Response to Reviewer #1

We greatly appreciate the reviewer for providing the very detailed comments, which have helped us improve the paper quality significantly. We have addressed all of the comments carefully as detailed below. The original comments are in black and our replies are in blue.

Line 7: I'm concerned about reporting the condensation mode MMAD of 0.21 um. This just reflects the midpoint of the diameter bin. If this is the case, why not just report the midpoint of diameter bins for all the modes? Reporting it like data is meaningless.

We agree with this comment and thus have deleted the MMAD for the condensation mode.

Line 19: How is "fine" defined here?

"Fine particles" are defined here as those with aerodynamic diameter smaller than 2.1 μm because the cutoff size of the instrument is at 2.1 μm.

Line 42: Define "IMPROVE" on first usage.

We have revised the text as follows: "the original and revised empirical formulas from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network".

Line 45: MSE are important parameters not just for the IMPROVE equation, but for any application relating mass to optical properties.

We have revised the text as follows: "MSEs of the chemical species are important parameters not only for building the relationships between chemical species and b_{sp} (Hand and Malm, 2007), but also for relating particle mass to its optical properties (Lin et al., 2015; Titos et al., 2012)".

Line 49: Include "based on an assumed size distribution" after "formula…"

Text added as suggested.

Line 60: Yes, the second IMPROVE algorithm was developed for rural (very clean) areas, so it isn't a surprise that it doesn't perform well in urban areas.

We agree with this comment. However, the majority of the studies in China still used the revised formula in urban environment likely because the original IMPROVE formula evidently underestimated b_{sp}. It is thus needed to further assess the uncertainties in these formulas when applying to urban environments.

Line 69: I think the reference here should be “Malm and Hand, 2007”. Also, the efficiencies used in the second IMPROVE algorithm are based on an assumed size distribution and composition.

Reference replaced as suggested.

Line 73: Consider removing “According to Mie theory” because Mie theory doesn’t specifically speak to the factors hindering the IMPROVE formulas. Line 75: Also, what about assumed hygroscopic growth curves in the IMPROVE algorithm?

We have deleted the text “According to Mie theory”. In this study, we only discussed closure of b_{sp} under dry condition, not wet condition.

Line 81: I think the authors mean “inline” when they say “online” data?

We have replaced “online” with “inline” in all the places.

Line 88: Especially in urban areas.

We have revised the text as follows: “Knowledge gained from the present study will improve the assessments of air-quality and climate impact caused by atmospheric particles, especially in urban areas.”

Line 116: It would also help to include the other measurements in Table 1, such as the size distributions and nephelometer measurements.

We have added the relevant instruments information in Table 1.

Line 123: Do the authors mean “blank” instead of “background”?

Revised as suggested.

Line 136: What is the expected size cut of the nephelometer? Are there expected size-resolved losses from the tubing from the inlets that affect the size distribution and nephelometer measurements?

We have added the following text in the revised paper to address this comment: “According to the method described in Kulkarni et al. (2011), particle losses in different sizes from the tube are plotted in Fig. S1. Generally, particle losses in the condensation (0.1-0.4 μm), droplet (0.4-2.1 μm) and coarse modes (2.1-10 μm) were less than 1.3%, 0.3% and 0.1%, respectively, suggesting that the particle losses from the tube were minimal. Ambient relative humidity (RH) and temperature were measured by an automatic meteorological station (Vaisala Company, Helsinki, Finland, model MAWS201) at the SCIES site, and the seasonal average of these two meteorological parameters were 53-75 % and 15-29 °C, respectively.”

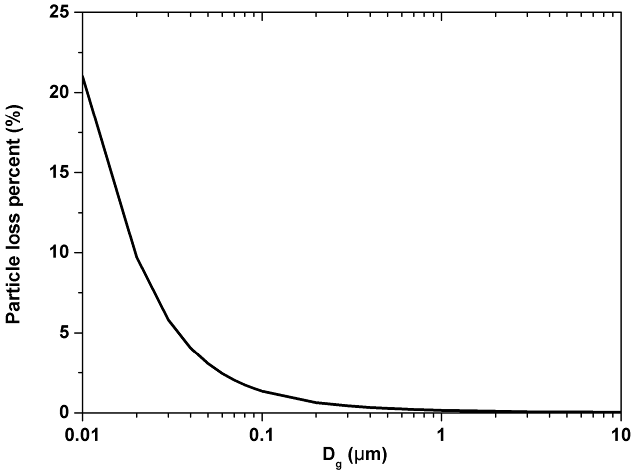


Fig. S1: The estimated particle losses in different size from the tube.

