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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Meteorological controls on the vertical distribution of bromine monoxide in the lower troposphere" *by* P. K. Peterson et al.

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We thank Anonymous Referee #2 for the constructive comments on this manuscript. In our response, we denote points that that the reviewer raises using bold text followed by our reply. Please also see our replies to Anonymous Referee #1.

1. The paper focuses on two possible meteorological controls for BrO vertical distributions: wind speed and temperature. Analysis of temperature (stability) and wind speed is not sufficient considering the particularities of the Barrow research site where air that arrives can be influenced different histories (originated from snow, ice, the city of Barrow, etc). As pointed out by the first

reviewer, the issues of airmass origin must also be considered in this analysis. For example, what is the influence of wind direction on BrO VCD,200m and BrO VCD,LT?

The reviewer points out that our analysis does not address air mass history, which has also been shown to influence BrO. Over the course of these measurements, all measured airmasses spent at least 25% of the past 72 hours in contact with sea ice while 90% of measured air masses spent over 75% of the past 72 hours over sea ice. While there is an influence of airmass history on the levels of BrO, given the high levels of sea ice contact observed throughout the campaign, it is unlikely that the variability of BrO observed during this campaign can be attributed to variations in air mass history. This is not to say that air mass history does not play a role, we simply point out that this data set is unsuitable for discerning that role. We are currently working on a separate manuscript that draws on observations from multiple years to look at the influence of air mass history on BrO observations at coastal sites. See Fig. 1 of this reply that shows statistics on the history of observed air masses. The left panel shows contact with sea ice areas in the previous 72 hours, calculated using methods detailed in (Simpson et al., 2007). The right panel shows the wind directions observed during the campaign. We have also added Section 3.3 to the revised manuscript to address these concerns.

2. It is concerning that in Figure 6 all of the area in gray is excluded from the correlation presented in Figure 7. Is there a known reason for the horizontal gradient in BrO during this period? A more in depth analysis (including airmass history) may provide insight onto these differences, which should be discussed in the paper.

This time period coincides with a convective plume from an open lead north of the

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BARC building that is visible on MODIS imagery. Because DOAS is a path-averaged measurement, it is possible that this convective lead plume would cause horizontal gradients in BrO. As this mechanism is purely speculative at this point, we felt it best just to show the existence of a horizontal gradient. We have modified Fig. 5 to show the existence of the lead in the DOAS view direction and added a discussion of the lead opening to Section 3.4. Please see page 17, lines 6-8 of the revised manuscript.

3. Figures 10 and 11 show the BrO VCD 200m as a function of temperature (Figure 10) and wind speed (Figure 11). However, other controlling factors (e.g. available sunlight and ozone mixing ratios) have not been considered as possible driving factors controlling BrO VCD,200m and VCD,LT. Although the influence of ozone has somewhat been taken into account by excluding very low ozone airmasses, it does not seem a fair comparison to consider all measurements together, without considering also cloud cover and/or time of day in order to consider the amount of available sunlight in these correlations.

We agree that simple univariate analysis is insufficient to fully elucidate the impacts of temperature and wind speed on BrO, however this data set is not large enough to provide an adequate sample size across the range of cloud conditions and daylight conditions proposed by the reviewer. Additionally, cloudy conditions make the use of MAX-DOAS to measure BrO problematic, given the relationship between AOD and the information content of the aloft layer.

4. In Figure 12, the authors show that BrO in the lowest 200m peaks at the beginning and end of the day. What does it mean that BrO peaks when there is no longer enough available sunlight to make MAX-DOAS measurements? Does it mean, that there is still enough available sunlight to drive Br2 <-> BrO photochemistry, but not enough to measure? Is this a real feature?

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Given the relationship between atmospheric stability and LT-VCD discussed in this paper, we find it reasonable that the diurnal profile of the VCD_{200m} is consistent with a surface source of BrO being diluted throughout an expanded boundary layer at midday. However, as discussed below, the growth of the LT-VCD is not consistent with purely dilution. Please see Section 4.1 of the revised manuscript for a discussion of this feature.

5. It is interesting that the VCD,LT-VCD peaks at a different time of day than the BrO VCD,200m. This may have to do with the combination of available sunlight and mixing processes. Has the balloon sounding data been used to look at the temperature profiles above 200 m in order to investigate the amount of vertical transport occurring at mid-day? Is the peak in the lowest 2 km from a surface source of reactive bromine, or is it from bromine already in the free troposphere? Airmass histories (FLEXPART or HYSPLIT) and additional balloon sounding data are needed to suggest the origins of the measured BrO within the VCD,LT.

Interestingly, the growth of the LT-VCD is not consistent with dilution, but indicates an increase in activation of bromine at midday, this could be due to increased sunlight around local solar noon, activation occuring aloft, or an increased BrO lifetime aloft. We find it unlikely that this systematic peak at a particular time of day would indicate increased transport of free-tropospheric BrO to the measurement site as there is currently no physical mechanism to explain that. Additionally, given the poor resolution and lack of sensitivity of MAX-DOAS at higher altitudes, we do not feel these data allow us to unambiguously differentiate between boundary layer BrO and free tropospheric BrO. Thus, we do not feel it is appropriate to discuss the existence, or lack thereof, of free tropospheric BrO at the measurement site. We have added a discussion of possible explanations for this behavior to the discussion, last paragraph of section 4.1

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of the revised manuscript.

