

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 #1

*This paper focuses mobile measurements of submicron particles in the city of Zurich. PMF was used to derive the components contributing to the organic mass. Three factors representing HOA, WBOA, and OOA are found, with OOA dominating organic mass, consistent with AMS studies conducted in other parts of the world. Measurements at various locations in Zurich suggest that WBOA, OOA, and inorganic components are uniformly distributed, and the background concentrations contribute up to 97% of the WBOA and OOA, and more than 94% of the inorganic components. This result of uniform distribution of pollutants is not surprising, given that all the measurement points are confined within an area smaller than a 10km * 10km region. A strong point of this paper is the discussion of separating the “background concentration” from the “local emissions”. This is an important paper that evaluates the major organic contributions in an urban area in winter; it merits publication but I recommend a number of clarifications to make the work, and the findings, well documented.*

The authors thank Referee #1 for his or her thorough review of the paper. We will address first the main critical points and then reply on the specific comments individually further below.

Main Points: The “background site” needs more justification/clarification. It can be seen from Fig. 6 that the “background site” lies in the middle of downtown and is very close to the on-road sites (perhaps a few hundred of meters from the roads), how is this site “shielded” from emissions?

Referee #1 is right, the background site lies in the city center. Important information justifying its qualification as “background site” was missing in the text. The background site is a courtyard, surrounded by walls, hence “shielded” from direct emissions. In addition, the roads on the four sides of the courtyard, on the other side of the walls, are in residential areas, with only minor traffic density. The site has been characterized in previous studies as “urban background” for PM₁₀, PM_{2.5}, and PM₁ and additional air quality parameters (Gehrig and Buchmann, 2003; Hueglin et al., 2005; Szidat et al., 2006; Bukowiecki et al., 2010; Lanz et al., 2008). This information was added in the manuscript.

On page 12345, line 7, it says “the local contributions amount to 20 and 10% for NO_x and CO”, indicating the “background site” is nearly the same as the on-road site in terms of NO_x and CO pollutions, which suggests this site may not be as good as a background.

We consider average local contributions of 20% for NO_x and 10% for CO to be significant. It has to be taken into account that “on-road” measurements do not only include major traffic arteries, but also roads in residential areas with little or no traffic volume. For CO, which is more inert than NO_x, higher background levels and hence lower local contributions are expected. Referee #1 is right that the local contributions from NO_x seem low, especially when compared to the local contributions from HOA and BC, also emitted from traffic. Important to note here is the fact that CO and NO_x measurements were only added in December 2008 (part 2), days with generally lower local contributions than during the first period of measurements. Mean local contributions

of HOA and BC for this same period only are ~20% and ~30%, respectively, lower than their mean values for the whole data set. This important information was added to the caption of Fig. 8.

Also, on page 12341, line 16, it says on “its PM1 chemical composition is different from that measured on-road”, but the composition of this site (Fig. 6) looks a lot like several on-road sites, e.g. the “main station 23” site.

As was stated earlier here and in section 2.1 of the manuscript, “the route was chosen such that major traffic arteries, residential areas, suburbs, and surrounding hills were covered”. We can therefore expect sites of higher altitude or residential areas with fewer emission sources to be dominated by background air and thus show a PM₁ composition similar to the urban background site. E. g. the “main station” site has higher contributions of BC (21% versus 15% at the urban background site) and HOA (12% versus 6% at the background site).

It seems papers suggesting this site is a background site are based on PM10 and elemental measurements, from which dust could contribute large fractions, while PM1 could be significantly different from dusts in many aspects. (Was dust measured?)

We agree that dust could be significantly different from PM₁ in many aspects. Dust concentrations are expected to be low in Zurich during winter time due to frequent rains (~30% of PM₁₀ (Richard et al., 2011), much lower than e. g. in Barcelona (Amato et al., 2011)). Dust was not measured during this campaign. Earlier papers suggesting this site as a background site were focusing on both PM₁₀ and PM_{2.5} (Gehrig and Buchmann, 2003; Hueglin et al., 2005; Szidat et al., 2006; Bukowiecki et al., 2010). Lanz et al. (2008) were the first ones that used AMS measurements and thus described PM₁.

