

The authors would like to thank the referee for his thorough review and very useful comments that helped improve the clarity and the relevance of this paper. The referee's comments are repeated in blue font and the answers are given in black.

Comment: The Abstract should mention that the analysis of the SP2 mass sensitivity is based on the Aerosol Particle Mass Analyzer (APM).

The following text was added to the abstract: "The sensitivity of the SP2 was determined, using an Aerosol Particle Mass Analyzer, for rBC from thermodenuded diesel exhaust..."

Comment: Page 666, line 7: How is refractory BC mass defined? Is it defined as the carbonaceous particle mass that shows a LII signal? Please give a brief description.

The following text was added: "This technique indirectly quantifies the mass of rBC in single particles by measuring the intensity of the thermal radiation emitted at the rBC boiling point. The rBC is therefore defined as the most refractory fraction of carbonaceous material retained on the particle until incandescence is reached at ~4000 K."

Page 668, line 24: Please add", and the applicability of a concentric coated sphere optical model." at the end of the sentence starting with "However the accuracy ...".

Sentence changed to:

"However, the accuracy of this approach depends on the accuracy of the detectors's calibration as well as on further assumptions such as e.g. the applicability of a concentric coated sphere Mie model or the refractive indices of rBC core and coating"

Page 669, lines 15 to 17: Strictly, a differential scattering cross section is measured by the SP2 which is defined by the solid angle of the scattering detector.

This has been clarified in the revised manuscript by adding the following text at the first appearance of the scattering cross section: "The scattering signal measured by the SP2 is proportional to the differential scattering cross section of the recorded particle into the solid angle covered by the detector optics. For conciseness, we will hereafter just refer to scattering cross section. Figure 1d (cyan line) shows the scattering cross section of an ambient particle at different times throughout the laser beam."

Page 669, lines 24: The exact laser intensity profile is not shown in Fig. 1a. What is shown is the normalised laser intensity profile $I(t)=I(t_{\text{centre}})$ that can be determined by a statistical analysis of a set of time resolved single particle scattering signals from purely scattering particles. I suggest to introduce the normalised intensity profile here and use this definition later in Eq. (4).

We prefer keeping the factor $I(t_{\text{centre}})$ in Eq. 4, in order to keep it valid independent of the approach used to normalize the scattering signals (they can for example be normalized by their area or by their amplitude). However, we modified the text to explain in more detail how the laser profile is determined and that the scattering signals have to be normalized for this purpose:

"The exact laser intensity profile as shown in Fig. 1a can therefore be determined with statistics of normalised scattering signal shapes obtained from purely scattering particles."

Page 670, Eq. (4): The authors might want to substitute t by t_{α} to be more consistent with the argumentation in this paragraph.

This substitution was not made because Eq. 4 describes the calculation of the scattering cross section at any time t . The time t_{α} , is used in Eqs. 2 and 3 since these relationships are illustrated in Fig. 1 for this specific time. However, we have added an extra sentence above Eq. 4 in order to lead over from the specific time t_{α} to free time parameter t : “The laser intensity for any other time t can be calculated in the same way, and the scattering cross section $\sigma(t)$ is obtained by dividing the measured scattering amplitude by the corresponding normalised laser intensity (Fig. 1d)”

Page 671, lines 3 and 4: substitute $I(t)$ by the normalised intensity profile $I(t)=I(t_{\text{centre}})$ to be consistent with Eq. (4) (see my comment above).

“ $I(t)=I(t_{\text{centre}})$ ” does not make sense since t is a free parameter, while t_{centre} is a fixed time. The referee probably meant defining the normalized laser intensity such that $I_{\text{norm}}(t_{\text{centre}})=1$. Anyway, we argued in the previous reply, why we prefer keeping $I(t_{\text{centre}})$ explicitly in Eq. 4.

Page 675, line 21: Please give the OC content found in the work by Chirico et al.

Added: „This results is consistent with the study by Chirico et al. (2010), who reported organic mass fractions of ~15% for the same diesel car“.

Page 679: The authors argue that the observed increase in the incandescence signals of thermodenuded Aquadag and fullerene soot particles at equal mass is due to a small amount of non-refractory coatings on the untreated particles. Now it is well known from laboratory studies (e.g. Mennella et al. (1995) and references therein) that the annealing of hydrogenated amorphous carbon particles results in a change of the electronic structure of the material and to a relative increase of the sp^2 hybridisation. This would be an alternative explanation for the observed changes in the calibration and scattering cross section curves. The authors should address this alternative explanation.

Answer: Mennella et al. (1995) studied the effect of annealing graphitic carbon at different temperatures and a duration of 3h on the sp^2 hybridisation. They showed that the sp^2 hybridisation starts to increase when heating to 400°C. We here argue that the annealing of hydrogenated amorphous carbon particles is unlikely to be the reason for the observed behavior for several reasons. First, in this study, the residence time of the particles in the thermodenuder is only 4s. Second, in order to see a measurable effect in the calibration curve, the sp^2 hybridisation would have to increase throughout the bulk of the BC and should not only be a surface effect. In the light of Mennella’s study it is unlikely that 4s at 400 °C would be enough for the sp^2 hybridisation to happen throughout the bulk of the BC.

The following discussion was added to the revised manuscript:

“Mennella et al. (1995) showed that carbon black’s chemical structure and absorption properties are altered by heating the sample at 400°C for several hours.

The thermodenuder effect on the SP2’s sensitivity to Aquadag® and fullerene soot is

unlikely due to this phenomenon as the particles are only heated for a few seconds. On the other hand, a small remaining coatings on the particles can shift the calibration.”

Fig. 3: Why does the thermodenuded Diesel exhaust BC show a more uniform scattering distribution, and why is the scattering signal on average higher for those particles?

The following sentence was added to the revised manuscript in order to clarify this point: “The number of data points in Fig. 4 cannot be compared between different panels for several reasons: Sampling durations are different, and panels a-c are from polydisperse samples, while panels d-f are from multiple monodisperse measurements.”

Technical corrections

Page 671, line 12: Remove the redundant “thus”.

Done.

Page 671, line 28: Remove “only”.

Done

References

Mennella, V., Colangeli, L., Blanco, A., Bussoletti, E., Fonti, S., Palumbo, P., Mertins, H.C., A dehydrogenation study of cosmic carbon analogue grains, *The Astrophysical Journal*, 444, 288-292, 1995