

We are very grateful to the numerous constructive suggestions and very detailed comments !

- In general,  $f(\text{RH})$  is called the “particle light scattering enhancement factor” (or short scattering enhancement factor) and I would like to encourage you to use this term instead of “hygroscopic enhancement factor” which is too general. Hygroscopic growth in general can influence/increase or enhance the particle size, the light back scattering, and other parameters.

Reply: The terms have been corrected per your suggestion.

- In the abstract: I would suggest to re-phrase the sentence starting in line 18 to something like: “The effect is measured by the particle light scattering enhancement  $f(\text{RH})$ , where RH denotes the relative humidity, which is found to be ...”

Reply: Revised.

- Line 31: Replace “ambient” with “elevated”.

Reply: Revised.

- Line 32: Add “usually” before  $\text{RH} < 40\%$ . The 40% RH is a recommendation from WMO/GAW for aerosol monitoring and dependent on the instrument and set-up.

Reply: Done.

- Line 61: In our 2015-paper we actually added further measurements from Hyytiälä (Finland) and the correlations to sulfate and nitrate were actually much higher (see Table 3 and e.g. Figure 6 in <https://acp.copernicus.org/articles/15/7247/2015/doi:10.5194/acp-15-7247-2015>)

Reply: The pertinent finding of your paper is referred to in the revised manuscript: Zieger et al. (2015) also found that the mass fraction of sulfates was strongly correlated with  $\gamma$ , while the mass fraction of nitrates had a low correlation in Hyytiälä, Finland.

For the measurements at different sites, a common linear behavior has been found for inorganic and organic matter mass fractions, while individual inorganic matter such as nitrates or sulfates may show different functional dependencies (L. Zhang et al., 2015; Zieger et al., 2014, 2015). So more field measurements including the  $f(\text{RH})$  and chemical composition of aerosol in different seasons and regions should be added to obtain a more reliable estimate of  $f(\text{RH})$ .

- Section 2.1: More information on the actual inlet needs to be added here (Inlet with cut-off? Length? Was it heated? Drying applied? Average RH within the sampling line? What were the flows? etc).

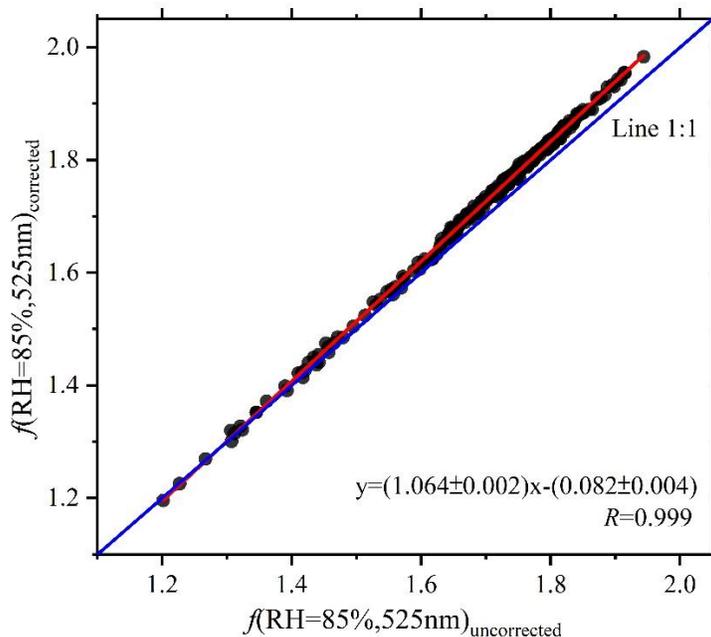
Reply: We appreciate the suggestion. The instruments used in this field experiment include a dual-nephelometer system (Aurora 3000, Ecotech), an aerosol chemical speciation monitor (ACSM; Aerodyne Research Inc.) and a seven-wavelength aethalometer (AE33, Magee Scientific). They were all located in a mobility container on the ground. There are two air conditioners inside the container whose temperature was maintained at about 23 °C. Sample air (16.7 lmp) went through a  $\text{PM}_{2.5}$  cyclone inlet at about 4m above the ground, which only allowed particles with an aerodynamic diameter smaller than 2.5  $\mu\text{m}$  to enter, and then dried by a Nafion dryer (MD-700-36F-3, Perma Pure LLC). The average RH within the sampling line was

about 30%. The sample air was not heated.

