

Review of “Aerosol classification by airborne high spectral resolution lidar observations” by Groß et al.

We would like to thank the anonymous reviewer the useful comments and suggestions which help to improve the clarity and scientific quality of this paper. The answers to the comments are given in a direct response (bold, italic).

This paper presents a very useful dataset of airborne HSRL aerosol measurements. The authors show how these measurements can be used to help qualitatively identify and in some cases classify aerosol types and some aerosol mixtures. There are relatively few such measurements and so these represent a significant addition to the database of such HSRL measurements. Current and future satellite lidar aerosol retrieval algorithms can benefit from such aerosol classification schemes. The paper is well written and easy to follow. However, there are some significant issues the authors should address before the paper is published.

These include the following:

1. The authors imply that this classification can be used for EarthCARE processing but do not address the fact that EarthCARE will employ only a single wavelength (355 nm) that differs from the wavelengths used here (532 and 1064 nm).

We revised the conclusion according to these suggestions.

2. The Burton et al. (2012) paper also described extensive airborne HSRL measurements of the same aerosol intensive parameters of very similar aerosol types. The authors point this out in the introduction but then repeatedly fail to mention or reference these prior HSRL measurements when describing in detail the values of aerosol intensive parameters of specific aerosol types. It is important to point out the similarities and differences in these measurements to provide the reader some indication of the variability of intensive parameters associated with these aerosol types. As additional measurements are made over other locations, one often finds additional aerosol types and/or wider ranges of the intensive parameters measured by the lidar. This complicates such classification schemes.

We already included the Burton et al. paper in the discussion on certain aerosol types. We now extended this discussion to all comparable aerosol types included in the classification scheme.

3. The different aerosol types described in Figure 5 were not classified using the lidar data but by other means. It appears that the strategy used was to use external information (e.g. backtrajectories) to select specific cases of specific types, then to infer the lidar intensive properties appropriate to those types, and finally to codify the results by coming up with an algorithmic classification scheme that would approximately achieve the same separations. This procedure is not incorrect, but the authors should clearly indicate how the classification scheme was developed. Furthermore, they should indicate the extent they used backtrajectories for developing and then evaluating the classification scheme.

We revised this paragraph to make our strategy more clearly. Furthermore we extended the description about the procedure on how we used for backward trajectory analysis to identify different aerosol types and source regions.

4. There is little if any information provided to allow the reader to determine the uncertainty in the classifications made using these lidar data. The ability of users to make use of such information depends on the uncertainty associated with these classifications. Some of the classifications made here can be made more confidently than others. Note that there is significant overlap in all three dimensions among some of the types shown in Figure 5 (black and dark green; red and orange) which indicates that the three lidar parameters are not entirely sufficient to separate the various types. The authors should at least comment on the uncertainty of the classification scheme presented here.

We included a discussion about the uncertainties of the aerosol classification scheme.

5. At least two of the aerosol types presented here (e.g. Figure 8) are mixtures, not pure types. This is mentioned in section 3.2, but should be mentioned in the abstract and discussion.

We changed the discussion and abstract as proposed by the reviewer.

Other comments that should be addressed before publication:

1. (page 25984, line 6-7) African biomass burning and marine aerosol are mixtures.

We do not classify marine aerosols as a mixture, but we changed 'African biomass burning aerosols' to 'African biomass burning mixture'.

2. (page 25984, line 12) Does backward trajectory analysis really validate the classification scheme or does it rather indicate that the aerosol type or mixture is consistent with an air mass that came from a particular location? I think the latter is more correct. Coincident in situ size and composition measurements of a particular aerosol type or mixture constitute validation. The statement made in line 18 one page 25986 "...supported by trajectory analysis and validated with in situ measurements" is better.

We changed that.

3. (page 25985, line 7) completed sixth year.

We changed that.

4. (page 25985, line 9) Should be changed to "...lacks the ability of direct extinction measurements"

We changed that.

5. (page 25985, line 11) Should be changed to "...aerosol lidar ratio S_p normally has to be assumed". There are occasions of cases of elevated aerosol layers where scattering from aerosol-free regions above and below an elevated layer can be used to derive the lidar ratio of the elevated aerosol layer.

