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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Black carbon physical properties and mixing state in the European megacity Paris" *by* M. Laborde et al.

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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: Δcoat and D0 should be briefly defined (e.g., coating thickness (Δcoat) was. . .) in abstract

25123, 1: “boxdetectable”

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

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?

25125, 23: This statement is only true if BC-free particles are more hygroscopic than

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BC, which for some particle types such as primary organic aerosol may not be the case. Minor clarification may be needed.

25125, 25: Where does the 47% figure come from?

25126, 3: Change “2nd” to “the second”

25126, 18: Change to “Boulder, Colorado, USA”

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).

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

25129, 22-25: Please state the criteria for classifying a BC-containing particle as thickly coated for the checks with the mobility diameter measurement.

25132, 20: omit “C”s

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.

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.

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...?

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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.

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.

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.

25143, 4: What was the rough time for transport from central Paris to the measurement site from observed winds?

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

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?

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.

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.

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

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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.

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.

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. 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.

References

Liu, D., et al., Carbonaceous aerosols contributed by traffic and solid fuel burning at a polluted rural site in Northwestern England, Atmospheric Chemistry and Physics, 11, 1603-1619, 2011.

McMeeking, G. R., et al., Influences on the fraction of hydrophobic and hydrophilic black carbon in the atmosphere, Atmospheric Chemistry and Physics 11, 5099-5112, 2011.

Schwarz, J. P., et al., Measurement of the mixing state, mass, and optical size of individual black carbon particles in urban and biomass burning emissions, Geophysical Research Letters 35, L13810, doi:10.1029/2008GL033968, 2008.

Schwarz, J. P., et al., The detection efficiency of the single particle soot photometer, Aerosol Science and Technology 44, 612-628, 2009.

Shiraiwa, M., et al., Radiative impact of mixing state of black carbon aerosol in Asian outflow, Journal of Geophysical Research, 113, D2410, doi:10.1029/2008JD010546

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