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Atmospheric
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Discussions

Interactive comment on "Aerosol particle measurements at three stationary sites in the megacity of Paris during summer 2009: meteorology and air mass origin dominate aerosol particle composition and size distribution" *by* F. Freutel et al.

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This manuscript reports ambient measurements of air pollutants from three stationary sites of the MEGAPOLI summer campaign which took place in the Paris metropolitan area in July 2009. The authors analyzed the influence of air mass origin and meteorology on aerosol loading, composition, and size distribution as well as on O3 and NOx concentrations. Based on these results, discussions are made on the relative

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importance of the impact of Paris emissions on local air quality in comparison to the influence of regional, advected pollutants. This work is comprehensive and of good quality. It yields new information on the chemical characteristics and the broad source contributions of air pollution in the greater Paris region. The manuscript is also well written. I therefore recommend it for publication after the authors respond to following comments.

The PMF factor results show signs of factor mixing. The time correlations between factors and tracer species are a bit low with R2 in the range of 0.2 – 0.3, so are the R2 of 0.8 for comparisons to the reference mass spectra. Performing PMF on the high-resolution spectra can significantly improve the separation of factors and reduce rotational ambiguity. The authors may consider to report the PMF results from analyzing the HR-MS.

The reviewer is correct in that the HOA factors indeed are likely subject to mixing: as stated in the manuscript (Section 3.1), the HOA from the 2 factor solutions at least for the Downtown site can be separated into two HOA components, due to the sources of cooking and traffic. The traffic component then correlates much better with time series and corresponding mass spectra, while the cooking factor does not, due to the different sources. For the Sub NE site, these two sources as well are likely (see Figure S7), however, they could not be retrieved from the ensemble measurements via PMF due to the instrumental noise as mentioned in the manuscript (e.g. Fig. S4), and HR-PMF is not applicable here since only a C-ToF-AMS was used. For the Sub SW AMS dataset, both components traffic and cooking indeed could be retrieved via HR-PMF, which are presented in (Crippa et al., 2012). So in conclusion, there is mixing in the selected PMF solution, however this mixing represents the combination of different HOA-like aerosol components rather than mixing fractions of factors that should better be separated. However, since the aim of the present paper is to compare measurements between the three different sites, we need comparable PMF factors at all three sites, so using

HR-PMF results which would be only available for two of the three sites would be insufficient. Furthermore, we only need to distinguish between organics due to primary emissions and those which are more aged, which can be well accomplished also with the HOA factor comprising a mixture of organics due to two different sources (traffic and cooking). For the reference of the reader, however, we have now inserted a statement where to find the results of HR-PMF analysis, if he/she is interested (Section 3.1):

“For Sub SW, the two aforementioned HOA factors can be distinguished via PMF of the high resolution mass spectra (Crippa et al., 2012). However, within the framework of this paper, where we compare measurements at the three different stationary sites, only the two-factor solutions from PMF of unit mass resolution mass spectra are used for all sites for better comparability.”

The mass spectra of factors shown in Fig. S8 should be processed consistently, i.e., the H2O-related signals should be scaled against 44 using the same approach.

We agree with the reviewer’s suggestion and therefore in the final version of the manuscript, we calculated m/z 17, 18 and 28 for the Downtown site mass spectra (Fig. S8) in accordance with the other sites mass spectra. Additionally, we changed the colour coding to increase contrast.

It is mentioned on Page 22211 that the Sub NE AMS may have smaller ion transmission at larger m/z’s than the other AMS’s during this study. The mass spectral patterns of this instrument can thus be quite different. I am curious about how this issue affects the PMF results and the comparisons of PMF factors among sites?

As can be seen in Figure S8, the PMF factors among sites are still surprisingly similar (high R2 values, and similar fragmentation pattern). Likely, this is due to the fact that the mass spectral patterns are dominated by the smaller m/z’s, and larger m/z’s contribute less to the overall pattern (and therefore, correlation coefficient). However,

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as can be seen also in Figure S8, the relative intensity of larger m/z's is smaller in the Sub NE HOA factor compared to the other HOA factors. The pattern itself is similar, however, only the relative intensity of m/z decreases overall for larger m/z, which is consistent with the lower ion transmission for larger m/z's. See also reply to comment #1 by reviewer #1 regarding the f44 value comparability between the different AMS.

