De Bilt, 18 February 2015

Improved spectral fitting of nitrogen dioxide from OMI in the 405 – 465 nm window — Van Geffen, et al.

Response to reviewer #1 of version 2 (publ. 21 Oct. 2014)

We would like to thank the reveiwer for carefully reading the and providing useful comments and suggestions that enable us to improve the paper.
Below we answer the reviewer's comments, quoting his/her remarks in a slanted font, which is followed by an overview of the main changes to the paper.

> *The paper "Improved spectral fitting of nitrogen dioxide from OMI in the 405-465 nm window" by Geffen et al. presents various improvements of the spectral analysis of NO2 compared to the "OMNO2A" retrieval, which is so far the basis for the standard NO2 product (NASA) as well as DOMINO (TEMIS). The paper is well written and matches the scope of AMT. I recommend publication after dealing with the following items:*

General remarks

1. *The authors present a lengthy comparison of OMI stratospheric column densities to other instruments. However, while the observed discrepancy between OMI and other measurements was the initiation for working on an improved spectral analysis, this part (section 2.3, section 3) is not directly related to the topic of the paper and should be shortened. I propose to move most parts of the comparisons to the supplement.*

We agree with the reviewer that the comparision with ground-based stratospheric NO_2 data is not the key point of this paper. The comparison has been moved to the Supplement, because (a) the comparison clearly shows that the bias between OMI and GOME-2/SCIAMACHY data is actually due to issues in the OMI NO_2 data, and (b) the comparison with OMI data has not been published elsewhere yet.

2. *The presented algorthm is going to be applied in a "forthcoming OMNO2A SC reprocessing". While the authors have clearly shown that the current OMNO2A has some shortcomings and present some clear improvements, I evaluate the presented data to be too sparse to be a sound basis for a reprocessing. The authors present the results of their new set-up for only one orbit, and test the other orbits of the same day for consistency. But no results are shown for other days (e.g., after the row anomaly). Effects like "striping", or the slit function, are very likely time-dependend. I thus urge the authors to include more days (different seasons, different years) in the analysis and check if the settings (optimized for 1 July 2005) work as well for other days, before starting a reprocessing of the complete OMI timeseries.*

We have reprocessed the OMI data for the complete year 2005 to evaluate the consistency of the reported changes. Results of this comparison have been added to the paper (Sect. 5.4) and the Supplement (Sect. S7).
To give an idea of the robustness of the results, the data of the four test days from 2005 and 2013 selected for the comparisons involving different fitting approaches and data of more years in the EU FP7 project QA4ECV (www.qa4ecv.eu), has been analysed; the results are presented in a paragraph of Sect. 5.1 and Fig. 5.

3. *The authors anticipate that the algorithm changes basically remove the stratospheric bias. However, some of the changes might effect the tropospheric columns as well (e.g. the update of the H2O cross section). Thus, a map of the NO2 SCD difference has to be shown, and possible spatial patterns should be discussed.*

Maps of the NO_2 SCD and other quantities have been added to the paper in a new Sect. 5.4.

4. *In the new algorithm, some cross-sections are updated or added, while other absorbers (e.g. Glyoxal) or Pseudo-absorbers (e.g. Vibrational Raman; spectral signatures of sand (Richter et al.) etc.) are not considered. The authors should motivate their selection of the absorbers they have added, and check the effect of the others they are still omitting (or give reasons/references why it is justified to skip them).*

The short text on these issues in the introduction has been adapted/extended to account for this and the reviewer's remark regarding "page 10622 lines 13-22" below. Since this additional information has too many details for an Introduction, it has been moved to Sect. 4.1.1. The conclusion there is that glyoxal, sand and VRS absorption signatures need not be taken into account at the moment. We believe that we should only include well-established absorption signatures in the operational OMI NO_2 retrieval at this moment.

Further comments

– *Abstract lines 5-6: The SCDs of OMNO2A are not only used in the current DOMINO and NASA products, but in all versions before as well.*

The reviewer is right that this should be mentioned; the text has been adapted.

– *Abstract line 23-24: ... independent NO2 retrievals from other instruments ...*

Thank you.

– *Abstract page 10620 line 28ff: This is a reasonable anticipation, but the authors should check this (e.g. by showing a map of the difference in NO2 SCDs between the fit versions).*

The requested map has been added to the new Sect. 5.4 and the possible changes in the tropospheric NO_2 column are discussed.

