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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Global observations of tropospheric BrO columns using GOME-2 satellite data" *by* N. Theys et al.

Anonymous Referee #1

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This paper reports on application of a stratospheric BrO climatology to the determination of background tropospheric BrO columns. It appears to be a carefully done piece of work, and the manuscript is well written. A very useful result of this work is represented in Figure 14, i.e. latitude and seasonal dependence of the tropospheric BrO columns from GOME-2, amounting to, on average, about 1.5x10ˆ13 cmˆ-2. This is high quality work, and it is very timely, as there is currently substantial interest in this topic. The paper concludes that the tropospheric "hotspots" seem to correlate with low tropopause heights, but that they cannot be completely accounted for on that basis. As far as can be determined from reading the paper, the methodology is rigorous and represents the current state of knowledge. I believe this paper should be published, but only after correcting a number of significant problems. Specifically, I find that the authors draw some definitive conclusions that are not supported by the data presented

in the paper. Most importantly, the paper concludes that they can confirm that the blowing snow hypothesis can account for some of the large tropospheric BrO columns. Indeed, on page 22 it is stated that "... the results of Fig. 13 confirm the existence of the mechanism of bromine release from blowing snow events,...". However, I do not believe this is the case in any way. Figure 13 compares the TOMCAT model with tropospheric BrO retrievals, when the TOMCAT model incorporates a blowing snow mechanism. This then runs the risk of having a model that is highly parameterized to support a particular mechanism, without all the details of the chemistry and meteorology to properly assess that mechanism. Since TOMCAT includes a blowing snow parameterization that results in reactive bromine release, it is not too surprising that the results are consistent with this hypothesis. It is very important to be circumspect about the blowing snow hypothesis. Surface observations show high BrO levels and ozone depletion events when the surface is thermodynamically stable, and with radiation present. In contrast, during blowing snow events, there would be enhanced vertical scale turbulence, and decreased radiation, both of which should slow bromine photochemistry. The paper should discuss if indeed the observations are consistent with stable air AFTER a blowing snow event, when presumably the surface is more saline, as discussed in the Jones et al. paper. While the authors refer to "BrO emissions", BrO is not emitted, but is photochemically produced in a process that is believed to be propagated by the BrO + BrO reaction. This turbulent mixing should substantially slow the process. The paper should state how TOMCAT parameterized BL turbulence and BL structure around the lows. Does TOMCAT parameterize both emission of sea salt, and appropriate BL turbulence and reduced radiation during the blowing snow events, and then do detailed chemistry? If so, knowing those details would make the paper more interesting, and also make this result more believable. This relates to the fact that there is very sparse evidence for the blowing snow mechanism, and that evidence does not include in-situ measurement of sea salt aerosol or BrO, or BrO precursors. Regarding what was discussed in Figure 13, we do not even know that the enhanced BrO observed by GOME-2 is in the boundary layer! It stated on page 22 that "The results of Figure 12 provide a strong indication that the release of bromine by blowing snow events is probably playing an important role in the bromine explosion phenomenon,...". But, in fact, Figure 12 says nothing whatsoever about blowing snow. Thus the authors statement of confirmation of the hypothesis is troubling. What would "confirm" the hypothesis would be observations of enhanced BrO at the onset of, or immediately after, a blowing snow event, along with observations of enhanced SSA (or enhanced salty surface snow). At the top of page 13, the authors say that "the results presented here call for further modeling developments...". Actually, what they call for, if anything, is in-situ measurements during blowing snow events. However, in their defense, the Conclusions are indeed much more circumspect and, I think, appropriate, stating that "...no firm conclusions can be drawn..." about this mechanism, only referring to it as plausible. Thus the Conclusions are a bit in contrast to or in contradiction with the text, and the latter should be tempered. I provide more specific and more minor suggestions for corrections/changes below, in the order they arose in the manuscript.

1. The end of the first paragraph of the Introduction should cite Salawitch et al., 2010.

2. You might consider citing Sirois and Barrie, JGR, 1999 regarding inorganic bromine seasonal cycles in the Arctic at the bottom of page 2.

