

Interactive comment on “Balloon-borne limb measurements of the diurnal variation of UV/vis absorbing radicals – a case study on NO₂ and O₃” by L. Kritten et al.

Anonymous Referee #1

Received and published: 12 March 2010

This paper describes a new method for the retrieval of the diurnal variation of UV-vis absorbing species like O₃, NO₂, BrO, OCIO from balloon-borne limb scattered light observations. This method, based on a Bayesian minimization technique, is applied to observational data obtained from a mini-DOAS instrument aboard balloon gondolas during flights around Teresina in Northern Brazil in June 2005. The inferred profiles have been successfully compared to in-situ ozone sondes and ENVISAT/SCIAMACHY limb observations, demonstrating the validity of the retrieval method presented here.

The paper is well written and clearly structured. I recommend its publication in AMT after addressing the following comments:

We are grateful to the referee for his/her constructive comments and the appreciation of our work. Below we respond to the specific/technical comments point-by-point.

Specific comments:

- 1.) Page 433, line 20: Balloon flights are listed in Table 1 and not in Table 4. Can the new Bayesian minimization algorithm be applied to all flights listed in Table 1? If not, it should be mentioned somewhere in the Table which flights are useful for this study.

Reply: We corrected the numbering of the table and added a column explaining the worthiness of a specific flight for our study.

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- 2.) Page 435, line 25: It is not necessary to put again the list of UV/vis absorbers since this one appears already in the Introduction.

Reply: We agree and delete the additional list.

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- 3.) Page 437: You should mention somewhere how the choice of the reference spectrum is done in practice. You should also show a plot of the retrieved and modeled O₃ DSCDs as a function of time since it is a paper dealing with NO₂ and O₃.

Reply: We changed the sentences (page 437, line 2 – 7)

“It is noteworthy that, in principle, any measured spectrum I_i can serve as reference spectrum I_{ref} . The choice of I_{ref} translates into a common offset for all ΔSCD_i ($i = 1, \dots, N$), which in particular implies that a negative ΔSCD_i is physically reasonable. Practically, a spectrum with low absorption is chosen as I_{ref} in order to increase the relative absorption and therefore decrease the relative error.”

to

“It is noteworthy that, in principle, any measured spectrum I_i can serve as the reference spectrum I_{ref} . It is not necessary that the absorption or SCD is zero, as shown before in Eq. 2 to 4. The choice of I_{ref} translates into a common offset for all ΔSCD_i ($i = 1, \dots, N$), which in particular implies that a negative ΔSCD_i is physically reasonable. Practically, a spectral analysis is performed using an arbitrary spectrum as I_{ref} in order to define a spectrum with low absorption, which is then chosen as final I_{ref} in order to increase the relative absorption and

therefore decrease the relative error. Accordingly this choice may differ for different absorbers “

We added a plot of retrieved and modeled O₃ ΔSCDs, as the referee suggested.

- 4.) Page 439, line 8: Referring to Schofield et al. (2004) is not relevant here since they also retrieve the chemical variation as in the present study (chemical modeling in the forward model is only needed if the chemical variation of the absorbing radical is not retrieved but provided a priori).

Reply: In the Schofield et al. (2004) study the modeled change of concentration with SZA for the changing SZA along the light path is used in the forward model of the retrieval. As in the case of our scattered light measurements the change in SZA along the light path is less than 1.21° for all viewing geometries and the relative change of the measured gases along 1.21° SZA does not exceed 1% during daytime, at a certain altitude we assume a constant concentration along the light path and thus modelling a change of concentration along the light path is dispensable.

- 5.) Page 443, line 9: A reference or a short description for the Labmos model should appear.

Reply: We added a short description of the Labmos model and refer to Bösch et al. 2003. “*A priori* profiles used here are inferred from predictions of our photochemical model Labmos (Bösch et al., 2003). This 1-dimensional photochemical model simulates the temporal evolution of certain molecules by executing a set of gas-phase, heterogeneous and photolytic reactions. Initial values are taken from the 3-dimensional chemical transport model SLIMCAT (Chipperfield, 2006).”

- 6.) Page 444, line 3: How do you calculate the number of degrees of freedom? By using the trace of the averaging kernels? Also according to the y-axis of Fig. 4, you have about 85 degrees of freedom. It seems to me to be an unrealistic large number.

