We thank the anonymous reviewers for their extensive comments and suggestions. Both reviewers suggested significant revision of the manuscript. We have rewritten much of the Section 4 to include a more detailed discussion of the sensitivities of the technique to the various input variables, including a more detailed consideration of the systematic errors. We have also attempted to provide further justification for our use of the term “accuracy”. These two themes were recurrent during both sets of comments, and detailed discussions of both are presented separately at the end of this document under the titles “Discussion of uncertainty” and “Use of the term accuracy”. We refer to these comments during our responses.

We present the reviewers’ comments in bold, our responses in standard font and the changes to the text in italics.

Anonymous Reviewer #2

General comments

This paper promises to assess the sensitivity of SP2-derived black carbon (BC) aerosol coatings to assumed density and refractive index. The analysis presented is decent and good arguments are made for using certain values of density and refractive index over other values; however, the results are not really presented as a “sensitivity.” Overall, the paper falls far short of a complete sensitivity analysis as is promised by the title and abstract. I hope the authors do a detailed sensitivity analysis and majorly revise the manuscript to fit the title, rather than scale back the scope of this study. I believe there ought to be sufficient data in this dataset to perform such an analysis.

We thank the reviewer for their constructive criticism and have sought to improve our analysis accordingly. Some of the more substantial modifications are given at the end of this document in ‘Discussion of uncertainty’. However, we must point out that there is no universally-accepted definition of what constitutes a ‘complete sensitivity analysis’, so the exact criteria of what would qualify is somewhat subjective.

A more complete sensitivity analysis should include a more systematic variation of the input parameters rather than choosing just six values from the literature. The refractive index should be separated into real and imaginary parts and the sensitivity to each part should be assessed.

The reason we chose literature values (specifically, those use in previous analyses using the same technique) was so one may assess how much of the variation in coatings reported by different studies may be due to real variation and how much may be due to simply using different values for density/refractive index.

Were we attempt to vary the core refractive index systematically, we would struggle to know what numbers to use, as it is not clear how (if at all) it varies. Bond and Bergstrom (2006) listed several possible values (for use at visible wavelengths) that lie in a straight line when the imaginary and real parts are plotted against each other, but suggest that there is in fact a single value for BC density and refractive index, and they were uncertain which it was. The values discussed here that were not taken from Bond and Bergstrom (2006) do not fall on this line. Real and imaginary refractive index are generally considered to co-vary according the Kramers–Kronig relation (Bohren and Huffman, 1983), but Bond and Bergstrom (2006) note that this is difficult to apply in the case of soot. Consequently, we have not made this addition to the paper.

We have however performed a more systematic exploration of the effects of refractive index and density, varying them separately- this is discussed below in “Discussion of uncertainty”

In addition, sensitivity to the coating refractive index should be assessed. The constraint on coating refractive index from co-located measurements of composition by the SP-AMS can be used to assess accuracy, but it is still of interest to assess the sensitivity to this parameter as well.
We now have, see comment below “Discussion of uncertainty”

Speaking of accuracy, throughout the current manuscript, it is reported that an accurate determination of mixing state can be made with proper input parameters. However, accuracy is never proven. To prove parameter accuracy, another independent measurement is needed, such as the co-located bulk optical property measurements. A forthcoming JGR paper by the same authors (Taylor et al., 2014) may address some of my concerns about accuracy, but this was impossible to assess in this review. If a discussion of accuracy is to be kept in this study, some of the optical property data seems necessary to include in this manuscript.
Taylor, J. W., J. D. Allan, D. Liu, M. Flynn, P. L. Hayes, J. L. Jimenez, B. L. Lefer, N. Grossberg, J. Flynn, J. Gilman, J. A. de Gouw, and H. Coe (2014) Comparison of measured black carbon mass absorption coefficient to modelled values during CalNex 2010, J. Geophys. Res. Atmos., in preparation.
We think the reviewer was suggesting something like plotting mass absorption cross-section (MAC) vs thermodenuder temperature (for the periods we have defined as the freshest airmasses) , which would ideally show a flat section at the higher temperatures, meaning all nonrefractory material had been removed. We tried this, but didn’t have enough signal/noise on our absorption measurement for the data to be statistically meaningful. We were able to perform a similar analysis using E_{Sca}, which is discussed below in the comment “Use of the term accuracy”

Similarly, a detailed discussion of the sensitivities within the SP2 itself should be added. This discussion is partially addressed with the section on notch location, but this could be much more complete. For example, taking the minimum detectable change in scattering intensity (2.44 mV from the APD), what is the minimum detectable difference in scattering cross section? All else being equal, how does this translate to core size or coating thickness?
We have added a discussion of the sensitivities of the instrument (incandescence vs scattering noise and calibration uncertainty), see below in the section ‘Discussion of uncertainty’. For the part about the minimum detectable change, this depends entirely on how the instrument is set up (e.g. the gain on the detectors), it would be different for every instrument. It would be particularly different for the D-model SP2s. See Laborde et al (2012b).

The paper is decently written for the most-part, but there are quite a few technical corrections listed below that are meant to make the wording more precise and clear. For example, use of the word “this” as the noun in a sentence should be avoided. While I find no major errors in the analysis presented, I do find it a bit lacking for a complete “sensitivity analysis” and, thus, I recommend a major revision before publication.

We hope that the clarifications and modifications presented here merit the paper as suitable for publication.

Specific comments

Title : To me, the wording is a bit confusing. In my opinion, it would be better to move "to density and refractive index" to immediately after "sensitivity." Also, because sensitivity is not rigorously explored in this paper, consider a new title if the scope of this paper remains largely unchanged throughout this review process.

Regarding the change of the title, the suggested changes would leave the title functionally identical to how it was before. A straw poll in our department (Manchester) showed no obvious winner, and most people said either would be fine. Given that changing the title could lead to confusion down the line, we think the title should stay as it is.

We have added more discussion of sensitivity below in comment "Discussion of uncertainty"

pg 5492 - line 16 : In many places throughout the manuscript (I won't list them all), you use the wording "coating thickness distribution" and then quote a single number with a standard deviation. Is this really a "distribution?" It seems like it would be more accurate to say that this number is a mean value plus-minus one standard deviation. If, instead, you mean that this value is the peak location of a Gaussian and the plus-minus value is the width of the Gaussian then it must be made clear in the text what you are reporting.

There is a distribution, as shown in Figure 6. However, we have clarified in the manuscript when we are talking about the mean/standard deviation/median. The shape of the distribution is not always a perfect Gaussian (see updated Figure 6, panels (c) and (d) which show extra coated particles on top of a main Gaussian mode). We also note that the peak and width (σ) of a Gaussian distribution are the same as a mean and standard deviation, but we had to use a fitted Gaussian so the result was not skewed by particles with $E_{Sca} < 1$.

5492 - 17 : How do you know the determination of mixing state is accurate? Do you have an independent measure of mixing state that you can directly compare to your results? In the manuscript, it seems like you assess accuracy as a situation that makes sense or sounds reasonable (that is, fresh BC that passes through a thermodenuder ought to have little if any coating) and whatever parameters gives this most-likely situation is "accurate." To truly assess accuracy, you need to have an independent measurement that you can quantitatively compare to the derived mixing state in this paper. This independent measurement could be those co-located optical property measurements, which I assume are all presented in Taylor et al., 2014. To keep these "accuracy" statements in this manuscript, you will need to bring in some of the data from your other pending paper.

See comment below "Use of the term accuracy"

5492 - 19-20 : The fact that the precision remains the same regardless of what parameters are used in the core/shell model simply means that whatever variation or noise exists in the retrieved coating thicknesses is a result of noise in the raw measurement. This point should be made clear in the paper. Also, it would be nice to assess the

sensitivity of the retrieved coating thickness to incandescence versus scattering noise.

We have added the following text to Section 3.6

"The fact that the spread in E_{Sca} *is similar regardess of the core paramters used means the main source of per-particle noise is in the raw data.The spread in the data is due to a combination of random error in the LEO fits, particles passing through different parts of the laser, and uncertainty in* D_C*. The per-particle statistical uncertainty in rBC mass is ± 30%, (equivalent to* $^{+9\%}_{-11\%}$ *in* D_C*). As scattering scales roughly with diameter to the sixth power, this uncertainty suggests a per-particle uncertainty in* E_{Sca} *of* $^{+70\%}_{-50\%}$ *from incandesence noise. The per-particle noise in the LEO fits during PSL calibration was ±40%, meaning the main source of per-particle noise in* E_{Sca} *and any derived coatings is the incandesence channel."*

5497 - 5 : Put your calibration with glassy carbon spheres in context of the fullerene/aquadag discussions found in Baumgardner et al., 2012 and Laborde et al., 2012. I see you mention it briefly in the supplemental section, but I think its important enough to mention in the main manuscript and maybe expand the discussion in the supplemental to include an estimate of the bias you would expect by calibrating with glassy carbon spheres versus the other materials.

See comment below "Discussion of uncertainty"

Baumgardner, D., O. Popovicheva, J. Allan, V. Bernardoni, J. Cao, F. Cavalli, J. Cozic, E. Diapouli, K. Eleftheriadis, P. J. Genberg, C. Gonzalez, M. Gysel, A. John, T. W. Kirchstetter, T. A. J. Kuhlbusch, M. Laborde, D. Lack, T. Müller, R. Niessner, A. Petzold, A. Piazzalunga, J. P. Putaud, J. Schwarz, P. Sheridan, R. Subramanian, E. Swietlicki, G. Valli, R. Vecchi, and M. Viana (2012) Soot reference materials for instrument calibration and intercomparisons: a workshop summary with recommendations, Atmos. Meas. Tech., 5, 1869–1887, doi:10.5194/amt-5-1869-2012.

Laborde, M., P. Mertes, P. Zieger, J. Dommen, U. Baltensperger, and M. Gysel (2012) Sensitivity of the Single Particle Soot Photometer to different black carbon types, Atmos. Meas. Tech., 5, 1031–1043, doi:10.5194/amt-5-1031-2012.

5497 - 8 : This wording makes it sound like your SP2 was plus-minus 10% of NOAA's SP2. However, in the supplemental section, you more clearly state that there was a systematic offset, which is what I would expect given your calibration material. Also, in the supplemental section, you further develop the uncertainty estimate to 14%, but never mention this in the main manuscript. You need to more clearly state your total uncertainties and biases in the main manuscript to bring the discussion in line with your supplemental section.

