Response to Referee 3

We thank the referee for their comments.

The referee makes a general criticism to our introduction and seems to think that it is unclear. Referee 2 on the other hand has positive comments on the introduction and we believe that it is important to retain key background information. This is a comment on style and is as much about personal taste rather than content. However, we will review the introduction and include some discussion on the size distribution in the revised manuscript.

The following details our responses to the specific points raised by the referee.

Lines 50-54: is the MAC value(and those it is compared to) appropriate for all the air sampled, or only the FT air?

Please refer to the answer to the comment of referee 1 [Page 8777: Line 24].

- Line 64: The statement that non-refractory materials are thickly coated "around the BC core" is likely overly specific. The incandescence method used cannot determine physical arrangement.

"Around the BC core" will been removed in the revised manuscript.

- Line 66: The term "enhancing BC mixing state" is not clear, but I believe the point of this sentence is not explicitly stated: as it is self-evident that condensation will lead to decreasing BC mass fraction in particles, I read this as an indication that, in the air masses sampled, condensation was a more significant route than coagulation or cloud processing in accomplishing this decrease. A similar sentence should be revised in the conclusion/summary.

The text will be amended in the revised manuscript.

-BC aerosol is removed through wet and dry deposition: i.e., it is not an "ideal tracer". Please correct the discussion

It will be corrected.

- - The introduction, which may be too far ranging, includes brief description of many questions "related to the size distribution and mixing state of BC". What is the subset of these are addressed in this paper? (Line 129). Providing an answer in the text is a step to addressing my larger comments above. - The introduction did not include much discussion of the importance of the BC size distribution – what is it?

The referee 2 has positive comments on the introduction and we believe that it is important to retain key background information. However, we will review the

introduction and include some discussion on the size distribution in the revised manuscript.

-Line 203: Please add "mixed" to the appropriate extended weather type in table 1.

Corrected.

-Line 236: is the laser intensity measured here, or is this the value published in Stephens et al? Please clarify in the text.

The position of the reference Stephens et al. will be changed.

Discussion of lines 240-252: if any strong absorber (line 240) emits thermal radiation, why is this sufficient to identify particles as BC or not (line 252)?

This was discussed clearly in the ACPD manuscript on pg.8772 lines 20-25: "Incandescence radiation is measured using two detectors, one has a broad bandwidth and the other is set at a narrow bandwidth close to the maximum of the blackbody radiation emitted by incandescing rBC. The ratio of signals from these two detectors provides information on the temperature of the incandescing particle and hence its composition.", and was also discussed on pg.8773 line14-16: "The composition of the absorbing component of ambient particles was determined from the ratio of the two incandescence signals, in turn rBC particles were identified."

L252: Calling a particle "BC" when an incandescence signal was observed suggests that the mass of the particle is then the mass of BC, which is not the case. I suggest calling these particles by a more general term (e.g., BC-containing) when specifically referring to an internally mixed particle containing BC . Then when you refer to "BC", it will be clear that you refer only to this component of the aerosol. (The discussion later in the paper of "when a particle has an absorbing core" is what brought this to my attention).

Please refer to our response to the first comment of referee 1.

Line 263: what was the minimum calibration mass used? What is the detection range of the SP2? Please clarify in the text.

The minimum calibration mass used was 12fg, the size range for rBC detection is 190-720nm, which will be included in the revised version.

L273: What is the meaning of "a range of uncertainty"? Does this value represent all the uncertainty with the mass determination, or just the extraction from the incandescent signal?

The ±5-9% is the uncertainty when converting the *M* from the calibration curve.

Line 275: the shape of the BC does not affect its void-free volume.

Thanks to the referee, this will be corrected in the revised version.

L283: presumably variation in laser intensity results in a proportional change in the scattering signal, which probably scales very strongly with particle diameter. Why does diameter vary so much 6% for such small (10%) changes in laser intensity?

The calibration shows the scattering signal is proportional to the optical diameter to the power of about 2.5, and the scattering signal is linearly proportional to the laser intensity, the calculated error is about 6% for a variation in laser intensity of 10%.

L284: the Baumgardner 2007 citation is not included in the references. (I assume it is the B. 2007 paper on Mexico City measurements.) This paper does not present a confirmation of lack of refractive index dependence (See figure 2 for a narrow range of RI). Additionally Figure 8 of the Gao 2007 citation shows a strong dependence of scattering signal on index of refraction. Please correct/clarify in the text.

Baumgardner et al. (2007) will be added in the references. The text will be changed to: "point out the uncertainty in the derived optical diameter is within ±30nm, assuming the coating is sulphate".

L291: Is the scattering signal of the absorbing material measured at the laser center?

Please refer to pg.8774 line 8-11 of the original manuscript.

L309: how is uncertainty for the mass fraction only 23% if the uncertainty in "Do" is as high as 55 %? (l296)

The uncertainty of Do ranges from 6-55% mentioned in pg.8774 line15 of the manuscript, which largely depends on the shape of distorted scattering signal. The uncertainty discussed in the mass fraction is the averaged uncertainty for the project.