Reply: We calculate the number of degrees of freedom by using the trace of the averaging kernels, as recommended on page 445, line 25. Since we infer ten profiles at once, the number of degrees of freedom of 101, actually translates to meaning 10 degrees of freedom for a single profile. We have clarified this in the text .

- 7.) Page 451, line 2: Do you have any explanation for this off-set of about 2km between SCIAMACHY and balloon NO₂ profiles above the concentration maximum?

Reply: We wouldn't call that an offset, because the maxima of the respective profiles are at the same altitude. Above 35km the mini-DOAS retrieval is to some extent depending on the a priori profile used, as is illustrated by the area and spread of the averaging kernels.

We add the sentence:

“The difference above 35km altitude may be due to decreasing sensitivity of the mini-DOAS measurement above balloon float altitude, which is illustrated by a decrease of the area of the averaging kernels.”

- 8.) Page 451, line 9: “. . . the balloon-borne retrieval takes into account the full range from 0 to 70 km”. For me, this sentence is a bit misleading because according to Fig 9, the information content from the balloon retrieval is only significant in the 10-35 km altitude range.

Reply: This statement is to discriminate our approach against other approaches where for the characterization of the retrieval only regions of positive averaging kernels are used. It means that no additional assumptions for the boundary of the retrieval range are necessary.

We complement the sentence to “Specific to the employed retrieval method the averaging kernels for satellite measurements (and therefore their area and spread) are only representative in a range from 11 to 42km, while for the characterization of the balloon-borne retrieval the range from 0 to 70 km is taken into account.”

9.) Page 456, line 7: Are the Johnston's cross sections available somewhere (web link)?

Reply: We are referring to Johnston et al. for the description of the I_0 -effect, not for a cross section. Unfortunately the manuscript is not available on the internet; therefore we change the reference to a published paper:

“Aliwell, S. R., Van Roozendael, M., Johnston, P. V., Richter, A., Wagner, T., Arlander, D. W., Burrows, J. P., Fish, D. J., Jones, R. L., Tørnkvist, K. K., Lambert, J.-C., Pfeilsticker, K., and Pundt, I.: Analysis for BrO in zenith-sky spectra: An intercomparison exercise for analysis improvement, *J. Geophys. Res.*, 107, 4199, doi:10.1029/2001JD000329, 2002.”

Technical corrections:

10.) Page 435, line 26: According to my copy of the Platt and Stutz's book, the year of edition is 2008 and not 2006.

Reply: We corrected the year of edition from “2006” to ”2008”.

11.) Page 438, line 20: Since it is the first time that 'RTM' is used, the meaning of this acronym should appear.

Reply: We agree and add the meaning of the acronym: “...is derived from RTM (Radiative Transfer Model) calculations...”

12.) Page 450, line 8: 'Envisat' -> 'ENVISAT'

Reply: Corrected for.

13.) Page 457, line 24: 'NO-2' -> 'NO2'

Reply: Corrected for.

Anonymous Referee #2

Received and published: 9 April 2010

General Comments

This is a nice paper that describes a novel technique for the retrieval of time dependent trace gas profiles from balloon-borne limb scatter measurements. It is generally well written and is certainly suitable for the scope of AMT. I recommend publication, especially if the following minor comments could be addressed.

We are grateful also to referee #2 for his/her constructive comments and the appreciation of our work. Below we respond to the specific/technical comments point-by-point.

Specific Comments

- 1.) I wonder if the current title is the best choice for this work. It seems that the novel work here is *in the presentation of the time dependent retrieval methodology for the balloon measurements*, and not in the measurements of the diurnal variation of NO₂ and O₃. I would suggest rewording the title to indicate this.

Reply: We change the title from “Balloon-borne limb measurements of the diurnal variation of UV/vis absorbing radicals – a case study on NO₂ and O₃” to “Time dependent profile retrieval of UV/vis absorbing radicals from balloon-borne limb measurements – a case study on NO₂ and O₃”

- 2.) The "proper choice" of reference spectrum is referred to several times. It would be helpful to discuss how this is chosen in practice. For example, is it always the largest elevation angle? Does it vary with species (i.e. what about for O₃?) according to the height of the number density peak and the float altitude?

Reply: See our reply to comment 3.) of referee #1.