6. These type of diurnal profiles in BrO (peak in the morning and afternoon) along with a dip at mid-day have been observed already at Summit (See Figure 6 of Stutz et al., 2011). This was used in part to suggest a small snow source of reactive bromine even far from the coast. The authors should discuss if the same type of diurnal profile in their VCD,200m (albeit at a different site and time of year) is similar evidence for a snow source of bromine.

We agree that our findings constitute similar evidence for snow-sourced bromine. We have included a discussion of this work in the discussion section, page 20, lines 24-26 of the revised manuscript.

7. The conclusions section of the paper should be expanded to include a better perspective on what this paper adds to our understanding of bromine sources and chemistry. In comparison to the paper, the conclusion is much too brief and should include a better synthesis of the science results and their context.

We have expanded the conclusions to address these concerns. Please see page 25, lines 1-9, of the revised manuscript for additions to the conclusions.

8. The title of the paper should be changed to "Temperature and wind speed controls" unless further analysis of the meterology during the campaign is presented in the paper.

The proposed change misrepresents the information contained in the paper, as

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many conclusions of the paper are based on atmospheric stability, which would not be implied by the proposed revised title. While we do not address every aspect of meteorology, this work is a substantial analysis of the response of BrO to various meteorological factors.

9. The statements about using bromine source parameterizations based on wind speed in 3D models are currently not totally supported by the data presented in the paper. Specifically - I object to the text which states, "BrO may show an apparent relationship to wind speed" on Page 23969. The authors have not done enough analysis to determine how bromine gets out of the boundary layer into the free troposphere, where it is likely to be detectible by satellites, which may very well correlated with wind speed. The authors should be more careful in their wording, because this may not be just an apparent, but an actual correlation of BrO in the free troposphere with surface wind speeds. Although, wind speed may not represent the first step in the bromine release process.

Our objective in the second paragraph of section 4.2 was to convey two points.

1. That our data do not support an increase in activation of BrO at high wind speeds.
2. That the discrepancies in the conclusions of our study vs previous work relying on satellite measurements may be due to differing viewing geometry.

We agree that our wording should be improved and have modified the text to convey these points more clearly. Please see modified text at the end of Section 4.2 of the revised manuscript.

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References

Simpson, W. R., Carlson, D., Hönninger, G., Douglas, T. A., Sturm, M., Perovich, D., and Platt, U.: First-year sea-ice contact predicts bromine monoxide (BrO) levels at Barrow, Alaska better than potential frost flower contact, Atmospheric Chemistry and Physics, 7, 621–627, 10.5194/acp-7-621-2007, http://www.atmos-chem-phys.net/7/621/2007/, 2007.

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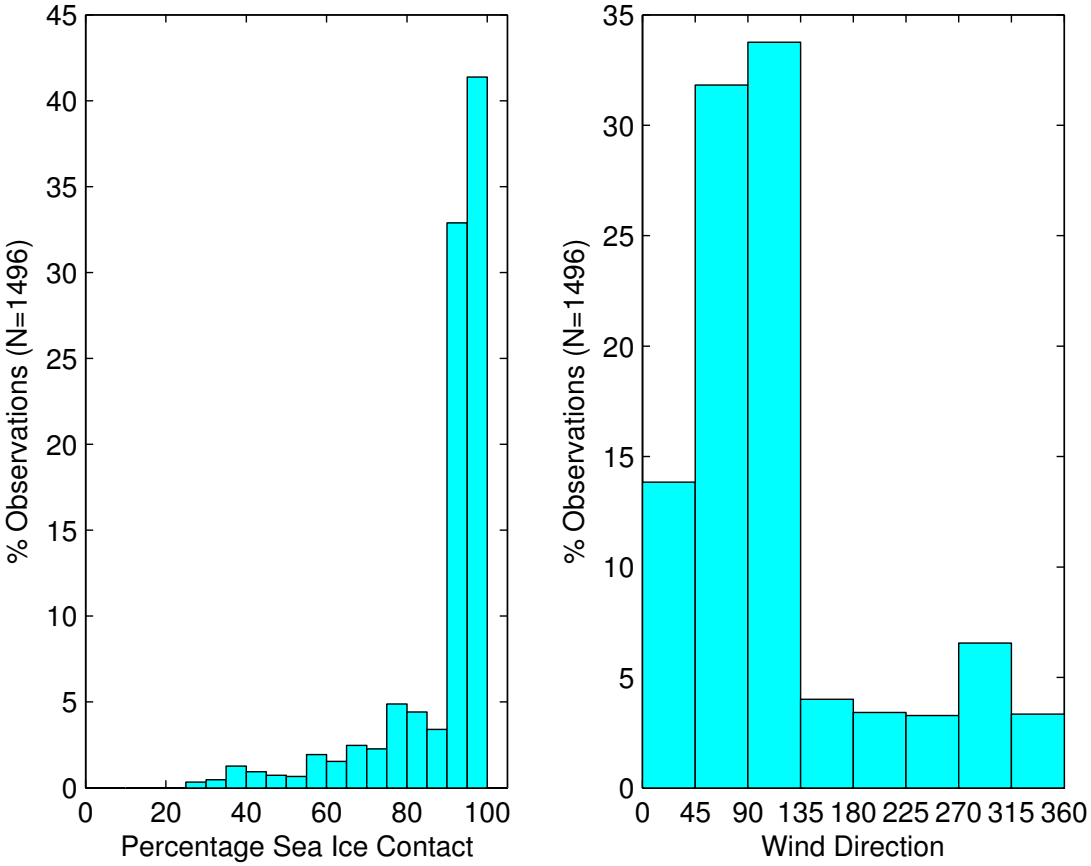

Fig. 1. Sea ice contact and local wind direction of sampled air masses

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