Paragraph at L329-345: I am surprised that there are such significant particle losses associated with diffusion at such large sizes: typically diffusion loss is at smaller particle diameters. This raises several questions: First, how good is the SMPS at quantifying particle number? Presumably the SMPS assumes an efficiency for the electrostatic neutralizer, but if it was not as efficient as expected (i.e. an older polonium source) this would quickly impact number determinations. Also, diffusion losses would lead to a shift in the size distributions measured by the SP2: large particle number would not be affected as much as small particle number. This would determine the impact on the BC mass concentrations determined by the SP2 [which I presume is the main reason to include this analysis]. Please comment on the feature of the sample line that suggests the losses are caused by diffusion.

Please refer to the answer to the comment made by referee 1 [Page 8775: Line 23]. We will remove the section regarding the comparison between the SMPS and SP2 and report the data directly from the SP2 measurement in the revised manuscript.

In lines 334-336, this statement is incorrect: the slope of the scatter-plot does not indicate the total number ratio. The discussion of the paragraph is aimed at the efficiency of the SP2 in counting total aerosol in the accumulation mode, but does not deal with the question of its counting of BC particles (and hence the implications for bias in the "bc number fraction" [l341-342]). I suggest adding an estimate of this bias, if possible, and explicitly stating that it is not addressed by the previous discussion. Ah. Now I see some of this discussion in the results section. Perhaps it is more appropriate here?

Please refer to our answer to the comment made by referee 2 [*L. 362-371*].

L367 Please explicitly state that the total number fraction of BC observed is based on the assumption of all BC in a single mode.

Yes. It will be stated in the revised manuscript.

The log-normal fits in figure 9 look like they were constrained, perhaps the center position value? If is this is the case, this should be stated, and the rational for choosing a center value explained. In either case, please clarify in text.

This will be clarified in the revised version.

L402: I don't understand: in the Slowik study, MAAP measured more absorption (with some intermediate MAC that's reasonable?), so would this not suggest that high MAC is correct?

The MAC is derived from the measurements MAAP/SP2, so if the MAAP is biased high, MAC will be overestimated.

L412: Delayed with respect to the peak of the scattering signal?

Yes. The peak intensity of incandescence signal will occur later than the peak of the scattering signal. The text will be amended to clarify this confusion.

P423: Please state that this analysis assumes a laser with uniform intensity, and thus there is no expectation that the bare BC core can scatter more light than the coated particle.

It will be stated.

L454: Which Moteki et al citation is appropriate? In the Schwarz et al reference, bounding assumptions were made about the coatings on undetermined particles – not what is stated here. Please correct.

The citation of Moteki et al. here is removed. It will be corrected.

L464 "more likely to be thickly coated.": : : if they are detected?

Some fraction of small rBC can be detected if they acquire considerable coating to scatter sufficient light.

L494 How does the pollution levels in an air mass change, if it's history doesn't change?

The text is amended to clarify this confusion.

Figure 7: please consider overlaying smoothed/averaged with slower time resolution BC,BC ME, and CO/BC ratio to make average values of these quantities easier to make out.

This has been done, and is summarized in Tab.3 and Fig. 8, median, mean and 10%, 25%, 75%, 90% percentiles are given for each parameter.

For discussion of BC/CO ratio, it seems that CO background has not been removed for the discussion/analysis. As CO has such a large background compared to its enhanced values, this hides the BC/CO relationship shown in Figure 8, and discussed. Perhaps you can plot BC vs CO to show the correlation between the two species. At present, I don't think this is convincingly displayed. Please improve.

Please refer to the answer to comment made by referee 1 [Page 8780: Section 4.3].

L582: please comment on the bimodal distributions associated with SE winds in Figure8.

The bimodal distribution results from the superposition of a range of polluted enhancements on a variable background and is discussed in the text.

L626: BC is not soluble

Corrected.

L635: .23 = 23%

It is 0.23.

L717 BC not BE. In this section it seems that there is mixed discussion of ice phase clouds: CCN and IN are different, and the impact of coatings of BC on its IN/CCN activity will be different. This discussion should thus be edited to remove this confusion.

Corrected.

The discussion in this paragraph was intended to provide an overview of the potential ways that BC may be incorporated into the ice phase. The text will be revised to avoid confusion.

L721 – I don't understand how mixed particles homogenously nucleate.

BC particles with a thick coating of soluble material will act as efficient CCN and will readily form water droplets once a supersaturation with respect to water is encountered. When temperature is low enough, the droplet could be converted into ice phase via homogenous nucleation. The text will be amended to clarify the confusion.

Figure 5: is the histogram on the right normalized for the different log-widths of the vertical axis? Please include the answer in the paper, as it makes it easier to know whether the area under curve scales as total particle number or not (for the different regions).

Yes, the y-axis is the particle counts, thus the area underneath the distribution will be equal to the total number of particles. This will be clarified in the figure caption.