

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 useful comments. In response to the comments made by the two reviewers and the editor, we have performed substantial additional analysis with respect to a possible stratospheric influence on the event and reconsidered our interpretation of the observations. 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.*

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- *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. The low tropopause height would be in agreement with such an interpretation but backward trajectories from the starting point of the event do not show evidence for strong downward movement into the troposphere as would be need.

On the other hand, 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 the individual points made by the reviewer.

Overall, this manuscript describes an interesting BrO event detected by satellite and followed over a roughly two-week period. The satellite BrO observations are compared to a FLEXPART

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model and also to fields of Potential Frost Flower (PFF) calculations to try to understand transport, recycling, and sources of bromine activation events. There are four main regions, where the manuscript needs work, as well as a number of smaller issues. In addition, the use of English should also be improved so that some unclear arguments may be understood.

We have tried to address all the points made by the reviewer.

Major Point 1: A valuable contribution of this manuscript is to show that initializing a transport model with a BrO distribution in the boundary layer (matching the satellite VCD enhancements) appears to show similar transport to the satellite-observed BrO enhancements. One expects that winds in the free troposphere (FT) and upper troposphere / lower stratosphere (UTLS) would be different, and thus placing this BrO higher in the atmosphere would lead to a divergence from the observed BrO distribution, which would then indicate that the satellite-detected event was probably in the boundary layer. However, the analysis does not include these other scenarios of FT or UTLS BrO events. The authors should do this FLEXPART modeling to see how much divergence there is between the model and the FT and UTLS BrO scenarios. In this way, they could enhance the picture that the satellite-detected BrO enhancement is in the BL. On the other hand, if there is not a divergence between the satellite observations and the FT and/or UTLS scenarios, the authors case that this event is a BL event is weakened.

In response to the suggestion made by both reviewers, additional FLEXPART model runs have been done and show broadly similar results up to 9000 m height. During the event, no large vertical wind shear was modeled below this altitude, limiting our ability to firmly determine the vertical position of the BrO. If a start altitude of 12 km is assumed in FLEXPART, then very different model BrO distributions result.

However, closer inspection of the results shows that the higher altitude simulations (above 3 km) show some rapid transport to lower latitudes not observed in the measurements. Correlation analysis indicates that best agreement is found for a layer of BrO situated between 1 and 3 km. We therefore conclude that the BrO was probably not exclusively located in the BL but rather in an extended layer in the lower troposphere.

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Major Point 2: There is not a rigorous testing of the PFF hypothesis in this manuscript. The authors point to what appear to be weak PFF events from 20-25 Mar as causing the 26 Mar satellite-detected BrO events. Yet, they do not discuss other times when there appear to be larger PFF events that do not lead to production of enhanced BrO detected by the satellite. Specifically, looking at the maps on 28 and 29 Mar, there are major PFF events in the E. Siberian Sea and the Laptev Sea (in the region 110 to 160 degrees East latitude and 75 - 90 degrees North latitude). These PFF events are more intense than those that purportedly started the 26 Mar event, but they show no enhancement in satellite-detected BrO on those days are nearly thereafter. To make claims about PFF, the authors need to make a rigorous comparison of the relationship between BrO events and PFF (and possibly other things like UV radiation and/or windspeed). Such a comparison would include all four possibilities: PFF detected (or not), BrO detected (or not). What they have done at this time is taken a large Br event and simply said that some weak PFF events prior to is caused it. They ignored a clear case of PFF detection without production of enhanced satellite BrO (28 and 29 Mar).

A main limitation of the potential frost flower (PFF) method is the accuracy of the sea ice concentration data. The sea ice concentration in the central Arctic exhibits errors that are larger than the actual ice concentration variability. Traditional passive microwave ice concentration data over the high concentration Arctic sea ice exhibit errors and biases that are about one magnitude larger than the true variability (Andersen et al., 2007). Although larger coastal polynyas are reasonably represented in 85 GHz SSM/I sea ice products, leads and openings in the central Arctic can not be resolved. Thus, the interpretation of PFF data based on SSM/I ice concentration is very difficult with the present data. One has to know that the coastal values are more accurate than the values at the pole, which is a problem especially in the potential source region of the discussed BrO event.

