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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Balloon-borne Limb profiling of UV/vis skylight radiances, O_3, NO_2 and BrO: technical set-up and validation of the method" *by* F. Weidner et al.

F. Weidner et al.

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We are grateful to the overall positive comments and suggestions given by the reviewer. Please find our point-to-point reactions in italic below. For a version of this comment including all figures go to http://www.iup.uni-heidelberg.de/institut/forschung/groups/atmosphere/stratosphere/publications/pdf/response_to_referee1.pdf.

Anonymous Referee #1

Received and published: 10 February 2005

1) Does the paper address relevant scientific questions within the scope of ACP?

Yes.

2) Does the paper present novel concepts, ideas, tools, or data?

Yes.

3) Are substantial conclusions reached?

Yes.

4) Are the scientific methods and assumptions valid and clearly outlined?

No, see below.

5) Are the results sufficient to support the interpretations and conclusions?

see below

6) Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)?

Not really – no description of profiling technique and hard to get information on RT code.

7) Do the authors give proper credit to related work and clearly indicate their own new/original contribution?

As far as I can tell, yes.

8) Does the title clearly reflect the contents of the paper?

Somewhat – see comments regarding the word "validation".

9) Does the abstract provide a concise and complete summary?

Yes.

10) Is the overall presentation well structured and clear?

Yes, but additional information in the figures would make life easier. See below.

11) Is the language fluent and precise?

Yes

12) Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?

Yes

13) Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated?

No

14) Are the number and quality of references appropriate?

Yes

15) Is the amount and quality of supplementary material appropriate?

N/A

Overall

My overall impression is that this is potentially an interesting and valuable paper but that it needs considerable work. I see three major issues that need to be addressed:

"Validation of the method", from the title. I do not think a method can be validated with just a description and a couple of favorable comparisons, particularly since the limb scatter technique is so new. Furthermore, there is no description of the method! There appears to be a total of three sentences describing the method – completely unacceptable.

We feel somewhat differently.
First we think we carefully demonstrated, validated (and cross-checked) each step of

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the method and the involved retrievals, however we admit for only a few gases and profiles. For the reasons see below. For example for ozone, we show that the measured SCDs agree very well with the forward modeled SCDs using otherwise measured ozone profiles (Figure 7). Next, we show that the averaging kernels of the profile inversion (as they turn out to be) when using the a posteriori solution technique described in Rodgers. At this point, we concur that a concession to the reviewer's criticism (and to the readability of the manuscript) should be made, c.f. by explaining in more detail (and once more) a well-established inversion method, which is frequently used in natural sciences (e.g. tomography). So we will add some text here.

That is, the method of retrieving profiles involves spectral fitting to obtain SCDs which is fairly standard and there is some discussion of this.

This statement is largely in contrast to our own experience, c.f., when reviewing studies on remote sensing of atmospheric trace gases such as BrO, or even on NO_2 (we only partly concur when it comes to ozone measured in the visible spectral range).

However, the inversion of these to obtain profiles is new and difficult and to requires a thorough discussion of the technique, the assumptions, uncertainties, etc.

In order to state it very clearly already at this point: The a posteriori method requires
(1) to assume an a priori profile, to which the weight can be set zero when the measurements are very sensitive to the parameters which are going to be inferred. This is the case in our investigation. In consequence need not assume anything for any of the inferred profiles in the height range traversed by the balloon. This sensitivity towards profiling is a particular strength of the balloon vs. c.f., ground-based or satellite measurements. In our study assumptions are only made for the profile shapes above balloon float altitude. This altitude range is however anyway not within the scope of our study, since balloon measurements are hardly sensitive to profile shapes above balloon float (see Figure 8).
(2) to include an error matrix before inversion. In our case, the matrix elements of the

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error matrix contain the uncertainties and errors that come from each individual observation i.e., mostly the least square errors of the spectral fitting. We understand that the small errors of our profiles are somewhat puzzling for non-ballooners, but please inspect (a) the spectral features and its likely error, (b) the many observations (measurements per inferred profile), and (c) last but not least the largely constraint profile information obtained from in-situ limb technique (Figure 8). All these factors render the balloon observations to become so accurate. Finally, in order to assess the total error of the measurements, the errors of the inversion, the uncertainty in the absorption cross section, and the contribution of the residual absorption in the Fraunhofer spectrum (to which all measurements are referred to) are Gaussian added. These errors are shown in the Figures.

