Atmos. Chem. Phys. Discuss., 5, S5200–S5210, 2005
www.atmos-chem-phys.org/acpd/5/S5200/
European Geosciences Union

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Inter-comparison of stratospheric O_3 and NO_2 abundances retrieved from balloon borne direct sun observations and Envisat/SCIAMACHY limb measurements" *by* A. Butz et al.

A. Butz et al.

Received and published: 24 January 2006

We are grateful to the referee's overall positive comments and suggestions. Please find below our point-to-point reactions in italic.

General comments

1. This paper involves comparisons between a number of different data sets (LPMA FTIR, DOAS UV-Vis, as well as SCIAMACHY data retrieved by 3 different institutes).

Full Screen / Esc
Print Version
Interactive Discussion
Discussion Paper

EGU

For validation purposes, a key issue is to assess the accuracy of the correlative balloon measurements, as identified by the authors. The study presents a good analysis of the comparisons, however a proper estimation of the errors on each technique (independent from the intercomparison) is somewhat lacking. Typical uncertainty estimates should be provided for each technique prior to the comparison. This is especially important since the two techniques apparently have rather different error budgets. Since from the comparison one can only conclude on the agreement within combined error bars, it is essential to provide independent estimates of these error bars. In the present manuscript, these can only be roughly estimated by inspection of Fig. 4. Even there, error bars on raw (unsmoothed) DOAS results are not given.

The contributions to the errors of the DOAS and LPMA SCDs are listed in section 2.1, p.10754, l.11ff and p.10755, l.25ff, respectively. An example of the resulting error bars is shown in Fig. 1, where the relative errors of O_3 and NO_2 SCDs are explicitly plotted. The errors of the vertical trace gas profiles are calculated from the errors of the SCDs according to equation (3), where $\mathbf{S}_\epsilon$ explicitly contains the squares of the SCD errors.
However, as correctly observed by the referee, we do not state a general number for the accuracy of the DOAS and LPMA measurements since the actual accuracies depend on the observation conditions. Some examples:
- The DOAS errors for O_3 are governed by the error coming from the determination of the overhead amount of absorber. Hence, this error contribution will depend strongly on the altitude of the actual balloon flight and on the relative attitude of the O_3 profile.
- The importance of the fitting errors as a contribution to the LPMA and DOAS NO_2 errors depends strongly on the actual abundance of NO_2. For the flight from Kiruna in February 1999, for example, NO_2 abundances were very low and sometimes even close to the detection limit of the LPMA instrument. The corresponding error bars are dominated by the fitting error while for the flight from Kiruna in March 2003, Fig. 1, the fitting errors are a minor contribution.

- Since the pre-flight optical alignment of the LPMA instrument has been continuously improved, the analysis of earlier balloon flights usually yields larger SCD - errors than analysis of the recent ones.
Following the suggestion of the referee, the paper is supplemented by some numbers for the accuracies of the LPMA and DOAS measurements. According to the argument above, these are rough estimates, only:
p.10754, l.20: The errors of the retrieved O_3 SCDs are dominated by the latter error contribution while, for NO_2, fitting errors become important when NO_2 abundances are very low. In total, typical accuracies of the DOAS O_3 and NO_2 measurements are better than 5% and 10%, respectively.
p.10756, l.1: Since the pre-flight alignment and in-flight stability of the LPMA instrument improved during the suite of considered balloon flights between 1996 and 2003, the analysis of earlier balloon flights usually yields larger errors than analysis of the more recent ones. Typical errors of the retrieved O_3 SCDs range between 10% and 15% and are dominated by the accuracies of the spectroscopic parameters and the estimated accuracy of the instrumental line shape function. The errors of the NO_2 SCDs range between 10% and 25%. As in the case of the DOAS error budget, fitting errors become important for NO_2 when its abundances are very low e. g. for the flight from Kiruna in February 1999, where NO_2 SCDs are close to the detection limit of the LPMA instrument.
Fig. 4: The figure is changed to show error bars of all data sets plotted.

