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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.

Anonymous Referee #2

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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 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.

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 tro-

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posphere / 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.

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).

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

ACPD

9, C7594–C7599, 2009

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. 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). 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?

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

ACPD
9, C7594–C7599, 2009

are among the weaker events and stronger PFF events apparently don't create BrO activation.

———-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.

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.

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 Roozendael, M., Errera, Q., Hendrick, F., Daerden, F., Chabrillat, 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.

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).

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9, C7594–C7599, 2009

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.

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

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.

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 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".

p20417, line 16: The authors claim that BrO ".....has not been produced in situ." Noth-

ACPD

9, C7594–C7599, 2009

ing 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 maintainance of levels similar to what is seen in the FLEXPART analysis, which assumes no losses, would require in-situ production.

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.

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

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 20407, 2009.