

Anonymous Referee #1

This manuscript presents the results of an intensive field campaign of two week duration in an area south of Beijing. The manuscript focuses on the study of the effect of aerosol hygroscopicity in the aerosol light scattering coefficient. Despite the short measurement period, the data collected are interesting and the results are of scientific significance. Nevertheless, I have some minor comments that need to be clarify by the authors.

Line 78: Briefly describe the humidifying scheme by Carrico et al. (1998)

Reply: The humidifying scheme by Carrico et al. (1998) is described in the manuscript as: “The water vapor controlled by the temperature of the liquid water in the outer annulus of the tube passes through a Teflon membrane, humidifying aerosols in the inner tube (Carrico et al., 1998). The temperature of the liquid water was controlled by adjusting the power of the water baths.”

[Lines 84-86]

Line 80: Was the tandem nephelometer calibrated with ammonium sulphate or other salt of known hygroscopicity? This is highly recommended to assure that the system is functioning correctly and that the RH inside the nephelometer chamber is correct (see Burgos et al. (2019) and Fierz-Schmidhauser et al. 2010b).

Reply: The tandem nephelometer in this study was calibrated with ammonium sulphate, whose deliquescence relative humidity (DRH) was $79.9 \pm 0.5\%$ at 298 K. The DRH of pure ammonium sulfate aerosols generated in the laboratory was 80.367%, measured by our high-resolution humidified nephelometer system (Fig. R1). This shows that the RH inside the nephelometer chamber was correct and that the system was functioning properly.

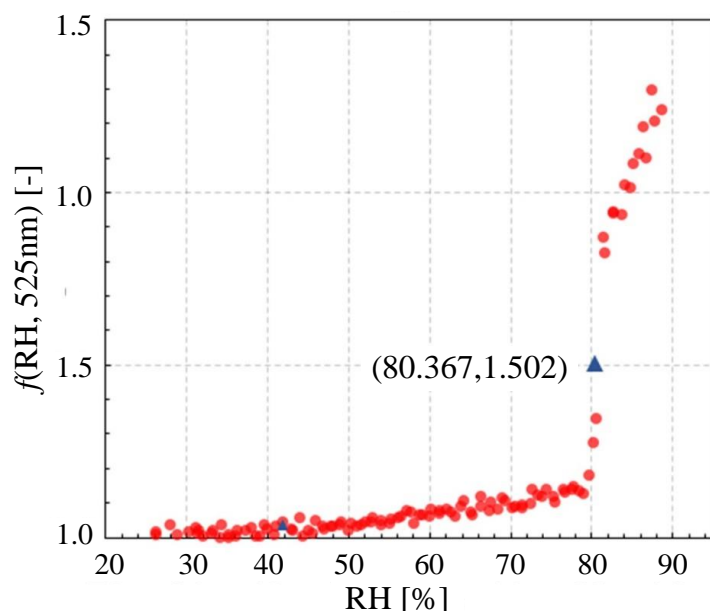


Figure R1: Deliquescence results of the pure ammonium sulfate aerosol generated in the laboratory.

[Lines 108-111]

Line 80: Were the nephelometers operated with or without the kalman filter option?

Reply: Since the RH of aerosols inside the nephelometers was constantly changing, and real measured data at every moment was needed, the nephelometers operated without the Kalman filters.

[Lines 91-92]

Line 90: The equation to calculate the dew point temperature is not the most common one. I looked in the references provided by the authors but Kuang et al. (2017) doesn't state which formula they use to calculate T dew point and the reference of Liu and Zhao (2016) is in Chinese. Please, use appropriate references for this formula.

Also, it would be interesting to see the comparison between RH_{in}, RH_{outlet} and RH_{calculated}.