We changed that.

6. (page 25985, line 14) Some of the lidar ratio values and ranges of values do not necessarily agree with other measurements and so are not necessarily "typical". For example, the same Müller et al., 2007 reference and the Burton et al. 2012 reference indicate that the maritime lidar ratio is 23 ± 5 sr. Other references (e.g. Burton et al., 2012, Liu et al., 2008) indicate that dust lidar ratio is in the range 45-51 sr (25-75%). The authors should indicate that the lidar ratios for these various aerosol types have a greater range and variability than reported here. Liu, Z., et al. (2008), CALIPSO lidar observations of the optical properties of Saharan

dust: A case study of long-range transport, J. Geophys. Res., 113, D07207, doi:10.1029/2007JD008878.

We do not see that the lidar ratios for maritime aerosols reported by Müller et al., 2007 of 23 +/- 5 sr and by Burton et al., 2012 of 15-25 sr (significantly) differ from the reported values of 18 +/- 2 sr (Groß et al., 2011) and from our findings of 18 +/- 5 sr. To improve clarity, we included the results of Müller et al., and Burton et al. in our discussion.

Concerning mineral dust: It appears that the values of 56 +/- 5 sr for Asian dust (Sakai et al., 2002) and for Saharan dust originating mainly from the Bodele depression of 62 +/- 5 sr (Groß et al., 2011) represent the natural variability of the lidar ratio as a result of the natural variability in the microphysical and chemical properties of the aerosol particles from different source regions. Esselborn et al., 2009 already reported about the differences in the optical properties depending on the source region. Groß et al., 2011 found differences in the source region of SAMUM-2 dust layers compared to SAMUM-1. One of the dominant source regions during SAMUM-2 was the Bodele depression, which 'unusual nature' was already discussed in Todd et al., 2007. Additionally aging and modifications during transport may broaden this variability range. To distinguish different aerosol types by measurements of their optical properties, this natural variability has to be taken in account. The classification scheme presented in this study (Figure 8) considers this variability range. Both, source region location and measurements are tainted with uncertainties which prevent a more precise discrimination of the aerosol particles with respect to source region.

7. (page 25986, line 21) This study uses the HSRL measurements acquired at 532 nm and not the 355 nm wavelength to be used by EarthCare. The optical properties at 355 nm are not necessarily the same at 355 as at 532 nm. Therefore, this study illustrates the possibility of using EarthCare to distinguish these same aerosol types, but does not demonstrate that it can.

We removed the reference to EarthCARE at this point.

8. (page 25988, line 3) Do the authors mean SAMUM-1 was one of the first coordinated missions to study dust aerosols or aerosols in general? If dust aerosols, this should be stated. If aerosols in general, this is not true. There have been several missions much earlier that have done this. For example, see the JGR special sections regarding the TARFOX mission (http://www.agu.org/journals/jd/special_sections.shtml?collectionCode=TARFOE1&journalCode=JD and the overview paper at <http://www.agu.org/pubs/crossref/1999/1998JD200028.shtml> See also the MILAGRO special issue at http://www.atmos-chem-phys.net/special_issue83.html

Of all previous studies, SAMUM-1 was the most comprehensive dust closure experiment with a strong emphasis on the vertical profiling of optical and microphysical properties of mineral dust (see also Ansmann et al., 2011). We clarified this in the text.

9. (Page 25988, line 15) "where" should be "were"

We changed that.

10. (page 25989) It would be helpful to provide some basic information regarding the HSRL measurements. For example, what are the nominal temporal (spatial) and vertical resolutions of the retrieved backscatter, extinction, and depolarization profiles? What are the uncertainties of these profiles? What is the approximate detection limit?

We extended the description of the HSRL system with respect to temporal and vertical resolution, measurement uncertainties and detection limit.

11. (Page 25990) Were there any in situ scattering measurements acquired on the aircraft?

