

Response to Xia Ke's comments:

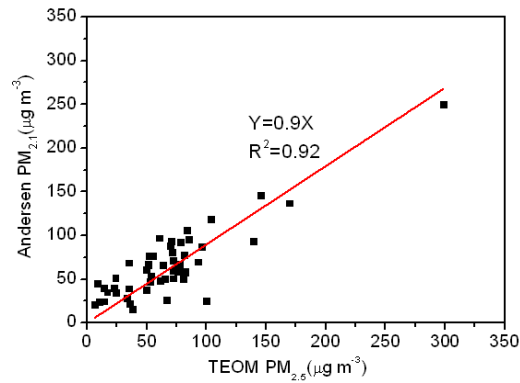
This study presents data of size-resolved aerosol chemical components in Beijing and analysis of their sources during the four seasons. While I see its scientific value, I also feel that there are some critical issues that need to be addressed.

1. The study used PM_{2.1} and PM_{2.1-9} data to represent fine and coarse parties, respectively, instead of the traditional PM_{2.5} and PM₁₀. How will this choice affect the final results? Uncertainty assessments can be easily done using known mass size distribution data.

Reply: Traditionally, fine and coarse mode particles were defined as particulate matter (PM) with aerodynamic diameter less than 2.5 μm (PM_{2.5}) and between 2.5 and 10 μm (PM_{2.5-10}), respectively. Note that coarse particle was defined as PM_{2.5-10} rather than PM₁₀.

In general, however, there was no uniform definition for fine and coarse particle during the field observation, which depends on the cut off size of the individual sampler. In previous studies, taking (Matsumoto et al., 2012) for example, PM samples with diameters smaller than 2.0 μm and of 2.0–10 μm were defined as fine and coarse particle, respectively. For PM samples collected by MOUDI, PM_{1.8} was used to represent fine particles (Wang et al., 2012; Sun et al., 2013). Similarly, when Andersen samples were collected, PM_{2.1} was usually defined as fine particles (Wang et al., 2011; Li et al., 2011).

The definition of fine and coarse particles did not significantly affect the final results in this study. For example, we have compared the mass concentrations of PM_{2.1} by weighting the filters before and after sampling with that of PM_{2.5} which measured by the commercial instrument of TEOM. The figure below shows that there was a good linear correlation between PM_{2.1} and PM_{2.5} ($R^2=0.92$, $P<0.05$), and the concentrations of PM_{2.1} were only 10% lower than that of PM_{2.5}.



- Why chose the weighing condition of RH as 10%? Cellulose filter or even quartz filter should be taken with static at such dry condition. Although the filters were eliminated static, the results of microbalance should not be stable during the multiple weighing processes. Thus, the uncertainty of aerosol mass should be addressed.

Reply: Thank you for your comment. We are sorry that we make a mistake here and the weighing condition of RH is $22 \pm 3\%$. During the whole weighing process each quartz filter was eliminated static (AD1683, A&D company, limited, Japan). In the lab the microbalance was stable and the balance time for each weighing process was less than 10s.

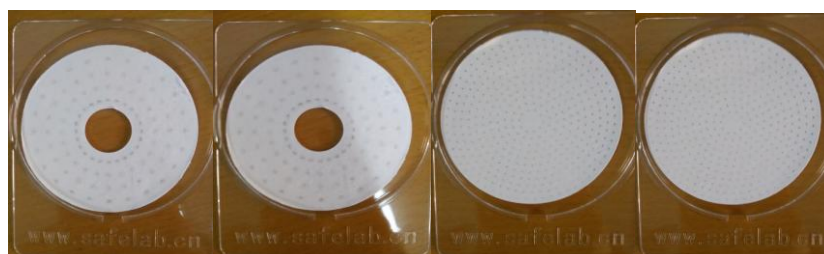
Similar equilibrium conditions for weighing have been widely used in previous studies. For example, the mass of particles on each quartz filter was weighted after 48 h equilibration in a glass chamber (25 ± 0.5 °C, $35 \pm 2\%$ RH) when the dust storm was studied in Xi'an based on Andersen samples (Wang et al., 2015). During the observation in Nanjing, the glass fiber filters was baked at 500°C for 2 h in order to eliminate organic species, conditioned in a desiccator for 24 h before weighing (Wang et al., 2003).

