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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol light absorption and the role of extremely low volatility organic compounds" *by* Antonios Tasoglou et al.

Anonymous Referee #2

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Review of Aerosol light absorption and the role of extremely low 1 volatility organic 2 compounds, by Tasoglou et al., for ACPD, 2020

Overview: The paper is focused on presenting observations from a ∼3 week measurement campaign at a Cretan site with little direct local influence. Measurements of rBC, aerosol light absorption, gas phase tracers, and bulk aerosol composition were complied to answer a primary focus topic: connecting observed aerosol absorption and potential BrC influences via the proxy of low volatility compounds.

Publication of this kind of work falls within the aims of ACP, and contributes to the broad understanding of aerosol absorption.

The manuscript does not yet make a strong enough case for its conclusions. The major

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reasons for this are 1) the lack of uncertainty analysis 2) un-discussed conflict between conclusions and observations, and 3) lack of details provided about interpretation and work such that the reader is not sure what was done and what it means. In my opinion, the paper is a good start towards a publication ready manuscript, but more analysis work and explanation is still required to make a solid contribution to establishing distilled conclusions from the observations.

General Comments:

1) Uncertainty: throughout the manuscript consideration and discussion of uncertainty should be expanded. Comparison of MAC from theory and measurement depends critically on the uncertainties of the various approaches. To the extent that there is validating information about the PMF, this could also be considered in terms of uncertainty in drawing associations between bulk composition (which is all that the PMF is indicating) and the BC sources/microphysics (which also can affect MAC).

2) Some issues have not been sufficiently addressed. They include: a) how is the observation of AAE of $\sim$1 consistent with expectations for significant BrC absorption? In fact, can any significant absorption be attributed to low volatility spiecies? b) how is absorption of internal mixtures with rBC separated from bulk phase absorption? c) How is the PMF validated? Did rBC concentration (not deviation in MAC) correlate or anti-correlate with either factor? d) How are coating thicknesses constrained for rBC? e) How high is confidence, based on measurement and theoretical uncertainties, that the range of MAC is not solely associated with 1) rBC and PAS uncertainties or 2) lensing enhancements?

3) The reader needs more information about what was done in order to understand the results: a) How did the PAS and Aethalometer compare? Which was used to generate MAC405? How significant were the differences between them? b) SP2 - laser intensity? size range of detection for rBC? Correction to account for total rBC accumulation mode concentrations? Type (8 channel?). Precision of cal with CPMA?

Total uncertainty estimate in concentration (noting that different BC does have different response in SP2)? Calculation of coating thicknesses? c) Sampling: was the aerosol dried? What temperature was the laboratory? If it was not dried, the whole analysis is likely questionable. How large were the corrections for diffusion and thermophoresis losses in the denuder (I expect likely negligible for Accum. mass). d) Mie theory tests: what constraints on coating thickness were used in the Mie Theory evaluation of index of refraction? Was this derived from SP2 data? How? This is a critical question if there is to be any conclusion drawn from this analysis. Note that SP2 determination of coating thicknesses are highly uncertain for thin coatings (e.g. Ohata et al.,(2016), Hygroscopicity of materials internally mixed with black carbon measured in Tokyo, J. Geophys. Res. Atmos., 121, doi:10.1002/2015JD024153.).

Line-level and specific comments L37 - averaged over a population. The literature review 34-83 - lensing enhancement is well written, but might give the reader the incorrect idea that similar work was done here. As only the AMS and SMPS were behind denuder this is not the case.

(Note that it is a nice feature of your data set that initial brown carbon bleaching/evap should have already substantially occurred)

TD - please provide more info about the temperature set points so that the reader isn't surprised by the lack of data between 200 and $\sim$375 °C.

L197 Bond and Bergstrom, 2006 suggest (1.95,-0.79) for the complex index at 1.8 g/cc. Not as stated.

The paper by China et al., 10.1002/2014GL062404 will be useful for justifying use of Mie theory shell and core.

Please consider if Figure 2 is unnecessary given figure 4Âă? Combine?

Wavelength of Mie theory calcs? Note that RI for BC is likely low. Were the calculations carried out for observed core size dist? coating thickness? On what time scale were

those calculated?

Figure 7: negative Measure-Predicted not included - meaningless without all the data. It appears that some positive M-P data is also missing (based on the number of data points in figure 7 vs those in figure 6a).

L249: “average calculated saturation concentration of 0.016 ug/m3”

Time series not very helpful for understanding more than trends in extensive properties (amounts). Scatter plots would be more useful.

L264: “similarly” - actually Babs and Bscat appear poorly correlated.

L273 - please include the uncertainty on this value. L286: Not clear how coating thicknesses were constrained. Big issue./ L281: AAE only described for 523 to 405... how the range calculated, averaged?

L289: is 13% meaningful in the context of uncertainties? L313 - I think you mean “no periods of enhanced BB influence”

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