

## **Response to anonymous reviewer #2 (C2263)**

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We thank the anonymous referee #2 for his/her valuable comments, most of which we agree with. They have helped us to improve the revised manuscripts in many places, we believe.

1. The paper is quite lengthy and sometimes lacks of a clear structure. The authors should try to shorten the manuscript, in particular for the parts which are not really new (e.g. that the reference sector method has problems at high latitudes in winter) and focus on the new results.

We agree that the manuscript lacked a clear structure in some places. We have streamlined the manuscript, trying to give more structure, especially in the Methods section. To these means, we have split Sec. 2 into two sections, namely “Datasets used in this study” and “Stratospheric correction algorithm”. In several places, we deleted references to the reference sector method.

2. The authors point out that they use limb measurements corresponding to the respective nadir state for their stratospheric correction, and claim that this (in contrast to Beirle et al. (2010)) results in a more accurate tropospheric product. This argument has to be strengthened by providing single-day results - preferably for a day shown in Beirle et al., 2010.

In the revised manuscript, we included daily plots of the four days shown in Beirle et al. (2010).

3. Both limb and CTM are applied for stratospheric correction, but have to be corrected for the reference sector first. Thus, there are different stratospheric datasets (with and w/o reference sector removal), and it was not always clear to me, which dataset is discussed or shown in the figures. So please make this clear, e.g. by defining abbreviations, or stating clearly that only the corrected datasets are shown.

In the revised manuscript, we have added formulas to the Methods section, to give a better explanation of our algorithm. In the following parts, we have reused the nomenclature which we introduced in the formulas. We believe that this makes it now clearer throughout the manuscript, which dataset is referred to at which point.

Title: Two stratospheric corrections are discussed in this study, without a clear conclusion which should be preferred. This equality should be also reflected in the title.

We agree that the title should reflect the focus on stratospheric NO<sub>2</sub> and therefore renamed the manuscript to “Improvements to the retrieval of tropospheric NO<sub>2</sub> from satellite—stratospheric correction using SCIAMACHY limb/nadir matching and comparison to Oslo CTM2 simulations”.

5044, 13: see 2.

see our answer to 2.

5044, 20-24: Given the high overall offset, I recommend to avoid to state “remarkably well agreement” before the offset is removed.

In the revised manuscript, we have rephrased these passages accordingly.

5046, 15: What does “originally” mean? “A widely used method was the relatively simple “reference sector method” “...”

This has been corrected in the revised manuscript.

5048, 7: See 2.

see our answer to 2.

5051, 24: add “e.g.” in reference list

This has been corrected in the revised manuscript.

5053: Try to clarify this section. I am confused by lines 19-20. What do you mean by limb state? In my understanding, all four limb line-of-sight directions belong to one limb state. What does “four distinct values for every nadir state” mean?

This confusion was based on a misconception on our part. We have rephrased the section accordingly.

Is the stratospheric correction for top left and bottom left pixels of the nadir state the same, or is there also a latitudinal interpolation applied within one nadir state? If not, systematic errors are introduced for states with strong gradients.

The stratospheric correction for top left and bottom left pixels of one nadir state are not the same.

As stated in Sec. 2.3.1 (Sec. 3.2 in the revised manuscript), we consider the limb columns for all four viewing azimuth directions as a function of latitude, and then for each nadir pixel, we interpolate one limb value for each of the four viewing azimuth directions, by latitude (step a). From these, the ‘final’ limb column for the nadir pixel’s location is interpolated in line-of-sight (step b). As the top left and bottom left pixels of the nadir state have different latitudes, they will be assigned different values in step a).

Why is the across-track interpolation done linearly? What would happen if e.g. spline interpolation would be applied?

We have seen that the choice of interpolation in across-track direction does not significantly impact the results. Therefore, we chose linear interpolation because this is the most simple interpolation.

5054, 2/3: Interpolation is cubic in location and linear in time - why? What would be the impact of a different interpolation scheme?

We assume that the temporal variability can be accurately approximated using linear interpolation. On the other hand, our experience has shown that cubic interpolation in geolocation usually yields more plausible results than linear interpolation.

