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

March 12, 2024

**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 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.

Response to Reviewers 2

**Comment:** There are still quite a few concerns of mine for this study.

**Authors Response:** We thank the reviewer for taking the time to do a second careful review, which raised several opportunities to further improve our manuscript. We trust that the reviewer will agree that we have done so.

**Comment:** The conclusion about the SP2-only method is not able to measure some smaller particles, which depends on a proper retrieval technique and needs more detailed discussions.

**Authors Response:** We have given a much more detailed description of our SP2-LEO analysis than is standard, and have cited this more extensively than in a typical SP2 paper. We have cited three different studies (Taylor et al., 2015; Zhao et al., 2020; Pileci et al., 2021) which are focussed on the detection limits of the SP2 and its implications, and one additional study which went into considerable detail on the topic (Dahlkötter et al., 2014). A repeated consideration of the detection limits of the SP2 would not be a novel contribution to the literature.

**Comment:** In addition, smoke may be the worst case for the comparison between both methods, because the particle is large and heavily coated.

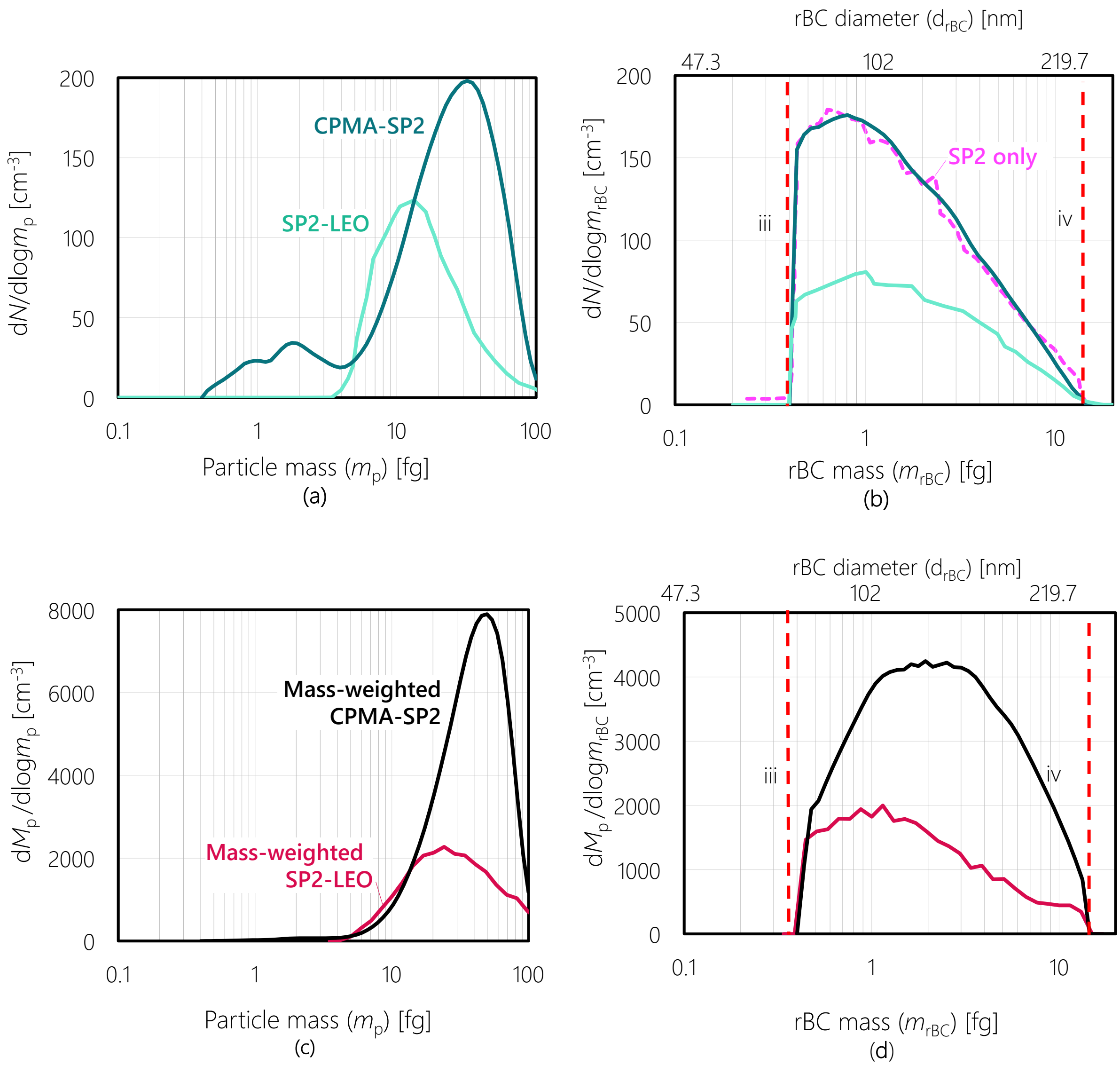
**Authors Response:** We agree that large, heavily coated smoke particles (Case III) may not represent all atmospheric conditions. However, our Case I included small, thinly coated urban particles from a highway. So we believe we have covered a representative range of atmospheric conditions. Further studies will, of course, be valuable.

