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Interactive comment on “Spatial variation of chemical composition and sources of submicron aerosol in Zurich: factor analysis of mobile aerosol mass spectrometer data” by C. Mohr et al.

Anonymous Referee #1

Received and published: 1 June 2011

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

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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.

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? On page 12345, line 7, it says “the local contributions amount to 20 and 10% for NOX 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. 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. 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?) 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. 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. 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. 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

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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 PMF solutions need more justifications. Some discussions are not very clear. 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.
2. Page 12324, line 1: should specify the diameter is “vacuum” aerodynamic diameter.
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”.
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.
5. Throughout: SO₄, NO₃ etc. should be “SO₄2-” and “NO₃-”, or “sulfate” and “nitrate”, the same for other inorganic components.
6. Page 12329, line 9: how the PAH is measured should be introduced.
7. Page 12329, line 27 to page 12330 line 2: this should be removed since measurements from this instrument is not discussed.
8. Section 2.4.2 should be moved to Section 3.2 “PMF results” since it describes the preparation of PMF inputs.
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.
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?
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

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the mathematical aspect of the PMF solutions, but should justify why the “HOA” factor is HOA, etc. 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. 13. The figures of supplemental materials are not referenced in order. 14. Page 12332, line 20, the sentence starting with “Forcing one factor. PMF solution” is not understandable. 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. 16. Page 12333, line 1: what is the “model error”? 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? 18. Page 12334, line 19: the “breakup of the inversion”, how is this justified and does this “breakup” occur on other days? 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. 20. Page 12336, line 3: what is “factor-respective fractions”? 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? 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? 23. Page 12339, line 12339: “During that period until December 2008” not clear which “period” is referred to. 24. Page 12343, line 15-19, I don’t understand what the author is presenting here. 25. Page 12344, line 2: 30% of PM₁ or OM? 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.

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Technical suggestions:

27. Page 12324, line 6: capitalize the first letter of “matrix” and “factorization”. 28. Page 12325, line 4: grammar problem of sentence starting with “effects”. 29. Page 12325, line 5: “smogchamber” should be two words. 30. Page 12331, line 6: grammar mistake of “any a”. 31. Page 12337, line 1: add “by” after “done”. 32. Figure SI-1: what do the squares and triangles represent? 33. Figure SI-4: it would be better if the x-axis of the figure shows time rather than “Point number”. 34. Page 3 of supplemental information, line 11: specify the correlation. 35. Figure 1: the sampling time periods are too small to see. 36. Figure 9: I had a difficult time distinguishing the thin and thick lines in the plots.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 12323, 2011.

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