Reply: We have revised the references in the manuscript. Equations (1) and (2) are from a Ph.D. thesis in Chinese (L. Zhang, 2017), explained as follows. Because the RH levels measured by the probe built into the optical chamber of the wet nephelometer (RH_{chamber}) was imprecise, two calibrated RH and temperature probes were placed at the inlet and outlet of the wet nephelometer, obtaining 1-min averages of RH and temperature (Fig. R2). We used Vaisala HMP110 probes with accuracies of $\pm 0.2^\circ\text{C}$ for the 0–40°C temperature range and $\pm 1.5\%$ RH and $\pm 2.5\%$ RH for the 0–90% and 90–100% RH ranges, respectively. As shown in Figure R2 (b), the temperatures measured by these probes were different. However, in principle, the dew point temperatures ($T_{\text{dew-point}}$) at these positions are all the same. Since the RH and temperature probes at the outlet of the wet nephelometer (RH_{outlet} and T_{outlet}) were less affected by the humidifier, RH_{outlet} and T_{outlet} were used to calculate $T_{\text{dew-point}}$ at this position using Eq. (1):

$$T_{\text{dew-point}} = \text{RH}_{\text{outlet}}^{\frac{1}{8}} (112 + 0.9T_{\text{outlet}}) + 0.1T_{\text{outlet}} - 112. \quad (1)$$

We assume that $T_{\text{dew-point}}$ was approximately the same as that in the optical chamber of the wet nephelometer. Based on the temperature in the optical chamber (T_{chamber}) and $T_{\text{dew-point}}$, the actual RH in the optical chamber (RH_{chamber}) can be calculated by rearranging Eq. (1), i.e.,

$$\text{RH}_{\text{chamber}} = \left(\frac{112 - 0.1T_{\text{chamber}} + T_{\text{dew-point}}}{112 + 0.9T_{\text{chamber}}} \right). \quad (2)$$

[Lines 94-107]

Figures R2 (a) and (b) show time series of RH_{inlet}, RH_{outlet}, and RH_{chamber} and T_{inlet} , T_{outlet} , and T_{chamber} , respectively. Temperatures measured by the three probes are significantly different. Since the RH and temperature probes at the inlet of the wet nephelometer is very close to the humidifying tube, T_{inlet} is influenced by the humidifier. Also, T_{outlet} is lower than T_{chamber} . Figure R1 (a) shows that RH_{chamber} is lower than RH_{outlet}.

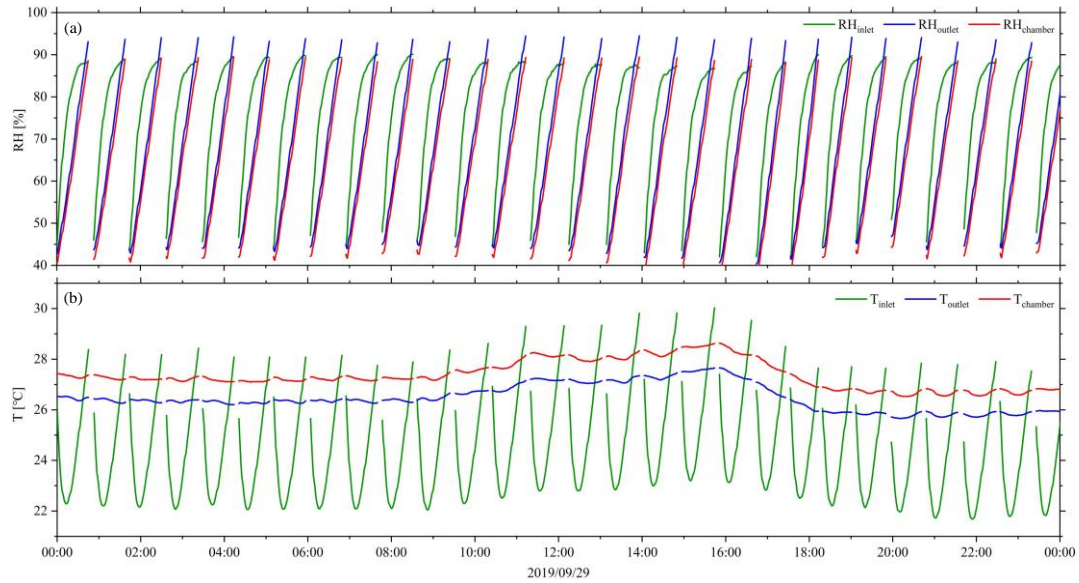


Figure R2: Time series of (a) RH_{in} , RH_{outlet} , and $RH_{chamber}$ and (b) T_{in} , T_{outlet} , and $T_{chamber}$ on 29 November 2019. Times in this figure are in the hour/minute format.

