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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characterization and source apportionment of aerosol light scattering in a typical polluted city in Yangtze River Delta, China" *by* Dong Chen et al.

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We thank very much for the valuable comments and suggestions from the reviewer, which help us improve our manuscript significantly. The comments were carefully considered and revisions have been made in response to suggestions. Following is our point-by-point responses to the comments and corresponding revisions.

0. The current manuscript presents a comprehensive study of the influential factors and source apportionment of aerosol light scattering at three sites in Nanjing, representative for suburban (NJU), urban (PAES) and industrial areas (NUIST) respectively. The data obtained in this work show interesting details on the linkage between chem-

istry composition and light scattering of aerosols, and help better understanding the effects of various sources on visibility degradation at the city scale. Overall I think the work provides reasonable analysis and the paper is clearly written. Before it can be published in Atmospheric Chemistry and Physics, however, I have some concerns that should be further addressed, and minor revisions are accordingly suggested as below.

Response and revisions:

We appreciate the reviewer's positive remarks on the importance of the work.

1. In line 130, QA/QC procedures of aerosol sampling process are missed in this manuscript, which are important for a scientific paper presenting the first-hand data. For example, the MOUDI sampler could be blocked during heavy pollution conditions, and the collected samples might not be evenly distributed. This phenomenon would affect the chemical analysis, particularly for OC and EC (choice of spots). How did the authors treat such kind of problems or estimate the uncertainty from sampling?

Response and revisions:

We thank the reviewer's comment. To prevent the blocking by particles during sampling, the MOUDI samplers were first cleaned using an ultrasonic bath for 30 min before each sampling. In addition, the sampling flow rate was calibrated before each sampling and was also monitored with the flow meter during the whole sampling period. Those quality control measures assured that the MOUDI samplers were not blocked during the sampling period. Even for heavily polluted days with the PM1.8 concentration measured at 128 μgâĂćm-3, the particles sampled by MOUDI were evenly distributed. We have added the explanation in lines 138-144 in the revised manuscript, and added a new Figure S2 (Figure R4 here) in the revised supplement, illustrating the size-resolved particle filter samples collected on 25 Dec 2015 at NJU.

2. In Line 136, were field blanks obtained during the sampling campaigns? And, why were the sampling periods different at the three sites? Similarly, in Line 128, why was

the sampling size at NJU larger than another two sites? The sampling strategy should be described more.

Response and revisions:

We thank the reviewer's comment. Yes we applied field blanks to correct the possible bias in the analysis of aerosol chemical species. Totally 19 sets of size-segregated blank filters (10, 4 and 5 for NJU, PAES and NUIST, respectively) and 35 daily blank PM2.5 filters (25, 6 and 9 for NJU, PAES and NUIST, respectively) were obtained at the three sites. All the blank filters were put in the samplers without inlet air flow for 24 h when the field campaigns finished. We have added the information in lines 152-157 in the revised manuscript.

Attributed to weather condition and aerosol sampler maintenance, the sampling periods for the three sites were different. Simultaneous samplings were conducted at the three sites from one week to ten days in each season from summer 2016 to winter 2016-2017. For the remaining time, two MOUDI samplers were applied to collect Teflon and quartz filter samples simultaneously at one of the three sites. As the Cavity Attenuated Phase Shift Albedo monitor (CAPS) was only installed at NJU and large amounts of data on aerosol optical and chemical information were needed to examine the influence of relative humidity on aerosol light scattering (Section 3.3), the sampling size at NJU was larger than another two sites. We have added the explanation in lines 144-152 in the revised manuscript.

3. In Line 145, Sunset analyzer was able to measure thermal EC and OC, and optical EC and OC. The author should clarify it carefully in the paper. Why choose them for the analysis?

Response and revisions:

We thank the reviewer's comment. The Sunset analyzer provides both thermal and optical concentrations for carbonaceous aerosols, and thermal EC and OC were used in

this study. The instrument estimates the optical EC by measuring the light attenuation (ATN). As ATN was determined not only by EC but also by brown carbon (BrC), the optical method may overestimate the EC and thus underestimate the OC (Cui et al., 2016; Massabò et al., 2016). Therefore, we applied the measured thermal EC and OC in this study. We added the explanation in lines 169-173 in the revised manuscript.

4. In Line 183, what software did authors use to run the multiple linear regression? If this model has been developed or used in other studies, the references should be provided.

