

***Interactive comment on “Stratospheric and tropospheric NO<sub>2</sub> variability on the diurnal and annual scale: a combined retrieval from ENVISAT/SCIAMACHY and solar FTIR at the Permanent Ground-Truthing Facility Zugspitze/Garmisch” by R. Sussmann et al.***

R. Sussmann et al.

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Final Response to Anonymous Referee #2

We like to thank the anonymous referee very much for his effort to carefully go through the manuscript and make valuable suggestions for improvements to this paper, and expressed this in the acknowledgment.

In final response, we thereafter provide positive point-to-point replies to all specific

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referee comments, and a list of all related manuscript changes we have performed.

"-page 2385, row 8: such conclusion can not be pointed out by eye on the plot considering also that actually it is recognizable from the same plot some differences in the slopes. The authors spent a lot of words in the previous paragraphs on the error analysis and I think that here a detailed calculation of the increasing rate of the stratospheric NO<sub>2</sub> considering also error bars is mandatory also because of its importance from a scientific point of view. As stated by the same title of the paper this is one of the main point of the work and I think it deserves more accurate calculations."

We agree, and added a new Figure (3b) showing the increasing rates retrieved from FTIR plotted for the different months of the year together with their error bars (retrieved from Fig. 3a), and supplemented the text accordingly, p2385, l5-9 (new):

"Figure 3a shows all individual FTIR columns of the data set, but now plotted as function of the hour of the day, and separated for the 12 different months by colors. For each month a linear fit is performed to all the individual columns of this month.

Fig. 3b gives evidence, that there is no significant seasonal change of the daytime increasing rate of stratospheric NO<sub>2</sub> within the FTIR error bars."

The caption for the new Fig. 3b added is (p2416): "(b) The 12 different diurnal increasing rates obtained from the fits performed to the monthly FTIR data sets in (a). The plotted slope error bars (2 sigma) are obtained from the linear fits in (a), and the red line gives the average increasing rate, i.e.,  $1.02(6)E+14 \text{ cm}^{-2}/\text{h}$ ."

We also added the statistical error to the retrieved average daytime increasing rate at page 2385, line 17: "The resulting daytime NO<sub>2</sub> increasing rate is  $1.02(6)E+14 \text{ cm}^{-2}/\text{h}$ ."

"-Figure 4 is not necessary."

Figure 4 describes the concept of "virtual coincidences". It presents - without many words - a simple but nevertheless crucial new concept for "validation and synergistic use". Therefore, and due to didactic reasons, we like to keep this figure within the

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paper.

"-page 2390: the procedure explained in the Step 2 has not be discussed from a scientific point of view. Where does the factor 2 come from?"

This is an ad hoc approach. We added the following explanantion to the manuscript (p2390, l25): "Using 2 times the average as a cutoff criterium is an ad hoc approach to achieve our goal of eliminating few but extremely spiking maximum values from an ensemble with rather uniform minimum values."

"Besides at page 2402 row 21 it is stated that this pollution-clearing scheme does not work well (because it keeps info on the PBL NO<sub>2</sub>) and few rows below authors come to a conclusion that is exactly the opposite! A clarification is required."

We understand this had not been explained in a clear way. However, after we have introduced a cloud clearing now within the revised version of the manuscript, the problem of a bypassing of our pollution clearing scheme by extraordinarily high pollution enhancements has disappeared. This is because the highest pollution enhancements (due to pollution above snow or clouds) are already filtered out by the cloud clearing. As a consequence for the revised manuscript, we removed Fig. 12b, as well as the second and third paragraph of Section 5.6.

"-For the reasons explained above, paragraphs 4 and 5.5 have no-sense in their present form."

In fact we present two completely new concepts to satellite validation of NO<sub>2</sub> by ground FTIR, i.e., i) the concept of "virtual coincidence" and ii) a "pollution clearing scheme", furthermore we have presented a quantitative intercomparison of day-to-day scatter. So clearly, the paper starts with developing concepts for satellite validation. These are then applied to enable the synergistic use, but provide at the same time the basis for more detailed upcoming validation studies. So, although this paper does not provide final conclusions on "validation" we think that it provides significant contributions to

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both "validation" and "synergistic use".