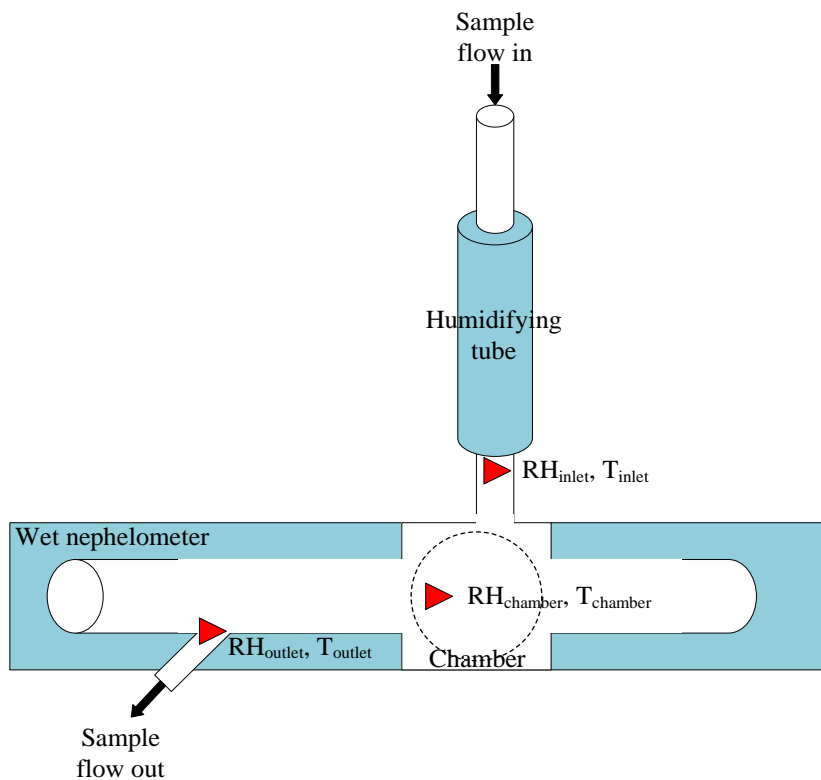


Figure R3: Diagram of the structure of the wet nephelometer. The three red triangles show the locations of the three sets of RH and temperature probes.

Line 105: I really don't understand how the $f(RH)$ is calculated. Why $f(RH)$ is normalized? What is the reason behind this? Also, $f(RH > 40\%)$ is averaged over what? Whole dataset, each scan? Then, in line 108 it is said that $f(RH > 40\%)$ is 1. This is true for all the observations? Is it exactly 1? This calculation needs clarification.

Reply: In this study, we assumed that the aerosol is in the dry state when $RH < 40\%$. So $f(RH)$ should theoretically equal 1 when RH is lower than 40%. This was what was meant by the sentence: “Here, $f(RH < 40\%)$ equals 1.” However, due to systematic errors and the differences in RH measured synchronously by the dry nephelometer and the wet nephelometer, respectively, measured $f(RH < 40\%)$ have small fluctuations and are not equal to 1. This is why $f(RH)$ was normalized. Not $f(RH > 40\%)$, but the corrected coefficient ($f(RH < 40\%)_{averaged}$), which was averaged over the whole dataset of $RH < 40\%$. $f(RH > 40\%)$ was then normalized using Eq. (4).

[Lines 124-129]

Line 112: The absorption coefficient is measured at 7 wavelengths, the absorption coefficient at 520 nm is more appropriate than using the absorption coefficient at 880 nm and then convert it to 525 nm.

Reply: Done. The single-scattering albedo ($\omega_{0(525nm)}$) has also been recalculated. Also updated were Figs. 2b, 2c, 3a and Table 1.

[Lines 130-132]

Eq 6: So, only $f(RH=85\%)$ is used to calculate gamma? If the f_{rh} measurements are performed at scanning RH it can be retrieved from a potential fit using the whole RH range, which will have less errors than using a single RH point (see Zieger et al. 2010, Titos et al., 2016).