– *Introduction page 10621 lines 12&15: For very polluted places, the tropospheric column can be higher than 90% of the total column, and higher than 10e15 molec/cm2.*
– *Introduction page 10621 line 15: Stratospheric columns can be much higher than 2e15 molec/cm2, in particular at higher latitudes.*

The text has been adapted accordingly.

– *Introduction page 10622 lines 13-22: Please extend this paragraph by adding some historical context: "Early satellite retrievals of NO2 focus on the dominant absorbers, i.e. the Ring effect, O3, and H2O (refs). During recent years, continuous progress was made by accounting for weaker absorbers (O4, liq. water, CHOCHO, vib. Raman) (refs) or pseudo-absorbers (e.g. sand, Richter et al.)."*

The text of introduction has been adapted, as suggested by the reviewer, and moved to Sect. 4.1.1.

– *2.1.2 page 10624 line 19: "The standard OMI NO2 SCD..."*

Indeed; corrected.

– *2.1.2 page 10624 line 26: Refer to the updates you are going to introduce: "In the modified fit presented in this study, these standard settings are modified/exteded by ...; for details, see sect. ..."*

Given that a column listing the updated OMI settings is added to Table 1 (see remark further down), it suffices to add a line to the table caption.

– *2.2: Shift the equation more to the beginning of the section, and add references.*

The order in which the elements are presented in this section is changed to be able to place the equation near the beginning of this section, and the text was streamlined and somewhat shortend. To our knowledge, the fit equation has not been published before in a peer reviewed paper, so a reference cannot be given.

– *Page 10631 line 9: "a" → "at"*

This typo has been corrected.

- *Page 10632: For the discussion of differences in the solar spectrum and the cross sections, total differences are only of minor relevance. Much more important are differences of the high-frequent spectral structures, in particular for the solar spectrum. It would thus be interesting to add plots showing the difference new vs. old.*

We now include a graph showing the differences between the high-resolution and the convolved solar spectra in the Supplement (Fig. S5).

- *Page 10634 line 12: If the cross section is given with a spectral resolution below that of OMI, convolution with the OMI SF is not appropriate (rather than "not strictly necessary"), as this further smooths the - already too wide - absorption structures.*

This is indeed the case, but for consistency sake all spectra are treated the same; the effect on the resulting fit parameters appears to be negligible.

- *Page 10635 line 22: I do not agree with this statement; a squeeze/stretch can be regarded as a wavelength dependend shift. From table 4, there are clear indications of a change of the shift with wavelength. Thus, it is not appropriate to assume the shift being constant over the whole wavelength interval, in particular for such large intervals as in wcF. The authors should thus test how far allowing for a squeeze/stretch improves the spectral calibration.*

The wavelength shift following from the calibration differs indeed for different window. Whether this means that a squeeze could be fitted as well cannot be directly concluded from this. If a squeeze was to be included in the calibration, then it would only make sense to use the full trace gas fit window for the wavelength calibration (so as to avoid extrapolation of the calibration results). Whether including a squeeze would in the end improve the NO_2 fit results is difficult to say.
Unfortunately the operational OMNO2A processor does not allow for a squeeze to be added to the wavelength calibration routine. Adding the option would require a major overhaul of the operational software at the NASA SIPS, which has a rather inflexible basis for changes in the DOAS procedure. For the forthcoming TROPOMI instrument we plan to start off with fitting only a shift but during the commissioning phase we will investigate the usefulness of adding a squeeze. See the next point for more remarks.

- *4.2.2: The authors discuss artefacts in the NO2 SCD for inaccurate spectral calibration. These effects have been discussed before (e.g., Stutz and Platt, Appl. Opt., 1996; Beirle et al., AMT, 2013). These studies should be referenced. The authors conclude that these findings "pose firm requirements on the accuracy of the wavelength grid" of future satellite missions. Please comment if the accurate, but very time-consuming wavelength calibration done for OMI is foreseen to be done operationally in future missions, e.g. for TROPOMI. A faster solution might be to account for the spectral structures caused by a small shift in the wavelength calibration by including a "shift spectrum" as pseudo-absorber in the fit (Beirle et al., 2013).*