We have clarified in the supplement, which now reads

"rBC modal mass measured by Manchester's SP2 was approximately 10% higher than that reported by the NOAA instrument when comparing several sizes of monodisperse fullerene soot [Anne Perring, private communication, 2012]. Convolving this with the 10% uncertainty due to the SP2's response to fullerene soot relative to diesel (Laborde et al., 2012a) gives a systematic uncertainty estimate of average rBC per-particle mass of ±14%. This corresponds to a difference in derived core diameter of

$^{+4.5\%}_{-4.9\%}$ *Assuming any error due to flow calibration is negligible, the error on the derived rBC mass concentration was also ±14%."*
We note that while NOAA's fullerene calibration was more repeatable, it is not clear whether or not it was more accurate.
In the main text we have removed reference to the NOAA comparison, and just said
"(2010). The accuracy of the incandescence calibration was estimated at ± 14%, corresponding to accuracy in average D_C *of* $^{+4.5\%}_{-4.9\%}$ *"*

5497 - 14-15 : Quote the average correction factor (4.6% from the supplemental section) here. Also clarify by stating in the manuscript that the correction was done by fitting the measured mass distribution to the sum of two Gaussian distributions. In fact, exactly how this correction was done was not very clear in the supplemental section A2 and should be improved as well.
The 4.6% the reviewer is referring to was a correction on the mass concentration, not the per-particle mass. We have clarified in section A2 that the 4.6% was based on the lognormal fits
"Based on the lognormal fits, the fraction of rBC mass outside the detection range of the SP2 was estimated as 4.6% of the total. rBC mass concentration measured by the SP2 was corrected for this factor."
We have also performed the fits in a more standard way (using lognormal fits rather than Gaussian fits in log-space), which gives precisely the same result. We have clarified the caption in Figure A1 to reflect this:
"Figure A1 - Average rBC mass-equivalent core diameter mass and number distributions measured by the SP2 during CalNex. A lognormal distribution was fit to $dM/dlogD_C$ *in the dark grey region. This fit was then subtracted from the measured data, and a second lognormal distribution was fit to the residual data in the light grey region. The discontinuity around 330 nm is due to the switch in detectors as the narrowband detector reached saturation, and was not used for the fits. The centres of the fitted modes and lognormal widths were 130nm ± 23% and 426nm ± 41%."*

5497 - 28 to 5498 - 1-3 : The rBC convention is also used for the SP2, as you state on page 5494 - line 18. Why do you say the SP-AMS and SP2 aren't measuring the same material? By using the same laser, they should be measuring the same material. If you mean that the laser powers may not be equal and therefore the incandescence temperatures reached in each instrument is different, then you need to clearly explain this. If you mean that the sum of the carbon clusters in the mass spectra are not equivalent to the incandescence signal in the SP2, then you need to clearly explain this.
We have clarified
"it is not possible to unambiguously determine whether the sum of the carbon clusters measured by the SP-AMS is equivalent to the material detected by the SP2."

5498 - 24-25 : Why do smaller particles have higher rBC to coating mass ratios?
We have clarified

"…the smaller particles that escape detection are likely to have a higher rBC to coating mass ratio. The addition of non-BC material would make them grow and become more spherical, and therefore stand a higher chance of being detected."

5498 - 29 : Why didn't you study the sensitivity to shell refractive index, especially since you recognize a known bias? I think that this should be examined in order to fully understand all the sensitivities involved in the core/shell retrieval technique with the SP2.
We now have, see comment below "Discussion of uncertainty"

5499 - 2 : Can you argue that accumulation mode particles are more important for light scattering than smaller particles and so your derived shell refractive index may turn out to explain bulk optical properties pretty well anyways?
We can't justify this argument in this instance- larger particles dominate bulk scattering, but we are using the shell refractive index in a per-particle sense. As an extreme example, if we sample one giant particle and 10 small ones, the SP-AMS would be mostly influenced by the giant one (as it had the majority of mass), but the SP2 would be more influenced by the small particles as they make up the majority of particle number. We would be unable to tell if the small particles had a different refractive index to the large one.

5503 - 25 : To be clear, are you talking about the notch when the signal changes direction or do you mean when the signal crosses zero? Gao et al., 2007 shows a notch in the signal that is different than the smooth crossing of zero.
Gao, R. S., J. P. Schwarz, K. K. Kelly, D. W. Fahey, L. A. Watts, T. L. Thompson, J. R. Spackman, J. G. Slowik, E. S. Cross, J.-H. Han, P. Davidovits, T. B. Onasch, and D. R. Worsnop (2007) A novel method for estimating light-scattering properties of soot aerosols using a modified single-particle soot photometer, Aerosol Sci. Technol., 41(2), 125–135, doi:10.1080/02786820601118398.
Our text reads
"The signal on one side is inverted, producing a clear notch as the particle passes the center of the detector and the signal switches from negative to positive."
Gao et al state
"The center of the notch in the time series from the TEAPD detector […] is given by the zero-crossing time"
The two are the same

5504 - 11 : Looking at the time-dependent scattering cross section is a different technique than the leading-edge only method described in the rest of this section. If you used this other technique, you need to describe it in a lot more detail, at least in the supplemental section if not in main manuscript.
We have clarified that we only used this to determine what percentage of laser power to use for the leading-edge fits.

5504 - 18 : Is "baseline" really the zero signal line? If this is true, then instead of using the zero-crossing position are you using 20 units above zero (positive signal)? I don't

think this is the case because it doesn't makes sense based on the ambiguity that you describe that you are trying to resolve. Clarify this paragraph.

The text already states in the previous paragraph

"For each LEO fit, the baseline is calculated using the mean of the first six data points 5 (1.2 µs). This is subtracted from the data…"

We have modified the paragraph as follows, which we hope will satisfy the reviewer's concerns

"Rather than rising above the baseline as the particle passes across the split in the TEAPD, these particles' signals merely fell back to the baseline (crossing it due to electronic noise), giving no indication of position within the laser. To remove this ambiguity, in this dataset we redefine the notch position based on the point where the scattering signal crosses 20×2.44 mV units above the baseline (much greater than the typical electronic noise) rather than the baseline itself, such that particles that evaporate before the split are not counted for the purposes of particle size measurements. In practice the data point at which the signal crossed the baseline and 20x2.44 mV units above is almost always the same, so the effect on particles with measurable notch position is minimal."

This proved to be an effective way of eliminating the particles that evaporated too early, whilst having minimal effect on the others.

5506 - 18 : Are there any references that support this claim? From Figure A1, 135 - 200 nm does not cover the "majority" of the size distribution of the cores.

We have changed this sentence to read

"Whilst this does not cover the entire rBC mass distribution, Figures 2b and 2c show minimal size-dependence, other than the measurement limitations discussed previously, meaning this range is likely representative of the majority of ambient absorbing aerosol."

5507 - 12 : Do you know for sure the APD's collection angles and alignment well enough for a 1 nm accuracy on your measurement? I would be surprised if this was true - if it is, you need to prove it in the manuscript or provide a reference to someone who has proven this accuracy. When assessing the sensitivity of your retrieved core/shell parameters, it is important to recognize the physical limitations of the instrument itself and not overstate its theoretical accuracy.

We have clarified that the 1nm resolution is only to reduce truncation error. It is clear from the rest of the manuscript that this does not imply a 1nm accuracy for a single particle. However, this has highlighted the precision with which we should be reporting measurements, and we have changed all reported coating thicknesses to the nearest nm.

5507 - 24-26 : What happens if you adjust the density and refractive index of the core such that the negative coatings are eliminated and the Mie lookup tables give a coating thickness of zero? What would the density and refractive index need to be? Please add a discussion of these numbers and their plausibility for ambient BC.

It is not clear what the reviewer means. If they are asking what the density and refractive index would need to be so that 100% of particles had a positive E_{Sca}, the question does not make sense- in order for that to be the case, all other values of E_{Sca} would also have to increase greatly and the resulting coatings would be wildly implausible. In a perfectly calibrated system measuring externally-mixed particles and using a perfectly precise optical model, 50% of particles would have $E_{Sca} < 1$, with a median of 1. Using Moteki et al's core parameters, we achieve results that are reasonably close to this in our least coated case.

5509 - 4 : Doesn't Figure 4b show that the fraction of particles with Esca <1 is actually not strongly dependent on density?
We have changed this so it just says it is strongly dependent on the refractive index

5509 - 6-7 : What is the "similar technique" you are referring to? Did you actually measure density and refractive index? If so, what method did you use?
We have clarified
"a similar technique using the SP2's scattering measurement"
At no stage in this paper have we claimed to have measured refractive index or density

5509 - 16-18 : There should be more analysis here. If you truly intend to present the "sensitivity" to density and refractive index, then you need to quote a sensitivity, which is Esca / (density) and Esca / (refractive index). From Figure 4b, the sensitivity to density is small, and from Figure 4c, the sensitivity to refractive index will be larger - but we need numbers here. This is also a good place to quantify the width of the distribution in order to prove that density and refractive index are not changing the shape of the distribution, just the offset.
The sensitivity to density and refractive index are interdependent, meaning this is not a trivial calculation to make. We refer back to our response to the reviewer's first comment- this would not make any difference to the conclusions of the paper. The sensitivity of E_{Sca} (and the width of the E_{Sca} distribution) are much less important than the sensitivity of the derived coating properties, as E_{Sca} exists only in the context of the Mie modelling, whereas the derived coating properties are (ideally) real properties. We have investigated the sensitivity to the real variables, including quantifying the width of the distribution, in Section 4 of the manuscript.

In Section 3.6, you derived a density and refractive index for fresh BC by finding literature values that made the Mie model "behave" like you think it should. This analysis does not prove that these are the absolute best values to use for fresh BC in this study - these values are merely the "most appropriate" (5509 - 10) of the values you tested. Also, it is incorrect to say that "all other parameters lead to an overestimation of Esca" (5509 - 13-14) because you did not test all other parameters, just several that you found in the literature.
To be clear, we did not derive a density and refractive index; that work was carried out by Moteki et al (2010), as we have stated in the manuscript several times. We have changed the text so it now reads
"all other parameters tested led to an overestimation…"

Several outstanding questions remain: How applicable are these parameters to aged BC that has been structurally rearranged? Does the thermodenuder do anything to fresh, uncoated BC that might change these results versus sampling fresh BC without the thermodenuder?

As we have stated in Section 3.1, the SP2 operates in the Rayleigh optical regime, meaning shape is not thought to have a large influence (Moteki et al., 2010). So if the BC were structurally rearranged by the thermodenuder the effect would be minor. For aged BC, we have already covered this in Section 3.1:

"For thickly coated particles (which are quasi-spherical in shape due to compaction of the primary BC and filling in voids between spherules), laboratory measurements have shown this method may be used to accurately determine the size of the coated particle (Shiraiwa et al., 2010). Laborde et al. (2013) also verified accurate sizing using this method for ambient particles with <35% rBC volume fraction when compared to particle mobility diameter."

