Author reply to review by anonymous referee #3:

We would like to thank referee #3 for his useful comments, which helped clarifying and improving this paper. The review comments are repeated in black font and the author replies are highlighted in blue font (*"italic style is used for modifications to the manuscript"*).

In this publication, the authors describe their finding that the SP2 fails to detect spark‐generated soot. The authors explain their finding by the morphology of the PALAS soot particles which behave like "the sum of very small individual primary particles" in the SP2's laser. This is an interesting result worth publishing. The experiments are well described and the paper is well written.

After addressing the following minor points, I recommend this paper for publication:

General comments:

I personally would recommend not to use so many acronyms (e.g. LDL, LII etc.) because those acronyms make it difficult to read the text.
LDL and LII are now avoided, while the acronyms BC, SP2, PALAS etc. are retained.

Are aged PALAS‐soot particles detectable with the SP2? It would be interesting to perform additional lab tests in order to proof the explanation suggested by the authors. However, I am not sure whether such tests are beyond the scope of this study. During such additional tests, the authors could send spark‐generated soot particles through a thermal denuder or through high relative humidity in order to collapse the fractal structure of the particles. Is the SP2 after this treatment able to detect the PALAS soot particles?
We would indeed expect that collapsing the PALAS soot, achieved by e.g. exposure of the PALAS soot sample to high RH before measurement with the SP2, would make it detectable by the SP2, or at least decrease the threshold laser power required for proper detection. Unfortunately we were not able to add such follow-up experiments to the original schedule of the BC-Act measurement campaign.

Do the authors have SEM/TEM images of the spark‐generated soot which is not detected by the SP2? If so, it would be helpful to show those images in this publication.
Such pictures are not available. However, previous literature (see references in the manuscript) consistently reports very small primary particle sizes and a low fractal dimension for PALAS soot.

Specific comments:

p. 4908, l. 15: For which reason did the authors use two bipolar chargers?
This is now explained:
"The sample taken from the vessel was further diluted and neutralized using ^{210}Po bipolar chargers (two chargers were applied in series in order to ensure charge equilibration at the relatively high flow rates applied in this study)."

p. 4910, l.8/9: „The DMA and the APM were operated at sufficiently high resolution such that *V*peak was not biased by the influence of doubly charged particles." => Could you please give a little bit more detail (e.g. what is "high resolution")?
The resolution of the DMA is determined by the sample-to-sheath air flow ratio and the resolution of the APM depends on the APM geometry, the revolutions per minute, the flow rate, the selected mass, the effective density of the particles, the temperature and the pressure (Ehara et al., 1996). The critical resolution below which interferences from multiply charged particles play a role does depend on the selected mobility diameter, the shape of the particle number size distribution, the charging state, the size dependence of the effective density and the homogeneity of the density distribution at the selected size. In our experiment we could use the SP2, which allows distinguishing between singly, doubly and triply charged particles, to verify that V_{peak} was not significantly biased by the interference from multiply charged particles. This is illustrated with Figures AR3 and AR4, which are representative of a "medium" and "strong" interference, respectively, relative to all measurements. The potential bias is always smaller than the accuracy of the measurement. We have added the following statement to the manuscript:
"The DMA and the APM were operated at sufficiently high resolution such that the potential bias of V_{peak} due to interference from multiply charged particles was smaller than the measurement accuracy. This was verified using the SP2 data, which make it possible to distinguish between singly and multiply charged particles."