**Comment:** My main concern is about the results in Fig. 4, as the rBC measurement should look similar between both methods. The measurement efficiency of rBC by the SP2 has been well established, which should be nearly unit above 0.8fg. If this is the case, that means something wrong with the SP2 operation. If you have only shown the BCc with successful LEO retrieval (the retrieval fraction could depend on many things which is rather difficult to be discussed), which should be applied with a retrieval fraction rather than reporting only the uncorrected one.

**Authors Response:** There seems to be a misunderstanding here. Figure 4 does not show the SP2 measurement efficiency of rBC. The reviewer is probably looking at the distance from the dark blue line to the light blue line in Fig 4b – this is not an SP2 counting efficiency, but a LEO measurement efficiency. The reason that LEO did not succeed for many rBC-containing particles is that they were either too small or too thickly coated, i.e., they lay outside of lines (i) and (ii) in Figure 2b. We have modified Figure 4 and its caption to clarify this as follows:

- Fig 4a shows the distribution of rBC particles as a function of total mass (not rBC mass-equivalent) according to the CPMA setpoint (CPMA-SP2) or light scattering (SP2-LEO).
- Fig 4b shows the distribution of rBC mass which the reviewer was referring to. Here, we show that the SP2 incandescence only (pink line) and CPMA-SP2 data agree. We also show that the SP2-LEO method was only successful on a fraction of particles.
- Fig 4c and 4d show Fig 4a and 4b weighted by mass.

We take responsibility for this miscommunication and have revised Fig. 4 to clarify these points.




We have also rewritten the Figure 4 caption:

Comparison of rBC–containing particle number and mass concentrations in Case II as functions of total particle mass (a and c) as functions of rBC-core mass (b and d). The figures shows data from both the LEO analysis and the CPMA-SP2 method. The detection-limit lines (iii) and (iv) from Figure 2 are reproduced in (b) and (d) here, to emphasize why the LEO results differ from the CPMA-SP2 results. The detection-limit line (i) from Figure 2 is the reason that the LEO data here drop to zero for small particles, relative to the CPMA-SP2 data. The pink dashed line in (b) illustrates the consistency between rBC distributions measured by CPMA-SP2 and SP2-only (standard measurement).

**Comment:** I am also a bit worried about Fig. 2 a and c, the LEO retrieval has been very different with CPMA-SP2 results. For example, how the SP2-only retrieval is so far from the lower limit dashline i, shouldn't it be close to the limit, otherwise it can't be deemed as a limit. There may be something wrong with the LEO fit. It would be very useful to check out if the coating/rBC ratio in bulk is consistent between both methods. This is important because even if SP2 is unable to detect them all, it can at least give the bulk information.

**Authors Response:** The Reviewer is correct that we could not have used the data in Fig 2a to define the lower limit (i). We did not. Instead, we calculated these lines from the light-scattering detector (LSD) and broadband incandescence detector (BID) limits of detection, as discussed on Page 13. To better clarify the meaning of these important lines, we have expanded our discussion on Page 13 to explicitly state the definitions of Lines i, ii, iii, and iv in words. We note that we took the concept for these Lines from Dahlkötter et al. (2014), which was cited in the text but not on Page 13. We have now added that missing citation:

Figure 2a and c demonstrate the relationships between rBC diameter and coating thickness derived from the CPMA–SP2 and the LEO analysis, respectively. The detection limits of the LEO method, inspired by the concept introduced in Dahlkötter et al. (2014), are indicated by red lines in Figure 2, and are defined as follows.