However, an ice concentration and PFF error field has not yet been derived. This was the reason why the central Arctic region was not discussed in Kaleschke et al. (2004). An improved sea ice concentration dataset may help to calculate more accurate PFF values in the whole Arctic. A step forward in this direction is a new lead detection technique from AMSR-E data

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which was recently developed (Röhrs and Kaleschke, 2010).

Because an improved sea ice concentration and PFF dataset isn't available yet, we decided to omit the PFF topic from the present work.

Andersen, S., R. Tonboe, L. Kaleschke, G. Heygster, and L. T. Pedersen (2007), Intercomparison of passive microwave sea ice concentration retrievals over the high-concentration Arctic sea ice, *J. Geophys. Res.*, 112, C08004, doi:10.1029/2006JC003543

Röhrs, J. and Kaleschke, L.: An algorithm to detect sea ice leads using AMSR-E passive microwave imagery, *The Cryosphere Discuss.*, 4, 183-206, 2010.

Major Point 3: The conclusions are much stronger than the arguments in the manuscript justifies. They need to be re-written to be more directly from the data and comparisons made in this manuscript. Specifically, the conclusions are written as if they have shown that these satellite-detected BrO events were actually tropospheric, which they have not shown. They have shown a general agreement between successive daily satellite observations of BrO enhancements and motion of a passive tracer in the BL in the FLEXPART model. By demonstrating significant deviations between BL, FT, and UTLS FLEXPART modeling, the authors could enhance the strength of their argument that these events are BL events; however, they have shown this analysis.

The reviewer raises two points:

- 1. that the conclusions are stronger than justified from the results.*
- 2. that the BrO event could also have been stratospheric.*

In response to these comments, we have

- a) rewritten the conclusions section and*
- b) extended the analysis to include FLEXPART runs at different altitudes.*

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As discussed above, the differences are not very large but for runs above 3 km, parts of the BrO are transported to low latitudes in contrast to observations which show that they are confined to the region over the Arctic Sea ice. The discussion in the paper was changed accordingly.

Similarly, the "conclusion" they claim of recycling on aerosols being more important than interaction with the surface is only one possible explanation for the actual observation from their study. The actual observation is that the FLEXPART model with no losses appear to give similar BrO magnitudes to the satellite enhancements. One could get that result either by highly efficient recycling or surface interaction (a combination of losses and production).

See very last point of the comments.

In addition, the FLEXPART model may not be capturing interactions with the surface properly. The springtime Arctic atmosphere is often stable, hindering interaction with the surface. However, open leads cause local convection that can overcome this static stability. It is likely that neither of these situations is well captured in the convection parameterization of FLEXPART. Can the authors indicate their confidence in FLEXPART for modeling surface interactions?

The reviewer is correct that FLEXPART may not simulate transport near the surface particularly well in the Arctic. Leads are much too small to be seen in the ECMWF analysis data used for driving the transport simulations. The convection associated with leads could indeed lead to additional mixing that the model does not capture.

On the other hand, the depth of the convection above leads is usually not very high, so mixing should be limited to the lowest part of the atmosphere and the subsequent transport error should not be very large, especially since we show that transport simulation results in this case are relatively insensitive to the exact height of transport in the lower and middle troposphere.

However, the stable situation should be handled relatively well by FLEXPART since the 91-level ECMWF model data used has a very high vertical resolution in the lower troposphere and the wind shear associated with the inversion layer should be captured quite well. There will be very little mixing by turbulence in such a stable situation (and only a shallow layer will be

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concerned) regardless of the actual exact strength and depth of the inversion.