Page 22206, what's the flow rate for AMS sampling, was the flow inside the 3 m 1/8"' tube laminar? If the flow was turbulent or the residence time too long, is particle loss a concern?

The flowrate inside the 1/8" tube was about 1.3 cm3/s. The particle losses for this specific setup were estimated using the Particle Loss Calculator (von der Weiden et al., 2009), and were found to be lower than 10% for the AMS-relevant size range between 0.1 and 1μm (mean value: $\sim 6\%$). Therefore, particle losses were not considered an issue for the AMS sampling. We have added this information in Section 2.1:

"Particle losses for the AMS sampling line were estimated using the Particle Loss Calculator (von der Weiden et al., 2009), and were found to be below 10 % for the relevant size range (0.1 - 1 μm; mean value: $\sim 6\%$)."

Page 22230, to see the difference between the size distributions of HOA and OOA, another approach is to compare the size distributions of AMS tracer ions such as m/z 57 and m/z 44 (Zhang et al., Atmos. Chem. Phys., 5, 3289-3311, doi:10.5194/acp-5-3289-2005.)

We agree with the reviewer that this would be a good approach. Therefore we tried this in our data analysis, but did not see any clear structures, likely due to the fact of the mentioned slight mixing between the two modes, and possibly also due to the fact that the HOA observed in our case is a mixture of traffic- and cooking-related HOA. Due to this fact, m/z57 might be less enhanced for the smaller particle size mode than

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expected, while m/z 44 could be enhanced, both due to the mass spectral structures of the cooking-related mass spectrum (compare Figure S5). We inserted a sentence on this in the revised manuscript (Section 3.3.2):

"Also the size distributions of the mass spectral markers m/z 57 (traffic-related HOA) and m/z 44 (OOA) do not show a clear structure. This might be due to both the aforementioned potential slight mixture as well as to the influence of cooking-related HOA, which shows less significant signal of m/z 57, but a larger influence of m/z 44 than traffic-related HOA (compare Fig. S5)."

Figure 9, instead of assuming same particle density for all sizes, it's probably better to use size-resolved particle density, which can be estimated using the chemically resolved size distribution data from the AMS. In addition, the four types of curves are hard to differentiate in the current plots. It may be worthwhile to consider using different line thickness or color shades to improve clarity.

We now adapted a size-resolved particle density for the calculation of AMS volume and number size distributions (Fig. 9 and corresponding explanations in the figure caption and in Section 3.3.2). For the other instruments' distributions, however, no size-resolved particle density can be calculated, since no size-resolved composition measurements for the larger particle sizes are available, and since the contribution of BC to the different size bins is unknown. Therefore, we use the average density for these also in the final manuscript, to not introduce further uncertainties.

We also changed the appearance of the different lines for more clarity, as suggested.

In Figures 11 and 12, showing the data of nitrate might be interesting too.

We omitted nitrate and ozone from the figure to not "overload" it; however, the corresponding data are given in Tables 4 and 5. Therefore, we added a reference to these tables in the figure caption of Figures 11 and 12 to refer the reader directly to these.

References:

Crippa, M., El Haddad, I., Slowik, J. G., DeCarlo, P. F., Mohr, C., Heringa, M. F., Chirico, R., Marchand, N., Sciare, J., Baltensperger, U., Prévôt, A. S. H.: Identification of marine and continental aerosol sources in Paris using high resolution aerosol mass spectrometry, submitted to JGR, 2012.

von der Weiden, S.-L., Drewnick, F., and Borrmann, S.: Particle Loss Calculator - a new software tool for the assessment of the performance of aerosol inlet systems, Atmos. Meas. Tech., 2, 479-494, 10.5194/amt-2-479-2009, 2009.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 22199, 2012.

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