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 #2 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.

Summary

> *Geffen et al. present results of OMI NO2 columns derived with an improved version of their NO2 slant column retrieval algorithm. They show column values from the old and new algorithm, and present comparisons with NO2 from independent satellite observations. The improved OMI NO2 data remove the known high-bias in the stratospheric columns that the OMI NO2 product has been suffering from.*
> *In principle, the results presented in the paper are well worth publishing, particularly since OMI NO2 is a high-impact data product frequently used in air quality studies. Making the user- and general science-community aware of improvements in this product is important. However, the manuscript is unpublishable it its current form. As I already pointed out in my initial "quick review", the manuscript is way to lengthy and unfocused. It resembles an ATBD (Algorithm Theoretical Basis Document) much more than a science publication. The latter is supposed to highlight important aspects of scientific development and not, like the former, present an unfiltered log book of all the gruesome details of algorithm development and retrieval elements. In fact, I wouldn't be surprised if a good portion of the writing here was taken from van Geffen (2014), the TROPOMI ATBD.*

We agree with the reviewer that publishing improvements in the OMI NO_2 slant column data processing system is important. In this paper we have tried to incorporate all relevant points regarding the improvements as well as some processing system issues relevant for the data users but not yet published in a peer-reviewed paper (see also below at the first point of the "Suggestions for revision" section).
Our paper is certainly not a translation of the existing ATBD of (TROP)OMI NO_2. The (not yet publicly available) ATBD TROPOMI NO_2 discusses all aspects of the NO_2 processing chain, but here we focus on NO_2 slant column retrieval only. In fact, the ATBD refers to this study for the specifics of both OMI and TROPOMI NO_2 spectal fitting, which have not been published in the peer-reviewed literature before.

> *To compound the problem, the authors have thought it a good idea to add a "Supplement" with even more details. Sure, electronic real estate is cheap these days, and why not cram all there is ever to know about an algorithm development into a PDF and data-dump it online, unabridged. But neither a reader's nor a reviewer's time is that cheap, and authors should be mindful of conveying essential information in a journal publication, so that key elements are available for easy grasp and reference. By all means, have a 50 page Supplement, if you must, that can also serve as ATBD. But do not waste readers'/reviewers' time with a manuscript that is unable to concisely make its point.*
> *It its current form, I recommend rejection of the paper. If the manuscript is SEVERELY shortened, it may be resubmitted for a new review process. The results are important, so I strongly encourage the authors to revise the paper for eventual publications. The comments below include some suggestions for revision.*

We thank the reviewer for encouraging us to revise and therewith improve the paper. The changes we have made to the paper and the supplement reflect the issues raised by both reviewers.

Suggestions for revision

> *What I expected from this manuscript was a clear description of the (1) problem (OMI NO2 high bias), (2) the steps that were taken to remedy that problem (concise summary of old and new algorithm approach), and (3) selected results (comparison of old and new OMI NO2; comparison with independent measurements) that highlight the improvements. All of these elements are in the current manuscript, but buried in between lengthy descriptions of algorithm elements that, while being essential for the retrievals, do not need to be spelled out in detail. An prime example is the descriptive list of crosssections: In a paper like this, it is completely pointless to show images of convolved cross-sections, old and new, and describe at length what they are.*

Though the observerved high bias in the OMI NO_2 stratospheric data is the direct motivation for this study, that is actually only one of *six* reasons the study into improvements and the paper were started:

- the need to understand and repair the observed high bias in the OMI data;
- the fact that some reference spectra were outdated (ozone, water vapour);
- the need to incorporate the wavelength and row dependency of the OMI slit function in the convolution of the reference spectra;
- the fact that the choice of the wavelength calibration window used in OMNO2A has never been studied in detail;
- the need to investigate whether including the absorption by O_2-O_2 and liquid water would improve the fit results;
- the need to document the OMNO2A slant column retrieval, including some aspects that where never documented publicly before, in a comprehensive way in a peer-reviewed paper, so that OMI NO_2 data users can refer to it easily, both regarding the previous and the updated versions of OMNO2A.

Apart from the latter point, the issues were mentioned in the abstract and the introduction, but admittedly in a round-about way. From the reviewer's remarks we must conclude that the points were not conveyed clearly enough. The introduction has therefore been adapted & expanded to outline these points.
Regarding the descriptions of the reference spectra: the text of Sect. 4.1 and the figures have been moved to the supplement, leaving only a list of the source of the (updated) reference spectra.

