Comparison of the LEO and CPMA-SP2 techniques for black-carbon mixing-state measurements (Response to Reviewers)

December 21, 2023

Dear Reviewer,

Thank you for providing us with your valuable feedback on our manuscript titled ***Comparison of the LEO and CPMA-SP2 techniques for black-carbon mixing-state measurements***, submitted to Atmospheric Measurement Techniques. We sincerely appreciate the time and effort you have dedicated to reviewing our work, and we are grateful for the opportunity to incorporate your constructive feedback into our revisions. After carefully considering all of your comments, we have made revisions throughout the manuscript to address them. To clearly indicate our responses to the reviewer's comments, we have highlighted them in red text text, while changes made to the manuscript are highlighted in blue text. We believe that these revisions significantly improve the quality and clarity of our manuscript. We are thankful for your valuable input, which has helped us enhance this work. We hope that our responses and revisions adequately address your concerns, and we eagerly await your positive consideration of our revised manuscript.

Comment: Naseri et al. compare several techniques to derive the mixing state from black carbon measurements obtained by the single particle soot photometer (SP2). Their results show that coupling the SP2 with a centrifugal particle mass analyzer provides the best results. Overall the paper is well written and clearly explains the origin and application of the different methods. This manuscript presents a valuable addition to groups that seek to experimentally determine the mixing state of black carbon using similar techniques. I therefore recommend publication.

Minor Comments:

1) "normalized derivative of the scattering signal is used in the ND approach to obtain analogous information" -> Please explain what the normalized derivative is here.
   Authors Response: We acknowledge the reviewer's comment. In response to this comment, we added a brief description of the ND method to the beginning of section 3.1.2 :
   The normalized derivative (ND) approach, as introduced by Moteki and Kondo (2008), offers a methodology for assessing the time-dependent solid angle scattering cross-section ($\Delta C_{\mathrm{sca}}(t)$) to identify individual particles traversing through a Gaussian SP2 laser beam. This technique hinges on the concept that the normalized derivative of the scattering signal (S'/S), as detected by the SP2, can be broken down into the normalized derivatives of the incident irradiance (I'/I) and the scattering cross-section ($\Delta C'_{\mathrm{sca}}/\Delta C_{\mathrm{sca}}$). In the realm of evaporative particles, the equality $S'/S = I'/I$ holds true until evaporation initiates at a specific point within the laser beam. The incident irradiance $I(t)$ for individual particles is deduced from I'/I, extracted from S'/S. Consequently, $\Delta C_{\mathrm{sca}}(t)$ is derived from $I(t)$ and $S(t)$.The ND approach to evaluating rBC mixing states is similar to the LEO approach in that it extrapolates the initial particle size from the first portion of the scattering signal, except a different methodology is used to estimate the undisturbed particle diameter.

2) "The upper coating thickness limit of LEO (line ii), was $\sim$ 285 nm in our study, due to saturation of the scattering detector"-> Was it not possible to utilize the low gain scattering detector? This should extend this upper limit further.
   Authors Response: We appreciate the reviewer's thoughtful comment. We would like to clarify that we utilized a low-gain channel for our analysis. We realize that our phrasing "due to saturation of the scattering detector" might have been unclear. Therefore, we have modified the sentence to enhance clarity as follows:

The upper coating thickness limit of LEO (line *ii*) was $\sim$ 285 nm in our study, due to saturation of the LSD before there is sufficient data below the 3% threshold of the maximum laser intensity to fit the leading edge

3) "morphological assumptions" -> please rephrase to "assumption regarding the morphology" or something like that.
Authors Response: Noted and revised the manuscript as:

To make a general comparison between the LEO results and those derived directly by the CPMA–SP2 measurements without making any assumptions regarding and density and morphology (Figure 2*b*),...

We believe that these revisions have improved the clarity of the manuscript. We hope that our responses and revisions adequately address your concerns and that you find the revised manuscript acceptable for publication in Journal of Atmospheric Measurement Techniques.

Thank you once again for your valuable feedback, which has helped us enhance the quality of our work. We look forward to your positive consideration of our revised manuscript.