

Firstly we would like to convey our gratitude to both the referees for the important and encouraging comments. The following details our responds to the specific comments.

The original referee comments are in *italic font*, and our comments are in regular type.

Response to Referee 1:

The abstract sentence, “The BC incorporation/removal process therefore plays an important role on modulating the radiative properties of aerosols at the site under the influence of fresh sources” is not fully substantiated by the results of this work. First, there is no direct measure of the removal processes here. Second, the mixing state measurement by the SP2 instrument is a nice handle, but does not seem to be more refined (at this point, anyway) than a binary indicator for ‘fresh’ vs ‘aged’ particles (i.e. BC mixing efficiency < 0.4 and > 0.4, respectively, which – by the way - is also observed in the variance in SSA in time). Suggest a minor change in tone: “The BC incorporation/removal process therefore may play a role in modulating the radiative properties of aerosols at the site under the influence of fresh sources.”

We thank for the comments and will adopt the suggestions in the revised manuscript.

Page 25252 lines 22-24, how is the “enhancement of CN from 10:00-20:00” a “clear indication of fresh emissions”? Couldn’t enhanced CN could also be due to photochemical SOA formation?

The CN correlates with the BC concentration as can also be seen in figure 3 indicating primary aerosol. Furthermore, there is unlikely to be substantial secondary new particle formation in wintertime at 53N. The substantial accumulation mode particle concentrations will further inhibit new particle formation. This sentence will be revised more carefully.

Page 25254 Equation 2 (and same page lines 22) and throughout rest of manuscript: For some reason the unit of ‘rBC’, specifically in ‘ug rBC’, is dropped in preference to “ugC”. This is incorrect and needs to be changed back to ‘ug rBC’.

This is corrected in revised manuscript.

Page 25255 line 2 change “10:00 and 12:00” to “10:00 and 20:00”, line 21 change “19:00” to “16:00”, and page 25256 line 6 change “BC” to “rBC” to agree with results shown in Figure 5.

We wanted to guide the reader to the first peak so have retained the time but will make the text clear in the revised version The latter time and the rBC will be corrected.

** Page 25257 lines 5-8, what was the temperature range during the study? **

6±2.5 °C, which will be inserted into the revised manuscript.

Page 25257 lines 18-20, the statement, “The presence of brown carbon introduces a bias: : :” is too strong, as the effects of brown carbon are very wavelength dependent and, thus, measurements at longer wavelengths may be unaffected by molecular organic absorption.

The texts will be modified more carefully.

** Pages 25258-25259 lines 22-26 and 1-13, discussion here focuses on comparing OOA/HOA and OOA/POA to Zhang et al. 2007 and Allan et al., 2010 results. It should be noted (and is*

currently not) that the OOA/HOA in Zhang et al. must be compared with OOA/POA (i.e. POA = HOA+SFOA) and not OOA/HOA from this study, as Zhang et al. did not classify HOA into subclasses. Thus, the initial comparison here is incorrect.

In the revised manuscript, we clarify the comparison of OOA with POA and HOA.

** Page 25259 lines 14-21 compares OM/rBC to OC/EC, without noting that while related, these ratios are not the same.*

We agree with the reviewer. We did not intend to compare the ratios OM/rBC and OC/EC directly. However, we wanted to use the OM/rBC in this study to indicate the presence of secondary organic formation in an analogous way to previous work using OC/EC. The text will more clearly state this in the revised manuscript.

** Last paragraph in Summary should be inverted, so as to down play the implied association (or remove readers confusion on the issue) between the results here and BC removal processes, which were not measured. The paragraph could note the observations of the results of this work (i.e. SSA changes with measured mixing state) and end by hypothesizing that mixing state measurements such as these may provide insights into BC removal processes in the future.*

The suggestions will be adopted. The last paragraph will be inverted and a sentence covering further work will be added.

The layout issues: Figures 2, 3, 4, 5, 6, 7a, and 9. Definitely need to increase font size of numbers on axes and labels and minimize label text (with perhaps explanations in captions).

We will solve this when typesetting the manuscript.

