

## ***Interactive comment on “Towards a climatology of stratospheric bromine monoxide from SCIAMACHY limb observations” by N. Sheode et al.***

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Review of Sheode et al., Towards a climatology of stratospheric bromine monoxide from SCIAMACHY limb observations, ACPD-2006-0093.

This paper reports retrievals of BrO profiles throughout the stratosphere, for two years, from SCIAMACHY limb radiances. Comparisons of the SCIAMACHY BrO profiles to balloon profiles are shown, and an estimate of the contribution to stratospheric BrO from very short lived bromocarbons is presented.

Overall, the paper is in good shape and should soon be suitable for publication in ACP.

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I have the following "major" comments, none particularly pressing but all of which hopefully will be addressed by some change to the paper. These are followed by many "minor" comments.

Major comments:

1. The comparison of SCIAMACHY BrO to the balloon BrO profiles is quoted in the abstract and conclusion as +17% to -42% (a bit more detail is given in the conclusions). These comparisons would be improved if a mean value of SCIAMACHY relative to the balloons were given, in addition to these ranges, and if it were made crystal clear whether +17% means SCIAMACHY is higher than the balloon value, or vice versa. I know +17% means SCIAMACHY is higher than the balloon, but I based this on looking at the figures to confirm I was interpreting the text properly. Assuming the mean value for the difference of SCIAMACHY vs balloon BrO will be a negative number, could add a phrase such as "-X% means SCIAMACHY BrO is lower than the average balloon BrO", or something like this.

2. Now that the Sioris et al. paper is published (JGR, 111, D14301, doi:10.1029/2005JD006479, 2006), there should be more substantive discussion of the results of this paper, which also examined retrievals of BrO from SCIAMACHY limb radiances. I do not mean to imply exhaustive discussions. But, Sioris et al. found higher values of BrO than reported in the paper under review and consequently found a value for Bry from VLSL of  $\sim 8.4$  pptv, considerably larger than the value of 3.5 pptv reported by Sheode et al. Also, Sioris et al. compared to balloon profiles of BrO. Unfortunately, even though Sheode et al. show comparisons of SCIAMACHY and balloon BrO for 4 balloon flights, and Sioris et al. show comparisons for 3 flights, there is only an overlap for one date (23 March 2003). So, relative evaluations of SCIAMACHY BrO versus balloon BrO from these two studies are difficult to assess. But, for this date, the Sheode et al. retrievals are closer to the balloon BrO profile than are the Sioris et al. retrievals, which exceed the balloon BrO profile by considerable amount.

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Statement of simple facts such as those above would add value to the paper. Other differences between Sheode et al. and Sioris et al. that should be noted:

a) Sheode et al. use BrO cross sections from Fleischmann et al., whereas Sioris et al. use cross sections from Wilmouth et al. Since (according to Sioris et al.) the Wilmouth cross sections tend to be larger than the Fleischmann cross sections, the use of these different cross sections goes in the opposite direction of explaining the differences between the Sheode et al. and Sioris et al. retrievals of BrO. A simple statement to this effect would also be of value.

b) although I do not fully understand the details of how the tangent height correction is handled, it appears that Sheode et al. and Sioris et al. are using different approaches to handle the "substantial error in the SCIAMACHY pointing as provided by ESA" (page 6440, lines 8-9). While both studies claim the tangent height uncertainty is not a dominant source of error (i.e., see paragraph 50 of Sioris et al.), a brief statement about these different approaches should be added.

c) I've always thought a key test of whether Bry from VLSL is important is whether or not Bry is near zero in the tropical, lowermost stratosphere (LMS). On page 6442, it is stated (and Figure 7 shows) that SAOZ balloon profiles of BrO exhibit much higher values in this region than are found based on the Sheode et al. retrievals of BrO. The statements about what F. Stroh is finding in the tropical LMS are fine. It is my understanding, however, that Dorf et al. (2005) see significant, non-zero BrO in the tropical LMS. Even if the Dorf DOAS values of BrO are less than SAOZ, as is stated in the paper, I am concerned the statement about DOAS BrO in the tropical LMS given on page 6442 is somewhat misleading because the DOAS values are still higher than "near zero". Most importantly, for March 2003, Sioris et al. also report non-zero BrO in the tropical LMS.

