

Author reply to short comment by Kim Prather:

We would like to thank Kim Prather for her useful comments, which helped clarifying and improving this paper. The comments are repeated in black font and the author replies are highlighted in blue font (*“italic style is used for modifications to the manuscript”*).

This is an interesting finding. The authors attribute the inability of the SP2 to detect soot from the Palas generator as being due to a very low effective density of soot. Would they expect a wavelength dependence in this effect? It might be useful to discuss other laser-based techniques that have detected individual Palas generated soot particles. For example, Su et al. and Spencer et al. (below) detected Palas single particle MS which using on-line laser desorption with a laser at 266 nm.

The wavelength of the laser used to heat the PALAS soot particles is essentially unimportant as elemental carbon is known to absorb light across the whole NIR-VIS-UV range with high efficiency (Schnaiter et al. 2003). The mass specific absorption cross section of PALAS soot is of course a factor of ~5-10 higher at a wavelength of 266 nm (ATOFMS) compared to 1064 nm (SP2). However, the key difference between the two methods is that the ATOFMS operates at ~3 orders of magnitude higher power density ($\sim 3 \cdot 10^8$ W/m²; Su et al., 2004) than the SP2 ($\sim 1.7 \cdot 10^5$ W/m²; Schwarz et al., 2010).

The following paragraph has been added to the revised manuscript:

“PALAS soot can be brought to incandescence by using a pulsed laser (Roberts et al., 1988) and it can be vaporised by the desorption/ionization laser of the aerosol time-of-flight mass spectrometer (ATOFMS; Su et al., 2005). These two methods apply laser pulses of much higher intensity than that of the continuous-wave laser applied in the SP2. This indicates that sufficient laser intensity plays an important role for reliable detection of PALAS soot by laser-induced incandescence, as further detailed in Sect 3.3.”

Is it possible there is a difference in the timing of the signal(i.e. do they gate the time when they look for incandescence)?

Timing issues can be excluded, as the signals from all detectors were stored for every PALAS soot particle (with $D_{\text{mob}} = 305$ or 500 nm) during the whole transit time through the laser beam. The peak of the incandescence signal, if at all present, must occur within the laser beam, as there is no heating of the particle outside the laser beam. The paragraph discussing potential data acquisition issues has been modified, also in response to referee #2:

“The SP2 records the signals from a particle only if at least one signal crosses the trigger threshold for data storage, which is set in the data acquisition software. Failure of signal triggering or any other issue with the data acquisition software can be excluded. The position sensitive detector’s signal triggered storage of the signals from all detectors for all PALAS soot particles with $D_{\text{mob}} = 305$ or 500nm. This proves that neither coincidence nor duty cycle problems caused the low counting efficiency. On the contrary, the signal from the broadband incandescence detector was recorded for all particles but it contained only baseline noise without a true incandescence peak for most PALAS soot particles.”

If they sent them through high RH and collapsed their structure, would they then be able to detect them?

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.

It would be worth adding a bit more discussion as to why the soot particles are not detected by their particular set-up.

This technical note is about identifying the reason why the SP2 fails to reliably detect PALAS soot (and to show that this can generally be expected for any operational SP2 in standard configuration). The conclusion is (p. 4917, l. 21 ff): "...the PALAS soot particles remain undetected as the SP2's laser intensity is insufficient to heat the primary particles to their vaporisation temperature because of their small size ($D_{pp} \sim 5-10$ nm)." The role of laser intensity and conductive heat loss for the detection of BC particles is discussed in detail on p. 4913, l.12 ff for the SP2 in general and on p. 4915, l. 17 specifically for PALAS soot particles. Further discussion on the effect of laser intensity has been added in response to other comments.

References:

1. Su, Y.X., M.F. Sipin, K.A. Prather, R.M. Gelein, A. Lunts, and G. Oberdorster, ATOFMS characterization of individual model aerosol particles used for exposure studies. *Aerosol Science and Technology*, 2005. 39(5): p. 400-407.

2. Spencer, M.T. and K.A. Prather, Using ATOFMS to determine OC/EC mass fractions in particles. *Aerosol Science and Technology*, 2006. 40(8): p. 585-594.

Schnaiter, M., Horvath, H., Möhler, O., Naumann, K.-H., Saathoff, H., and Schöck, O. W.: UV-VIS-NIR spectral optical properties of soot and soot-containing aerosols. *J. Aerosol Sci.*, 34, 1421-1444, 2003.