Atmos. Chem. Phys. Discuss., 11, C6033–C6038, 2011
www.atmos-chem-phys-discuss.net/11/C6033/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Variations in time and space of trace metal aerosol concentrations in urban areas and their surroundings" *by* T. Moreno et al.

Anonymous Referee #3

Received and published: 6 July 2011

Recommendation: Major revision.

I have not looked at the comments by the other referees to avoid bias in my reviewing. I'm sorry beforehand if there should be large overlap between my comments and the comments of the other referees.

General comment:

The authors present an analysis of metals in aerosol particles at a rural background station (MSY) close to Barcelona, Spain, and at an urban background station in Barcelona (BCN) in the DAURE campaign. The authors have 1-h data from winter and therefore they are able to analyze diurnal profiles of the trace metals, which are

rarely published elsewhere. The analysis is promising, and the authors have found several sources for their elevated metal concentrations. However, the analysis is not quantitative and the detection of sources is speculative. More sources might exist than the authors have found. Here, I stress performing a source/receptor run, which should give further clues which sources should contribute and what are their relative contribution to metal levels.

Language: Excellent.

Outline: Excellent.

Specific suggestions:

The source/receptor run should be made with for example PMF to start with on the 1-h resolved winter data. With this factor analytical engine the authors are able to try different number of sources to see how many should contribute to high metal levels or certain metal tracers. The modeling should be made without PM levels to start with, since the main focus of the paper is the source contribution to metal levels and not PM. The modeling should be made for MSY and BCN separately, since their source contributions are different during different times of the day as the authors have pointed out. The modeling should be made with the PM1, PM2.5-PM1 and PM10-PM2.5 elemental concentrations in the same modeling run or in different runs depending on the success of either approach. Using the knowledge from this first PMF run, the authors can continue to steer their modeling into a direction with more constraints on some of the source profiles (closer to a chemical mass balance model) with for example the multi-linear engine (ME-2) or by adjusting the different parameters of the PMF model. The co-authors of the current paper have proven their ability in this area before (Amato et al., 2009). The modeling would yield an improved quantitative estimate of how the different sources contribute as function of time for the current winter period. The results and source profiles acquired should be displayed in tables. The analysis should also be compared to either wind roses or trajectories to be able to understand the origin of

the different sources. The DAURE campaign atmospheric interpretation can be of help here (Jorba et al., 2011).

This is not the main focus of the current study, but in my opinion the following should also be performed given the unique opportunity that the authors have: You should include also PM levels to be able to perform another source/receptor run, where you quantify the contribution to PM1 and the coarse fraction of PM10 (for example PM10-PM2.5) for each hour of the day for the two sites separately using the 1-h winter metals as tracers for different sources. Then, also PTRMS, AMS, and 14C data should be used to assist the identification of additional sources of PM for those sources that the metal tracers are not able to catch. It might turn out that the results from the second modeling are not far away from the first source receptor modeling without PM. However, this is impossible to predict. If the authors are able to quantify source contributions from the second attempt, also the magnitude of PM1, PM2.5 and PM10 during the current winter period should be compared to the average winter data from the continuous measurements in urban background Barcelona and at MSY. This data is available (see for example Pey et al., 2009, and Amato et al., 2009). The motivation for this comparison is that it is important to see whether the current winter PM and metal levels are unusually high or low.

I completely agree with the authors that the seasonal variation is important to study. However, the continuous data sets from MSY and from urban background Barcelona are much better suited for this purpose (both the elemental concentration and PM2.5 and PM10 are available on a continuous basis as seen in Pet et al., 2009 and Amato et al., 2009). The average and maximum values from the current winter and summer data available from the DAURE campaign are not necessarily representative of a typical summer and winter. Hence I suggest leaving out or significantly shortening the first to fourth paragraph of the 3.1. section. This should be a topic of a different paper, since it includes different data. In the fifth paragraph on the other hand, a discussion is made about the 12-hour sample variation, which cannot be retrieved from any other data set,

and hence should be included in the paper.

