Journal: ACP
Title: Sources and mixing state of size-resolved elemental carbon particles in a European megacity: Paris
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MS No.: acp-2011-665
MS Type: Research Article
Special Issue: Megapoli-Paris 2009/2010 campaign

General Comments: This paper investigates black carbon mixing state and concentrations from a single particle (ATOFMS) and bulk perspective (aeth, MAAP, sunset ECOC, etc.) for the purpose of source apportionment in Paris during the MEGAPOLI campaign. The authors have drawn substantial conclusions regarding the properties and origins of black carbon through comparison with meteorological conditions and FLEXPART Lagrangian modeling. The paper thoroughly documents the analysis and scaling decisions made regarding the different data sets, which helps explain where the correlations are strong (or not strong in places). There are a few areas within the paper that could use revision or further analysis (discussed below) regarding aging and scaling. However, overall this paper is a solid contribution to the literature regarding sources and aging of black carbon and should be accepted after minor revisions. I hope the comments below are helpful.

Specific Comments:

- The paper makes the assumption in a number of places that intense peaks of $^{46}NO_2^-$ and $^{62}NO_3^-$ in the negative mass spectrum are ammonium nitrate. I am not sure the data supports being this definitive given the weak $^{18}NH_4^+$ signal and other potential cations for nitrate in the aerosol phase. This argument could be strengthened by showing that high $ECOCNO_x$ time periods have increased NH_4^+ from the HR-AMS relative to $ECOCSO_x$ time periods or through the comparison of m/z 18 intensities for the EC particles themselves for the 4 different EC types discussed in the paper. Absent that, the label could be changed to “nitrate” instead of “ammonium nitrate” or a more thorough qualifier could be added.
- The bimodal nature of the EC particles is a particularly interesting finding. One question regarding these two modes is whether the modes are indicative of two real EC populations (as the authors discuss) or whether these populations are due in part to instrumental effects from the ATOFMS or the scaling methods used to convert from raw ATOFMS counts to mass size distributions? In particular I am curious as to whether the mode below 300 nm may be due in part to non-spherical/fractal soot particles.[1-2] The increased scattering of these particles relative to their small aerodynamic diameters can lead to a mode at the smallest sizes of the ATOFMS size range. The potential shift from fractal to spherical, even if it cannot be addressed numerically should be discussed to provide context to Figure 3. This issue of sphericity might also impact the assumption of spherical particles used to convert from D_{va} to D_{ve}. Thoroughly addressing this issue would be beyond the scope of this specific paper, but its potential importance could be addressed.
- Figure 4: The very tight and strongly westerly PES for Period 1 is quite interesting. Could the oceanic time period here (and in period 3 to a lesser degree) be influenced by ship emissions, specifically those leaving the English Channel? Would that help explain the higher $ECOCSO_x/ECOCNO_x$ ratio during this time period?

- Page 30348 Line 5: The contribution of other combustion sources is discussed here; I would be interested in a slight expansion of this section to discuss what other sources may be present in the Paris region and how they were determined to have minimal impact.
- Page 30348 Line 11: Given the small number of bins (and likely low counts) at the smallest sizes used for the fit of the mode greater emphasis should be placed on the roughness of this assumption.
- Figures 7 and 8: The agreement here is remarkable; an expanded discussion of why there is divergence on Feb 10 between the methods would be of interest.
- The supplemental information is a strong aspect of the paper and supports the findings nicely.

Technical Comments:

- Figure 1: The labels on these peaks are really small in my version of the figure; please increase their size for the next version.
- Page 30335 line 27: This sentence might be reworded to clarify that BC has this effect if it is internally mixed.
- Page 30340 line 1-2: Was testing done to determine if semi-volatile species such as ammonium nitrate were lost due to the denuder (or controlled RH system described above)? Heating has been shown to drive off nitrate from aerosols in denuder systems,[3] could that help explain lower aeth concentrations?
- Figure S2: The TEOM mass is often times lower than the AMS mass, even though the AMS does not measure much above a micron and doesn't measure refractory material such as sea salt and dust that contribute to $PM_{2.5-1,}$ is there an explanation for the seemingly low TEOM values?
- Figures 7 and 8 could be combined for space considerations at a 3 level plot as they contain similar information.

References:

1. Moffet, R. C.; Prather, K. A., In-situ measurements of the mixing state and optical properties of soot with implications for radiative forcing estimates. *Proceedings Of The National Academy Of Sciences Of The United States Of America* **2009,** *106* (29), 11872-11877.
2. Ault, A. P.; Gaston, C. J.; Wang, Y.; Dominguez, G.; Thiemens, M. H.; Prather, K. A., Characterization of the Single Particle Mixing State of Individual Ship Plume Events Measured at the Port of Los Angeles. *Environmental Science & Technology* **2010,** *44* (6), 1954-1961.
3. Pratt, K. A.; Prather, K. A., Real-Time, Single-Particle Volatility, Size, and Chemical Composition Measurements of Aged Urban Aerosols. *Environmental Science & Technology* **2009,** *43* (21), 8276-8282.