A global aerosol classification algorithm incorporating multiple satellite data sets of aerosol and trace gas abundances by M. Penning de Vries et al.

Authors' reply to the interactive comment by anonymous referee 2

First of all, we would like to thank the referee for his/her very positive review of our manuscript. We used the referee's comments to improve our manuscript further; our replies and the changes made to the manuscript are given below, colour-coded for clarity:

Green: Referee comment. Black: Authors' reply. Red: Modified text in manuscript.

Referee comment: 1. What happens if both the MODIS deep blue algorithm and the MODIS dark target algorithm provide an AOD (Collection 5.1, not combined in the MODIS product)? Does GACA average both values or use the deep blue values to fill gaps of the dark target algorithm?

Authors' reply: Indeed we had to combine AOD data from the dark target (DT) and Deep Blue (DB) algorithms, because the reprocessing MODIS Collection 6 data, where the results of all AOD algorithms are combined, was not completed at the time of writing. We assumed that where the DT algorithm provided an AOD, this value was more accurate than that retrieved by the DB algorithm, because the former algorithm is more rigourously validated than the latter. Hence DB data were only used to fill gaps in the DT data set. To clarify our approach, we changed the sentence starting on page 13556, line 24 ("The retrieval algorithms...") to:

The retrieval algorithms for aerosols over ocean and dark land are described in Remer et al. (2005), and Levy et al. (2007b), respectively. For bright surfaces where no AOD value is available from the dark target algorithm (mainly deserts), the Deep Blue product (Hsu et al., 2004) is used.

Referee comment: 2. How different is the angstrom exponent in the MODIS product from the angstrom exponent calculated for GACA?

Authors' reply: The Ångström exponent (EAE) used in GACA is determined from the gridded, monthly mean AOD at 470 and 660 nm, which are the only two wavelengths common to all MODIS aerosol retrieval algorithms. To illustrate the effects of these two differences, in the figure below we have plotted the over-ocean EAE in three different ways: in panel A we present the Level-3 EAE (mean of daily mean, determined from AOD values at 865 and 550 nm). Panel B shows EAE, determined from monthly mean AOD values at 865 and 550 nm; panel C contains EAE as used in GACA, i.e., EAE determined from monthly mean AOD values at 660 and 470 nm. The colour scale was chosen so that EAE associated with large aerosols (EAE < 0.75) in GACA appear in blue, those for small aerosols (EAE > 1.25) in yellow-red tones, and medium-sized aerosols (intermediate EAE) in gray.

The differences between the three panels, although present, are minor, and are not expected to affect GACA results significantly, therefore we decide to leave the EAE input unchanged.



Referee comment: 3. Which datasets are used to estimate the thresholds in Table 3?

Authors' reply: We thank the referee for this question; since referee 1 addressed the same issue, it is clear that it requires some clarification. The determination of the thresholds is certainly an important part of GACA, as mentioned at the beginning of Sect. 5.1.: "It is clear that GACA results depend on the choice of thresholds and criteria for aerosol type and source determination.".

As explained in the manuscript on page 13561 (lines 17-26), the UVAI and EAE thresholds used for aerosol type assignment are motivated by Fig. 1. Trace gas thresholds and source assignment criteria were determined in a more subjective way, i.e., by starting out with some simple assumptions (e.g., biomass burning is associated with HCHO and CO emissions; NO₂ pollution mostly originates from anthropogenic (urban/industrial) activities) and adjusting the criteria and thresholds iteratively until consistent seasonal maps were obtained.

To make this procedure more clear, we inserted the following statements on page 13565, line 15, between "(...) of which the values were chosen empirically." and "The ΔCO threshold, (...)":

The source assignment criteria were chosen based on textbook knowledge (e.g., that biomass burning is associated with HCHO and CO emissions), as detailed for each source type in Sects. 3.3.1-3.3.8, and were adjusted iteratively to obtain consistent results. The quantitative understanding of aerosol-trace gas relationships, however, is currently not sufficient to derive trace gas thresholds in a systematic way, hence the trace gas thresholds were determined in a more empirical fashion. The thresholds were empirically chosen high enough to exclude noise (or natural variability), but low enough that the associated sources are recognized. The thresholds were chosen independent of region and season to keep the algorithm globally consistent. A future development of GACA may be the adoption of threshold climatologies to better account for regional and seasonal variability of trace gas and aerosol emissions (see Sect. 5.4).

And we replaced the sentence starting on page 13575, line 12, ("Nevertheless, ...") by:

Most source assignments are rather robust and altering thresholds only causes small shifts of borders between different sources. Beyond being rooted in textbook knowledge, our criteria are justified by the consistency of the obtained results and the good general agreement with MACC model results.

Incidentally, we corrected the typo in the range of ΔCO in Table 3 to: $0 - 40 \times 10^{17}$ molec/cm².

