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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.

Anonymous Referee #2

Received and published: 23 December 2004

********** General comments **********

This is a very poor paper in its current form, hardly worthy of being published in ACP, one of the pre-eminent journals of our field. However, it is also a very important paper if indeed accurate BrO profiles can be measured by this balloon limb scattering instrument. Thus this work could make an extremely valuable contribution to the science of remote sensing of atmospheric chemistry after major revisions and additions are made and additional validated limb scanning profile measurements are included. These measurements should take place far from large chemical gradients (e.g. vortex

edge) so that the validity of the limb scanning measurements can be assessed without the need to excuse disagreements on the basis of atmospheric variability. The authors are encouraged to take the time required to effectuate these improvements and additions to their work.

A fairly good description of the instrument is provided. However, there is no discussion of the straylight characteristics of these instruments. Ocean Optics instruments, even with a simple cross Czerny-Turner design, are known to have significant straylight and this source of error will probably dominate over hot pixels and shot noise. An analysis of this source of error is needed in the next version of this paper as well as a description of any algorithmic remedies for this problem.

The description of the inversion process is completely lacking. It is not enough to cite Rodgers. The level of detail must be sufficient that I could repeat your analysis myself. Such details include the a priori profile and covariance matrix, number of retrieval iterations, retrieval vertical range, convergence criteria, etc. if optimal estimation is used. (It is unclear which inversion method is used) Particularly for BrO, the a priori covariance matrix and profile are not trivial since a good BrO climatology is lacking. Your results should show the a priori used in both retrievals (occultation and limb scattering) and indicate whether the same a priori was used with both sensing methods. At present, it is impossible to judge whether the BrO profiles agree simply because the first guess profile for limb scattering agrees well with the retrieved profile from occultation (combined with a lack of measurement sensitivity for BrO from limb scattering). The spectral detection of BrO is quite clear and this is quite encouraging.

The results presented here are insufficient. In particular, the results from the limb scanning do not provide a validation at all. In fact, the measurements or the model could well be invalid based on the small sample of discrepant results shown here. In fact, there appears to be a problem with the radiative transfer model because the authors suggest that 1/cos(SZA) could lead to difficulties and state that this is a "notorious problem" with RT models. The fact of the matter is that this is not a problem for pseudo-spherical

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limb radiative transfer models, let alone fully spherical models such as Tracy (supposedly?) because path lengths through shells have finite distances even at SZA=90deg in a spherical atmosphere. Any plane parallel approximations in the model should be eliminated before publication.

The measurements made at positive viewing elevation angles can only broadly be considered limb viewing since they are not tangential. Most of the vertical profile information in this paper comes from these near-horizontal measurements. The only figure showing limb scanning data from multiple tangent heights is Figure 12 and thus, the authors have failed to demonstrate the ability to accurately retrieve profiles of any of the species from limb scanning. I also would like to suggest that it would be more scientifically useful to show profiles of BrO, NO2, and O3 all from the same flight (namely Mar. 24, 2004).

I am concerned in general with the assumptions made regarding the absorbers above float altitude. The statement "the residual trace gas absorptions should be minimal" is especially not true for NO2 at the float levels you achieved. The number density peak altitude (˜30 km) probably lies near your float altitude. The reader needs to be convinced that residual absorption signatures in the reference are not significant and are adequately handled by using an independent, ground-based solar spectrum. Please confirm whether NO2 absorption has been removed from this solar reference spectrum by Kurucz et al. Quantify or illustrate the fit residuals from this step.

Since O4 and H2O profiles are not shown, they should not be mentioned in this paper. Their scientific quality from limb scattering is not well established in your work or elsewhere.

The discussion contains plenty of unsubstantiated speculation. Suggestions are made below to test some of these hypotheses. This paper should include some estimate of forward model input errors since the method is somewhat novel (especially for O3 and BrO profiling from balloon limb) The two obvious examples, almost suggested by the

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authors themselves in their discussion, are: 1) to perturb the SAGE3 aerosol extinction profile by the uncertainty profile and examine the effects on the retrieved ozone (,NO2, BrO) profile. 2) to perturb the sonde O3 profile by its uncertainty profile (if available) and examine whether this can explain the model-measurement O3 SCD discrepancy.

Some work is needed to improve the grammar despite the efforts of the acknowledged M. Long. It is unlikely that all co-authors have proofread this manuscript based on some of the expressions used.