5055, 6-8: please reword.

This has been reworded in the revised manuscript.

5056, 7: "originally"?

This has been corrected in the revised manuscript.

5056 13 "... by averaging all ..."

This has been corrected in the revised manuscript.

5057 23 "... observed for all ..."

This has been corrected in the revised manuscript.

Section 3.2.2:  
- Refer to previous studies, which have reported the same phenomenon.

We have added a reference to Beirle et al. (2010) to the revised manuscript. As both Sioris et al. (2004) and Sierk et al. (2006) do not mention this effect, we are not aware of any further studies exploring the offset between limb and nadir measurements.

- Try to avoid repetitions on the reference sector method.

We have deleted some repetitions about the RSM in the revised manuscript.

5060, 21: "in many regions": please specify. I don't see a striking global land-sea contrast. There are dominating "clouds" of positive as well as negative DVCD extending over land and sea. Thus, I cannot comprehend the following discussion.

Along the S-American West coast, a strong land-sea contrast is visible in Fig. 8 (Fig. 9 in the revised manuscript). E.g. in the annual mean and in October, negative values are visible West of the N-Chilean coast. We have specified this region in the revised manuscript, replacing the phrasing "in many regions".

5061, 15: How far does a cloud study improves our knowledge on surface spectral reflectance? This has to be discussed more in detail, and should also be listed in the error evaluation of the nadir column.

Not the cloud study would improve our knowledge on surface spectral reflectance, but rather the more detailed "understanding of the systematic differences between limb retrievals and model simulations". Knowledge about the influence of clouds on the limb retrievals and model simulations could improve our understanding of these systematic differences. And from these differences, it might be possible to draw conclusions about the influence of, among others, surface spectral reflectance.

Section 3.2.4: I found this section hard to read. Fig. 9a shows uncorrected VCDs, 9b corrected VCDs, and 9c corrected VCDs for a different latitude - this is quite confusing.

I suggest to

- move 9a into a separate figure, which could be shown in the method section as illustration for the need of offset correction.
- remove the unimportant parts of the caption (like "strat. AMF applied")
- label each subplot with the respective latitude range.

These points have been accounted for / added in the revised manuscript.

I would appreciate if the authors could focus the study of zonal variations to one or two months exemplarily. For each month, all latitudinal bands could be shown as subplots (top: northernmost) of a single figure. Other months could be added in the supplement.

There are 36 individual 5-degree latitude bands per month. Choosing a larger latitudinal binning would reduce the level of detail visible in the plots. Even when considerably shrinking the size of the individual plots, it would be hard to fit them on one page.

We want to convey the multitude of different situations to the reader and believe that this is best accomplished by cherry-picking the most interesting months and latitude bands. Therefore, we chose to keep the exemplary zonal variation plots shown in the revised manuscript. However, we have added additional plots to the supplement, namely all zonal bands for January and August 2006.

5062, 5-10: A terrain effect or 3d effects would not only show up in October.

Note that the figure captions in the original showed a wrong month; in reality, Fig. 9 (Figs. 4 and 10 in the revised manuscript) shows data for August 2006. The effect of very low VCD<sub>strat</sub> over Greenland is visible throughout Northern Hemisphere summer; we have chosen to show August 2006 as an example.

The area of Southern Greenland is known to be special for multiple reasons. There exists strong tropopause folding activity (Elbern et al., 1998) and thus troposphere-stratosphere exchange (Sprenger and Wernli, 2003). Furthermore, a local maximum in the density of polar low pressure systems exists to its East (see Zahn and von Storch, 2008), and Greenland's high surface altitude and high surface reflectance (due to ice cover) stand in clear contrast to the surrounding Atlantic Ocean. While all these factors might contribute to the observed summer lows in VCD<sub>strat</sub> NO<sub>2</sub>, the underlying mechanism remains unclear at the moment, and it is hard to clearly attribute this phenomenon to one of them.

We have amended the revised manuscript accordingly.

5062, 24: This should not be a problem any more after applying the offset correction?

