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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 1 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 first reviewer are written in *italics* and our reply to them is written in non-italics.

Major comments:

1. The authors start the paper with a sensitivity analysis on factors and corresponding errors impacting the inferred BrO concentrations. This approach is highly acknowl-

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edged. However when further inspecting the details of the study it becomes clear that only the least problematic conditions were considered, i.e. BrO limb measurements in the tropics implying the smallest solar zenith angles (SZA), where multiple scattering, the curvature of the Earth, refraction et cetera play the least role.

We would kindly like to point out that in the previous paper by Rozanov et al. (2005b) a similar error analysis was already performed for a high latitude profile. We have decided to include in our current manuscript a sensitivity study for a low latitude profile because it may be more representative for the majority of the conditions considered in our climatology and in fact the tropical conditions are more problematic for the retrieval of BrO than the high latitudes. The main reasons are as follows:

- Due to the characteristics of the SCIAMACHY orbit, the scattering angle at high latitudes is close to forward scattering in the northern hemisphere and to the backward scattering in the southern hemisphere. This increases the portion of the single scattered light falling onto the detector as compared to around 90° scattering angle specific to tropical measurements.
- The multiple scattering at smaller solar zenith angles, typical for tropical region, is higher than at larger solar zenith angles due to a stronger illumination of the lower atmospheric layers, where the multiple scattered light is typically coming from. For example, a simulation at 350 nm using the SCIATRAN radiative transfer model (Rozanov et al., 2005a) for a standard atmospheric conditions yields a fraction of the multiple scattered light of about 55% at a solar zenith angle of 30° for tangent heights between 10 and 30 km, whereas at a solar zenith angle of 80° this fraction reduces to about 45%.
- Tropical profiles of BrO exhibit much lower values below 21 km as compared to high latitude profiles complicating the retrieval at low altitudes.

Finally, the effect of the curvature of the Earth and refraction is the same for high and

low latitudes and of minor importance for the direct solar beam.

These arguments support the reasonability to use a tropical profile to investigate the error budget and are also included into the text in Section 3.2.

In that respect an extension of the sensitivity study to the SZA dependencies (within the range of SZAs used) of all considered factors is highly essential. Also, since the errors are likely to be a function of latitude and season, the error discussion of BrO Limb measurements needs to be concluded with latitude/altitude/season error panels (similar the panels shown in Figure 9).

We appreciate the referee's suggestion to perform the sensitivity studies for a range of different latitudes and seasons. However this would require a substantial amount of additional calculations at this point that according to our previous results is not really needed. The sensitivity study performed by Rozanov et al. (2005b) performed for a high latitude BrO profile gives results which are comparable to the results for a tropical profile shown in our current manuscript. Hence we decided to include the sensitivity analysis only for the tropical conditions. We have now included a comment in the text that the results of Rozanov et al. (2005b) for a high latitude profile give similar results.

For the error discussion, it is also worthwhile to argue on the precision of the measurement (by Gaussian weighting of all possible error sources) to which the accuracy error(s) has (ve) to be added (c.f., by adding the BrO cross section error).

As pointed out such an error analysis has been done by Rozanov et al., (2005b). The text now includes a text in Section 3.1: The study of the theoretical precision of the BrO vertical profile retrieval has been performed by Rozanov et al., (2005b) and is about 10 – 20% in the altitude region 18 – 28 km, decreasing to 20 – 40% above 28 km and between 14 – 18 km, and rapidly degrading below 14 km.

2. With respect to the comment (1) and the known difficulties of Limb observations under twilight or even (polar) night conditions, it is hard to accept the shown BrO data

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beyond 65° north and south for midwinter observations (shown upper left and lower right panel in Figure 9). So this data potentially need to be skipped. In addition, whether a boundary for the observation of 65° can be accepted, or if in fact it needs to be taken at a lower latitude can only be decided when the investigation suggested in (1) is completed, and a limit for an acceptable error is assumed (explicitly mentioned in the text) and used in the study further on.

For the construction of our climatology, retrieved BrO profiles were considered if the solar zenith angle was less than 95 degrees. In practice there are only few profiles between 90 and 95 degrees. Note that the stratospheric layer between 18 and 30 km altitude that is considered here is still illuminated by direct sun up to a solar zenith angle of approximately 95 degrees. We have included some statements in Sections 3.1 and 5 to clarify this point.