Line 140: Define “RH” at first usage (unless I missed it earlier).

Defined as suggested.

Line 141: Were RH and temperature monitored? What were typical values?

We have added the following text in the revised paper: “Ambient relative humidity (RH) and temperature were measured by an automatic meteorological station (Vaisala Company, Helsinki, Finland, model MAWS201) at the SCIES site, and the seasonal average of these two meteorological parameters were 53-75 % and 15-29 °C, respectively.”

Line 179: How were field blanks obtained?

We have added this description in the revised paper: “Moreover, 8 sets of blank samples were also collected for each of the size-segregated particle, $PM_{2.5}$ and PM_{10} samples during the whole sampling period. Two sets of blank filters in each category were put in the samplers without flow for 24 h when seasonal field campaigns finished. The aerosol-loaded filter samples were stored in a freezer at -18 °C before analysis to prevent volatilization of particles.”

Line 179: Why was OP so low from the 81mm filters?

Firstly, particle sample showed dot pattern (100-400 dots in every stage) in the size-segregated filters (81 mm filters). Secondly, carbon analyzer only analyzes one punch (0.526 cm^2), which contained 4-5 dots. Thirdly, total carbon loading in fine particle was not high (about 5-30 μg m^{-3}). If its concentration was distributed into 8 stages, then OC and EC concentrations in each stage would be very low. In addition, OP concentration was much lower than OC and EC. Thus, the uncertainties in OC and EC concentrations would be larger using OP to separate OC and EC in each stage.

Line 201-205: I am not sure of the rationale behind defining the condensation mode as the midpoint diameter of the smallest bin? If the MMAD of the mode is just assigned the midpoint diameter of the bin, what point is there in measuring any size distributions? The MMAD would just be the midpoint diameter of each bin which is meaningless. I don't think you can report the MMAD of the condensation mode if this is how you derive it.

As mentioned above, we have deleted the statement of MMAD for the condensation mode throughout the whole manuscript.

Line 209: I assume this discussion is with respect to the technique by Dong et al. (2004)? It might be helpful to provide more detail here regarding this method, since many of the results depend on it. For example, how were collection efficiencies incorporated into this inversion?

The reviewer is right. Here we indeed refer to the technique of Dong et al. (2004). We have added the key formulas in in section 2.4.

We have also revised the text as follows: "Continuous size-distribution profiles of major chemical species are needed in order to accurately calculate b_{sp} using Mie theory. To improve the resolution of b_{sp}, 401 bins were used for chemical species ranging from 10 nm to 100 μm, with a constant ratio between the adjacent size bins, defined as $\log_{10}(D_{a2}/D_{a1})=0.01$. Further increasing the number of size bins does not have any significant impact on the results, e.g., the changes in b_{sp} are smaller than 1% even if the above ratio of 0.01 is replaced with 0.001. Continuous size-distribution profiles of major chemical species are obtained from the inversion of the measured mass concentration distribution in the size bins of the Anderson 8-stage air samplers, using the technique described in Dong et al. (2004). The key formulas to calculate the normal distribution of density function (f (D, μ, σ)) were summarized as follows:

$$f(D,\mu,\sigma) = \frac{1}{\sqrt{2\pi}\sigma} e^{-\left(\frac{(D-\mu)^2}{2\sigma^2}\right)}$$

(6)

$$\mu = \bar{y} - \mu\bar{x} \qquad (7)$$

$$\sigma = \frac{n\sum xy - \sum x \times \sum y}{n\sum x^2 - (\sum x)^2} \qquad (8)$$

Where D is log (D_a), and μ and σ are the mean and standard deviation, respectively, of the log (D_a) in the different modes. x is the inverse function value of the cumulative probability of a standard normal distribution in each bin, y is logarithm of D_a lower limit (e.g. 0.43, 0.65, 1.1, 2.1, 3.3, 4.7, 5.8 and 9.0 μm) in each bin. An example of the calculation process was demonstrated in supplementary.

However, this approach is not applicable for the condensation mode because there is only one size bin in this mode. To obtain the number concentrations of all the concerned chemical species in the condensation mode, MMADs ($=10^{\mu}$) of this mode are calculated according to:

$$\text{MMADs} = (D_{a1} \times D_{a2})^{0.5} \tag{9}$$

Where D_{a1} and D_{a2} represent the lower (0.10 µm, limits of detection of Anderson 8-stage air sampler) and upper (0.43 µm) boundaries of this size bin, respectively.

Line 215: Is this size resolved mass from the thermodynamic model on the binned data or the fit data?

