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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Classifying aerosol type using in situ surface spectral aerosol optical properties" *by* Lauren Schmeisser et al.

Anonymous Referee #1

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GENERAL COMMENTS

The paper addresses the important topic of aerosol type classification through aerosol spectral optical properties. Authors explore how existing schemes reported in the literature may be applied and possibly improved. The paper demonstrates that there still are several uncertainties in all classification schemes proposed here, and discusses major ambiguities and limitations . No significant improvement is, however, proposed.

The topic is fully within the scope of ACP, and I recommend this paper to be published in ACP after the following issues are addressed.

SPECIFIC COMMENTS

1. *The meaning of AAE as indicator of aerosol properties still contains uncertainties that have not been formally assessed in the scientific community*:

(i) AAE=1 as indicator of BC.

Although AAE =1 has often been attributed to BC, several studies have demonstrated that AAE depends on both aerosol composition (and in particular, to the BC-to-OA ratio more than to the BC mass concentration alone), and size (e.g., Lack and Langridge, 2013; Saleh et al., 2014; Costabile et al., 2017). Caution should therefore be taken in interpreting AAE=1 as indicator of BC only. As an example, if AAE=1 indicates BC only, cluster 1 will indicate BC (pag.9 lines 35-38); however, authors ackowledge that "the generally low scattering coefficients at these sites indicate the presence of anthropogenic aerosols is infrequent" (pag.10, lines 6-8) .

(ii) AAE>2 as indicator of brown carbon.

Although several studies have indicated that AAE values from 2 to 6 may indicate brown carbon (e.g., Andreae and Gelencsér, 2006; Moosmüller et al., 2011), authors do not consider brown carbon at all in this paper. In fact, the contribution of BrC aerosol with large AAE values (2-6) may be less significant at the regional remote/rural sites analysed here, as discussed at pag.18, line 17. Therefore, the paper should mention that the proposed analysis may fail at urban sites and more polluted sites. Indeed, authors ackowledge (pag.8, line 28) that Cazorla et al.'s scheme may fail for sites in proximity to aerosol sources.

(iii) AAE of carbonaceous aerosols.

The paper mentions that carbonaceous aerosols have AAE $\sim$ 1.2 (e.g., pag.10, line 10). However, carbonaceous aerosols include both BC, OA and BrC, and their relative contribution in particle with different size can cause a larger variability in AAE values.

2. *Overall presentation structure*.

Results section (Sect.5) is too long and a little bit confusing. In particular:

- cluster analysis results (pag. 9,10,11) are somehow ambiguous, as authors ackowledge (pag.11, lines 20-24);

- description of multivariate cluster analysis (pag.9, lines 1-25) should be moved to a separate paragraph (e.g., in a data analysis section).

Discussion section (Sect.6) is the most interesting part of the paper, but it should be better clarified in some parts. In particular, I recommend to address:

(i) Uncertainties in the AAE attribution method mentioned above;

(ii) Differences between columnar and in situ surface spectral optical properties.

Authors apply thresholds from Cazorla et al.'s scheme - obtained by columnar measurements (AERONET) - to interpret in situ surface data. Although results may be consistent, in principle this is not completely correct. In fact, aerosol spectral optical properties measured by columnar and in-situ ground instruments may in principle differ significantly. Relevant thresholds seem, however, to be consistent in Tab.1. Please, add a more comprehensive discussion (this is only sketched in the discussion section, pag. 18, lines 1-10).

(iii) Classification of measurement site.

Authors demonstrate that none of the classification schemes applied here can classify measurement sites. None of the sites has indeed only a single dominant aerosol type (pag.12, lines 6-8). Only continental polluted sites are well classified. This is a reasonable conclusion, as these classification schemes should classify aerosol type, not measurement site. Please, discuss this point more clearly (this is roughly discussed in Sect.6).

3. *Sampling line description is not clear enough*:

- Are all sites but SUM equipped with PM_{10} sampling heads?

- Heated inlets might cause losses in organic aerosol and volatile compounds. This can influence aerosol spectral optical properties. Is heating performed at all sites?

Please, add more details.

TECHNICAL CORRECTIONS

- Pag.8, line 2: is

- Tab.4 : check log (σ)

- Pag.12, line 20: "is"

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