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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 Prof. Paul Seakins,

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: 1. Introduction - I confess to some self interest here, but there are other publications on mobile measurements that have not been cited. Some are possibly too specialized, but we do have a publication on aspects of the PUMA campaign (looking the Birmingham region, ref 3 below) which would be very relevant to this work. References to consider are: (1) Estimations of Primary Nitrogen Dioxide Exhaust Emissions from Chemiluminescence NOx Measurements in a UK Road Tunnel, W.A. Simmons and P.W. Seakins*, Science of the Total Environment, 2012 438, 248-59. (2) NO and NO2 interconversion downwind of two different line sources in suburban environments, Alison Chaney, David C. Cryer, Emily Nicholl and Paul W. Seakins, Atmospheric Environment, 2011, 45, 5863-5871, Doi 10.1016/j.atmosenv.2011.06.070 (3) 'Measurement and modeling of air pollution and atmospheric chemistry in the U.K. West Midlands conurbation: Overview of the PUMA consortium project.' R.M. Harrison, J. Yin, R.M. Tilling, X. Cai, P.W. Seakins, J.R. Hopkins, D.L. Lansley, A.C. Lewis, M.C. Hunter, D.E. Heard, L. J. Carpenter, D.J. Creasey, J.D. Lee, M.J. Pilling, N. Carslaw, K.M. Emmerson, A. Redington, R.G. Derwent, D. Ryall, G. Mills and S.A. Penkett. Science of the Total Environment 2006, 360, 5-25 doi 10.1016. (4) 'Mobile Laboratory reveals new issues in urban air quality' P.W. Seakins*, D.L. Lansley, N. Huntley and A. Hodgson. Atmospheric Environment 2002, 36, 1247-8.

Answer: Thank you for these suggestions. We checked the four publications you suggest for citation and found that paper No. 3 (Harrison et al., 2006) and No. 4 (Seakins et al., 2002) are best suited for citation in the introduction.

Comment: 2. Intercomparisons - Given the nature of the publication (AMT vs ACP), I was surprised that more detail wasn't provided on the intercomparison between the two mobile laboratories. Some of the text could more usefully by replaced and certainly would be enhanced by figures showing timeseries comparions and/or regression plots (I appreciate statistics are given in the table, but are hard to visualize and may be distorted by spikes). Some issues need following up on - e.g. CO2 measurements ' During the time.....and reliably calibrated....' Does this mean that there were other

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times when the agreement was not good? How do we know when the measurements were reliable? Does this cast doubt on the measurements reported on CO2? Another area to address would be the NOx data. A difference of 30% would be outside most random calibration errors suggesting a systematic source. NO:NO2 is also an important parameter (like O:C ratio, giving some evidence on air mass processing, but also as a useful indicator of fresh/local emissions) and I would suggest that both NO and NO2 as well as total NOx is reported. It is quite likely that this paper will be referenced in several upcoming studies, so it is important to get this details fixed.

Answer: Since the actual publication is already quite long we tried to condense the intercomparison section (and other sections as well) as much as possible. For better illustration of transient effects now the new Figure 1 shows the concentration time series of particle number, black carbon, CO2 and O3 recorded by the devices in MoLa and MOSQUITA during the summer and winter intercomparisons. The CO2 device installed in MOSQUITA did not work during the winter intercomparison, so only the summer data are shown in Fig. 1. During the second summer intercomparison (23 July 2009) a strong enhancement of CO2 mixing ratio was measured between 16:00 and 17:00 local time by the MOSQUITA device. The reason for this enhancement could not be found. Possibly, an internal instrument error or problems during calibration occurred. So only during the first summer intercomparison (11 July 2009) both CO2 instruments were reliably calibrated. We included this information to the manuscript. Unfortunately, MOSQUITA had problems with the NO and NO2 measurement device, so there are only NOx data available for the intercomparison times. We also included this information to the manuscript.

Comment: 3. Data interpretation and video analysis (2.3, 2.3.1). Ref 3 above includes discussion of local influences including from opposite carriageway (criteria 2) and Ref 1 on tunnel measurements. Presumably there is potential for future automation where proximity devices could be used to screen data (criteria 2 vehicles < 150 m).

Answer: Actually, we already have made some effort to automate the local pollution

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detection. It is now possible to note via mouse click the time period and the kind of pollution event occuring during the measurement. With this information a contamination mask is created, which can be used to analyze the data. We added this information to the manuscript.

Comment: 4. Fig 1 - I found this figure difficult to interpret (and impossible in b/w printed output). It is not clear where Paris is, what the scale is etc. It might be helpful to have two figures side-by-side, one with a map and the other with the predictions.