Secondly, separation of the local emissions from the background is based on the assumption that “secondary formation and primary emissions of SO₄ are negligible during the course of one round trip (~1h)”. If sulfate is formed slowly (by OH oxidation only) as assumed, it would be probably more regional and may not be a good local tracer.

There seems to be some confusion. According to the concept of the background estimation described in section 3.5.1, we assume that “secondary formation and primary emissions of SO₄ are negligible during the course of one roundtrip (~1 h)” and that “differences in SO₄ between Kaserne and measurement point *p* are purely due to meteorological effects”. Our background estimation is the difference of measurement at point *p* to measurement at the background site, corrected for above mentioned meteorological effects using SO₄. SO₄ is not meant to serve as a local tracer, but indeed as a background tracer.

Also, sulfate could be formed much more rapidly in aqueous phase oxidation, especially given the high RH during the study (70% on average), the assumption of fast sulfate formation may not be applicable.

Referee #1 is right that aqueous phase oxidation of SO₂ by aqueous reactions with hydrogen peroxide, ozone, organic peroxides and a number of other oxidants is much more rapid (Kreidenweis et al., 2003). However, a pre-requisite is the existence of a

condensed phase, which was not given during the measurement drives (measurements in fog/clouds were avoided).

Thirdly, this method of separating background concentration may be more applicable for primary emissions but not very good for secondary components. Studies have shown that more O₃ and SOA could be formed in rural sites, thus the “background concentration” may be resulted from fast oxidation rather than real “background” values. This explains that the background HOA is 60% and nearly all secondary components are from “background” contributions.

We agree with Referee #1 that methods for separating background concentrations may be more easily applicable for primary emissions. However, we think that the concept as it is presented here applies also for secondary compounds. Opposed to most other studies investigating urban emissions, our goal was not to assess emissions of the city and their fate (i. e. to follow the plume), but rather to differentiate between direct emissions from the city itself (including more rapidly formed secondary compounds) and from emissions/compounds in the background. To this background the city's emissions of course also contribute, but on a longer timescale than one hour (the duration of 1 round trip), therefore they belong to “urban background” as well. With this concept in mind we agree that more O₃ and SOA could be formed in rural sites and that almost all secondary components are from “background” contributions. Compare also responses for Referee #2.

Lastly, on page 12339, line 22, it says the OOA factor identified in this study is “rather fresh, minimally processed OOA”, this combined with the argument “nearly all OOA is from background” suggests the background OOA is “fresh”, this does not make sense for background aerosols, which are in concept more aged since they exist for longer time.

The OOA factor found here resembles the LV-OOA spectra found in other studies (e. g. Ng et al., 2010). However, its f_{44} suggests it to be “rather fresh, minimally processed OOA”. We are not referring here to the SV-OOA produced immediately after the primary compounds were being emitted into the air (Chirico et al., 2010; Heringa et al., 2011), but to OOA which had time to process and age for several hours up to several days and to be distributed in the Swiss plateau air basin. Nevertheless, Swiss winters are dark and wet, thus degrees of oxidation which can be found e.g. at the coast of Greece (Hildebrandt et al., 2010) will not be reached.

We deleted “rather fresh” from the sentence as it might be misleading and changed “minimally processed” to “moderately processed”.

The PMF solutions need more justifications. Some discussions are not very clear.

Indeed, a thorough discussion on how the factors were identified is missing. Additional justifications for the chosen PMF solution were added in the manuscript (see responses to point 8 in specific comments below).

Specific comments are listed below:

Specific comments:

1. The title should specify “in winter”, since all measurements were conducted in winter as this is a relatively unique and important aspect of this study.

Title was changed to "Spatial variation of chemical composition and sources of submicron aerosol in Zurich during wintertime using mobile aerosol mass spectrometer data"

2. Page 12324, line 1: should specify the diameter is "vacuum" aerodynamic diameter.

PM₁ is defined with the aerodynamic diameter, we prefer not to redefine the term. In section 2.3, where the AMS is explained, "vacuum aerodynamic diameter" was added.

3. Page 12324, it says "domestic wood burning is more important for organic" because the contribution is 8-15%. This is nearly the same as traffic emission contributions 7-14%, I don't see how wood burning is "more important".