We have not performed light-scattering measurements aboard the research aircraft, because we realized from earlier measurements (e.g., Petzold et al., 2002), that the cut-off characteristics of the aerosol inlet ($D_{max} = 2.5 \mu\text{m}$) interferes with the light scattering measurement, because the coarse mode which is essential for dust measurements is not transferred completely through the inlet. Instead we decided to measure light absorption properties of the sub- $2.5 \mu\text{m}$ mode and calculate light-scattering properties according to the method described in Petzold et al. (2009).

12. (Page 25991, Measurement strategy) There are not many details here. How long (time, distance) were the various legs? How long were the flights? Were the in situ results averaged over the entire horizontal legs or were multiple in situ results obtained on each level leg? What were the typical altitudes of these legs?

We clarified this point by adding the following section to the manuscript: "As is shown in Fig. 2; vertical in-situ flight patterns consisted of a series of constant altitude flight sequences. Each sequence lasted for at least 5 min. The altitudes of the flight sequences were adjusted from previous lidar measurements. In homogeneous single aerosol layers (e.g. SAMUM-1) minimum two constant-altitude runs were conducted inside the aerosol layer with one close to the lower boundary of the layer or the surface, respectively, and another one close to the top of the aerosol layer. Depending on the vertical extension of the dust layer, a third run was conducted in the center of the aerosol layer, if manageable. In case of multi-layer structures and thin aerosol layers one constant-altitude run within the aerosol layer was performed. During data analysis, each flight sequence was checked for data homogeneity. Particle size distribution data were then averaged over sections of the respective flight sequence with homogeneous aerosol conditions."

13. (Page 25991, line 23) Were these modified trajectories computed as a check on reliability?

Yes, the modified trajectories were computed to check the reliability. We extended this paragraph to improve clarity, and refer to a former publication describing the procedure of trajectory analysis in more detail.

14. (Figure 3) What altitudes were these trajectories computed? How were the altitudes chosen?

The trajectories were calculated for the altitudes of interesting aerosol layers indicated by lidar and listed in Table 1. We modified the text and the figure legend to improve clarity.

15. (Page 25992, line 18) The backscatter ratio defined here is incorrect. I believe the authors mean $\text{BSR} = b/b_m$ where b_m is the molecular backscatter and b is total backscatter.

This was a typing mistake which we have corrected.

16. (page 25993, line 10) The motivation for aerosol type classification is weak. Some applications (e.g. radiative forcing estimates at the surface, and to some extent at the Top-of-Atmosphere) do not require vertically-resolved discrimination of aerosol types and so can

make use of column-integrated aerosol properties. (In contrast, radiative heating profiles do require vertically resolved measurements).

We modified the text to strengthen our motivation for aerosol type classification.

17. (page 25993, line 18) The authors should also include the paper by Sasano and Browell (Applied Optics, vol. 28, No. 9, 1989, p. 1670) when referencing how the color ratio can be used to discriminate particle types.

We included this reference.

18. (page 25993, line 28) When referencing “mixed Saharan dust layers”....what were these mixtures made of? Likewise, what was mixed in the “African biomass burning aerosol”? It would seem likely that dust was mixed with the biomass burning to obtain a depolarization value of 26%.

In the ‘mixed Saharan dust layers’ either marine aerosols or biomass burning aerosols were mixed with Saharan dust. In the layers of African biomass burning mixtures a contribution of Saharan dust was found. This is discussed in Section 3.3.

19. (Page 25994, line 16) The range (1.4-16.2) of CR for biomass burning aerosols seems much too large when compared to other measurements such as Burton et al. (2012) and Müller et al. (2007). I think this large range is produced by large uncertainties when scattering levels are small. It would be good for the authors to review the uncertainties at low scattering levels and determine the minimum backscatter level for trustworthy computations of CR.

As stated in the text, the uncertainties are large when the optical thickness of the aerosol layers is low. We agree with the advice of the reviewer and confined the range for biomass burning aerosols.

20. (page 25995, discussion of Figure 6). The lower thick line which is supposed to represent the minimum range of the Saharan dust-Biomass Burning mixing looks like it could also comfortably represent the upper range of Saharan dust-urban (Pollution) aerosol). How would one discriminate between such mixtures?