********** Specific comments **********

The title should be changed to "...preliminary validation..." since the results are not sufficient to claim that both techniques (ascent and scanning) are definitely valid, to the point where your balloon limb scatter observations and/or retrieval algorithm can be used to validate future constituent profile measurements.

p. 7633 SCIAMACHY measurements do not require novel methods. OSIRIS measurements (e.g. Sioris et al., 2003 as referenced in your paper) require all of these steps (spectral retrieval, forward modelling and inversion). The inversion and spectral retrieval for limb scattering are clearly not novel. Limb radiative transfer modelling including multiple scattering and atmospheric sphericity has been done for ˜30 years. Backward Monte Carlo code papers were published in the early 1970s by Collins et al. e.g. Appl. Opt. 11, 2684, 1972.

line 24- DOAS does not give profiles. DOAS in combination with optimal estimation does.

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The period of a limb scan (e.g. time to scan from TH=30 km to some minimum tangent height) should be given. The expected tangent height precision (between successive elevations steps) and accuracy should be given. It should also be noted whether the CCD is integrating during elevation steps to give a sense of the worst-case vertical

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resolution.

p. 7636 Using the ~2000 pixel detector to achieve a sampling ratio of ~8 is commendable.

p. 7637 Please write whether Tracy includes the SZA variation in 3-D and whether a Henyey-Greenstein approximation is used for aerosol (and cloud) scattering. The Hasekamp et al. study is almost irrelevant since you are using a skylight I0 from float altitude. Your I0 will have similar degree of linear polarization as your spectra of interest, leading to a cancellation of scalar approximation errors to first order (see Sioris et al. Adv Space Res. 34: 780-785, 2004).

p. 7638 "'conventional' profile inversion". Is this onion peeling? Do you assume that concentrations of O3, NO2, and BrO are zero above float level in obtaining your path length matrix? I'm confused. It appears optimal estimation is used from the statement in line 5 but line 9 suggests a more conventional technique is used. Please clarify. Also note that onion peeling is inappropriate (unacceptable) for non-linear retrievals problem such as these. An iterative approach is absolutely required to overcome non-linearities due to strong Rayleigh scattering at low (tangent) altitudes.

p. 7639 The hot pixels are a major issue for BrO. I expect that one hot pixel in your retrieval range could bias your BrO retrievals significantly based on simliar previous experience in dealing with radiation hits in the south Atlantic anomaly. Could you estimate the magnitude of this source of BrO error by perturbing the measured radiance at a hot pixel by the magnitude of the largest pixel-to-pixel variation in the fitting window and then re-perform the retrieval? Your argument that noise is inversely proportional to the square root of the number of scans does not seem logical. For a single spectrum, it is true that the noise is dominated by random sources, but for co-added spectra, the instrumental performance will be limited by those pixels which have high dark current, since this is systematic, fairly large (on average, 41 electrons for 10 second integration or 0.082% according to your numbers) and not reduced by co-adding. This dominates

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shot and offset noise at N= 100 spectra. You should state that your calibration of the spectra includes removal of the offset and dark current if this is the case. You should also report how often you record dark current spectra (i.e. following every "bright" spectrum?) in flight.

p. 7640 An absolute radiometric calibration of 35% at 380 nm is quite poor. Please discuss why this accuracy worsens so significantly in the field.

p. 7642 I suggest that you include some very small aerosols to see if this can account for the wavelength dependent bias (i.e. difference at 360 nm) and back up your speculated cause.

line 28 you should remove O4, OClO and H2O since you do not show profiles of these species. OClO may not be detectable given your instrumental issues.

p. 7643 Fitting windows etc. should be put in the method section along with plenty of other missing algorithm information. It is not a result or a discussion topic.

The Voigt et al. 203 K O3 cross section is very noisy and far from the best available (see also J. Orphal: A critical review of the absorption cross-sections of O3 and NO2 in the 240–790 nm region, J. Photochem. Photobiology A: Chemistry 157, 185-209, 2003). I do not understand why you have chosen it, nor how you combine it with the Burrows et al. O3 cross sections in the analysis. Please provide the necessary details of your spectral fitting approach.

(Partly a technical comment) Why do you write "NO2 (T=217 K) from Harder et al. (1997) (T=217 K and 230 K)" and then show fitted differential optical depths for T=230 K. Is fitting two O3 or NO2 cross-sections simultaneously better than fitting cross-sections at different temperatures sequentially and interpolating the SCDs from each temperature to the local effective temperature. Or how does your approach compare with fitting cross-sections interpolated in temperature to the local effective T? My experience in the Huggins bands (BrO fit window) is that your method of simultaneous fitting

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of O3 cross-sections at different temperatures produces slightly larger residuals and lower adjusted-r2-of-the-fit than even a single appropriate temperature cross-section, presumably due to strong correlation between the cross sections at similar temperatures. Please provide evidence that this is not the case for you in any window in which you use absorption cross-sections of the same molecule at different temperatures. You also should quantify the sensitivity of BrO to your approach in handling the O3 temperature dependence by contrasting BrO profiles retrieved with different T-dependence approaches.

