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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Validation of SCIAMACHY tropospheric NO_2-columns with AMAXDOAS measurements" *by* K.-P. Heue et al.

K.-P. Heue et al.

Received and published: 4 March 2005

General remarks

First of all we thank the reviewers for their helpful comments and suggestions. Before we discuss the comments in detail we would like to give some general information concerning our new version.
1. We have discovered an error in the old correction of the spectrograph stray light. Unfortunately such errors can hardly be discovered in the DOAS analysis and we did not recognize them in the analysis of the old version. The updated data stray light correction is based upon the 2D information of the non illuminated parts of the CCD-chip. This approach has already been used successfully for similar systems and is described in detail in *Wagner et al. 2004. We found that the differential slant columns are about

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10% higher compared to the old version.
2. The AMF for both instruments AMAXDOAS and SCIAMACHY were recalculated with the same radiative transfer model. We added a sensitivity study on the mixing layer height as suggested by the first reviewer.
3. The criteria for matching SCIAMACHY and AMAXDOAS were defined in a different way. The important criterion is whether most of the AMAXDOAS pixel is within a SCIAMACHY ground pixel or not. Following the reviewers remark we average the AMAXDOAS measurements covered by one SCIAMACHY ground pixel.

Anonymous Referee 2 Received and published: 24 December 2004

GENERAL COMMENTS

The authors managed to convince the reader that AMAXDOAS measurements, when correctly analysed, are of great importance for the quality assessment of satellite DOAS retrievals, in this case for tropospheric NO_2 columns from SCIAMACHY. However, both retrieval techniques rely heavily on the atmospheric assumptions made in the air mass factor calculations. The authors argue that when the same assumptions are chosen for both methods, the products are comparable. And indeed they are comparable and it is valuable that they are compared, but this study cannot be called a "validation" study because of the following reasons: [...]

We are aware that a comprehensive validation has to go far beyond our study. However a complete validation study for a tropospheric NO_2 product won't be possible in the near future (if at all) because neither an official nor a standard scientific product exists. On the other hand we are confident that our study
1. provides a very well suited data set for a comparison with satellite data,
2. reports on observations with large dynamics in the tropospheric NO_2,
3. uses good settings for RTM based upon independent measurements.
It is true that both observations are based on the same concept and rely on the same assumptions concerning the RTM. Nevertheless these assumptions are based on in-

dependent observations and can be further adjusted as soon as newer information is available. It should also be noted that the comparison of similar observations can yield very valuable information. Just imagine we did not find such a good agreement between AMAXDOAS and SCIAMACHY. This would very well provide valuable information. AMAXDOAS observations are one method allowing such an extended spatial comparison on both slant and vertical columns. Because of these reasons we will keep the term "validation" in the title as well as in the text.

1) The product to be validated is SCIAMACHY tropospheric NO_2 vertical column, described by Richter et al (2004). This product is adapted for this study by using particular AMF settings for aerosol, NO_2 profile and ground albedo which should be representative for the particular situation. So the authors are not validating the original product, but a specially adapted version.

As pointed out by the reviewer (comment 17) an official product of SCIAMACHY tropospheric columns does not exist. As a comparison for the slant columns is shown as well, we think this objection only holds partly. Besides we added a sensitivity study on the NO_2 profile, and found only a weak dependency on this parameter within the studied range. This is a specific result for the high aerosol load. We also added a comparison to the Bremen tropospheric-near-real time NO_2-product.

2) The AMAXDOAS product "tropospheric NO_2 vertical column" with which the SCIAMACHY product is compared is not fully described or referenced, and also a validation of this product is not fully described or referenced. Therefore it is not clear what the validatity of the validation measurements is.

We pointed out that the reference (Wang et al 2004) to other AMAXDOAS measurements includes a comparison to ground based data and a comparison with ground based in situ data is added as well. A sensitivity study for the mixing layer height (MLH) is included and good agreement with ground based data in Bologna was observed for MLH between 200 m and 300 m above ground (AGL). This is in agreement

ACPD

4, S3791–S3804, 2004

to situations in February 2000 and 2001 when the MLH was between 300 m and 600 m ASL (Petritoli et al. 2004). On the specific day 19/02/2003 the soundings from Linate and San Pietro (http://weather.uwyo.edu/upperair/sounding.html), showed no clear inversion at 12:00 UT and about 200 m (ASL) in the night (0:00 UT) and morning hours (6:00 UT Linate only). So our result of 200 m to 300 m (AGL) seems to be realistic. More details about this comparison to the grond based data are encluded in the final version

