Interactive comment on “Validation of SCIAMACHY tropospheric NO$_2$-columns with AMAXDOAS measurements” by K.-P. Heue et al.

Anonymous Referee #2

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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:
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.

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 validity of the validation measurements is.

3) 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).

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 validity and the limitations of the methods used in this paper, and estimate the error budgets.
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.

2) References:

a) As far as I know, there are currently at least four groups, retrieving tropospheric NO\textsubscript{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.

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 geo-physical parameters, and several independent measurement techniques. The fact that the Po-Valley/high Alps combination provides a large range of tropospheric NO\textsubscript{2} values, makes it a good region for studying the behaviour of the error in the tropospheric NO\textsubscript{2} product with respect to the amount of tropospheric NO\textsubscript{2}, not for validation in general.

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.

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 ver-
tical 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.

6) 7515-19: "In contrast to these observations...": What is the contrast: they didn’t measure NO₂, or it was not tropospheric NO₂, or it was not for satellite validation?

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

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.

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.

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

11) 7518-15 and figure 3: It appears to me that the reference spectrum here is a nadir spectrum, 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=I₀ there. A clarification would be in place.
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 tropospheric slant column, \( SCD_s \) and \( SCD_{s,0} \) are the stratospheric slant columns for the measurement and the reference measurement respectively. \( SCD_s - SCD_{s,0} = VCD_s \cdot AMF_s - VCD_{s,0} \cdot AMF_{s,0} \), 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., \( (a + b\phi)/\cos \theta_0 \), \( \phi \) is latitude, \( \theta_0 \) 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.

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

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.

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

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

17) 7520-11/12: Operational SCIAMACHY NO\(_2\) columns are available and they are 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.
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 8% difference is very much! In any case a reference should be given here, or the comparison should be explained in the paper, illustrated, and discussed.

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

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

21) 7521-10/18: Here it is argued that the AMFs used for SCIAMACHY and AMAXDOAS 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 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. 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 NO₂ density in the valleys inbetween the snow-capped mountains.
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.

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.

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

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

26) 7526-5: There is no overestimation of the SCIAMACHY data. Using the fitted line formula for the slant columns ($A = 0.95 \cdot S + 1.1$) it follows that for (in $10^{15}$ molec/cm$^2$)

$S = 0 \Rightarrow A = 1.1; \ S = 10 \Rightarrow A = 10.6; \ S = 20 \Rightarrow A = 20.1.$

And for the vertical columns ($A = 0.89 \cdot S + 1.0$):

$S = 0 \Rightarrow A = 1.0; \ S = 10 \Rightarrow A = 9.9; \ S = 20 \Rightarrow A = 18.8.$

So in the range where most measurements are (below $10^{16}$ molec/cm$^2$) 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 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.

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 NO₂ 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 NO₂ 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.

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.

TECHNICAL CORRECTIONS
7515-18: Pertitoli -> Petritoli
7515-26: its mission are described -> 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 atmospheric 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.'