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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.

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Dear authors,

you reported on multiple atmospheric pollutant measurements across the city of Montreal using a mobile laboratory. The material is original and of sufficient new interest. The subject and scope of the paper clearly fall within the scope of ACP.

After balancing the ups and downs of the manuscript, however, my opinion on this manuscript is still equivocal.

On the one hand, you provide a very extensive body of new data, showcasing multi-

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parameter mobile measurements across an urban atmosphere. The data set collected by you team is, to my knowledge, unusually comprehensive and unique. I am not aware of any similar data set that has been collected across an urban landscape. The data was processed with care, and is certainly useful to assess the various aspects of complex pollutant exposure across a wide stretch of urban landscape. To sum up, the experimental material that has been generated in this work is a real achievement.

On the other hand, I see several hurdles that need to overcome in order to make this an ACP publication.

1) Overall, the paper remains rather descriptive. It lacks a certain scientific rigor, i.e. in the way of condensing the descriptive results into something that is of a wider relevance to the scientific community.

Part of the problem is that the results gained from in Sects. 3 and 4 do not really make it into the Conclusions section.

Another part of the problem is that the results sections themselves are not well balanced. Sect. 3.1, for example, is rather lengthy but yields only some modest results. Sect. 3.2 is interesting, but the discussion remains too much on a descriptive and anecdotal level. I like Sect. 3.3 very much, and that section, for example, could be further expanded.

In addition, I need to state that you did already extract a major part of their campaign results, notably the air pollutant cross-correlations, into another journal article (Levy et al., 2013). Spreading results over several journal articles can, in general, be a good idea. In the current situation, however, I feel that "picking out the cherry" of the data analysis did not particularly help this ACPD article. This article looks a bit like a loose collection of the less intuitive and less attractive results remaining from the entire work in Montreal.

2) The Introduction part gave a fuzzy impression to me. It lacks overview and clarity. I

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will give some detailed suggestions to improve this below.

3) The Conclusions section contains very little in terms of novel results. This is a major point that needs to be fixed.

4) There are certain technical issues (processing methods) that need to be clarified. I see a particular problem with the wind speed correction to the PM measurements.

5) Presentation of the geographical setting and the data can be improved.

As a whole, publication in ACP seems possible once these (major) issues have been addressed.

Specific comments

1) Overall compilation and organization of results

As stated above, the paper provides many descriptive results, but little in terms of condensed results that may be remembered by a reader. The major statement lasting in my memory, after reading the article for quite a while, is that pollution gradients and time development in the urban atmosphere are overwhelmingly complex. This is something that a lot of people know or could have guessed already.

Section 3.1 is rather long, essentially establishing a certain representativeness of the mobile measurements towards a long-term average. I am wondering whether this could be condensed.

The results in Fig. 2 are not really impressive. While I clearly understand that from an experimental point of view, there are many reasons why the data clouds are scattered, I am not sure how this can really "prove" that the mobile measurements conducted during ca. 30 days are representative for the entire year.

Section 3.2 present extensive information on the spatial variability encountered, but much in the form of single anecdotes. It would be helpful to compile a systematic list, what pollutant profiles are associated with which kind of pollution sources (point

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sources and/or line sources). This would also better help justify the manuscript title "Elucidating multipollutant exposure...".

Section 3.3 is very interesting because it highlights the problem of how to experimentally determine the spatial pollution gradients. I wish this section to be expanded a little, yielding a bolder statement on which pollutants are more and less difficult to capture with respect to spatio-temporal variability.

2) Introduction

This article deals with the relevance of ambient particles upon human health. Therefore I am missing some passages which, at least briefly, touch on the following matters:

The relevant dose affecting human health is best approximated by what we call "personal exposure". In certain cases, exposure to outdoor pollutants might make up a minor part of total personal exposure only. Much of this personal exposure by indoor exposure.

Regulatory issues. In your paper you present data on a lot of atmospheric parameters. Which of them are of regulatory concern, and why? Which of them are the currently more problematic pollutants, i.e. those whose ambient parameters exceed legal threshold values often in urban and/or hotspot areas?

Particle epidemiology. I see that you make, with your work, a move towards the determination of long-term exposure. This is perfectly right. Nevertheless, the Introduction would greatly benefit from an brief outline of what type of epidemiological studies exist that could make use of chronic exposure data.

p. 31588, l. 9: "These primary pollutants react among themselves...", unspecific — you speak about reactions of pollutants without having mentioned a single one explicitly.

l. 11: "The quantity formed depends on ambient conditions (e.g., photochemistry...)". Would you call photochemistry an "ambient condition"? Be more specific here, stating ambient conditions that might foster photochemical processes.

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l. 17ff: This paragraph is confusing. You start with a mention of AQ monitoring, then move to LUR modelling, back to monitoring data interpolation before you speak about multipollutant numerical models. Right after that statement you turn to “saturation campaigns” again. According to my impression, the Introduction would become much clearer to a reader if you started, for example, with a clear statement on the health relevance of ambient pollutants, then move over to state-of-the art experimental observations, then to spatial data interpolation, and lastly to exposure modelling (LUR vs. numerical dispersion models).