- Sect 2.2: Did you apply the truncation and illumination correction for your humidified nephelometer measurements?

Reply: In the paper, the truncation and illumination correction has been applied to the scattering coefficient with the Mie scattering calculation using the observational data of aerosol size distribution. However, there were some mistakes in the observed data of aerosol size distributions. So now, we use the AO98 method by Anderson and Orgen (1998) to correct the scattering coefficient again in the new revision, and all the relevant variables, such as scattering coefficient ( $\sigma_{sp,525nm}$ ), single scattering albedo ( $\omega_{0(525nm)}$ ) and scattering Ångström exponent ( $\alpha_{(450nm-635nm)}$ ) are replaced with the newly corrected data in the current revision.

For the  $f(RH)$  calculations, there are no truncation and illumination correction applied to the scattering coefficients of both dry and humidified nephelometer. The comparison of the deviation between corrected and uncorrected  $f(RH=85\%,525nm)$  is shown in Figure R1. The linear least square regression slope  $\pm$  standard deviation is  $1.064\pm 0.002$ , the intercept  $\pm$  standard deviation is  $-0.082\pm 0.004$  and  $R$  is 0.999. The fitted line is very close to the line of 1:1. The uncorrected  $f(RH=85\%,525nm)$  is a little lower than the corrected  $f(RH=85\%,525nm)$ .



**Figure R1: The relationship between the uncorrected  $f(RH=85\%,525nm)$  and corrected  $f(RH=85\%,525nm)$ . The solid red line represents the linear least square regression. The blue line is the line of 1:1. The linear regression function and the Pearson's correlation coefficient ( $R$ ) are given in the bottom-right corner of the panel.**

- Line 114: Please state the actually measured mean  $\pm$  STD of the sample RH here (“usually < 40%” is not sufficient).

Reply: Done. The actually measured mean  $\pm$  standard deviation of the sample RH was  $28.75\pm 5.50\%$ .

- Line 119: You state that calibrations were performed once a month but you only present 14 days of data. Please be more specific here by stating when which calibrations were performed.

Reply: Manual ‘full calibration’ and zero check and span check of the two nephelometers were performed at 10:30 on September 19<sup>th</sup>, 2019.

- I am still a bit puzzled (similar as reviewer #1) about the calculations of the dew point temperatures (Eq 1). The current equation 1 would need a proper reference. Why have you not used the Magnus formula?

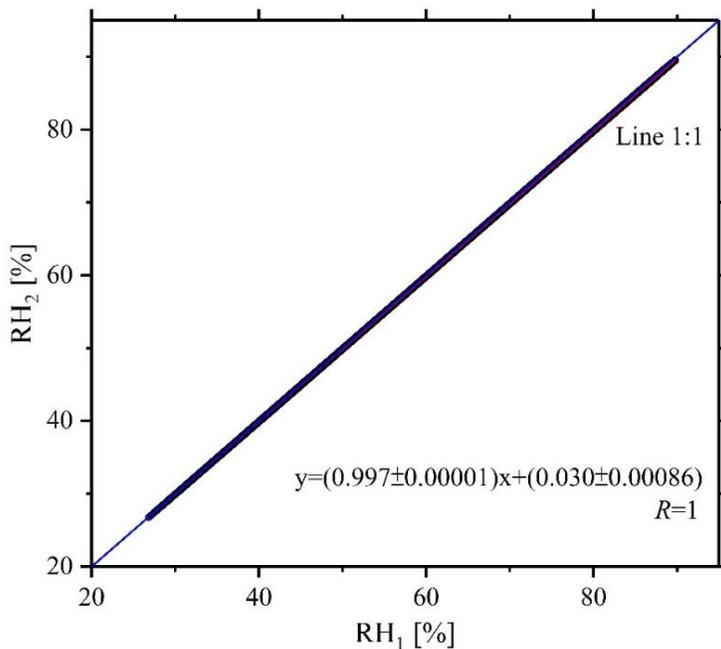
Reply: Equation 1 was from the book “Hydrology: Water quantity and quality control” by Martin P. Wanielista, Robert Kersten and Ron Ealgin in 1997. James D. Johnston et al. (2015) also referenced this equation in their paper too.

