Atmos. Chem. Phys. Discuss., 11, C14487–C14492, 2012
www.atmos-chem-phys-discuss.net/11/C14487/2012/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Airborne measurements of trace gases and aerosols over the London metropolitan region" *by* G. R. McMeeking et al.

Anonymous Referee #2

Received and published: 18 January 2012

The paper by McMeeking et al. is an overview of flight data in terms of VOC emission ratios, aerosol physical properties, and aerosol chemical composition from a series of flights following the M25 road encircling the megacity of London. The measurements are of high quality, and the context of the paper is of interest on a broad scale with data from this developed megacity adding to the literature on megacity emissions on regional concentrations. As it is written, the paper does a good job of presenting results for London, but falls short of putting these measurements in better context of observations in other locations, e.g. Paris, Los Angeles, Tokyo, and Mexico City. In general, more comparison of these result to other locations will strengthen the paper considerably.

How were background concentrations and areas of "background" decided? The only

clue to the reader comes on page 30685 line 10, but gives no exact criteria delineating what is a plume, what is background, and what is upwind. This is extremely important to be clear about since the conclusions of the paper regarding the importance of regional aerosol are dependent on how exactly it is defined. Is background aerosol in the air mass upwind of the city and into which London emits (thereby increasing the concentration), or is it aerosol away from the london plume, and may not be indicative of the air mass in which London adds emissions? For example, Figure 9 shows the comparison between background and london plume concentrations. From this figure there is little difference in background concentrations and on 24 June the background concentration is higher than the plume concentration. An extreme interpretation of this is that the london area actually cleaned the regional plume (which would clearly be at odds with our understanding of urban vs regional emissions). Is it possible that some of the "background" concentration is due to recirculation of london area plumes from previous days? The authors provide synoptic views of the region on flight days, but this does not provide the reader with an understanding of the history of the air masses. The authors MUST give detailed discussion on how plumes were located, how plume vs background was decided, and background concentrations were determined for species such as CO and C2H2 used in ratios and analysis.

Emission Ratios calculations (30677-30678): Using the plume and non-plume samples with various ages, and a variety of sources adds significant uncertainty to the measurements. The authors note correlations are stronger "in plume" which suggests that the authors should report those numbers (at least in addition to the other numbers). While the plumes may have fewer points, they likely "better" represent the same general sources, and are younger. The use of plume data, avoids uncertainties in plume age (and photochemical loss) and more importantly reduces the much greater uncertainty associated with mixing of different sources with different emission ratios.

Specific Comments (in the format Page/line)

30667/9-12 indicate what the VOCs emission ratios were compared to e.g. "Emission

ratios of VOCs to C2H2" (and elsewhere in the paper)

30673/Section 2.2: Give more details about the data products from the lidar. I.e. what is it measuring, and what processing is required to give you e.g. $b_{ext}(UsedinFigure13)$

30678/17-20: Because the VOC / C2H2 ratios were almost systematically a factor of 2 off (except for the alkanes at a kerbside monitoring site which can be easily explained), and C2H2 is the one species across all measurements, is it possible that the C2H2 measurement is either incorrect, and off by a factor of 2 aloft or on the ground or that C2H2 is simply higher in concentration by factor of 2 aloft? That would seem the most reasonable explanation rather than different source regions since all of the measurements are made in and around London.

30679/19-25: While VOC emission ratios are within a factor of 2 of those reported in the NE, it does not follow that VOC emissions are generally consistent. Source strength needs to be accounted for in addition to emission ratios which do not give any indication of source strength. It is recommended that the authors rephrase to limit the statement to emission ratios and not "VOC emissions" in general. In addition see previous comment, and potential impact of C2H2 concentration variability on the similarity between NE US and London VOC emission ratios.

30681/Section 3.3: Other studies have looked at the evolution of size and number concentrations of particles in a MEGACITY (e.g. Kleinman et al. 2009). The Kleinman study finds little change to the location of the size distribution, but an increase in the number of particles in the accumulation mode, corresponding to the increase in mass also observed. Could the authors put the results of this work into context of this and other previous studies?

30682/14-17: Shinozuka et al. (2007) compiles a range of optical properties of aerosols over North America and finds a range of 3.6 +/- 1.3 m2/g. The paper also examines f(RH) from aircraft, and those results should be compared with results of this study.