We have revised the text as follows: "The ISORROPIA II model was run at the reserved mode (Fountoukis and Nenes, 2007) with input data of K^+, Ca^{2+}, Mg^{2+}, NH_4^+, Na^+, SO_4^{2-}, NO_3^-, Cl^-, RH (40%), and temperature (25°C), to estimate the size-resolved mass concentrations of NaCl, $NaNO_3$, Na_2SO_4, $NaHSO_4$, NH_4Cl, NH_4NO_3, $(NH_4)_2SO_4$, NH_4HSO_4, K_2SO_4, $KHSO_4$, KNO_3, KCl, $MgSO_4$, $Mg(NO_3)_2$, $MgCl_2$, $CaSO_4$, $Ca(NO_3)_2$, $CaCl_2$ and H_2O. Several of these chemical species had extremely low mass concentrations and were thus excluded from the calculation of b_{sp}. Generally, only NaCl, $NaNO_3$, Na_2SO_4, NH_4NO_3, $(NH_4)_2SO_4$, K_2SO_4, $Ca(NO_3)_2$, $CaSO_4$ and H_2O were used to estimate b_{sp} in this study."

Line 217: A section on the DMA and APS size distribution analysis is needed. How was the APS calibrated? How was aerodynamic diameter converted to mobility diameter (or vice versa)? What is the response of the APS to particles of different density? How was density calculated?

The measured particle number concentrations by SMPS and APS were used to assess the accuracy of the estimated particle number concentrations of chemical species in section 3.2.2. We have added the calibration procedure of APS in section 2.2 as follows: "APS was calibrated using 5 sizes solid spheres (polystyrene latex monodisperse)."

We have added the convert formula in section 2.4: "The measured particle number concentrations using SMPS in D_p (similar to D_g) were converted to the particle number concentrations in aerodynamic diameter according to:

$$D_a = D_p/(\rho)^{0.5} \tag{6}$$

$$\rho = \frac{\sum_{chemical\ species} m_i}{\sum_{chemical\ species} \frac{m_i}{\rho_i}} \tag{7}$$

Where, ρ represents the daily average densities of particle, m_i is chemical species mass concentration in a bin, ρ_i is chemical species density. The seasonal average densities of particle were calculated in Fig. S4."

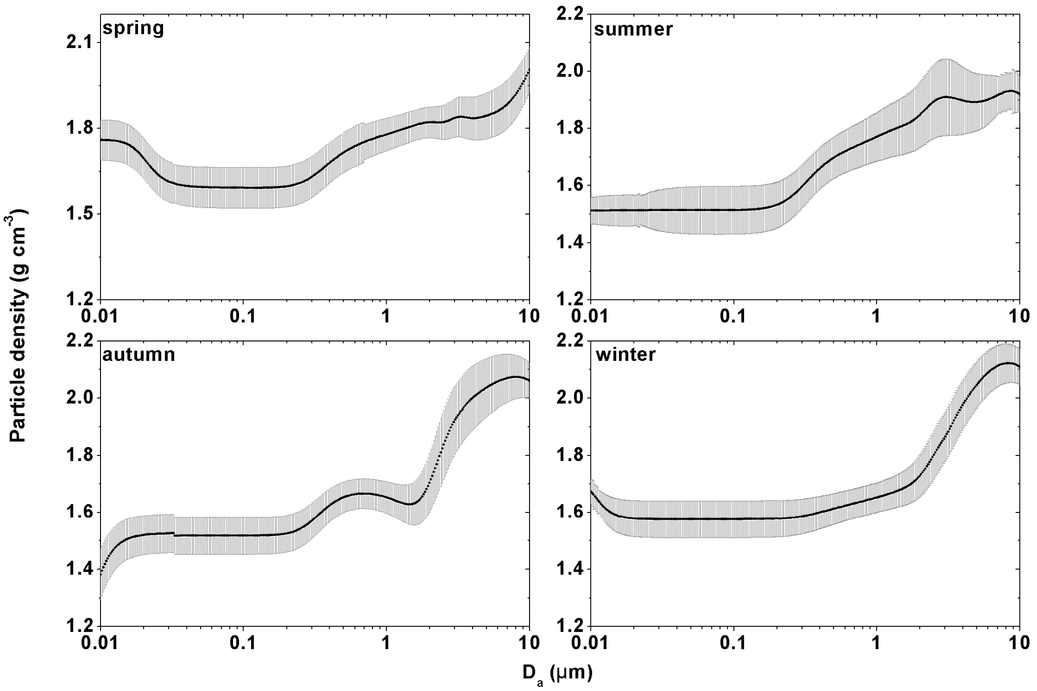


Fig. S4: Continuous log-normal size distributions of seasonal average densities in four seasons.