This is correct. Therefore for specific cases further (independent) information, e.g. from backward trajectories, are crucial. We have modified the text concerning describing possible uncertainties in our classification scheme to emphasize these potential ambiguities.

21. (page 25996, line 15) Can the authors provide reference(s) for these statements?

The reference is the following: ESA 2009, ICAROHS - Inter-Comparison of Aerosol Retrievals and Observational Requirements for Multi-Wavelength HSRL Systems; Task Report 1, Contract No 22169/NL/CT, Noordwijk, NL, 62 pp., 2009.

22. (page 25997, line 5) What supporting evidence is there that the linear depolarization of 14% for African biomass burning aerosol is due to supermicron dust particles?

In-situ measurements reported by Lieke et al., 2011 and Weinzierl et al., 2011 showed that large dust particles of different amount were found in the corresponding aerosol layers. We added the two references to support this statement.

23. (page 25998, line 8) In Table 4, why not also list the lidar ratio (~ 41 sr at 532 nm) of dust of Liu et al. (2008) ? These were derived close to the source, in an elevated layer well above the surface so this can be considered pure dust also (see Figure 6 in this paper).

We did not include the lidar ratio values of Liu et al., 2008 as they do not show direct measurements but indirect evaluation from other methods.

24. (page 25998, discussion of Table 5) Why only list measurements from ground-based lidars in Table 5? Why not also list the measurements from the airborne HSRL from Burton et al. (2012)?

We included the results of Burton et al. in the discussion of the analyzed aerosol types.

25. (page 25999, line 2) Here again, Table 6 omits any reference to the extensive airborne HSRL measurements of smoke reported by Burton et al. (2012). The authors are aware of this publication, but seem to avoid presenting results from this paper...why? Moreover, Burton et al. (2012) found considerably lower lidar ratio (~ 40 sr) and depolarization (5%) values for fresh smoke than reported here; so in fact the measurements reported in this paper do not necessarily agree very well with former findings.

The results of Burton et al. are added. Furthermore, we changed our aerosol type 'African biomass burning (fresh)' in 'African biomass burning mixture' as a certain amount of Saharan dust particles was found in these aerosol layers (see SAMUM publications referred in the text). As this is not a general feature for fresh biomass burning layers, we renamed this class and changed the discussion accordingly.

26. (page 25999, line 10) Here again, in Table 7, no mention of the marine aerosol measurements from Burton et al. (2012). "Prior" would be a better word than "former".

See above.

27. (page 25999, line 15) Burton et al. (2012) clearly report the HSRL measurements of ice particles, so the authors are incorrect in stating that such measurements are unavailable.

This was a misleading formulation; the correct statement is "as no own HSRL measurements of ice particles and pure volcanic ash are available so far".

28. (page 25999, description of Table 8.) The ice observed by Burton et al. (2012) was not associated with cirrus clouds as indicated in Table 8. These were extensive layers (often several kilometers thick) of ice particles that appeared more as a haze and sometimes appeared to be precipitating. In many cases, cloud camera images showed no evidence of any clouds and cloud clearing techniques employed by AERONET did not detect these as clouds. Perhaps the closest description of these is "altostratus nebulosus" as described by Sassen and Wang (Clouds of the Middle Troposphere, Surveys in Geophysics Volume 33, Numbers 3-4 (2012), 677-691, DOI:10.1007/s10712-011-9163-x).

We thank the reviewer for this clarification. The text was changed according to this hint.

29. (Figure 8) Is there some classification for cases where the depolarization is between 10 and 20% and the lidar ratio is below 30 sr? If not, then it looks like the last decision for selection of marine (depolarization below 10%) is not required.

There is no classification for cases where the depolarization is between 10 and 20% and the lidar ratio is below 30 sr yet. However, this could be the case e.g. for dry marine particles.

Since we do not have measurements of dry marine particle so far, we prefer to keep this selection.

30. (Page 26000, line 10) Over what part of the flight were the in situ measurements shown in Figure 10a acquired?