> *In essence, everything before Section 5 is nonessential. In the Intro to Section 5, the authors repeat the lists of changes (4 bullet points). Start from there and fill in the blanks with concise descriptions what has gone into them. Don't use 17 pages to do that! Some ways to shorten:*
> *Section 2.1.1 has three paragraphs, each an almost verbatim copy of the other, differing only in the name and characteristics of the satellite instruments (OMI, GOME2, SCIAMACHY). Include only that information which is actually relevant to the work, and/or put the values sidebyside in a table, if you must. If spectral sampling is not specifically used/mentioned in the comparisons, then there is no point in listing it.*

The wording of the section has been changed and non-essential information is removed (wavelength ranges, spectral resolution and spectral sampling).

> *Sections 2.1.2/2.2 can be combined and shortened. DOAS is a pretty well known technique by now. First describing model function elements over two paragraphs, and then showing the same again in an equation is repetitively useless.*

Sect. 2.1 – formerly 2.1.1 + 2.1.2 – describes the satellite instruments and the main settings used in the NO_2 retrieval, in as far as relevant for this study. Sect. 2.2 then describes, independently of the source of the spectral data, elements of the DOAS technique necessary for the content of the remainder of the paper. The text of Sect. 2.2 has been revised, to present the issues in a more logical order, and thereby reducing its length.

> *Section 3 wastes two pages, two figures, and one table to establish the fact of the OMI NO2 stratospheric bias, when it has already been published in literature (Belmonte-Rivas, 2014). It also does so without mentioning that this is entirely about the old version of the retrievals. This section should be deleted altogether, including figures 1 and 2. Elements of figure 1 can be used for a graph that shows old and new OMI with GOME and SCIAMACHY.*

The study of Belmonte-Rivas et al. (2014) does not contain the same information as this paper. For example, GOME-2 data is not used in that paper, nor does it present a comparison against ground-based measurements at Jungfraujoch, which is relevant as it provides the evidence that OMI is biased high against an independent non-satellite reference. However, since this comparison is not a key point of the paper, it has been moved entirely to the Supplement.
With this change, all of Sect. 3 can be combined into one section, which leads to the essential conclusion, that the high bias in the OMI data is rooted in the OMI NO_2 slant column processing, not in the subsequent steps leading to the stratosphere/troposphere separation. In order to shorten the paper further, Fig. 1 is reduced to one panel only.

> *Section 4.1 can be condensed to a few sentences that list the changes in crosssections for the new NO2 retrieval. Figures 3, 4, and 5 should be deleted or. Alternatively, wavelengthdependent fitting residuals could be shown that demonstrate the effect of changing from the old to the new crosssections. That would actually provide some information for the retrieval expert. A mere sidebyside display of old and new crosssections, at least in the context of this manuscript, is silly.*

We agree with the reviewer that Sect. 4.1 contains rather many details. They were included here in view of the need to be complete and document elements of the processing not published before in one peer-reviewed paper. In order to improve readability of the paper and still retain the full description, most of Sect. 4.1 has been moved to the Supplement, including the figures, leaving only a list of the updated cross sections.

> *Section 5 makes two references to the Supplement - for the "spectral residual of the NO2 retrieval" and for a "OMNO2A - QDOAS" comparison. Both of these may actually be better served if included in the main text. After all, the authors want to convince us that their new retrievals are an improvement over the old ones. A concise description should fit well within the flow of the manuscript.*

We agree with the reviewer and now include a large part of the "OMNO2A - QDOAS" comparison in the main text; the rest remains in the Supplement, so as not to make the main paper too long with too many details.

> *In general, a Supplement distracts the reader and detracts from the main text. A Supplement should be used to provide relevant information in support of claims that can't be elaborated on in the framework of the main text (GRL page limits come to mind). Given that this work does not, in fact, describe new science in as much as it describes an improvement of a data set, I seriously challenge the necessity of having any Supplement at all.*

In order to document the current and updated OMI NO_2 spectral fitting, we think a Supplement is very useful, thus providing easily referable and essential documentation to OMI NO_2 data users.

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.