Line 225: Is PM_{10} here the bulk gravimetric PM_{10} or the summed data from the impactor? Does this include water at the RH of the PM_{10} gravimetric measurement? Particle bound water can still exist for 40% RH.

Here, PM_{10} mass was the sum of the size-segregated mass concentrations. We revised the text as follows: "On annual average, 10±2%, 48±7% and 42±8% of total mass in the size-segregated samples were in the condensation, droplet and coarse modes, respectively, with the average MMADs being 0.78±0.07 μm in the droplet mode and 4.57±0.42 μm in the coarse mode." Yes, particles content a small amount of water even at RH=40% according to the ISORROPIA II model (as shown in Fig.2).

Line 226-227: Units for MMAD?

Units added for MMAD in the revised paper.

Line 226: How were MMADs calculated for the 'continuous' lognormal data?

We have added the key formulas for calculating MMADs in section 2.4. The formula in the droplet and coarse modes : MMADs ($=10^{\mu}$). The formula in the condensation mode: MMADs $=(D_{a1}\times D_{a2})^{0.5}$, where, D_{a1} and D_{a2} represent the lower (0.10 μm, limits of detection of Anderson 8-stage air sampler) and upper (0.43 μm) boundaries of this size bin, respectively. Here MMADs in the condensation mode were only used for estimating the continuous lognormal chemical species mass and number concentrations.

Line 226: Again, reporting an MMAD for the condensation mode is meaningless.

We have deleted the statements on the MMADs for the condensation mode in this manuscript. However, the MMADs in the condensation mode were still used to estimate the continuous lognormal chemical species mass and number concentrations.

Line 229: Define PRD.

We have added the text “the Pearl River Delta (PRD) region”.

Line 237: Close to what?

We have revised the text as follows: “Seasonal average particle mass concentrations were evidently lower in summer than in the other seasons for the condensation and droplet modes, and were similar during spring, autumn and winter for all the three modes.”

Line 289: What about K^+ in the fine mode?

We have added the size distribution of K^+ in Fig. S5.

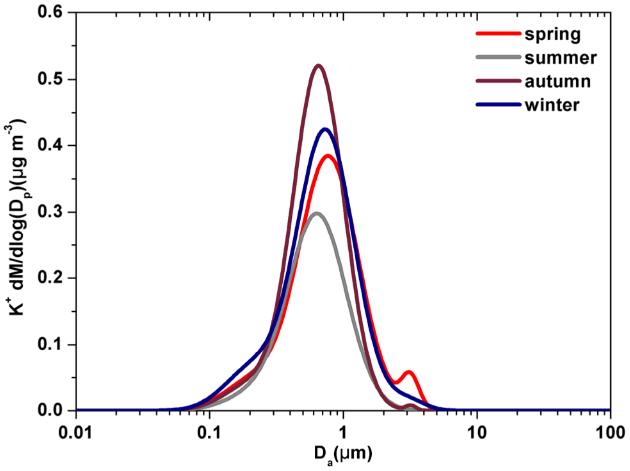


Fig. S5: Continuous log-normal size distributions of K^+ in four seasons.

Line 336: Coarse mode mass fractions also depend on other species. Do the authors mean their absolute concentrations rather than relative concentrations?

Yes, coarse mode mass fraction also depended on chemical species especially OM and crustal element oxides. Here, PM_{10} mass concentration was the absolute (not relative) concentration. We have revised the text as follows: “Annual average PM_{10} concentrations (46 μg m^{-3}) in 2015-2016 in the PRD region were about 40% lower than that (76 μg m^{-3}) in 2006-2007, which further supported the above hypothesis.”

Line 339: Change title to “Closure of particle mass, number concentration, and bsp”

Revised as suggested.

Line 345: Was sulfate fully neutralized for the duration of the study?

Sulfate was fully neutralized by NH_4^+, Na^+, K^+ and Ca^{2+} according to the ISORROPIA II model.

Line 355: Was 5% used here?

We used 5.3% as stated in the supplementary. We have added such explanation in the revised paper: “Alternatively, crustal element oxides mass concentration was estimated from Ca^{2+} mass concentration because of their good correlations (slope=0.053, R^2=0.79) as was found in a previous study (Fig. S6) (Tao et al., 2017b). It was suggested that Ca^{2+} accounted for 5.3% of crustal element oxides in $PM_{2.5}$ in urban Guangzhou, a value that is close to the content of Ca^{2+} (5.0%) in soil dust source profiles (representing crustal element oxides) in $PM_{2.5}$ in cities of southern China (Sun et al., 2019). Because $CaSO_4$ and $Ca(NO_3)_2$ were mainly from the reactions between calcium oxide and acids (e.g. H_2SO_4 and HNO_3), the estimated mass concentration of crustal element oxides needs to deduct those of $CaSO_4$ and $Ca(NO_3)_2$.”

