Atmospheric Measurement Techniques Discussions

Interactive comment on "Extending differential optical absorption spectroscopy for limb measurements in the UV" *by* J. Puķīte et al.

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Response to Referee #2

We thank Referee #2 for its generally positive review and very helpful suggestions for improving the article.

General comments:

The paper present a modified DOAS retrieval that account for the slant column densities wavelength dependence using a Taylor-series expansion. The authors demonstrated improvement to minor species

profile retrieval (BrO in this case), mainly using simulated profiles, and to some extent, real observation using SCIAMACHY limb scatter measurements. The paper is some how too long and some parts of the text are poorly written. I encourage the authors to consolidate sections 1, 2 and 3 into one or two shorter sections.

We agree with the Referee that the paper is (too) long and thank for the detailed suggestion to reduce the paper. We followed these suggestions and reduced parts of section 1, 2 and 3, mainly by skipping redundancy. Also, some of the studies could be combined and a part of the text and some of the equations are moved to the Appendix (i.e. Sect. 2.3). However we also think that the remaining detailed studies are relevant to fully describe the approach (e.g. we should first demonstrate the improvement in the fit of ozone SCDs and then show the related improvement for the profile retrieval of the minor trace gas BrO). We reduced some redundancies in the text (especially in the introduction and by rewriting Sect. 2.2 and Sect. 3). We also moved the description of the Taylor series approach for the AMFs (previously Sect. 2.2.2 and 2.3) to the Appendix. We also restructured Sect. 5 and excluded the side studies w.r.t. the different cross-sections from Sect. 5.3.

In general I was disappointed to see a limited discussion for the retrieval results using real observations, which was presented in section 5.3, as compared to the simulation results which were well presented and discussed. This section is poorly written and somehow difficult to follow. The paper could benefit from proofreading preferably by an English native speaker.

In our opinion the paper is meant to present a new method to account for the wavelength dependency of the SCD in DOAS retrievals. The verification of this new approach can be demonstrated best for simulated spectra, since for real measurements combined effects of many factors like measurements noise, other instrumental issues, impact of different geophysical parameters, etc., and even unknown causes usually can make the interpretation ambiguous.

Moreover, improving the retrieval is complicated by the fact that the true profile is gen-

erally not known. Also, when comparing to observations performed with another instrument, additional problems arise, in particular for the retrieval of the minor trace gas BrO which has only small optical depth and for which there are still large uncertainties regarding its amount in the lower stratosphere. Therefore the main focus is on the retrieval from simulated spectra. However, after verifying the approach for the simulated spectra we want to show that the method can be applied successfully also for real observations. Therefore we included as an example a comparison to all available LPMA/DOAS measurements performed for the validation of the SCIAMACHY BrO limb profiles.

These studies show that 1) the difference between the results for the new approach and standard DOAS is very similar as for the simulations and 2) for 3 of 4 cases the new approach is in better agreement with the validation measurements. We agree that these findings were not presented clear enough in the original article (mainly because the discussion on those factors like different BrO cross sections and the temperature effect was distracting the reader and moreover was also in itself confusing). In order to make these findings more clear we rewrote the section on the real measurements and strongly reduced the discussion of the points which are out of the main focus of the article and caused the confusion (see also comment to point on the BrO cross sections and the temperature effect below).

We revised Sect. 5, 5.2 and 5.3 where we now focus more on the improvement by the Taylor series approach with respect to standard DOAS. First we discuss the differences between both approaches for SCIAMACHY itself, include the comparison of them in the left panel of Fig. 10 (Fig. 9 in the new manuscript). Then we extend the comparison to balloon measurements. We also mention already in the introduction of Sect. 5 that for the comparison of retrievals of minor absorbers from different instruments, discrepancies can be expected.

Major revisions of the text are required for the manuscript to be published.

We revised the manuscript as suggested by this (and a second anonymous) Reviewer i.e. with respect to the length (by reducing its redundancy and also eliminating or moving some of the studies to the Appendix), improving Sect. 5, making additional proofreading for English, as well as considering the specific comments by the Referee.

Specific comments:

Page 2922, line 10: "... e.g. ozone, clouds, aerosols, albedo etc.)" Replace it with "e.g. ozone, as well as clouds, aerosols, albedo etc.)"