Reply: We appreciate the suggestion. Previously, only $f(RH = 85\%)$ and $f(RH = 40\%)$ were used to calculate γ , which has now been revised. The parameter γ is now retrieved from the following $f(RH)$ parametrization scheme: $f(RH) = (1 - RH)^{-\gamma}$ (i.e., Eq. (8)), using the whole RH range (generally from $\sim 40\%$ to $\sim 90\%$). Figures 7 and S7 have been updated accordingly.

[Lines 146-147]

Line 125: Include a reference to Zieger et al. 2010, who firstly introduced the hysteresis index.

Reply: The reference is added.

[Line 142]

Eq9: Actually, what it is here called g , it is usually referred as gamma.

Reply: Thank you.

[Lines 143-151]

Eq. 8: The RH range used to identify deliquescence is very narrow and can miss deliquescence processes occurring at slightly different RH . Maybe consider the procedure of Zieger et al.

Reply: We appreciate the suggestion. The deliquescence observed in this study mainly resulted from the ammonium sulfate in ambient aerosols. The deliquescence RH (DRH) of pure ammonium sulfate aerosols generated in the laboratory was 80.367%, measured by our high-resolution humidified nephelometer system (Fig. S8). Forty-seven cycles of $f(RH)$ (16% of all cycles) in this study show clear deliquescence, and the DRH of all deliquescence are mainly distributed between 78% and 80%. So the RH range used to identify deliquescence in this paper ($78\% < RH < 82\%$) does not miss deliquescent processes. In addition, if we use a larger RH to identify deliquescence, for example, $75\% < RH < 85\%$, less data would be available to calculate $\gamma_{>85\%}$ on each hydration branch.

Fig7: Why not consider all measured species, including NH₄⁺, Cl⁻ and BC? Is the organic mass fraction defined differently than in Figure 6?

Reply: Since chloride ions (Cl⁻) accounted for less than 1% of submicron aerosols during the entire measurement period, the influence of Cl⁻ on the hygroscopic enhancement factor of aerosols is essentially negligible. In addition, chemical species consisting of Cl⁻ are hardly determined. Therefore, the influence of Cl⁻ on γ has not been considered in Fig. 7. As for BC, its influence on $f(\text{RH} = 85\%, 525 \text{ nm})$ has been analyzed in Fig. 5d. Since γ is the parameter that can replace $f(\text{RH})$ over a wider RH range, it is not necessary to discuss the correlation between BC and γ again in Fig. 7. Considering the importance of the ammonia ion (NH₄⁺) on the $f(\text{RH})$ of aerosols in previous studies (Zieger et al., 2010; L. Zhang et al., 2015), it is a helpful suggestion to take NH₄⁺ into account. So an ion-pairing scheme was conducted to calculate the mass concentration of ammonium sulfate ((NH₄)₂SO₄), ammonium bisulfate (NH₄HSO₄), and ammonium nitrate (NH₄NO₃) in aerosols on the basis of the molar numbers of all ions (Gysel et al., 2007). The following is the ion-pairing scheme:

$$\begin{aligned} n_{\text{NH}_4\text{NO}_3} &= n_{\text{NO}_3^-} \\ n_{\text{H}_2\text{SO}_4} &= \max(0, n_{\text{SO}_4^{2-}} - n_{\text{NH}_4^+} + n_{\text{NO}_3^-}) \\ n_{\text{NH}_4\text{HSO}_4} &= \min(2n_{\text{SO}_4^{2-}} - n_{\text{NH}_4^+} + n_{\text{NO}_3^-}, n_{\text{NH}_4^+} - n_{\text{NO}_3^-}) \\ n_{(\text{NH}_4)_2\text{SO}_4} &= \max(n_{\text{NH}_4^+} - n_{\text{NO}_3^-} - n_{\text{SO}_4^{2-}}, 0) \\ n_{\text{HNO}_3} &= 0, \end{aligned} \quad (9)$$

where n denotes the number of moles. Figures 7a-c now show γ as a function of F_{org} (Eq. 6), where C_i represents the sum of (NH₄)₂SO₄ and NH₄HSO₄, NH₄NO₃, and the sum of (NH₄)₂SO₄, NH₄HSO₄, and NH₄NO₃ mass concentrations, respectively. Figure 7 has been revised accordingly. The organic mass fraction defined in Fig. 7 is the same as that in Fig. 6.

