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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Elucidating multipollutant exposure across a complex metropolitan area by systematic deployment of a mobile laboratory" *by* I. Levy et al.

Anonymous Referee #1

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Levy et al.: "Elucidating multipollutant exposure across a complex metropolitan area by systematic deployment of a mobile laboratory" (acpd-12-31585-2012)

General comments:

This study presents some interesting data and a potentially useful analysis but, in my opinion, it is not publishable in its current form.

First of all the authors should take a clear decision whether they will focus this paper on the exposure issues only or whether they want to discuss it also in relation to applying the new instrument in future epidemiological research. I suggest focusing the paper on the exposure issues only. So far, I didn't learn from this paper how to use the

data in an epidemiological study. In my opinion the only way for using the data in epidemiological research is the estimation of the long term average for specific locations (such as annual averages) and using the estimates for further modelling, for example LUR modelling. However, the estimation of annual averages for this approach could be also done by satellite monitoring sites (as already done in the past) and there is only a limited necessity to change the methodology. Thus, the exchange of the satellite monitoring sites by CRUISER would by nice, but it is not really crucial.

Moreover, the estimation of the annual averages could be conducted by CRUISER only in near-road environments and not in urban background locations (where the study population may also live). This problem doesn't exist for the satellite monitoring sites, which could be located at almost all relevant locations. Furthermore, I have severe doubts whether the described design really allows the estimation of annual averages as concluded in this study (see specific comments).

With respect to short-term epidemiological studies, I don't see any possibility for application of the data in such studies. If the authors really want to postulate the using of CRUISER in epidemiological studies, a clear description of how to use the date in which studies is needed. In this case, also a deeper discussion of the current stage of exposure assessment in epidemiological research is needed.

The problem of air pollutant variability between and within a city is well known in the epidemiology and it was evaluated in many studies (Jerret et al., 2005, Marshall et al., 2008, Brauer 2010, Boogaard et al., 2011, Cyrys et al., 2012, Eeftens et al., 2012). While the small scale variability is well characterized for some pollutants (especially for PM10 or PM2.5), it is not for ultrafine particles. This is the reason that no long-term studies on UFP and heath were conducted until now. It shows clearly that the epidemiologists are aware about the necessity of sufficient characterization of large and small scale temporal and spatial variability for all air pollutants under study. However, some sentences in the manuscript suggest rather the opposite: "Nearby microenvironments may have a wide range in average pollution levels varying by up to 300 %, which may cause large misclassification errors in estimating chronic exposures in epidemiological studies". Without any further evaluation and discussion such sentences are misleading and should be deleted.

The manuscript is very long and it is difficult for the reader to catch the main massages. The whole manuscript should be definitely shortened. Some parts of the results section should be moved to the method section (see specific comments).

The authors state that in this study a number of hypotheses can be explored, for example:

(1) measurements taken by a monitoring network are not representative of all areas within a city and underestimate maximum exposures;

(2) predictions from numerical air quality models at fine grid resolution cannot account for the variability in pollution levels existing within a neighbourhood scale. Both hypotheses are trivial and don't need any further exploration.

Specific comments:

Abstract:

Page 31586, line 10: it is not true that 23 pollutants were measured: 20 pollutants were measured, 3 were calculated (please correct)

Page 31586, lines 14 -17: "This approach allowed linkage of the mobile measurements to the network observations and to generate average maps that provide reliable information on the typical, annual average spatial pattern" this sentence is not true (see comments below)

Page 31586, lines 19 -23: "Nearby microenvironments may have a wide range in average pollution levels varying by up to 300 %, which may cause large misclassification errors in estimating chronic exposures in epidemiological studies" this sentence is not true (see general comments)

2.2 Measurements

Speed correction: apart from the vehicle speed also wind speed should influence the airflow in the inlet. How did the authors adjust for it? What was the R2 for the linear regression between PM (corrected) and PM (original) as stated in the equation? 2.3 Mobile measurement strategy

Page 31592, lines 19-20: The number of measurements per km of road (more than 2000) is really very impressive. But what does it mean? Given that the CRUISER travelled with an average speed of 25 km h-1, it needed about 144 seconds per km and consequently 144 every second measurements were conducted. It means that for achieving of the (apparently) huge number of 2000 observation, only 14 measurement days (or trips) were needed (2000/144=13.9). The authors should consider how to express the number of observations per point or per route in a more common way, for example how many times the route (including the specific road segment) was completed.

I assume that the huge number of observation was achieved only for pollutants measured every second. The number of observation for pollutants measured every 2 minutes is much smaller. It should be indicated in the manuscript.