After probing the atmosphere with the HSRL from an altitude of about 9 km, the in-situ measurements shown in Figure 10a were acquired during descent into the aerosol layers. During descent, the Falcon stayed at constant altitudes for about 5-10 minutes at 3.3, 2.0, 0.7 and 0.6 km a.s.l. During this particular flight, the Falcon performed a box-like pattern in the vicinity of Santiago/Cape Verde so that the in-situ measurements were performed in close proximity to the HSRL measurements although there is a time difference of about 15-30 minutes between the HSRL and the in-situ measurements.

Are these at a single time or averaged over a flight leg?

To derive the absorption Angstrom exponents, we averaged over one constant altitude sequence, because the aerosol properties were very homogenous at the individual constant altitude sequences.

What does the dashed line at 1.5 km represent?

The dashed line indicates the border between the Saharan dust and the African biomass burning aerosol mixture.

If the Angström exponent represents the fine mode, how does this necessarily correspond to coarse mode dust detected by the lidar?

Figure 14 in Weinzierl et al. (2012) shows that absorption Angstrom exponents of ~1.3-1.4 typical for biomass burning mixtures during SAMUM-2 corresponded with effective diameters smaller than 1 μm , while absorption Angstrom exponents between 2-5 typical for mixed Saharan dust corresponded with effective diameters larger than 2.5 μm . In the case shown in Figure 10a, the effective diameter in the dust layer below 1.5 km was 2.45 and 2.75 μm respectively. In contrast, the effective diameter in the biomass burning mixture between 1.5 and 4 km was 0.66 and 0.83 μm , respectively.

31. (page 26000, line 24) Besides its role in the retrieval of aerosol profiles from CALIPSO and EarthCare satellite, what other applications require aerosol classification?

We revised the conclusion, and refer now to CALIPSO for the remote sensing applications.

32. (page 26001, line 2) What retrieved quantities are referred to here?

We meant e.g. refractive index, size distribution, and absorption. However, we revised the conclusion and deleted this reference.

33. (page 26001, line 5) Should be three, not two, aerosol intensive properties.

Changed.

34. (page 26001, line 14) Since EarthCare will measure only two intensive properties (lidar ratio and depolarization) and at a different wavelength (355 nm), the authors should comment on how these differences will impact the aerosol classification.

We revised the conclusion to emphasis this point.

35. (page 26001, line 15) What high level inversion methods are referred to here?

We were talking about the inversion of the lidar signals. To avoid confusion, we removed this statement from the conclusion.

36. (Figure 3) Figure 3c shows 7 day back trajectories assigned to fresh African biomass smoke. How is this smoke necessarily “fresh” after 7 days? This may explain some difference between the characteristics between the “fresh” smoke reported here and by Burton et al. (2012). In the case of Burton et al. (2012) the smoke was directly coming from fires seen by the pilots of the aircraft and so was minutes (not days) old.

We changed ‘African biomass burning (fresh)’ to ‘African biomass burning mixture’. See above.

37. (Figure 5) The different aerosol types described here were not classified using the lidar data but by other means. This should be indicated here. Are these distributions normalized in any way? What does each point refer to; a single lidar observation or some kind of average?

We changed the text to describe our strategy for aerosol type identification. The distributions shown in Figure 5 are not normalized. Each point refers to a single lidar observation. We clarified this point in the Figure caption.

38. (Figure 8) This figure is a bit confusing in that it shows paths to specific types as well as mixtures. There should be some explanation of this and indication that this chart does not necessarily represent the paths to specific aerosol types. Also, what comprise the “dust mixtures” shown in this chart? Also, can some kind of confidence estimate be provided for the classifications produced using this chart? For example, given the logic of this chart, the classification of mineral dust would be much more confident than the classification of pollution and aged Canadian biomass burning.

We added a paragraph in Section 4.2 discussing the uncertainty and confidence of the aerosol type classification scheme.

39. (Figure 8) Figure 5 shows considerable overlap in the color ratios of pollution and Canadian biomass burning. Given this, and the similar overlap between the color ratios of smoke and pollution shown in Figure 10 by Burton et al. (2012) and in Figure 1 by Müller et al. (2007), it is doubtful that the color ratio can be used to make a definitive distinction between smoke and pollution.

See above.