The thermodenuder's main effect is the removal of the primary coatings, which would otherwise bias the measurements. It is questionable whether "fresh, uncoated BC" exists outside of a laboratory (e.g. Toner et al., 2006; Adler et al., 2010); the thermodenued fresh BC in our paper are less coated than fresh ambient particles. We refer the reviewer to the new Section A5 (shown at the end of these comments) to see the effect the thermodenuder has on removing the primary coatings. If the reviewer was referring to charring, we have modified a sentence in the supplementary section A2 to read

"The average distribution was nearly identical through the thermodenuder, meaning charring is unlikely to have affected the particles."

5510 - 1 : How do you get from mass density to refractive index? More details are needed in this section.

We think the reviewer may have been confused by a full stop (period) that was inadvertently added during the typesetting process. The full sentence should read

"This yields a refractive index of organics of 1.46, which is in the range typically assumed for organics, though it has been shown that oxidation of organics can increase both density (Kroll et al., 2009) and refractive index (Cappa et al., 2011), with opposing effects on scattering calculations."

So we did not make a logical step from density to refractive index, we referenced two experimental studies that measured these changes.

5510 - 7-8 : How big were the changes in refractive index? Were these changes significant?

Though we previously quantified the variation using a mean and standard deviation, we have made it clearer by including the maximum and minimum values

"The refractive index for the ambient data calculated with volume mixing was 1.46 – 1.50 (mean 1.48)"

The standard deviation seems small, but there's no information provided that helps us assess whether a 0.01 change is significant or not. Most importantly, there is no work done to show the sensitivity to the coating refractive index of the derived coating thicknesses in Section 3.9. Constraining your coating fits with co-located measurements is the right thing to do, but you should also present the sensitivity to the coating refractive index and an estimate of the error on this number based on any assumed errors in the SP-AMS measurements.

We have now done this- see comment below "Discussion of uncertainty"

5510 - 24 : How do you assess "accuracy?" Use of this word implies that you know the

morphology and size of BC for certain. Supplementary Section A1 does not list "~3%" anywhere - it is listed as "~2.5%." Also, ~30% is not the statistical uncertainty but the absolute uncertainty. Statistical uncertainty is the variation over a time period, which the supplementary section quotes as ~2.5%. Absolute uncertainty is the uncertainty away from the true BC mass for any given measurement and exists because we don't know the morphology, density, and refractive index of the calibration material relative to ambient BC.
What about uncertainties from the chosen calibration materials? You should discuss the uncertainties resulting from use of glassy carbon spheres.
For the use of the term accuracy, we refer to the section below "Use of the term accuracy".
For the 3%/2.5%, we realised this was calculated incorrectly, and the actual value for D_C is $\pm^{4.5}_{4.9}\%$ which is explained in the supplementary material. Previously, it was calculated as 14%^(1/3), but the correct calculation is 1.14^(1/3) or 0.86^(1/3) converted to a percentage. 14% is the absolute uncertainty in the mass measurement, which scales to the power 1/3 when converted to D_C.
30% is the statistical uncertainty, which refers not only to variation over a time period but any set of repeated measurements that do not give precisely the same answer. If we measured the same rBC particle 10000 times, we would expect a 30% standard deviation in the measured rBC mass. When we sample enough particles that the statistical uncertainty becomes negligible, the average mass would be subject to an absolute uncertainty of 14%, and the average D_C of $\pm^{4.5}_{4.9}\%$, due to the calibration. We have made the following clarification in the text:
"As discussed in Sect. A1, the statistical uncertainty in the incandescence signal is ±30%, which corresponds to statistical uncertainty in D_C of $^{+9\%}_{-11\%}$. The accuracy of the rBC mass calibration is ±14%, which corresponds to absolute uncertainty in D_C of $^{+4.5\%}_{-4.9\%}$."
For the question regarding the calibration materials, this is addressed below in the section "Discussion of uncertainty"

5511 - 12 : Did you work out the overall uncertainty? If you did it is not clear. In supplemental Section A1, it looks like you convolve the 10% systematic difference between your SP2 and NOAA's SP2 with a 10% uncertainty based on your chosen calibration material, but you never use the 30% per-particle mass uncertainty.
This is because the per-particle uncertainty is averaged out when using a larger number of particles. We have though added this to Section 3.6
" The per-particle statistical uncertainty in rBC mass is ± 30%, (equivalent to $^{+9\%}_{-11\%}$ in D_C). As scattering scales roughly with diameter to the sixth power, this uncertainty suggests a per-particle uncertainty in E_{Sca} of $^{+70\%}_{-50\%}$ from incandesence noise."

5511 - 24 : How do Gaussian fits to the data yield accuracy and precision?
See comment below "Use of the term accuracy"

5512 - 4-5 : Again, how do we know this technique is accurate?
See comment below "Use of the term accuracy"

5512 - 8-9 : See note above for page 5492 - lines 19-20.

See response above to page 5492 - lines 19-20

5512 - 10 : The way Figure 6 is presented may be systematic in refractive index but not in density. Again, you haven't really explored sensitivity in a systematic way - these would need to be presented much like Figures 4b and 4c, where you hold one parameter constant and vary the other. You should also vary the values systematically and not just use six selected values.
Please see our response above to the reviewers first comment. This approach would not change the conclusions of the paper

How does Figure 6 look if you separate fresh and aged air masses rather than use the whole campaign?
See discussion below "Discussion of uncertainty"

5513 - 13 : The mention of CCN activity seems out-of-the-blue here. What connection does any of the rest of the manuscript have to CCN activity?
We previously mentioned this in the introduction
"BC is co-emitted with primary organic aerosol and secondary aerosol precursors such as organic gases and NO_x *which, through coagulation and condensation, form an internal mixture that increases BC's cloud nucleation activity (Khalizov et al., 2009)"*

5513 - 16 : Actually, you have not demonstrated anything about optical properties and have only pointed to another "in preparation" paper you are working on. To make this claim, you need to take your derived coating thicknesses and calculate optical properties and show the differences resulting from the different coating thicknesses retrieved.
We have changed the word "optical" to "coating"

5513 - 17 : Also, your data do not show a strong sensitivity to rBC core density.
We have changed "refractive index and density" to "parameters"

5513 - 24 : Again, you have not proven that the SP2, even with the proper input parameters, can accurately determine mixing state.
See discussion below "Use of the term accuracy"

5514 - 6-8 : This sentence presents a new idea. You should explain how a new SP2 will improve precision.
We have moved this sentence a paragraph earlier, and it now says
"On the newer D-series SP2 the single particle precision may be improved by the higher resolution, but the systematic error on the median coating properties can only be improved with a more accurate incandescence calibration."

Technical corrections
pg 5492 - line 17 : "This" means what exactly? Be more specific and precise in your wording. Below, I will list many more places in the manuscript where you used "this"

as a noun, but I may not have caught them all. Please carefully proofread your final manuscript to remove all use of "this" as a noun.
The sentence in question now reads
"This work demonstrates that using this technique the SP2 can accurately determine the average mixing state (externally or internally mixed) of ambient soot within the precision of the instrument calibration."

5493 - 12 : Is the "concentric core/shell model" the Mie model? Need a reference here.
We have clarified "the concentric core/shell Mie model" and referenced Bohren & Huffman (1983)

5493 - 20 : "Californian" should be "California"
Done

5493 - 22 : "or" should be "and"
Done

5493 - 25 : Need to add the year to the Cappa et al. reference.
That would make it "Cappa et al (2013)'s" so we've reworded the sentence
"Cappa et al. (2013) showed that even when taking into account the limitations of thermodenuders, the calculated absorption enhancements exceeded their measurements."

5495 - 11 : Should reference the CalNex overview paper, Ryerson et al., 2013. Ryerson, T. B., A. E. Andrews, W. M. Angevine, T. S. Bates, C. A. Brock, B. Cairns, R. C. Cohen, O. R. Cooper, J. A. de Gouw, F. C. Fehsenfeld, R. A. Ferrare, M. L. Fischer, R. C. Flagan, A. H. Goldstein, J. W. Hair, R. M. Hardesty, C. A. Hostetler, J. L. Jimenez, A. O. Langford, E. McCauley, S. A. McKeen, L. T. Molina, A. Nenes, S. J. Oltmans, D. D. Parrish, J. R. Pederson, R. B. Pierce, K. A. Prather, P. K. Quinn, J. H. Seinfeld, C. J. Senff, A. Sorooshian, J. Stutz, J. D. Surratt, M. Trainer, R. Volkamer, E. J. Williams, and S. C. Wofsy (2013) The 2010 California Research at the Nexus of Air Quality and Climate Change (CalNex) field study, J. Geophys. Res. Atmos., doi:10.1002/jgrd.50331.
Done

5495 - 14 : Replace "campus, Pasadena" with "campus in Pasadena"
Done

5495 - 16-17 : Is this chemical, photochemical, or physical processing?
We have specified photochemical

5495 - 26 : "This" means what exactly?
The sentence now reads
"These emissions caused short, but strong spikes in the SP2 time series, which were removed from the dataset as they were not deemed representative of the ambient aerosol"

5496 - 2 : Remove "these"

See previous comment

5496 - 4 : Strictly speaking, the PASS-3 is not a "BC instrument." Need to make the wording here more accurate.
The sentence now reads
"The University of Manchester instrumentation consisted of…."

5496 - 5 : Add the aethalometer manufacturer and model number.
We have added
"Aethalometer AE-31 (Magee Scientific, Berkeley, CA, USA)"

5496 - 9 : What is meant by "NOAA" in this sentence? Did NOAA setup and operate the inlet you tapped into, or is this a specific inlet built and used many times on NOAA field campaigns? Please clarify and add a reference if it is a specific inlet that has been used previously.
We have removed the word NOAA so it just says "a common inlet"

5496 - 17 and 19 : "PASS" should be "PASS-3." There may be other locations in the manuscript where this mistake is made.
We have changed every instance of "PASS" to "PASS-3"

5496 - 23 : "light" should be "laser"
Done

5497 - 12-14 : Be more accurate in your wording - the detectors measure single particles with a mass between those numbers you quote.
This sentence now reads
"During CalNex, the high-gain broadband detector measured particles with mass between 0.2 – 27 fg (67-305 nm mass-equivalent diameter at core density ρ_C *=1.8 gcm*$^{-3}$*) and the low-gain narrowband measured particles with mass 0.6 – 130 fg (84 – 520 nm)."*

5497 - 25 : Be more accurate in your wording - the laser vaporizes refractory species (most of which is BC).
The sentence in question reads
"rBC-containing particles absorb the infrared laser radiation and are heated to incandescence, emitting visible light."
The sentence is accurate as-is: all parts of rBC-containing particles are vaporised, and not all refractory species are vaporised (e.g. sea salt is not)

5498 - 6 : I think "a rBC" should be "an rBC"
Done

5498 - 16 : Clarify that "other components" are in the aerosol.
Done

5498 - 21-22 : Clarify this sentence - the relative concentrations are BC to organics and inorganics within the particle and are not relative in time.