[Lines 273-283]

Fig8: Use same color for WD from north (360° and 0°)

Reply: We have revised Fig. 8b accordingly.

Line 220: Don't understand the reasoning, which marine aerosols do the authors refer to?

Reply: The absolute values of both slopes and corresponding standard deviations (0.80±0.04 and 1.00±0.06 for $f(\text{RH} = 85\%, 525 \text{ nm})$ as a function of inorganic and organic matter mass fractions, respectively) shown in Fig. 6 were similar to those reported in Lin'an, China (0.96±0.02 and 1.20±0.04, respectively; L. Zhang et al., 2015) but much lower than those observed at Melpitz, Germany (2.2±0.078 and 3.1±0.1, respectively; Zieger et al., 2014). This might be because the $f(\text{RH} = 85\%, 525 \text{ nm})$ measured in Melpitz, Germany, was much higher than that in Lin'an and Beijing. Ambient aerosols in Melpitz, Germany, were affected by sea salt, like sodium chloride, transported from the North Sea and highly hygroscopic. Marine aerosols have a higher hygroscopicity than aerosols influenced more by human activity.

[Lines 234-240]

Line 245: Do the authors refer to an instrument artefact due to water depletion?

Reply: Yes, we do, because the amount of water vapor passing through the Teflon membrane to humidify aerosols in the inner tube was finite at a specific temperature. The amount of water vapor may be insufficient when the proportion of inorganic matter is very high.

Line 260: It is not that in the previous studies the role of NO₃⁻ was not as important as in the present study. Quinn et al. (2005) didn't look at NO₃⁻, their organic mass fraction was calculated using only SO₄²⁻ as inorganic component. Why do the authors don't include NH₄⁺? Previously they stated the importance of ammonia, but here it is not included. See the relationships obtained by Zieger et al. and Zhang et al.

Reply: The unclear statements have been revised. As we stated in the response to the suggestion given by the reviewer for Fig. 7, an ion-pairing scheme was conducted to calculate the mass concentrations of ammonium sulfate ((NH₄)₂SO₄), ammonium bisulfate (NH₄HSO₄), and ammonium nitrate (NH₄NO₃) in aerosols on the basis of the molar numbers of all ions (Gysel et al., 2007). The following is the ion-pairing scheme:

$$\begin{aligned}
 n_{\text{NH}_4\text{NO}_3} &= n_{\text{NO}_3^-} \\
 n_{\text{H}_2\text{SO}_4} &= \max(0, n_{\text{SO}_4^{2-}} - n_{\text{NH}_4^+} + n_{\text{NO}_3^-}) \\
 n_{\text{NH}_4\text{HSO}_4} &= \min(2n_{\text{SO}_4^{2-}} - n_{\text{NH}_4^+} + n_{\text{NO}_3^-}, n_{\text{NH}_4^+} - n_{\text{NO}_3^-}) \\
 n_{(\text{NH}_4)_2\text{SO}_4} &= \max(n_{\text{NH}_4^+} - n_{\text{NO}_3^-} - n_{\text{SO}_4^{2-}}, 0) \\
 n_{\text{HNO}_3} &= 0,
 \end{aligned} \tag{9}$$

where n denotes the number of moles. The updated Figs. 7a-c show γ as a function of F_{org} (Eq. 6), where C_i represents the sum of (NH₄)₂SO₄ and NH₄HSO₄, NH₄NO₃, and the sum of (NH₄)₂SO₄, NH₄HSO₄, and NH₄NO₃ mass concentrations, respectively. Overall, γ and F_{org} are negatively correlated. The coefficient of determination between γ and F_{org} (Org/(Org+NH₄NO₃)) (Fig. 7b) was higher than that between γ and F_{org} (Org/(Org+(NH₄)₂SO₄+NH₄HSO₄)) (Fig. 7a). The coefficient of determination between γ and F_{org} (Org/(Org+ (NH₄)₂SO₄ + NH₄HSO₄+NH₄NO₃)) was the highest (Fig. 7c). This suggests that nitrate played a more significant role than sulfate in affecting aerosol hygroscopic growth during the study period in Beijing.