We correct it as: (e.g. ozone), as well as clouds, aerosols, albedo etc.

Page 2923, line 27: "The method is applied for simulated and measured spectra for different fit windows." The different fit windows were only shown for simulated profiles, not real measurement.

To correct this misunderstanding we changed the text to: "The method is applied for simulated spectra for different fit windows to investigate its importance and performance for different spectral regions. Also, to verify that the approach can be applied for improved retrievals from actual measurement data, we compare the obtained BrO profiles to those from correlated balloon observations performed for the validation of SCIAMCHY limb measurements".

Page 2938, section 3.2.2: can you state the retrieval altitude range where it is independent of the a priori?

In section 3.2.2 we write that the measurement response is close to unity for altitudes above 13 km. This means that the impact of the a-priori is minimized for these altitudes. We modify the respective sentence to make it clear: "...is close to unity at altitudes between 13 to 35 km for all of the three approaches ... i.e. thereby minimizing the impact of a-priori."

Page 2948, line 5: "The agreement is within around 25% for .." Change to "The agreement is within 25–35%.."

For the Kiruna comparisons, the agreement is between 0-40% depending on altitude

and profile, showing a rather large fluctuation. In the article we wanted to provide the mean value for the agreement, which is certainly better than 30%. We changed the sentence to: "For wide altitude ranges, the discrepancy is within 25%, for certain altitudes also below 10%, and always within 40%."

Also change line 10 to "top) and 10% at Aire sur l'Adour ..."

We changed as suggested.

Page 2948 line 15-19: "We found that the later one is by 10% larger ... Therefore, by a similar factor higher concentrations would be expected to be retrieved from the balloon if they were analysed using the Fleischmann et al. (2004) cross section." Can you support this claim? i.e showing a 10% bias of the retrieved profile after multiplying your cross sections by 10%?

The quantity fitted in the DOAS retrieval is the optical depth τ, which is the product of the SCD S and the cross-section $\sigma : \tau = S \cdot \sigma$.

Therefore, a simple multiplication of the cross section will lead to a similar change in the retrieved SCD (and this is also the case for simulated and measured spectra).

The complication in the comparison study is that three different BrO cross sections were used (Wahner, Fleischmann, Wilmouth). Moreover, the retrievals are not performed for the same wavelength ranges. The difference between these cross sections is not a simple multiplication, but wavelength dependent, i.e. for one wavelength the peak of cross section a is larger than b while for another wavelength the peak of the cross sections b is larger. Moreover, also the spectral resolution of the cross sections is different, which also affects the retrieval. Therefore, the fit of the cross sections in one fit window can not be extrapolated to the actual retrieval results in another fit window. Although this possibility was not claimed in the article, we wanted to give at least some rough estimate of the effect of the different cross sections applied in the different retrieval algorithms. However, since the discussion of this effect is actually far out of the scope of the article, we now note in the introduction of Sect. 5 that: "...in compari-

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son to the simulation studies larger errors are expected for real measurements. These include effects of the temperature dependency of cross-sections, the spectral calibration, the Ring effect and instrumental problems. For the comparison of measurements from different instruments, trajectory modelling and photochemical correction for balloon measurements contribute to additional uncertainties. Also note that for the balloon retrieval, different cross-section (Wahner et al., 1988) for the DOAS fit was used and the retrieval was performed in another fit window..."

Page 2948, line 24–29: you are trying to support the same argument above using wilmouth cross sections that are 10% larger, and claiming a 10% lower retrieval compared to the retrieval using Fleishmann cross sections. Figure 10 shows only a difference of 2–3% at most, not the 10% claimed in the text, which contradict your argument.

Please revise those two paragraphs.

Please see answer to comment above.

Our opinion is that this study and discussion is leading away from the main subject of the article and is not affecting the main conclusions of our study. Therefore we decided to skip this side study.

Page 2949, line 24, page 2950, line 3: "We also found that ..." to the end of the paragraph. How difficult is it to use the temperature dependant cross sections in your retrieval? Assuming you already have a good estimate of the temperature, can't you just apply it for the 4 cases and present the results instead of guessing the temperature that gives the best agreement with the balloon measurements? This paragraph is at best confusing!