Response and revisions:

SPSS 16.0 was used to conduct the multiple linear regressions. This information was added in line 216 in the revised manuscript and relative references were provided (Cheng et al., 2011; Tian et al., 2016).

5. In Line 190, considering the light absorption of methanol soluble organic carbon (MSOC), the optimization of the US IMPROVE algorithm is quite interesting. How did the authors estimate the MSOC concentrations in fine and large size modes?

Response and revisions:

The estimations of fine MSOC and large MSOC concentrations were the same as the calculations of ammonium nitrate (NH4NO3), ammonium sulfate ((NH4)2SO4) and organic carbon (OM), following the previous studies (Pitchford et al., 2007; Cheng et al., 2015). The concentration of large MSOC was estimated by dividing the total concentration of the component by 20 μg/m3 (e.g., if the total MSOC concentration was 4 μg/m3, the large mode concentration was calculated to be one-fifth of 4 μg/m3 or 0.8 μg/m3, leaving 3.2 μg/m3 in the small mode). If the total MSOC concentration exceeds 20 μg/m3, all of it was assumed to be in the large mode.

6. In Line 190, why did the authors only include those eight variables in this equation? How about other species like coarse particles, sea salt, and soil dust?

Response and revisions:

We thank the reviewer's comment. In this study, the optimization of the US IMPROVE formula was for PM2.5, thus the optimization process did not include coarse particles. As indicated in Figure R2 (the response to Question 3 of reviewer 1), the scattering coefficients by sea salt and soil dust accounted for less than 10% of the total PM2.5 scattering, suggesting that the two species should have limited contribution to the total scattering coefficient. In order to be concise in the IMPROVE formula optimization and to ensure the stability of the multiple linear regression, therefore, only ammonium sulfate, ammonium nitrate and organic matter were used as independent variables in the regression. We have added the explanation in lines 459-465 in the revised manuscript.

7. In Line 214, the authors need to explain the special reason why they applied PMF model in their analysis.

Response and revisions:

We thank the reviewer's comment. The source apportionment technologies include emissions inventory, chemistry transport model and the receptor model, and the receptor model based on aerosol chemistry analysis has been widely used because it is not limited by the uncertainty of emission inventory or meteorology simulation. Principally the receptor models contain two categories, i.e., the models in which source profiles are needed, such as the chemical mass balance (CMB) method, and those in which source profiles are not needed, such as the positive matrix factorization (PMF) method (Yin et al., 2015). Due to lack of sufficient local measurements, it is generally difficult to build comprehensive and accurate source profiles of various aerosol components for individual cities in China. In this work, therefore, we applied the PMF model to exclude the uncertainty of source profiles which were commonly developed based on literatures or measurements across the country. We have added the explanation in lines 247-252 in the revised manuscript.

8. In Line 292, the author stated that "the sum of NO3-, SO42- and OC for the heavily polluted period was 10.7 and 2.9 times greater than those for the lightly polluted and clean periods". Something seems wrong here. In which period was the concentration higher, lightly polluted or clean period? From Table 1, moreover, it seems that SNA was elevated more than OC in the heavily polluted period compared to the clean days. Any reason for this difference?

Response and revisions:

We thank the reviewer's comment. We are sorry for the mistakes and corrected the text as "the sum of NO3-, SO42- and OC for the heavily polluted period was 2.9 and 10.7 times greater than those for the lightly polluted and clean periods" in line 328 in the revised manuscript.

It is correct that SNA concentration was elevated more than OC in the heavily polluted period compared to the clean days in this work. During heavily polluted episodes, enhancement of sulfate and nitrate levels could be more significant than organic matter because the high relative humidity and precursor emissions (i.e., SO2 and NOx) promoted the generation of SNA (Tian et al., 2014). During clean periods (commonly in summer), ammonium nitrate (NH4NO3) would dissociate to NH3 and HNO3 at high temperatures, while secondary organic carbon (SOC) concentration may be increased due to the high levels of O3 and solar radiation. Those factors caused the result that SNA was elevated more than OC in the heavily polluted period compared to the clean days. Similar result was found in Beijing, where the mass fraction of SNA in fine particles increased from 19% on non-haze days to 31% on haze days, while that of OM decreased from 38% on non-haze days to 31% on haze days (Tian et al., 2016). We have added the above explanation in lines 334-342 in the revised manuscript.