You should refer to the method used in calculating your Ring spectrum rather than giving a historical reference. If a theoretical Ring spectrum is used, describe how it was calculated with respect to the temperature dependence of the rotational Raman scattering cross sections (i.e. which temperature profile was assumed?) Also, your analysis consists of a ratio of spectra both containing Ring (I and I0 have Ring)? How are you accounting for this in your Ring modelling? Is a different approach taken to account for the Ring effect in the quantification of the residual absorption above float since the Kurucz spectrum is used as I0? How is this residual absorption included in the profile inversion for the limb scanning case? Note that the residual absorption optical depth depends on viewing geometry (e.g. for limb scanning) and will even vary to some extent in the fixed observation geometry due to SZA changes and multiple scattering considerations. Also diurnal changes in the residual absorption should be quantified for the ascent case where the SZA is changing significantly.

p. 7645 line 5 "It is a result..." -> To what does "It" refer? Why does solar occultation have smaller averaging kernels than limb scattering? This would be useful to show in a figure, along with a sound explanation of the physical cause.

“or a better visibility” -> “(i.e. greater visibility)”

Discuss the possibility that relative and/or absolute errors in tangent height determination are responsible for some or all of the differences shown in Fig. 12.

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p. 7646 The agreement in limb scanning geometry is not “reasonably good”. This statement should be altered. I consider the agreement to be poor to moderate. The “reasonably well understood” statement at the end of the paragraph is also an exaggeration based on your results.

Your measurements do not provide direct information on photolysis frequencies since nowhere near 4 pi steradians of solid angle are observed (i.e. your instrument has a narrow FOV). I suggest you revise this sentence and focus on the ability of the model, which if validated, can be used to directly answer open questions on photolysis frequencies.

The larger sensitivity from balloon limb than space limb is likely, but the statement as such is merely unsubstantiated speculation in the Conclusions section of this paper. The authors are encouraged to use their model to demonstrate the magnitude of the increase in sensitivity achieved by placing the sensor in the middle stratosphere rather than above the atmosphere, particularly for BrO and OClO. This demonstration would provide a valuable conclusion to this paper. Without such a study, the statement probably has no place in the paper since the increased sensitivity may well be only marginal.

p. 7663 Fig. 10 shows better agreement with the correlative measurement in the lower stratosphere than the previous version of the paper which I “pre-reviewed”. However, the method section has not changed. The reason for the better agreement should be discussed in the method section. In the caption, the SZA range should be given or the reader should be referred to Table 1.

Fig.10-11 Photochemical scaling of the profiles is required to match the slight differences in local time. This is critical for BrO (and of less importance for NO2), otherwise validation can only be considered qualitative.

********** Technical comments **********

Title: Limb should not be capitalized in mid-sentence. The title needs rewording. "limb"

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should immediately precede "radiances" instead of describing "profiling". How about the following: Balloon-borne profiling of O3, NO2, and BrO from UV/vis limb scattering: ...

ABSTRACT

No need to use symbols for temperature and pressure in the abstract. "and otherwise measured profiles" -> "correlative profile measurements"

INTRODUCTION p. 7632 You need to use the term "limb scattering" rather than "Limb" at least once in the introduction, because "limb" is too vague (e.g. limb IR emission, limb occultation, etc.) UV/vis should be spelled out at first (ultraviolet/visible) METOP-IASI is not a limb scattering instrument. If it is, please provide a reference. The Kerr et al. reference is not relevant. It relates to occultation. Some reference should be made to SME (Solar Mesosphere Explorer) which was the first space-borne limb sensor of the stratosphere and has contributed to our knowledge of stratospheric NO2.

p. 7633 line 2- "...scattered and reflected skylight..."-> "...skylight..." since skylight implies scattering. line 5- excessive and irrelevant references to Rozanov et al. and Kaiser and Burrows. The other three references are more than enough. In fact, the Eichmann ref is probably the only appropriate one in this context. It seems like some of the Bremen authors are trying to build up their "cited" statistics by self-referencing. This is in poor taste.

line 7 "The UV/vis Limb measurements..." -> "Profile retrievals from SCIAMACHY limb scattering measurements..."

line 8 "...methods and techniques..." -> "...methods..."

line 10 "...modelling and the verification..."-> "...modelling..." (see previous sentence, since the modelling requires verification, the verfication does not require verification).

line 12 "...pressure..."-> "...pressure,..."

