

## Author reply to referee #1 (G. McMeeking):

Referee comments are repeated in plain black font, author replies are provided in plain blue font and modified/additional excerpts of the manuscript are provided in quoted *italic blue font*.

The manuscript provides results from an investigation of the effective density of two materials (Aquadag and fullerene soot) commonly used for calibrating the response of the single particle soot photometer (SP2) to the mass of black carbon (BC) in individual particles. Two different research groups measured mass distributions of electrical mobility-selected BC particles using different aerosol mass analyzers (the APM and CPMA) coupled to condensation particle counters and in one case, an SP2. They then determined effective densities relating particle mass to particle “volume” as defined by the mobility diameter for a range of sizes and provide coefficients for a polynomial fit relating effective density to mobility diameter for both materials. As shown previously by Moteki and Kondo (2010), the effective densities deviate strongly and non-linearly from the bulk values. The results can be applied by groups operating SP2 instruments that do not have access to aerosol mass analyzers for calibration of the SP2.

The relationship between effective density and electrical mobility has been described previously, but the manuscript makes several valuable contributions that merit publication. First, they examined the reproducibility of the behavior reported by Moteki and Kondo (2010) by repeating experiments for different batches of Aquadag and tested how varying the particle generation methods and concentrations affected the results. Second, they provide coefficients needed by other investigators to correct mobility-based calibrations using these materials adding utility. Third, they also report a new finding that Aquadag is not 100% BC as is normally assumed. For these reasons and also because the manuscript is within the scope of AMT, well written and will be useful to the SP2 community, I recommend its publication if the following minor comments can be addressed.

We thank the referee Gavin McMeeking for his constructive comments which helped improving our manuscript.

## General comments

The results from the thermal-optical analysis should be described in more detail. At minimum the sampling method and analysis protocol should be provided. Was the Aquadag size selected before being sampled on the filter or is this a bulk measurement? Were the thermally denuded particles also sampled onto filters for OC/EC analysis? If so it would be worth presenting how the EC fraction changed in addition to the higher BC mass fraction observed by the SP2. If any information regarding the size-dependence of the contamination is available it would be valuable.

The thermal-optical OC/EC analysis was done on bulk samples of Aquadag and thus no information is available on the size dependence of the contamination. The EC content of thermally denuded Aquadag particles was not determined nor did we investigate it further, as Laborde et al. (2011) recommend using non-denuded calibration standards in a follow-up study.

We clarified that:

*“Thermal-optical analysis of bulk samples of dried Aquadag revealed ...”*

Some specific attributes of the mass analyzer systems are referred to in the paper (e.g., voltage, RPM, mass-voltage relationship), but the basic approach used by these instruments is not given. It would help readers unfamiliar with these instruments if the manuscript had a sentence or two describing the instruments.

The following sentence has been added:

*“Particle mass analysers select particles by their mass-to-charge ratio by balancing the electrostatic and centrifugal forces which act on particles introduced into a thin annular space formed between rotating cylindrical electrodes.”*

On a related note, Figure 2 would be easier to interpret if the concentrations were reported with respect to mass rather than voltage in the APM.

The plot shown in Fig. 2 is convolution of the DMA and CPMA transfer functions and the voltage displayed on the x-axis of Fig. 2 does not directly correspond to particle mass. The true distribution of the particle mass (i.e.  $dN/d\log(m)$  vs  $m$ ) cannot be shown without a proper inversion of the data to account for the width of the transfer functions, multiple charging, particle losses etc. However, that is not what is important here. To determine the mass (or the number-weighted average mass) of the DMA-selected particles all that is needed is the peak of the number concentration vs voltage curve (Fig. 2), which can then be used in Eq.2. Therefore, the data should not be reported with respect to mass (i.e. applying Eq.2 to each voltage in Fig. 2) because it would be misleading. It is already explained in the text how the mass is determined from the raw DMA-APM-CPC measurement. In addition we indicate the voltage corresponding to the peak number concentration in the revised Fig. 2 and we briefly explain how the mass is obtained in the modified figure caption:

*“Classification of fullerene soot particles with mobility diameter  $D_{mob} = 456$  nm with the DMA-APM system. Raw number concentration is shown as a function of voltage applied at the APM. The mass,  $m = 25.6$  fg, of the monodisperse particles, is obtained from the voltage,  $V_C$ , where the fitted number concentration is at a maximum (APM operated at 2000 rpm).”*

The summary section could benefit from a paragraph discussing some of the implications of these results for other products of the SP2 commonly reported, such as total mass concentration, mass size distributions, and coating thicknesses. Are these likely to result in minor changes in these parameters, or larger differences that merit a re-visiting of previously published results?

Using correct effective density data for the evaluation of DMA-based SP2 calibration measurements is one important factor for accurate SP2 measurements. The other one is choosing a calibration standard which gives similar SP2 response per unit mass as the BC material in question. This study only provides effective density data of the calibration standards. The SP2's sensitivity to different BC materials has previously been investigated by Moteki and Kondo (2010) as well as in a follow-up study by Laborde et al. (2011), which has just been submitted to AMTD.

Here we added the following statement at the end of the conclusions:

*“The accuracy of SP2 measurements depends both on using a calibration standard suitable for the investigated samples and on using correct effective density data for the evaluation of DMA-based SP2 calibrations.”*

A more detailed statement about the implication of using different calibration standards and effective density assumptions on the accuracy on previous and future SP2 measurements is made in the manuscript by Laborde et al. (2011).

Finally, it would be helpful to solicit a comment for the manuscript's interactive discussion from an investigator involved in the Moteki and Kondo (2010) or related studies regarding the discrepancy observed at larger mobility diameters.

Actually, we got in contact with the investigators of the above-mentioned study before submission of this manuscript to AMTD with the very goal to find possible reasons/explanations of the observed discrepancy between the two studies. Unfortunately, we did not succeed in identifying potential causes. However, in response to a comment made by the other referee, we clarified in the revised manuscript that the discrepancy observed at larger diameters is of minor relevance to atmospheric BC measurements as the BC mass size distribution typically peaks below  $\sim 10$  fg BC per particle.

## Specific comments

Page 4945, line 9: “. . . which shows that a DMA combined with an SP2 can be used for fast effective density measurements of pure BC particles, if an accurate calibration of the SP2 has been done. . .” Moteki and Kondo (2010) found that it is likely that the refractive index of the BC

particles can affect the SP2 response, which has implications for effective density measurements made using only a DMA and SP2 that should be addressed here.

The need of material specific SP2 calibration is now emphasized:

*“Differences of effective density data measured by the two methods are well within experimental uncertainty, which shows that a DMA combined with an SP2 can be used for fast effective density measurements of pure BC particles. It has to be emphasized that this is only possible if an accurate calibration of the SP2 for the BC material in question has been done using an APM or CPMA, given the fact that the SP2’s sensitivity differs substantially between different BC materials (Moteki and Kondo, 2010; Laborde et al., 2011).”*

Page 4948, line 11: in reference for Laborde et al. in prep omit “Which journal”

Done.

**Additional references:**

Laborde, M., Mertes, P., Zieger, P., Dommen, J., Baltensperger, U., and Gysel, M.: Sensitivity of the Single Particle Soot Photometer to different black carbon types, Atmos. Meas. Tech. Discuss., submitted, 2011.