August 3, 2015

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 1

We thank the referee for his/her constructive review of our manuscript; particularly for the literature references that we had overlooked. Our replies to the referee comments 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. Authors need to note relevant work in the past. There have been studies to classify aerosol type either by multiple channel algorithm (e.g., Higurashi and Nakajima, 2002; Lee et al., 2010), or by using aerosol parameters such as AI, AE, FMF from different satellite instruments (e.g. Jeong and Li, 2005; Kim et al., 2007).

Authors' reply: We thank the referee for the list of references. We added several references to the introduction as follows below; the study by Higurashi and Nakajima was already referenced on page 13554, line 6 (and later on in the manuscript). The paper by Lee et al. introduces an aerosol retrieval method and as such does not fit very well in our introduction; the paper by Mielonen et al., mentioned in referee comment 9, was added instead.

To further discriminate between aerosol types, differences in absorption can be exploited (as e.g., in Higurashi and Nakajima, 2002; Jeong and Li, 2005; Kim et al., 2007; Mielonen et al., 2009).

Referee comment: 2. It is not clear how the threshold values are determined. Those should be based on physical reason and/or previous work, other than stated as 'empirical'. As authors have dealt with all the satellite dataset, they can look into the details.

Authors' reply: The referee has pointed out an important question; since referee 2 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; NO_2 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.

Referee comment: 3. Introduction: There have been studies to classify aerosol type using aerosol index from TOMS (or OMI) and AE (or FMF) from AVHRR (or MODIS) as listed above. Thus, it is appropriate to mention such work, which are relevant to the current studies.

Authors' reply: Please see our answer to comment 1 above.

Referee comment: 4. Lines 284-285: What is the threshold value of HCHO/NO2 and how is the value determined? I have similar questions on other threshold values as listed in Table 3 for example. Although the values were stated to be 'empirical', it is desirable to have reference or physical reason. Are the values global or regional?

Authors' reply: All threshold values are given in Table 3; for the determination of those values, please see our answer to comment 2 above.

We added a reference to Table 3 on page 13563, at the end of line 25: (given in Table 3).

Referee comment: 5. Lines 329-339, Table 3: The threshold values of SO2 and HCHO suggested in this table is below the measurement uncertainty. How can these be justified in classifying aerosol type other than volcanic SO2?

Authors' reply: The measurement uncertainty for single measurements is higher than the chosen threshold, but the statistical error of the mean is much smaller for gridded, monthly mean data. The SO_2 data are, indeed, only used to detect volcanic sulfate (VOG), as they do not pass the threshold value of 1 x 10^{15} molec/cm^2 for any other sources (except in Eastern China and South Africa; see Fig. 8 and the accompanying text on page 13570, lines 22-25).

Referee comment: 6. Lines 311-313, 398-401: It is not physical to allow SS classification over land other than coastal area. Furthermore, AOD of SS tends to be very low, as indicated in the manuscript, thus is beyond the detection limit, especially over land where the AOD retrieval uncertainty is higher than over ocean.

Authors' reply: The referee is right in saying that the SS classification over land is not physical. We state this on on page 13565, on lines 5-7 ("GACA does not discriminate between grid boxes located over land and ocean, therefore the SS type is also regularly found over land and may be interpreted as a generic background type."), and again on page 13568, on lines 23-26, where it is mentioned that "The misclassification of SS aerosols over continents in the high latitudes is most apparent in fall (lower-most panel). These grid boxes show no enhanced trace gas concentrations and have mean AOD$<$ 0.15, corresponding to the definition of SS in GACA.". The referee also correctly points out the increased uncertainty of aerosol retrieval over land. However, in the regions classified by GACA as being SS-dominated, the AOD is systematically over 0.05 (in contrast to regions classified as "na"). One solution would be to define a separate class over land and ocean, but we choose not to, preferring to keep the algorithm general and global.

To clarify the interpretation of SS occurrence over land, we added a sentence after lines 23-26 on page 13568 (quoted above):

These aerosols may be regarded as background aerosols of which the source cannot reliably be determined by GACA.

Referee comment: 7. Lines 357-363: What about the aerosol type over western U.S. during summer when wild fires are frequent? The algorithm seems to detect such features. Please add statements.

Authors' reply: Over the Southwestern USA, GACA finds mainly large absorbing (LA) aerosols and some contributions from MA, LN, and MN aerosols (Fig. 4), which points to desert dust as a main source and possibly other, minor sources as well. However, the trace gases associated with fire, i.e. HCHO and CO, are not significantly enhanced over this region, hence the biomass burning source type is not selected. As wildfires occur at irregular intervals in time and space, data points associated with smoke may have been screened out by the removal of outliers that is performed as a first step in GACA (see Sect. 3.1). Yet, even if outliers are not removed, BB does not appear as a major source for aerosols in North America (see Fig. S4 in the supplementary material).

As a recognition of these facts, we added the following statement to page 13567, after the sentence ending

on line 12:

Despite the fact that wildfires occur frequently in summer in North America, BB is not selected as a major source there. This is because forest fires occur at irregular intervals, so that their signal is suppressed as a consequence of averaging data in time and space.

Referee comment: 8. Lines 409-414: Unusual behavior in smoke plume in Southwestern coast of Africa can be a mixture of small and large particles such as dust lifted together from ground surface by large buoyancy with fire.

Authors' reply: The referee makes an interesting point here, however we have found no evidence for this hypothesis in literature on measurements of aerosols by different satellite instruments (MODIS, CALIOP, OMI, SCIAMACHY) in this region.

Referee comment: 9. Section 5.3. There have been studies to classify aerosol type from AERONET beyond the fine/coarse mode, as listed below for example. At least it should be noted as previous works in classifying aerosol type from AERONET.

Authors' reply: We thank the referee for pointing out these studies; we included the paper by Lee et al. in the discussion section on page 13578, as follows:

The most established empirical climatologies are derived from AERONET data (e.g., Dubovik et al., 2002; Omar et al., 2005; Levy et al., 2007a; Lee et al., 2010). At a first glance, the agreement between GACA-source and AERONET-derived climatologies (e.g., Fig. 2 in Omar et al., 2005, Fig. 3 in Levy et al., 2007a, and Fig. 2 in Lee et al., 2010) is good. However, due to large differences in spatial sampling and the limited information available from AERONET, the informative value of such a comparison is limited.

The paper by Mielonen et al., however, does not fit to the discussion section, as the results cannot easily be compared to GACA. We included the paper in the introduction section instead (see our answer to comment 1).