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line 16 "...thus of the whole method, involved in the UV/vis Limb technique." -> "...thus of the whole UV/vis limb technique."

line 23 "as a function of height" -> "as a function of tangent and sensor height"

line 29 "the latter being used" -> "the latter to be used"

p. 7634 line 8 "companying"-> "accompanying" or "companion"

line 10 "radiative"-> "RT"

METHODS

line 19 "a stable optical imaging"-> "stable optical imaging"

line 20 "While the former characteristic" To what the "former" is referring is somewhat ambiguous. I assumed it was "stable optical imaging" but it could have been "low weight".

line 25 no need for reciting your previous experience to make the point that weak absorbers require high S/N. It comes across poorly...see comments above.

p. 7635

line 5 "thermo-stated" is not a word. Try "temperature-stabilized".

p. 7636

line 8 the slit dimensions need not be reiterated line 8 "collimated"->"collimating"

p. 7637

line 11 "P"->"p"

line 13 "full"-> "fully"

line 22 "...if available profiles of the atmospheric aerosol and cloud cover and ozone profiles..." -> "..., if available, profiles of ozone and aerosol and cloud particle num-

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ber density...". Does Tracy require particle number densities or simply extinction and absorption coefficient spectra?

line 26 1994-> 1984

line 28 "simulated wavelength..."-> "wavelength..."

INSTRUMENT PERFORMANCE AND CALIBRATION

p. 7638

line 26 "noise"-> "noise level" (occurs twice in this line). You should call this "electronic offset noise" if that is what it is.

p. 7639 lines 27-8 "...1987) in combination ... Labsphere is employed."-> "... 1987) is employed in combination ... Labsphere."

p. 7640 line 9 "into the optical axis given by light intake of the Limb" -> "into the optical axis of the Limb..." or reword.

line 27 "(490 nm)/(360 nm)^4" -> "(490 nm/360 nm)^4"

OBSERVATIONS AND FLIGHTS

p. 7641 line 9 "instrument" -> "instrument and method" Sections 3 and 4 can be combined with section 2. Sections 3 and 4 would become sub-sections 2.3 and 2.4.

RESULTS AND DISCUSSION

line 23 "actual" -> "actually"

p. 7642 line 4 “reasonably reproduced well” -> “reproduced reasonably well”

p. 7643

line 21 "29 746 km" -> "29.746 km" (The decimal was lost in my printout of this version. Perhaps I am missing a font set?) A decimal point appears to be missing in the captions of Figures 4-6.

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p. 7644 line 22 "...(on a 1 km...(see Fig. 8)". You need a second right parenthesis.

line 26 "described" Hopefully some description will be present in a revised version.

line 27 "aboard" -> "onboard". This error is repeated below.

p. 7645

line 11 "excellently compares" -> "compares excellently"

line 12 "compelling confidence" -> "confidence" (redundant)

line 13 "inter compare" -> "intercompare"

line 18 " Noteworthy is however that" -> "However, it is worthy noting that"

p. 7646 lines 1-2 “Therefore the low-in-ozone polar vortex air masses are coming more and more into theĚ” -> “The ozone-poor polar vortex air masses were increasingly occupying theĚ”

CONCLUSIONS

line 11 “Overall a reasonably good agreement” -> “Overall, reasonably good agreement”

line 18 “to be calculated” -> “to calculate”

line 22 Limb scattering measurements are not “quasi in situ” and should not be presented as such. If the authors can demonstrate that the path length through a 1 km shell is less than ˜0.5 km for limb scanning, they can justifiably claim the “quasi in situ” nature of their measurements, if they so desire. The long path lengths provided by remote sensing techniques provide information on large volumes of air which cannot be provided by in situ. Thus, in this sense, trying to claim a “quasi in situ” character is not self-serving.

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line 6 “various methods” -> “various steps”

Ackowledgements lines 15-16 “for English proof-reading the manuscript” -> “proof-reading the manuscript with regard to the English”

References p. 7648 line 33 “abd” -> “and”

p. 7651 Voigt et al. comes before Von Friedeburg alphabetically.

p. 7656 (partly non-technical) Fig. 3 is difficult to understand and of poor quality. The x-axis should be labeled “SZA at sensor” if this is the case. It is important to clearly distinguish between SZA at the observer and at the tangent point for tangential observations. The tangent point SZA is actually the conventional quantity for limb scanning observations. If the caption mentioned the azimuth difference angle (90 deg), the SZA definition would not be critical, since the SZA should be almost equal at the sensor and at the tangent point. Also, the error bars drawn by your plotting software are strange. The positive error bar starts at the bottom side of the square and the negative error bar starts at the top end of the square symbol. The software should plot error bars originating from the centre of the symbol. Also, there should be a description in the method of how the error bars are calculated for the simulations. This was omitted entirely. Four points does not qualify as a limb scan in my opinion. Is the scanning mechanism failing to perform as expected here?

p. 7657-9 Figs. 4-6 The tangent height of these measurements should be given.

p. 7664 Fig. 11 The axes on this new figure should have labels.

Sorry for my delay in reviewing this paper.

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