**Responses to interactive comment on "Spatial variation of chemical composition and sources of submicron aerosol in Zurich: factor analysis of mobile aerosol mass spectrometer data" by Referee #3**

*The manuscript by Mohr et al. presents an interesting study of the aerosol composition in and around Zurich. On-road and stationary measurements were made to explore the spatial variation in the aerosol composition and factor analysis was used to identify distinct organic aerosol factors that could be correlated with traffic emission sources (HOA), wood burning and heating (WBOA), and oxygenated organic aerosol (OOA). The measurement and analysis methodologies used in this work are of interest and I recommend publication after the following questions are addressed:*

We thank Referee #3 for his or her comments. Many points that were referred to have also been mentioned by Referee #1 and Referee #2. We will address them here again for completeness.

*General Comment:*
*1) Discussion of Background versus local contributions:*
*- The authors' state that they are using "urban background" interchangeably with "regional background", but this is confusing. The measurements that are presented can be used to asses the spatial variability of chemical composition and "local" sources in and around Zurich above the urban background value in Zurich. However, I do not think that this can be automatically extended to assess the "regional background" which would require more measurements both north and south of the city in background rural sites. The PM10 comparisons made between Zurich Kaserne and Payerne and Tanikon are not necessarily reflective of the much smaller PM particles measured with the AMS.*

The corresponding paragraph in section 3.5.1 was changed as following: "Lenschow et al. (2001) and Querol et al. (2004) showed that in a city area, the background PM concentration is elevated compared to the regional background PM concentration ("urban background"). However, the meteorological conditions during the period of mobile measurements favored a uniform distribution of pollutants throughout the Swiss plateau and thus a homogenization of background air being it urban or rural".
We agree with Referee #3 that $PM_{10}$ does not necessarily reflect $PM_1$ measured with the AMS. As was mentioned in the responses to Referee #1's comments, dust could be significantly different from $PM_1$ in many aspects. However, dust concentrations are expected to be low in Zurich during winter time due to frequent rains (~30% of $PM_{10}$ (Richard et al., 2011), much lower than e. g. in Barcelona (Amato et al., 2011)). Unfortunately, $PM_{2.5}$ data are only available in Payerne and Zurich Kaserne (there starting from 1 January 2008 only), and $PM_1$ data are only available for the reference station Payerne. Also $PM_{2.5}$ data indicate that the assumption of an homogeneous background pollution for the Swiss plateau is appropriate: Average values for 14 – 16 December 2008 (the days of mobile measurements) of are 24.3 µg m$^{-3}$ in Payerne and 25.5 µg m$^{-3}$ at Zurich Kaserne.
The $PM_{2.5}$ comparison was added to the manuscript.

*- Top of page 12342, the authors state "it seems that even though the city of Zurich is a major emitter of air pollutants, the beforehand mentioned meterological conditions and the high, well distributed emissions of the densely populated Swiss plateau area lead to a homogenization of background air being it urban or rural" Are the authors trying to say*

*that the "urban background" in Zurich is due to regional sources outside Zurich? It is not clear that the case for this has been made by the authors.*

This sentence was deleted as it was misleading. As discussed for several points made by Referee #2, we refer to Zurich Kaserne as an urban background which, for specific components of $PM_1$, is enhanced compared to the Swiss plateau background (while the total PM concentration is remarkably similar over this area).

*- Have the authors tried using CO as the inert dilution tracer in place of SO4 in equation 4? If so, what were the results? If not, it would be useful to mention why not.*

As stated also in the responses to the main points of Referee #2's comments, it was not our intention to normalize secondary products to primary components such as CO to account for dilution. This approach is usually used for the characterization of city plumes, to investigate what happens with emissions and pollutants downwind of a city (see e.g. DeCarlo et al., 2010). As stated by Referee #2, we are using sulfate, which is being formed more slowly than other secondary components, to normalize out inhomogeneities due to small-scale meteorological effects. Primary emissions and chemistry are then the only two reasons left for differences in background pollution and "local" pollution. The quantification of the background fraction and the locally emitted or formed fraction of $PM_1$ is the main goal of this study.

*Specific Comments*
*1) Since a significant amount of the analyzed data was obtained with mobile on-road measurements, one would expect that there was significant contamination from single vehicles. It is not clear from the manuscript how the authors dealt with these contaminations in the data stream. Were the contamination datapoints removed before PMF, for example? This would be an important step to describe in the preparatory data analysis section 2.4.2, particularly since the Q-AMS would suffer from particle counting statistics as mentioned in line 8 on p. 12338.*

We do not expect significant contamination from single vehicles for 2 reasons: 1. As shown in Fig. 2, the main inlet of the mobile laboratory is above tailpipe emissions. 2. The original data acquired with a time resolution of 6 s were averaged to 1 min, smoothing out possible contamination points.