We agree that this paragraph is very confusing and excuse for the related misunderstanding. In the article (section 5.2.) we wrote that the retrieval using "the ozone cross section at 203 K as second term" (instead of 243 K) "gives better results for cases with colder temperatures". This was the case for the Air sur l'Adour measurement, where temperatures close to 200 K occurred.

However, this was obviously not written clear enough.

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For the temperature range relevant for the retrieval of stratospheric profiles, ozone cross-sections measured by Bogumil et al. (2003) are available for the temperatures 203, 223 and 243 K. In the studies of chapter 5, we applied in our retrieval algorithm different combinations of cross sections and showed (for the example of the Air sur l'Adour measurement) that better agreement with balloon measurements is found when in fact the temperature best agreeing with the actual temperature profile of the observation is used (taken from ECWMF data). Thus, the approach suggested by the referee (applying the temperature profile of the individual measurement) is already performed.

On the other hand, the inclusion of a full temperature dependent retrieval is not possible within the time of the article resubmission but could be implemented in the future. In case studies, however, we found that the effect of this correction is small and far less than the improvement obtained by the Taylor series approach.

In the article we now write: "In the first step, the retrieval of SCDs, we apply the same retrieval settings as described in Kühl et al. (2008). The fit window ranges from 338.01–357.25 nm and two ozone cross-sections (at 223 and 243 K) by Bogumil et. al, (2003) are included in the fit in order to account for the temperature dependency of ozone cross-section. In the current study, the cross-section at 243K is replaced with the one at 203K for cases when temperature drops below 210K in the lower stratosphere in order to apply the cross-section best fitting to the actual temperature profile."

Page 2949, line 4-14: "For the tropical case ... ...to standard DOAS"

An attempt to explain the poor comparison of the retrieval with the balloon tropical measurements leave the reader only wanting more! The claim that Rozanov's retrieval sees similar bias is not a real explanation, and the authors need to discuss the sources of this bias properly.

Unfortunately we have no explanation available for the disagreement for this case. We only note that there are still considerable uncertainties regarding the observations of BrO writing: "Currently we cannot provide an explanation for this discrepancy, except that there are still large uncertainties regarding the observations of stratospheric BrO.

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In particular, only very few measurements have been performed so far for the tropical lower stratosphere (Dorf et al., 2008).". In this respect, the agreement with the retrieval algorithm by Alexei Rozanov, which intrinsically does not encounter the systematic errors due the wavelength dependency of the SCDs, indicates that at least the two different SCIAMACHY retrievals are consistent.

As suggested by Referee #1 we discuss that the improved agreement is expected, because the global fit approach intrinsically correctly considers the wavelength dependence of the SCDs: "...for this case, also other SCIAMACHY retrieval algorithms retrieve lower concentrations from the SCIAMACHY measurements (Alexei Rozanov, personal communication, 2009). Note that for all compared BrO profile retrievals the agreement with the profiles obtained by the IUP Bremen full retrieval approach (Rozanov et al., 2005) is improved when applying the Taylor series approach compared to standard DOAS (compare with the results presented in Sheode et al., 2006). The improved agreement is expected, because the full retrieval approach by itself correctly considers the wavelength dependence of the SCDs."

The statement afterward "Note that for all compared BrO profile retrievals the agreement with the profiles obtained by the IUP Bremen global fit approach ..." Should be deleted unless the authors can provide a valid reference to support it, or show a proper comparison.

There is a manuscript by Rozanov et al. in preparation that will show and discuss these comparisons, and this was meant to be cited here. We regret that because of a typing & text formatting error it was referenced in the manuscript incorrectly as Rozanov et al., 2005. For some of the balloon comparisons, BrO profiles obtained by the IUP Bremen approach are shown in Sheode et al., 2006 which we now add as a reference.

Figure 1: Need to change the color of the right y-axis of the right panel, not sure of which color.

We make it darker. We also add red and cyan for the left y-axis in order to more easy distinguish the relations between the plotted lines and the axis.

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There are many grammatical errors and sentences that need to be rewritten. As I motioned above, the manuscript needs a proofreading preferably by an English native speaker.

We thank Reviewer #2 again for the comments which helped to improve the article. We rewrote many parts of the article (see points above) and also additional proofreading by an English native speaker was performed.

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