In addition, as aforementioned figure, the mass concentrations of $PM_{2.1}$ were comparable with that of $PM_{2.5}$, the latter of which is the appropriate alternative proposal of standard method recommended by US EPA. The results indicated that the weighing method is sound and reasonable for further mass closure analysis.

3. QA/QC procedures of sampling process are missed in this manuscript, which are important for a scientific paper presenting the first-hand data. The Anderson sampler should be blocked during heavy pollution conditions, and then the collected samples were not evenly distributed. This phenomenon should affect the chemical analysis, especial for OC and EC (choice of spots).

Reply: Thanks for the suggestions; we have added detailed QA/QC procedures of sampling process in the revised manuscript.

To prevent the sampler from blocking by particles during sampling, in this study the samplers were first cleaned using an ultrasonic bath for 30 min before each sampling. In addition, the sampling flow rates were calibrated before each sampling and also monitored by flow meter during whole sampling. These control measures assured that the Andersen sampler was not blocked during the sampling period. Even at heavily polluted days with PM_{2.5} values of 169.8 $\mu\text{g m}^{-3}$, the filter sampled by Andersen was evenly distributed and the spots were clearly obvious, as shown below.



>9 μm 5.8-9 μm 4.7-5.8 μm 3.3-4.7 μm



2.1-3.3 μm 1.1-2.1 μm 0.65-1.1 μm 0.43-0.65 μm <0.43 μm

4. Meteorological parameters seemed to be collected, but was not shown in the manuscript. Aerosol should bound when RH<40%, and thus, samples under these

conditions should be removed.

Reply: Thank you for your advice. In the revised manuscript, we added a figure in the supplementary material to describe meteorological parameters.

Additionally, we use visibility and RH together to determine the haze/no-haze days: sampling days with visibility < 10 km and RH < 90% were defined as haze days, while sampling days with visibility > 10 km and RH < 90% were defined as non-haze days. During the observation period, 12 sets of size-resolved PM samples were collected during non-haze days and 19 sets during haze days (marked in Fig. 2). Of the remaining 21 sets, 15 sets were collected during rain, snow or fog days and 6 sets were observed during dust days (visibility < 10 km, RH < 40%). These sets were excluded from the dataset when we discuss the differences between haze and non-haze days. The table below shows how 52 weeks samples were divided into different types, according to the visibility and RH.

	Visibility	RH	Sample quantity
Haze	<10 km	40%<RH<90%	19
Non-haze	>10 km	no rain, snow or fog	12
Dust	<10 km	RH<40%	6
Other events		rain, snow or fog	15

To check whether the aerosol bound or not when RH<40%, we have compared the mass concentrations of PM_{2.1} with that of PM_{2.5} measured by commercial instrument of TEOM at different RH conditions. There was a good linear correlation between PM_{2.1} and PM_{2.5} under both RH>40% (slope=0.93, R²=0.94, P<0.05) and RH<40% days (slope=0.87, R²=0.92, P<0.05). After a review on the literatures, we found that Andersen samplers can be used to collect dust storm samples when RH was much lower than 40% (Wang et al., 2015; Wang et al., 2014). In the revised paper, samples with RH lower than 40% and visibility less than 10km were defined as dust samples, and were removed from the haze days.

5. In general, results generated from PMF model could be questionable if less than 100 samples were used in the model.

Reply: In the revised manuscript, the PMF analysis was performed respectively for the fine mode (the input data included the mass concentrations and chemical species in particles with size bins of <0.43, 0.43-0.65, 0.65-1.1 and 1.1-2.1 μm) and coarse fractions (the input data included the mass concentrations and chemical species for particles in size fractions of 2.1-3.3, 3.3-4.7, 4.7-5.8, 5.8-9 and >9 μm) (Contini et al., 2014). The samples used in model were 208 and 260, respectively.

