The authors would like to thank the referee for his/her 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.

Question: The method and results from the new technique to identify coatings presented in the manuscript needs to be given greater weight in the abstract and introduction, given its importance for interpreting the results. In the current manuscript this new approach is not mentioned in the abstract, but would be of interest to the SP2 community.
The following sentence was added to the abstract:
"The amount of remaining non-refractory matter was estimated to be below 30% by mass, according to a comparison of the scattering cross sections of the whole particles with that of the pure BC cores."
In addition, the following sentence was added to the introduction:
"A new method using the time-resolved scattering cross section measurement for the identification of thin non-refractory coatings is also presented."

Question: The introduction should also include some background material on previous attempts to identify coatings on BC particles.
Some existing methods for the identification of coatings on BC particles were already discussed in Section 2.1 of the AMTD manuscript. This paragraph has been complemented with:
"Moteki and Kondo (2008) introduced an alternative method to determine the time-resolved scattering cross section of the particles, which is essentially similar to that of Gao et al. (2007), except that the PSD is not required."
We prefer keeping the discussion of alternative approaches in Sect 2.2 of the revised manuscript, as it is tightly linked to the new method. However, we have added additional subsection titles in Sect. 2 for better guidance of the reader.

Question: The paper should also provide a more detailed description of the method, including information regarding the size range of BC-cores and total particles sizes over which coating information can be obtained, as well as much more details regarding the experimental uncertainties. They are occasionally referred to but not quantified, nor is there much description of how they were determined.
The new Figs. 3 and S1 and Section 2.2.2 were added to provide a detailed sensitivity analysis including the previous time-delay approach. However, information on the size ranges for which the method can be applied is not given, as these size ranges are not of general nature. In contrast, they are different for each SP2 and they also depend on user selectable instrument parameters.

Question: This material is probably deserving of its own expanded section and should also include a comparison to the previously applied time-delay and optical estimates for identifying coatings.
See previous comment.

The following sentences were added in order to clarify the method used to obtain the laser beam shape:

"The intensity profile is normally stable over time-scales of days, unless contamination of laser optics or alignment drifts occurs."

and

"In this study, the laser intensity profile was determined on a daily basis by analysing the signals of several thousands purely scattering particles from an ambient sample."

Answer: We tried to obtain an unbiased calibration using thermodenuded ambient BC particles by restricting the analysis to the data points for which the scattering cross section of the whole particle agrees with the one of the bare BC core. Unfortunately, no significant difference in the calibration curve could be obtained. This absence of difference can be explained by the fact that the experimental noise of individual data points is similar or larger than the actual heterogeneity of the particle properties in our data set. Nevertheless, this approach could in principle work if a better signal-to-noise ratio could be achieved.

A second approach would be to correct the calibration curve for the estimated mass fraction of the coating. A detailed sensitivity analysis presented in the new Fig. 3 Section 2.2.2 shows that the residual coating is on average most likely below 30% by volume (slightly less by mass) for the thermodenuded ambient BC. The corrected ambient calibration would still remain closer to the diesel and fullerene soot calibration curves than to the Aquadag calibration curve.

However, we promote the diesel calibration curve as the primary reference rather than correction of the ambient calibration, considering the uncertainties of the estimated coating mass fraction

This statement was included:

"…However, this observation remains to be confirmed with
measurements in different locations featuring contributions by possibly different BC sources such as diesel exhaust, biomass burning, trash burning."

Question: The paper states that diesel is likely the most important BC source in "most locations" but this should be clarified, since biomass burning emissions are estimated to contribute over half of the globally-emitted BC (Bond et al., 2004). Do they mean most urban locations? Or locations where vehicular emissions are predominant?

This was clarified as follows:

"This diesel exhaust calibration is expected to be representative of the SP2's sensitivity to ambient rBC where fossil fuel combustion is the dominant BC source. This applies for most urban locations (e.g Szidat et al., 2006), whereas different sources such as biomass burning, burning of biofuel or urban waste may be dominant in other locations (Bond et al., 2004)".

Specific comments (Pg-Line)

664-2: odd wording here, suggest either omitting "nowadays" or replacing with "now"
Replaced with now

664-27: ": : :agrees with the one of: : :" suggest re-phrasing
Changed to "agrees with the SP2 sensitivity to rBC from Diesel exhaust"

665-2: suggest changing "curve" to "response" here and throughout the manuscript
We agree that using "response" might be better here. However, at other instances the term "curve" is more suitable. For consistency reasons we keep the term "curve" throughout the whole manuscript.