Getting back to the title, I suggest changing the title to "...set-up and description of the method" or "...set-up, description of the method, and sample applications"

Probably, at this point there might exist some confusion. It is clear that the philosophy of a validation is (and has to be) different when it comes to balloon compared to satellite observations for the following reasons:
While the former observations usually have in total much less profiles at hand, these tend to be intrinsically more accurate than obtained by satellite observations (see above). This is simply because balloon payloads dive through the profile of interest which results in much more sensitive profile information (see figure 8) than obtained by 'outside observers'. We know that this fact is hard to accept by many remote sensing people (but well accepted by funding agencies when they have to spend a lot of money for balloon flights). Conversely, satellite measurements tend to be less precise at the sake of obtaining much more observations (which some use to do statistics for validation purposes but conversely remote sensing ballooners would never call such an approach a validation).
Therefore, since in our case one-to-one inter-comparison of inferred profiles is possible, we feel it would be useless to show comparisons for all the performed measurements

because this would not provide more information (since the comparison are equally good than showed) to the reader than what is already provided.
In all these respects we can understand the comment of the reviewer, but nevertheless we feel we should stick to the title, and the chosen approach.

2. Essential to retrievals using limb scattered light is the RT code. There were a couple of unnerving characteristics about the Tracy model – see points below. The only reference to it was a PhD thesis and it was not readily found on the internet;
I suggest making the salient parts of it accessible (and give a link). I would strongly encourage the authors to make detailed comparisons with a proven limb RT model to diagnose some of these discrepancies.

We also apologize for inconvenience the missing information could have caused!
Here is a www link http://www.iup.uni-heidelberg.de/institut/forschung/groups/atmosphere/stratosphere/publications/pdf/Dissertation_CvFriedeburg.pdf from which the PhD thesis of Von Friedeburg can be downloaded. We will also include the same link into the revised manuscript. In the Von Friedeburg study, c.f., you will find a detailed description of Tracy, and comparison exercise with the Bremen SCIATRAN, or former version of Tracy (Marquardt et al.) validated against spherical DISTORT.

3. Uncertainties and Error bars. Where do these come from? Figures 2, 3, 7, 9, 10, 11, 12 all show error bars on the observations and/or model calculations yet there is no mention of them anywhere.

(see our response above)

Other points

- For NO_2 and BrO how are diurnal effects handled; that is, while they are minimized along the LOS due to the viewing geometry, the fact that the observations are made for SZA_=90° means the incoming light has passed through air at smaller SZAs and hence probably less NO_2 and more BrO. Is this accounted for and if not what are the potential errors?

We clearly see the point of the reviewer, but the following facts should be kept in mind: We only show profiles from balloon ascents conducted at lower SZAs than 90°, where photochemical changes are not yet important for both radicals. This information is drawn from our own photochemical model that is well validated against the SLIMCAT CTM (Chipperfield, 1999, and please check to our studies cited in the manuscript). Also these simulations show that the (still very small) photochemical changes during our observations affect both observation modes (limb and direct Sun) equally, and thus are irrelevant in the inter-comparison study. Conversely, because of this difficulty with spectroscopic radical observations at twilight, we are still refraining to do more complicated (and awesome) inversions for the limb scanning observations of radicals, performed at twilight. This will be the scope of a future study. Also, at this point the reviewer should know that our observations were largely dictated by the wishes and needs of the other instruments (LPMA-IR and UV/vis DOAS spectrometer) deployed on the host payload. These measurements intentionally go for lower sun observations (solar occultation), while a particular strength of the limb technique should be the higher sun observations. For the latter, however we already allocated flights on different payloads with better Limb observation suited trajectories than in the past.

BoxAMF: AMFs usually imply a RT calculation at a single wavelength, is this the case here and if so it is dangerous to do so over a 50 nm fitting window (as for NO_2) due to non-linearities in the growth of the optical depths as a function of tangent height/altitude. I would suggest you forward simulate the entire fitting window at some reduced resolution.

We are aware of this problem, and actual one of the authors (e.g. Pfeilsticker et al., JGR, 104, 4101–4116, 1999.) is using this 'non linearity' effect for inferring photon path length in cloud sky RT. However, when it comes to spectroscopy in optically thin media (absorbers) the non-linearities are not (very) important for the RT, at least not to a degree, which matters here.

We also simulated the NO_2-SCDs at several wavelengths (see figure http://www.iup.uni-heidelberg.de/institut/forschung/groups/atmosphere/stratosphere/publications/pdf/fig11.pdf). It shows no significant change over a quite wide spectral range, so using the center wavelength of the fitting window should be a very good approximation:

Nadir: it is mentioned several times but no results are shown.

Nadir observations will be reported elsewhere.

