Response to Anonymous Referee #2

We would like to thank anonymous referee #2 for the useful comments. We have responded to each specific comment in detail as well as updated the manuscript according to the suggestions, which we believe has helped to strengthen and clarify portions of the study. Comments are in blue and italics, and our responses are in black.

Some questions: Which are the criteria for choosing 1.5 SAE as the limit value for Coated large and EC dominated particles? (Figure 1). Is there any reference in the literature for this? Which are the criteria for choosing 1.5 AAE as the limit value for Dust / OC/Dust / OC dominated and Dust/EC /mix / EC/OC mixt? (Figure 1) Figure 1.

In the supplemental material of Bahadur et al. (2012), a threshold value of AAE=1.5 was found to demarcate the dust dominated region fairly well, containing 72% of all measurements in dust-dominated regions, but only 17% of measurements in fossil fuel dominated regions. Therefore, we use AAE=1.5 to delineate only aerosols that have an enhanced absorption at shorter wavelengths (i.e. dust and OC); and their external mixtures. All smaller values of AAE are then considered to have an influence of EC leading to more complex mixtures. Similarly, SAE of 1.5 was found to reasonably delineate the fine mode aerosols (EC and OC); and their external mixtures. All smaller values of SAE are then considered to have an influence of larger particles (such as dust or coated large particles), again leading into the mixture containing regions of the phase space.

In section 2.1, when presenting the Angström matrix we included the following clarification:

"This partition is based on a simplified division published by Bahadur et al. (2012). In the supplemental material of Bahadur et al. (2012), a threshold value of AAE=1.5 was found to demarcate the dust dominated region fairly well, containing 72% of all measurements in dust-dominated regions, but only 17% of measurements in fossil fuel dominated regions. Therefore, the condition of AAE>1.5 has been retained to delineate the aerosols that have an enhanced absorption at shorter wavelengths (i.e. dust and OC) with smaller values of AAE considered to have an influence of EC leading to more complex mixtures. Similarly, SAE of 1.5 was found to reasonably delineate the fine mode aerosols (EC and OC) with smaller values of SAE considered to have an influence of larger particles (such as dust), again leading into the mixture containing regions of the phase space. In our partition, the inclusion of mixtures changes some of the phase boundaries. Thus, the phase boundaries for large particles and "EC dominated" particles were relaxed to the more intuitive value of 1.0."

The "coated large particles" quadrant in Figure 1 is mainly related to measurements carried out at "dust dominated regions". These regions (listed in Table 2) are mainly located in Northern Africa and Middle East, areas with a high impact of dust storms. Could this association be related to the absorption by free iron present in dust?

The studies of Alfaro et al. (2004) and Fialho et al. (2006) find that were absorption associated primarily with free iron in dust, we would expect an Angstrom exponent in the range of 2-3. Therefore, we are led to the conclusion that the points in Figure 1 (in North Africa and the Middle East) that fall within the "Coated Large Particles" domain are more likely associated with dust that has been polluted with a stronger absorber, such as EC. We expect that the large dust core is responsible for the small SAE value. We do however expect that the free iron in the mixture is responsible for part of the absorption. As the relative contribution of the iron increases, we would see a corresponding increase in the AAE transitioning into the "Dust/EC mixture" and finally "Dust Dominated" regimes. Also as stated in the discussion section, the misclassification of dust in the California case are most likely humic-like substances (HULIS) species formed by fog or cloud processing.

We modified the sentence where the Fig. 1 is presented:

"Figure 1 illustrates the division of the Ångström matrix with labels that represent the three aerosol absorbing species and their mixtures. The lower-left quadrant has been labeled as "coated large particles" indicating that it contains a species with AAE<1 but larger in size. Lack and Cappa (2010) showed that black carbon particles with a sulfate coating might present those optical properties, and polluted dust with strong absorbers might also present the same spectral response."

Figure 1. Biomass burning influenced aerosols are mainly allocated in the finest quadrants (SAE>1.5) whereas "Fossil fuel" related aerosols are concentrated in both (fine and coarse fractions). Is there any explanation for this? Could this be related to the presence of secondary aerosols (mainly scattering) such as sulfate?