The values given here are the ranges of percentages for various sites within the city (see Figure 6). The average values of all percentage values (10.5% for WBOA, 8% for HOA) were added, and the sentence was changed to "[...] domestic wood burning makes up between 8 – 15% of PM₁ for different sites within the city (average all sites 10.5%)".

4. Page 12326, first paragraph: it seems very sudden the author starting to talk the health effects. This paragraph is suggested to be moved before the second paragraph on page 12325.

The introduction was changed accordingly.

5. Throughout: SO₄, NO₃ etc. should be "SO₄²⁻" and "NO₃⁻", or "sulfate" and "nitrate", the same for other inorganic components.

We agree with Referee #1 that this would be the chemically correct notation. However, e. g. nitrate measured in the AMS can be both of inorganic and organic origin (organonitrates are added to the nitrate fraction in the AMS), and thus NO₃⁻ would not be the correct terminology. Thus we decided to introduce the chemical notation in the beginning, but will then use the "AMS acronyms" for the inorganic species measured with that instrument.

6. Page 12329, line 9: how the PAH is measured should be introduced.

This sentence was misleading, PAHs were assigned to total organics by the analysis software of the AMS and hence not determined separately. The corresponding part of the sentence was deleted.

7. Page 12329, line 27 to page 12330 line 2: this should be removed since measurements from this instrument is not discussed.

The corresponding lines were removed.

8. Section 2.4.2 should be moved to Section 3.2 "PMF results" since it describes the preparation of PMF inputs.

We agree with Referee #1 that section 2.4.2 already uses some information from the PMF results. However PMF is an iterative process, and results from previous runs

helped preparing the final PMF input matrices. We therefore decided to keep the order of the sections as it is now, but changed the title of section 2.4.2 to “Preparatory data analysis for PMF” for clarification.

9. Page 12331, line 19: the CE of 0.85 is derived from comparing the Q-AMS measurements with the HR-AMS and SMPS/TEOM. The HR-AMS has the same CE issue as the Q-AMS and cannot serve as a “standard” for determine the CE. Are these two measurements off by 15%? SMPS and TEOM measure total PM₁, while the AMS only measures the “non-refractory” components, how the comparisons were done? Details should be given how CE is determined.

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 PM₁ 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 PM₁ 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”.

10. Page 12331, line 20: it says “a lack of drying the sample air”, while on page 12329, line 19, it says “thus functioning as a drier for the purpose of this study”, are these at odds?

We changed the sentence to “due to a lack of a specific drying unit in the sample air”. Since the temperature inside the van was considerably varying, and not monitored, the drying of the sample air cannot assumed to be constant and/or steady. In this sense we also changed the description of the drying of the sample air due to temperature differences to “However, the temperature difference between outside and inside the mobile laboratory has a (varying) drying effect on the sample air, as it reduces relative humidity (RH) inside the van (e. g. for campaign average values from 79% RH at 2°C outside to 33% RH at ~15 °C inside)”.

11. There is no discussion how the factors are identified. Since the majority of the analysis is based on the PMF factors, the justification of the factors should be emphasized. There is a lot of discussion of the mathematical aspect of the PMF solutions, but should justify why the “HOA” factor is HOA, etc.

The following paragraph was added in section 3.2: “The HOA factors show high signals of the ion series C_nH_{2n+1}⁺ (*m/z* 29, 43, 57, 71,...) from saturated alkanes, and C_nH_{2n-1}⁺ (*m/z* 41, 55, 69,...) from alkenes plus cycloalkanes. Diesel exhaust is typically dominated

by recondensed engine lubricating oil and consists mainly of n-alkanes, branched alkanes, cycloalkanes, and aromatics (Canagaratna et al., 2004). Correlation of the HOA factors found here with the standard HOA mass spectral profile from Ng et al. (2011) yields R^2 values of 0.98 and 0.9 for part 1 and part 2, respectively, showing their high similarity. The factors identified as OOA have their dominant signal at m/z 44, which mostly represents CO_2^+ from the thermal decomposition of organic acids (Alfarra, 2004). This is in accordance with previous findings suggesting that the fraction of m/z 44 in the organic spectra increases with photochemical aging (Ng et al., 2010). Comparisons with the standard OOA spectrum from Ng et al. (2011) show high similarity (R^2 of 0.97 and 0.98 for part 1 and part 2, respectively). m/z 's 17, 18, and 28 were removed from the fit due to differences in the frag table used here (Aiken et al., 2008). The presence of significant signal at m/z 60 in the WBOA spectra helped identify their origin, since m/z 60 is attributed to $\text{C}_2\text{H}_4\text{O}_2^+$, a fragment of levoglucosan which in turn is a pyrolysis product of cellulose (Alfarra et al., 2007). R^2 of 0.83 and 0.63 were found for the correlations of the WBOA factors with the standard biomass burning (BBOA) spectrum from Ng et al. (2011) for part 1 and part 2, respectively".

