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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characterizing the impact of urban emissions on regional aerosol particles; airborne measurements during the MEGAPOLI experiment" *by* E. J. Freney et al.

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The manuscript describes aircraft-based measurements of aerosol composition performed in the vicinity of Paris during the MEGAPOLI campaign in 2009. The paper focuses on a characterization of non-refractory particulate matter (NR-PM) in the Paris plume as a function of photochemical age. The NR-PM measurements are compared with measurements performed in similar environments near large urban areas and also used to examine the ability of recently reported organic aerosol (OA) formation yields to predict OA loadings from simultaneously-measured volatile organic compound (VOC) mixing ratios.

The paper is well written and the topic within the scope of ACP. The field measurements reported in this paper are a valuable addition to the growing collection of SOA formation studies in the atmosphere. I recommend it for publication once the following minor revisions have been addressed.

General comments

One general comment is that it would be interesting to see more detailed comparisons to the ground measurements in Freutel et al. (2013) as well as some of the results (e.g., OA/CO) we reported for London during a similar period (McMeeking et al., 2012).

While the writing is in general quite clear, there are minor grammatical and/or typographical errors in places that need to be corrected prior to publication (e.g., "in the New England, USA" [24903, line 25]; "OOA missions" [24906, line 22]).

Please state somewhere if concentrations have been corrected to standard temperature and pressure.

Specific comments

24890, 9: Please give an estimate of the upper size range sampled by the PSAP and C-ToF-AMS, including transmission through the aerodynamic lens in the case of the AMS.

24890, 15: Please state whether the standard corrections were applied to the PSAP data (e.g., Bond et al., 1999)? If not, please estimate the magnitude of any biases that may result from using the un-corrected measurements and their potential impacts on defining the plume and background removal. This is especially important for the results discussed later in Section 3.2.2, since changes in the aerosol composition could affect aerosol optical properties and the PSAP measurements of BC used to normalize the other aerosol components.

24890,18: An 84-second SMPS sample time translates to a large horizontal distance, so changes in aerosol concentrations will lead to errors in the measured size distribu-

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tions if they occur on similar timescales. How homogeneous was the aerosol around Paris, both inside and outside of the plume? Did the validation of the AMS collection efficiency include both plume and non-plume SMPS samples?

24891, 16-23: The observed slope of $\sim$1 implies that the majority of aerosol mass/volume was below 500 nm, assuming the AMS measured efficiently up to the typically-reported aerodynamic diameter of about 1 μm. Do the SMPS volume distributions confirm this? The text also states that aerosol volume concentrations measured by the SMPS and AMS were compared, but Figure S1 is labeled as a mass concentration comparison. If BC volume was included in the volume comparison please also provide the density used.

24892, 25: I am a little confused on how plume boundaries were defined. The text states plume boundaries were defined based on when concentrations increased above the background level by more than 0.15 μg m-3, but the next sentence states the differences varied from 0.15-0.58 μg m-3. Do these values represent different values used to define the plume boundaries or the range of observed differences with respect to the background? Also suggest changing "Subtracting too large background. . ." to "Subtracting incorrect background. . ." in the last sentence of the section.

24893, 12: define MONA acronym

24893, 15: "inox tube cover of gold"

24894, 14-16: Please also change "northerly, north-easterly, and easterly direction" to "northern, north-eastern and eastern directions" here and throughout the manuscript to avoid confusion with wind direction (e.g., the "easterly" research flights were associated with westerly winds).

24895, 7-12: It may also be worth commenting on the different photochemical environments in the two locations (Paris versus Mexico City).

24896, 26: The wording here is a little confusing "SO4 mass concentrations appear

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to decrease as soon as they encounter the urban plume". I believe the authors mean SO4 concentrations are observed to decrease when the aircraft encounters the urban plume, not the air mass itself, which originated to the west, not north, of the plume in the example being discussed. It is difficult to see the changes in sulfate relative to other aerosol components in Figure 4d. The sulfate and OA plumes appear to be in slightly different locations based on this graph. A separate figure showing OA and sulfate concentrations versus latitude might help.

24901, 11: Can the authors comment further on the low differences between background and local CO mixing ratios? Was this true even for the flight leg closest to Paris? Were observed excess BC / excess CO ratios consistent with similar measurements downwind of other urban regions?

24905, 21-25: The similar concentrations of isoprene inside and outside of the Paris plume suggest Paris itself does not represent a major biogenic VOC source (at least for isoprene), but the regional biogenic emissions may still play a role in SOA formation in the plume through interactions with anthropogenic VOCs (e.g., Spracklen et al., 2011).

24906, 24-26: It is also worth commenting on different temperatures at the surface compared to aircraft sample height given that some of the OA is semi-volatile.

General comment on tables and figures: Both "OA" and "org" are used in the Tables and Figures. . .should be consistent unless there is a reason for the distinction.

Table 1: Units for the total aerosol mass concentration are listed under the OA percentage column label. Also give units for BC.

Table 2: Give units for mass concentrations. Picky point, but first column is date, not the research flight numbers listed in Table 1.

Table 3: Could also list OA/CO values reported for London here as a column in this table as it is probably the most comparable study location.

Figure 3: Including error bars on the right-hand side panels would help show if ob-

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served differences between plume and non-plume are significant.

Bond, C. M., Anderson, T. L., Campbell, D., and Bond, T. C. (1999). Calibration and intercomparison of filter-based measurements of visible light absorption by aerosols. Aerosol Science and Technology, 30(6), 582–600.

Spracklen, D. V., Jimenez, J. L., Carslaw, K. S., Worsnop, D. R., Evans, M. J., Mann, G. W., ... Forster, P. (2011). Aerosol mass spectrometer constraint on the global secondary organic aerosol budget. Atmospheric Chemistry and Physics, 11, 12109–12136. doi:10.5194/acp-11-12109-2011

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 24885, 2013.