[Lines 273-287]

References

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- Zhang, L.: Observation and model study of relative humidity effects on aerosol light scattering at regional background site in the Yangtze delta region, Ph.D. thesis, Chinese Academy of Meteorological Sciences, China, 107pp., 2017.
- Zhang, L., Sun, J., Shen, X., Zhang, Y., Che, H. C., Ma, Q., Zhang, Y., Zhang, X., and Ogren, J. A.: Observations of relative humidity effects on aerosol light scattering in the Yangtze River Delta of China, *Atmos. Chem. Phys.*, 15, 8439–8454, <https://doi.org/10.5194/acp-15-8439-2015>, 2015.

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Anonymous Referee #2

The effects of aerosol chemical composition on the relative humidity dependence of light scattering are presented for a site in Beijing. Parameterizations of $f(\text{RH})$ are developed for different observed conditions (e.g., very clean, moderately polluted, polluted based on measured light scattering levels). The paper is very well written and the figures (with one exception) clearly convey the results of the study. I only have minor comments – see below.

Line 39: change to “that REDUCES the amount”. Also, please add a brief description of how SO₂ control reduces the amount of sulfate.

Reply: Revised. L. Zhang et al. (2015) studied the relationship between the scattering enhancement factor and chemical composition in Lin’an, China, finding that nitrate has a stronger effect on aerosol hygroscopicity than sulfate has, partially due to the rigid control of SO₂ that reduces the amount of sulfate and increases the content of nitrite (Morgan et al., 2010). Apart from sea salt emissions and gypsum dust emissions during construction containing sulfate, sulfate is mainly formed by the oxidation of its gaseous precursor, SO₂, in the atmosphere. In recent years, SO₂ emissions have been reduced substantially through a series of effective measures taken in China, like controlling the burning of loose coal and desulfurizing industrial equipment (Q. Zhang et al., 2019). Reducing SO₂ in the atmosphere thus directly affects the reduction in the sulfate content of aerosols. The saturated vapor pressure of nitric acid (HNO₃) is higher than that of sulfuric acid (H₂SO₄), so the availability of ammonia (NH₃) is key to the partitioning of HNO₃. HNO₃ is often neutralized by NH₃ after H₂SO₄. Therefore, a reduction in SO₂ means that more NH₃ can be used to neutralize HNO₃, leading to higher nitrate concentrations, such as ammonium nitrate (NH₄NO₃), in aerosols (Monks et al., 2019).

[Lines 36-46]

Lines 109 – 111: Why is the absorption coefficient at 880 nm transformed into those at 525 nm? Doesn't the 7-wavelength aethalometer have a measurement wavelength near to 525 nm?

Reply: The absorption coefficient at 520 nm measured by the 7-wavelength aethalometer is more appropriate than using the absorption coefficient at 880 nm. So we chose absorption at 520 nm then converted it to 525 nm. The single-scattering albedo ($\omega_{0(525\text{nm})}$) was also recalculated. Also updated were Figs. 2b, 2c, 3a and Table 1.

[Lines 130-132]

Lines 163 – 164: It is stated that “the proportion of organic matter and BC with weak hygroscopic abilities was low” from the southeast sector. Figure 3d indicates that mass fractions of BC were high in the southeast sector which seems to contradict this statement. Please clarify in the text.

Reply: The text has been revised as: “Figure 3c reveals that strongly hygroscopic aerosols with high values of $f(\text{RH} = 85\%, 525 \text{ nm})$ primarily came from the southeast sector. The proportion of secondary inorganics with strong hygroscopic abilities in aerosols from this direction was high, while the proportion of organic matter with weak hygroscopic abilities was low (Figs. 3e-f). Figure 3d indicates that the mass fraction of BC with weak hygroscopicity was slightly low in the southeast sector when wind speeds were lower than 4 m s⁻¹. However, when wind speeds were higher than 4 m s⁻¹, the mass fraction of BC was relatively high in this direction. Of all data associated with

southeast winds, identified were only three cases with wind speeds higher than 4 m s⁻¹, likely winds of short duration so not representative.”

