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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Wintertime aerosol chemical composition and source apportionment of the organic fraction in the metropolitan area of Paris" *by* M. Crippa et al.

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This manuscript reports the source apportionment results from three aerosol mass spectrometers and two black carbon analyzers deployed at three stationary sites in the Paris metropolitan area during winter 2010. It presents a thorough analysis of the chemical characteristics and the sources of aerosol particles in the Paris region during winter season. The subsequent discussions on the effect of Paris on local and regional air quality are very interesting. The overall quality of this work is good and the manuscript is well-written. I thus recommend its publication after the authors respond to a few comments.

- I'd like to suggest that the authors are more specific about the spatial contexts of the regional and local sources. For example, what region (spatial coverage) does "local" correspond to? Does it refer to the city center and nearby vicinity only or to the broad metropolitan area? Speaking of Paris emissions, it seems that the latter is more relevant. According to Wikipedia: "A metropolitan area is a region consisting of a densely populated urban core and its less-populated surrounding territories, sharing industry, infrastructure, and housing." (http://en.wikipedia.org/wiki/Metropolitan_area). So, what if all three sites are located within the Paris metropolitan area and neither GOLF nor LHVP is a background site strictly speaking? The similarities in the time series of primary aerosol species (HOA, COA, BBOA, and BC) seem to suggest so. Their diurnal patterns correlate very well with human activities (e.g., rush hour, meal times, and wood burning for heating) and there are no time shifts in the temporal variation profiles among the three sites. If urban emissions are the main sources of primary aerosols in Paris, the fact that they together constitute a significant fraction ($\sim$30% or more according to reading Fig. 12) of the aerosol loading indicates emissions from Paris is a significant contributor to aerosol pollution in its metropolitan region. It was mentioned on page 22562 that aethalometer measurements at a remote rural site located 58 km east/northeast from the center of Paris show no significant differences from the BC levels detected at GOLF and LHVP. I agree with the authors that this could be interpreted as an evidence for the regional infuences on Paris aerosol. However, another interpretation is also possible – Paris emissions control BC loading in the region. These points need to be clarified.

The urban core of Paris is $\sim$20 km in diameter, while the greater Parisian metropolitan area is $\sim$ 40 km in diameter; the SIRTA and GOLF sites are located near the edges of this metropolitan area. While we agree with the reviewer that homogeneity in terms of chemical composition, mass concentration and sources among the three sites could theoretically indicate a dominant impact of Paris on its surroundings, the following evidence indicates the opposite conclusion. 1. The MEGAPOLI project showed that the yearly impact of Paris emissions is on average equal to only 30% of PM2.5, while the

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remaining 70% is advected to Paris and impacted by long-range transport of continental pollution (Beekmann et al., 2012). These findings included one year of PM2.5 chemical composition measurements at 4 rural sites, covering a spatial distribution of 140km (and overlapping with measurements performed during the summer phase of MEGAPOLI). Together with meteorological measurements that indicate which of these rural stations is upwind of Paris, these measurements indicate the dominance of regional sources over the Paris plume. 2. PM1 levels measured at the GOLF site were much higher for air masses coming from central Europe than for air masses with SW winds, which contain the urban emissions from Paris (Beekmann et al., 2012). 3. During the MEGAPOLI summer 2009 campaign, a marine aerosol factor was identified in PMF analysis of high-resolution AMS data. This marine aerosol contributed on average $\sim$16% of total OA in Paris, even in the urban core (Crippa et al., 2012b). The ability of the Atlantic Ocean to significantly influence air quality in the Paris urban core (200 km from the ocean) suggests that particulate emissions from Paris are unlikely to be the major regional influence. 4. Finally, major increases in PM1 concentrations occurred when the Paris area was influenced by transported air masses (e.g. during the event on the 26-28th of January 2010, the high pollution event at the end of the winter campaign and the continental period observed in the beginning of the summer 2009 campaign (Freutel et al., 2012)). The strong effects of these events, coupled with the predominance of secondary/aged organic components during these periods, further demonstrate the dominant effect of regional air quality effects relative to primary and secondary sources in Paris.