The reviewer's suggestion to investigate the coating/rBC ratio "in bulk between both methods" is an important one. This is the purpose of our Figure 4, which shows the overall size distributions for total particle mass (Fig 4a, 4c) and for rBC only (Fig 4b, 4d) from the two methods.

We note that even for the CPMA-SP2 method, a fraction of the smallest and largest rBC particles is not quantified; this has been discussed in detail by Pileci et al. (2021).

Thanks to the reviewer's comments, we added the above sentence to our discussion surrounding Figure 4d.

**Comment:** It looks CPMA-SP2 has a cut off at larger particle mass, which could be a limitation for such method but advantage for SP2-only. This should be discussed.

**Authors Response:** The CPMA is able to transmit larger particles than our SP2 was able to measure. This is an SP2 limitation (line (iv) in Figure 2b) and not a CPMA limitation. We therefore discussed this implicitly as appropriate. Nevertheless, the Reviewer's point is important, and was addressed by the new citation to Pileci et al. (2021) above.

**Comment:** I can't see the point to show Fig. 3 as they all look similar to Fig. 2. This should be at least mentioned in the figure caption. How much uncertainty when deriving from the uncoated and coated BC mass distribution from the CPMA-SP2, considering it is a 2D inversion to get this information. This should be discussed in addition to the disadvantage of the SP2-only method.

**Authors Response:** The reviewer is correct that we duplicated Fig 2b and 2d in Fig 3b and 3e. This was to show the full range of our data set. However, we reconsidered following this comment.

Indeed, we can omit Fig 3b and 3e (Case II) since the other panels in Fig 3 show the extreme minimum (Case I) and maximum (Case III) of wildfire smoke influence. We revised the figure.
We also revised the caption of Fig. 3 to be clearer and briefer.