The sentence now reads

"…the relative concentrations reported (i.e. the ratios of the different aerosol species)…"

5499 - 11 : "PASS" should be "PASS-3"

Done

5500 - 17 : "from" should be "for"

Done

5500 - 21 : "Whilst" should be "While"

Although "Whilst" is perfectly valid, we have changed it to "while" to benefit any non-British English readers

5501 - 8 : "there is question" should be "there is a question"

Done

5502 - 13 : What is "this" technique? Rayleigh scattering or Mie scattering? Clarify this section.

The sentence now reads

"Coating thicknesses derived using the core/shell Mie technique…"

5503 - 22 : "twin-" should be "two-"

Done

5504 - 9 : By "laser intensity" do you mean "scattered intensity"? You are considering data up to 5% of the peak scattered signal for that particle, correct?

No we mean 5% of the peak laser intensity. For particles that contain no BC, the scattering signals are Gaussian and map the Gaussian laser profile. You then use this to work out where the 5% laser power position is. See Laborde et al (2012a) (who we have referenced) for their excellent description of this calculation.

5504 - 10 : "This" means what exactly?

"This" has been replaced with *"The 5% threshold"*

5505 - 19-21 : Note that the thermodenuder settings have nothing to do with the age of the particles - need to reword this sentence and other places in the manuscript and figure captions to clarify this point.

The text is accurate as it is, and the thermodenuder temperature profile is defined in the experimental section.

5506 - 17 : "Whilst" should be "While"

Done

5507 - 2 : The scattering model is the Mie model, not the Gao model. This reference needs to be changed.
We have removed the Gao reference from this sentence

5507 - 12 : Reword the sentence so that it does not begin with "600 nm."
Changed to:
"We were unable to reliably sample calibration particles larger than 600 nm, so this size is considered the upper limit."

5507 - 17 : Isn't the "most basic product" of Mie theory the scattering cross-section? Consider rewording this sentence.
No, the scattering cross-section is measured and requires no optical model

5508 - 24 : What is a "significant change" in coating thickness? Please give numbers.
We have added a reference to Section 4 here, as the coating thickness numbers are given there

5509 - 6 : The word "encouraging" is a matter of opinion - reword this sentence to be objective.
The sentence now reads
"Using the density and refractive index recommended…."

5509 - 11 : Remove the comma after "dataset" and remove the word "likely"
Done

5509 - 15 : Remove the comma after "enhancement"
Done

5510 - 4 : "this approach" is what exactly?
We have replaced *"this approach"* with *"volume mixing"*

5510 - 18 : By "laser power" do you mean "scattered intensity"?
No, see above 5504 – 9

5511 - 14 : What does "spread" mean? Is it error, uncertainty, or width of a Gaussian fit to the data?
We have replaced the last 2 sentences in Section 3.8 as they didn't add anything to the analysis, and replaced them with
"We have attempted to minimise and quantify all sources of uncertainty. The sensitivities to the various input parameters are discussed further in the next section."

5512 - 4 : "This" means what exactly?
The sentence now starts
"The proximity of this result to externally-mixed soot demonstrates…"

5512 - 13-16 : This sentence is confusing and needs to be reworded.

The sentence in question has been lost in the revisions to section 4.

5512 - 16 : “This” means what exactly?
The sentence in question has been lost in the revisions to section 4.

5513 - 1-3 : This sentence is confusing and needs to be reworded.
We have removed this sentence, and reworded the start of the next one as
“It has consistently been observed that, in regions where…”

5513 - 13 : “This” means what exactly?
“This potential bias”

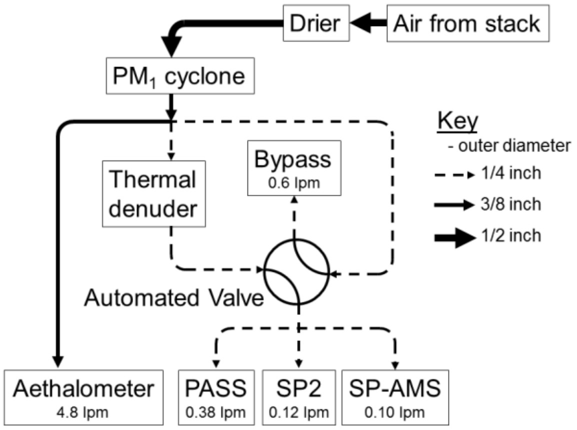
5514 - 12 : “increase” should be “increased”
Done

5514 - 14 : “refractive” should be “refractive index”
Done

5514 - 15 : Remove the comma after “datasets”
Done

5514 - 18-21 : This sentence is confusing and needs to be reworded.
We have removed this paragraph

5525 - Figure 1 : Consider using color and different types of dashed lines instead of different weights of solid black lines. The difference in 1/4 inch and 3/8 inch is especially hard to see. Also, what is “- outer diameter?”
We have changed it like this so it is clearer:



Outer diameter refers to the diameter of the outside of the tube (rather than the inside, the difference between the two being double thickness of the tube), this is standard terminology.

5526 - Figure 2 : Consider using a thick dashed while line to represent the saturation of the scattering detector; the black line can be hard to see amidst all the color.

We plotted this but it made it more difficult to see

Supplemental section comments

page 2 : How was the stability of the incandescence channel assessed during the experiment?
We have changed this to
"Repeated calibrations showed that the response of the incandescence channels was stable to within ±2% during the course of the experiment."

page 2 : Is the 30% per-particle mass uncertainty used in the assessment of errors in the rest of the paragraph?
Not in the absolute error as it averages out when you consider thousands of particles. We have made this clarification in the main text- see "Discussion of uncertainty" below

page 2 : Is the 10% uncertainty due to calibration material a systematic error as well? How do you know the diesel soot type you need to match to Is the same as in Laborde et al., 2012?
Laborde, M., P. Mertes, P. Zieger, J. Dommen, U. Baltensperger, and M. Gysel (2012) Sensitivity of the Single Particle Soot Photometer to different black carbon types, Atmos. Meas. Tech., 5, 1031–1043, doi:10.5194/amt-5-1031-2012.
We have already been clear
"Convolving this value with the 10% uncertainty due to the SP2's response to fullerene soot relative to diesel (Laborde et al., 2012a) *gives a systematic uncertainty estimate…"*
There is no evidence to suggest the SP2 responds differently to different diesel engines' emissions

page 2 : Does the systematic uncertainty of 14% mean that your SP2 is measuring 14% more per-particle mass than you believe is actually there in the ambient BC? Do you correct for this bias when determining the proper input parameters in the main manuscript? What effect would this bias have on your retrieved coating thickness?
No- the 14% is a ±14%. We have now investigated the sensitivity of the derived coatings to this uncertainty, discussed below in "Discussion of uncertainty"

page 4 : Is "average" actually "campaign average?"
We have clarified that this is the case

page 4 : What is "high-HDDV" mean?
We have clarified *"Heavy-Duty Diesel Vehicle"*

page 4 : Was rBC mass concentration ever reported in the main manuscript?
No but it was used in the supplement to compare to the EC and SP-AMS rBC concentration.

page 9 : By "enhance" do you mean that you expect to have more or fewer problems with the particle focusing in your AMS chamber?
We have clarified *"worsen any focusing problems in this study"*

page 12 : The fractional composition at higher thermodenuder temperatures is a bit concerning. It sounds like what is happening is that at higher temperatures, the rBC is changing somehow such that its not showing up in the mass spectra where it should be. If the rBC is, in fact, changing chemically, doesn't it make sense that it might be changing optically as well? If so, this could affect your "most accurate" results for fresh BC, especially because you derived this result at the highest temperature settings on the thermodenuder.

We have already noted the focusing problems and the possibility of refractory CO_2^+ mixed in with the EC within the black carbon cores (ie not a coating). We have added reference to Corbin et al (2014) who studied the refractory CO_2^+ in detail. We think also the reviewer might have thought the plots were for just the freshest airmasses when they were campaign averages, so we have added the sentence at the start of the paragraph:

"We note that this plot represents the average for the whole campaign, as the signal/noise was not high enough to split into different photochemical ages."

There is no suggestion that the BC itself is changing chemically, rather that the nonrefractory elements are being removed. We have previously noted in Section A2 that the rBC core size distribution was almost identical after passing through the thermodenuder as it was in ambient air, meaning the thermodenuder had no effect on the rBC cores, just on the nonrefractory material mixed with them.

Anonymous Referee #3

General comments:

In this paper the authors discuss the effects of assumptions of density and refractive index in the determination of thickness of rBC combining SP2 and SP-AMs data of incandescence and scattering, and applying a core/shell model and Mie model. These analyses are interesting and important in rBC retrievals. However, some aspects of the analysis and discussion are not fully explained or clear to me, and in my opinion they require a significant revision and some addition of information.

The conclusion that a given set of density and refractive index is the most appropriated is based only in the fitting data of the least coated particles. It would be interesting to add a discussion about the sensitivity to density and refractive index in the case of the most coated particles. A diagram showing the products obtained and the stages of this analysis, including the experimental phase and modeling, would benefit this work and help the understanding of the applied method. The authors should also avoid the excessive use of parenthesis along the text.

We thank the reviewer for their comments.

Regarding the sensitivity of the most coated particles to density and refractive index, we have included a comparison in Figure 6, discussed further below in "Discussion of uncertainty". We didn't want to compare all the possible values because the plot would be too messy. We have also explored the sensitivity of the median coatings to a number of parameters, including core density and refractive index, in Table 2. This table is also discussed below in "Discussion of uncertainty"

A diagram of the process is an excellent suggestion, and we have now added this as the new Figure 2:

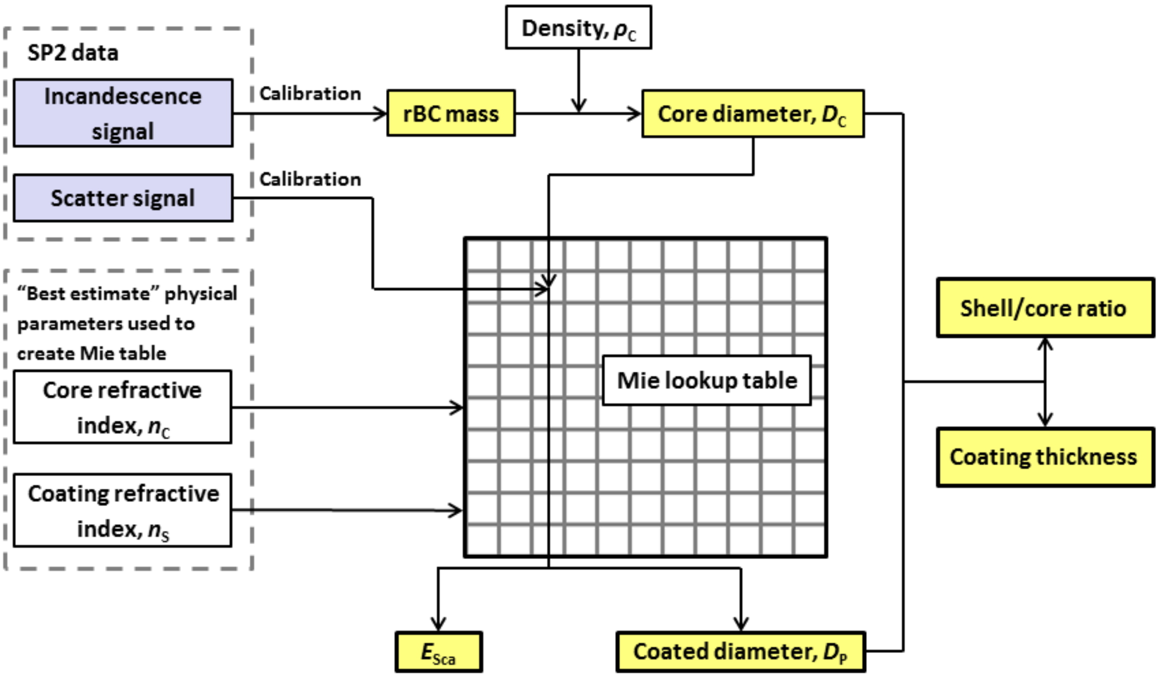


Figure 2 - Overview of the input data/parameters and steps involved in calculating coating properties using SP2 data. Blue boxes represent the raw measured data, white boxes represent assumed parameters, and yellow boxes represent derived parameters.