2. Along the same line, I think that the discussion of satellite validation results could be improved by adding more explicit comments on whether the observed differences fall or not within the combined uncertainties of transported and photochemically corrected balloon data and satellite measurements. Looking at the results, my impression is that satellite retrievals (at least those from Bremen and the 3 profiles from Heidelberg) are satisfying above 20 km, but not below. However it is difficult

ACPD
5, S5200–S5210, 2005

Interactive Comment

to realize whether the discrepancies at lower altitudes can be considered as due to errors in satellite retrievals or to problems related to the validation process itself (photochemical correction, horizontal smoothing of actual spatial inhomogeneities, etc.). Maybe it is not possible to conclude on this issue based on available data, but then this should be made clear and suggestions should be provided on how to improve on this in future studies.

The referee's observation that the satellite retrievals are satisfying above 20 km altitude and insufficient below, is underlined by the structure of sections 4.2.1 and 4.2.2. For both trace gases a paragraph is dedicated to the altitude range above and below 20 km. In addition, abstract and conclusion distinguish explicitly between the two regimes which are unambigously identifiable in Fig. 7 and 8. The latter two figures are supplemented by the combined error bars of satellite and balloon borne data in order to facilitate the comparison.
Reasons for the discrepancies at lower altitudes are discussed in detail in sections 4.2.1 and 4.2.2 (p.10773,l.28ff and p.10775,l.23ff) and summarized in the conclusion where the observed discrepancies are attributed to "the lower sensitivity of the satellite retrieval, uncompensated horizontal variations of the trace gases and in the case of NO_2*, modelling uncertainty" (p.10777,l.6ff). The relative importance of the listed contributions is hard to assess. An estimate of the sensitivity of the satellite retrievals to low altitudes can be inferred from the averaging kernels shown in Fig. 4c and d, which indicate that there is only very little information on the trace gas profile below 15 km. The impact of horizontal variations of the trace gas abundances strongly depends on the meteorological situation as outlined in the paper, but can be estimated by comparing* O_3 *profiles inferred from balloon ascent and solar occulation measurements of the same balloon flight. While the high resolution* O_3 *data from balloon ascent in Kiruna in 2004, Fig. 5e, excellently agree with the in-situ sonde data the corresponding solar occultation measurements, Fig. 5f, reveal some discrepancies with respect to the in-situ data below 20 km. This is a clear hint for sampling horizontally inhomogeneous*

air masses. The uncertainty of the photochemical model, should be well represented by the modelling error inferred from sensitivity runs.

Clearly, the validation strategy could be improved by performing dedicated balloon flights where the instruments sample nearly the same air masses as SCIAMACHY. For direct Sun measurements performed on a balloon which is subject to ambient meteorological conditions, this is impossible.

Fig. 7 and 8 are updated by the combined error bars of the satellite and balloon borne measurements. The following statements are added/changed.

p.10773,l.25ff: The combined error bars of the balloon and satellite borne observations are on the order of the observed standard deviation of all coincident measurements in the 20 to 30 km altitude range. However, a number of data points differ by more than the combined error bars which might point to a systematic error as suggested above.

p.10774,l.19ff: Fig. 7 reveals that the combined error bars cannot explain the observed discrepancies below 20 km altitude.

p.10775,l.17ff: In the 20 km to 30 km altitude range the agreement between the balloon borne NO_2 profiles and the satellite observations is on the order of 20%. $\rightarrow$ In the 20 km to 30 km altitude range the agreement between the balloon borne NO_2 profiles and the satellite observations is on the order of 20% and most often well represented by the combined error bars. The latter amount to about 1.5 to 3 times the observed standard deviation between the two data sets for all coincident datapoints in the considered altitude range.

p.10776,l.4: The combined relative errors shown in Fig. 8 increase dramatically with decreasing altitude since, there, the absolute abundances of NO_2 are very low. The relative errors of SCIAMACHY NO_2 measurements below 15 km typically are larger than 50%. Adding the rather large modelling error and the error of the balloon borne measurements, the combined error bars are often on the order of the observed deviation. Despite the large combined error bars, a systematic underestimation of the balloon by the satellite borne data is obvious.

p.10777,l.7: Since the origin of the discrepancies observed at low altitudes cannot *be unambiguously attributed to the satellite retrievals or the validation strategy, it is important for future studies to keep the spatial and temporal mismatch between satellite and validation measurements as small as possible.*

3. Given the complexity of the comparison methodology, I think that a graphical illustration of the technique would be useful. This requires an additional figure which could be inserted e.g. before Fig. 1, or before Fig. 4.

