

## Author reply to referee #2 (J. Schwarz):

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*.

I believe that the manuscript is a valuable resource for experimentalists interested in convenient single-particle black carbon mass standards for calibration and measurements (not only for users of the SP2), and is close to the form it can be published in. I have some suggestions to expand discussion and interpretation of the results to improve the impact and value of the paper:

We thank the referee Joshua Schwarz for his constructive comments which helped improving our manuscript.

1) The impact of the differences between this work and Moteki and Kondo 2010 (for effective material density at larger mobility diameters) should be put in context for typical ambient BC measurements with the SP2, where the center of the mass distribution is typically below 10fg. This will help the typical SP2 user better understand the impact of that discrepancy on their results.

This is indeed an important point. It is addressed in the revised manuscript by addition of mass scales to Figures 3 and 4 as well as by addition of the following statement to the discussion of results for Aquadag and fullerene soot:

*“However, this difference at larger particle mass is of minor relevance for atmospheric measurements, as the BC mass size distribution typically peaks below ~10 fg BC per particle (e.g. Schwarz et al., 2010).”*

2) The paper does not touch upon the very fundamental question of SP2 response as a function of mass (other than in discussion of OC/EC/non-C fraction in the two materials). It's clear that the data is there to make at least a basic statement about the relative SP2 response per unit mass of the two materials. This discussion, too, will inform the SP2 community about the level of discrepancy that they can expect for ambient BC measurements due to their choice of calibration material, as typically used.

We fully agree with the referee that the very fundamental question of SP2 response as a function of mass for different calibration standards compared to pure ambient BC remains a major issue in the quantification of BC using the SP2. This issue has also been identified as an urgent action item in a joint discussion of several SP2 users at the “Workshop on Black Carbon Reference Materials” held in Vienna on June 30, 2011. In order to address this issue we carried out a series of experiments to determine the SP2's sensitivity to pure BC from ambient particles, Diesel exhaust, wood burning exhaust as well as fullerene soot and Aquadag. A separate manuscript by Laborde et al. (2011) has been prepared with highest priority in order to make the results of this comprehensive and important set of experiments public as soon as possible.

The last paragraph of the conclusions has been modified:

*“Additional tests revealed that the Aquadag and fullerene soot calibration particles generated by atomization and diffusion drying contain substantial amounts of non-EC components. This raises the question whether the SP2 calibration curves should be corrected for the mass of the non-EC components. The SP2's sensitivity to pure ambient BC compared to untreated and thermodenuded Aquadag and fullerene soot was investigated in a follow-up study by Laborde et al. (2011). They concluded that the calibration standards should be used in the untreated form. Furthermore, fullerene soot has been suggested as a preferred calibration standard for ambient purposes, in agreement with previous results for ambient BC in Tokyo reported by Moteki and Kondo (2010). 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.”*

### Specific comments:

1) Line 13 page 4939: I do not believe that the differences in SP2 response to different BC materials are “subtle”. It is effectively a factor 2! Please specify the “ways” in which fullerene soot has been found to behave like ambient soot (i.e. SP2 response per unit mass, index of refraction, and effective density).

More emphasis has been put on this issue and an additional reference, showing the similarity between fullerene soot and ambient BC in terms of SP2 response per BC mass unit, has been added. It reads now:

*“It has been shown that significant differences exist in the SP2’s sensitivity to different BC materials and that fullerene soot behaves most similarly to ambient BC in terms of SP2 response per unit mass of BC, refractive index and effective density (Moteki and Kondo, 2010; Moteki et al., 2010; Kondo et al., 2011; Laborde et al., 2011).”*

2) Please change the sentence at line 19 on page 4941 to read “. . .relating the properties of BC calibration materials to those of ambient BC denuded at 400 degrees C in Tokyo (Moteki. . . . .” or similar.

Done, this sentence reads now:

*“This is the way it has been done in previous studies relating the properties of BC calibration materials to those of ambient BC thermally denuded at 400°C in Tokyo (Moteki and Kondo, 2010; Moteki et al., 2010) and it is common calibration practice among the SP2 users.”*

3) Line 5 of page 4942 – is “Collison type” meant here rather than “collision type”?

Corrected (we noticed this mistake only after manuscript publication in AMTD).

4) Line 9-11 page 4945: please weaken this statement by commenting on the additional uncertainty due differing SP2 response to different BC materials (per unit mass) from Moteki and Kondo 2010, or specify that the SP2 calibration need be valid for the particular material in question.

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).”*

5) In figure 3 it appears that the spread of Aquadag effective density from different bottles at some mobility diameters is on order  $\pm 10\%$ . Why does the text describe this as “insignificant” when the level of agreement between the two laboratories, based on fullerene soot, appears better than this?

Figure 3 shows agreement within  $\pm 10\%$  for two different batches of Aquadag and independent measurements by two different laboratories. Only one batch of fullerene soot has been investigated by PSI with different methods and fullerene soot suspension treatments (Figures 4+5). Therefore it is not possible to make conclusive statements whether Aquadag or fullerene soot is preferable in terms of batch-to-batch variability of effective density. However, the SP2’s sensitivity to different calibration standards compared with the sensitivity to thermodenuded ambient BC is the most important criterion for choosing a calibration standard. This question is addressed in detail in a follow-up study by Laborde et al. (2011) showing that fullerene soot is a better SP2 calibration standard for ambient purposes than Aquadag, thereby confirming previous results from Tokyo (Moteki and Kondo, 2010). These facts have been added to the manuscript with the modifications done to address the 2<sup>nd</sup> main comment provided by this referee.

6) Thanks for a nice paper with easy-to-use results: I am already planning on citing it – Shuka  
Good to hear that the manuscript is useful. That’s exactly what it is meant for.

**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.

Schwarz, J. P., Spackman, J. R., Gao, R. S., Watts, L. A., Stier, P., Schulz, M., Davis, S. M., Wofsy, S. C., and Fahey, D. W.: Global-scale black carbon profiles observed in the remote atmosphere and compared to models, *Geophys. Res. Lett.*, 37, L18812, 10.1029/2010GL044372, 2010.