Specific comments:

Page 5492, Line 2: "The optical properties and cloud condensation nuclei (CCN) activity of soot depend on the amounts (both relative and absolute) of BC and nonrefractory material in the particles." , you also wrote on Page 5494, Line 48, that "SP2 quantifies refractory black carbon (rBC) mass...". It might be good to briefly mention what is refractory black carbon and how it is relate to the total black carbon content in soot.

We have added this sentence to Section 2.3:

"rBC here is defined as the fraction of a soot particle that is thermally-stable enough to incandesce before vaporizing (Petzold et al., 2013). Though sp^2 carbon bonds are linked with both thermal stability and light absorption, it is not clear whether any non-carbon fraction of BC (such as refractory oxygen species) is included in rBC."

Also, you should make clear in the text what are the assumptions in the distribution of refractory black carbon inside the particles for the shell/core model, and for each one of the techniques you used: SP2, SP-AMS, and thermal denuder. For instance, when you say that SP2 measures only refractory BC, are you assuming that rBC is present in the shell, core, or both?

We have clarified in the introduction regarding the core/shell model

"…focusing light onto the BC core"

We have already stated *"The amount of light detected is linearly proportional to the mass of black carbon in a particle, regardless of particle coatings or morphology"* so it is clear what the SP2 detects.

We have clarified for the SP-AMS

"…selectively vaporize BC-containing particles and measure their average composition as an ensemble"

For the thermal denuder, no clarification is needed as it doesn't measure anything.

Page 5492, Line 17: "This demonstrates that using this technique the SP2 can accurately determine the mixing state (externally or internally mixed) of ambient soot ", Your previous sentence cannot demonstrate what you are claiming. Discuss why or how this is demonstrated?

See discussion below "Use of the term accuracy"

Page 5496: Line 18: "From 18 May 2010, the SP2, PASS and SP-AMS sampled through a line, alternating every 10 min between an ambient inlet and a line drawn through a thermodenuder" . What is the importance of this date in the whole campaign? Were all data presented in this study taken under the same condition or not? Clarify it.

This is just when we set up the thermodenuder. We have added an additional sentence afterwards

"Before this date, only the ambient inlet was used."

Page 5497, Line 26: "During CalNex, the tungsten vaporizer was removed and only BC-containing particles were vaporized". It is not clear why the tungsten vaporizer was

removed and neither why you say that "only BC-containing particles were vaporized". What is the temperature that the particles reach in this case? Why do you consider that the part analyzed is only BC-containing?

We removed the vaporiser so only BC-containing particles were vaporised. In other words, if a particle containing BC entered the laser it was vaporised (in its entirety). If a particle containing no BC entered the laser it was not vaporised. The removal of the tungsten vaporiser eliminates any interference from particles containing no BC.

We have modified the text slightly

"the tungsten vaporizer was removed and so only BC-containing particles were vaporized"

Page 5498, Line 29: "For the purposes of estimating the shell refractive index (ns), the reported mass fractions are used." What are the "mass fractions" you refer to? Where did you report them?

We have clarified

"the relative mass fractions of the non-rBC SP-AMS species were used"

Page 5501, Line1: "the data presented here were taken after 26 May 2010, when the SP2's detectors were re-aligned." Do you have data from the period where the SPS's detector was not aligned? What is the relevance of this sentence in the context?

We do have data from before the realignment but we haven't used it as the detector was not properly aligned. We have moved this sentence to Section 2.3 discussing the SP2.

Page 5501, Line 23: "this regime is appropriated for externally-mixed BC (i.e. with a single refractive index)". Add at least one more sentence about the meaning of externally-mixed BC and why it is appropriated in this case.

The text says the regime is appropriate, not appropriated.

This now reads

"For a wavelength of 1064 nm used in the SP2, this regime is appropriate for externally-mixed BC <10^{-20} m^3 in volume (equivalent to 267 nm diameter) (Moteki et al., 2010). Externally-mixed BC here means particles containing only BC- the mixing of more than one compound within single particles requires a distribution of refractive index, which is not accounted for in the simple Rayleigh-Debye-Gans approximation."

Page 5503, Line 3: "The refractive index of the coating was calculated using the chemical composition of the coating material measured by the SP-AMS." In the section 2.4 (Page 5497, Line 25) the authors also state " SP-AMS [: : :] selectively vaporize BC–containing particles and measure the composition of core and coating". Did you measure composition of core and coating separately? How were these measurements done? What were the results obtained in each case?

We have clarified (as discussed above)

"*selectively vaporize BC-containing particles and measure their average composition as an ensemble"*

Page 5503, Line 10: "By examining the size dependence of these factors, a range of core diameters for which the scattering data was considered reliable was identified." What criteria were used in this identification?

The criteria are discussed in some detail in Section 3.4- we have added a “see Sect. 3.4” to the end of this sentence.

Page 5503, Line 3: “: : :and allowed to vary during the course of the campaign”. What was allowed to vary? How?
We have reworded this sentence
“The refractive index of the coating was calculated using the time-dependent chemical composition of the coating material measured by the SP-AMS”

Page 5503, Line 4: “A number of look up tables were generated for a range of different core and coating refractive indices, and thermally denuded data was used to constrain the most appropriate core density and refractive index, such that the measured scattering matched the modelled scattering if the cores were assumed to be uncoated”. This description is not sufficient for the understanding of your procedure. You might want to say (as a minimum) why and how the data of denuded particles were used to constrain the core density and refractive index. It is not clear how “the measured scattering matched the modelled scattering if the cores were assumed to be uncoated.”
The paragraph in question is in a section titled “Overview of technique”. We have discussed in great detail how we matched the scattering data to calculated scattering by uncoated cores in the following sections.

Page 5504, Line 26: “In this analysis we avoid this issue by choosing the core diameter range in which particle coatings are considered such that these particles only make up a small fraction of particles, and do not affect derived coating properties. ” Even though you say that this will be better explained in the next section, some more elaboration is needed in this paragraph.
It makes no sense to repeat ourselves- we have referenced the next section, in which we state in some detail how this works

Page 5507, Line: 11: Please justify why you chose 80nm as minimum as well.
We have added
“The 80 nm lower limit was chosen as it is close to the lower cutoff diameter of the incandescence measurement”

Page 5509, Line 21: What was assumed for the imaginary part of the refractive index? Why? How important is the refractive index of the coating in your analysis?
We have added just before this sentence
“n_S has only a real component at 1064 nm, as any coating material found in the atmosphere is nonabsorbing at this wavelength.”
For a discussion of the impact of the shell refractive index, see below “Discussion of uncertainty”

Page 5511, Line 12: “For the purposes of this work it is most important to work out the overall uncertainty, not diagnose different sources of it.” I disagree with this point of view. Diagnoses of different sources of uncertainty are essential to identify possible systematic errors.

We have removed this paragraph. See “Discussion of uncertainty” below

Fig 3: The plot is missing the blue continuous line and it has two blue dashed lines.
Actually its that the solid blue and dashed green lines overlap, so it looks like a solid green and dashed blue. We have added to the legend *“The solid blue line and dashed green line overlap.”* but would like to hear the editor’s advice on making this more clear.

Fig 4: Why don’t you apply the sensitivity study for a case of most coated particles?
We have done this now, see “Discussion of uncertainty” below

Technical corrections:
Page 5493, Line 17: Consider replacing “, but” by “ . In opposition, : : :.results:”
Done, but instead of “In opposition,” we used “However,”

Page 5495, Line 22: Consider replacing “: : :emissions, and biomass” by “ : : :emissions. Biomass burning”,
Done

Page 5496, Line 10: Consider writing the meaning of “a.g.l.” (above ground level?).
Done

Page 5498, Line 5: Define “m/z” channels.
Done

Page 5498, Line 27: Explain secondary and primary coatings
This is now 2 sentences
“The concentrations of coating materials relative to each other are likely to be more accurate, but it is likely to be biased slightly towards secondary rather than primary coatings. Primary coatings, such as those co-emitted with the BC, will be prevalent on the missed particles and be almost purely organic as opposed to secondary coatings (those formed after emission) which are more likely to be mixed organic and inorganic.”