To verify the accuracy of corrected results by equation 1, we used the Magnus formula (Eq. (R1)) to calculate the RH in the optical chamber of the wet nephelometer again.

$$e_w(t) = 611.2 \times e^{\frac{17.62t}{243.12+t}} \quad (R1)$$

The parameter  $t$  is the temperature in °C. The parameter  $e_w$  is the saturation vapor pressure at  $t$ °C. The results of the calculated RH using these two methods are compared in Figure R2.

As shown in Fig. R2, the linear least square regression slope  $\pm$  standard deviation is  $0.997 \pm 0.00001$  and the intercept  $\pm$  standard deviation is  $0.030 \pm 0.00086$ . The Pearson’s correlation coefficient ( $R$ ) between the  $RH_{\text{chamber}}$  corrected by these two different methods equals 1. It illustrates that the calculated results using these two methods are highly consistent.

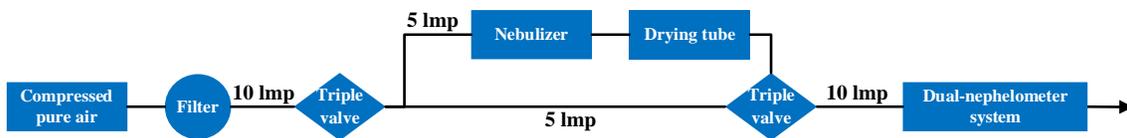


**Figure R2: The consistency of the  $RH_{\text{chamber}}$  corrected by equation 1 ( $RH_1$ ) and equation R1 ( $RH_2$ ). The solid red line represents the linear least square regression. The blue line is the line of 1:1. The linear regression function and the Pearson’s correlation coefficient ( $R$ ) are given in the bottom-right corner of the panel.**

- Line 150 and SI (supplementary information): There is something wrong in your figure S8 (which should be referenced here as well): The values of the y-axis do not make sense and they are too low. Please check. Why is the humidogram of pure ammonium sulfate already increasing up to the deliquescence RH? Was the solution contaminated? Did you use mono-disperse aerosol? Please give more technical details on how the calibration was performed. I can recommend to have a look at the paper by Fierz-Schmidhauser et al (2010) who give details on the calibration of humidified nephelometers using known salts.

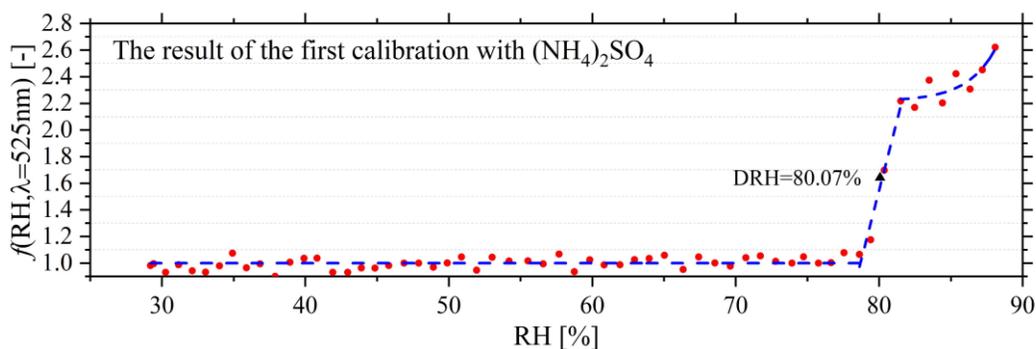
Reply: Thank you so much for noting the mistake we made in Fig. S4 (the original figure is Fig. S8), and we have replaced the plot with new one in the revised manuscript.