2.4 Spatial analysis

I have doubts whether a spatial analysis for a given study area could be done based on measurements done not simultaneously and without any adjustment on the temporal variation. First of all, if the measurements are conducted at different times of the day, the results should be different even for the same site, due to the diurnal pattern of air pollutant concentrations. Furthermore, the day-to-day variation of all air pollutants is (in almost all areas around the world) very strongly influenced by meteorological conditions. I assume that this is the case also for Montreal. Therefore measurements conducted at different days and different times of the day are not comparable without any adjustment for the temporal correlation. This adjustment could by done by using a reference site operated continuously during the whole study period. This approach was already very often used and it is described sufficiently in the literature.

3.1 Representativeness of the mobile measurements

Obviously different measurement methods were used by CRUISER and VdM. If so, a direct comparison of the measurement (side by side) is needed before and after the study period. A strong correlation is needed for any further comparison. Without such site by site comparison the interpretation of the results is somewhat crucial, as the authors stated on page 31595, lines 18-19 or page 312596, lines 16-17.

In Figure 2 some scatter plots of CRUISER's vs. VdM measurements are shown. Some scatter plots are showing surprising low correlation between the measurements. For example, the R2 for PM2.5 measurements is 0.60. Given that the CRUISER was operated in close proximity to the AQ sites, it is very low. In our network we observed R2 of 0.90 for both traffic and urban background sites located 3-4 km apart from each other.

I wonder that in the scatter plot for SO2 also values below the limit of detection (1 ppb?) are displayed.

Page 31597, lines 13-26: The whole paragraph should be moved to the method section.

Page 31598, lines 1-10: It is difficult for me to believe that the annual averages could be estimated based on very few and rather short term measurements – some studies on this issue were already published and support this finding. Cyrys et al. (2006) showed that "monthly means" based on 6-7 measurements distributed over a two-week measurement period for each month substantially over- or underestimate the "true" monthly mean values.

The requirement in this study is that "typical days" should be chosen for the measurements (page 31597, lines 28-29). How to find it? What is the definition of the "typical

days". What is "typical" for winter and what for summer season, which days are "typical": rainy, sunny, with low wind speed or rather stormy? I see that the differences between the estimated annual means and the "true" annual means in Montreal are not very big. However, what is the reason for it and could it be expected also in other cities around the world. May be the rather low concentrations (and probably low day-to-day or season-to-season variation) make it possible for Montreal, but in this case the authors should discuss the unique situation in their study region. It might be also helpful to see the time series of the pollutant under study for the whole year 2009 (with indicated time periods of CRUISER measurements).

3.2 Intra-urban variability observed by CRUISER

We know very well that the concentrations of air pollutants in the vicinity of strong local sources are elevated. The whole section is showing that and could be significantly shortened.

Table 2: A/B and A/C might be more interesting for the reader as C/D and B/D (those relationship could be calculated for any monitoring network, without CRUISER).

Figure 3: The differences between the "daytime averages" and "daily averages" are really very small. Taking into consideration the mostly common diurnal pattern of air pollutants, it is somewhat surprising. Are there any explanations for it?

4 Discussion

Page 31606, lines 14-16: It might be interesting for readers from other countries to get to know the requirements for monitoring site location in other parts of the world. So for example clear criteria for siting of the measurement stations are provided by the EU. With respect to the protection of human health, all Member States are required to provide data on the areas with highest concentrations (hot spots) as well as on those being representative for the exposure of the general population (urban background). All parts of the discussion and conclusions related to exposure assessment in epidemiological studies should be corrected by any expert working on this field.

Literature

Boogaard et al.:" Contrast in air pollution components between major streets and background locations: Particulate matter mass, black carbon, elemental composition, nitrogen oxide and ultrafine particle number" Atmospheric Environment 45 (2011) 650-658

Brauer: "How Much, How Long, What, and Where Air Pollution Exposure Assessment for Epidemiologic Studies of Respiratory Disease" Proc Am Thorac Soc Vol 7. pp 111–115, 2010

Cyrys et al.:" Evaluation of a sampling strategy for estimation of long-term PM2.5 exposure for epidemiological studies" Environmental Monitoring and Assessment (2006) 119: 161–171.

Cyrys et al.:" Variation of NO2 and NOx concentrations between and within 36 European study areas: Results from the ESCAPE study" Atmospheric Environment 62 (2012) 374-390.

Eeftens et al.: " Spatial variation of PM2.5, PM10, PM2.5 absorbance and PMcoarse concentrations between and within 20 European study areas and the relationship with NO2 - Results of the ESCAPE project" Atmospheric Environment 62 (2012) 303-317.

Jerret et al.:"A review and evaluation of intraurban air pollution exposure models" Journal of Exposure Analysis and Environmental Epidemiology (2005) 15, 185–204.

Marshall et al.: "Within-urban variability in ambient air pollution: Comparison of estimation methods", Atmospheric Environment 42 (2008) 1359–1369.

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