665-10: "nowadays"
Replaced with now

665-13: "earth" to "earth's"
Done

665-15: "being" to "beings"
Done

666-16: "that one" to "that"
Done

666-25: should also add the CPMA as well as APM here, or refer to the method more generally (e.g., single particle mass analyzers, or something similar)
Done

666-26: though thermodenuders are fairly common in the aerosol community a general reference to the technique is probably still needed here, or else could change to "Heating ambient particles to remove non-BC components using a thermodenuder may provide: : :"
Changed to: "Heating ambient particles to remove non-BC components using a thermodenuder may provide nearly pure ambient rBC,…"

667-23: suggest adding wavelength info somewhere here
The wavelength has been added to the methods section: "…through a high-intensity continuous intra-cavity laser beam (λ=1064nm) in order…"

668-19: "early in the laser" suggest re-wording (e.g., "determining the coated particle's optical diameter from the initially unperturbed scattering signal when the particle enters the laser beam"
Done.

668-24: the coating thickness estimate also depends on the assumed density of the rBC core since this determines the rBC volume/diameter
The main message of this sentence is that calculated coating thickness values are susceptible to instrument calibration and several assumptions, while providing a complete list of all major and minor uncertainties would just dilute the main message. The assumptions made on BC's bulk density are only a minor uncertainty. We modified this sentence to emphasize the fact that the list is not exhaustive:
"However, the accuracy of this approach depends on the accuracy of detector calibrations 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."

672-8-9: "within experimental uncertainty" As stated in the general comments, a description of how the experimental uncertainties were determined should be provided, along with an estimate of typical values for ambient particles. What is the critical difference in scattering cross-section needed to be observed to classify the particle as having a coating?
The new Fig. 3. and Section 2.2.2 provide a detailed sensitivity analysis.

672-9: The approach described here is also similar to that described by Gao et al. (2007) and Schwarz et al. (2008) in that those investigators identified classified a BC particle as being coated if there was a decrease in the scattering signal prior to the onset of incandescence. The approach described in the manuscript is exploiting essentially the same feature in the scattering signal, but in a more quantified way.
The authors totally agree, which is reflected in the following statements already made in the AMTD manuscript: "In Fig. 1 we present an approach that builds up on the method introduced by Gao et al. (2007)." (page 668, line 29) and "This approach is essentially equivalent to a leading edge fit as described in Gao et al. (2007)." (page 671, line28).
The following sentences were also added;
"Moteki and Kondo (2008) introduced an alternative method to determine the time-resolved scattering cross section of the particles, which is essentially similar to that of Gao et al. (2007), except that the PSD is not required."

668-25: The text describing the method used to identify thin coatings should be a separate sub-section within the methods section.
Additional subsection titles have been added.

Also, please give range of operating temperatures used with the thermodenuder in the study

The temperature and the description of the thermodenuder is described in details in previous section 2.3 (now 2.4). In addition the temperature is now also mentioned in Sect. 2.2.1 at the first instance of mentioning the thermodenuder in the experimental section: "In this study, a thermodenuder (heated to 400˚C) was used to obtain pure rBC cores"

669-3: Is the particle being used an example an ambient BC particle, from wood smoke, or from diesel exhaust?

The example particle is from ambient air. This sentence reads now: "Fig. 1d shows the scattering cross section of an ambient particle at different times throughout the laser beam (cyan line)."

671-2: what is the physical meaning of the normalization factor? Also, brackets at end of "center" are sub-script.

The formatting has been corrected and the following sentence has been added: "This normalisation assures that absolute values of the scattering cross section can directly be obtained by multiplication of the normalised sigma(t) with the calibration factor of the scattering detector."

671-27: there is an extra "/" at the start of the in-line equation

This is a capital "I" and not a slash and it belongs there.

673-3: is this just the engine?

No, the entire car was operated at idling and the sampling line was connected to the exhaust pipe as described by Chirico et al. (2010).

673-21: can any previous ambient sampling studies at the PSI facility be cited here to support this?

No paper was published on this. However PSI, originally a nuclear research center, was built in the middle of forests and agricultural fields with only small villages (<10'000 inhabitants) nearby, and it is 10 km away from the nearest motorway and 35 km away Zurich, the nearest major city.

675-20: there could also be an absence of coatings if the organics and BC are externally mixed, though this is highly unlikely in this case.

Agreed. However, only the properties of the BC containing particles are relevant in our case. We modified the sentence to: "…which indicates essentially no coating and a low OC to BC ratio of the BC particles."

Suggest changing "consequently" to "consistent with".,

Sentence was reworded in response to the above comment.

676-2: What is the potassium sulfate boiling point? Potassium chloride is also emitted

and tends to be the majority of the inorganic composition, so may be worth including its boiling point as well. Text should clarify that these materials will be vaporized in the SP2 due to the very high boiling point temperatures of the BC.