Why the percentage contributions for chemical species estimated from the six source profiles are not shown in the manuscript? (Figure 5).

Reply: We have redraw Figure 5 and the percentage contributions were shown in the figure in the revised version.

6. Figure 1 and Figure 3 are likely wrong due to the samplers with cut points of 0.43-9.0 μm .

Reply: Thanks for the comments. We have redraw Figure 1 and Figure 3 in the revised manuscript.

7. Discussions in section 4.4 seemed to be out of the scope of this manuscript.

Reply: The main idea of Sect. 4.4 was to investigate the effect of the chemical species, especially in the different size fractions, and meteorological factors on the visibility. This part may aid in the reconstruction of the relationship between the visibility and sources of particulate matter (Sect. 4.3.3). In the revised paper we have added more discussions on this topic. This may improve our understanding of the mechanism of haze formation and the results will help policy-makers designing emission control strategies to reduce the effect of haze.

References

Contini, D., Cesari, D., Genga, A., Siciliano, M., Ielpo, P., Guascito, M. R., and Conte, M.: Source apportionment of size-segregated atmospheric particles based on the major water-soluble components in Lecce (Italy), *Sci Total Environ*, 472, 248-261, 2014.

Li, J., Wang, G., Zhou, B., Cheng, C., Cao, J., Shen, Z., and An, Z.: Chemical composition and size distribution of wintertime aerosols in the atmosphere of Mt. Hua in central China, *Atmos Environ*, 45, 1251-1258, 2011.

Matsumoto, K., Masuda, N., and Nakano, T.: Partitioning of atmospheric fixed nitrogen species among gas phase, fine particles, and coarse particles, *J Aerosol Sci*, 54, 49-58, 2012.

Sun, Z., Mu, Y., Liu, Y., and Shao, L.: A comparison study on airborne particles during haze days and non-haze days in Beijing, *Sci Total Environ*, 456-457, 1-8, 2013.

Wang, G., Wang, H., Yu, Y., Gao, S., Feng, J., Gao, S., and Wang, L.: Chemical characterization of water-soluble components of PM₁₀ and PM_{2.5} atmospheric aerosols in five locations of Nanjing, China, *Atmos Environ*, 37, 2893-2902, 2003.

Wang, G., Chen, C., Li, J., Zhou, B., Xie, M., Hu, S., Kawamura, K., and Chen, Y.: Molecular composition and size distribution of sugars, sugar-alcohols and carboxylic acids in airborne particles during a severe urban haze event caused by wheat straw burning, *Atmos Environ*, 45, 2473-2479, 2011.

Wang, G., Cheng, C., Meng, J., Huang, Y., Li, J., and Ren, Y.: Field observation on secondary organic aerosols during Asian dust storm periods: Formation mechanism of oxalic acid and related compounds on dust surface, *Atmos Environ*, 113, 169-176, 2015.

Wang, G. H., Cheng, C. L., Huang, Y., Tao, J., Ren, Y. Q., Wu, F., Meng, J. J., Li, J. J., Cheng, Y. T., Cao, J. J., Liu, S. X., Zhang, T., Zhang, R., and Chen, Y. B.: Evolution of aerosol chemistry in Xi'an, inland China, during the dust storm

period of 2013 – Part 1: Sources, chemical forms and formation mechanisms of nitrate and sulfate, *Atmos Chem Phys*, 14, 11571-11585, 2014.

Wang, X., Wang, W., Yang, L., Gao, X., Nie, W., Yu, Y., Xu, P., Zhou, Y., and Wang, Z.: The secondary formation of inorganic aerosols in the droplet mode through heterogeneous aqueous reactions under haze conditions, *Atmos Environ*, 63, 68-76, 2012.