

### Response to Anonymous Referee #3

We would like to thank anonymous referee #3 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.

#### *Main Comments*

*1. Figure 1, where the methodology is explained and also obtained results for 33 globally distributed AERONET stations are shown, displays the so-called Angstrom matrix. More specifically, the different aerosol types are discriminated through scatterplot analysis of AAE and SAE, setting specific thresholds to both of them. However, a question arises here as to the choice of these thresholds:*

*(i) how these thresholds were defined/decided? This is not discussed in the text.*

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.”

*(ii) what are the uncertainties/errors in the measurements used and how these can affect the classification?*

While the AERONET AOD measurements (responsible for SAE evaluation) are fairly robust, the AAOD measurements (responsible for AAE evaluation) rely on the SSA retrieval, which can be uncertain. Based on the analysis shown in Bahadur et al. (2012) it was found that between 70-76% of all measurements performed in either the dust or fossil fuel dominated areas fell within the corresponding phase regions. In the more general matrix used in this work however, these points would be classified as mixtures containing dust/EC. The effect of the specific choice of threshold would be to reclassify these 24% (outliers) points into a new mixture category. In fact the comparison with the in-situ chemical measurements presented in this work is our first effort to better constrain and reduce this uncertainty by attempting to compare the rough classification of the Angstrom matrix to actual composition trends.

*As stated in lines 16-17 of page 3454 of the manuscript, these 33 AERONET sites have dominant aerosol species. Nevertheless, it should be taken into account that apart from dominant aerosol types influences by other types are possible as well. For example, it is known that such influences are seasonally dependent. Has any care been taken in order to minimize this influence, for example through screening procedures applied to data and excluding specific seasons?*

No data screening was employed in the development of the Angstrom matrix. Rather a probability distribution of the large volume of data spanning different seasons and sites was utilized to establish the “most likely” SAE and AAE values for Dust, OC and EC independent of location and season.

*2. I find that the organization of the paper can be improved. In its current form, the methodology and data section already gets into the presentation and discussion of obtained results. This can bring the reader into a little confusion, though a more traditional methodology presentation, referring to existing knowledge and clarifying the added new one here and focusing to the specification of decided thresholds (as stated above), would be preferable.*

We reorganized the manuscript with a more classical methodology section. Now, the methodology includes the explanation of the remote sensing and in-situ measurements and there are separated sections for the results, discussion and conclusions.

### *Minor Comments*

1. *Sub-section 2.1, page 3457: it is reported that, and presentation of results is based on that, data are divided by seasons, grouping together winter and spring, and summer and autumn. It should be explained why such a grouping is applied and what are the criteria for this selection. There should be specific reasons related to specific aerosol regimes in the study region, but these are not discussed at all.*

2. *Related to the previous point, probably it is useful to say a few words at the end of sub-section 2.1, on the consistency of the first findings. This will be helpful to the readers who are not familiar with the prevailing aerosol regime in California, and does not need to be detailed but just based on knowledge from existing literature referring to natural and anthropogenic aerosol emission sources and transport processes in the region.*

The limited availability of AERONET data is the reason for grouping the seasons in order to have statistically significant results. The following line has been added to the manuscript:

“Due to the limited availability of Level 2.0 AERONET data, seasons were grouped using winter and spring in one season and summer and autumn in another season.”

3. *Sub-section 2.2, page 3458, second paragraph: “PSAP data were corrected ... “. Please explain what the corrections were applied for.*

We modified the sentence as follow:

“PSAP data were corrected for scattering aerosol and spot size based on Bond et al. (1999) and Ogren (2010) ...”

4. *Sub-section 2.2, page 3459, 2nd paragraph: “Spectra are grouped ... “. A few words about this will be helpful.*

The beginning of the paragraph has been modified:

“Spectra from individual particles, i.e. their chemical signature, are grouped into chemically ...”

5. *Sub-section 2.2, page 3459, lines 16-17: Please explain the units in axes x and y.*

The spectra shown in Fig. 4 represent the mass-to-charge ratio ( $m/z$ ) on the x-axis, where  $m$  is the atomic mass and  $z$  is the number of charges, and the intensity on the y-axis (in arbitrary units). The positive part of the spectrum corresponds to

positive ions ( $z$  is a positive charge) and the negative part correspond to negative ions.

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

“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.

*6. Sub-section 3, page 3463, line 3: replace “... burning sources respect to the” by “... burning sources with respect to the”.*

The line has been corrected.

*7. Sub-section 3, page 3463, lines 12-13: here it is stated that dust is not expected to make large contributions in urban areas. However, this is not true under specific conditions, e.g. areas in proximity of great deserts or undergoing dust transport. Also, it should be taken into account that the results refer to the whole atmospheric columnar aerosol loading, and it is possible to have transported dust aloft. Some reference to this is being made at the end of this paragraph. Nevertheless, for making statements like the one at this part of the paper, such dust transport cases should be excluded. This can be done based on prevailing synoptic conditions or findings in existing literature.*

We should clarify that we have made no effort in this paper to quantify the relative absorption of dust in urban areas, rather we expect the frequency of “events” where significant absorption could be attributed to dust to be small. The reviewer is correct in pointing out that in column-integrated measurements (such as AERONET) it would be impossible to get a truly dust-free measurement. This influence is may be a significant factor in the spread of the FF measurements along the SAE axis. However, we note that these events are classified into either the “EC/OC/Dust Mixture” or “Coated Large Particles” category and excluding them from the analysis is not necessary.

*8. Sub-section 3, page 3465, line 4: replace “... indicating that the those were ...” by “... indicating that those were ...”.*

The line has been corrected.

9. *Sub-section 3, page 3466, line 27: replace “absorption due OC, which ...” by “absorption due to OC, which ...”.*

The line has been corrected.

10. *Table 3: with regards to the 3 aircraft campaigns, outlined in this Table, it should be noted that all three only partially cover the year, e.g. from late winter to early summer. A comment should be made with respect to this, namely on whether this is a problem or not as to the representativeness of regional aerosol regime, in terms of validation of proposed methodology.*

Aircraft campaigns are very limited in time and we are aware of the limitations when comparing with long-trend results. We tried to make this clear in the results section:

“Also each campaign, because of the location and dates, can be associated with a region and season. Taking into account the limitation of such comparison (the campaigns are punctual in time compared to the long-term AERONET data), CalNex corresponds with southern California during the summer, CARES is northern California also during the summer, and CalWater is northern California during the winter.”