

Interactive comment on “Identification of key aerosol populations through their size and composition resolved spectral scattering and absorption” by F. Costabile et al.

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(Itemized Italic text refers to the referee's comments, while authors reply (AR) indicates the answer to referee's comment. Each AR is followed by the corresponding new text of the revised manuscript, labeled by NTRM. Here, quoted text refers to the old version of the manuscript, while bold quoted text is new text added to the revised manuscript.)

We do thank the referee # 1 for the valuable review, which we have fully addressed as detailed below.

- This is an interesting paper and should be published. It's a bit difficult to read*

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due to the large number of abbreviations and acronyms. However the figures do show a separation of the data into different aerosol types which could be useful. The authors use a new parameter of dSSA times AAE which has the effect of differentiating between increasing SSA (dust) and decreasing SSA (BC). It would be useful if they would also plot the data as AAE and EAE following to show the difference between the two different ways of analyzing the data.

AR: To show relevant differences between our paradigm and the versions suggested, we have extended the former fig.1 and added a supplementary figure (Fig.S1).

The new figure 1 now includes EAE and SAE in the y axis, and AAE, dSSA and dSSA·AAE in the x axis based on experimental data. The other figure (Fig.S1), added as supplementary material shows the corresponding numerical simulations. With respect to EAE vs AAE used by previous studies (e.g. Fig.5 of Russel et al. (2010)), we believe both new figures show the advantages provided by the SAE vs dSSA·AAE plot: a comprehensive data separation, which fits in previous classifications based on EAE vs AAE relation, but put the observations into a more general framework. Reasons for this clearer separation are discussed in the revised manuscript, including the (strong) inverse relation between SAE and dSSA based on their common dependence on particle size, the (weak) inverse relation between AAE and dSSA based on their common dependence on particle composition, and the fact that separating SAE from AAE (instead of combining them in EAE) helps in separating particle size from particle composition effects.

Lastly, to improve the readability we have made clearer some acronyms (Tab.2 of the manuscript): the former AM (Aitken mode) has been substituted by AKM, the former SM (soot mode) by STM, the former MM (Marine mode) by CMM (coarse marine mode), and the former DM (dust mode) by CDM (coarse dust mode).

NTRM: Text added at the very beginning of the Section 3 (sect.Results):

“An aerosol population can be considered as a set of particles being “homogeneous” with respect to their sizes and composition (and shape and mixing state). The variation of this “homogeneity” translates into a different spectral variation of aerosol scattering and absorption, SAE (or EAE) and AAE varying with varying particle size and composition. In this work, our aim is to find proper combination of the intensive parameters SAE, AAE, SSA and dSSA by which key aerosol populations can be optically separated. Similar classifications have been proposed before (cf. Introduction section). Some of those classifications (e.g., Russell et al (2010)) were based on AAE vs EAE plots. The advantage of using EAE is that it takes contemporarily into account SAE and AAE; but this is also a disadvantage as EAE fails in separating particle size effects (mainly linked to SAE) from particle composition effects (mainly linked to AAE). To make this separation more evident (i.e., to separate particle size from particle composition effects on the spectral variables) SAE was used instead of EAE.

The SAE vs AAE relation was first experimentally analysed (Sect.3.1). Since SAE represents the scattering spectral variation, and AAE represents the absorption spectral variation, one might expect their combination to correlate to the SSA spectral variation (SSA being a mathematical combination of scattering and absorption). To investigate the way SAE and AAE shall correlate to dSSA (the SSA spectral variation) we combined the relevant experimental data, and found significant inverse correlation between SAE and dSSA, and between SAE and dSSA·AAE (Fig.1). Those behaviors observed on experimental data were reproduced by numerical simulations (Sect.3.2), thus providing a theoretical support to the observations. A classification scheme (hereafter referred to as “paradigm”) resulted. The relevance of the paradigm is discussed in the Discussion section (sect.4.1) based on both experimental results (Fig.1) and numerical simulations (Fig.3).”