- 3.) Page 438: Although more detail is provided in later sections, it would be useful to expand upon the calculation of K in terms of L and C in the introduction to 2.3 as it seems unclear how they are related after reading this section.

Reply: We bring forward Eq. 11 to the introduction of 2.3 and refer later to that Eq..

- 4.) Page 443, line 14: Can you comment on why the correlations in DSCD's are (or can be) neglected and what effect this may have?

Reply: We changed the sentence

“The diagonal elements of the measurement covariance S_e are the squared DOAS fitting errors and non diagonal elements are zero since correlations of Δ SCDs are neglected.”
To

“The diagonal elements of the measurement covariance S_e represent the uncorrelated errors of the Δ SCDs and are the squared DOAS fitting errors. Non diagonal elements represent correlations of Δ SCDs and are set to zero, according to the findings in Sect. 2.2.”

Accordingly we added in Sect. 2.2 (page 437, line 23):

“The squared 2σ error of the spectral retrieval represents the measurement error in the retrieval of profiles. Since the residual of the DOAS fit shows only minor systematical structures, systematical errors are not taken into account for the profile retrieval. Possible sources of error are a principle offset on all Δ SCDs or a drift with time. Both aspects are tested by the use of different reference spectra in consecutive retrievals.”

- 5.) Page 443, line 23: Is the technique for choosing the diagonal elements of the a priori covariance matrix new to this work? If not, please provide a reference.
Also, how is the number of degrees of freedom calculated in this case? I realize that the time dependence has a contribution in this case but the numbers seem very large. More detail regarding the degrees of freedom calculation and interpretation would be helpful.

Reply: We added Schofield et al.(2004) as a reference to the manuscript. See reply to referee #1, comment 6 for the degrees of freedom question – as you rightly postulate the retrieval of 10 temporal profiles accounts for the large total number of degrees of freedom – 10 per profile.

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- 6.) Page 447: Can you clarify what you referring to as Type I and Type II oscillations with specific reference to Figure 6? The statement referring to 13:15 UTC and 14:15 UTC may be confusing as it is written. It is not clear why the numerical calculation of K_{EA} would fail. Can you comment on this please? Also, I completely understand the blurring and shifting interpretation of the effect of the two types of oscillations discussed on pages 447-448; however, I cannot see these effect as the authors state are illustrated by the profiles in Figure 6. Finally, the authors state this effect is more pronounced in the O₃ retrieval. Then why not show the O₃ retrieval results in the figure?

Reply: We added an additional description to Fig. 6 explaining type I and type II oscillations. We cancel the statement “However, since K_{EA} is highly non-linear, its numerical calculation fails and we choose an alternative approach. We estimate the error covariance S_{osci} due to EA pendulum oscillations of the gondola via a sensitivity study.” We replace it with “However, since K_{EA} is highly non-linear, its numerical calculation would require an iterative approach. Alternatively we estimate the error covariance S_{osci} due to EA pendulum oscillations of the gondola via a sensitivity study.”

Sorry, the colours in the original plot were confused. We corrected it by exchanging green and black in the lower left and lower right panel. The black profiles show now the blurred and shifted profiles as they are seen on an oscillating gondola.

- 7.) I would suggest that the authors carefully consider the use of the word "validation" for the work presented in 3.1-3.2. This is not necessarily a "validation" in the sense typically used in this field.

Reply: We change:

Page 448, line 14

“Results and validation of the method”

to

“Results and inter-comparison to results from other methods”

Page 449, line 1

„O₃ validation“

To

“Inter-comparison of O₃ profiles from mini-DOAS to an *in-situ* measured O₃ profile”

Page 449, line 1

“NO₂ validation”

To

“Inter-comparison of NO₂ profiles from mini-DOAS to collocated measurements of the SCIAMACHY instrument“

- 8.) It would be useful to explore how the independent retrievals of single balloon scans (performed in a typical sense) before and after the SCIAMACHY measurement compare to the result obtained at the SCIAMACHY measurement time using this new technique.

Reply: We performed a retrieval leaving the differential aspects, with the same settings regarding covariance. The comparison of the profile derived by the new technique to those specific two profiles from scans before and after 13:15 UT is shown in Figure 1. The resulting profiles are same within the error bars but more oscillating in shape.