We agree with the reviewer that the problem should generally be solved by applying the offset correction. Nonetheless, in some instances it seems that the Oslo CTM2 overestimates stratospheric NO<sub>2</sub> over the Pacific Ocean, which would then lead to problems in the retrieved tropospheric NO<sub>2</sub> elsewhere in the same zonal band.

In the revised manuscript, we have rephrased this paragraph accordingly.

5063, 9-20: Shorten.

In the revised manuscript, we have shortened this passage.

5064, 1-12: This potential uncertainty has to be added to section 3.5.1.

In the revised manuscript, we have added this to Sec. 3.5.1 (4.4.1).

Section 3.2.5:

The 31-day running mean introduces artefacts after gaps. If the running mean would be calculated in opposite direction, i.e. starting on 31st December, running back in time, Fig. 15 would look significantly different.

We are using a centered 31-day moving average, so the direction of the averaging does not change the outcome. This is clarified by introducing the word “centered” in the revised manuscript.

The calculation of  $c_v$  for limb is problematic, as the interpolated limb VCDs for each nadir ground pixel are not independent! The precision of the limb measurements should be derived based on limb data per state.

We have repeated the calculation of  $c_v$  with the ‘raw’ model and limb data, i.e. before the spatial interpolation. The areas had to be extended to approx.  $5 \times 5$  degree to yield a sufficient number of limb measurements / model values per box. The discussion has been adapted accordingly.

5066, 1-6: How does the stratospheric look like for the 60% deviation, and what is the SZA? Please add this profile to Fig. 5.

Due to a problem in our processing system, Figs. 16 and 17 in the original manuscript were affected by sampling issues leading to the comparison of ‘apples and oranges’.

We have repeated the sensitivity study about stratospheric air mass factors using all retrieved limb profiles between Jan 2003 and Dec 2010. In the revised manuscript, Figs. 16 and 17 are combined into one figure (17), showing that the influence of the limb profile on the stratospheric AMF is indeed negligible. At maximum, using the U.S. Standard Atmosphere’s  $\text{NO}_2$  profile instead of the actual vertical profile leads to an over-estimation of  $\text{AMF}_{\text{strat}}$  by 4%.

5068, 20: “delicate”?

This has been reworded in the revised manuscript.

5069, 10-18: I can see enhanced values in the marked area in the Indian Ocean for both products, so I can not comprehend this discussion.

This point, together with Fig. 19, has been removed from the revised manuscript.

5074, 1: The “very accurate representation” has not been demonstrated on daily basis.

The revised manuscript contains daily plots of trop.  $\text{NO}_2$  for those days which are also shown in Beirle et al. (2010).

5074, 7: "agree surprisingly well" - after offset correction!

This has been reworded in the revised manuscript.

5075, 14-16: I don't see this point supported by Fig. 19.

This sentence has been removed in the revised manuscript.

5076, 8-9: Repetition.

This has been reworded in the revised manuscript.

Fig. 3: The colorbar has smooth transitions, but in the map, only 7 distinct colors are shown.

This has been corrected in the revised manuscript.

Fig. 19: The inclined rectangle shape of the strange patterns (in both subplots) close to the South Pole indicates that they are caused by single nadir states.

Fig. 19 has been removed from the revised manuscript.

Supplement: It would make more sense to subtract the stratosphere from the total (=nadir) column; then the continents would look reddish, reflecting the tropospheric residue.

In the revised supplement, we have followed the reviewer's suggestion as to make the plots more intuitive.

## References

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- Sierk, B., Richter, A., Rozanov, A., von Savigny, C., Schmoltner, A. M., Buchwitz, M., Bovensmann, H., and Burrows, J. P.: Retrieval and monitoring of atmospheric trace gas concentrations in nadir and limb geometry using the space-borne Sciamachy instrument, *Environ. Monit. Assess.*, 120, 65–77, doi:10.1007/s10661-005-9049-9, 2006.

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- Zahn, M., and von Storch, H.: A long-term climatology of North Atlantic polar lows, *Geophys. Res. Lett.*, 35, 22, L22702, doi:10.1029/2008GL035769, 2008.