3. The last paragraph of the Introduction section is unnecessary.

4. Are brackets needed in the numerator in Equation 1?

5. I think some discussion about the VSL bromine species referred to at the bottom of page 7 is needed. If indeed there is an important contribution from such VSL species, and they are indeed very short lived, then they must be highly spatially variable, depending on where there is deep-convective injection of tropospheric air into the stratosphere, and thus there must be a great deal of uncertainty and variability in this component (which hasn't been measured). How does this translate into uncertainties in the stratospheric climatology?

6. There should be a brief explanation of how/why NO2 is an indicator of BrO/Bry, or provide a reference.

7. At the bottom page 18, it is stated that it is assumed that the BrO/O3 regression slope is fully controlled by the stratospheric component. But, the large offset between the stratospheric and total BrO regressions makes one wonder if this is a valid assumption. There should be a discussion of the impact of that assumption.

8. The units for the weighting function in Equation 2 should be stated. It is cmˆ2?

9. Re the first sentence in section 3.3.3, you might cite Neuman et al., ACP, 2010.

10. Bottom of page 13 - make it clear that those uncertainties are absolute uncertainties, not relative. I also think it is always better to use the word uncertainty, as the word "error" implies a systematic uncertainty.

11. The first sentence of Section 4 is a strange way to begin a section, as it reads as if it is a conclusion. The word "verified" is a poor word to use, as it implies some sort of correctness. It would be better to just say that a comparison with SCIA has been done and here you discuss the results. Along these lines, at the end of this section, the paper refers to the agreement as "satisfactory". That is a fully subjective term that depends on your criteria, and it should be changed. The discussion in section 4.2 also contains some subjective comments, such as "the agreement between GOME-2 and ground-based data is generally good...". The average reader has no idea what that might mean in quantitative terms. It would be best if the subjective evaluations are removed, and they are simply replaced with quantitative comparisons. For Figure 7, it might be more interesting/useful to discuss the times when the two are systematically different, in quantitative terms.

12. Mid page 14 - delete the "scientific product".

13. Section 4.2 - describe the sites discussed, in terms of local characteristics. For the last paragraph in this section, it would be helpful to remind the reader about the time of

day for the satellite-ground comparisons.

14. On page 17 and on page 20 the text refers to "BrO emissions". It is important that both the authors and the readers are aware that BrO is not emitted; rather, precursors, like Br2 or organobromine compounds are emitted, and then photolyze. There is considerable scientific interest in the drivers of those emissions! The top paragraph on page 17 refers to correlations of BrO with the coast lines of the Arctic and the Antarctic, and sea ice. However, it is clear in Fig. 8 (and in other places in the literature) that the enhanced BrO in the Arctic does not follow the coast lines. It is also the case that the last sentence of this paragraph is not in any way supported by the data presented! Just compare March and April for the Arctic in Figure 8. In both cases the areas in red are well into the ice covered region. There is much more to the story than just BrO following the ice retreat.

15. Mid page 18 and Fig. 10 - it seems from looking at Fig. 10 that the tropospheric (rather than stratospheric) columns may correlate better with tropopause height; however, staring at the plots is not the best way to decipher correlations. Can you be more quantitative about the correlations?

16. The statement on page 19 that (for the Hudson Bay) "...one can see that the retrieved tropospheric BrO columns rarely exceed the surrounding tropospheric background by more than this limit" (later defined as 3.5x10ˆ13) is very odd and unsupportable for two reasons: a) there are only three cases presented. So, what does "rarely" mean? And, b) in every case presented in Figure 10 that limit is exceeded, for at least part of Hudson Bay! From the data one can only conclude the opposite of what was stated.

17. Top of page 20 - I simply cannot see how the data presented in Figure 11 could in any way "confirm the effect of a weather pattern specific to this area". That sentence should be deleted.

18. Figure 15 is a very nice representation of the impact of the dependence of tropo-

spheric column BrO on cloud top pressure, and is a convincing case for the determination of tropospheric background BrO.

19. Figure 4 and caption - did you plot Air Mass Factors, or are they weighting functions, as indicated in the caption?

20. Figures 6 and 7 - it is hard to tell the difference between black and green.

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