** Run-on sentence, abstract lines 9-14, "Besides the oxygenated organic aerosol (OOA), this site was found to contain a considerable fraction of primary organic aerosols (POA, mass fraction 50-70% within total mass of OM). The sources of POA are attributed as traffic emission and solid fuel burning and are identified as hydrocarbon-like organic aerosol (HOA) and solid fuel organic aerosol (SFOA), respectively." * Abstract line 16, "a" single particle soot photometer (rather than "the"). * Page 25245 line 6 delete 'the' in "of the secondary aerosols". * Page 25245 line 7 'focused' rather than 'targeted' * Page 25245 line 16 comma before respectively * Page 25245 line 19 semicolon after OOA before "for example" * Page 25245 line 24 ": : : particulate mass. Furthermore, BC is highly..." * Page 25246 line 2 concurrent instead of combined * Page 25246 line 3 OA rather than OOA * Page 25248 line 15 semicolon needed, "cloud droplets; thus, when cloud" * Page 25250 line 7 need period, ": : : USA). The MAAP..." * Page 25250 line 8 remove comma * Page 25252 line 11 remove "shown to be" as the time dependence are shown in the plots, not the direct correlations. * Page 25256 line 18 ": : : relative similarities of the distributions of precursors: : : " * Page 25257 lines 24 and 26, change "lambda" to "lambda's" or "lambdas". * Page 25259 line 23 remove "significant" before "increase" to agree with results shown in Figure 8 (all variations in Dgm are within the variance of measurement scatter and therefore there cannot be a significant change). * Page 25260 line 2 example in parentheses should be "low OM/rBC" not "low rBC"? * Figure 7 Needs "A" and "B" labels in Figure to correlate with caption and change "BBOA" to "SFOA" in Figure.*

All of these will be corrected accordingly.

Referee 2:

For the determination of the submicron mass, obviously only the AMS data and the SP2 data were used. No info is given for the size detection limits of the AMS (what was the lowest detectable particle size?), and no mention is made that the data cannot be directly compared to usual published data from filter samples. The DMPS data could have been used to calculate total submicron mass. Reasonable assumptions about density are given in the MS anyway, so why not do it? It would be very instructive to see how the mass concentrations deduced from the AMS (and the SP2, see below) and those obtained from the DMPS compare.

The smallest size of particles transmitted into the AMS are aerodynamic diameters of between 60-70nm. The volume fraction of particulate matter contained below these sizes is very small. This will be noted in the revised manuscript. We are unsure of the referee's point here. In the first paragraph of section 3.1 in the original ACPD article we calculated the total submicron volume as derived from the AMS measurement by accounting for the composition-specified particle densities. We also calculate the volume concentration of sub micron particles as derived from the DMPS and show this in figure 2. The comparison is also discussed in the first paragraph of section 3.1. Figure 2 covers the points raised by the reviewer.

What I find really problematic is the definition of rBC mass, which is only deduced from the SP2 measurements and then used for comparison with literature values. The instrument has a lower cut size of ca. 200 nm, which means that about half of BC cannot be determined (for mass size distribution of BC see e.g. Berner et al., 1996; JGR 101, D14, 19559-19565), so BC mass is severely underestimated by rBC mass.

Thanks to the reviewer for identifying an important point, which we will add in the revised manuscript. Whilst we accounted for the rBC mass below the SP2 lower detection limit based on an approach used in previous studies, which have estimated the total rBC by extrapolating the mass size distribution according to a single lognormal mode (Schwarz et al., 2006), we neglected to state this in the proposal. This methodology has also been applied for our instrument, which is detailed described in Liu et al. (2010), where about 40-60% mass of the entire rBC population has been detected by the SP2. The mass of rBC presented in this paper has been corrected for these effects.

A MAAP had been running during the experiment, anyway – why wasn't it used for determining BC mass, for submicron mass and the comparisons with OC/EC ratios or the BC fraction of the aerosol? There is no mention of MAAP data in the whole MS.

The MAAP and PSAP, both of which are used in this study, directly measure absorption, not black carbon mass and we have only used the directly measured absorption coefficient derived from these instruments in this paper. The "BC mass" from these filter-based absorption measurements is converted to BC mass using the measured absorption coefficient by empirically assuming a mass absorption cross section (MAC), however the SP2 provides a direct measurement of rBC mass. The MAC in the real atmosphere is variable and is influenced by the presence of brown carbon. In section 4.2.1 we discuss the MAC and its variability and discuss both the MAAP and PSAP data to investigate the possible uncertainties of the filter-based measurements on the BC mass determination by comparing with the directly measured rBC mass. We do not understand why the reviewer believes that there is no mention of the MAAP in the paper.

Turpin et al., who are quoted on p. 25259 that values of OC/EC give indication of secondary particle formation use OC/EC ratios obtained from total filter samples, which of course contain all the small particles. Using OC/rBC values here is questionable. OC determined with the AMS probably has a lower cut size around 50 or 60 nm, while rBC refers only to particles > 200 nm. The values cannot be comparable to those obtained from thermal analysis of filter samples even if the usual uncertainties in the OC/EC cut point in thermal protocols are disregarded.

We refer the reviewer to the answer to the question above: an extrapolation methodology has been applied to calculate the total rBC mass. Whilst the size transmission of the instruments is a little different to that of filter based technologies these effects are rather small. Furthermore, the specific values of OC/EC from literature and OM/rBC from this work have not been compared in the manuscript for the above reasons.