And the caption of Fig. 3 was corrected to:

Relationship between $1^{\circ} \times 1^{\circ}$ monthly mean values of AOD and trace gas columns (in molecules/cm²) for a region in central Africa ($2^{\circ} - 4^{\circ}$ S, $18^{\circ} - 20^{\circ}$ E; left panel) and in the East Pacific Ocean ($16^{\circ} - 18^{\circ}$ N, $162^{\circ} - 164^{\circ}$ W; right panel) for July-August 2007-2011. Dots depict NO₂ (blue), HCHO (green), and SO₂ (red) VCDs and excess CO VCDs (light blue, scaled by a factor 0.01) and their respective thresholds. The threshold values of NO₂ and SO₂ are identical (dotted blue and red lines). Note the differences in y-axis scales.

Referee comment: 4. Which criteria are used to screen out sun-glint conditions? Is the sun glint filter applied to all instruments separately? The authors correctly identify a wind-speed related problem in the MODIS data. This is caused by the wind-speed dependence of the glint effects. I would guess that for SZA < 50 between 30% and 50% of the MODIS swath are potentially ill-posed for the retrieval of the AE.

Authors' reply: Only aerosol data are screened for sun-glint effects: the MODIS aerosol retrieval is not performed on pixels satisfying certain sun-glint criteria (e.g. scattering angle and wind speed; see Remer et al., 2005 for details), which, as the referee points out, causes the loss of a large amount of data at low latitudes. GOME-2 UVAI from pixels under potential sun-glint conditions (sun-glint angle $< 18^{\circ}$ or sun-glint angle $< 11^{\circ}$ and cloud fraction < 0.3 and/or no high clouds) were discarded from the data set, as recommended in the "ATBD for the GOME-2 Aerosol products" by de Graaf et al. (EUMETSAT document O3MSAF/KNMI/ATBD/002, 2013). The effects of sun-glint on trace gas retrievals is minor and was ignored in this work in favor of better statistics.

We added the citation to the revised manuscript on P. 13558, line 7 (after "and solar eclipses"): as recommended in the "ATBD for the GOME-2 Aerosol products" by de Graaf et al. (2013).

Referee comment: 5. The high UV index at 60° south could be caused by geometric limitations dependent on the scattering angle (is the scattering angle close to 90° or lower?).

Authors' reply: The unphysically high UVAI in the Southern Oceans might certainly be caused by the small scattering angle. Following the recommendations given in the ATBD for the GOME-2 Aerosol Products (see above), we discarded UVAI from pixels with scattering angles $< 90^{\circ}$, which eliminates the major fraction of affected pixels, but not all. Using a tighter constraint, e.g. requiring that the scattering angle $> 100^{\circ}$, further reduces the number of affected pixels, but also causes the loss of unaffected data. As a compromise, we leave the constraint at $< 90^{\circ}$, but note that for future studies, better criteria for the exclusion of scatter-angle affected pixels should be found.

We added this explanation to the manuscript on page 13569, by changing lines 4-6 to:

the apparent band of desert dust at 60° S is caused by a few data points with unrealistically high AOD, as mentioned above, in addition to erroneous (high) UVAI values that are probably caused by small scattering angles ($90^{\circ} - 100^{\circ}$) encountered in this region.

Referee comment: 6. The authors mention that GACA underestimate the amount of desert dust compared to a model. It would be interesting to see if the results change if the GOME UV aerosol index is replaced by the correspondent index from OMI or the GOME PMD. However, this is probably out of the scope of this paper.

Authors' reply: The reason that GACA possibly underestimates desert dust from the source is related to an instrinsic property of the UVAI: its dependence on the altitude of the aerosol layer. As a layer of absorbing aerosols (e.g., desert dust) descends, its UVAI decreases — even if all other properties remain constant (see, e.g., de Graaf et al. (2005) or Penning de Vries et al. (2009)).

UVAI from GOME-2 and OMI generally agree very well and apart from a small offset, UVAI from GOME-2 PMD is in excellent agreement with UVAI from the nominal science channels. We do not expect GACA results to change significantly if another UVAI dataset would be used, although the UVAI thresholds might need to be shifted slightly.

Referee comment: Specific comments: p. 13558: Maybe it would be helpful to add a few sentences on the basic methods used for the trace gas retrievals instead of giving just a reference.

Authors' reply: We agree with the referee; the manuscript was changed accordingly by adding the following lines to the beginning of Sect. 2.2.3 on page 13558:

Total column densities of SO_2 and HCHO, and tropospheric column densities of NO_2 are retrieved by DOAS analysis (Differential Optical Absorption Spectroscopy, see e.g., Platt and Stutz, 2008; Richter and Wagner, 2011) of GOME-2 spectra in the UV-visible range.

Referee comment: p. 13559 line 14: analyses Authors' reply: Corrected.