12. Page 12332, line 15, it says "m/z's directly proportional to m/z 44 were not downweighted, since downweighting of those variables led to no feasible PMF solution (see Supplement SI, Fig. SI-3)." I found the figure but there is no explanation. I don't see why this solution is not "feasible". Not downweighting the m/z 44 related m/z will give more weight of m/z 44, how is the issue dealt with? Keeping or not downweighting m/z 28 also resulted in the difficulty in separating m/z 28 and m/z 29 as mentioned on Page 12335, the last paragraph.

The following text was added to supplementary information, section 1: "When downweighting m/z 's directly proportional to m/z 44, the PMF solution does not fully separate OOA and WBOA. m/z 's 60 and 73, markers for BBOA (Alfarra et al., 2007), show up in Factor 2, which resembles OOA with the dominating signal at m/z 44 (see also section 3.2 in the manuscript). As for the corresponding time series (not shown), both Factor 1 and Factor 2 follow periodically the time series of inorganic secondary components, but no consistent comparison can be done".

We agree with Referee #1 that not downweighting m/z 's directly proportional to m/z 44 gives more weight to m/z 44. The following information was added to section 2.4.2: "Increasing the weight of m/z 44 by a factor of 2.24 (the square root of the number of m/z 44-related ions, Ulbrich et al., 2009) relative to normally calculated errors was just sufficient a priori information to yield an atmospherically relevant PMF solution".

13. The figures of supplemental materials are not referenced in order.

The order of the figures of supplemental material was changed accordingly.

14. Page 12332, line 20, the sentence starting with "Forcing one factor. . . . PMF solution" is not understandable.

See response to comment by Referee #3, point 20: The sentence was rephrased to "Increasing the weight of m/z 44 by a factor of 2.24 (the square root of the number of m/z 44-related ions, Ulbrich et al., 2009) relative to normally calculated errors was just sufficient a priori information to yield an atmospherically relevant PMF solution".

15. Page 12332, line 26: it says to run PMF separately because of the time gap and “varying instrument performance”, please specify how the performance varies.

Compare section 3.2, the major difference was due to a bad resolution in the quadrupole for the second part. This additional information was added.

16. Page 12333, line 1: what is the “model error”?

The model error refers to an additional error term in the data uncertainties estimation which is proportional to the signal. An explanation was added.

17. Page 12333, line 21: the decrease of number concentration could be explained by “different traffic conditions on that day”, but could “different traffic conditions” result in different PM₁, PAHs, and CO₂ concentrations as well?

We agree with Referee #1 that different traffic conditions would also result in different CO₂ concentrations. We deleted “or different traffic conditions on that day” from the sentence and explain increasing PM₁ concentrations with coinciding decreasing number concentrations by particle growth through coagulation/condensation only.

18. Page 12334, line 19: the “breakup of the inversion”, how is this justified and does this “breakup” occur on other days?

Temperature differences between urban background site Kaserne and Üetliberg (altitude 800 m a.s.l., compare Fig. 6) do not show daily inversion breakups, but a clear inversion breakup on 16 December 2008 around 4 pm due to weather change. As is stated in the manuscript, on 16 December 2008 we observed a change in meteorological conditions (the start of a frontal passage including rain). We added the information on the frontal passage in the text.

19. Page 12334, line 23: this paper mentioned several times PM₁₀ measurements in the text and the supporting information, should specify how PM₁₀ is measured. As mentioned above, PM₁₀ measurements could not represent the PM₁ measurements.