3a) This study performs a comparison for a specific region, on a specific day. A validation study should also be concerned with the behaviour of the error of the product with respect to several relevant geophysical parameters (e.g. season, clouds, latitude, aerosol). It is not necessary that all relevant parameters are covered, but a validation study should specifically describe the range of validity for some of these parameters. Objective of the paper: This paper is not a "validation" paper. However, if the specific comments below are taken into account, it can be
1] a valuable demonstration of the potential of SCIAMACHY to properly quantify tropospheric NO_2 columns with the method developed by Richter et al (2004), or
2] an independent quantitative verification that the two instruments can measure (approximately) the same amounts of tropospheric NO_2, although they are measuring at very different heights. The first can only be achieved when independent ground-based measurements are added in the analysis (see specific comment 27 below).

We keep to using the term "validation", see general comments 1). A comparison to local ground based in situ measurements is added, as mentioned above and therefore we demonstrated the potential of SCIAMACHY to properly quantify the tropospheric NO_2 columns.

3b) There are two major sources of uncertainty in this paper: the determination of stratospheric slant columns, and the calculation of AMFs. These two sources are not fully discussed and/or appropriately referenced (see specific comments, below). The authors should discuss the validaty and the limitations of the methods used in this

ACPD
4, S3791–S3804, 2004

paper, and estimate the error budgets.

We extended our discussion on both potential error sources. These two major sources of uncertainties are now objects of two sensitivity studies included in the final version:
1. Different ways to separate tropospheric from stratospheric are discussed.
2. Different AMFs have been calculated based on independent observations.

SPECIFIC COMMENTS 1) Title and Abstract: The word "validation" and "validated" should be replaced by "comparison" and "compared". The complete title may be changed to cover the (new) scientific objective of the paper, see general comments. The abstract of course needs to be rewritten after all comments have been taken into account.

The demonstrated work is our contribution to the worldwide efforts taken for the validation of SCIAMACHY. See also general comments.

2) References:
a) As far as I know, there are currently at least four groups, retrieving tropospheric NO_2 from SCIAMACHY (IUP Bremen, IUP Heidelberg, KNMI/BIRA, and SAO). The authors should refer to the relevant publications, websites or other of these groups in the introduction.
b) Besides the authors' institutes, several other institutes are currently developing SCIAMACHY products, publishing about the retrieval techniques and making the products available. Therefore the reference to Frankenberg et al (2004) on 7514-19 should be accompanied by a few more references, reflecting this properly.
c) references are missing in 7515-16, and 7518-20/21, and 7519-21, 7519-23.

References to the following institutes are added:
* www.temnis.nl (KNMI and BIRA),
* wdc.dlr.de (DLR),
* cfa-www.Harvard.edu/saohome.html (SAO),
The reference to Frankenberg was removed and replaced by a more

ACPD

4, S3791–S3804, 2004

general reference to the Proceedings of the ACVE 2 in Frascati 2004 http://envisat.esa.int/workshops/acve2/contents.html

3) 7515-4/6: "In Europe the ... et al., 2004).": The validation of a satellite product requires not only a large range of values, but also a large range of several other geophysical paramaters, and several independent measurement techniques. The fact that the Po-Valley/high Alps combination provides a large range of tropospheric NO_2 values, makes it a good region for studying the behaviour of the error in the tropospheric NO_2 product with respect to the amount of tropopsheric NO_2, not for validation in general.

It is not clear to us what the reviewer means with "validation in general". In most locations on earth the tropospheric NO_2 column is close to 0. Thus a meaningful validation must focus on polluted areas. The high concentrations and the strong gradients make the Po basin a region ideal for a relation between AMAXDOAS and SCIAMACHY columns.
See also general comments of reviewer 1.

4) 7515-10/11: "As the conversion ... a major uncertainty": The conversion itself is not a major uncertainty, but the conversion introduces or adds uncertainties, because of the assumptions that go into the air-mass factor calculations.

Corrected.

5) 7515-7/16: It is not clear what the authors mean here: a) one should compare slant columns because AMFs introduce uncertainties or b) one should compare vertical columns, because differences in solar zenith angle introduce differences in slant columns. It would be much clearer if the line of logic would be: We compare both slant columns and vertical columns and we list for both methods the main sources of errors expected, and quantify them if possible.

We reorganised our final version following the suggested line of logic.