One possible explanation for the difference between FF and BB observations is that the so-called fossil fuel dominated areas correspond to large urban agglomerations that tend to have a more polluted and complex mixture of aerosols (for example, while urban areas certainly have large fossil fuel emission sources, they also have dust emissions from asphalt, roads, and construction). This factor results in a much larger spread along the SAE (size) axis for the "FF" regions compared to either the dust or BB regions that tend to be located in more remote locations.

The reviewer is completely correct in pointing out that an influence of mainly scattering aerosols (such as sulfate) could also be responsible for the large spread in SAE. While this study does not focus on measuring purely scattering particles, based on the phase space we have defined we expect the majority of these points to again fall into the “coated large particles” region.

I would acknowledge having some more explanation about the ATOFMS results. Which are the negative spectra for primary fossil fuel and for dust? Does the negative spectrum presented in Figure 4 correspond to Primary fossil fuel? Or does it correspond to Secondary fossil fuel? does it correspond to PFF or to SFF? Actually, ATOFMS results are expressed as sources more than as chemical composition. Is it possible to present what is the contribution of the chemical species to the sources distinguished? What is the mean contribution of EC and OC to the FF and BB sources deduced form the ATOFMS?

All spectra shown in Fig. 4 range from -250 to 250 m/z, i.e. both positive and negative spectra are shown in the same graph. For example, for primary fossil fuel the negative spectra contain mainly carbon clusters, C^-_n (shown on the left), or sulfate and/or nitrate markers, HSO^-_4 and/or NO^-_3 (shown on the right); and for dust the negative spectra presents silicates on the negative spectra, SiO^-_2 and SiO^-_3.

The ATOFMS provides the spectrum of every single particle, not from a mixture. All the particles detected are classified based on the markers shown in their spectra. Based on the experience from previous studies, we know that particles that show a specific pattern in their spectra correspond to specific sources. In this sense, there is no contribution of EC or OC particles in the primary or secondary fossil fuel classification, but rather all the particles in the different sources present a similar chemical signature.

In this study we tried to combine the actual source detected with the ATOFMS with a more general classification that is typically provided when estimating aerosol type with optical properties.

For clarification, we modified the paragraph where the figure containing the spectra is presented:

“Spectra from individual particles, i.e. their chemical signature, are grouped into chemically similar clusters using the ART-2-a algorithm (Song et al, 1999). The initial clusters are then manually grouped in a small set of clusters based on the identification of the mass spectral peaks that correspond to the most probable ions for a given mass-to-charge ratio (m/z) based on previous lab and field studies. These clusters are then classified into an absorbing source: primary fossil fuel, secondary fossil fuel, primary biomass burning, secondary biomass burning and dust, excluding other non-absorbing sources. Figure 3 shows a representative mass spectrum for each aerosol source where the mass-to-charge ratio (m/z) is on the x-axis, and the intensity of the ion peaks is on the y-axis (in arbitrary units)."

Note that with the new reorganization Fig. 4 is now Fig 3.

Page 3465; line 4 delete "the" in "that the those"

The line has been corrected

Caption from Figure 5. The values depicted in Figure 5 and Table 4 did not refer to chemical composition. The results obtained from ATOFMS are expressed as dominant sources, as deduced from chemical composition. Please, replace "chemical composition" by "aerosol sources deduced form ATOFMS results".

The caption has been corrected as suggested.

References

Alfaro, S. C., Lafon, S., Rajot, J. L., Formenti, P., Gaudichet, A. and Maillé, M.: Iron oxides and light absorption by pure desert dust: An experimental study. J. Geophys. Res., 109, D08208. 2004.

Fialho, P., Freitas, M.C., Barata, F., Vieira, B., Hansen, A.D.A. and Honrath, R.E.: The Aethalometer calibration and determination of iron concentration in dust erosols. Journal of Aerosol Science, 37(11), 1497-1506. 2006.

Lack, D. A. and Cappa C. D.: Impact of brown and clear carbon on light absorption enhancement, single scatter albedo and absorption wavelength dependence of black carbon. Atmos. Chem. Phys., 10, 4207-4220, 2010.