PM₁₀ was gravimetrically determined by the Swiss National Air Pollution Monitoring Network (NABEL) using quartz fiber filters in HIVOL samplers (Digitel, Switzerland). Unfortunately, PM_{2.5} data are only available in Payerne and Zurich Kaserne (there starting from 1 January 2008 only), and PM₁ data are only available for the reference station Payerne. Also PM_{2.5} data indicate that the assumption of a homogeneous background pollution for the Swiss plateau is appropriate: Average values of PM_{2.5} for 14 – 16 December 2008 (the days of mobile measurements) 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.

20. Page 12336, line 3: what is “factorrespective fractions”?

The fractions of *m/z* 29 of each factor. The sentence was changed accordingly.

21. Page 12336, line 13: it seems the choice of the WBOA factor is based on *m/z* 60, but the *m/z* 60 fraction in the selected solution seems very low, can it be used to justify the WBOA factor?

See paragraph added in response to point 11. As stated by Ng et al. (2010), the presence of m/z 60 suggests wood (or biomass) burning organic aerosol. The R^2 values for the correlation of the WBOA spectra with the standard BBOA spectrum from Ng et al. (2010) (0.83 and 0.63 for part 1 and part 2, respectively) confirm the identification.

22. Page 12337, line 28: “Ambient temperature shows an anti-correlation with OOA”, does it suggest this OOA factor is more like the SV-OOA factor from other AMS studies?

No, the factors are LV-OOA like. The corresponding paragraph was changed to: “Ambient temperature shows a slight anti-correlation with OOA for part 1 (27 November 2007 – 08 January 2008: slope = -0.4, $R^2 = 0.3$, see Fig. SI-16 and SI-18), indicating the importance of thermal inversions (with low boundary layer temperatures) for the build-up of secondary particle mass”.

23. Page 12339, line 12339: “During that period until December 2008” not clear which “period” is referred to.

December 2008. The information was added for clarification.

24. Page 12343, line 15-19, I don't understand what the author is presenting here.

A third method for the estimation of the background concentrations was tested, based on Bukowiecki et al. (2002). Background concentrations were calculated using a moving 20-min lower 5%-percentile of each PM_{10} component. Further information was introduced for clarification.

25. Page 12344, line 2: 30% of PM_{10} or OM?

The percentages denote the fraction of local PM_{10} . The information was added for clarification.

26. Page 4 of supplemental information, line 2: it says “led to a split of the HOA factor”, I don't see how this “split” occurs, please explain.

There is no clear HOA factor distinguishable anymore, but 2 factors with a high signal at m/z 43.

This information was added to the text in the SI.

Technical suggestions:

27. Page 12324, line 6: capitalize the first letter of “matrix” and “factorization”.

We are used to not capitalize those, in agreement with style e.g. in Environ. Sci. Technol. So we prefer to leave this as is.

28. Page 12325, line 4: grammar problem of sentence starting with “effects”.

Section was changed to “[...] and impair human health (Pope and Dockery, 2006). These effects make them [...]”

29. Page 12325, line 5: “smogchamber” should be two words.

Word was changed accordingly.

30. *Page 12331, line 6: grammar mistake of “any a”.*

It should read “a priori”, latin for "from the former" or "from before". The sentence was not changed.

31. *Page 12337, line 1: add “by” after “done”.*

Sentence was changed accordingly.

32. *Figure SI-1: what do the squares and triangles represent?*

Squares and circles represent a time axis (see below map on the left), the red triangles the receptor site Zurich Kaserne. This information was added to the figure caption of SI-1.

33. *Figure SI-4: it would be better if the x-axis of the figure shows time rather than “Point number”.*

We agree with Referee #1 that it would be more convenient for the reader if the x-axis of figure SI-4 showed time. The software works such that it plots only temporally continuous time series, which would lead in this case to huge data gaps (between the measurement drives) and thus would make it impossible to distinguish data values for the measurement periods. The solution for this would be to cut the data into “measurement pieces” and to plot the pieces individually. But this would require much more space and would significantly complicate analysis, therefore we chose to plot the data versus point numbers.

34. *Page 3 of supplemental information, line 11: specify the correlation.*

Sentence was changed to “similar to”.

35. *Figure 1: the sampling time periods are too small to see.*

The labels were made bigger.

36. *Figure 9: I had a difficult time distinguishing the thin and thick lines in the plots.*

The distinction was made clearer.

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