Line 362: I am not sure what is meant here by the “total”? How was “total” derived in this context?

We have deleted the word “total”. We originally referred to the sum of the condensation, droplet and coarse mode mass concentrations.

Line 371: Same comment as the previous.

The word “total” is now deleted.

Line 382: Please provide more details regarding this method.

We have added the formula for calculating number concentration of chemical species in section 2.4, which reads: “$N_{i,j}$ is number concentration of chemical species calculated by the formula (3).

$$N = \frac{6C}{\pi \rho D^3} \tag{3}$$

Where, N is chemical species number concentration, C is chemical species mass concentrations, ρ is density of chemical species (Table S1), D is geometric diameter (D_g) of chemical species.

The particle number concentrations in aerodynamic diameter (D_a) were converted to the particle number concentrations in D_g (similar to D_p) according to:

$$D_a=D_g/(\rho)^{0.5} \quad (4)$$

$$\rho = \frac{\sum_{chemical\ species} m_i}{\sum_{chemical\ species} \frac{m_i}{\rho_i}} \quad (5)$$

Where, ρ represents the daily average densities of particle, i is chemical species, m_i is chemical species mass concentration in a bin, ρ_i is chemical species density. The seasonal average densities of particle were calculated in Fig. S4."

Line 384: This would be expected because of the diameter-cubed dependence between number and mass.

We agree with this comment and we deleted the irrelevant statement.

Line 386: What is the difference in the definition of the estimated NMAD of the number concentrations of individual species and the NAMD of particle number concentrations? (individual versus particle?). I think an issue here is that the constant 0.21 m value is meaningless.

The sums of the individual species number concentrations were the particle number concentrations. We have revised the text as follows: "The estimated number mean aerodynamic diameters (NMADs) of the number concentrations of individual chemical species mainly distributed in the range of 100-120 nm. The estimated NMADs of particle number concentrations (sum of individual chemical species number concentrations in the same size bin) were close to about 100 nm in the four seasons, which was larger than the NMADs (30-70 nm) of the simultaneously measured particle number concentrations by the SMPS and APS (Fig. 4)."

Line 392: What densities were used?

The densities of the individual chemical species are listed in Table S1.

Table. S1 The refractive indices and densities of chemical species.

Chemical species	refractive index	density(g cm^{-3})	Chemical species	refractive index	density(g cm^{-3})
NaCl	1.54-0i	2.16	$Ca(NO_3)_2$	1.53-0i	2.50
$NaNO_3$	1.59-0i	2.26	H_2O	1.33-0i	1.00
Na_2SO_4	1.48-0i	2.68	OM	1.55-0i	1.40
$(NH_4)_2SO_4$	1.53-0i	1.76	EC	1.80-0.54i	1.50
NH_4NO_3	1.55-0i	1.73	crustal element oxides	1.56-0.01i	2.66
K_2SO_4	1.49-0i	2.66	unidentified fraction	1.58-0.01i	2.00
$CaSO_4$	1.57-0i	2.61			

Line 397: The reasoning here isn't clear. The size segregated chemical mass species concentrations should be dry. Unless the authors mean that particle bound water was associated with a gravimetric measurement, the individual species mass do not include water.

The size segregated particle mass concentrations and chemical species mass concentrations were weighted and estimated under a dry condition (temperature = 25°C and relative humidity = 40%). Besides chemical species, water was also resolved in the size segregated samples according to ISORROPIA II model (Fig. 2). In contrast, the particle number concentrations were measured under dry condition (relative humidity < 30%). Moreover, little water was resolved in the size segregated samples according to ISORROPIA II model. To some extent, chemical species likely internally mixed with chemical species in the real world and resulted in the larger diameter of chemical species than the measured ones under dry condition.

Line 418-420: I am unclear as to why scattering efficiencies are being discussed here?

We agree with this comment and have deleted this part.

Line 421-423: This is the first discussion of these design flaws – are the authors referring to the single bin for the condensation mode?

Yes, it was because we cannot get the MMAD in the condensation mode and cannot accurately estimate the number concentration especially those of <100 nm.

Line 426: How much higher?

We have added this text: "On annual average, the estimated particle number concentrations in the range of 430 nm-10 μm based on the size-segregated chemical species mass concentrations were 33±42% higher than those measured by the SMPS and APS."

Line 429: How do the authors know that EC was internally mixed with OM or inorganic salts during this study?