Page 5500, Line 5: “the variable collection efficiency of the SP-AMS complicated factors”. Please be more specific about what do you mean.
Complicated factors -> *“influenced some of the measurements”*

Page 5500, Line 11: define “PANs”.
Done

Page 5501, Line 15: define “SOA”.
Done

Page 5501, Line 24: “: : :this regime is appropriate for externally-mixed BC (i.e. with a single refractive index) < 10-20m3 in volume (equivalent to 267 nm diameter).” The text above is confusing. The authors might consider removing the excess of parentheses

and rewrite the information clearer.
"*For a wavelength of 1064 nm used in the SP2, this regime is appropriate for externally-mixed BC* $<10^{-20}$ m^3 *in volume (equivalent to 267 nm diameter) (Moteki et al., 2010). Externally-mixed BC here means particles containing only BC- the mixing of more than one compound within single particles requires a distribution of refractive index, which is not accounted for in the simple Rayleigh-Debye-Gans approximation.*"

Page 5502, Line 16: Specify what "this" refers to.
This -> "this amount"

Page 5510, Line 3: Please remove the "." after "refractive index".
Done, this made its way in during the typesetting process

Page 5511, Line 21: Explain where the range was discussed.
We have added "in Sect. 3.4"

Page 5509, Line 17: separately.
Done

Fig. 4: Consider changing the legend entry "Recommended by review paper". You can add the references from where you took the values instead.
Done

Discussion of uncertainty

Several of the reviewers' comments wished to see more detail discussing our handling of errors, the sensitivity to coating refractive index and to our calibrations. Our thinking behind not including the discussion to coating refractive index was that, based on previous work (Shiraiwa et al., 2008, which we cited), it was not thought to be a large source of error, and one that we had effectively eliminated by calculating it from the SP-AMS data.
We have performed a more sophisticated analysis, making clearer the distinction between per-particle statistical errors and systematic errors in the median coatings, which are not affected by the statistical errors as long as a sufficiently large number of particles are measured. We have explicitly investigated the sensitivities to the instrument calibrations, and to coating refractive index. We have made changes to Section 3.8, Section 4, Figure 6, and added Table 2 summarising our main results. We have edited parts of the abstract/conclusions to reflect the changes made to section 4 and also added a section in the supplementary material section A5 on the use of the term accuracy. A revised version of each of these sections is provided below.

“3.8 Sources of uncertainty

Coatings reported for individual particles are subject to both systematic and random errors, whereas average coating properties are subject only to systematic errors as long as enough particles are sampled. As discussed in Sect. A1, the statistical uncertainty in the incandescence signal is ±30%, which corresponds to statistical uncertainty in D_C of $^{+9\%}_{-11\%}$. The accuracy of the rBC mass calibration is ±14%, which corresponds to absolute uncertainty in D_C of $^{+4.5\%}_{-4.9\%}$.Calibrations of the optical size of particles using 200 nm PSLs showed the per-particle uncertainty was ±30%, but increased to ±40% when using LEO fits due to the decreased signal. After the TEAPD was realigned on 26 May, the modal signal of 200 nm PSL calibrations throughout the campaign were all within ±3%, demonstrating the stability of the laser power and scattering detectors. The per-particle uncertainty of the scattering and incandescence measurements causes the spread in E_{Sca} seen in Figure 5 and, similarly, a width to the distribution of reported coatings.

The uncertainties in the calibration values are due to variation between the mean values of multiple calibrations, and the different instrument response between ambient soot and glassy carbon. These uncertainties represent an estimate of possible systematic errors in the measurements. Other possible systematic uncertainties relate to the core properties n_C and ρ_C, which it is the purpose of this paper to investigate, as well as n_S, though this sensitivity is thought to be small (Shiraiwa et al., 2008). In Section 4.1, we quantify the effect each of these parameters has on the derived coatings.

The FWHM and notch position have minimal size-dependence for small particles, but for larger particles some variation is expected as these particles take longer to traverse the laser beam. However, 200 nm and 300 nm PSLs show similar distributions of FWHM and notch position, meaning for particles <300 nm in total diameter (which represent the vast majority of rBC number) this sensitivity is small. The optical size of particles could be sensitive to the fraction of peak laser power used to generate the LEO fits, however Laborde et al. (2012) demonstrated the scattering cross-section of coated particles is stable before they start to evaporate in the laser. As discussed in Sect. ***Error! Reference source not found.****, inspection of single particles showed that 5% of the peak laser power was within this plateau.*

Scattering at 1064 nm is not thought to be sensitive to particle shape for the sizes of externally-mixed particles considered here. For coated rBC particles, shape may affect scattered light intensity but, as we have already discussed, when soot is not in a core/shell configuration the derived coating thicknesses are only a qualitative indicator of mixing state.

We have attempted to minimize and quantify all sources of uncertainty. The sensitivities to the various input parameters are discussed further in the next section.

4. The effect of core density and refractive index on derived coating thickness

4.1 Systematic uncertainties

Using the coating refractive index calculated from the SP-AMS, the absolute and relative coating thicknesses of BC-containing particles were calculated for particles with rBC core diameter in the range discussed previously in Sect. 3.4. Figure 1 shows the distributions of coating thickness under the same conditions as ***Error! Reference source not found.****, using the combinations of core density and refractive index that resulted in the smallest and largest* E_{Sca} *in* ***Error! Reference source not found.****.*

Gaussian fits to the least coated distributions yield the single-particle precision. Using a core density $\rho_C = 1.8$ *g cm*$^{-3}$ *and refractive index* $n_C = (2.26 - 1.26i)$ *generated a distribution of shell/core ratio with mean of 1.04 and standard deviation of 0.21, corresponding to an absolute coating thickness distribution with mean of 2 nm and standard deviation of 17 nm. Here, the mean and standard deviation are estimated from Gaussian fits to the data so they are not biased by particles with* $E_{Sca} < 1$*. For comparison, using a density* $\rho_C = 2$ *gcm*$^{-3}$ *and refractive index* $n_C = (1.76 - 0.44i)$ *generated a distribution of shell/core ratio with mean of 1.40 and standard deviation of 0.19, corresponding to an absolute coating thickness distribution with mean of 30 nm and standard deviation of 15 nm. In Figure 1, the difference in coatings using the different core parameters is larger than the difference between fresh, thermodenuded and aged particles.*

The SP2 is capable of measuring a large number of particles rapidly and therefore can obtain data with good counting statistics. The distributions shown in Figure 1 were generated using many thousands of particles. It is most common to use the median values of the properties of the coatings to measure changes in BC mixing state. When taking the median of a sufficient number of particles, the precision of any individual particle becomes unimportant. Only systematic errors can skew the median coating properties.

In order to assess possible systematic uncertainties in the calculated median coatings, we systematically varied each parameter involved in the calculation. The results of these calculations are shown in Table 2. The standard parameters were $n_C = (2.26 - 1.26i)$*,* $\rho_C = 1.8$ *gcm*$^{-3}$ *and time-dependent* n_S *calculated from the SP-AMS composition data. It is clear by comparing the results of changes in core density and refractive index that the differences in the latter were responsible for the majority of the difference between the red and black traces in Figure 1. However, by choosing the* *most appropriate values based on the results of Sect. 3.6, we have, as much as is possible, eliminated this sensitivity as a possible source of systematic error.*

Though we have used the SP-AMS composition to calculate a time-dependent n_S to try to eliminate any bias this parameter may have, this measurement is not always available. We therefore performed the same calculations, using n_S of 1.45 and 1.5, which span the range calculated using the SP-AMS data, and 1.59, the refractive index of PSLs. The results of these calculations are shown in Table 2. This sensitivity was small- the maximum change was a shift in shell/core of 0.02 (corresponding to 1 nm absolute coating thickness).

The only systematic uncertainties we were not able to effectively eliminate were the uncertainties associated with the calibration of the core diameter ($^{+4.5\%}_{-4.9\%}$) and the scattering detector ($\pm 3\%$). To investigate this uncertainty, we calculated the median coatings using the same particles as those in Figure 1, but with calibrations 1 standard error away from the mean. Changing the scattering calibration by $\pm 3\%$ caused a 0.01 shift in median shell/core ratio, and a corresponding 1 nm shift in coating thickness. Changing the D_C calibration by $^{+4.5\%}_{-4.9\%}$ caused a ~0.11 shift in shell/core ratio, and a corresponding ~8 nm shift in absolute coating thickness. With the increased D_C, the median E_{Sca} was 0.81, but the coatings were assigned to that of externally-mixed particles. When combining this value with the scattering uncertainty in quadrature, the scattering uncertainty is insignificant, and the systematic uncertainty associated with the instrument calibration is the same as the uncertainty associated with the D_C calibration.

In the least coated conditions, the median measured shell/core ratio was 1.02 ± 0.11, corresponding to an absolute coating thickness of 2 ± 8 nm. The proximity of this result to externally-mixed soot demonstrates that when using these parameters for density and refractive index, this technique can accurately determine the median mixing state (externally vs internally mixed) of ambient BC-containing particles within the precision of the instrument calibration. We note however that the precision will depend on the specific setup of the instrument (e.g. alignment, detector gain settings). If used with different ρ_C and n_C the precision is similar, as is the uncertainty associated with the instrument calibrations, but the average values would not be accurate for ambient soot. Using the same least-coated measured data, but assuming ρ_C = 2 gcm^{-3} and refractive index $n_C = (1.76 - 0.44i)$, the median shell/core ratio was 1.39 ± 0.11, and absolute coating thickness 30 ± 8 nm. Here the uncertainties are those associated with the instrument calibration, but they do not represent the systematic uncertainty due to the use of the different values for ρ_C and n_C. The uncertainty associated with the core refractive index is larger than any of the other sensitivities listed in Table 2.

4.2 Time-dependent variation

***Error! Reference source not found.** shows the effect of systematic offsets associated with n_C and ρ_C on the measured shell/core ratios over the whole campaign. The density and refractive index recommended by Moteki et al. (2010) were shown in Sect. **Error! Reference source not found.** to be the most appropriate for this dataset and result in the lowest coating thicknesses. This result emphasises that comparisons between different data sets can show differences that can be significantly influenced by the choice of core parameters and misrepresent any underlying similarity or difference between the data sets.*

While the use of different core density and refractive index values may systematically change reported coating parameters, there is also variation in coatings due to real changes in the ambient particles. ***Error! Reference source not found.*** *compares changes in the median shell/core ratio, using the refractive index and density combinations that result in the thickest and thinnest coatings, as a function of photochemical ageing. Changes in coatings were captured regardless of the parameters used, but there was a significant offset between the data using the different sets of density and refractive index of the rBC core. The difference between the minimum and maximum coating thicknesses reported does have a dependence on these parameters, but this range is small compared to the difference in the absolute values. When using the density and refractive index recommended by Moteki et al. (2010), in the 'most coated conditions' of Figure 1, the median shell/core ratio was 1.28 ± 0.11, though there were some data periods on Fig. 8 with values up to ~1.5 ± 0.11. These values are several times larger than the systematic uncertainty associated with the instrument calibration, meaning the SP2 is able to resolve absolute differences in mixing state, though for coated particles there may also be an additional uncertainty associated with the morphology of internally-mixed soot particles (Liu et al., 2015).*

Coatings were thickest in the most processed airmasses, similar to results reported from urban plumes in the Western Pacific, (Moteki et al., 2007; Shiraiwa et al., 2007) and in California, including some in the Los Angeles area (Sahu et al., 2012). It has consistently been observed that, in regions where secondary aerosols also constitute a large fraction of submicron particulate matter, condensation of secondary material increases BC coating thicknesses with age. Even using the core parameters that result in the thinnest coatings, the freshest ambient BC-containing particles were also associated with some coating material.