The scientific community has hopes that AQ models will do a good job in the future to assess spatial variability, because this seems the only way to provide a spatially complete picture of pollutant exposure. Also, your data shows great potential to serve as a reference data set for the validation of pollution models. Therefore, as a reader I would appreciate some more information on the current state and capabilities numerical pollution modelling here as well.

It is, of course, neither possible nor necessary to cite all previous relevant works. Nevertheless, I feel that this paper omits some of the more relevant contributions to the topic of spatial variability of urban pollutants, particularly concerning PM, particle number and size distributions. Therefore, I am suggesting you to consider the following list of studies for possible inclusion in the references.

Avery et al.: Estimating error in using ambient PM2.5 concentrations as proxies for personal exposures: a review, Epidemiology, 21, 215–223, 2010.

Costabile et al.: Spatio-temporal variability and principal components of the particle number size distribution in an urban atmosphere, Atmos. Chem. Phys., 9, 3163–3195, 2009.

Freiman et al.: Urban-scale variability of ambient particulate matter attributes, Atmos. Environ. 40, 5670–5684, 2006.

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Hudda et al.: Inter-community variability in total particle number concentrations in the eastern Los Angeles air basin, Atmos. Chem. Phys., 10, 11385–11399, 2010.

Pinto et al.: Spatial variability of PM2.5 in urban areas in the United States, J. Air Waste Managem. Assoc. 54, 440–449, 2004.

Pirjola et al.: Dispersion of particles and trace gases nearby a city highway: mobile laboratory measurements in Finland, Atmos. Environ., 40, 867–879, 2006.

3) Conclusions section

In its current state, the Conclusions section contains only little in terms of concrete and really novel results. The current text appears like a confusing mixture of study results and rather general statements. I have read this section many times, and arrived at extracting the following three significant conclusions:

p. 31609, l. 6ff: A lot of variability in pollutant concentrations was observed, both spatially, and also among pollutants.

p. 31609, l. 11ff: Mobile measurements collected in particular places during ca. 20 visits per year agree with corresponding annual averages. From this it is concluded that the mobile measurements, although recorded only during limited periods of time, can be used with confidence to approximate longer-term averages.

p. 31610, l. 5ff: More studies to characterize the spatial variability of air pollutants are needed, also using new observation technologies.

The body of these conclusions is not particularly impressive, many of them being common knowledge. In their current state, these conclusions are not sufficient to warrant an ACP publication.

Therefore, in my opinion, the Conclusions section needs to be completely rewritten. I suggest to start by building up some very concrete results from this study, for example mentioning distinct features of the spatial particle distribution, the profiles of particle

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sources, certain peculiarities found with individual pollutants, and highlighting the ups and downs when sampling from a driving car. See also my suggestions related to the Results sections above. In the second half of the section, these results can then be interpreted in the framework of the more general knowledge, and lead into recommendations of what could be done in the future.

4) Technical issues

Major points

p. 31590, l. 14, caption of Fig. 4 and others: : "annual season", "annual mean pollution levels": Calling the total campaign average "annual" average is misleading and should be avoided. I truly accept that you wish to generate from their data something approaching the annual average but as a matter of fact, even the three campaigns cover only part of the entire year (in fact, less than 5% of the time), and nomenclature should in any case respect that situation. We all know how changeable, for instance, PM concentrations can be over the course of a year as a function of synoptic weather. Better candidates might be "campaign average", or "total average".

p. 31590, l. 23ff and Appendix A2: Wind speed correction to the PM measurements

First, the physical units are missing for the factors given on p. 31611, l. 24–25. Therefore I am unable to judge the magnitude of the effect.

Second, I find the concept of this correction quite adventurous! One basic reason is that for its derivation, you use PM data collected up to 500 m apart from the corresponding stop site. This bears some serious contradiction: One the one hand, you are not getting tired in this manuscript stating over and over again how variable airborne pollutants are in this urban environment. At the same time you seem to be happy to pick values as far as half a kilometer from a stop site, possibly recorded half an hour earlier or later, and consider them as equivalent to those recorded at the stop site, with the exception of being recorded at a given driving speed. If I accepted your method as is, you would

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need to agree that your whole experiment should not warrant the discussion of spatial parameter variations at a resolution of higher than 500 meters at all.

Another reason is that the relative particle losses depend on the particle size distribution, which may vary from time to time. At one time you may have more coarse particles compared to fine particles present in the atmosphere, and vice versa. In these two cases, the relative particle mass losses will necessarily be different even if you encountered the same wind speed. The latter reason implies that the current correction can, in the best case, correct for some crude overall effect, and may be highly invalid in single cases.

On the basis of the information currently available, I would strictly decline the usability of this correction. At the same time, the magnitude of the correction might not be overly high. I would be willing to reassess the issue if you provided, for example, a graph of the data cloud upon which the correction was established. Moreover, I would expect a critical assessment of the method, such as an uncertainty involved, and a discussion of the consequences on the interpretation of the measurement results, for example, in the shape of error bars that depend on wind speed.