Major Point 4: The abstract is similarly too strong compared to the findings reported in the study. Specifically, the abstract says, "...could be well reproduced by FLEXPART calculations for a passive tracer indicating that the activated air mass was transported all the way from Siberia to the Hudson Bay without further activation at the surface." Again, couldn't there be involvement of the surface in recycling Br? In the manuscript, they indicate that recycling on the surface (or pre-conditioning of the surface) are possibilities.

Yet the abstract picks one possibility without mentioning others or a solid argument for the validity of their choice. The abstract says "No direct link could be made to frost flower occurrence and BrO activation but enhanced PFF were observed a few days before the event in the source regions." this sentence seems to indicate that the PFF hypothesis was tested, yet not all possibilities were tested. Additionally, the PFF linkage in the manuscript seems quite weak because the identified PFF events are among the weaker events and stronger PFF events apparently don't create BrO activation.

Following the comments of the reviewers and in light of the results from the additional analysis, both the abstract and conclusions have been rewritten and now give a less selective view.

—Minor points— P20410, line 4: Satellite BrO is generally related to sea ice areas, but we have few observations of ozone depletion in those areas. Therefore, this sentence should be split into one that talks about ground-based BrO and ozone and one that talks about satellite BrO observations.

Changed as requested.

P20410, line 7: Many ground-based observations show multiple day ODEs or multi-day BrO events (i.e. at Alert and Barrow). Thus, we do not only rely on satellites to inform us that halogens must be reactivated to keep the events going.

Agreed.

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P20411, line 14: Can the authors more fully discuss the degree of uncertainty due to the selection of a constant BrO amount? Theys (ACP, 2009) indicate that stratospheric BrO varies with ozone column density. How important is the background level of stratospheric BrO on the interpretation? The authors claim it is small, but show no analysis indicating that it is small.

Theys, N., Van Roozendaal, M., Errera, Q., Hendrick, F., Daerden, F., Chabrilat, S., Dorf, M., Pfeilsticker, K., Rozanov, A., Lotz, W., Burrows, J. P., Lambert, J.-C., Goutail, F., Roscoe, H. K., and De Mazière, M.: A global stratospheric bromine monoxide climatology based on the BASCOE chemical transport model, Atmos. Chem. Phys., 9, 831-848, 2009.

We have now incorporated the BrO climatology data from Theys et al. into our analysis. For the period of the event, stratospheric BrO values reach from 2.5 up to $4.5^{13} \text{ molec/cm}^2$. Compared to observed GOME2 tropospheric BrO values, which reach up to $2 \cdot 10^{14} \text{ molec/cm}^2$, the variations are small. Nevertheless, we have applied the stratospheric correction in the revised manuscript to reduce the uncertainty introduced by stratospheric variability. The values of the main event are largely unchanged but tropospheric BrO columns over the Hudson Bay area are reduced.

P20412, line 4: The BrO was presumably initialized to be the boundary layer, but no details at this time as to the initial vertical distribution are given. Details are given later - p20414, Line 14. Please clarify early on or refer here to the later section.

p20412, line 9: The surface area of frost flowers was measured to be similar to snow (Domine et al., 2005, Obbard et al., 2009), not the assumed large values from early work. Thus their surface area is not large (as compared to snow).

Domine F., A.-S Taillandier, W.R. Simpson, K. Severin (2005), Specific surface area, density and microstructure of frost flowers, Geophys. Res. Lett., 32, L13502, doi:10.1029/2005GL023245.

Obbard, R. W., H. K. Roscoe, E. W. Wolff, and H. M. Atkinson (2009), Frost flower surface area and chemistry as a function of salinity and temperature, J. Geophys. Res., 114, D20305, doi:10.1029/2009JD012481.

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Agreed.

p20412, line 24: Note that these inferred values of BrO would indicate roughly 160ppt BrO in a 400m boundary layer, which is much higher than ground-based observations (which support up to 40ppt). This huge value could indicate something might be wrong with their calculation of tropospheric BrO VCD.

