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12, C11858–C11863, 2013

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Interactive comment on "Ambient black carbon particle hygroscopic properties controlled by mixing state and composition" by D. Liu et al.

D. Liu et al.

dantong.liu@manchester.ac.uk

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We thank both references' reviews and inputs. The original comments are in *italic* font, and the reply from the authors is in normal font.

Referee 1:

The subject of the manuscripts fits into the scope of ACP. The manuscript presents data from rather short measurements campaign with novel instrumentation. Although the science and conclusions are rather thin paper presents some nice results. The manuscript gives and impression that there were also other measurements conducted at the same time. If this is the case it would be good to present at least some numbers or overview of these measurements to get broader picture. The scientific methods are

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valid and clearly outlined. The rather small amount of data does not give possibility to very firm conclusions on the overall faith of BC, just some case studies on one season. However the conclusion drawn here is supported by the measured data. The language of the manuscript is fluent. Although the analysed measurement period is short which leaves the scientific output and usability of the data rather shallow, I recommend this to be published in ACP.

An overview of similar measurements from other literatures is given in page 28958-28959 of the original manuscript. While a very wide range of other composition and meteorological measurements were taken, they were not seen as relevant to the microphysical processes under investigation here, so will be presented in future publications dealing with the broader themes. We deemed the trace gas data sufficient to give an indication of the levels of pollution experienced. The referee may have considered a long-term experiment covering a wider range of temporal scale, however the instrumentations as used in this study were not suitable for long-term measurements. Intensive operating periods (IOPs) using research-grade instrumentation are standard practice in the atmospheric composition community when studying detailed processes (e.g. Gysel et al., 2007).

page 28960 line 6, Penkett et al, 1999; is this on the reference list

The reference is added in the revised manuscript.

page 28961, line 13; what other absorbing substance are, and how much compared to BC

The other absorbing particulates possibly include some absorbing organic carbon (brown carbon) and dust. However, these are not relevant to this study, as the SP2 only detects BC, which is generally recognised as the main optical absorber in polluted environments.

page 28962, line 29; DeCarlo et al, 2004; is this on the reference list

ACPD

12, C11858–C11863, 2013

> Interactive Comment

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page 28964, line 5; DeCarlo et al, 2006; is this on the reference list

The reference is added in the revised manuscript.

Is McFiggans, G., Artaxo, P., Baltensperger, U., Coe, H., Facchini, M. C., Feingold, G., Fuzzi, S., Gysel, M., Laaksonen, A., Lohmann, U., Mentel, T. F., Murphy, D. M., O'Dowd, C. D., Snider, J. R., and Weingartner, E.: The effect of physical and chemical aerosol properties on warm cloud droplet activation, Atmos. Chem. Phys., 6, 2593–2649, doi:10.5194/acp-6-2593-2006, 30 2006. refered in the text

This reference is used in the revised manuscript.

in the text McMeeking et al., 2011 is referred but on the reference list the year is 2010 It is corrected in the revised manuscript.

ACPD

12, C11858–C11863, 2013

> Interactive Comment

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Referee 2:

Specific comments: 1. page 28960, first paragraph: It would be helpful to add a few sentences on how the categorization of air masses was done (even though the procedure is described in Fleming et al., 2012). I assume that the pathway of an air mass would often pass over several sectors. How is the air mass assigned in such a case?

The following sentence is added in the revised manuscript:

For each region the percentage of time spent in the region out of all the regions was calculated and when the percentage was greater than the 40th percentile of that region's average percentage it was deemed significantly influenced by this region. The time periods with significant influence from each region are shown in the top panel of Fig. 3. The averaged values in Table 1 are for all periods when each region was deemed significant so for some periods with an overlap of regional influences, those periods would contribute to the averages for all the regions.

2. It would be interesting to see how large the fraction of BC-containing particles is, and how this varies over the course of the experiment. I suggest that you consider adding such a time series to Figure 3, for example.

The mass fraction of rBC in time series has been added in Fig. 3 of the revised manuscript.

3. Is it possible to separate the whole data into day versus night datasets? For example, it would be interesting to see if the hydrophobic BC mode is stronger during nighttime or not. (Difficult to tell from Figure 5). Likewise, it would be interesting to see if the coating material of the BC particles shows a day/night difference.

The nitrate was preferentially formed at midnight (0-6am) in this study and there were a few events when the nitrate formation was strong and BC particles significantly mixed with nitrate coincided with a elevated hydrophilic fraction, however the diurnal analysis of the mixing state/coating/hygroscopic properties of BC particles have not shown ap-

ACPD

12, C11858–C11863, 2013

> Interactive Comment

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parent day/night differences on these parameters within the statistics of the datasets for the entire experimental period.

This statement is added in the revised version.

4. The role of nitrate for BC aging has been pointed out by a few modeling studies on the mesoscale as well as the process scale, for example by Riemer et al. (2004) and Riemer et al. (2010). I suggest including these references in the manuscript.

Both references have been included and addressed in the summary section.

5. p. 28964, I. 10: "Note that like the standard AMS, the SP-AMS quantifies particulate matter in bulk, as opposed to analysing individual particles": This statement is somewhat confusing because, in general, both instruments are capable of producing size-resolved composition information. Later in the manuscript you mention that the SP-AMS was in fact not used in size-resolving mode because of signal to noise problems, but I suggest that you rephrase this sentence on page 28964 to: in bulk or at best size-resolved.

Thanks for referee to point out this. The sentence has been rephrased in the revised manuscript.

6. Fig 4: What do you mean by Âa° raw number concentration?

Additional explanations are added: raw HTDMA-CPC counts (before the HTDMA inversion is applied).

7. Fig. 5 and 6: The phrase $\hat{A}q^{\circ}$ inverted concentration $\hat{A}q\pm$ as y-axis label sounds strange (although it may be a common phrase in the HTDMA community?). I assume you want to indicate that the this is a result from applying the inversion method, but you do mention this in the text, so simply $\hat{A}q^{\circ}$ concentration $dC/d(gf)\hat{A}q\pm$ sounds much clearer. The same comment applies to the term $\hat{A}q^{\circ}$ inverted $gf\hat{A}q\pm$ later in the text.

The words "inverted" are removed from Fig. 5 and Fig. 6 in the revised manuscript.

ACPD

12, C11858–C11863, 2013

> Interactive Comment

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8. Fig. 6, top graph: Are these fractions averaged over all BC-containing particles (hydrophobic and hydrophilic)?

Yes, there fractions are averaged over all BC-containing particles for each full HTDMA scan. This has been added to the text.

9. What about the contribution of sodium chloride? I would assume that sea salt contributes to the inorganic mass of the particles. If it does and if it is not included in the growth factor modeling, this will also contribute to the differences between observation and model result.

We have assumed that the BC-containing particles observed do not contain sodium chloride. As sea salt and BC are both primary aerosols, it is not possible for them to become internally mixed through secondary processes, although we concede that it may be possible for them to mix through coagulation. However, the lack of a high growth factor mode in the HTDMA-CPC (e.g. doi:10.5194/acp-9-9299-2009) would suggest that sea salt is not an important contributor to the number concentrations at the dry sizes under investigation here. As the AMS does not measure sea salt, it is difficult to critically assess whether including this improves closure. This caveat has been added to the text.

Technical comment: Page 28973, line 16: Should read as 2.1.

This is corrected in the revised manuscript.

ACPD

12, C11858–C11863, 2013

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