ACPD

4, S3791–S3804, 2004

6) 7515-19: "In contrast to these observations...": What is the contrast: they didn?t measure NO_2, or it was not tropospheric NO_2, or it was not for satellite validation?

All these Airborne DOAS measurements have been made for different aims: Some for tropospheric trace gases, some for stratospheric trace gases, but none of these measurements was used for a validation of any satellite for tropospheric species. We concentrate on tropospheric NO_2 for the validation of SCIAMACHY. We clarified the text.

7) 7516-3/6 "But only light ... the tropospheric gases" contradicts with 7524-3/10 "As expected, a ... by tropospheric absorbers."

This sentence clearly simplifies the real situation. We clarified the text.

8) 7517-4: The horizontal resolution of the AMAXDOAS measurements should be 6.3km=0.057°! Later, in 7525-18, a grid of 0.075° x 0.075° is used to find colocations. If this is a (long,lat)-grid, then this should be 0.057/cos(lat) x 0.057. Apart from this, making such a grid is not the way to find the proper colocations. Each AMAXDOAS measurement has a SCIAMACHY ground pixel which is closest to it. Just look at the coordinates of both observations and calculate the distance.

Compared to the pixel-size of SCIAMACHY (30 km x 60 km) an AMAXDOAS measurement (6.6 km x 0.08 km) is very small. The most meaningful criterion is whether or not most of the measurement is inside a SCIAMACHY ground pixel or not. This paragraph, the complete text and all the figures are changed accordingly.

9) In section 2.1.2 the selection criteria for the solar reference spectrum are listed. The first criterium is "use of the same telescope". It should be explained how this is practically implemented. From section 2.1.1 I understand that the telescopes are mounted inside housings outside the aeroplane. I don't understand how the same telescope for nadir and zenith measurements can be used on the same flight. 11) 7518-15 and figure 3: It appears to me that the reference spectrum here is a nadir spectrum,

ACPD

4, S3791–S3804, 2004

not a solar spectrum. It is not clear to me whether the AMAXDOAS data shown in this paper is analysed using nadir measurements or direct solar measurements. When the Alps spectrum would be used as the reference, there would not be a stratospheric contribution in the Alps, since $I = I_0$ there. A clarification would be in place.

A spectrum from the same telescope and therefore the same line of sight is used as reference. The reference spectrum chosen here was observed over the Alps. Of course this reference spectrum contains both the stratospheric and the tropospheric signal which is very small over the mountains. The reference contains only the stratospheric absorption. For longer periods and distances it has to be considered that, the stratospheric signal changes, due to the change in the vertical column along the flight track and due to the change of the solar zenith angle. The information was added to the text.

10) The criteria listed from 7517-24 to 7518-8 should be quantified. What are the quantitative selection criteria used for this study?

The used reference spectrum is described in detail.

12) In both cases, solar spectrum as reference or nadir spectrum as reference, the stratopsheric contribution will still be in the retrieved slant column:
SCD$_m = SCD_t + SCD_s - SCD_{s,0}$,
where SCD$_m$ is the calculated slant column, SCD$_t$ is the tropospherics lant column, SCD$_s$ and SCD$_{s,0}$ are the stratospheric slant columns for the measurement and the reference measurement respectively.
SCD$_s - SCD_{s,0} = VCD_s * AMF_s - VCD_{s,0} * AMF_{s,0}$,
where VCD$_s$ and VCD$_{s,0}$ are stratospheric columns, and AMF$_s$ and AMF$_{s,0}$ are stratospheric air mass factors for the measurement and the reference measurement respectively. The authors assume that the vertical stratospheric columns are varying only very slowly, so I suspect that the main source of variation of the SLANT stratospheric columns will be the solar zenith angle. It should be argued why a "linear function" in 7518-25 would be a good choice. A more obvious choice in my opinion would be, e.g.,

ACPD

4, S3791–S3804, 2004

(a+b*phi)/cos(SZA), phi is latitude, SZA is solar zenith angle. Clarify the subtraction of the stratosphere with a figure, e.g. such as figure 8, with AMAXDOAS total slant column versus latitude, for all AMAXDOAS measurements, and the assumed stratospheric contribution as a line. Discuss the limitations of this method.

We agree that the variations of the stratospheric slant column have to be considered. However from sensitivity studies we found that for our observations at moderate SZA a linear interpolation is sufficient. The information was added to the text.

13) 7519-1: I assume that "tropospheric" AMF is meant here?

Corrected.

14) 7519-12: Although the SCIAMACHY nadir and limb viewing mode is alternated every other minute, the limb measurement over a specific area is about 7 minutes prior to the nadir measurement over the same area.

