

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

N. Sheode et al.

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Reply to the comments of reviewer 2 for the manuscript: acpd-2006-0093

We thank the reviewers for their useful comments. We have taken them into consideration and have made changes in the manuscript accordingly.

In the following, the comments of the second reviewer are written in *italics* and our reply to them is written in non-italics.

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.

As suggested, we have added in the abstract and also in conclusion the following text: 'The mean difference between the SCIAMACHY BrO and balloon BrO is found to be -15% (i. e. the SCIAMACHY BrO retrievals are on an average 15% lower than the corresponding balloon-borne BrO profile).' This hopefully also makes clear that -15% means that SCIAMACHY BrO is lower.

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 VLSLs of 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 the are the Sioris et al. retrievals, which exceed the balloon BrO profile by considerable amount. Statement of simple facts such as those above would add value to the paper.

We compare our BrO retrievals with balloon-borne BrO profiles in Section 3.3. In the same section now we also discuss the comparison of Sioris et al. (2006) BrO retrieval for the DOAS balloon profile on 23 March 2003.

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

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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.

In Section 3.3 we have now mentioned that we use the BrO absorption cross sections of Fleischmann et al. (2004) whereas Sioris et al. (2006) use the cross sections of Wilmouth et al. (1999) in the retrieval of BrO. Further we also state that the Wilmouth et al. (1999) BrO absorption cross sections are larger than the Fleischmann et al. (2004) BrO absorption cross sections. The use of different cross sections thus goes in the opposite direction of explaining the differences between our retrievals and Sioris et al. (2006).

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". 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.

Section 3.1 now includes an explanation about the way the tangent height (TH) correction is applied. Section 5 states that our tangent height correction is a post retrieval process in comparison to an independent retrieval of BrO from SCIAMACHY radiances by Sioris et al. (2006), who apply the TH correction during orbit by orbit retrieval process.

c) I have 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). 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,

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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 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 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.

We thank the referee for making this important comment. The points raised by the reviewer have been noted and stated now in Section 3.3.

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. This estimate truly comes from the work of Montzka et al., GRL, 2003. Appropriate citations to these organic measurements, upon which all estimates of Bry from VLSLs rely upon, should be added upon revision. Also, when an estimate of Bry from VLSLs is deduced by subtracting 15 pptv from Br_{total}^{mean} , there is an explicit assumption of no tropospheric loss of CH₃Br. We know that, based on the short lifetime of this species, that significant loss in the troposphere should occur, with values at the tropopause being perhaps 7% lower than global average surface values (Montzka et al., 2003). It is okay to assume no tropospheric loss of CH₃Br, especially because it is possible that the inorganic species produced following the tropospheric decomposition of CH₃Br make it to the

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

The citation of Montzka et al. (2003) is indeed needed. The citation has now been added in Section 6. Further, the explicit assumption of 'no tropospheric loss' has been stated in the same section.

Minor comments

1. *With so many citations given in Section 1 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.*

Kreher et al. (GRL, 1997) and Pfeilsticker et al. (GRL, 2000) have been added at the suggested places in Section 1.

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*

The citations have been corrected.

3. *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).'*

Changed to '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. *Does the retrieval algorithm assume that BrO varies diurnally? This is important*

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under some conditions according to Sioris et al. :) Somewhere, perhaps here, it should be stated whether or not this is taken into consideration.

The retrieval algorithm does not assume the diurnal variation of BrO. This is now stated in the Section 3.1.

5. 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? Also, it is unclear why the lower limit for Figure 1, right hand panel, is -45 and not a value closer to -70, as used for Figure 2. As drawn, Figure 1 right hand panel is clearly truncating information. Please clarify upon revision.

It is now explained in Section 3.2 how the shaded region is obtained. Section 3.2 now states: 'The grey area represents the total estimated retrieval uncertainty obtained as RSS of uncertainties due to retrieval parameters and measurement errors. This error estimation does not include uncertainties of BrO cross sections.' Further, the limits of Figure 1 have been changed and made similar to Figure 2.

6. 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.

The tangent height correction is now explained in Section 3.1.

7. 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.

A sentence informing the meaning of positive and negative percentage difference between SCIAMACHY and balloon profile has been added in the Section 3.3.

8. I do not 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

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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.

The sentence 'The reason for the discrepancy.....is still not clear.' has been removed.

9. *I understand this material, but it could be confusing to some that there is a 'decrease in BrO'. How about stating 'decrease in BrO/Bry'.*

As suggested the text now states 'decrease in BrO/Bry' in Section 4.2

10. *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.*

The text has been reorganised as suggested. The latitude band discussion has been moved appropriately closer to Figure 11.

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. correction would be helpful.*

The text now is more clear and states that 'Prior to climatology construction'. Tangent height correction has been explained properly in Section 3.1 now.

12. *Sioris et al. discuss the relation between BrO and NO₂ more extensively than any of the citations that are given. Perhaps a citation in this section, to Sioris et al., is warranted.*

The citation for Sioris et al., 2006 has been added.

13. *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'.

The citation for Sioris et al., 2006 has been added and we mention the differences between our results and Sioris et al., (2006).

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.

As suggested, the uncorrected balloon profiles have been removed from Figures 5-7. Also, all the SCIAMACHY values of BrO are shown in 'blue' and the photochemically corrected balloon profiles are shown in 'red'.

15. Figure 8 is unreadable and should be improved.

The font size has been increased. We hope that it will be possible in the final paper to reproduce the figure at a larger size.

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

In the caption the text has been changed to 'The blue points....'.

Minor suggestions related to grammar and trivial issues

All the other minor suggestions related to grammar and trivial issues have been taken

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into account.

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