Author reply to Review by Gavin McMeeking:

We thank Gavin McMeeking for his valuable comments which helped the authors to better understand previous publication and improve the clarity and the exactitude of the manuscript.

Referee comments are repeated in black font, author replies are given in red font.

Overview

The authors present observations of black carbon, light absorption and aerosol hygroscopicity made using an SP2, an Aethalometer, and an HT-DMA during the winter in an urban site to the southwest of the Paris city center. They report physical properties of black carbon and aerosol growth factors that are consistent with previous observations in urban regions in Europe. They find somewhat higher BC mass absorption efficiency relative to literature estimates, but consistent with previous studies performing a similar analysis using the SP2 and a filter-based absorption method. They expand their analysis by examining periods dominated by specific sources and observed differences in the BC physical properties, aerosol hygroscopicity, and optical properties during these periods. Finally, they report measurements from a period of the study when they operated the SP2 downstream of the HT-DMA to examine BC hygroscopicity and found that BC generally had low GF and that BC with higher GF tended to be more coated compared to low-GF BC.

The experimental section is described in sufficient detail and the measurements were carried out to a high standard. The paper is well written with the exception of a few minor grammatical errors that can be addressed in copy editing. The work represents a significant contribution to the growing set of information regarding BC physical properties, particularly with respect to its hygroscopicity. It could be improved by examining some of the more intriguing findings in more detail, such as the discrepancy between the relative role of biomass burning and traffic to BC in Paris and Manchester as reported by Liu et al. (2011). I also echo the comment by C. Cappa and D. Lack regarding uncertainties in the Aethalometer correction procedures. I recommend the manuscript be published in Atmospheric Chemistry and Physics once the following comments have been addressed.

Specific comments refer to (page, line number):
25122: coat and D0 should be briefly defined (e.g., coating thickness (_coat) was:) in abstract
DONE

25123, 1: “boxdetectable”
Changed to detectable

25123, 19: not all greenhouse gases are long-lived (e.g., ozone). Please qualify this statement.
Changed to “most greenhouse gases”

25123, 27: I believe the authors support this statement using the finding from Vignati et al. (2010) that removing below-cloud removal of BC had a very minor impact on BC burden in a transport-chemistry model. Did Vignati et al. (2010) separate the activation process from coagulation between BC and existing cloud droplets?
Indeed, Vignati et al. (2010) did not explicitly model the incorporation of BC into cloud droplets through droplet nucleation and coagulation with existing droplets. We modified our statement as follows:
“The incorporation of BC-containing particles into cloud droplets through droplet nucleation or coagulation with existing droplets is a main removal mechanism of BC (Vignati et al., 2010). The ability of BC-containing particles to act as cloud condensation nuclei (CCN) depends …”

25125, 23: This statement is only true if BC-free particles are more hygroscopic than BC, which for some particle types such as primary organic aerosol may not be the case. Minor clarification may be needed.

This sentence reads now: "As a result, BC is expected to be much less CCN active than most BC free particles of equal size (at equal supersaturation)."

25125, 25: Where does the 47% figure come from?
The link to the WHO website was added.