Here we only tried to interpret the possible reasons of the overestimation of the particle number concentrations by the size-segregated chemical species mass concentrations. We have deleted this statement in the revised paper.

Line 433-434: The reasoning here is unclear. What are the estimation errors and models?

This refers to the estimated particle number concentrations by the size-segregated chemical species mass concentrations using the inversion technique and ISORROPIA II model. We have revised the text as follows: "To some extent, the intercepts represent the measurement errors of SMPS and APS and estimation errors of the inversion technique and ISORROPIA II models."

Line 448: The authors need to provide more details on how they derived bsp. What refractive indices did they use, how did they calculate them, which number size distributions did they use, etc.

We have added description of several key input parameters of Mie model for estimating b_{sp}, which reads: "Daily b_{sp} was estimated using Mie model (in section 2.4) with input parameters including refractive indices, densities and number concentrations in 401 bins of chemical species (NaCl, $NaNO_3$, Na_2SO_4, $(NH_4)_2SO_4$, NH_4NO_3, K_2SO_4, $CaSO_4$, $Ca(NO_3)_2$, H_2O, OM, EC, crustal element oxides and unidentified fraction). The refractive indices and densities of above chemical species are summarized in Table S1."

Line 455: Why "especially the inversion technique method"?

We agree that this statement is a bit confusing and we have deleted the word "especially".

Line 460: What do the authors mean that OC was underestimated by the OC/EC protocol?

This is because OC of size-segregated samples is defined as OC1 + OC2 + OC3 + OC4 rather than OC1 + OC2 + OC3 + OC4 + OP due to the low OP concentration in each bin.

Line 468, 472: Do the authors mean "inline" data?

We have replaced the word as suggested.

Line 502: What did the authors use for refractive indices for the "unidentified fraction"?

We have added the refractive indices and densities of chemical species in supplementary (Table S1). The refractive index of the unidentified fraction is 1.58-0.01i.

Line 517, 521: I am not sure what is meant by "particle and chemical species". What is the distinction?

Particle MSE was estimated by sum of b_{sp} from individual chemical species divided by sum of particle mass concentration. MSEs of individual chemical species was estimated by b_{sp} using Mie model according to its particle number in 401 bins, refractive index and density divided by its mass concentration. Thus, we have clarified this part as follows: "Here, only the MSEs of $(NH_4)_2SO_4$, NH_4NO_3, OM, EC, crustal element oxides and unidentified fraction in the condensation, droplet, fine (sum of condensation and droplet), and coarse modes were estimated (Table 3), considering these chemical species accounted for more than 90% of the estimated b_{sp}. However, particle MSEs in the condensation, droplet, fine and coarse modes were estimated by sum of b_{sp} from individual chemical species divided by sum of particle mass"

Line 540: This points back to the previous comments as well. Was "particle MSE" estimated by summed bsp from individual species divided by summed particle mass, or was bsp calculated for "particle", which then would require a "particle" refractive index? It would help if the authors provided details for how these things are calculated (see comment for line 448).

See our clarification in the previous comment.

Line 577: Define MMGD

We have redefined the GMMD as geometric mass mean diameters (MMGD) of chemical species ($(NH_4)_2SO_4$, NH_4NO_3 and OM), which was converted from MMAD and its density according to the formula (6) in section 2.4.

Line 626-627: Sentence is unclear.

We have simplified this part as follows: "Different from the approach used for fine particle MSE, the MSEs of $(NH_4)_2SO_4$, NH_4NO_3 and OM in the droplet mode were determined using measurement-based their mass size distributions prescribed as log-normal size distributions. MSEs of these chemical species strongly depend on their size-distributions, which were defined here as log-normal distributions with three parameters including mass concentration (in the range of 0.43 - 2.1 μm), MMAD and standard deviation (σ)."

Line 630: How were sigma values calculated?

We have added the key formulas in section 2.4, which included the calculation method of sigma and MMAD.

Line 653: What does "bulk particle" mean?

The bulk particle means the sum mass concentration of the condensation, droplet and coarse modes. We have revised the text as follows: "and particle mass, NO_3^-, OC, Na^+, Ca^{2+} and Cl^- in both droplet and coarse modes."

Line 670: Sea salt in the IMPROVE formula is assumed to have a mass mean diameter of 2.5 um, so it is assumed to be in the coarse mode with the tail extending into the PM2.5 mode. Figures and Tables: Table 2: Define size range of condensation, droplet and coarse modes. Again, reporting 0.21 um for all condensation mode MMAD is meaningless. Define “MMAD” in the caption.