Thank you for this hint. We added this information here.

15) 7519-25: The validity of this method, and its limitations should be discussed here (see Boersma et al, 2004).

From our point of view it is not necessary to discuss the limitation in detail here. This was already done in the mentioned reference in an excellent way, so we refer to this work. However we added a sensitivity study on the stratospheric tropospheric separation as we did for AMAXDOAS. For our purpose we found hardly any difference between the three corrections we used: 1. the reference sector method, 2. the linear interpolation, 3. the method suggested for AMAXDOAS in comment12). The information was added to the text.

16) 7520-6/8: Explain this (see also my comments 12 and 14) and quantify.

An estimate of the influence of the reference method is given.

17) 7520-11/12: Operational SCIAMACHY NO_2 columns are available and they are

ACPD

4, S3791–S3804, 2004

being validated by other groups. The authors might want to refer to the ACVE-2 proceedings (Lambert et al, 2004) for the latest status. However, they can also leave the sentence out. There is no tropospheric NO_2 column in the operational product.

We point out that an official tropospheric product is not planned.

18) Section 2.3 "Slant and vertical columns" (pages 7520/7522) is confusing. The tropospheric AMF calculation is introduced here. However, the definition given for the AMF includes the total vertical column. Is the calculation done by Friedeburg (2003) and Honninger et al (2004) for total atmosphere AMFs or for tropospheric AMFs? Is the "comparison between both programs" (7521-7) used for calculating total atmosphere AMFs for SCIAMACHY? In that case 8should be given here, or the comparison should be explained in the paper, illustrated, and discussed.

For the conversion of tropospheric slant column densities to tropospheric vertical column a tropospheric air mass factor is applied. It is calculated for a purely tropospheric NO_2 profile. We clarified the paragraph. In the revised version we calculated all AMF with the same RTM (Tracy). This leads to a better consistency of the comparison.

19) 7520-17/18: "For a better ... calculated:" change in: "The vertical column density(VCD) is defined as:"

Changed.

20) 7521-9: "even better": quantify

This is no longer necessary, as the AMF are now calculated using only one program. The comparison between different ray tracing programs is definitely beyond the aims of our work.

21a) 7521-10/18: Here it is argued that the AMFs used for SCIAMACHY and AMAX-DOAS should be calculated with the same settings to make the vertical columns comparable. (See also remark 5.) If tropospheric slant columns would be compared you would indeed expect that the differences would be caused mainly by the different light

ACPD

4, S3791–S3804, 2004

paths of the two measurement methods. Therefore one should compare slant columns divided by an AMF, calculated for one particular atmosphere (the particular settings are not so important here), but for correct viewing geometries (as is done in the paper). If a difference is found now the main suspects should be: difference in slant column retrieval, differences in the RTMs, the different method of stratospheric correction. For all three suspects one should be able to study, quantify or exclude the effect, giving either proper references or discuss it satisfactory.

We adapted our text accordingly.

21b) Apart from these three, another important cause of differences can be the variability of the atmosphere at spatial scales which are resolved by AMAXDOAS, but not by SCIAMACHY. The albedo differences in the Alps are an example of this. Tropospheric AMFs are known to be very sensitive to albedo, 10s of percents for 0.2 albedo difference (Boersma et al, 2004). The albedo variability could also cause the high variability in figure 8 ("Adige" and "Apennine"), instead of a possible higher NO_2 density in the valleys in between the snow-capped mountains(7525-26/29).

If the change in the albedo caused the observed high SCDs, it would require that there is NO_2 above the snow. While we cannot exclude this possibility we think it is more probable, that the NO_2 is in the valley. The different spatial resolution is of course a significant difference between AMAXDOAS and SCIAMACHY, and even averaging all data points covered by one SCIAMACHY pixel does not completely solve this problem. See also comment 2) of reviewer 1.

22) 7523-23: Beirle et al (2004) is not the proper reference for this statement. The authors might want to rewrite this sentence and the next and only talk about tropospheric NO_2 concentrations. If not, the authors should find the proper references.

We pointed out, that only tropospheric pollution is meant, therefore the reference to Beirle et al. is well suited here.

ACPD

4, S3791–S3804, 2004

23) 7524-26/7525-5: This paragraph should be rewritten in view of my remarks 5 and 21. The sentence 7525-2/4, "Compared to validation ... the AMAXDOAS instrument." ,is wrong, see my general comments. Ground-based measurements are absolutely essential for the validation of both AMAXDOAS and SCIAMACHY.

