# **Referee comment on**

# "Primary and secondary aerosols in Beijing in winter: sources, variations and processes"

by Sun et al., Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-255, 2016

### Anonymous referee #1

### 1. General comments

This manuscript reports results obtained during a field campaign performed at Beijing in Winter 2013/14. The authors deployed an Aerodyne high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) to measure the particle concentration, chemical composition and size distribution, sampled particles on filters for subsequent extraction and analysis by gas chromatography/mass spectrometry (GC/MS), and measured gaseous species and meteorological data. A source apportionment of organics was performed by positive matrix factorization. The effect of the relative humidity on the particle concentration and chemical composition was studied through several case studies.

This manuscript is very descriptive, but well written and interesting. Moreover, with the severe pollution events occurring regularly at Beijing, it is very important to perform this kind of study in order to better understand sources and processes of particles impacting this megacity. Thus, I recommend its publication after the authors address the following comments.

## 2. Specific comments

Page 4, line 9: I think that the reference Canagaratna et al. (2007) is much more appropriate here than Canagaratna et al. (2015).

Section 2 "Experimental methods": the authors give later in the manuscript some results from back trajectory analysis with the HYSPLIT model. They should describe this analysis in the "Experimental methods" section rather than in the caption of Figure 14. By the way, the back trajectory analysis reported here concerns only a short period (Jan 15<sup>th</sup>-17<sup>th</sup>). The absence of a complete analysis for the entire study is maybe the main weakness of this manuscript.

Section 2 "Experimental methods": it seems that all the dates and time are given in local time. The authors should mention that somewhere in this section.

Page 6, lines 26-27: a constant collection efficiency of 0.5 was used for this dataset. The authors need to justify this choice in the manuscript, in particular by giving some information on the particle acidity and on the presence or absence of a dryer in front of the AMS. Concerning the chemical composition, Figure 1f suggests that particles were never dominated by ammonium nitrate, so this point should be mentioned as well.

Section 3.1 "Mass concentrations and compositions": there is a long discussion on the  $SO_4/NO_3$  ratio, without any information under which form these two species are present. So here also, a few words on the particle acidity would be helpful to clarify this point.

Page 10, lines 20-21: the authors claim that cooking organic aerosols (COA) are mainly in the ultrafine range (< 100 nm). This is in contradiction with results obtained by Ge et al. (2012), who showed that their COA factor had a very broad size distribution peaking at 450 nm (in  $D_{va}$ ). However, according to other studies, the sizes of cooking-related particles vary widely, depending on cooking types, operations, and distance from the cooking sources. The authors may include this discussion in the manuscript.

Page 10, lines 26-28: the fact that two species have similar size distributions does not necessarily mean that they are internally mixed. This kind of information cannot be obtained with the AMS, which does not perform single particle analysis (unless the instrument is equipped with a light scattering module).

Page 14, line 11: concerning the Paris megacity, the authors can also mention the more recent study performed by Fröhlich et al. (2015), who also found a higher contribution of COA (15.0% of the total organics) than HOA (14.3%).

#### **3.** Technical corrections

Page 3, line 3: "is of a-great concern".

Page 3, line 5: "concentration of PM<sub>2.5</sub> in Beijing was-decreased from".

Page 4, line 15: please define the "SIA" abbreviation.

Page 7, line 4: "hydrogen-to-carbon (H/C), nitrogen-to-carbon\_(N/C), and".

Page 16, line 21: "levoglucosan <del>/</del>O/C of 0.062".

Page 17, line 10: "OOA was highly correlated to\_the oxygenated ions series".

Page 19, line 4: "are of a great concern".

Page 21, lines 4-5: "two episodes, i.e., E3 and E5 showed much higher SOA contributions (67 - 77%) than the other episodes".

Page 21, line 8: "enhanced the oxidation stageslevels of OA".

Page 40, Figure 12b: this figure is very crowded, the different size distributions are a bit hard to identify. In particular, the colors of E1, E2 and E4 are almost similar, as well as those of M3 and M5. The authors may try to use different markers, it will be easier to identify the different size distributions.

Page 41, caption of Figure 14: "(Stein et al., 2015).\_The background picture is\_the MODIS image on January 16". This sentence is incomplete, I'm not sure whether that's what the authors wanted to say.

#### 4. References

Canagaratna, M. R., Jayne, J. T., Jimenez, J. L., Allan, J. D., Alfarra, M. R., Zhang, Q., Onasch, T. B., Drewnick, F., Coe, H., Middlebrook, A., Delia, A., Williams, L. R., Trimborn, A. M., Northway, M. J., DeCarlo, P. F., Kolb, C. E., Davidovits, P., and Worsnop, D. R.: Chemical and microphysical characterization of ambient aerosols with the Aerodyne aerosol mass spectrometer, Mass Spectrom. Rev., 26, 185-222, 10.1002/mas.20115, 2007.

Canagaratna, M. R., Jimenez, J. L., Kroll, J. H., Chen, Q., Kessler, S. H., Massoli, P., Hildebrandt Ruiz, L., Fortner, E., Williams, L. R., Wilson, K. R., Surratt, J. D., Donahue, N. M., Jayne, J. T., and Worsnop, D. R.: Elemental ratio measurements of organic compounds using aerosol mass spectrometry: characterization, improved calibration, and implications, Atmos. Chem. Phys., 15, 253-272, 10.5194/acp-15-253-2015, 2015.

Fröhlich, R., Crenn, V., Setyan, A., Belis, C. A., Canonaco, F., Favez, O., Riffault, V., Slowik, J. G., Aas, W., Aijälä, M., Alastuey, A., Artiñano, B., Bonnaire, N., Bozzetti, C., Bressi, M., Carbone, C., Coz, E., Croteau, P. L., Cubison, M. J., Esser-Gietl, J. K., Green, D. C., Gros, V., Heikkinen, L., Herrmann, H., Jayne, J. T., Lunder, C. R., Minguillón, M. C., Močnik, G., O'Dowd, C. D., Ovadnevaite, J., Petralia, E., Poulain, L., Priestman, M., Ripoll, A., Sarda-Estève, R., Wiedensohler, A., Baltensperger, U., Sciare, J., and Prévôt, A. S. H.: ACTRIS ACSM intercomparison – Part 2: Intercomparison of ME-2 organic source apportionment results from 15 individual, co-located aerosol mass spectrometers, Atmos. Meas. Tech., 8, 2555-2576, 10.5194/amt-8-2555-2015, 2015.

Ge, X., Setyan, A., Sun, Y., and Zhang, Q.: Primary and secondary organic aerosols in Fresno, California during wintertime: Results from high resolution aerosol mass spectrometry, J. Geophys. Res., 117, D19301, 10.1029/2012jd018026, 2012.