Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-176-AC1, 2020

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.

Dong Chen et al.

yuzhao@nju.edu.cn

Received and published: 8 July 2020

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 paper by Chen et al. systematically investigates the characteristics and sources of aerosol light scattering through measurements at three different functional sites in a typical polluted city in the Yangtze River Delta, China. Aerosol scattering is important for both visibility degradation and air pollution, and is also complex due to aerosol

chemical composition and hygroscopic growth. In this study, the US IMPROVE formula for aerosol scattering calculation was optimized using online and offline measurements at different functional sites in Nanjing with complicated sources of air pollutants. The influence of aerosol size distributions and pollution levels on the aerosol scattering was quantitatively evaluated based on a comprehensive analysis of the size-specific chemical compositions of particles at various sites. In general, this manuscript is well organized and easy to follow. I would recommend its acceptance after some necessary corrections suggested as follows:

Response and revisions:

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

1. Line 87: "NH4NO3 and (NH4)2SO4" need to be defined at their first mention in the manuscript. The manuscript has similar problems with other chemical species as well. Please go through the manuscript and change all of them.

Response and revisions:

We thank the reviewer's reminder. As suggested, the two species were defined at their first appearance (lines 88 in the revised manuscript). We have also checked through the manuscript and revised all other items that need to be defined.

2. Line 149: What is the mass fraction of the methanol soluble organic carbon in the total organic carbon mass? Did you try the water extraction?

Response and revisions:

We thank the reviewer's comment. We explored the relationship between the total organic carbon (OC) and methanol soluble organic carbon (MSOC) concentrations in this study. The average MSOC was 8.23 $\pm$ 4.84 μg/m3 and accounted for 88% of the total OC mass in all samples. This result was similar to the fraction of 85% estimated by Cheng et al. (2016). Considering that a large fraction of brown carbon (BrC) absorption comes from OC insoluble in water, water extraction (WSOC) method was thus not

applied in the current study. We have added the discussion in lines 174-181 in the revised manuscript, and provided a new Figure S4 in the revised supplement, illustrating the relationship between the total OC and MSOC concentrations

3. Line 186: In the process of formula optimization, why did the authors subtract the scattering coefficients by sea salt, soil dust and coarse particles from the measured scattering coefficient? Does it mean that the light scattering of those species has little impact on the optimization of IMPROVE formula?

Response and revisions:

We thank the reviewer's comment. We calculate the ratios of the collective scattering coefficients of the sea salt and soil dust to the total PM2.5 scattering at the three sites. The ratios were 0.083, 0.093 and 0.081 at NJU, PAES, and NUIST, respectively, i.e., the scattering coefficients by sea salt and soil dust accounted for less than 10% of the total PM2.5 light scattering. Therefore the impact of the two species on the optimization of IMPROVE algorithm should be limited. In order to be concise in the algorithm optimization and to ensure the stability of the multiple linear regression, the independent variables contained (NH4)2SO4, NH4NO3 and OM in the optimized formula, and the light scattering of sea salt and soil dust was subtracted from the measured scattering coefficient of PM2.5. We have clarified the methodology in lines 217-220 in the revised manuscript, and have added the discussion in lines 459-465 in the revised manuscript. A new Figure S10 has also been provided in the revised supplement, illustrating the ratios of the collective scattering coefficients of the sea salt and soil dust to the total PM2.5 scattering at the three sites.

4. Line 201: Mie theory is very sensitive to the size distribution of aerosol chemical species. However, the size distribution data obtained from a high-flow MOUDI impactor can usually be influenced by the particle bounce. This is particularly concerned in case where filters, instead of metal foils with grease coating, are used as the substrate. I suggest the authors make an uncertainty evaluation upon the size distribution

measurement in this study.

