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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Satellite observations of long range transport of a large BrO cloud in the Arctic" *by* M. Begoin et al.

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First of all we would like to thank the reviewer for his critical and thought provoking comments. They motivated us to perform substantial additional analysis with respect to a possible stratospheric influence on the event and to reconsider our interpretation in the light of the new results and the comments made by the reviewers and the editor. More specifically, we have

- *rerun the FLEXPART model for different altitudes from the surface to the stratosphere to investigate in more detail the most probable altitude of the BrO observed,*
- *improved our satellite BrO analysis by implementing the stratospheric correction*

developed by Theys et al.,

- *investigated the connection between BrO and ozone fields,*
- *compared variations in tropopause height to BrO and ozone columns.*

The main new results are

- *that the best agreement between FLEXPART and measurements is achieved if BrO is assumed to be between 1 and 3 km,*
- *that there is little vertical shear in the airmasses during the BrO event making firm conclusions on the vertical position of the BrO layer difficult,*
- *that the influence of variations in stratospheric BrO on the retrieved tropospheric columns is relatively small according to the model of Theys et al. and cannot explain the observed BrO enhancement,*
- *that the very large BrO columns are observed in areas with low tropopause but even lower tropopause altitudes elsewhere do not coincide with enhanced BrO.*

Based on this additional information, the possibility of a stratospheric intrusion cannot be excluded as explanation for the observed sudden enhancement of the BrO column. However, activation of BrO on the surface as result of blowing snow at high wind speeds (see Jones et al.) in combination with upward lifting in the low pressure system is also a possible mechanism. This is now discussed in detail in the revised paper.
In the following, we will answer to the individual points made by the reviewer.

The manuscript compares GOME2 satellite observations of Arctic tropospheric BrO enhancements to transport publications based on passive tracer studies using the FLEXPART model and source region predictions based on Potential Frost Flower (PFF) maps. Qualitatively good agreement is found between the FLEXPART tropospheric BrO column predictions and the GOME2 observations, suggesting that long-range transport of BrO from remote regions may contribute to local BrO enhancements and subsequent ozone depletion events (ODEs). Given the short lifetime of BrO in the troposphere, the fact that FLEXPART passive BrO tracer predictions agree with GOME2 BrO observations leads the authors to conclude that BrO must be rapidly recycled within the advected air mass. Significant PFF levels were predicted nearly 5 days prior to the observed BrO enhancements, implying that Frost Flowers must have a lifetime of at least 5 days to be a potential source of the observed BrO enhancements. I find the authors arguments regarding BrO recycling during long-range transport within the Arctic boundary layer to be rather unconvincing. The editor's comments regarding some quantification of the sources and sinks of BrO along the FLEXPART trajectories would help. However, it seems quite likely to this reviewer that a much simpler argument could account for the qualitative agreement between the FLEXPART passive tracer predictions and the observed BrO. Namely, that the large-scale BrO observed by GOME-2 during the study period is in the upper troposphere/lower stratosphere and not within the Arctic boundary layer as assumed by the authors. I have two reasons to suspect this:

1) The GOME-2 tropospheric BrO retrievals used in the study are constructed by removing a constant ($4.5 \cdot 10^{13} \mathrm{molec/cm^2}$) stratospheric contribution during a period of significant dynamical variability in the lower stratosphere. This is likely to introduce significant errors in the inferred "tropospheric" BrO column.

We agree that variations in the stratospheric BrO column have an impact on the retrieved tropospheric columns if a simple offset is subtracted as was done in the original manuscript. In response to the reviewer's comment, we have implemented a correction for the variation in stratospheric BrO based on the model developed by Theys et al. The results show that the stratospheric variations of BrO during this period were small compared to variations shown in GOME2 BrO measurements. The BrO climatology data from Theys et al. show stratospheric BrO values between 2.5 and $4.5 \cdot 10^{13} molec/cm^2$, introducing an uncertainty of

$2 \cdot 10^{13} molec/cm^2$ which is more than a factor of 5 smaller than the observed BrO enhancement. In the revised paper, we have corrected the BrO columns for the stratospheric variability removing this source of uncertainty. The values in the main event are largely unchanged but BrO columns over the Hudson Bay area are lower after correction.