Coatings measured through the thermodenuder were thinner than ambient coatings, though this difference was smallest in the least processed airmasses as the ambient particles were less internally-mixed. When the core parameters that result in the thickest coatings were applied, even the thermodenuded particles appeared to have significant coating material. Taking these data in isolation could therefore lead to misleading conclusions about the optical properties of BC and its CCN activity. This potential bias must be considered if BC coating data generated in this way are to be used quantitatively.

Conclusions

We have demonstrated that coating properties of black carbon particles derived by core/shell modelling of SP2 data show a strong sensitivity to assumed parameters of the rBC core. For a given measured scattering signal and core mass, a larger refractive index (real and imaginary) and smaller core density result in a larger scattering cross section due to the core, meaning less is attributed to a coating. This shift results in thinner calculated coating thicknesses for the same measurement data.

An assessment of the coating properties of freshly-emitted, thermodenuded ambient particles demonstrated that, when using the appropriate core density and refractive index, the SP2 incandescence/scattering technique can accurately determine the mixing state (externally or internally mixed) of ambient rBC within the precision of the instrument calibration. Using a core density of 1.8 gcm^{-3} (Bond and Bergstrom, 2006) and refractive index of (2.26 – 1.26i) (Moteki et al., 2010) generated a distribution with median shell/core ratio of 1.02 ± 0.11 corresponding to an absolute coating thickness distribution of 2 ± 8nm. The main source of statistical error in the *measurement of single particles was random variation of the incandescence measurement. Other than the sensitivity to core refractive index, the incandescence calibration was the main source of uncertainty in the average coatings. The refractive index of coatings was found to have only a minor influence. On the newer D-series SP2 the single particle precision may be improved by the higher resolution, but the systematic error on the median coating properties can only be improved with a more accurate incandescence calibration.*

The effective density of soot particles is subject to considerable variability due to complex, varying soot morphology in different atmospheric conditions. However, at a near-infrared wavelength of 1064 nm, light scattering is less sensitive to the particle geometry as the wavelength is large compared to the measured size of rBC. The bulk density of black carbon of 1.8 g cm^{-3} *as suggested by Bond and Bergstrom (2006) is appropriate to derive the rBC volume based on the direct mass measurement from the SP2. However, the use of the bulk density may not be appropriate at visible wavelengths, where shape has much more of an influence on optical properties. The core refractive index measured using the SP2 (Moteki et al. 2010) is larger than that recommended by Bond and Bergstrom (2006) for use at visible wavelengths. The wavelength-dependence of black carbon's refractive index is highly uncertain, and caution should be taken when using this value of BC refractive index for optical calculations at different wavelengths.*

The use of different core parameters caused a systematic difference in the reported coating data. The majority of these differences were due to the change in the refractive index of the core. Calculations performed using the same measured data, but with the core parameters that resulted in the thickest coatings generated a median shell/core ratio of 1.39 ± 0.11, corresponding to an absolute coating thickness of 30 ± 8 nm. Here, the quoted errors are due to the instrument calibration, and do not capture the systematic uncertainty associated with the use of the different core refractive index and density.

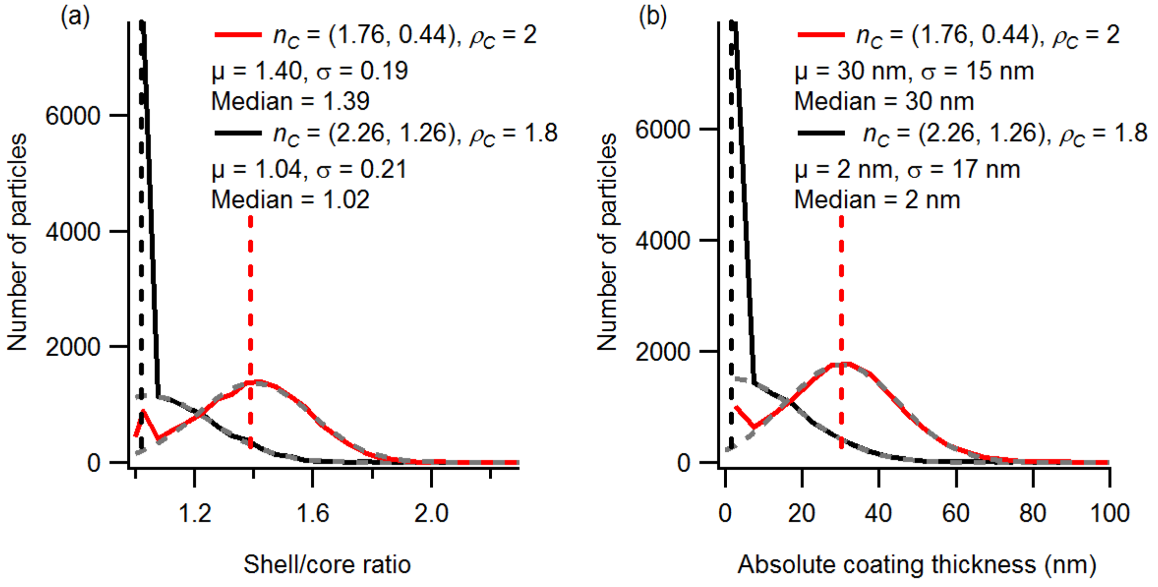
Temporal changes in coating thickness were also measured, which were several times larger than the uncertainty associated with the instrument calibration. Coatings were thickest in the most processed airmasses. This change is similar to previous results from California and the Western Pacific, where a large fraction of submicron aerosol is also secondary, and suggests a consistent picture of increased coating by condensation of secondary aerosol. While these relative changes in coating thickness were captured well regardless of the density and refractive index used in the Mie model, the sensitivity to assumed core properties must be taken into account when comparing different datasets or when using SP2 coating data to compare with large scale models. Omitting information of this sensitivity could lead to misleading conclusions regarding the optical properties of BC and its CCN activity.

We have determined the most appropriate values of BC density and refractive index to use to measure mixing state at 1064 nm where particle morphology has only a minor effect, but appropriate values to use for optical calculations of nonspherical particles at visible wavelengths may be different. At present, there are significant uncertainties in the absolute values of the refractive index and density of BC particles. Without similar constraints as those provided here, constraining the behaviour of BC particles in models using field data will be subject to large systematic measurement uncertainties.

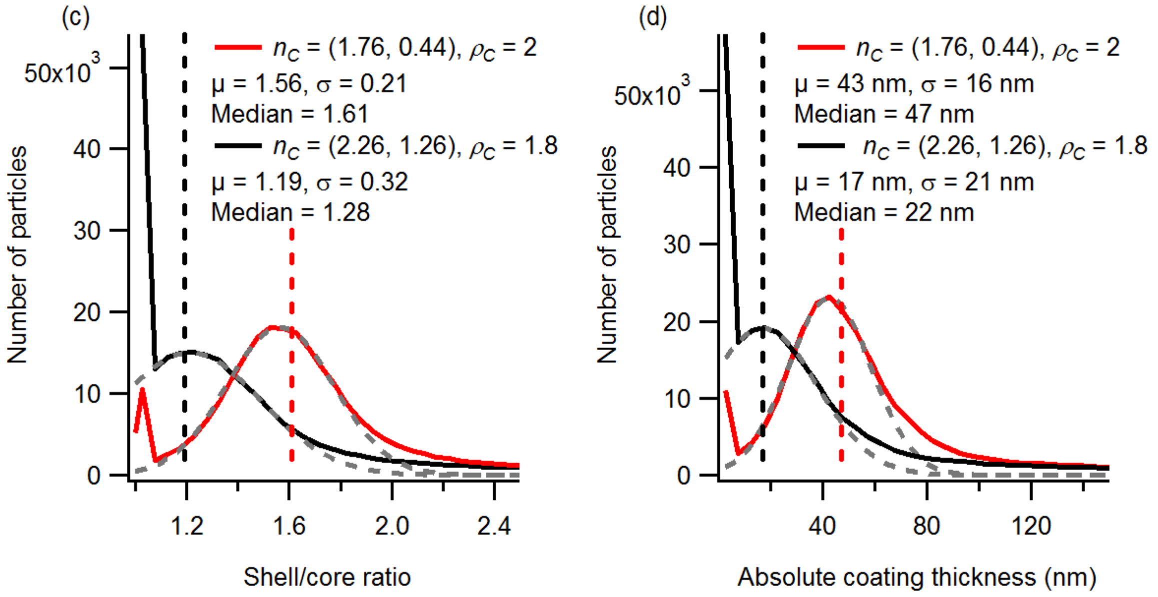
Table 2 - Median shell/core ratios and absolute coating thicknesses with varied assumptions about ρ_C, n_C and n_S and varied instrument calibrations. All other parameters were held at their best estimate values, described in the text. For clarity, increasing the scattering calibration refers to increasing the signal of a 200nm PSL used to scale the Mie table, whereas increasing the D_C calibration refers to increasing D_C.

	Shell/core ratio		***Coating thickness***	
	Least coated	*Most coated*	*Least coated*	*Most coated*
Best estimate	*1.02*	*1.28*	*2*	*22*
n_C				
$(2-1i)$	*1.12*	*1.39*	*10*	*31*
$(1.95-0.79i)$	*1.20*	*1.45*	*15*	*35*
$(1.87-0.56i)$	*1.27*	*1.50*	*22*	*40*
$(1.76-0.44i)$	*1.33*	*1.54*	*26*	*43*
ρ_C				
1.7 gcm^{-3}	*1*	*1.24*	*0*	*19*
1.9 gcm^{-3}	*1.06*	*1.33*	*5*	*25*
2.0 gcm^{-3}	*1.10*	*1.37*	*7*	*28*
n_S				
1.45	*1.02*	*1.29*	*2*	*23*
1.5	*1.02*	*1.28*	*2*	*22*
1.59	*1.02*	*1.26*	*1*	*21*
Scattering calibration				
3%	*1.01*	*1.27*	*1*	*22*
-3%	*1.03*	*1.29*	*2*	*23*
D_C calibration				
+ 4.5%	*1*	*1.18*	*0*	*15*
- 4.9%	*1.13*	*1.40*	*10*	*30*