Table 1, "UFP": It is not acceptable to call a total particle number concentration measurement "UFP", even if some publications might have done so. As you know, the condensation particle counter measures not only UFPs (D $<$ 100 nm) but also other particles up to a maximum diameter defined by the inlet system. I accept the view that total particle number is often dominated by the number of UFPs but still, UFP number and total particle number are not the same! Therefore, please use "total particle number concentration", N_tot or a similar term throughout the entire manuscript.

Minor points:

p. 31586, l. 6 and five more occurrences in the text: "saturation monitoring campaign". I do not completely grasp the meaning of "saturation" in this expression. I just speculate that you mean that somebody measures as long as needed to determine

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some representative state of the atmosphere. However, experience tells that the state of the atmosphere can be very variable when looking at pollutant concentrations. It can, in fact, take years in the atmosphere to determine a climatologically representative average. Therefore, I suggest to replace “saturation monitoring campaign” by the less controversial “intensive monitoring campaign” unless explained in more detail why there should be any “saturation”.

p. 31590, l. 14: “three week deployment”. Please note the periods of the measurements explicitly by date.

p. 31590, l. 17: “23 different species”, replace by “23 different parameters”

p. 31591, l. 17: “(1) East Montreal; (2) Central and West Montreal”. I was trying hard to identify in Fig. 1c which routes you mean. In my eyes, the Island of Montreal seems actually aligned much in a direction from the south to the north. After combining some thoughts I guess that the area in Fig. 1b is what you mean by East Montreal. But this seems to be neither noted in the map, nor in the figure caption, nor in the description of the study area (Sect. 2.1). Therefore, please add labels to the maps where “East”, “Central” and “West” Montreal are. See also below my suggestion to improve Fig. 1.

p. 31592, l. 6: “Figure 1c shows that a large majority of the sampling was conducted in residential areas...”. Sorry, but this cannot be grasped from Fig. 1c. It could be easily seen if there were a topographic map and a concentration map side by side showing exactly the same area. See also below my suggestion to improve Fig. 1.

Table 1, “Response time”: Do you mean time resolution? Please clarify. In environmental technology “response time” is usually associated with the delay time of a particular sensor to react (“respond”) to a change in sample concentration, emphasizing potential limitations in the physical measurement process. “Time resolution” would simply refer to the frequency at which data points are collected.

Fig. 2: Changing “VdM measurements” to “VdM fixed site measurements” would make

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a reader grasp immediately that these are the fixed site measurements.

5) Presentation of the geographical setting and data.

Figure 1: This Figure plate is nicely compiled, but confusing in some aspects.

Fig. 1a and 1b are inconsistent. They use quite different color shades (red) to mark roads. Second, Fig. 1a features black as a color code for certain roads that neither appears in the legend, nor in Fig. 1b. This is quite annoying to a reader and should be fixed.

Fig. 1b contains a corrupted road label “25 & 40”. Please fix this, or delete it.

The legend is confusing in that “CRUISER’s stops sites” and “AQ site” appear below the headline “NPRI emission sources”. These need to be filed under a dedicated headline, such as “Experimental points of interest” or similar.

I guess that the terms “East”, “Central” and “West” Montreal refer so specific urban areas. I found these descriptions in the text, but was unable to locate these areas on the map. Hence, please add corresponding labels to the maps.

There is another issue I discovered: On the one hand, you put a focus on North Montreal in that you zoom into that region (Fig. 1b). On the other hand, this interest is not reflected in a corresponding separate map for particle concentrations. Fig. 1c does this job for Fig. 1a, but it would be insightful to see North Montreal as a dedicated concentration map covering the same area and displayed at the same scale. This would certainly improve the visual orientation on the pairs of maps (geography/concentrations). I like the maps, but had real trouble associating the driving patterns in Fig. 1c with the geographical features displayed in Fig. 1a.

To improve the situation for the reader I suggest the following: Do assemble two Figures, each containing a topographic and a concentration map side by side. The first Figure would cover the area of Fig. 1c, which is the area of interest for the exposure measurements, or maybe a bit larger. The topographic map would be much like Fig.

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1a, the concentration map much like Fig. 1c. This new Figure would provide the overall situation for the entire campaign. Try to make sure that both maps cover exactly the same area. This is important for visual orientation!

A second Figure would provide the situation for the focus area in north Montreal, containing a topographic map similar to Fig. 1b and a concentration map (new) side by side. The advantage over the large-scale map would be the identification of industrial point sources, which are already omitted from Fig. 1a. I believe that the production of two separate Figures would be of excellent service to the readership.

To save some space, the overview map showing Montreal on the continental scale could be dropped. Knowledge of the location of Montreal might be widespread enough.

References:

Levy, I., Mihele, C., Lu, G., Narayan, J., and Brook, J. R.: Evaluating Multipollutant Exposure and Urban Air Quality: Pollutant Interrelationships, Neighborhood Variability, and Nitrogen Dioxide as a Proxy Pollutant. Environ. Health. Perspect. http://ehp.niehs.nih.gov/1306518/, 2013.

Wolfram Birmili, December 18, 2013

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 31585, 2012.