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Atmospheric Measurement Techniques Discussions

Interactive comment on "Application of mobile aerosol and trace gas measurements for the investigation of megacity air pollution emissions: the Paris metropolitan area" *by* S.-L. von der Weiden-Reinmüller et al.

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Dear Referee,

Thank you very much for your helpful comments on our manuscript "Application of mobile aerosol and trace gas measurements for the investigation of megacity air pollution emissions: the Paris metropolitan area"! We tried to include all your suggestions for improvement. In the following we answer in detail to the individual points.

Comment: . . . The important discussion of methods to identify and remove data dominated by nearby emissions sources so they don't unduly corrupt studies of larger scale ambient background variations should be moderately expanded and made less pessimistic.

Answer: Since the manuscript is already quite long we tried to condense the necessary information as much as possible. We provided a detailed discussion of local pollution removal procedures in Drewnick, F., Böttger, T., von der Weiden-Reinmüller, S.-L., Zorn, S. R., Klimach, T., Schneider, J., and Borrmann, S.: Design of a mobile aerosol research laboratory and data processing tools for effective stationary and mobile field measurements, Atmos. Meas. Techn., 5, 1443-1457, 2012. We slightly extended the local pollution discussion in Sect. 4 and we hope that the discussion about local pollution is now less pessimistic. We also added the important point that data associated with local pollution contain valuable information about e. g., pollutant emission fluxes from point sources or on-road pollutant emission indices.

Comment: Several interesting trace gas and fine particle trends are presented in four complex data plot figures, but there is no quantitative attempt to demonstrate whether and/or how the various pollutant concentrations trends are related.

Answer: We obtained detailed (quantitative) analysis results about how the various pollutants are related, based on the MoLa data set. These analysis results will be presented in a separate publication which is almost ready to be submitted to Atmospheric Chemistry and Physics. To avoid content conflicts with that upcoming publication we decided not to present detailed analysis results in this more methodical manuscript.

Comment: While this is primarily a measurements techniques manuscript, if some of the data presented were actually analyzed, at least for a few illustrative examples, the scientific value of the paper would be greatly enhanced. For instance, other investigations of megacity plumes have used combinations of fixed site, mobile laboratory and aircraft measurements to demonstrate that odd oxygen ([Ox] = [O3] + [NO2]) pro-

duction is closely correlated with oxidized secondary organic aerosol production. The two mobile labs described measured both components of Ox in the Paris plume and have computed PMF oxidized organic aerosol (OOA) components from their on-board aerosol mass spectrometer data for axial plume transects and/or stationary plume intercepts, but this paper does present plots of [Ox] versus [OOA] to determine if key photochemical products in the Paris plume behave like other sampled megacity or near-megacity plumes. As another example, Figure 4 presents plume axial and background plots of hydrocarbonlike organic aerosol (HOA), particulate sulfate (SO4=) and black carbon (BC). Plots of [HOA]/[BC], as well as [OOA]/[BC], and [SO4=]/[BC], as a function of plume axial distance (or transport time) and their comparison with the same ratios in the out-of-plume background ambient might tell a very interesting story. They might reveal in-plume chemical conversion rates of primary pollutants like HOA and SO2 and how fast their secondary products reach typical background levels, assuming that deposition, not heterogeneous oxidation, is the primary BC loss process. I'm sure the distinguished authors of this manuscript could identify other illustrative mobile lab data analysis opportunities to help convince their readers of the value of their efforts.

Answer: Unfortunately, MOSQUITA had problems with the NO / NO2 device and there are no reliable data available for the relevant time periods (intercomparisons, parallel measurements with MoLa). The NO2 data recorded by MoLa are internally calculated by the Airpointer instrument from the NO and NOx measurements. Therefore, the NO2 data have a larger uncertainty than the NO and NOx data, especially when the NO and NOx concentrations are changing rapidly like during mobile measurements. Furthermore, the MoLa instrument (Airpointer) applies a molybdenum converter for the NOx measurement, causing additional uncertainty of the measured NOx and NO2 mixing ratios (Steinbacher, M., Zellweger, C., Schwarzenbach, B., Bugmann, S., Buchmann, B., Ordóñez, C., Prévôt, A. S. H., and Hueglin, C.: Nitrogen oxide measurements at rural sites in Switzerland: Bias of conventional measurement techniques, J. Geophys. Res., 112, D11307, doi:10.1029/2006JD007971, 2007). For the measurement example described in Section 3.4 (stationary measurement) one can see, that O3 is reduced