Response and revisions:

We thank the reviewer's comment. Although application of metal foils with grease coating could avoid the particle bounce, it might change the result of chemical species measurement. More, the metal foils substrate cannot meet the requirement of carbonaceous aerosol analysis, due to its special heating up program. In this study, therefore, we selected teflon filter for ion and element analysis, and quart fiber filter for carbonaceous aerosol analysis. Teflon filter membrane was generally applied for size-resolved particles sampling by MOUDI with excellent results (Contini et al., 2014; Gao et al., 2016; Guo et al., 2010). Taking NJU as an example, excellent agreement was found between the mass concentrations of PM1.8 collected with quartz fiber in MOUDI impactor and PM2.5 collected with TH-150 samplers. Therefore, the effect of particle bounce was expected to limited in this study. We have added the information in lines 157-161 in the revised manuscript, and provided a new Figure S3 (Figure R3 here) in the revised supplement, illustrating the correlation between the mass concentrations of PM1.8 collected with MOUDI impactor and PM2.5 collected with TH-150C sampler at NJU.

5. In Section 3.2, the US IMPROVE algorithm was optimized only within one city in the Yangtze River Delta with good performance. How did the authors consider the application of the optimized formula in typical regions such as cities in Beijing-Tianjin-Hebei or Pearl River Delta? Some discussions are recommended here.

Response and revisions:

We thank the reviewer's comment. In this study, the optimized IMPROVE formula was obtained based on the measured ambient concentrations of aerosol chemical species at three different functional sites in Nanjing, a typical polluted city in the Yangtze River Delta. As the chemical composition of aerosol (particularly SNA) was the key factor affecting its light scattering, the optimized IMPROVE formula could be applied in nearby

cities with similar composition of aerosols in eastern China including Shanghai and Jinan, as we stated in lines 465-474 in the revised manuscript. Moreover, for other regions with rapidly developing economy and fast industrialization in China including Beijing-Tianjin-Hebei or Pearl River Delta regions, the current work provides methodology and data support for the studies of aerosol light scattering in cities with relatively serious particle pollution. Given the fast changes in emission control and aerosol pollution in those regions, more campaigns on aerosol optical and chemical properties are recommended to further evaluate and improve the applicability of the optimized IMPROVE algorithm. We have added the explanation in lines 474-481 in the revised manuscript.

6. Line 352: The study did not mention if the scattering coefficients used for the US IMPROVE estimation at the three sites were measured by CAPS or nephelometer? According to Section 2.3, the scattering coefficients at PAES and NUIST were measured by two integrating nephelometers. Need some clarification on this issue.

Response and revisions:

We thank the reviewer's reminder and sorry for the error. The scattering coefficients used for the evaluation and optimization of the IMPROVE algorithm at the three sites were all measured with nephelometers. The relevant texts have been revised in line 399 in the revised manuscript.

7. Line 447: Due to the varied chemical properties of particles in different regions, the growth factors of particles (GF) can be different, and it would bring some uncertainty to the calculation of scattering coefficient in Section 3.3. It is recommended to measure and apply the local GF values in this work.

Response and revisions:

We thank and agree the reviewer's comment. Due to the lack of suitable instrument like hygroscopicity tandem differential mobility analyzer (H-TDMA), we did not measure

the local GF values directly, and it is a limitation of this study. Instead, we collected the GF data from the existing local studies in Nanjing (Table S1 in the supplement), and applied them in estimation of ambient scattering coefficient by Mie theory. To check the uncertainty of this application, the estimates were compared with those calculated with the scattering hygroscopic growth factor (f(RH)), as shown in Figure S12 in the revised supplement (Figure S8 in the original submission). A good agreement was found between the two methods (R2=0.95), indicating the limited uncertainty from the GF values applied in this study.

8. In Section 3.4, there was no clear description whether the scattering coefficients were estimated based on the assumption of dry or ambient conditions.

Response and revisions:

We thank the reviewer's reminder. The estimated scattering coefficients in Section 3.4 were based on the assumption of ambient condition. The relevant text has been revised in line 543 in the revised manuscript.

