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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Impact of anthropogenic and biogenic sources on the seasonal variation of the molecular composition of urban organic aerosols: a field and laboratory study using ultra-high resolution mass spectrometry" *by* Kaspar R. Daellenbach et al.

**Anonymous Referee #2**

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General comments

The authors present an analysis of Orbitrap ultra-high resolution mass spectra from organic aerosol (OA) filter samples collected over the course of 2013 in Zurich, Switzerland. These spectra are also compared to specific source spectra from laboratory wood burning experiments, and from ambient locations that are clearly influenced by a specific source (wood burning, biogenic emissions). The results show that summer OA in



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Discussion paper

Zurich is dominated by compounds of biogenic origin, whereas winter OA is dominated by compounds from wood burning emissions, and confirm the importance of non-fossil organic carbon for OA mass loadings in Central Europe.

This is a well and clearly written paper. Whereas the main message may not be completely new (it has been shown before by the same group that winter OA in Zurich is dominated by wood burning, and summer OA by biogenic secondary OA - the authors also refer to these studies), it is still of importance to corroborate it, as it has implications for air quality policy making. The ultra-high resolution spectra provide a new layer of information on the molecular composition of OA at this location. In addition to confirming the importance of wood burning and biogenic emissions for this location, it would also be interesting to use the detailed molecular information for improved estimates of OA health effects and/or physicochemical properties influencing climate effects.

I recommend this paper to be published after the following comments have been taken care of:

Specific comments

P. 1, l. 26: I suggest to add also the O/C ratio and average carbon number.

P. 3, l. 20 – 30: In Zurich, Magadino, and San Vittore, PM10 was measured, in Hyytiälä, PM1. The authors should explain if, and how these differences in size cut influence the measured chemical composition. The sampling time for the filters in Switzerland was 24 hours; what was the sampling time in Hyytiälä?

P. 3, l. 29 – 30: Why would temperature be the only parameter that can vary between years? Do the authors assume e.g. emissions to be the same, and why?

P. 4, l. 4: What kind of filters were used in the smog chamber experiments?

P. 4, l. 15: The authors should motivate their choice for using negative mode only.

P. 4, l. 18: Would it be possible for the differences in vaporizer temperature to produce artefacts? Please elaborate.

P. 7, l. 21: I realize this paragraph summarizes previous, already published studies, and is therefore not up for discussion here. However, it is not fully clear to me how WOOA and BBOA (Figure 1) are connected to the results presented here. WOOA is interpreted as being formed from anthropogenic VOC emissions – however, I am assuming emissions are similar between summer and winter. With the biogenic emissions becoming more important in the summer months I can see how the relative contributions WOOA become much less important in summer (Figure 1). What about the absolute concentrations, however? And if OA is really dominated by Wood Burning in winter, as the Orbitrap spectra suggest, then a large fraction of WOOA must be from Wood Burning as well. How come it correlates with NH4+? On p. 15, l. 5 - 7 the authors state that " a good correlation was observed between the relative signal contribution of the compounds with H/C between 0.7 and 1.1 and the relative contribution of the sum of BBOA and WOOA to OA" – does this refer to a correlation of time series? How does a potential mass closure look like? What are the compounds in the "white parts" of Figure 11? Meteorology (inversion episodes) play a major role as well in Zurich, especially during winter. How is that taken into account in the present study?

P. 7, l. 28 / Figure 2: What are the 9 / 6 summer / winter samples? Please give more details.

P. 8, l. 31 – 34, p. 9, l. 1 -3: It would have been interesting to compare to spectra not from the boreal forest, but from the temperate broadleaf forest dominating Central Europe. Would it be possible for the authors to add such a comparison?

P. 10, l. 16 – 19, p. 11, l. 4 - 8: Such ratios are highly dependent on plant species. Given the differences in biome between Hyytiälä and Zurich, I highly question if such statements can be made without further proof.

P. 14, l. 6 – 11: It was shown earlier that wood burning OA in Zurich is mostly of

regional origin, especially during inversion episodes (Mohr et al., ACP, 2011), which is in agreement with its longer lifetime compared to the Alpine valleys shown here. Since meteorology, and especially inversion episodes, play such an important role for winter air quality in Zurich, periods when there was such an episode should be marked in e.g. Figure 1.

P. 28, Figure 1: What is the reason for the increase in NOx emissions in the winter months, and the very high fraction of SC-OA in November?

Technical corrections

P. 5, equation 1: Define N

P. 11, 10: Influence on what? – Sentence should be rephrased.

P. 28, Fig. 1: It would improve readability of the graph if the sample days were just labelled "Jan, Feb. . .", and the dates were added in a table.

P. 32, Figure 5: This figure is hard to read and could potentially be left out.

P. 33, Figure 7a: What are the grey lines?

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