Since the difference between them will depend on how much the concentration is changing during the measurement time the comparison is not be representative for all possible situations, but the benefit will increase with a faster change.

The benefit of the proposed method does not only lie in the opportunity to derive a profile at any time during the measurement period, but also in the applied link between all measurements regarding the reference spectrum/measurement.

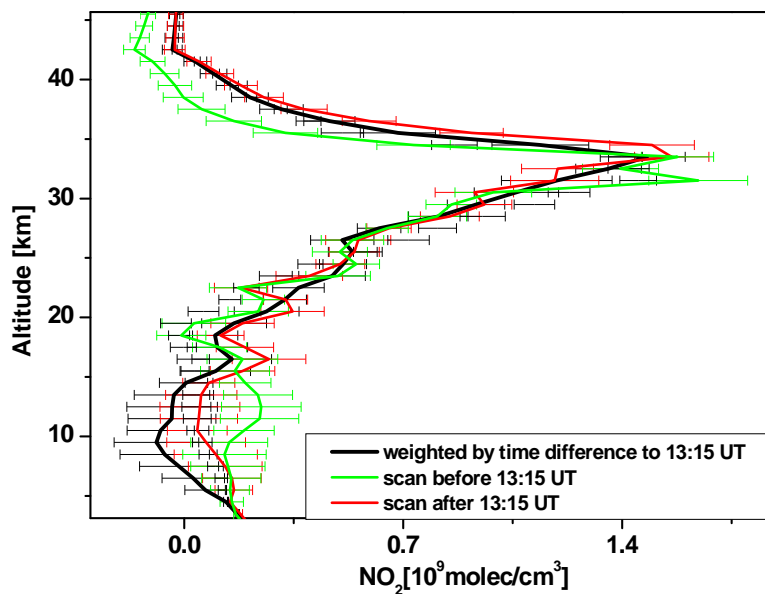


Fig. 1. Profile of NO₂ derived by the proposed method (black) from the limb scan before (green) and after (red) 13:15 UT.

Technical Corrections

p. 433 line 7: move the reference to Ferlemann et al. to a more suitable place later in this sentence

Reply: The according sentence is changed to “Balloon-borne solar occultation measurements have proven to be a particularly valuable tool for investigating stratospheric photochemistry (Ferlemann et al., 2000) and trends in stratospheric species, such as total bromine (Dorf et al., 2006).”

p. 433 line 20: do you mean Table 1?

Reply: This numbering was accordingly corrected to Table 1.

p. 436: Equations 2 and 3: I think the subscripts '0' and 'o' are getting confused.

Reply: We changed “o “ to “0” in equation 2 and the text above.

p. 440, lines 12-13: I would suggest "variability" rather than "variety", "background" rather than "common", and "influences" rather than "influence".

Reply: The new sentence is “This can be properly modelled in volcanically quiet periods, since the variability in the background stratospheric aerosol load barely influences the radiance distribution.”

p. 442: Equations 12 and 13: Is "!=" an accepted symbol?

Reply: We exchange "!=" by "=".

p. 443, line 443: Please be careful in what you mean when you state that the "a priori is a first guess".

Reply: We change “first guess” to “qualified first guess”.

Further changes (in black):

Page 447, line 23 - 28

“As illustrated by Fig. 6, EA oscillations with frequencies lower than the **inverse** time resolution of the measurements cause oscillations in the retrieved profile, a phenomenon similar to a blurred photograph. EA oscillations with frequencies larger than the **inverse** time resolution of the measurements lead to an enhanced contribution of light coming from lower atmospheric layers compared to the forward modelling assumptions.”

Page 449, line 7 - 8

As the ozone concentration is expected to vary little with time in the tropics, the time lag of **half a day** between the two measurements is negligible.

Page 449, line 9 - 11

For comparison, the higher resolution in-situ measured O₃ profile is degraded to the altitude resolution of the lower resolution mini-DOAS O₃ profile using the averaging kernel matrix **A** of the mini-DOAS.

Page 451, line 7

Both, the Bremen and the mini-DOAS retrieval are performed on a 1 km grid, while the Mainz retrieval is performed on coarser height grid of 3.3 km.

Page 451, line 20 - 21

Forthcoming studies will discuss implications of our measurements for stratospheric photochemistry.