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(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 SO_2 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, SO_2, in the atmosphere. In recent years, SO_2 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 SO_2 in the atmosphere thus directly affects the reduction in the sulfate content of aerosols. The saturated vapor pressure of nitric acid (HNO_3) is higher than that of sulfuric acid (H_2SO_4), so the availability of ammonia (NH_3) is key to the partitioning of HNO_3. HNO_3 is often neutralized by NH_3 after H_2SO_4. Therefore, a reduction in SO_2 means that more NH_3 can be used to neutralize HNO_3, leading to higher nitrate concentrations, such as ammonium nitrate (HN_4NO_3), 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 ($\omega_{0(525nm)}$) 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(RH = 85%, 525 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, SO_4^{2-}, and NO_3^- 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_4NO_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) Sun et al. (2004)
BJ-2	23.0	12.0	36.0	
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 NOx 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 SO_2 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 SO_2 emissions. H. Li et al. (2109) have reported that emissions of SO_2 and NO_x in 2017 dropped by 79.9% and 38.1%, respectively, from 2014 levels in Bejing, China. In 2020, SO_2 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(RH = 85%, 525 nm) and f(RH = 80%, 525 nm), i.e., f(RH = 85%, 525 nm) minus f(RH = 80%, 525 nm).

[Lines 347-348]

References

Gysel, M., Crosier, J., Topping, D. O., Whitehead, J. D., Bower, K. N., Cubison, M. J., Williams, P. L., Flynn, M. J., McFiggans, G. B., and Coe, H.: Closure study between chemical composition and hygroscopic growth of aerosol particles during TORCH2, Atmos. Chem. Phys., 7, 6131–6144, https://doi.org/10.5194/acp-7-6131-2007, 2007.

Li, H., Cheng, J., Zhang, Q., Zheng, B., Zhang, Y., Zheng, G., and He, K.: Rapid transition in winter aerosol composition in Beijing from 2014 to 2017: response to clean air actions, Atmos. Chem. Phys., 19, 11,485–11,499, https://doi.org/10.5194/acp-19-11485-2019, 2019.

Malm, W. C., Day, D. E., Kreidenweis, S. M., Collett, J. L., and Lee, T.: Humidity-dependent optical properties of fine particles during the Big Bend Regional Aerosol and Visibility Observational Study, J. Geophys. Res. Atmos., 108, 4279, https://doi.org/10.1029/2002JD002998, 2003.

Malm, W. C., Day, D. E., Kreidenweis, S. M., Collett, J. L., Carrico, C., McMeeking, G., and Lee, T.: Hygroscopic properties of an organic-laden aerosol, Atmos. Environ., 39, 4969–4982, https://doi:10.1016/j.atmosenv.2005.05.014, 2005.

Monks, P. S., Granier, C., Fuzzi, S., Stohl, A., Williams, M. L., Akimoto, H., Amann, M., Baklanov, A., Baltensperger, U., Bey, I., Blake, N., Blake, R. S., Carslaw, K. S., Cooper, O. R., Dentener, F. J., Fowler, D., Fragkou, E., Frost, G. J., Generoso, S., Ginoux, P., Grewe, V., Guenther, A., Hansson, H. C., Henne, S., Hjorth, J., Hofzumahaus, A., Huntrieser, H., Isaksen, I. S. A., Jenkin, M. E., Kaiser, J., Kanakidou, M., Klimont, Z., Kulmala, M., Laj, P., Lawrence, M. G., Lee, J. D., Liousse, C., Maione, M., McFiggans, G. B., Metzger, A., Mieville, A., Moussiopoulos, N., Orlando, J. J., O'Dowd, C. D., Palmer, P. I., Parrish, D. D., Petzold, A., Platt, U., Poschl, U.,

A.S.H. Prévôt, A. S. H., Reeves, C. E., Reimann, S., Rudich, Y., Sellegri, K., Steinbrecher, R., Simpson, D., ten Brink, H., Theloke, J., van Der Werf, G. R., Vautard, R., Vestreng, V., Vlachokostas, C., and von Glasow, R.: Atmospheric composition change – global and regional air quality, Atmos. Environ., 43, 5268–5350, https://doi.org/10.1016/j.atmosenv.2009.08.021, 2009.

Morgan, W. T., Allan, J. D., Bower, K. N., Esselborn, M., Harris, B., Henzing, J. S., Highwood, E. J., Kiendlerscharr, A., Mcmeeking, G. R., Mensah, A. A., Northway, M. J., Osborne, S., Williams, P. I., and Krejci, R.: Enhancement of the aerosol direct radiative effect by semi-volatile aerosol components: airborne measurements in northwestern Europe, Atmos. Chem. Phys., 10, 8151–8171, https://doi.org/10.5194/acp-10-8151-2010, 2010.

Pan, X. L., Yan, P., Tang, J., Ma, J. Z., Wang, Z. F., and Gbaguidi, A.: Observational study of aerosol hygroscopic growth factors over rural area near Beijing megacity, Atmos. Chem. Phys., 9, 5087–5118, https://doi.org/10.5194/acpd-9-5087-2009, 2009.

Sun, Y., Wang, Z., Dong, H., Yang, T., Li, J., Pan, X., Chen, P., and Jayne, J. T.: Characterization of summer organic and inorganic aerosols in Beijing, China with an aerosol chemical speciation monitor, Atmos. Environ., 51, 250–259, https://doi.org/10.1016/j.atmosenv.2012.01.013, 2012.

Yan, P., Pan, X., Tang, J., Zhou, X., Zhang, R., and Zeng, L.: Hygroscopic growth of aerosol scattering coefficient: a comparative analysis between urban and suburban sites at winter in Beijing, Particuology, 7, 52–60, https://doi.org/10.1016/j.partic.2008.11.009, 2009.

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.

Zhang, Q., Zheng, Y., Tong, D., Shao, M., Wang, S., Zhang, Y., Xu, X., Wang, J., He, H., Liu, W., Ding, Y., Lei, Y., Li, J., Wang, Z., Zhang, X., Wang, Y., Cheng, J., Liu, Y., Shi, Q., Yan, L., Geng, G., Hong, C., Li, M., Liu, F., Zheng, B., Cao, J., Ding, A., Gao, J., Fu, Q., Huo, J., Liu, B., Liu, Z., Yang, F., He, K., and Hao, J.: Drivers of improved $PM_{2.5}$ air quality in China from 2013 to 2017, P. Natl. Acad. Sci. USA, 116, 24,463–24,469, https://doi.org/10.1073/pnas.1907956116, 2019.