A schematic drawing of the comparison and validation strategy is added before Fig.1.

Specific comments

p.10750,l.9: replace "..., exploiting that O_3 and NO_2 absorb electromagnetic ..." by "exploiting the fact that O_3 and NO_2 absorb electromagnetic ldots "

Corrected.

p.10760,l.26: it is unclear how the temperature dependence is treated here. Are there 6 cross-sections included in the least-squares process to account for NO2 and O3 absorption?

p.10760,l.26:Absorption cross sections included in the fitting process are NO_2 and O_3, both at T=203 K, T=223 K, T=243 K from Bogumil et al. (2003), and the collisional oxygen dimer (O_4) from Greenblatt et al. (1990). → Absorption cross sections included in the fitting process are NO_2 and O_3 from Bogumil et al. (2003),

ACPD

5, S5200–S5210, 2005

and the collisional oxygen dimer (O_4) from Greenblatt et al. (1990). The temperature dependence of the NO_2 and O_3 absorption cross section is handled by performing three runs with absorption cross sections corresponding to T=203 K, T=223 K, T=243 K (Bogumil et al., 2003). At each tangent height the run which exhibits the smallest errors is chosen for profile retrieval.

p.10761 and before: it is unclear from the description of the satellite algorithm whether cloud effects are only considered by one group, or if this aspect of the retrieval has been simply omitted by others in their general description. Please check for consistency here.

For SCIAMACHY limb observations the impact of clouds on the retrieved O_3 and NO_2 profiles is small, in particular at altitudes above the tropopause where the bulk of presented data orginiates from. The present paper is not intended to compare the different SCIAMACHY NO_2 retrievals or their sensitivity to retrieval parameters e. g. clouds. Rather, emphasis is put on providing a valuable validation data set.
For consistency we changed the paper as follows:
p.10759,l.10ff: The forward simulations of the SCIAMACHY limb measurements and the calculations of the appropriate weighting functions are performed employing the SCIATRAN radiative transfer model (Rozanov, 2004; Rozanov et al., 2005). $\rightarrow$ The forward simulations of the SCIAMACHY limb measurements and the calculations of the appropriate weighting functions are performed employing the SCIATRAN radiative transfer model (Rozanov, 2004; Rozanov et al., 2005), assuming cloud free conditions.
p.10761,l.19ff: The weighting function matrix $\mathbf{K}$ is calculated by the fully spherical 3-dimensional Monte Carlo radiative transfer model "Tracy" (von Friedeburg, 2003; Weidner et al., 2005), assuming a cloud cover at 10 km altitude. Sensitivity studies show that the impact of clouds on the retrieval of stratospheric NO_2 is negligible.

ACPD
5, S5200–S5210, 2005

p.10767,l.19: The temperature dependence of the NO_2 cross-section is quasi linear in the 440 nm wavelength region, hence extrapolation (linear?) should not be a major source of uncertainty here. Anyway this could even be checked using the Bogumil et al. data for which measurements are available down to 203 K.

As proposed by the referee we checked our (linear!) extrapolation of the NO_2 SCDs to temperatures below 217 K using the Bogumil et al. (2003) data. The cross sections of Harder et al. (1997) at T=217 K and T=230 K are convolved by a Gaussian slit function of width 0.52 nm which corresponds to the spectral resolution of the Bogumil et al. data in our retrieval range. A correlation plot of the T=230 K cross section versus the T=217 K cross section between 435 nm and 485 nm yields a slope of 0.52% K^{-1}. A similar analysis of the Bogumil et al. (2003) data for T=223 K and T=203 K results in a slope equal to 0.41% K^{-1}. Hence based on the Bogumil et al. (2003) data, the temperature dependence below T=217 K is overestimated by about 20% when extrapolating the Harder et al. (1997) data using a scaling factor inferred from the cross sections at T=217 K and T=230 K.
The impact on the retrieved SCDs is tested for flight from Kiruna in 1999, where the most extreme case of extrapolation is observed. Assuming an error of 20% of the NO_2 temperature correction results in 3.5% maximum error of the retrieved SCDs.
Clearly, 3.5% error cannot acount for the observed dicrepancy between LPMA and DOAS SCDs. Therefore the following statement is changed:
p.10767,l.16: In this case, stratospheric temperatures were well below 217 K where NO_2 absorption cross sections from Harder et al. (1997) are not available. Thus, the DOAS NO_2 data have to be extrapolated when adjusting the retrieved SCDs to the effective mean temperatures along the line-of-sight. The extrapolation might not reproduce the true atmospheric state. → In this case, stratospheric temperatures were well below 217 K and an extrapolation of the Harder et al. (1997) data has to be used when accounting for the temperature dependence of the NO_2 absorption