[Lines 180-186]

Figure 6: The inset figures showing organic mass fraction vs. f(RH) are difficult to read because of their size – especially if a reader is looking at a print version of the paper. I recommend putting the insets into a separate figure.

Reply: We have put the inset figures into a separate figure: Figure S6.

Lines 258 – 260: Please report the mass fractions of organics, SO₄, and NO₃ if they were provided in Malm et al. (2003), Pan et al. (2009), Quinn et al. (2005), and Yan et al. (2009). It is difficult to assess differences in the role of NO₃ versus SO₄ in determining f(RH) in these different regions without knowing the chemical composition reported in these previously published papers.

Reply: The proportions of organics, SO₄²⁻, and NO₃⁻ provided in Malm et al. (2003, 2005), Yan et al. (2009), Pan et al. (2009), and this study are listed in Table R1.

Table R1: Proportions of sulfate, nitrate, and organic matter reported in previous studies and this study.

Site	Ammoniated sulfate or SO ₄ ²⁻ /FM [%]	NH ₄ NO ₃ or NO ₃ ⁻ /FM [%]	OMC/FM [%]	Reference
BBNP	51.0		21.0	Malm et al. (2003, 2005)
GC	31	4.8	44	
GSM	63	0.8	25	
BJ-1	28.0	16.0	34.0	Yan et al. (2009)
BJ-2	23.0	12.0	36.0	Sun et al. (2004)
BJ-3	23.0	14.0	29.0	
XA#1	15.3	7.6	29.8	Pan et al. (2009)
XA#2	17.7	9.4	24.4	
XA#3	11.2	5.2	37.4	
XA#4	5.3	1.5	40.0	
XA#5	9.5	5.7	26.0	
XA#6	6.1	1.2	36.7	
XA#7	8.9	1.1	39.4	
XA#8	10.8	3.2	43.0	
BJ-CMA	19.0	21.0	39.0	This study

Noet: BBNP was the observation site at the Big Bend National Park, Texas.

GC was the site at the Grand Canyon.

GSM was the site at the Great Smoky Mountains.

BJ-1, BJ-2, and BJ-3 were, respectively, affected by traffic emissions, industrial emissions, and anthropogenic emissions.

XA was the site at the Xin’An weather operational station in Baodi County. The symbols ‘#1’ - ‘#8’ represent different sampling dates.

FM: The FM at the XA site represents PM_{2.1}. The FMs at the other sites represent PM_{2.5}.

Lines 261 – 263: Does this mean the Chinese government has made more efforts to control SO₂ emissions than other governments or has made more efforts to control SO₂ than NO_x emissions? Please clarify in the text.

Reply: To address this comment, the following has been added to the revised manuscript:

“In recent years, the Chinese government has made more efforts to control SO₂ emissions (Q. Zhang et al., 2019), e.g., adjusting and optimizing industrial capacities. Clean fuels have also been promoted in the residential sector, with trials for using clean energy in heating in northern China carried out in all "2+26" cities and in the Fenhe and Weihe River Plains. In addition, compliance with industrial emission standards has been strengthened. Desulfurization technology has been also applied to many heavy industrial facilities. However, China has many small-scale manufacturing enterprises, so it is much more difficult to regulate NO_x emissions than SO₂ emissions. H. Li et al. (2109) have reported that emissions of SO₂ and NO_x in 2017 dropped by 79.9% and 38.1%, respectively, from 2014 levels in Beijing, China. In 2020, SO₂ and primary PM_{2.5} emissions dropped to one million tons, while NO_x and volatile organic compound emissions were still ten million tons.”

[Lines 287-295]

Line 315: Please define “DF”.

Reply: Done. DF is the difference between $f(\text{RH} = 85\%, 525 \text{ nm})$ and $f(\text{RH} = 80\%, 525 \text{ nm})$, i.e., $f(\text{RH} = 85\%, 525 \text{ nm})$ minus $f(\text{RH} = 80\%, 525 \text{ nm})$.

[Lines 347-348]

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