9. The authors did not clearly explain the data of which site were used in Section 3.4. If it is based on the data of the three sites, the assumption in Section 3.4 that chemical particles were externally mixed will cause large uncertainty in the calculation of scatter-

ing coefficient at NUIST because the internal mixture was an important particle mixing state at NUIST (Figure 3c).

Response and revisions:

We thank the reviewer's comment. Assuming different aerosol species were externally mixed, the influences of size distribution and pollution levels on aerosol light scattering was analyzed based on the aerosol composition information at the three sites. Although internal mixing was identified as an important particle mixing state at NUIST, the Mie theory cannot simulate the scattering coefficients of individual aerosol chemical components based on the assumption of internal or core-shell mixing but external mixing (Cheng et al., 2015; Ding et al 2015). According to Figure 3c in the revised manuscript, the scattering coefficient estimated with the external mixing assumption was 1.07 ± 0.05 and 1.08 ± 0.06 times of those from the internal mixing state simulation and instrument measurement, respectively. Therefore, the overestimation was smaller than 10% by the aggregated scattering coefficient from those of individual chemical species with an assumption of external mixing at NUIST. We have added the explanation in lines 540-545 in the revised manuscript.

10. In Line 534, in general, results generated from PMF model could be questionable if less than 100 samples were used in the model. How did the authors consider this problem?

Response and revisions:

In this study, the PMF analysis was performed respectively at the three sites for the accumulation mode particles with the size bins from 0.18 to 1.8 μm. The samples used in PMF model were 300, 100 and 124 at NJU, PAES and NUIST, respectively. We thus believe that the sample size was sufficient for the PMF analysis.

References

Cheng, S. H., Yang, L. X., Zhou, X. H., Xue, L. K., Gao, X. M., Zhou, Y., and Wang, W.

X.: Size-fractionated water-soluble ions, situ pH and water content in aerosol on hazy days and the influences on visibility impairment in Jinan, China, Atmos. Environ., 45, 4631–4640, doi:10.1016/j.atmosenv.2011.05.057, 2011.

Cheng, Z., Jiang, J., Chen, C., Gao, J., Wang, S., Watson, J.G., Wang, H., Deng, J., Wang, B., Zhou, M.: Estimation of aerosol mass scattering efficiencies under high mass loading: case study for the megacity of Shanghai, China, Environ. Sci. Technol., 49, 831-838, doi:10.1021/es504567q, 2015.

Cui, X., Wang, X., Yang, L., Chen, B., Chen, J., Andersson, A., Gustafsson, Ö.: Radiative absorption enhancement from coatings on black carbon aerosols, Sci. Total Environ., 551-552, 51-56, doi:10.1016/j.scitotenv.2016.02.026, 2016.

Ding, J., Han, S., Zhang, Y., Feng, Y., Wu, J., Shi, G., Wang, J.: Chemical characteristics of particles and light extinction effects in winter in Tianjin, Res. Environ. Sci., 28, 1353-1361, doi:10.13198/j.issn.1001-6929.2015.09.03, 2015.

Massabò, D., Caponi, L., Bove, M.C., Prati, P.: Brown carbon and thermal-optical analysis: A correction based on optical multi-wavelength apportionment of atmospheric aerosols, Atmos. Environ., 125, 119-125, doi:10.1016/j.atmosenv.2015.11.011, 2016.

Pitchford, M., Malm, W., Schichtel, B., Kumar, N., Lowenthal, D., Hand, J.: Revised algorithm for estimating light extinction from IMPROVE particle speciation data, J. Air Waste Manag. Assoc., 57, 1326-1336, doi:10.3155/1047-3289.57.11.1326, 2007.

Tian, S. L., Pan, Y. P., Liu, Z., Wen, T., and Wang, Y. S.: Size-resolved aerosol chemical analysis of extreme haze pollution events during early 2013 in urban Beijing, China, J. Hazard. Mater., 279, 452–460, doi:10.1016/j.jhazmat.2014.07.023, 2014.

Tian, S. L., Pan, Y. P., Wang, Y. S.: Size-resolved source apportionment of particulate matter in urban Beijing during haze and non-haze episodes, Atmos. Chem. Phys., 16, 1-19, doi:10.5194/acp-16-1-2016, 2016.

Yin, J., Cumberland, S. A., Harrison, R. M., Allan, J., Young, D. E., Williams, P. I., and

Coe, H.: Receptor modelling of fine particles in southern England using CMB including comparison with AMS-PMF factors, Atmos Chem Phys, 15, 2139-2158, 2015.

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