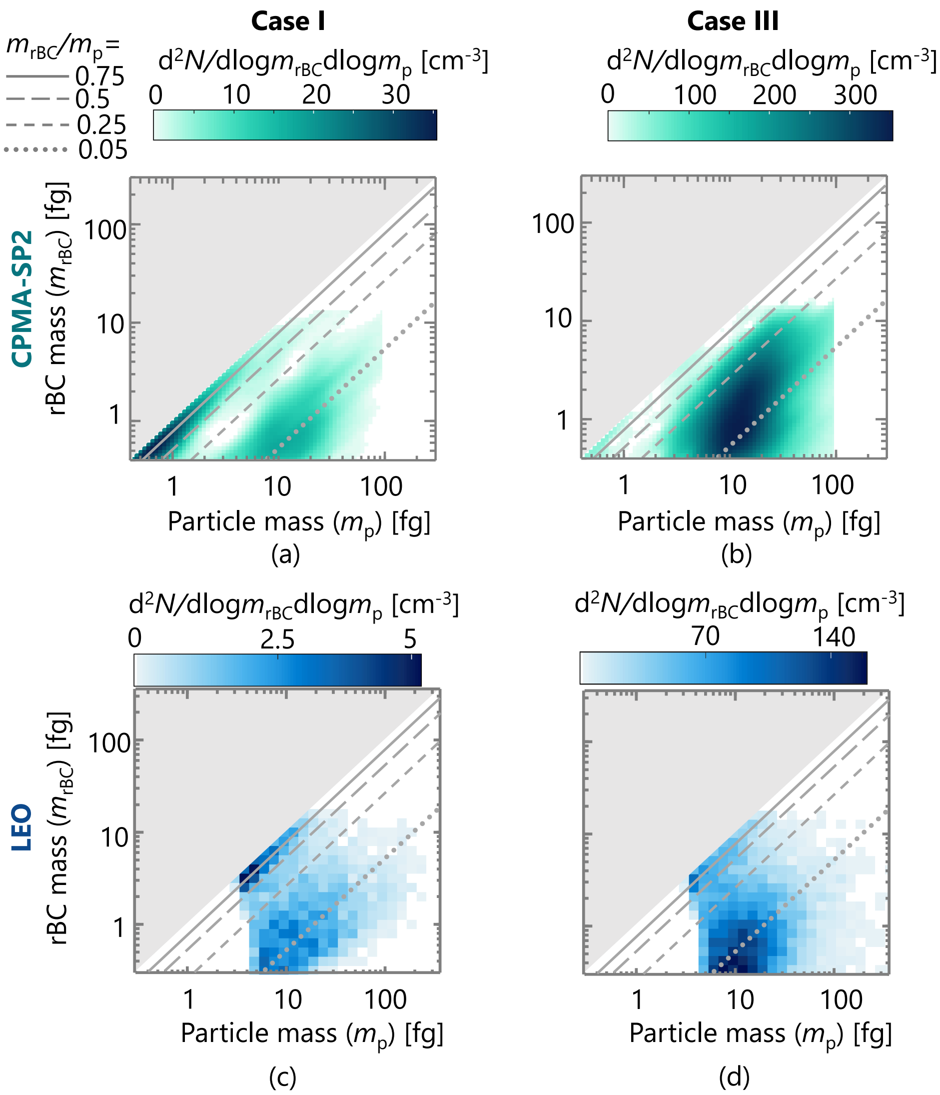
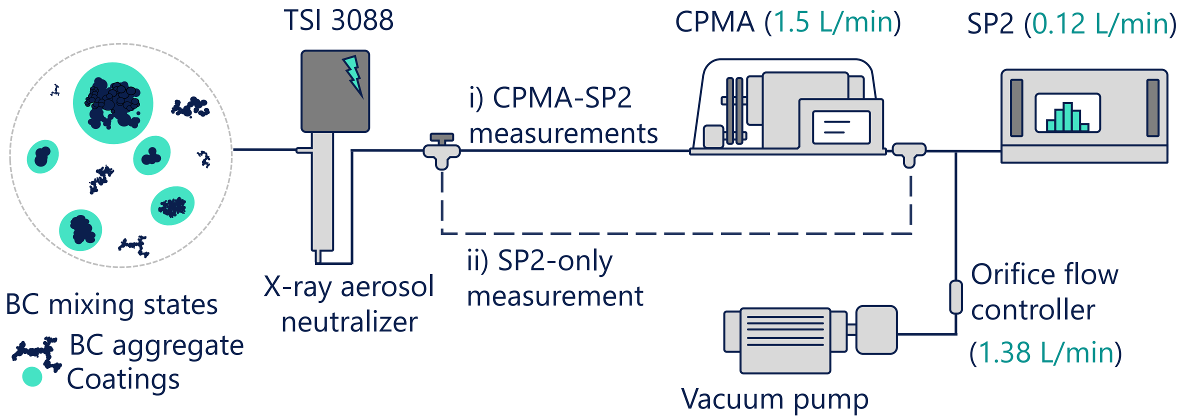



Figure 1: Distributions of number concentration for Cases I (clear visibility, thinnest rBC coatings, urban influence) and III (poorest visibility, thickest rBC coatings, wildfire influence), similar to Figure 2b and Figure 2d (Cases II). Case I represents the smallest influence of wildfires observed in our study; Case III represents the maximum. Solid diagonal lines indicate pure rBC particles with an rBC mass fraction of unity ($m_{\mathrm{rBC}}/m_{\mathrm{p}} = 1$), while parallel lines represent decreasing mass fractions of 0.75, 0.5, and 0.25, respectively.

**Other changes to the manuscript:** We revised some of the text and figure captions for clarity. We revised Figure 1 to show collapsed soot inside the mixed particles, according to recent work (Corbin, Modini, and Gysel-Beer, Aerosol Sci Technol., 2023, link to paper).




We are confident that these revisions further enhance the clarity and quality of our manuscript. We trust that our responses and modifications effectively address the issues raised, rendering the revised manuscript suitable for publication in the Journal of Atmospheric Measurement Techniques.

Thank you once again for your time and attention to our study.

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.