Given the recent results from F. Stroh, which he has kindly shared with me prior to publication, as well as the analysis of June 2005 DOAS measurements of BrO in the

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tropics (Terisina, Brazil) that M. Dorf is conducting, I am not sure anyone knows what the true values of BrO and Bry are in the tropical LMS. The Sheode et al. paper would be improved by noting that the retrievals of BrO presented by Sioris et al. show significant, non zero BrO in the tropical LMS for March 2003 and that understanding the true value of BrO in this region of the atmosphere is a critical research issue, for both the orbital and sub-orbital measurement communities.

In summary, to conclude major point #2:

even though Sioris et al. study is cited (page 6442, line 15), it is cited just once and in a fashion that conveys too little information. The Sheode et al. paper would be stronger if more detail is given regarding the findings of this other group, and how these findings compare and contrast to their results.

3. In general, the paper contains a large number of citations to previous work involving members of this group. While not really an issue, as they have been very active in this area, there is an omission of one key citation that must be addressed upon revision.

The estimates of Bry from organics are referenced to Sinnhuber et al. 2005 on line 25, page 6448. But, of course, this estimate truly comes from the work of Montzka et al., GRL, 2003. I had pointed out the need to cite Montzka et al. in a private email to the authors following the initial round of screening, but perhaps this communication was overlooked. Regardless, appropriate citations to these organic measurements, upon which all estimates of Bry from VLSL rely upon, should be added upon revision.

Also, when an estimate of Bry from VLSL is deduced by subtracting 15 pptv from  $\hat{Bry}_{mean\_total}$ , there is an explicit assumption of no tropospheric loss of CHBr<sub>3</sub>. We know that, based on the short lifetime of this species, that significant loss in the troposphere should occur, with values are the tropopause being perhaps 7% lower than global average surface values (Montzka et al., 2003). It is okay to assume no tropospheric loss of CHBr<sub>3</sub>, especially because it is possible that the inorganic species produced following the tropospheric decomposition of CHBr<sub>3</sub> make it to the stratosphere.

But, the assumption of no tropospheric loss should at least be explicitly stated.

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#### Minor comments

1. With so many citations given on page 6433 to prior measurements of BrO, perhaps Kreher et al. (GRL, 1997) should be added to the list of ground based BrO measurements and Pfeilsticker et al. (GRL, 2000) should be added to the list of UV-visible balloon measurements.

2. One of the 1996 papers by Lary is single authored, and should be cited as Lary 1996. The other should be cited as Lary et al., 1996. Looks like the two papers by D. Lary written in 1996 are mixed up in the citations given on pg 6435, line 4 and pg 6435, line 22.

Suggest checking all Lary et al. 1996 and Lary 1996 citations to be sure they are matched to the proper paper :)

3. pg 6436, line 6: suggest "in good agreement with field measurements of the upper limit of HBr (i.e., higher yields of HBr from this reaction would lead to profiles that exceed this upper limit) (Johnson et al., 1995)."

4. pg 6437, lines 22-28: Does the retrieval algorithm assume that BrO varies diurnally?

This is important under some conditions according to Sioris et al. :)

Somewhere, perhaps here, it should be stated whether or not this is taken into consideration.

5. pg 6439: it is unclear how the shaded region in Figure 1 is found.

Caption for Figure 1 states it is the total uncertainty.

Does this mean it is an RSS combination of the uncertainties shown in Figs 1, 2, and 3? Are other terms considered?

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Also, it is unclear why the lower limit for Figure 1, right hand panel, is  $\sim -45$  and not a value closer to  $\sim -70$ , as used for Figure 2. As drawn, Figure 1 right hand panel is clearly truncating information.

Please clarify upon revision.

6. pg 6440, lines 6-10: somewhat more information could be given about how the pointing correction was made, and most importantly, this info seems more appropriate to state in Section 3.1.

7. pg 6441 and 6442: simple definition of the sign of the difference would be helpful here, as well. Also, as noted above, would be useful to quantify overall average offset between SCIAMACHY and the balloon BrO profiles.

8. pg 6443, lines 23-26: I don't understand why this so-called discrepancy between measured and modeled BrO from Avallone et al. (1995) is being "called out". 40% compared to 50-60% is actually rather good agreement :)

Perhaps it is worth noting, as stated in several other recent papers, that the Avallone et al. results, taken at face value, imply no need for Bry from VLSL. In my opinion, this is the true importance of the Avallone study.