in the emission plume, but NOx (as well as NO and NO2) is enhanced. OOA is in our case especially in summer dominated by long-range transported organic aerosol that is not associated with the emission plume and therefore it is only slightly enhanced in the emission plume. Consequently, the correlation of odd oxygen (Ox) to oxidized organic aerosol (OOA) does not show a clear trend in this case. To provide further analysis results we added the average PM1 aerosol mass composition of background and emission plume air masses to Fig. 5 (stationary measurement example). From these average compositions the plume contribution was calculated as well as the increase in PM1 mass concentration. We also added the average HOA to BC ratios in plume and background air masses for the axial measurement discussed in Section 3.3. Further discussion of transformation processes in the emission plume would be beyond the scope of this (already very long) paper and will be presented in an upcoming publication which will be shortly submitted to Atmospheric Chemistry and Physics.

Comment: There are some minor flaws in the current manuscript that deserve attention:

Frist, I believe that Figure 1 introduces an unfortunate nomenclature choice. While the yellow arrows, representing cross plume trajectories are reasonably labeled "cross section" measurements (should be cross-sectional if you want to use the adjective form) are reasonable, the black double ended arrow along the plume's axis labeled "radial measurements" is misleading. While these transects are "radial" with respect to the city's geometry, they are "axial" with respect to the pollution plume's flow geometry, which is the natural reference frame for the reported measurements. Transects along the plume's axis should logically be called "axial;" radial plume measurements are those already termed "cross section." I suggest that axial replace radial everywhere in the manuscript. This includes the abstract, where "radially away from the city center," should be changed to "axially along the flow of the city's pollution plume."

Answer: We changed the nomenclature from "radial" to "axial", which is a quite better and precise choice.

Comment: Second: The important discussion of methods to identify and remove data dominated by nearby emissions sources in section 4.2 fails to clearly explain that in most mobile laboratory sampling modes pollution exhaust plumes from motor vehicles, and even many industrial point source emission plumes, produce data spikes lasting a few to a few tens of seconds that are much shorter in duration than intrinsic variations in background pollution concentrations. If pollutant sensors with real-time (_1s or less) response times are deployed these nearby pollutant source data spikes are relatively easy to recognize and remove. More importantly, they can be (and often are) separately analyzed to yield very useful emissions data, such as fleet averaged fuel-based pollutant emission indices for on-road motor vehicles or pollutant emission fluxes from individual fixed site point sources like factories, commercial operations or road maintenance activities. Of course, if traffic is too heavy or moving too slowly, especially in low wind conditions, it becomes too difficult to remove the influence of nearby discrete pollution sources and ambient background data has to be designated as contaminated, as noted in the manuscript.

Answer: We added the information that sporadic local emissions can easily be identified by concentration spikes in fresh pollution marker time series (e. g. CO2), if the temporal resolution is sufficiently high (around 1 s). In Drewnick et al. (2012) more details about the automatic local pollution removal procedures we tested are presented. These procedures are under certain circumstances (e. g. mobile measurements in remote regions) very useful and efficient to obtain uncontaminated data sets. We also added the information that the data points we remove as local pollution contain indeed valuable information about e. g., local emission indices and point source emission fluxes. Only for investigating larger scale phenomena like urban emission plumes it is necessary to remove these local emission data points.

Comment: Third, the current manuscript also needs some moderate copy editing beyond the “axial” for “radial” terminology swap noted above. For instance, “aircrafts” appears in several places (e.g. page 4 - line 8, page 7 – line 2, and page 9 – line 6);

however, the plural of aircraft is aircraft. There are also problems with prepositions; for instance: on page 13 - lines 19 and 26, where “with about” would normally be “at about;” and page 25 – line 24, where distance “to” should be “from” and “cross sections in” should be “cross sections at.” Also on page 25 – line 24, as noted above, “cross sections” is more properly “cross sectional transects.” Please note that I recognize the manuscript is generally well written and its English usage and grammar is far better than any manuscript that I could produce in either German or French.

Answer: We corrected the typos and grammatical errors – thank you for these hints.

Comment: After some modest condensation, clarification and copy editing the current manuscript will be a very good Atmos. Meas. Tech. paper. Adding a few selected analyses illustrating the scientific value of the some of the data presented would, in my opinion covert it to an excellent Atmos. Meas. Tech. paper. I recommend publication after the author’s have considered and addressed the suggestions listed above.

Answer: Thank you!

Interactive comment on Atmos. Meas. Tech. Discuss., 6, 7659, 2013.