We agree with this comment, and we also suspected that sea salt may distribute in the tail of $PM_{2.5}$. However, we cannot find NaCl in $PM_{2.5}$ according to the ISORROPIA II model. In fact, we found a large amount of Na_2SO_4 in $PM_{2.5}$, which would be related with aged sea salt. Here, we referred sea salt as NaCl rather than Na_2SO_4. We have deleted 0.21 μm for all condensation modes in Table 2. We defined the size ranges in the different modes in Table 2 and defined MMAD in the subtitle.

Table 3: Define size range of condensation and droplet modes. Again, reporting 0.21 um for all condensation mode MMAD is meaningless. Define “MSE” and “MMAD” in the caption. Include wavelength and relative humidity (Dry = ?%) in the caption or subtitle.

We have deleted 0.21 μm for all the condensation modes in Table 3. We have added the size ranges for the different modes in Table 3. Table caption revised as suggested.

Figure 1: Define “PRD” in the caption.

Revised as suggested.

Figure 2: Was CaSO4 and Ca(NO3)2 subtracted out of the soil formula when using Ca to calculate soil? These figures suggest that EC mass size distributions are larger than OM distributions? Are these stacked? If so EC » SO4 but mass concentrations in Table 2 suggests this is not the case. The presentation is somewhat confusing. Keep the y-axis the same for all seasons for easier comparisons.

We have clarified the relationships between the estimated soil mass concentration and calcium salts ($CaSO_4$ and $Ca(NO_3)_2$) in section 3.2.1, which reads: “Alternatively, crustal element oxides mass concentration was estimated from Ca^{2+} mass concentration because of their good correlations (slope=0.053, R^2=0.79) as was found in a previous study (Fig. S6) (Tao et al., 2017b). It was suggested that Ca^{2+} accounted for 5.3% of crustal element oxides in $PM_{2.5}$ in urban Guangzhou, a value that is close to the content of Ca^{2+} (5.0%) in soil dust source profiles (representing crustal element oxides) in $PM_{2.5}$ in cities of southern China (Sun et al., 2019). Because $CaSO_4$ and $Ca(NO_3)_2$ were mainly from the reactions between calcium oxide and acids (e.g. H_2SO_4 and HNO_3), the estimated mass concentration of crustal element oxides needs to deduct those of $CaSO_4$ and $Ca(NO_3)_2$.”

The mass size distributions of OM were in fact larger than those of EC in four seasons, although Figure 2 seems to show an opposite result, which was due to the overlap of chemical species. The annual average size distributions of the individual species of $(NH_4)_2SO_4$ OM and EC are plotted below. We have also revised the scale of the y-axis in all the figures.

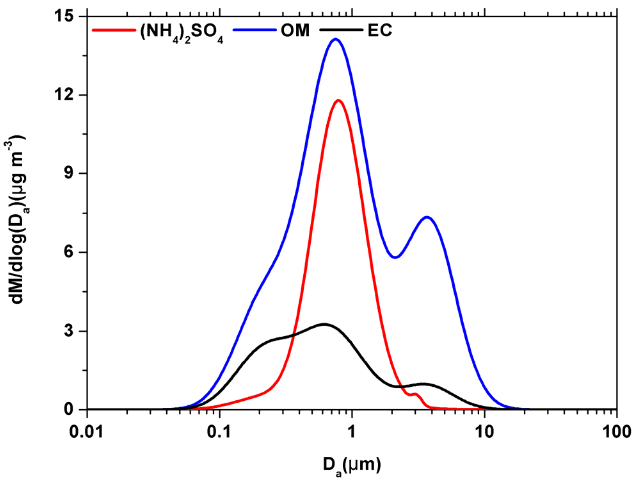


Fig. Suppl. Continuous log-normal size distributions of $(NH_4)_2SO_4$, OM and EC.

Figure 3: Similar comments as previous caption.

See our response in the previous comment.

Figure 4: It would help to plot the APS data in terms of mass or volume instead of number based on the size range- the larger modes would be more visible.

