

## ***Interactive comment on “Observations and the source investigations of boundary layer BrO in Ny-Ålesund Arctic” by Yuhan Luo et al.***

### **Anonymous Referee #1**

Received and published: 8 September 2017

The paper describes an interesting event of enhanced tropospheric BrO concentrations at Spitsbergen. During that event very rapid decrease of O<sub>3</sub> and mercury was observed.

In my opinion this is a very interesting case study and should finally be published in ACP. However, in its current form, the paper has a few major and many minor problems. They need to be addressed before acceptance.

#### Major problems

A) The introduction is not very clear. Please put your study in a better context of existing studies. For example, please make the following points more clear: - what are important current open research questions - what does this study contribute to answer

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these questions

B) The discussion of the role of long range transport is not clear. In my opinion, from the data you present (trajectory and satellite data), long range transport of air masses enriched in bromine can not be excluded. This should be clearly stated in the paper. Nevertheless, from other findings, I think you can very well conclude that this is probable a local event: -the sea ice occurred only for a very short period close to the measurements -exactly during this period, the enhanced BrO is observed Both findings indicate that the enhanced BrO is caused by a local source (connected to the sea ice). Here, it would be very important to have more information: -how large was the area in which the sea ice appeared? -how does this area compare to spatial scale determined by the wind speed (and direction) and the duration of the ozone destruction? -how long have the air masses been in contact with the sea ice before they reached the measurement site? -I think it is in general very important here to discuss the importance of transport compared to chemical processes.

C) You use a fixed Fraunhofer reference spectrum. In my opinion this is not a good strategy here, because you are interested in tropospheric BrO DSCDs. I strongly suggest to use a sequential Fraunhofer reference. The QDOAS software which you use offers this option. Alternatively, you could subtract the BrO DSCDs from the 90° measurement of each elevation sequence from the BrO DSCDs at low elevation angles.

D) You apply the method of Sinreich et al. (2013). However, this method should be applied to the tropospheric DSCDs, for which the stratospheric part was already removed (both BrO and O<sub>4</sub>), see the point above. In the current form, the derived O<sub>4</sub> light paths include the light paths in the free troposphere and stratosphere, which are misleading for your study.

E) From the dependence of the BrO DSCDs on elevation angle you conclude that the highest BrO concentration is located at the surface. If this was the case, then the differences of the BrO DSCDs for the low elevation angles should be much larger. In my

opinion, the highest BrO concentrations are probably not located directly at the surface. To clarify this important point I suggest to perform one of the following tasks: a) perform RTM simulations (for different assumed BrO profiles) and compare the RTM results to your measurement results. Then you can derive more robust conclusions about the BrO profile shape. b) it would be even better to perform a full profile inversion.

Minor points:

Page 1, line 24: Enhanced BrO was first detected by LP-DOAS observations. Please add the respective references.

Page 2, line 4: I think Monsoon and turbulence are not the correct terms here. Monsoon is a tropical to mid latitude phenomenon. Why not simply write 'wind'?

Page 2, line 19: I don't agree with this statement for observations over bright surfaces like ice or snow. I think for such conditions a general underestimation was not reported.

Page 2, line 21: You write 'Long-path DOAS measurement provides regional determination of BrO in PBL.' It is not clear what exactly you want to say with this. LP-DOAS usually has light path lengths between a few and 20 km.

Page 2, line 22: you write 'but few ground-based MAX-DOAS measurement of BrO has been performed in Ny-Ålesund.' I think you can not write this. To my knowledge, University of Bremen performs MAX-DOAS measurements there since many years. (Did you compare your results with the Bremen results?)

Page 3, line 9: Did you perform MAX-DOAS measurements also before and after the presented period? Have you observed similar events before or after, or in other years?

Page 3, line 9: What do you mean with 'wavelength adjustment'? Why do you have no measurements during such periods of wavelength adjustment?