Freshest airmasses, thermodenuded >220°C



Most aged airmasses



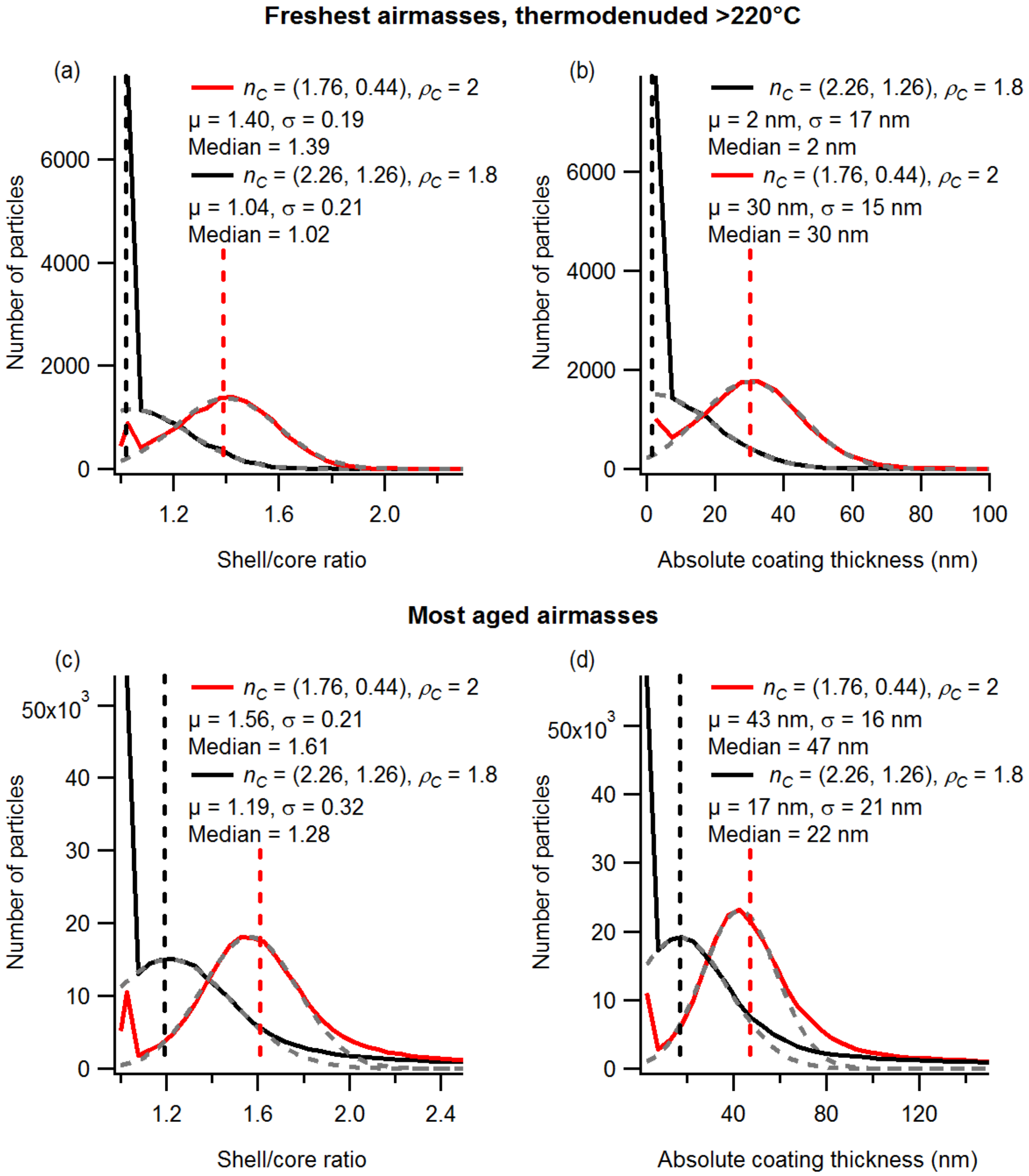


Figure 1 - Distributions of (a & b) relative and (c & d) absolute coating thickness, measured under the same fresh and aged conditions as Figure 2. The dashed grey lines are Gaussian fits to the primary modes, and fit parameters are listed in the legend. The first bin represents all particles with $E_{Sca} \leq 1$, *and this bin was not used in the Gaussian fits. The vertical dashed lines are the median values, also listed in the legend. These are often used to compare changes in coatings and are only biased by particles with* $E_{Sca} < 1$ *if the median value falls in this range.*

New abstract:

Black carbon (BC) is the dominant absorbing aerosol in the atmosphere, and plays an important role in climate and human health. The optical properties and cloud condensation nuclei (CCN) activity of soot depend on the amounts (both relative and absolute) of BC and non-refractory material in the particles. Mixing between these two components is often represented in models by a core/shell coated sphere. The single-particle soot photometer (SP2) is one of, if not the only, instrument capable of reporting distributions of both core size and coating thickness. Most studies combine the SP2's incandescence and 1064 nm scattering data to report coating properties, but to date there is no consistency in the assumed values of density and refractive index of the core that are used in these calculations, which can greatly affect the reported parameters such as coating thickness. Given such data are providing an important constraint for model comparisons and comparison between large datasets it is important that this lack of consistency is addressed.

In this study we explore the sensitivity of the reported coatings to these parameters. An assessment of the coating properties of freshly-emitted, thermodenuded ambient particles demonstrated that a core density of 1.8 gcm^{-3} *and refractive index of (2.26 - 1.26i) were the most appropriate to use with ambient soot in the Los Angeles area. Using these parameters generated a distribution with median shell/core ratio of 1.02 ± 0.11, corresponding to a median absolute coating thickness of 2 ± 8 nm. The main source of statistical error in the single-particle data was random variation in the incandescence signals. Other than the sensitivity to core refractive index, the incandescence calibration was the main source of uncertainty when optically determining the average coatings. The refractive index of coatings was found to have only a minor influence.*

This work demonstrates that using this technique the SP2 can accurately determine the average mixing state (externally or internally mixed) of ambient soot within the precision of the instrument calibration. Ambient coatings were measured up to a median shell/core ratio of 1.50 ± 0.11, meaning this technique is able to resolve absolute changes in mixing state.

However, when different core parameters were used, the core/shell ratio and the coating thickness were shown to be offset by amounts that could be larger than the atmospheric variability in these parameters, though the results have a similar precision. For comparison, using the core parameters that resulted in the thickest coatings (on the same thermodenuded fresh particles as before) generated a median shell/core ratio of 1.39 ± 0.11, corresponding to a median absolute coating thickness of 30 ± 8 nm. These results must be taken into account when comparing BC coatings measured using this technique, or if using these data for optical or CCN calculations.

We have determined the most appropriate values of BC density and refractive index to use to measure mixing state at 1064 nm where particle morphology has only a minor effect, but appropriate values to use for optical calculations of nonspherical particles at visible wavelengths will also be subject to similar, significant uncertainties. Without similar constraints as those provided here, constraining the behaviour of BC particles in models using field data will be subject to large systematic measurement uncertainties.

Use of the term 'accuracy'

Several comments mentioned the use of the term "accuracy" when referring to our results from the least-coated conditions. The main question was how we were sure our BC was externally-mixed (i.e. containing only BC), and so provided a robust test of our technique. We have added the following section to the supplementary material, which is referenced in Section 3.4 of the main text:

"A5. Removal of primary coatings

Figure A7 shows a thermogram of the median E_{Sca} *taken under the freshest ambient conditions. For temperatures <180°C, there is a trend of decreasing* E_{Sca} *with increasing temperature, suggesting the presence of some primary coating that was removed at higher temperatures. However, above 180°C there is no significant trend, meaning above this temperature, any primary coatings were removed, leaving BC that could reasonably be considered externally-mixed. By considering only data the freshest airmasses, we minimise the effects of any coating material the thermodenuder would not be able to remove.*

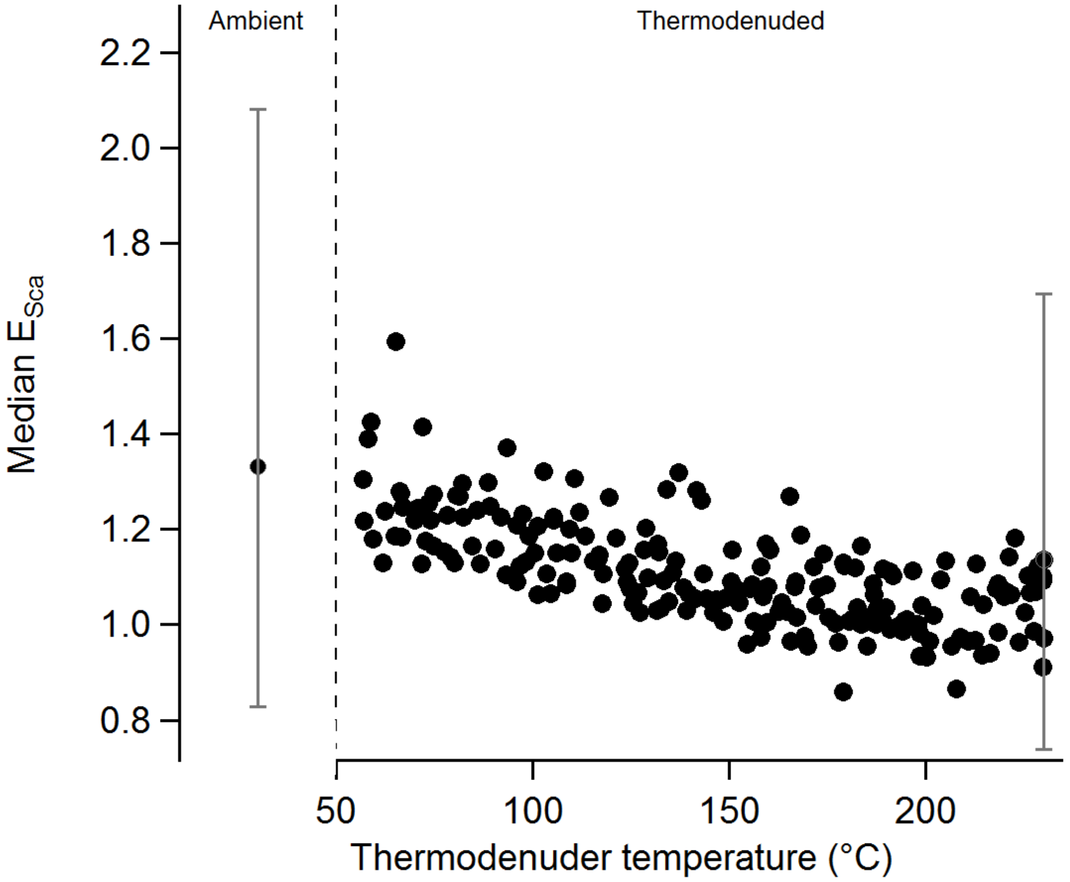


Figure A7- Median E_{Sca}*, from the freshest ambient airmasses, as a function of thermodenuder temperature. The average for ambient data in the same atmospheric conditions is shown for comparison. The data were calculated using, calculated using* $n_C = (2.26 - 1.26i)$ *and* $\rho_C = 1.8\ gcm^{-3}$

and are only take from particles with 135 ≤ D_C ≤ 200 nm. The data are 5-minute averages, and are all taken from the periods defined as the freshest ambient conditions ($-\log(\mathrm{NO_X}/\mathrm{NO_Y})$ *< 10th percentile). The grey error bars show the 25th and 75th percentiles of selected data points, to demonstrate the spread in the data."*

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