Page 7642, line 7–8: I have not heard before of a substantial amount of stratospheric aerosol in the 0.01 mm size range. At 360 nm, the size parameter is less then 0.2 which makes these Rayleigh-like and you would need a huge number of then to substantially alter the radiance. (Checking out the Hirsekorn reference it seems that the impact of these small scatterers Overall I would argue that this is an extra-ordinary claim and you need to base your ascertain on refereed literature.)
In any event, with the observation point at 12 km adding extra aerosol should lead to a decrease in limb radiance so I don't think this argument is even consistent. Also, should the larger difference not be at 490 nm then due to the larger relative role of aerosol scattering? Finally, why not try adding a small aerosol to the simulation just to see what happens.

In order make a very long story short please consider the following: We believe we do have two indications for a larger than previously thought fraction of small stratospheric aerosols (we prefer to call it suspicion and not a claim), however, we still need to tighten, confirm and substantiate our suspicion.
First our former diploma student M. Hirsekorn intensively worked on inferring the Mie scattering from our balloon-borne direct sun observations in the UV/visible spectral range. You may receive a copy of this thesis upon request.
The findings are the following:
(1) Compared to SAGE II and III, our Mie extinctions are up to a factor of 2 larger for

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collocated observations. At this point one should know, however that the most sensitive SAGE channels for aerosol detection are around 1022 nm. The SAGE Mie extinction in the visible spectral range is then inferred by using the little information on Mie extinction detected by the observations in the visible SAGE channels (little due to interfering larger absorption of NO_2, O_3, and H_2O) assuming a wavelength dependence for Mie scattering based on Mie scattering calculations of sulfuric acid/water aerosols. Conversely, we directly inferred stratospheric Mie scattering and its wavelength dependence within the wavelength range 420–650 nm from solar spectra observed from our balloon platform. Of course, we inter-compare the inferred Mie extinctions for the same wavelengths of both sensors.

(2) Further when inspecting the wavelength dependence of the Mie extinction measured by our instrument with those from the SAGE's and with Mie calculation assuming sulfuric acid aerosols, different curvatures for the wavelength dependence are found (see the figure: http://www.iup.uni-heidelberg.de/institut/forschung/groups/atmosphere/stratosphere/publications/pdf/fig12.pdf).

Accordingly, we believe that both findings can only be reconciled if a (somewhat) larger fraction of small aerosols is present in the stratosphere than what is generally believed. Worth noting is to recall here previous studies, which were indicating such a fraction of small (not necessarily made of sulfuric acid/water) aerosols (e.g., Turco et al., Stratospheric Aerosols: Observation and Theory, Review of Geophysics, 20, 233–279, 1982).

To come back to your remark: We repeated the RT calculation and concur to your statement that adding aerosols generally leads to a lower radiance compared to a pure Rayleigh scattering atmosphere with our measurement geometry (i.e. 90° to the sun). So aerosols cannot explain the observed discrepancies between measured and RT modeled radiance. We will change the respective passage in the manuscript.

Page 7642, line 19–20: If you are using a fully spherical, ray-tracing model the 1/cos(SZA) problem should not be an issue, and if they are you need to further explain, and would this not propagate into the retrieved profiles? Furthermore, the largest differences occur around 89.5° and agreement at 90° seems good. This does not seem to make sense.

First the error is a purely numerical and thus technical error, which comes into play when the RT calculations are approaching SZA=90°. In Tracy, the technical problem arises in a necessary approximation of LOS calculation (see Rodgers), which (almost) passes parallel to the boundaries of the different atmospheric layers. This error (or shortcoming of the model) is, however not really relevant in our study, except for the limb radiances measured and modeled at twilight which we honestly report on. Actually we were pleased to realize that the measured and modeled limb radiances for twilight conditions are found agree on an absolute scale to better than 20%. We would be very surprised if any of the known RT model came to such an agreement.

Figure captions: heights should be in metres, not km.

The decimal points were lost during the final editing of the paper.

Figure 2: it would be useful to attach some SZAs to the altitudes

We follow the reviewer's recommendation!

Page 7645, line 21: The fraction of multiple-scattered photons that is simulated, 5%, is too small. See, e.g. Oikarinen et al., JGR, 104, 31 261, 1999. Depending on SZA, wavelength, etc..., I would say a minimum of 15% and more likely 20–25%. The fact that you are floating at 32 km and the Oikarinen calculations were for a space-based instrument is not will impact this ratio only marginally.

First, thanks for providing the Oikarinen et al 1999 reference! We checked our results, and we are finding a fraction of 20±5% multi-scattered photons for the relevant observations in agreement to your statement. In the revised manuscript, we will change the number and add a reference to the Oikarinen et al., 1999 study! Thanks for the comment.

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Interactive Comment

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 7631, 2004.

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Comment