cross sections. However, an extrapolation error alone cannot account for the observed discrepancy which has been tested using the NO_2 cross section from Bogumil et al. (2003) at 203 K.

p.10771,l.22: replace “artificial” by “arbitrary”

Corrected.

p.10772,l.1: In fact none approach is optimal in this case. I guess the choice of applying a stacked calculation at the location of the balloon flight was made essentially for simplicity reason.

As stated in the paper, the air mass trajectory model is not able to identify a backward coincidence between the balloon and satellite borne observations for Aire sur l’Adour in 2003 applying the 500 km spatial mismatch criterion. When extending the maximum spatial mismatch to 1000 km a coincident satellite observation can be found in the east of the balloon borne measurements while air masses move slowly from west to east. Hence, air masses are not transported from the satellite borne toward the balloon borne observations but are actually transported in opposite direction. The coincidence found by the trajectory model is not due to the correct representation of the air mass movements but rather due to the overall spatial and temporal proximity of the balloon and satellite borne measurements. In this particular case, we indeed consider a stacked calculation a more realistic scenario than the calculation along air mass trajectories, although "none approach is optimal".

p.10772,l.26: add “in the absence of available DOAS measurements” at the end of the sentence.

Corrected.

p.10775,l.4: I would move the sentence in L13-14 of this section right after the sentence ending by "... Fig. 8", in the following way: "Although a detailed comparison of the different retrieval exercises is beyond the scope of the present study, we note that the internal agreement of the satellite data . . . "

Corrected.

p.10789, Fig.2 caption: replace "The 1032 data points are grouped . . . " by "The 1032 data points corresponding to the 6 flights of Table 1 are grouped ..."

Corrected.

p.10792-10794: Figures 5 and 6 are quite complex and hardly readable as printed here. Fonts are too small (and distorted) and the different curves are difficult to distinguish. Please resize and reorganize the various plots to improve readability.

Here, we ask the referee to re-think the issue. Fig. 5 and 6 are designed for ACP, not ACPD. Unfortunately, ACPD format is somewhat different from ACP format. In particular, ACPD is published in landscape design, while ACP uses portrait. Fig. 5 and 6 will be about two times as large in ACP than in ACPD and not distorted.
Resizing and reorganizing Fig. 5 and 6 would imply to tear panels a) to e) apart and to include separate figures for all panels (in total 12). In its present shape, Fig. 5 and

6 illustrate the validation study for the two considered trace gases on one page each and the different panels can be compared easily with respect to each other, which would not be possible when splitting the figures.
Moreover, a second point of view onto the validation study is presented in Fig. 7 and 8 which emphasize the differences between the data sets and provide quantitative conclusions on the agreement observed.

p.10796, Figures 7 and 8: Since balloon data are reference data here, I think it would be more logical to plot relative differences w.r.t. balloon data, i.e. $(O_{3SCIA}/O_{3Balloon}) - 1$. Please take this just as a suggestion, in case the plot can be easily redrawn.

In most cases O_{3SCIA} *is smaller than* $O_{3Balloon}$ *below 20 km. It is true that,*

$O_{3Balloon}/O_{3SCIA} - 1$ *tends to large numbers for small* O_{3SCIA}*, while*
$O_{3SCIA}/O_{3Balloon} - 1$ *tends to* -1 *for small* O_{3SCIA}.

Hence, the present choice of the abscissa illustrates large differences between SCIAMACHY and the balloon borne measurements on a larger scale than the abscissa suggested by the referee provided that the SCIAMACHY profiles are consistently lower than the correlative balloon borne data. We prefer to keep the present illustration since we want to emphasize the two regimes above and below 20 km.

ACPD
5, S5200–S5210, 2005

Interactive Comment