

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

December 21, 2023

**Dear Reviewers,**

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**, 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.

## **Reviewer 1 comments**

**Comment:** The manuscript gave the comparison results of the measured black carbon mixing states from LEO and CPMA-SP2 techniques. The articles fall well into the scope of AMT. In general, the results are interesting and important to the related researchers. The manuscript can be accepted after some major revisions.

### **Major Comments:**

- 1) The LEO detection limits results and the corresponding impacts of measured black carbon mixing states were mentioned before by Zhao, Shen et al. (2020). The new findings of this manuscript relative to that article should be detailed.

**Authors Response:** We acknowledge the reviewer's observation and appreciate the opportunity to clarify the relationship between our study and the work conducted by Zhao et al. (2020). In response to this comment, we have revised our manuscript to explicitly detail the Zhao et al.'s (2020) finding and compare it with the present study. The comparison between our findings and those presented by Zhao et al. (2020) is now elucidated in the revised version of our manuscript. Specifically, please refer to page 2, where the relevant content can be found in blue color between lines 55 and 60.

Second, the range of response of the SP2 detectors does not span all relevant scenarios. For example, very small (but significant in mass concentration) rBC particles do not scatter enough light to be detected, so the otherwise broad range of response of the SP2 incandescence detector cannot be fully exploited (Taylor et al., 2015; Zhao et al., 2020).

- 2) The Gassion fit of LEO fits of the LSD signals may still be useful even though the signals detected by the SP2 are saturated. If these particles are considered, the difference between the LEO and CPMA-SP2 methods would not be so different as shown in Figure 4.

**Authors Response:** We appreciate the reviewer's thoughtful comment. We would like to clarify that we indeed included all possible LSD signals in our analysis, We only excluded particles with scattering signals that saturated almost instantly, preventing the determination of the 3% threshold of the maximum laser intensity. In section 3.1 of the paper, we stated this as, "We defined the leading edge of the scattering signal as 3% of the maximum laser intensity based on scatter plots of LEO

and standard analysis for non-absorbing particles.”; therefore, we excluded signals which saturated at 3% of peak laser power, not signals which saturated at the peak laser power. To make this point clearer in the manuscript, bullet point 2 in section 4.2 (page 13), where the relevant content can be found is modified as:

Very large rBC-containing particles, with very large scattering signals (above Line *ii* in Figure 2), saturate the LSD even before the 3% threshold of the maximum laser intensity is reached.

- 3) Around line 360, iff the BC core were not compacted, ie the BC core is filled with some air, then the effective density of the BC core may be reconsidered as shown in (Zhang, Zhang et al. 2016, Zhang, Su et al. 2018). This effects should also be dissucssed.

**Authors Response:** We acknowledge the reviewer’s comment. Indeed, the detailed work of Zhang and colleagues has caught our attention. However, we avoided a detailed discussion of it because of the complexity of understanding the mobility diameters in relation to particle mass. Nevertheless, it should indeed have been cited. We now added the following statement to page 18 between lines 392 and 394.

Our conclusions here are also consistent with work relating soot-core morphology to coating thickness based on mobility-diameter based effective densities, which exploit particle mobility diameters to estimate particle volume in relation to SP2 scattering measurements Zhang et al. (2016, 2018b).

**minor comments:**

- 4) Line 209, it should be mentioned that some lag time of the detailed information can be found in Figure 8 or some following discussion

**Authors Response:** Noted and the below sentence is added to the end of the section 3.1 .

The detailed information pertaining to lag time can be found in Section 4.4, illustrated in Figure 8.

## Reviewer 2 comments

**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 ( $\Delta C_{\text{sca}}(t)$ ) 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 ( $\Delta C'_{\text{sca}}/\Delta C_{\text{sca}}$ ). 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,  $\Delta C_{\text{sca}}(t)$  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 2b),...

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.