Text added in the Experimental results section (Sect.3.1, block 17510, lines 9-13):

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“Figure 1 shows the observed data (more than 15500 data points) as either EAE (y-axis, upper panels) or SAE (y-axis, lower panels) versus either AAE, dSSA, and dSSA·AAE(x axis). As previously discussed, SAE vs dSSA·AAE data ($R^2=-0.66$) correlate more than SAE vs AAE data ($R^2=-0.07$). As well, SAE vs dSSA·AAE (lower panels, $R^2=-0.66$) correlate better than EAE vs dSSA·AAE (upper panels, $R^2=-0.58$). Data are color coded by SSA530...”

Text added in the Discussion section (Sect.4.1, block 17514, line 13):

“This is shown in Fig.4 where SAE vs dSSA·AAE relation is separately plot for all identified aerosol population. Figure S1 is same as Fig.4 but for the fact that it also shows relations between SAE (y-axis) and either AAE or dSSA (x axis).”

Text added in the Discussion section (Sect.4.3, Block 17519, lines 26-27):

“The paradigm in Fig. 4 fits in previous works. By way of illustration, it might be useful to compare Fig.4 (and Fig.1 and S1) to Fig.5 of Russel et al.(2010). The comparison reveals that both ways of analyzing data reach very similar conclusions and allow to optically classify:

- the “urban/industrial” aerosol type cluster (AAE=0.8-1.5, EAE=1.5-1.8) of Russel et al.(2010), here represented by the AKM aerosol population (AAE ~ 2 , SAE=1-2.5);
- the “biomass burning” aerosol type cluster (AAE=1-1.7, EAE=1.8-2) of Russel et al.(2010), here represented by the BBM aerosol population (AAE < 2 , SAE=1-3);
- the “desert dust” aerosol type cluster (AAE=1.5-2.5, EAE=0.2-1) of Russel et al.(2010), here represented by the CDM aerosol population (AAE ~ 2 , SAE < 0.5).

As well, the comparison shows that the paradigm in Fig.4 has the effect of putting

the aerosol observations into a more general framework.”

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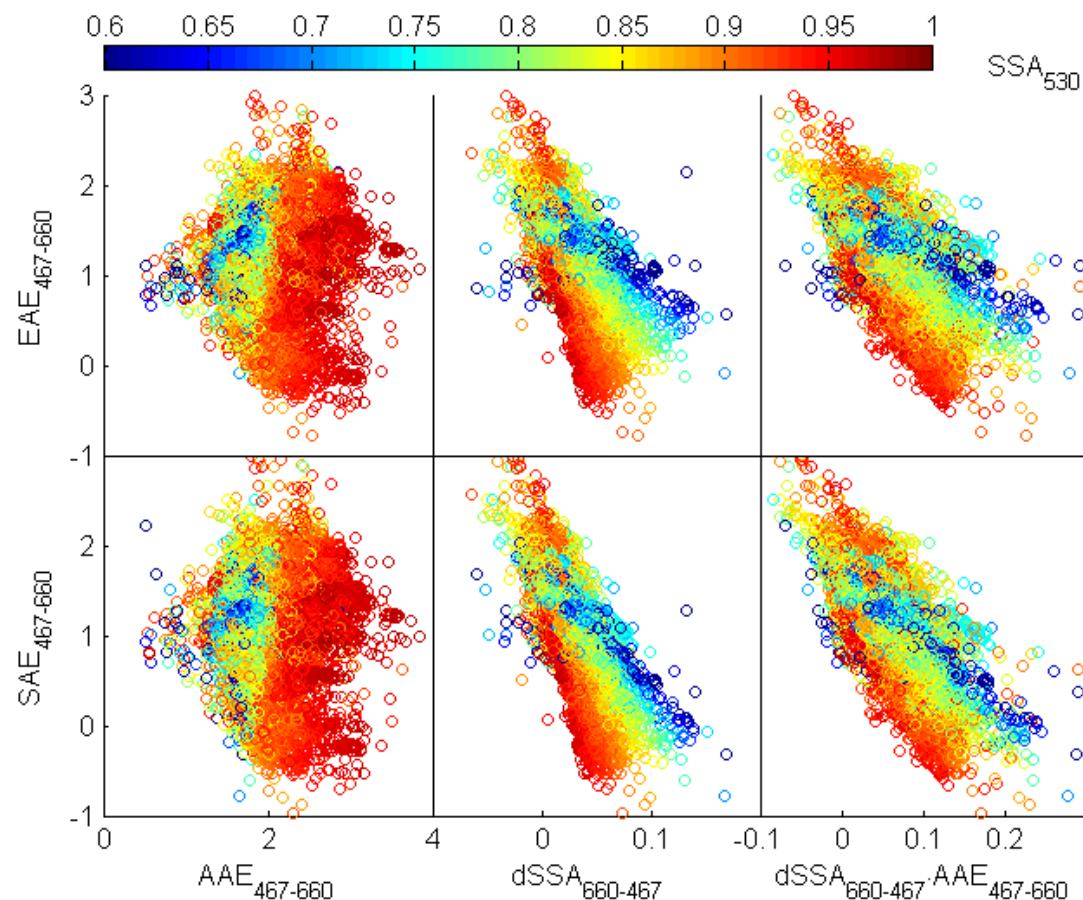
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Fig. 1. New Figure 1: “Inverse linear relation observed between EAE and SAE (y-axis) and AAE, dSSA, and dSSA.AAE (x-axis). The slope varies with varying SSA (color code)”.