25126, 3: Change "2nd" to "the second"
Done

25126, 18: Change to "Boulder, Colorado, USA"
Done

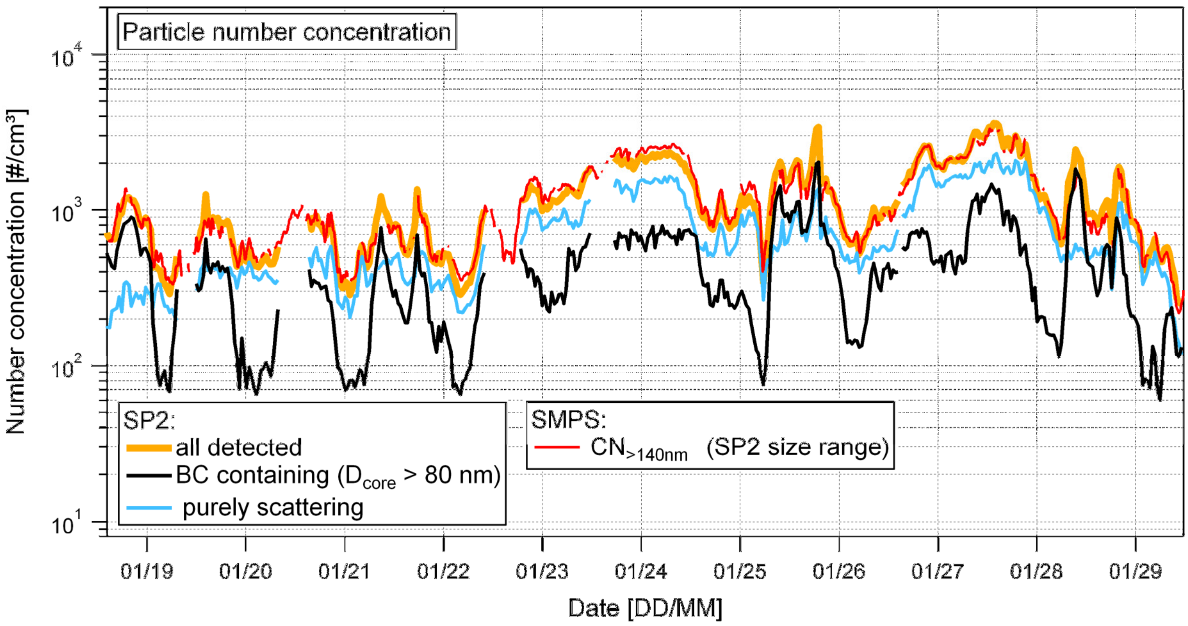
25127, 4: The detection efficiency may be lower for small BC particles with very large amounts of coating (see Fig. 11, Schwarz et al., 2009).
Fig. 9 in Schwarz et al. (2010) shows that the detection efficiency of BC particles is not decreased by thin and medium thick non-refractory coatings. Fig. 11 in Schwarz et al. (2010), which includes thickly coated BC particles does not give a clear picture whether or not the counting efficiency is affected by non-refractory coatings (it is too noisy; some data points indicate a small positive effect, some a small negative effect). Furthermore, Schwarz et al. (2010) do not make any statement in the direction that thick coatings on small BC particles would decrease counting efficiency. Therefore we do not include such a statement in our manuscript either but rather keep our general statement about the lower detection limit of the SP2.

25128, 11: Was the missing mass less than 10% of the measured mass or mass estimated from the log-normal fitting? Please clarify.
The word estimated has been added.

25129, 22-25: Please state the criteria for classifying a BC-containing particle as thickly coated for the checks with the mobility diameter measurement.
This reads now: "Thickly coated BC particles are also expected to be almost spherical. The optical sizing of BC-containing particles with <~35% BC volume fraction, derived with above assumptions for n_{coat} and n_{BCcore}, was also successfully verified against the mobility diameter of size-selected ambient samples…."

25132, 20: omit "C"s
done

25136, 21-26: It would be worth including the correlation coefficient and regression slope here. The excellent agreement between the SMPS > 140 nm and SP2 total also implies that the contribution to number from BC particles detected by the SP2 between its lower detection limit (50-80 nm) and the 140 nm cutoff is small. Is this the case? It would also be interesting to see how the number concentration of BC compared to the SMPS number above the SP2 lower size limit for BC rather than total number.
The correlation coefficient has been added to the manuscript: ($r2= 0.85$).
The following figure shows the comparison between the SMPS and the SP2 in more detail. A highly interesting feature is that good agreement is also achieved at times when the SP2 detected more BC particles than purely scattering particles (influence from fresh traffic emissions). This can be explained with the simple fact that the mobility diameter of fresh fractal-like BC particles is much larger than their mass equivalent diameter (and by chance the lower detection limit of the SP2's incandescence channel translates to almost the same mobility diameter as the lower detection limit of the SP2 for purely scattering particles). This detail-information is not added to the manuscript as it would dilute the general message.