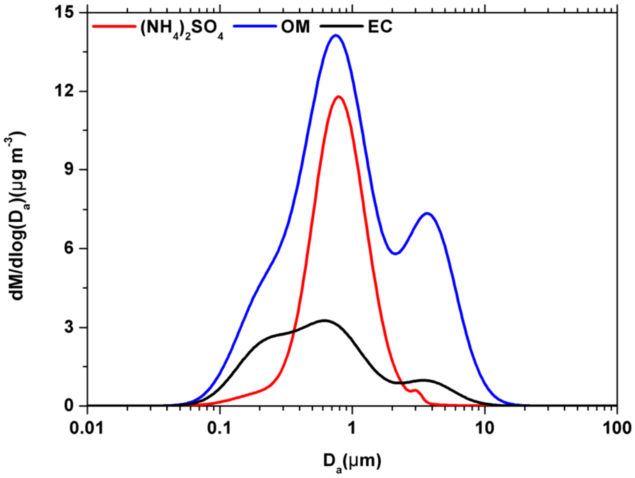
Although it may be more visible using mass or volume data than using number data, it is the number concentration that was directly measured by SMPS and APS. Moreover, the input data of Mie model also need number concentration of chemical species. Thus, we used the measured number concentrations by SMPS and APS to close the estimated number concentrations of chemical species.

Figure 6: Include wavelength, relative humidity conditions, and size range. Is this total bsp?

We revised the caption as follows: "Fig. 6. Correlations between the measured b_{sp} in TSP at wavelength of 520 nm under dry condition (relative humidity <30%) and estimated b_{sp} in PM_{10} at wavelength of 550 nm under dry condition (relative humidity =40%) in four seasons."

Figure 7: Similar comments to figure 2. I don't understand how the mass of sulfate can be so much higher than EC yet the EC scattering is greater?

Figure 7 shows the contributions of chemical species including NaCl, $NaNO_3$, Na_2SO_4, NH_4NO_3, $(NH_4)_2SO_4$, K_2SO_4, $CaSO_4$, $Ca(NO_3)_2$, H_2O, OM, EC, crustal element oxides and unidentified fraction to the estimated b_{sp} in the different sizes (0.1-10μm in 401 bins). The different size distributions between $(NH_4)_2SO_4$ and EC caused the higher contribution from $(NH_4)_2SO_4$ to the estimated b_{sp} despite its lower mass concentrations.



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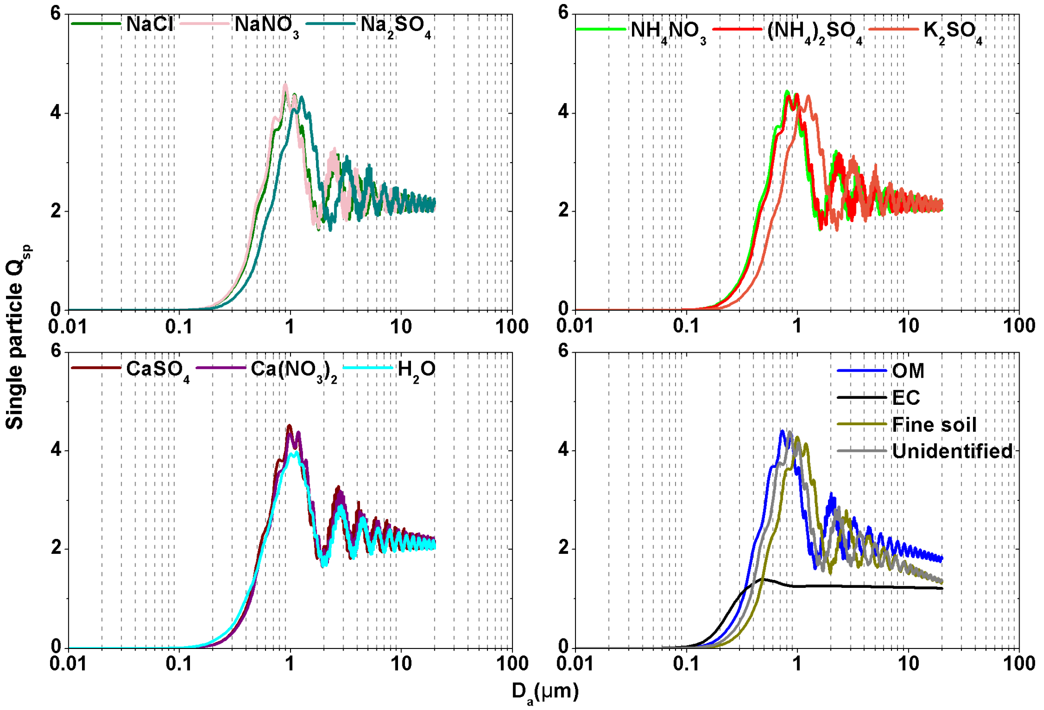

Fig. S3: Single particle scattering efficiencies of the dominant chemical species.

Figure 8: Define "MSE", "fine", wavelength, and relative humidity conditions in the caption.

Figure caption has been revised as suggested.

Figure 9: Caption doesn't include any information on MSE. Also include wavelength, and relative humidity conditions in the caption.

Figure caption has been revised as suggested.