2) The GOME-2 BrO retrievals used in the study were not cloud-cleared; consequently much of the BrO enhancements must be above the clouds, not in the Arctic boundary layer.

The reviewer is right in pointing out the fact that clouds affect the retrieval of BrO from satellite measurements. Over dark surfaces, clouds strongly reduce the sensitivity to absorption below the clouds which is the argument the reviewers makes. However, cloud sensitivity studies show that over bright surfaces the measurements are sensitive to the surface layer even in the presence of a cloud with optical depth 20 (see e.g. http://www.doas-bremen.de/posters/dpg_2010_zien.pdf*). Therefore, we do not think that this argument precludes that the BrO is situated in the lower troposphere.*

Regions of strong cyclonic motion (low pressure) associated with the analyzed transport event will have significant mid-tropospheric cloudiness and lower tropopause heights. The mid-tropospheric cloudiness will obscure boundary layer BrO while the low tropopause heights will lead to incorporation of stratospheric BrO in what the authors assume is "tropospheric" BrO column.

We agree that low tropopause heights are associated to the strong low pressure system and that this might increase the stratospheric BrO column. This point is discussed in more detail in the paper now. As mentioned above, no general correlation is seen between low tropopause height and high BrO, in contrast to the situation for ozone where the link is much clearer. Also, very low tropopause heights are observed in situations with no BrO enhancement. We therefore conclude that tropopause height changes alone cannot explain the observations. One possible explanation for the large BrO enhancement is a stratospheric intrusion bringing BrO rich stratospheric air down to the troposphere where it remains until the removal through mixing with surrounding air masses. With a mixing ratio of 12 ppt, a layer of 3 km height situated at

5 km could already explain about $5 \cdot 10^{13} molec/cm^2$ of enhancement or half of the observed columns, if BrO isn't converted into bromine reservoirs.

Figure 1 illustrates this point for March 26, 2007, which is the day used to initialize the FLEXPART BrO trajectories used in this study. The figure shows GOME2 BrO from figure 2 in the M. Begoin et al manuscript, MODIS Cloud Optical Depth produced using the Giovanni online data system, developed and maintained by the NASA GES DISC, and OMI total column ozone imagery obtained from ftp://toms.gsfc.nasa.gov/pub/omi/images/ on 26 March 2007.
The GOME-2 BrO enhancements are found within the Arctic polar vortex (the region with lower OMI total column ozone amounts) and are associated with high MODIS cloud optical thickness. This pattern is consistent with lower stratospheric BrO enhancements associated with stratospheric ozone loss processes. Assuming that the observed BrO is within the Arctic boundary layer is not justified based on the MODIS cloud fields, which would obscure low level BrO enhancements.

The reviewer suggests that the enhanced BrO is linked to low ozone within the polar vortex, indicating a stratospheric origin of the BrO. We do not agree with this argument for two reasons:

First, BrO is usually only slightly enhanced within the stratospheric polar vortex and shouldn't be able to destroy large amounts of stratospheric ozone in the absence of activated chlorine. Chlorine activation could be observed several weeks before the event, but not at the end of March 2007 (see e.g. http://mls.jpl.nasa.gov/data/gallery.php*).*

Second, low tropopause heights are usually linked to larger ozone columns just as the reviewer suggests for BrO. This has been demonstrated in many studies and in the measurements shown here, there also is a general anti-correlation between tropopause height and ozone. For this reason, we would expect to see higher, not lower ozone values in the region of enhanced BrO if the origin of the BrO is indeed from the stratosphere.

I suspect that FLEXPART trajectories initialized in the upper troposphere would show similar qualitative agreement with the GOME-2 BrO observations, particularly if the cyclone responsible for the transport is occluded. I recommend that the authors conduct FLEXPART BrO tracer

simulations initialized in the upper troposphere to test this possibility.

Following the suggestion of the reviewers, we have performed FLEXPART model runs initialised in different heights. The runs do show similar transport patterns up to heights of 9000 m with the best overall agreement between model and measurement for a layer in 1 - 3 km altitude. Because of the similarity of the patterns, no firm conclusion can be drawn on the most probable altitude of the BrO, but there is indication that it is the lower rather than the upper troposphere as the higher altitude runs of FLEXPART show some early transport to lower latitudes which is not observed in the measurements.