Answer: We agree completely that these figures are difficult to interpret. However, unfortunately, all plume prediction maps are only available in this format. For the winter campaign the scale of the prediction maps is different and the plume appears even much smaller. During the measurement campaigns we only had these prediction maps as forecast for the emission plume direction and strength, and the missing reference points were indeed problematic for measurement planning. We want to show this original prediction map here (1) to demonstrate the difficulties of measurement planning and (2) because it is a suitable map to demonstrate cross sections and radial trips through the in this case clearly visible emission plume. To make the map easier to interpret we included a scale and marked the Paris region.

Comment: 5. Section 3.1 (Fig 2) How does this description of the data with obvious influences of local pollution link to the earlier section (2.3.1) on video analysis. Have the data been screened? Possibly might be worth showing a 'before and after' figure.

Answer: The data shown in Fig. 2 have not been screened for local pollution. The purpose of this measurement example is to demonstrate the applicability of mobile measurements for the distinction between local / regional and long-range transported pollution. The peaks in the concentration time series should not only be seen as "data waste" (removal by video analysis). If the focus of the investigation is e. g., on fresh emissions, the concentration peaks would contain the major information and could be analyzed individually. The presented measurement data are meant to demonstrate the

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general applicability of mobile measurements for the investigation of pollution of different oxidation stages (ages). More details on different local pollution removal strategies and examples of "before – after" time series are presented 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.

Comment: 6. Section 3.2 (Fig 3) What was the role of the MOSQUITA system which appears to be making virtually all of its measurements outside the plume? Fig 3 works well, but might consider removing most of the MOSQUITA data which do not appear to transect the plume shown in Fig 3. This would allow you to expand the spatial scale of the MoLa transects.

Answer: The purpose of Fig. 3 is to show how useful a combination of as much data as possible could be to get a broad overview about the pollutant distribution. In this case the MOSQUITA data demonstrate the pollutant distribution mainly in background air masses, while MoLa crossed the emission plume. The MOSQUITA data help to distinguish between plume and background air masses. It is right, that the MoLa data are more focused on the pollution plume in this case. However, before the actual measurement the direction of the emission plume was not exactly known. The decision to measure in different sectors around Paris reduced the probability to miss the emission plume. Unfortunately, there are no measurements where the MoLa and MOSQUITA data can be combined in a more useful way (e. g. four cross sections through the entire emission plume in different distances to the city which could be performed in future campaigns with stronger coordination between the mobile measurements).

Comment: 7. Section 3.3 (p7685, line 14) What defines the boundary of the plume (shaded area in Fig 4)? On the outward journey the boundary is drawn at 60 km, but O3, HOA and BC all seemed to have reached a plateau closer to the centre.

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Answer: The boundary of the plume was defined manually taking into account all measured variables. In some cases the fresh pollution markers (e. g. black carbon, HOA) reach constant background levels at slightly different distances to Paris. Reasons for this are (1) different background levels, (2) different sensitivity of the measurement devices, (3) scattering of the data, and (4) different influence of local emitters. The gray shaded area in Fig. 3 is only an approximation for the extent of the emission plume. We corrected the gray shaded area in this figure, because the plume is visible in black carbon and HOA only up to 50 km to the city center. In the original graph the plume was misleadingly defined by the sulfate concentrations. However, in the text we argue that the enhancement in sulfate concentrations near the city is most likely not connected with the emission plume.

Comment: 8. Section 3.4 (Fig 5) The influence of the moving plume across the stationary measurement site is nicely shown. I think it would be useful to significantly expand the time scale to focus on crossing of the plume - a lot of the data after 13.30 don't change significantly. It would be interesting to see if there is any temporal variation (allowing for different sampling) between the different plume markers. Plotting NO and NO2 as well as NOx (and possibly NO:NO2) might be interesting too.

Answer: We think it is important also to show the recorded background concentrations after 13:30. This measurement example demonstrates that the emission plume is not related with enhanced sulfate concentrations, which might be suggested by measurements like presented in Fig. 4. Additionally, one can see which variables vary strongly also in background air masses (e. g. CO2, O3) and which variables are useful indicators for the emission plume (e. g. black carbon, PAH, NOx). Since the applied NOx device in MoLa (Airpointer) measures only NOx and NO and then calculates NO2, the NO2 concentration time series has a larger uncertainty than that of NO and NOx. We checked the time series of NO:NO2 for the presented measurement example. A small decrease in the NO:NO2 ratio can be observed during the plume crossing. However, there is large noise in the NO:NO2 ratio and we assume, that the NO / NO2 / NOx

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concentrations measured by MoLa are not precise enough for detailed analysis of the gas phase nitrogen compounds with high temporal resolution.

Comment: Minor editorial comments p7674 line 6 times whilst driving... p7677 line 8 should 'Apes' be fully capitalized? p7680 line 7 therefore rather than therewith p7682 line 14 In this section...

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

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