9. In Section 3.5, the assumption that the secondary components were proportional to the emissions of their precursors is subject to great uncertainty, as noted by the authors. Please be more specific on how to get better results with improved measurement or modeling methods.

Response and revisions:

We thank the reviewer's comment. As we stated in the manuscript, there was substantial uncertainty in the methodology in which source apportionment of secondary aerosols depends on the magnitudes of precursor emissions. It is a limitation of the present study. To further improve the source apportionment results, some specific tracers of secondary aerosols like semi volatile and low volatile oxygen-containing organic aerosols can be firstly observed with advanced technology such as aerosol mass spectrometry (AMS), and the observation data can then be combined with receptor models

to quantify the source contribution of secondary aerosols. Besides, air quality model that integrates particle source apportionment technology (PSAT) is recommended to be applied to evaluate and confirm the performance of the source apportionment of secondary aerosol with the receptor model. We have added the explanation in lines 678-685 in the revised manuscript.

10. Some minor comments: Line 31: Define “IMPROVE” on first usage.

Response and revisions:

We thank the reviewer’s reminder and the full name has been given in the revised manuscript.

Line 32: "OC" should not be abbreviated when it is mentioned for the first time.

Response and revisions:

We thank the reviewer’s reminder and the full name has been given in the revised manuscript.

Line 160: What is the wavelength of the integrating nephelometer at the three sites used?

Response and revisions:

We thank the reviewer’s reminder. The two nephelometers (Ecotech Pty Ltd, Australia, Model Aurora1000G) at NJU and PAES were operated at the wavelength of 520 nm. The integrating nephelometer (Model 3563, TSI, USA) used at NUIST can measure the light scattering at three visible wavelengths (450, 550 and 700 nm), and the scattering coefficient at the wavelength of 550 nm was adopted in this work. We have added the explanation in lines 190-193 in the revised manuscript.

Line 246: The operational symbol was missing in Eq. (3).

Response and revisions:

We thank the reviewer’s reminder and it is corrected in the revised manuscript.

Line 522: “Mien theory” should be “Mie theory”.

Response and revisions:

We are sorry for this mistake and thanks for the reminder. We have corrected it in the revised manuscript.

Line 562: “PME” should be “PMF”.

Response and revisions:

We are sorry for this mistake and thanks for the reminder. We have corrected it in the revised manuscript.

Line 970: SIA in the legend did not exist in Figure 8.

Response and revisions:

We thank the reviewer’s reminder and the SIA legend in Figure 8 has been removed.

Reference list: The format of references should be in accordance with the journal requirement.

Response and revisions:

We thank the reviewer’s reminder. We have checked the format of references and made it consistent with the journal requirement.

References

Cheng, Y., He, K.B., Du, Z.Y., Engling, G., Liu, J.M., Ma, Y.L., Zheng, M., Weber, R.J.: The characteristics of brown carbon aerosol during winter in Beijing, Atmos. Environ., 127, 355-364, doi:10.1016/j.atmosenv.2015.12.035, 2016.

Contini, D., Cesari, D., Genga, A., Sicilianob, M., Ielpo, P., Guascito, M. R., 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, 10.1016/j.scitotenv.2013.10.127, 2014.

Gao, Y., Lee, S. C., Huang, Y., Chow, J. C., Watson, J. G.: Chemical characterization and source apportionment of size-resolved particles in Hong Kong sub-urban area, Atmos. Res., 170, 112-122, 10.1016/j.atmosres.2015.11.015, 2016.

Guo, S., Hu, M., Wang, Z. B., Slanina, J., and Zhao, Y. L.: Size resolved aerosol water-soluble ionic compositions in the summer of Beijing: implication of regional secondary formation, Atmos. Chem. Phys., 10, 947–959, doi:10.5194/acp-10-947-2010, 2010.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-176, 2020.