After the major changes made, the structure of the discussion section and abstract should naturally be modified.

Other Specific comments or technical corrections:

1. Please define the REE acronym when it is written for the first time.

2. Section 3.2. A very important section. The “single morning peak” at BCN is attributed mostly to industrial metallurgical emissions and reversal of wind direction as the morning sea breeze recirculates pollution back to the city. Isn’t it possible that another explanation is that this activity is taking place during day hours, and therefore is not seen in the evening rush hour? The first source/receptor and meteorological analysis should shed more light to this question.

3. Section 3.2. I’m sorry I don’t understand your discussion about the Zn diurnal pattern relative to the the Cu and Sb pattern at BCN. They look the same to me (although Zn pattern is for PM1 fraction, whereas Cu and Sb pattern is for coarse particles).

4. Section 3.2. Ti, Zr, Sr and Rb do not conform to either pollution patterns as the authors point out (“single morning peak” or “double peak”). Again, the first source/receptor modeling run and a meteorological analysis might reveal why.

5. Section 3.3. Again, the first source/receptor run would improve our understanding and quantification of fine and coarser metal sources since it is made for the fine and coarser metal fractions (not for the summer data though, since it is not included in the source/receptor modeling). We would also get a more precise grouping of elements in the source profiles for the different size fractions.

6. Methodology section. Please put information in a table, which time resolution and which size fraction were measured in the winter and summer campaigns respectively. It is hard to understand this otherwise.

7. Section 3.4. Metalliferous pollution episodes: Would you please explain what kind of industry this comes from? Has this kind of pollution source been studied before? Would you mind try to see if there are any data in literature to compare with. Sorry to say this once again: The grouping of species in the source profiles would again be improved with the source receptor modeling.

8. You discuss ship traffic and V/Ni ratios which is very interesting. Reche et al. (2011) show that there is really an influence of shipping emissions on the aerosol concentrations. You claim that the average ratios of V/Ni is rather low indicating industrial smokestacks or shipping emissions close to land where fuel controls apply. But maybe occasionally the ratio is higher indicating the use of cheaper bunker oil. This could also possibly be resolved using the 1-h resolved source contribution estimates from the source receptor runs.

9. The tables 1 and 2 might become less important when you have the results from the source receptor runs. In any case, the tables should be larger and be shown over a whole page in landscape format.

10. Several of the figures will become obsolete after the source receptor run and replaced by other graphs. I'm thinking mainly about Figures 2,3, and 5. Anyhow, please avoid analyzing results in the figure captions, which you have done in figures 3 and 5.

References:

Amato, F., Pandolfi, M., Escrig, A., Querol, X., Alastuey, A., Pey, J., Perez, N., and Hopke, P. K.: Quantifying road dust resuspension in urban environment by multilinear engine: a comparison with PMF2, Atmos. Environ. 43, 2770–2780, 2009.

Jorba, O., Pandolfi, M., Spada, M., Baldasano, J. M., Pey, J., Alastuey, A., Arnold, D., Sicard, M., ArtinËIJano, B., Revuelta, M. A., and Querol, X.: The DAURE field campaign: meteorological overview, Atmos. Chem. Phys. Discuss., 11, 4953–5001,

doi:10.5194/acpd-11-4953-2011, 2011.

Pey, J., P′ erez, N., Castillo, S., Viana, M., Moreno, T., Pandolfi, M., L′opez-Sebasti′an, J. M., Alastuey, A., and Querol, X.: Geochemistry of regional background aerosols in the Western Mediterranean, Atmos. Res., 94, 422–435, 2009.

Reche, C., Viana, M., Moreno, T., Querol, X., Alastuey, A., Pey, J., Pandolfi, M., Prévôt, A., Mohr, C., Richard, A., Artiñano, B., Gomez-Moreno, F.J., Cots, N. Peculiarities in atmospheric particle number and size-resolved speciation in an urban area in the western Mediterranean: Results from the DAURE campaign, Atmospheric Environment (2011), doi: 10.1016/j.atmosenv.2011.06.05

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 14747, 2011.