3. *Taken together points (1) and (2) and the fact that BrO retrievals of SCIAMACHY from competing research groups (c.f. Sioris et al., 2006) come to different conclusions concerning an estimate of total Bry inferred from SCIAMACHY BrO, you need to address potential causes for this discrepancy.*

It is true that the recently published study by Sioris et al. (2006) comes to a much higher estimate of the total Bry inferred from SCIAMACHY BrO. This is consistent with differences in the retrieved BrO profiles between Sioris et al. and our results. Possible reasons for the discrepancies between the results obtained by different groups are still under investigation. This is being done in the framework of a joint international activity focused on the quality assessment and possible improvements of BrO profile retrievals from SCIAMACHY limb measurements. The statement to this effect has been added in the text in Section 3.3. We have included some additional comments how our results compare to Sioris et al. in Section 3.3. (See also our reply to referee 2.)

4. *The authors hastily ascribe the observed latitudinal variation in BrO (in particular at mid-latitudes) due to the photochemical interaction of BrO with NO₂ into BrONO₂,*

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while it is evident that the amount of stratospheric BrO is a function (a) release of bromine atoms from the precursor molecules (a process being presumably dominant in the tropics and at mid-latitudes), (b) (mostly vertical) transport (a process being presumably dominant in polar winter and spring), and (c) photochemistry (a process being presumably dominant at mid- and high latitudes). Since similar atmospheric processes are responsible for behavior of NO₂, and thus a radical/radical correlation (Figure 10) is in general insufficient to figure out which of the individual processes dominate where and in what season.

The points raised have been taken into account and have been included in the text. The text in Section 5 now states that: While the close anti-correlation between the observed BrO and NO₂ and the agreement with photochemical model suggests that NO₂ has indeed a strong control on BrO, it should be noted that transport processes also play an important role in the annual variation of BrO as well as NO₂. Although it is difficult to quantify the effect of the individual processes such as transport and chemistry, the observation of the anti-correlation between BrO and NO₂ confirms our present understanding of the gas phase bromine chemistry.

Specific minor comments:

A.) Section 2: change the main sinks of BrO is believed photolysis, reaction with O(3P), and reaction with NO₂ (instead of NO), or you should qualify the altitude region for which your statement holds true

Changed to 'and reaction with NO₂'

B.) Section 3.1: Provide a SZA range and error estimate for the following statement In the spherical mode the SCIATRAN.

The text has been added in Section 3.1, informing about the SZA range and error estimate for the statement, 'In the spherical mode of SCIATRAN....'

C.) Section 3.1: Provide information how you infer BrO in your Fraunhofer spectrum

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(taken at 33 km) and on the likely error of your estimate.

The information is provided in Section 3.1

D.) Section 3.1: Since your definition of y in equation (4) is different from usual, please provide additional an equation for your definition.

Equation (4) has now been explained in a more lucid manner with additional information.

E.) Section 3.1: Provide information of the SZA (or range of SZAs) and other atmospheric conditions for which you conducted the sensitivity study.

The text now includes the information of the SZA and other atmospheric conditions for which the sensitivity study was conducted.

F.) Change all Reaction (x , y or z) to reaction (x,y , and z)

As suggested, reaction (x , y or z) has been changed to reaction (x,y , and z).

G.) Change Dorf et al. (2005) to Dorf et al., (2006) throughout the text (Atmos. Chem. Phys., 6, 2483-2501, 2006)

Changed Dorf et al. (2005) to Dorf et al., (2006) (Atmos. Chem. Phys., 6, 2483-2501, 2006)

H.) Section 4.1: Skip the '!' behind BrONO₂

'!' removed

I.) Section 4.2: I doubt that BrONO₂ is 40% of Bry in the upper stratosphere (> 35 km) as stated at the end of the sentence.

The sentence has been removed. The text now states that BrONO₂ is between 10 – 20% in the UTLS region.

J.) Provide a figure for the averaging kernels before Figure 1.

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A figure for the averaging kernels is now provided along with text.

K.) *Figure 1 to 3: Provide information on the SZA and latitude of your simulation.*

The latitude of simulation is 3°N, 3°E and SZA = 37° at tangent point. This information is now provided in the caption of Figures 1 to 3.

L.) *Figure 8: The different lines and symbols are barely visible. Please plot the data on larger panels or color-code them. AND*

M.) *The same comment as made under L.) holds true for Figure 11.*

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

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

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