*2) p. 12331, line 19. The authors mention that they use a CE of 0.85 based on comparisons with other instruments. What value would be chosen if the AMS composition was directly used to determine a CE based on the previous lab results of Matthew et al.? What CE was used for the HR-ToF-AMS?*

Compare also point 9 by Referee #1 and point 17 by Referee #2: We agree that the HR-ToF-AMS data has the same CE issues as the Q-AMS, however, once CEs have been determined independently, comparing mass loadings of two AMS is still a valid quality check. The corresponding paragraph in the manuscript was changed as following: "The CE of the Q-AMS was determined to be 0.85, based on a comparison of the mass concentrations of the sum of Q-AMS and MAAP data (non-refractory $PM_1$ plus BC) with a tapered element oscillating microbalance (TEOM) monitor (Thermo). This comparison was performed in the time period of 27 November 2007 to 10 January 2008. An Aerodyne high-resolution time-of-flight (HR-ToF-AMS) was measuring at the background site during December 2008. The CE of the HR-ToF-AMS was determined to be 1

(Richard et al., 2011). Q-AMS (CE = 0.85) and HR-ToF-AMS (CE = 1) agreed very well (maximum difference 10% for all comparison periods). A high ammonium nitrate fraction (42% of total $PM_1$ mass on average) and possible high aerosol water content due to a lack of a specific drying unit in the sample air (see section 2.3) support the relatively high Q-AMS CE. Comparisons of various AMS datasets from all over Europe within the EUCAARI/EMEP project show that ammonium nitrate fractions of > 0.25 can already lead to a CE above 0.5 (Nemitz et al., 2011, in preparation). Interestingly, Hildebrandt et al. (2010) found, based on the method by Kostenidou et al. (2007), exactly the same CE for this instrument, when it was operated in Crete".

*3) p. 12332, line 20. Since this study did not use ME, which would allow fixing of mass spectral profiles, it is not clear what they authors mean by "forcing one factor (OOA) in the solution to have the mass spectral characteristics of the fragmentation table dependencies for m/z 44 was just sufficient a priori information to yield an atmospherically relevant PMF solution"?*

This sentence was deleted (compare also response to point 20 of Referee #2).

*4) Figure SI-9 and associated discussion. The reasoning for why the Fpeak=-0.1 solution was chosen should be clarified. Was the choice based on getting m/z 60 fractions to be in a range that is representative of burning spectra? How did the correlation between the components and component tracers (i.e. CO, NOx etc.) change with Fpeak?*

Since the separation of OOA and WBOA was difficult for PMF (compare also point 27 of Referee #2 or point 21 of Referee #1), Fig. SI-9 was in the first place one of the most important tools to find a meaningful solution. As there was a very limited choice of meaningful factors depending on *fpeak* (basically the solution presented here), correlations of time series for solutions with other *fpeaks* were not investigated.

*5) Figure SI-12: Why are the scaled residuals for m/zs greater than 200 so small compared to the rest?*

The "scaled" residuals are scaled to the uncertainties of the data matrix or input matrix. The signal-to-noise ratios decrease with increasing *m/z*'s, the uncertainties become relatively bigger than the residuals from the fit and thus the scaled uncertainties become smaller.

*6) Figure 1: It would be useful to see the overview values and statistics for all the aerosol species as well (SO4, NO3, OOA, HOA, BC, etc.)*

This information is already given in Fig. 3 and would therefore be redundant here.

*7) Figure 5: The f44 and f43 values measured for 14.12.2008-16.12.2008 do not seem to be well reproduced by the PMF components for part 2. Several of the measured points have f43<0.05, but none of the components have low enough f43 values to account for this. Are these low f43 points noisier (i.e. weighted less in PMF) than the others?*

Yes, the part 2 data (also the PMF input matrix) is noisier than the part 1 data (see section 2.4.2).

*8) Figure 6: It is hard to get an overview of how the absolute loadings vary with the type of site (residential, urban etc..) from this figure. It would be useful if the authors added a bar chart showing the loading vs. site with the sites arranged according to classification type. Separation of the bars according to primary and secondary organic and inorganic contributions will also be useful.*

Since in Fig. 6 it was more important to show the spatial distribution, we decided to visualize the information with pie charts on a map. In Fig. 7, bar charts are used.

*9) Figure 10: It would be useful to see how NO3, OOA, and SO4 vary in this figure as well. If I understand correctly from the discussion in the text, most of the BC appears to correlate with the vehicle emission source. So, why is the BC trend with ambient temperature and wind speed different from the HOA trend?*

As stated in section 3.5.2, correlations are weak. The BC trends are more distinct than the HOA trends. We assign this to the more inert nature of BC than HOA, and possible interferences from other sources than traffic to HOA.
Since local contributions from secondary species were so low, we refrained from plotting them in this figure.

**References**

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