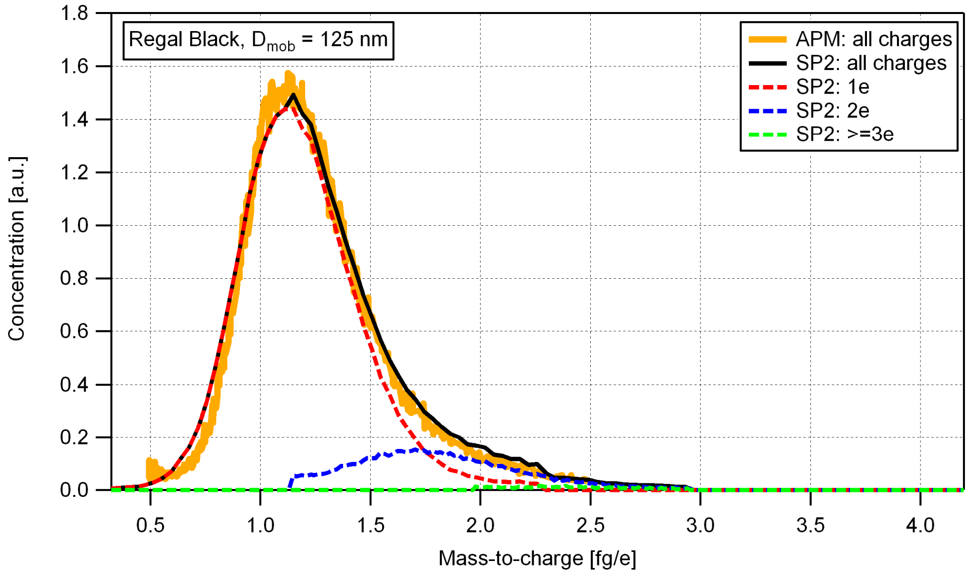

Figure AR3: Identification of the influence of multiply charged particles on the APM measurement for Regal Black particles with a mobility diameter of 125 nm.

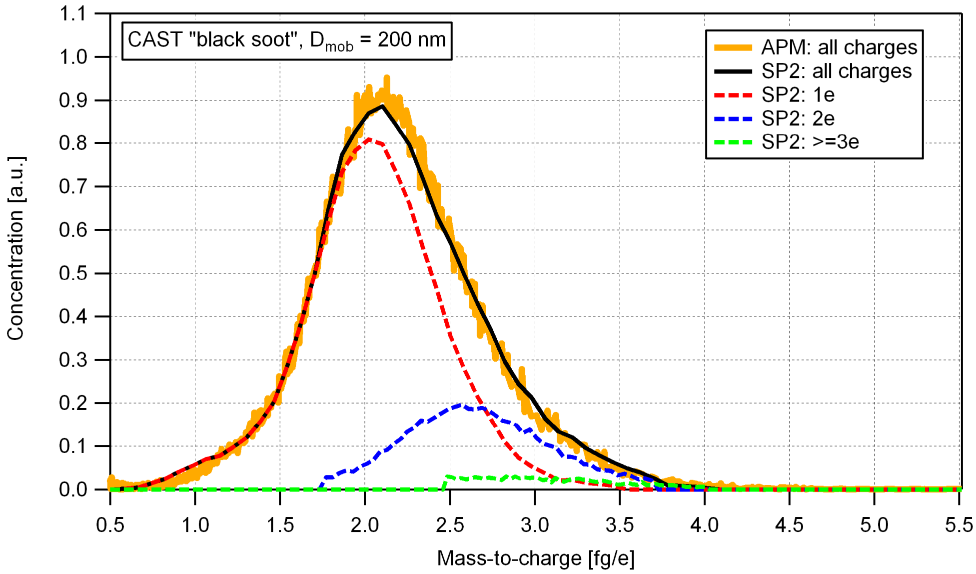

Figure AR4: Identification of the influence of multiply charged particles on the APM measurement for CAST "black soot" particles with a mobility diameter of 200 nm.

p. 4910/4911: How do the settings of the PALAS soot generator impact the particle morphology?
Previous literature (see references in the manuscript) consistently reports very small primary particle sizes and a low fractal dimension for PALAS soot.

p. 4915, l. 19ff: Explain fractal dimensions and the meaning of these numbers.
The reader is now referred to a textbook:
"The PALAS soot particles are fractal-like agglomerates (see e.g. Kulkarni et al., 2011, for details)…"

p.4916, l. 13: The authors mention biomass burning aerosol but they do not further discuss it. Either more discussion on detectability of biomass burning particles is given, otherwise the authors should not refer to biomass burning particles.
The remark on the biomass burning has been removed.

References:

Ehara, K., Hagwood, C., and Coakley, K. J.: Novel method to classify aerosol particles according to their mass-to-charge ratio - Aerosol particle mass analyser. J. Aerosol Sci., 27, 217-234, 1996.