The data on mass fraction of BC found in the literature (Tsyro, Schaap, etc.) are also based on total filter samples, so the values in the MS do not “agree well”, because both the definition of total submicron mass and rBC are quite different, and as no estimate is given for the total submicron mass and as rBC severely underestimates BC, the data cannot be compared.

We agree with the reviewer that the techniques for the EC or rBC mass determination are quite different, however a general consistency has been observed and stated in section 4.2.2. We will add a final clause in the sentence (reproduced here) to emphasize that different methodologies were used:” A modelling study by Tsyro et al. (2007) reported a varied EC mass fraction of 3-15% in PM 2.5 across Europe using the 2002-2004 emission inventory, and simulated higher mass fraction in the populated residential or commercial regions, which agrees well with the observations in this study despite the differences in the measurement methodologies.”

Minor points:

The sentence in the abstract starting on line 14 is very confusing – please separate the first part from the second one (“as the refractory BC component was characterized by: : :”)

Corrected.

p 25259: the diurnal variations of the geometric mean diameter of the size distribution are mentioned without reference to Figure 8. In Fig. 8, the hourly averages of the mean diameter have very large variations – is it really possible to describe a “pattern”? The same holds for the other parameters shown in the plot.

Whilst there is considerable variability in the data the median values do show distinct diurnal variation in most cases and the discussion reflects the broad changes. It should be borne in mind that the bars represent the 25th and 75th percentiles and not uncertainties. A diurnal variation in the 75th percentile indicates that during high concentration periods there is an enhanced trend compared to the median for instance.

p 25261, line 6: HOA is no “source” but a “component”

Corrected.

p 25262, line 25: even at this wavelength, there still is some influence of brown carbon, so B_{abs} of BC may still be lower than deduced from the measurement.

We agree. The manuscript stated: “more reliable estimates of the mass absorption cross section of black carbon are possible.” This statement is not excluding the influence by brown carbon.

p 25263, line 15: the statement that the diurnal variation of σ_{ap} showed no wavelength dependence cannot be deduced from the graph (which is much too busy), and is actually surprising enough to warrant a discussion. I suggest plotting the diurnal variation of the Angstrom exponent, and if no diurnal variation is found then, discussing why it was not found. From the discussion of the data there seems to be quite a variation depending on whether the traffic source or solid fuel combustion contribute most to the aerosol.

The modified manuscript will change this strong tone.

The Angstrom exponent is discussed in section 4.2.1 rather than in this section. The Angstrom exponent shows no diurnal pattern for the entire dataset. The influence of SFOA on the Angstrom exponent can only be reflected when the SFOA/HOA > 2.5 as Fig. 7B shows.

p 25263, discussion of SSA: if there is a concern about the influence of brown carbon on the SSA (which is discussed at length in the text) why did you use 550 nm instead of longer wavelengths? The nephelometer has a channel at 700 nm, which is quite close to the PSAP channel at 660 nm or the wavelength of the MAAP (670)?

The referee raises a valid point thus the SSA at 700nm derived from MAAP_{670nm} is reported in the revised manuscript. In section 4.2.1 the MAC values calculated using the MAAP at 670 nm were more consistent with literature values than those of the PSAP at 660 nm. Given this, we will present the SSA derived from MAAP measurement only in the revised SSA section.

p 25263, lines 15 and 20: if one looks at the definition of SSA ($=1 - \sigma_{abs}/\sigma_{ext}$), the fact that SSA is “modulated by the BC mass fraction rather than by absolute loadings” and the counter-current trend of SSA with the BC mass fraction are not surprising. Is there really a need for discussion for this overall picture? There might be a need for discussion, however, for the data given in lines 25/26: if the particles consist of a BC core and a non-absorbing shell, their mass absorption efficiency can be much higher than if they consisted of BC only. In this case, a straightforward dependence of SSA on the BC mass fraction is not to be expected.

The coating can somehow enhance the BC absorption, however this relationship in the real atmosphere was not observable during this experiment. The rBC mass fraction is the principal factor in influencing the overall ambient SSA, and this reported relationship provides important information on modeling the BC radiative effects.

Figures 10 a and b, though color-coded, should be either deleted or their contents should be plotted differently.

The reviewer offers no guide as to how he/she would like to see the figure plotted. From our point of view the figure content should be retained. However in the revised version we will change the image color scheme to more efficiently identify the continuum of related parameters.

p 25266, last paragraph + discussion next page: This issue is interesting, but the data don't seem to contain enough info to resolve it. Of course it is an interesting question, but it should be moved to the discussion section.

As the answer to the last comment from referee 1, the last paragraph is inverted to avoid the confusion to the reader and to point to further work.