The text was adapted: "alkali salts such as potassium sulfate or potassium chloride are known to be common constituents of WB (Dusek et al., 2005) and will not be removed in the thermodenuder due to their high boiling point (1689 °C and 1420 °C, respectively). In the SP2, however, the particles reach temperatures of typically ~4000 K (Schwarz et al., 2006) and these compounds will evaporate before the maximum of the incandescence signal is reached".

676-5: What is the detection uncertainty? It would be useful to have additional lines on the Figure 3 indicated the region of non-significant differences between the scattering cross-sections of pure BC and the whole particle.
This was addressed with the new Fig. 2, Section 2.2.2 and additional lines in Fig. 4 (prev. Fig. 3).

676-6: there are dramatically fewer BC particles detected after thermodenuding the wood smoke samples. What is the reason for this? Losses through the thermodenuder are probably not this large. How do the total numbers of BC-containing particles compare for the thermally denuded and non-treated samples? Are the differences in number due simply to different sampling times? If so perhaps it would be better to shade pixels by concentration rather than number:
This was clarified:
"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."
Besides we decided to keep absolute numbers instead of concentrations, as only the former provides information about the actual sample size.

676-10: Does the Kondo et al. (2011) reference refer to BC-containing particles or total ambient particles? How much of the particle volume would have to remain to explain the results in Figure 3e?
This was clarified:
„Kondo et al. (2011) showed that pure organic particles composed of saccharides, dicarboxylic acids or aromatic acids lost >95% of their volume removed when heated up to 400 °C, however, the removal efficiency was as low as 20-60% for humic-like substances."
Furthermore, the additional Fig.3 and new Section 2.2.2 present the estimated remaining coating volume fraction, which explains the result in Fig.4e (formerly Fig. 3e).

676-28: The Szidat paper describes measurements performed in Zurich, Switzerland so it is difficult to see how it supports the statement that BC mass is dominated by diesel exhaust in most locations. Emission estimates (e.g., Bond et al., 2004) suggest that over half of BC emitted globally arises from biomass burning of some type. Perhaps changing to most "urban" locations would be an improvement, but even then there are

many cities where other BC sources are likely important (e.g., Mexico City; Yokelson et al. 2007). The Szidat reference should be cited to state that the ambient sample referred to in the manuscript is likely to be dominated by diesel emissions, or at least fossil fuel emissions, based on the carbon isotope analysis.

The sentence has been changed to: "This applies to most urban locations (e.g. Szidat et al., 2006), whereas different sources such as biomass burning, burning of biofuels or urban waste may be dominant at other locations (Bond et al., 2004)."

And a bit further down we added the caveat: "This confirms that the diesel exhaust calibration curve can be considered to represent the SP2's sensitivity to ambient rBC where fossil fuel emissions are the dominant source of BC."

677-14: "An unbiased ambient calibration would likely:.." In the absence of any estimate of the magnitude of the bias this statement isn't particularly meaningful. If the bias were large enough the diesel calibration could potentially fall quite far from the ambient data on the other side of the 1:1 line. If the authors could provide an upper estimate of the bias it would help support the claim made here.

This was addressed with the additional Fig.3 and new Sect. 2.2.2 as well as the following statement added to Sect. 3.1.: "The volume fraction of the remaining non-BC matter is most likely below 30% on average, as indicated by the pink line in Fig. 4e, which is taken from the sensitivity analysis presented in Sect. 2.2.2 and Fig. 3. The corresponding mass fraction will be slightly lower, depending on the density of the coating."

And the following statement has been added to Sect. 3.2:

"The bias in mass was estimated to be most likely below 30% (Sect. 3.1). Consequently an unbiased ambient calibration would still fall close to the diesel calibration."

Figures
The aspect ratio of Figure 4 is quite horizontal and it would be easier to see the differences between the curves if it was made more square in the AMT version.

Done.

Error bars should be described in the caption as standard deviations or appropriate parameter.

Done.

It might also be useful to indicate an approximate range for ambient BC particle size/mass based on previous SP2 literature reports.

Answer: The following statement is already made in the discussion version of the manuscript: "…in the mass range 5–10 fg, where the atmospheric rBC mass size distribution typically peaks (Schwarz et al., 2008)…"

References
Bond et al., 2004, doi:10.1029/2006JD007315

Chirico, R., DeCarlo, P. F., Heringa, M. F., Tritscher, T., Richter, R., Prevot, A. S. H., Dommen, J., Weingartner, E., Wehrle, G., Gysel, M., Laborde, M., and Baltensperger, U.: Impact of aftertreatment devices on primary emissions and secondary organic aerosol

formation potential from in-use diesel vehicles: results from smog chamber experiments, Atmos. Chem. Phys., 10, 11 545–11 563, 2010