Thank you for pointing this out. The calculation of the tropospheric VCD was correct but the assumption that all the BrO was located in a 400 m BL is probably not correct. After analysis of the FLEXPART runs in different altitudes (see above) it is clear that the BrO was probably not (exclusively) located in the BL but rather over a larger altitude range which reduces the local mixing ratios to more realistic values.

Can the authors compare their observed values to those of active DOAS or other boundary layer BrO observations?

Unfortunately not. While we know of several ongoing studies linking ground-based and satellite observations, we are not aware of any ground-based BrO observations during this time in the region of interest.

p20413, line 12: In this section, wind speed is implicated as important for halogen activation, but couldn't the increased solar intensity (which is needed for halogen production and recycling) in the southern regions be important and the cause of this effect? Either add this idea or justify why you can eliminate solar differences.

We have added this point to the manuscript as suggested.

p20416, line 17: The authors mention PFF on 20 Mar, while the Figure 5 (and the enhanced figures supplied separately) show no data from 20 Mar, but instead start on 21 Mar, which shows particularly low PFF values. Later the authors continue: "'From this we conclude that for a direct initialization of the BrO event by frost flowers due to a wind induced release of sea salt aerosols to the gas phase a life time of frost flowers respectively their saline compounds of up to five days has to be assumed.'" This sentence is awkward, but appears to be based upon a

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prior belief that frost flowers release BrO, which is not proven and is supposedly being tested here. Possibly the authors are trying to say that frost flowers must live longer than 5 days, which is at odds with the findings of Perovich and Richter-Menge (1994), which indicates that frost flowers are covered by blowing snow "'within several days"'.

See comment under major point 2

p20417, line 16: The authors claim that BrO "'.....has not been produced in situ.'" Nothing in their analysis indicates that BrO could not be produced in situ. In fact, if BrO was being lost from the transported airmass (which is likely), then maintenance of levels similar to what is seen in the FLEXPART analysis, which assumes no losses, would require in-situ production.

We agree that this sentence was misleading. In-situ production of BrO is in fact needed to maintain BrO levels in the presence of loss mechanisms, and recycling on aerosols is one of the possible mechanisms. What we intended to say is that BrO observed on day 3 of the event was not necessarily originating from the ice / snow / frost flowers in the area where the BrO was observed on this day but was produced from bromine released 3 days earlier into the air masses more than 1000 km away. We have reformulated the sentence trying to make it more clear.

p20417, line 23: The authors refer to work discussing ozone depletion events. Because there are no significant ozone production methods in the lower Arctic atmosphere during springtime (outside of downmixing from aloft), one would expect that ozone depleted airmasses would transport over long distances, even if BrO were short lived. Therefore, the use of ODE observations to discuss lifetime of BrO is not relevant.

We agree that ODEs can be transported over long distances even if BrO is removed quickly and have added this point to the discussion. However, as long as the absence of BrO was not documented, the events discussed in the literature could well be similar to the one shown here. In fact, we are not aware of any publication showing evidence for strong ozone depletion with simultaneous measurements of low BrO.

p20418, line 23 on: At the end of a speculative section putting forward many hypotheses that

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are not testable by the current study's observations, the authors conclude, "However, the good agreement between transport calculation and observations as well as the relatively constant total BrO amount observed over several days and the high wind speeds involved suggest that at least for this event, recycling on aerosols within the air mass is more important than surface reactions." This conclusion is not justified by the broad discussion above it.

The reviewer is right that we do not have direct proof of aerosol recycling during the event. However, in our opinion it would be very difficult to explain the movement of the BrO "plume" by surface reactions, in particular as there is additional indication from the new FLEXPART runs that the BrO was not (exclusively) situated in the BL. Considering the use of "suggest" in the sentence we decided to keep it in the revised manuscript.