of the measurements, again for each individual spectrum, as twice the statistical error. For the spectrum in Figure 2 the detection limit is thus

0.6 ppb. It should be noted that this is not smallest detection limit found during the experiment. The description of the BrO error and the detection limit was already given on page 6712 lines 16-29.

Figure 3: top 2 panels: The text inlay (2007: LP-DOAS etc.) covers the highest data points in panel B, why not move it slightly higher since the ticks on the x-axis are anyway covered.

done

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 6707, 2011.