The references mentioned by the reviewer have been added to Sect. 4.2.1.
Regarding the plans for TROPOMI: using only a shift in the wavelength calibration is not expected to be very time consuming, as such a calibration can be done with a simple linear fit; it will therefore not be a problem for the TROPOMI processors. But as we also plan to test the need of a squeeze, we are investigating the options in terms of speed and of programming issues.
At the moment a wavelength calibration method inspired by the approach proposed by Beirle et al. (2013) is being implemented in the TROPOMI processor, so that we can test the need for a squeeze easily. Initial tests of the method on OMI level-1 radiance data using different calibration windows indicate that the approach works well. Preliminary results indicate that even for the large calibration window [405nm:490nm] the squeeze is very small, if not absent. It looks like the calibration works better if a (well-calibrated) Ring spectrum is taken into account (as is done in OMNO2A). Results of this investigation fall outside the scope of the present paper. A TROPOMI Technote will be prepared with details on the calibration procedure and results for tests using OMI data.

– *Table 1: I propose to add the updated settings to this table.*

A good suggestion; they have been added as additional column. The column on the SAOZ retrieval has been deleted from Table 1, as it is not relevant for the main paper now that the sections on ground-based data have been moved to the Supplement.

– *Figure 12: The liquid water is shown as global map. It would be interesting to have additional maps, in particular the NO2 SCD, the difference of NO2 SCD vs. the old set-up, and the fit results for all other aborbers (the latter probably in the supplement), in order to evaluate the fit performance and possible artefacts.*

See the new Sect. 5.4 in the main paper and Sect. S7 in the Supplement.

Main changes to the paper

The text of the main paper has been revised in the light of the comments and suggestions of both reviewer #1 and reviewer #2. To further strengthen the paper, some (parts of) sections have been moved to the Supplement. The main changes are the following:

– The abstract and introduction have been adapted in several places in line with the suggestions of the reviewers and other clarifications we deemed necessary. A paragraph regarding considerations for not including other absorbers in the fit has been extended, as requested by reviewer #1, and subsequently moved to a new Sect. 4.1.1.

– The previous Sect. 2.1.1 has been shortend by removing non-essential text and rewording the remainder, and it has been merged with the previous (somewhat adapted) Sect. 2.1.2 into one Sect. 2.1.

– The sections 2.3 and 3.2 in the previous version of the paper presenting the comparison between satellite and ground-based NO_2 data have been moved to the Supplement, because the information is not the key message of the paper. The results are, however, useful in that they clearly show that the bias in the stratospheric NO_2 between OMI on the one hand and GOME-2 and SCIAMACHY on the other hand find their origin in the OMI data (and not in the GOME-2 data). In addition, the comparison between the ground-based measurements and OMI data has not been published before.

– Sect. 2.2 has been reorganised, presenting the DOAS details in a more logical order, thereby shortening the section.

– With the removal of Sect. 3.2 (see above), the whole of Sect. 3 is merged into one section without subsection headings. Fig. 1 has been reduced to show only one panel (March 2007), since for other months the comparison looks quite similar.

– The subsections on the reference spectra are moved from Sect. 4.1 to the Supplement (Sect. S4.1–S4.2), which now includes the section on the water vapour absorption spectrum details that was already in the Supplement, as well as the plot showing the differences between the v2006 and v2014 solar reference spectra requested by reviewer #1.

– Now that the details of the reference spectra have been moved to the Supplement, a new section (S4.3) therein documents an instrumental issue affecting the Ring fit coefficient for OMI detector row 0 in the updated data version processing.

– The header of Sect. 4.2.1 has been removed, as it is not really necessary. What was Sect. 4.2.2 is now 4.2.1; the text therein has been reorganised, so as to make it better readable.

– The text of Sect. 5.1, 5.2 and 5.2.1 have been merged into one Sect. 5.1 (removing the non-essential equation for the geometric air-mass factor).

– A paragraph and a figure have been added to Sect. 5.1 to show the robustness of the changes in NO_2 SCD and RMS error, as requested by one of the reviewers.

- Part of the section presenting a preliminary comparison between OMNO2A and QDOAS results has been moved back from the Supplement to the main paper (now Sect. 5.3), following the request of reviewer #2. The QDOAS non-linear fit equation that was in the first part of this section has been moved to the DOAS section, Sect. S1.
- Some global maps of retrieved quantities and maps of the differences between the current and updated values of these quantities, requested by reviewer #1, have been added in the form of the new Sect. 5.4 in the main paper and the corresponding Sect. S7 in the Supplement.
- Sect. 5.5 has been deleted, as it is not essential to the paper.

With these changes, the main paper is reduced from 17 to 15 pages in AMT format.