In situ measurements are indeed a very good way to substantiate our results. In our final version we added a comparison. However we still believe that the comparison of the slant columns between AMAXDOAS and SCIAMACHY is very important in itself.

24) 7525-15: 5% is not correct from the figure, it looks more like 7%, but the authors should have the correct numbers.

Changed according to the new AMFs.

25) 7525-17: what is meant by "The zigzag ... was taken into account"?

It seems that the German word "ZickZack" English "zigzag" is no longer generally understood in English, so we removed the complete sentence.

26) 7526-5: There is no overestimation of the SCIAMACHY data. Using the fitted line formula for the slant columns (A = 0.95*S+1.1) it follows that for (in 1E15 molec/cm2)
S = 0 => A = 1.1; S = 10 => A = 10.6; S = 20 => A = 20.1.
And for the vertical columns (A = 0.89*S+1.0):
S = 0 => A = 1.0; S = 10 => A = 9.9; S = 20 => A = 18.8.
So in the range where most measurements are (below 10E15 molec/cm2) SCIAMACHY values seem to be on average less than AMAXDOAS values, both for slant columns and for vertical columns! The paragraphs 7526-9/23 should be removed or rewritten. The average difference <A-S> should be calculated separately, not as one of the parameters of the fit. In order to determine whether the difference in the slant column is a significant function of the slant column itself (that is what you do when you fit a straight line in such a plot), the authors should calculate the significance of the fit or the probability that one finds a slope differing from 1 by 0.05 or more for slant columns

ACPD
4, S3791–S3804, 2004

and by 0.1 or more for vertical columns, assuming the parent population has a slope of 1. I suspect that this is not very significant. Possible causes for the differences that are found are listed in 21.

The offset in the linear fit is mainly given by the different correction of stratospheric influence. The overestimation is mainly given by the slope of the linear fit. As the slope is less than one SCIAMACHY overestimates the tropospheric NO_2 columns.

27) 7526-24: Indeed it is very unfortunate that the authors didn't find any ground-based measurements. Ground-based measurements would demonstrate the potential of SCIAMACHY to properly quantify tropospheric NO_2 columns, which mainly depends on a proper knowledge of the atmosphere, reflected in the AMFs. What is really needed is some case studies for a well-measured atmosphere colocated with a SCIAMACHY pixel (use e.g. one of the balloon campaigns for SCIAMACHY validation), or with an AMAXDOAS measurement to show that both the stratospheric correction and the AMF calculation can be properly performed, so that the tropospheric NO_2 is correctly calculated. The case studies should be done both in clean and in polluted areas. This would strengthen the current paper considerably. See also my remarks on the objective of the paper in the general comments.

As already mentioned we added a comparison to in situ data. Nevertheless we are a cautious since the comparison of in situ and remote sensing is not straight forward. Especially close to sources strong gradients may influence the in situ measurements but are neither resolved by AMAXDOAS nor SCIAMACHY.

28) The conclusion should of course be rewritten after the changes have been made. In 7527-6/7 is stated that the correlation gets worse for vertical columns: this is not shown. The fitted slope is smaller, but this has nothing to do with the correlation.

Done.

TECHNICAL CORRECTIONS 7515-18: Pertitoli -> Petritoli

ACPD

4, S3791–S3804, 2004

7515-26: its mission are descibed -> its mission objectives are described
7517-14/16: use the subscript i with the cross section (sigma) and concentration (c)
7517-20: absolute atmospheric column -> total atmopheric slant column
7517-20: difference in the column -> difference in the slant column
7517-21: no comma
7520-14: analyses -> analysis
7519-4: 2.3 -> 2.2.1
7519-4: change title, e.g.: Description of the instrument and its measurement characteristics
7519-18: 2.3.1 -> 2.2.2
7520-13: 2.4 -> 2.3
7520-23: parameter -> parameters
7524-7: increasing -> increased
7526-4: derivation -> slope
7527-16: 19702/2003 -> 19/02/2003
Fig.5: "For comparison the flight ... is shown as well." -> "The flight ... is shown in red."

Thank you very much for these helpful technical comments, we considered them all.

*Wagner, T, Dix, B., Friedeburg, C. v. , Sanghavi, S., Sinreich, R. and Platt, U.: MAX-DOAS O4 measurements - a new technique to derive information on atmospheric aerosol. - Principles and information content, JGR 109, D22205, doi 1029/2004JD004904, 2004

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