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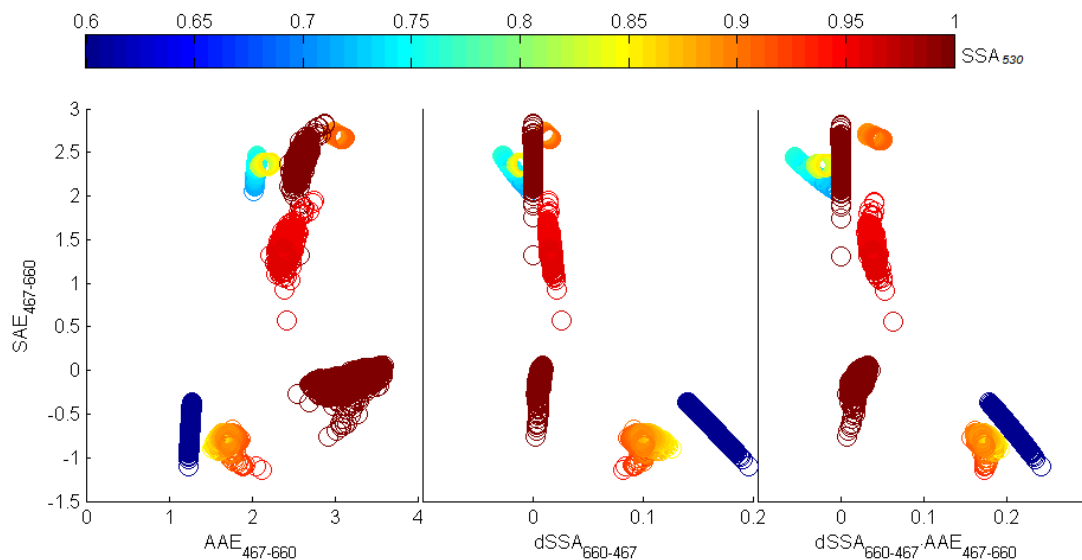
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Fig. 2. Supplementary Figure S1: “Results of the numerical simulations illustrating the “paradigm” proposed. The figure is same as the lower panels of Fig.1, but shows relevant numerical simulations”

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