9. pg 6444, lines 19 to 22. I understand this material, but it could be confusing to some that there is a "decrease in BrO" (line 19). How about stating "decrease in BrO/Bry".

10. pg 6445, lines 7 to 28: Some of the organization is a bit confusing, because topic sentence on line 7 introduces the five latitude bands, yet the first figure discussed (Figure 9) is clearly based on data averaged on a finer latitude grid. Perhaps move the latitude band discussion closer to where Figure 11 is discussed!

11. pg 6445, lines 10-11: the tangent ht discussion is confusing: text states "prior to this", and I wrote on my printout "prior to what" :)

This sentence should be clarified. As noted previously, a bit more info in the tangent ht

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correction would be helpful.

12. pg 6446: Sioris et al. discuss the relation between BrO and NO<sub>2</sub> more extensively than any of the citations that are given. Perhaps a citation in this section, to Sioris et al., is warranted?!?

13. pg 6448, lines 15-28: In lines 15-22, the findings of Sioris et al. for Bry from VLSL should be mentioned.

It is kind of "sweeping some differences under the rug" to conclude on line 28 that the 3.5± 4 pptv value is "in agreement with previous studies". The 3.5 pptv does agree with the estimates of Bry from VLSL by Pfeilsticker et al. and Salawitch et al., within respective uncertainties. But, 3.5 pptv is on the low side of these previous estimates. Also, the Sinnhuber et al. (2002) estimate is ~ 5 pptv.

I suggest a compromise between an exhaustive discussion (which is not needed; this is given in Chapter 2 of the 2006 WMO/UNEP Assessment, as some of the co-authors are well aware) and a discussion that "sweeps differences under the rug".

14. I found use of color in Figs 4 to 7 to be confusing.

Perhaps the figures would be clearer if all SCIAMACHY values for BrO were shown using the same color (say BLUE), and all balloon profiles of BrO were shown using a different color (say RED). Then, we would know to focus on RED vs BLUE to assess SCIAMACHY BrO vs balloon BrO. Is it really necessary to show the raw, uncorrected balloon BrO profiles? These data are shown in Dorf et al. (2005), after all? Including 4 lines on the right hand panels results in confusion, at least for me [I didn't like the manner of presentation of similar figures in Sioris et al., but I was overruled :)].

15. Figure 8 is unreadable and should be improved.

16. Despite what is said in the caption for Fig 10, I do not see any red points :)

Minor suggestions related to grammar and trivial issues

A. pg 6432, line 21: suggest "ozone losses at a per molecule level"

B. pg 6432, line 22: suggest striking "very", as there are now actually a fair amount of BrO measurements

C. pg 6433, line 9: suggest "have also been measured" rather than "have also been done"

D. pg 6433, line 14: strike "now"

E. pg 6433, line 15: add a comma after "two years"

F. pg 6433, line 18: no need to abbreviate Section

G. pg 6333, line 25: suggest "conclusions are given"

H. pg 6434, lines 13-14: add commas before "by" and after "stratosphere", so it is clear that is it the inorganic forms that participate in the ozone destruction cycles.

I. pg 6434, lines 15-16: suggest "are called reactive species" and "are called reservoir species".

Actually, I am not sure why all of this detail is being given. Reads like parts of a Ph.D. thesis. All of this is known by nearly everyone reading this far into the paper. No worries leaving as is, but if more space is needed to address Major Point #2, then some of the details in this section can be considerably shortened.

J. pg 6435, line 25: suggest "causes a rapid increase"

K. pg 6436, line 20: should define LT if this is the first place it is used.

L. pg 6437, lines 3-5: these sentences are awkward and repetitive.

M. pg 6437, line 23: suggest "personal communication", and the year should be given.

N. pg 6439, line 18: suggest "despite fairly high"



O. pg 6439, line 23: "demonstrated the changes" is awkward, and should be improved.

Q. pg 6446, line 6: no need to capitalize "A" in "A rise"

Sorry this review is arriving on deadline day :(

Sincerely,

Ross Salawitch

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Interactive comment on Atmos. Chem. Phys. Discuss., 6, 6431, 2006.

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