25137, 1-12: Were the Milan and London measurements also performed in similar "urban background" sites as in this study? We found lower percentages (5-7%) for an identical SP2-AMS analysis in our aircraft measurements downwind of London, though these took place aloft in summer rather than on the ground in winter.
The site in London was more precisely in north Kensington and is qualified by the authors as an "urban background" site (already stated in the manuscript). Three sites were taken into account in Milano, including one considered as a background site by the authors.

25137, 18-19: Please explain the reasoning for how the different GF modes are related to background and recently emitted aerosols. Is this based on measured composition, transport patterns when different modes dominate, previous studies?
The reasoning is an anticipation of the results presented in Sect. 3.3.5 and Fig. 12, which is also in agreement with previous literature (e.g. Swietlicki et al., 2008). The text has been reworded:
"The growth factor probability distribution function (GF-PDF), shown as contour plot in (Fig. 7a), reveals that the variability of the mean GF is mostly driven by varying relative contributions of a more hygroscopic mode with a modal GF of ~1.6 and a non- or slightly hygroscopic mode with a modal GF between 1.0 and 1.2. Moderately hygroscopic particles with GFs in the range of 1.2–1.4 are occasionally present. The more hygroscopic mode corresponds to aged background aerosol, the non- and slightly hygroscopic modes represent recent combustion emissions, and the moderately hygroscopic particles can most likely be attributed to moderately aged emissions of regional origin (see Sect. 3.3.5)."

25138, 11-12: This argument is stronger for longer events, such as those that would be associated with a change in air mass, rather than shorter events that are likely associated with a change in local sources. How long were the high events that are reported later in this paragraph? Did they correspond with increases in BC mass concentrations, HOA or any other parameters measured at the SIRTA site? If so this would considerably strengthen the argument regarding T/B ratios.
The events with high T/B ratios were a few hours long and Fig. 3 shows that high T/B ratios correlate well with high BC mass fraction. The modified manuscript reads now: "…The T/B values from downtown Paris can therefore be expected to provide meaningful information for the SIRTA site too. Indeed, high T/B values at the LHVP site coincide e.g. with increased BC mass fraction (Fig. 4) and increased HOA mass fraction at the SIRTA site, which indicate a strong contribution from traffic emissions….".

25139, 5-8: It would be worth comparing the periods of strong biomass burning influence to temperatures to see if they could be related to domestic wood burning.
The diurnal patterns of the BBOA indicated that domestic wood burning is most likely the dominant source of the biomass burning influence (this is discussed in Sect. 3.2).

25142, 2: Some of the patterns discussed in this section should be stronger on weekdays relative to weekends (e.g., rush hours, domestic burning). It would be worth doing a comparison or at least examining just weekdays in case there are not enough weekend samples to obtain decent statistics.
The diurnal pattern is indeed expected to vary between week days and weekend. However, including all data, which are mainly from weekdays, in the diurnal cycles still provides distinct patterns for the traffic vs biomass burning emissions and associated aerosol properties. The limited data coverage, after filtering by source type, does not allow to investigate the weekend effect quantitatively.

25143, 4: What was the rough time for transport from central Paris to the measurement site from observed winds?
The transport time from the city centre to the SIRTA site is ~1.7 h for a typical wind speed of 5 m/s. However, it is difficult to relate this number to the age of the BC particles encountered at the SIRTA site.

25144, 16: Benelux should be defined for readers unfamiliar with the term.
Done

25145, 14-15: The absence of precipitation along the trajectories during the continental periods could also help explain larger BC core diameters. Is it possible to obtain accumulated precipitation for the trajectories to confirm?
Absence of precipitation would likely explain larger BC core diameters compared to air masses with the same back trajectories and precipitation. However, it does not explain larger BC core diameters compared to the local sources, which were not exposed to precipitation either.