To make this clear we agree to reword our sections 4 and 5.5, and also performed several further related changes throughout the manuscript as follows:

p2392, l14 (new): "4 Intercomparison of SCIAMACHY versus FTIR column retrievals"  
paragraph 4.2. (new): "4.2 Intercomparison of absolute column levels

The difference of the time series of SCIAMACHY and FTIR is displayed in Fig. 5 (red curve). Clearly, the SCIAMACHY columns show significantly higher values throughout the full validation period. The difference ( $col_{SCIA} - col_{FTIR}$ ) is  $0.83E+15 \text{ cm}^{-2}$  on average, with a minimum of  $0.60E+15 \text{ cm}^{-2}$  and a maximum of  $1.24E+15 \text{ cm}^{-2}$ .

This kind of intercomparison of the direct-output  $\text{NO}_2$  column levels from two different remote sounding systems (satellite versus ground) has been performed in many previous papers, and the differences been interpreted in terms of errors of the satellite instrument. However, we would like to point out that this approach is only the first possibility out of two limiting (theoretical) cases: i) The observed differences are due to intrinsic errors in either of the two remote sounding data sets under the simplifying assumption that the two measurement systems have identical sampling characteristics or, ii) the observed difference can be attributed to the different sampling characteristics of the two instruments (differing averaging kernels) under the assumption that they are both working in principle without intrinsic errors. Reality will be in between these limiting cases. A theoretical framework to deal with this problem for the purpose of satellite validation has been given by Rodgers and Connor (2003).

We decided to thereafter follow the assumption ii) as a basis for our subsequent synergistic use of SCIAMACHY and FTIR data aiming at the retrieval of tropospheric  $\text{NO}_2$ . I.e., we assume the difference shown in Fig. 5 is dominated by the differing sensitivities of SCIAMACHY versus FTIR and not by intrinsic errors of SCIAMACHY (or FTIR) measurements. Clearly, this is a simplifying assumption for the purpose of this study,

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and future validation studies have to be performed in order to explore to which degree this assumption holds. Our concepts of "virtual coincidence" and "pollution clearing" can contribute to the required refined validation studies."

p2401, l1 (new): "5.5 Discussion of the retrieved tropospheric columns series in terms of validity"

p2401, l12-14 (new): "This is only a qualitative statement and it means that from our findings we can neither exclude nor find any evidence for an intrinsic principle error in the SCIAMACHY data set."

p2401, l19, we added: "A more quantitative validation of our new method could be performed in an upcoming study using collocated SCIAMACHY tropospheric columns retrieved with the Richter and Burrows (2002) method. In fact the two methods are independent (because now the stratospheric background to be removed is that of FTIR and not that retrieved by SCIAMACHY over the Pacific Ocean) so that useful indications on selfconsistency and/or validation of the new approach (or limitations of the old one) can be pointed out." (We also added this idea to the conclusions in a shortened version).

"-In the text figure 12 is mentioned before figure 11. So please, invert the figure numbers."

We changed the numbers accordingly.

"In fact the used parameterization has not been justify (or at least is not clear from the text) neither from a physical nor from a mathematical point of view. A better explanation would improve the comprehension."

We agree to motivate our retrieval constraint in some more detail and added the following text at p2399, l6: "Our retrieval constraint is set up from two parameters, i.e., it allows for retrieval of two independent degrees of freedom, because this is what can be expected from using two complementary input parameters, namely  $col_{FTIR}$  and

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*col<sub>SCIA</sub>*. The constraint for the stratospheric part is a simple scaling of the US standard profile as used also for the FTIR retrieval. For the lower part we use a scaling of a VMR profile that is constant with altitude. This is because only one degree of freedom is left for this lower part, and it is the zero-order approach in our case where no better a priori information on the vertical distribution of free tropospheric background NO<sub>2</sub> is available. Our approach of linking the two parts of the profile together just at the point where the tropospheric VMR matches the US standard profile is the simplest solution that avoids (unphysical) negative VMR gradients at the transition between the lower and upper part of the profile."

"-page 2404, row 17-19: it does not make sense. If authors think that an "horizontal map of tropospheric NO<sub>2</sub>" is required to explain better some concepts/results they are strongly encouraged to add it."

We explain why a "horizontal map" was not added to this paper, i.e., we explain the steps that will have to be performed before. We added the following sentence to p2404, l19: "For this, the averaging kernels have to be determined for each measurement independently using the best estimates for albedo, aerosols, clouds and NO<sub>2</sub> profile available (Eskes and Boersma, 2003)."

End of Response.

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