30683/21-30684/1: The authors should state the uncertainty associated with these regressions based on the discussion in the methods section. In addition figures should be included so that the reader has an idea of the scatter of the data, and the range of that the data covers. In the discussion of sub-micron mass to BC ratio, there is no correlation coefficient given, and terms like "roughly" and "as much as" with no quantitative information leaves the reader wanting either a figure so that (s)he can evaluate it or more precision in the language.

30685/20: Is it valid to assume no sulphate source for an urban area? If sulphate was purely regional, and homogeneous in concentration than the plume and upwind concentrations would be the same. From Fig. 9 it is difficult to tell, but it seems that in many cases the background concentrations of sulphate are higher than the plume concentrations. In that case can the authors justify the sulphate normalization?

30687/25-26: Nitrate to CO differences in Mexico City was observed and discussed by DeCarlo et al. 2008, who noted a rapid decrease in NO3/CO with distance from the urban area.

30687/20 - 30688/26: Is the CO a delta CO above background CO concentration or absolute CO concentration. This is not clear to the reader. From table 4, it appears to be a delta value, but there is no discussion of background values of CO, and what procedure was followed to determine them. If it is absolute CO concentration, then the analysis needs to be done subtracting a background CO value from the measured CO value for comparison to other studies which have done this.

30688/14: The measurements of OA/CO is discussed in Kleinman et al. 2008 for Mexico City and also in DeCarlo et al. (2010) with a comparison to several other field measurements, and it would be interesting to see the London results placed into the context of the other field campaigns discussed in these papers.

30690/9-12: The increase of nitrate with height in the PBL has also been observed in other studies in LA (Neuman et al. 2003) and Tokyo (Morino et al. 2006). How do their

results compare with the results of this study?

30692/3: Were the results the dry scattering coefficients or wet scattering coefficient?

30693/2-3: Check units on ratios, should be “ppmv” not “ppbv”.

30705 Table 4: Units are incorrect for OA/deltaCO, should be “ppmv” not “ppbv” Fig. 4: Flight tracks colored by concentration and shown on top model predictions would be useful for the reader to see general model performance. Also, the color scale legend is very difficult to read.

Fig. 5: Please label all axes.

Fig. 6: Seems to be mostly qualitative, and dashed lines are only visible on the G panel, why not on all panels?

Fig. 11: Nitrate appears to be a purple trace in the figure but blue in the legend. Please fix.

References: DeCarlo, P. F., et al.: Fast airborne aerosol size and chemistry measurements above Mexico City and Central Mexico during the MILAGRO campaign, Atmos. Chem. Phys., 8, 4027-4048, doi:10.5194/acp-8-4027-2008, 2008.

DeCarlo, P. F., et al.: Investigation of the sources and processing of organic aerosol over the Central Mexican Plateau from aircraft measurements during MILAGRO, Atmos. Chem. Phys., 10, 5257-5280, doi:10.5194/acp-10-5257-2010, 2010.

Kleinman, L. I., et al.: The time evolution of aerosol composition over the Mexico City plateau, Atmos. Chem. Phys., 8, 1559-1575, doi:10.5194/acp-8-1559-2008, 2008.

Kleinman, L. I., et al.: The time evolution of aerosol size distribution over the Mexico City plateau, Atmos. Chem. Phys., 9, 4261-4278, doi:10.5194/acp-9-4261-2009, 2009.

Morino, Y., et al.: Partitioning of HNO3 and particulate nitrate over Tokyo: Effect of vertical mixing, J. Geophys. Res.-Atmos., 111, D15215, doi:10.1029/2005JD006887,

2006.

Neuman, J. A.,et al.: Variability in ammonium nitrate formation and nitric acid depletion with altitude and location over California, J. Geophys. Res.-Atmos., 108, 4557, doi:10.1029/2003JD003616, 2003.

Royer, P., et al.: Comparison of lidar-derived PM10 with regional modeling and ground-based observations in the frame of MEGAPOLI experiment, Atmos. Chem. Phys., 11, 10705-10726, doi:10.5194/acp-11-10705-2011, 2011.

Shinozuka, Y., Clarke, A. D., Howell, S. G., Kapustin, V. N., McNaughton, C. S., Zhou, J. C., and Anderson, B. E.: Aircraft profiles of aerosol microphysics and optical properties over North America: Aerosol optical depth and its association with PM2.5 and water uptake, J. Geophys. Res.-Atmos., 112, D12S20, doi:10.1029/2006JD007918, 2007.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 30665, 2011.