We acknowledge the Reviewer for pointing out the importance of the definition of the local vs. regional sources in Paris. This topic is developed into several papers, therefore now we included a brief section in the manuscript to clarify the impact of Paris emissions and the regional sources.

“The observed homogeneity in terms of chemical composition, mass concentration and sources among the three sites could theoretically indicate a dominant impact of Paris

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on its surroundings, however the reduced impact of Paris itself on the surrounding air quality indicates the opposite conclusion. The urban core of Paris covers ∼20 km in diameter, while the greater Parisian metropolitan area is ∼ 40 km in diameter; the SIRTA and GOLF sites are located near the edges of this metropolitan area. Beekmann et al. (2012) showed that the yearly impact of Paris emissions is on average equal to only 30% of PM2.5, while the remaining 70% is advected to Paris and impacted by long-range transport of continental pollution. Moreover PM1 levels measured at the GOLF site were much higher for air masses coming from central Europe than for air masses with SW winds, which contain the urban emissions from Paris during the summer and winter MEGAPOLI campaigns (Beekmann et al., 2012;Freutel et al., 2012). Crippa et al. (2012b) showed also the ability of Atlantic Ocean air masses to significantly influence air quality in the Paris urban core (200 km from the ocean), suggesting that particulate emissions from Paris are unlikely to be the major regional influence. Our results agree with the conclusions of Sciare et al. (2010) who reported long-range transport to be the major source of PM2.5 secondary aerosol in Paris during springtime."

- The OOA2-BBOA factor is ambiguous and related discussions vague. Was PMF performed on the high-resolution mass spectra? The aerosol loading was high during this study, so the high resolution spectra should have good signal-to-noise ratios. Analyzing the high resolution spectra will likely give less ambiguous PMF solutions and better separated factors.

PMF was here performed using the unit mass resolution MS since the HR information was not available for all the three sites (e.g. the GOLF site deployed a C-TOF-AMS). From our analysis we were not able to determine whether the OOA2-BBOA at LHVP was a real secondary OA factor deriving from primary BBOA emissions, an atmospheric mixture of primary BBOA and secondary OOA, or a mathematical mixture due to the inability of PMF to clearly separate sources with similar temporal variation (i.e. peak concentrations at night). These uncertainties prevent us from precisely defin-

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ing this factor. However, PMF analysis of combined UMR AMS data and simultaneous PTRMS measurements at the LHVP site (Crippa et al., in preparation) provided clearly separated BBOA and nighttime SV-OOA factors. This suggests that the OOA2-BBOA factor presented in this study is mostly a mathematical mixture of factors that could not be completely separated by PMF. This is a limitation of AMS PMF and will be discussed in detail in the forthcoming publication.

- Page 22539, line 23, a comprehensive review of factor analysis approaches of aerosol mass spectrometry of ambient aerosol was published [Zhang et al., 2011]. It seems an appropriate reference to cite for this sentence.

This reference is now added to the manuscript.

- Page 22558, 2nd paragraph, for COA in Beijing, Sun et al. [2010] and Huang et al. [2010] should be cited since they actually reported the observations of cooking aerosols based on analyzing ambient AMS measurement data while He et al. [2010] primarily discuss the spectral profiles of various cooking OA. In addition, significant quantities of COA were also determined in New York City [Sun et al., 2011] and Fresno [Ge et al., 2012]. Fig. 1, it would be helpful to show the comparisons of the average loadings of total PM1 among three sites too.

Sun et al. 2010 and Huang et al. 2010 references are now cited instead of He et al. 2010 for the Beijing case and Sun et al. 2011 and Ge et al. 2012 for New York and Fresno, respectively are additionally cited. The average loadings of total PM1 are already reported in Fig.12 for the three sites, so we decided not to show it here again.

- Fig. 2 and 8, the medians are used for making the diurnal profiles at here. Often times, the means are also shown. It will be interesting to know how the diurnal patterns differ if the mean values are plotted. The difference between the median and the mean is usually larger for a set of data that is more deviated from normal distribution. Primary species are more influenced by spikes, thus might show bigger differences between the mean and the median.

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The mean values are now added in Fig.2 and Fig.8.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/12/C10523/2012/acpd-12-C10523-2012-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 22535, 2012.