25146, 7-10: The Schwarz et al. (2008) and Shiraiwa et al. (2008) studies also took place in early autumn and spring, respectively, with likely more active photochemistry as a result.
This point was added to the text.

25149, 6-11: Another possible source for higher observed MAC is an underestimation of BC mass due to contributions from particles outside the SP2 detection range and not accounted for by the log-normal fitting approach.
This is indeed another possibility for overestimated MAC values. However the BC particles above the detection limit of the SP2 should just give a minor contribution to the light absorption according to Mie theory. In addition, the good agreement between the mass concentrations measured by the Sunset and the SP2 mass concentration (Fig. page 10 of this document) highlights the fact that the SP2 is measuring most of the BC mass.

25152, 12-18: There is a misunderstanding of our results here. We found that about 25% of the low GF particles did not contain detectable concentrations of BC (see Figure 4 in McMeeking et al. (2011)) and speculated that these were likely HOA-dominated particles that have been measured in urban areas. We did not look at the GF_1 and GF 1.1-1.2 regions separately, but the results for GF <1.2 seem to be similar to what is reported here for Paris.
This discussion has been improved:
"McMeeking et al. (2011a), who conducted coupled HTDMA-SP2 measurements in urban Manchester (UK), also observed that the dominant fraction of the low GF particles does contain BC. However, they did observe a minor fraction of particles without a detectable BC core all the way

down to a GF of 1.0, which might possibly be explained by the presence of HOA-dominated particles in their case."

22153, 12-20: I don't follow the argument well here. If I understand correctly, the authors link the importance of the weakly hygroscopic mode observed in HTDMA data throughout the study to an important role for BBOA, but couldn't other oxidized OA species explain this observation? In addition, how do the limited observations of a "distinct biomass burning influence" reconcile with the finding that BBOA makes a substantial contribution to the whole campaign? Please explain further.
This paragraph has been rewritten:
"Crippa et al. (2013) showed that biomass burning emissions give a substantial contribution to the total aerosol mass averaged over the whole campaign, while the analyses presented in Sects. 3.1 and 3.2 indicate that biomass burning emissions only give a minor contribution to the BC mass. This is consistent with the observation that most particles from the biomass burning emissions, which appear in the GF range of ~1.1-1.3, do not contain a detectable BC core, whereas essentially all particles from traffic emissions, which appear at a GF of ~1.0, are BC particles without substantial coatings."

25155, 19-23: The conclusions should note that the findings for BC hygroscopicity refer to only a small portion of the measurement period. They should also stress that the conclusion regarding higher BC in the interstitial phase of clouds is valid only with respect to nucleation scavenging and ignores potential in-cloud aerosol scavenging.
The coupled HTDMA-SP2 measurements are indeed only done for a small portion of the measurement period. However, the similarity of the diurnal patterns of the BC number fraction and the number fraction of non-hygroscopic particles (Fig. 8B and 8D of the current ACPD version) gives strong evidence that the dominant fraction of BC particles was generally non-hygroscopic during the whole measurement period. The fact that BC hygroscopicity is only important for nucleation scavenging has been clarified. This paragraph reads now:
"In this study, the dominant fractions of the BC particles were found to be non- or slightly hygroscopic. Consequently they require a higher supersaturation for CCN activation compared to the majority of particles of equal dry size. It can therefore be expected that BC particles are enriched in the interstitial phase of liquid clouds, if nucleation scavenging dominates, thereby decreasing their wet removal efficiency and increasing their lifetime. This composition specific activation behaviour is important to be considered in global simulations modelling atmospheric BC, as the wet scavenging efficiency remains a major source of uncertainty."

Figures are referred to in the text in non-numeric order and should be re-ordered to match when they come up in the text.
Done