Page 3, line 19: Please give typical integration times

Page 4, line 17: please add information about typical uncertainties of this method,

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which are given in Sinreich et al., 2013. You might also have a look at the paper by Wang et al., 2015: Wang, Y., Li, A., Xie, P. H., Wagner, T., Chen, H., Liu, W. Q., and Liu, J. G.: A rapid method to derive horizontal distributions of trace gases and aerosols near the surface using multi-axis differential optical absorption spectroscopy, *Atmos. Meas. Tech.*, 7, 1663-1680, <https://doi.org/10.5194/amt-7-1663-2014>, 2014.

Page 4, line 20: To my knowledge, the formula is only valid for the tropospheric DSCDs. That means that a) the DSCD for 90° elevation of the individual elevation sequence have to be subtracted from the DSCDs of the low elevation angles (for analysis with fixed reference) before the formula is applied b) or the analysis has to be performed with a sequential reference. Since you use QDOAS, I strongly recommend to use this option.

Page 4, line 24: In my opinion it makes no sense to apply such a correction, because of two reasons: a) the differences of the light paths for such small wavelength differences are very small b) the errors of this method (Sinreich et al) are generally rather large. Thus the uncertainties caused by the different wavelengths are negligible. I suggest to remove this part (lines 23 to 31)

Page 5, line 25: The results in Fig. 5 indicate that enhanced BrO (above the background) is found until 4 May 2015.

Page 5, line 29: I suggest to remove the GOME-2 BrO VCDs from the figure. You show total BrO VCDs. It is unclear how they relate to the BrO mixing ratios from the MAX-DOAS. If you want to keep the GOME-2 BrO VCDs, then you should discuss, how large the stratospheric BrO VCD is. And you should indicate, above which value of the total BrO VCD you think they are affected by enhanced tropospheric BrO.

Page 6, line 1: Since you use a fixed Fraunhofer reference, these light path lengths include the light paths in the free troposphere and stratosphere. This makes no sense here. I strongly suggest that you should use a sequential Fraunhofer reference (see major point above).

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Page 6, line 4: at least on 27 April a slight enhancement is seen.

Page 6, line 9: The differences of the BrO DSCDs between 2° and 4° are very small (5 to 10%). In my opinion this indicates that the highest values are probably not directly above the surface. I suggest that you should perform RTM simulations (for different assumed BrO profiles) and compare them to the measurement results. Then you can derive more robust conclusions about the BrO profile. Alternatively also a full profile inversion could be done.

Page 6, line 12: how high is the boundary layer? Maybe the maximum BrO concentration is on the upper edge of the boundary layer? (see e.g. Wagner et al., 2007)

Page 6, line 28: In my opinion this can not be concluded. The trajectories pass over several regions with enhanced BrO (e.g. directly north of Spitsbergen). So, in principle air masses enriched in BrO might have been transported to Spitsbergen.

Page 7, line 15: The enhancement goes very slowly back to normal values (until 4 May). You might mentioned the period here.

Fig. 2b: to which reference (time period) are the anomalies calculated? Please give a reference for the map.

Technical suggestions / language improvement (please note that language improvement is urgently needed; here I list only a few points)

Page 1, line 11: 'difficulty of real-time observations' What exactly is meant here?

Page 1, line 17: separately => respectively?

Page 2, line 4: former => source?

Page 2, line 9: If you took this scheme from another publication, please add the corresponding reference.

Page 4, line 18: The sentence 'The diurnal variations of the clear-sky AMFs for BrO

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are in accordance with O4 in the boundary layer.' is not clear to me. Please clarify.

Page 5, line 13: Are you sure that the BrO maps are from NASA? In Fig. 8 you show results from Uni Bremen.

Page 5, line 26: unpredictable => unexpected?

Fig. 5: It should be mentioned that presented BrO DSCDs are obtained from measurements at 2° elevation angle.

Fig. 8: Not the tropospheric, but the total BrO VCDs are shown. Please correct the caption. Would it be possible to show additional BrO maps for the period after 27 April?

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