About the humidogram of pure ammonium sulfate increasing up to the deliquescence RH, we checked the calibration records, and found that the calibration result presented in the manuscript was wrong. Actually, we did twice calibrations of humidified nephelometers in September 21<sup>st</sup>, 2019, only the first calibration was well performed. The schematic diagram of experimental set-up for calibration of dual-nephelometer system is illustrated in Figure R3. Ammonium sulfate aerosol was generated by nebulizing the aqueous ammonium sulfate solutions with a nebulizer (model SH600) and then dried through a drying tube without size selections. Then the dried ammonium sulfate aerosol (usually RH<5%) was carried and mixed with the filtered compressed pure air at flowrate of 5 lpm into the dual-nephelometer system.



**Figure R3: the schematic diagram of experimental set-up for the calibration of high-resolution humidified nephelometer system.**

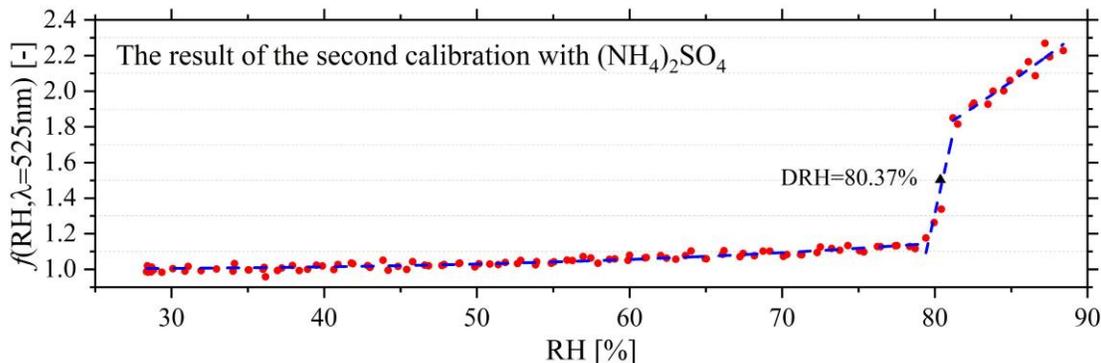
Figure R4 shows the measured humidogram of  $f(\text{RH}, 525\text{nm})$  for ammonium sulphate, where x-axis represents the RH in the optical chamber of humidified nephelometer. When RH is lower than 79.41%, the values of  $f(\text{RH}, 525\text{nm})$  are consistently remained about 1. The literature value for the deliquescence relative humidity (DRH) of ammonium sulphate is 80% at 298K (Cheung et al., 2015). Figure R4 shows that the measured phase transition occurs at RH=80.07%. It illustrates that the RH inside the nephelometer chamber is correct and that the system is functioning properly.



**Figure R4: The  $f(\text{RH})$  vs. RH of ammonium sulfate particles at  $\lambda=525\text{nm}$ . The phase transition at deliquescence occurred at about RH=80.07%.**

Figure R5 shows the result of the second calibration. However, the humidogram of pure ammonium sulfate is already increasing up to the DRH. It is probably because that we did not clean up the container completely

before reformulating the solution, the ammonium sulfate solution was contaminated.



**Figure R5: The  $f(\text{RH})$  vs. RH of ammonium sulfate particles at  $\lambda=525\text{nm}$ . The phase transition at deliquescence occurred at about  $\text{RH}=80.37\%$ .**

- Line 157: An aethalometer does not directly measure BC and it is common to use the acronym “eBC” or “EBC” for “equivalent black carbon” by convention.

Reply: Thank you for your suggestion. We have revised it.

- Equation 6 and Line 200-201: To calculate the organic mass fraction, do you now use the salts or the just the ions like sulfate, nitrate, etc? What do you mean with “among others”? As it reads now, you would count certain ions more than once.

Reply: We now just use the inorganic salts including  $(\text{NH}_4)_2\text{SO}_4$ ,  $\text{NH}_4\text{HSO}_4$  and  $\text{NH}_4\text{NO}_3$  to calculate the organic mass fraction ( $F_{\text{org}}$ ). The misleading sentence has been revised.

- Line 210: This should be more an if-statement, so “If the values of  $\gamma < 78\%$  and  $\gamma > 82\%$  are about the same, then  $\eta$  will be close to 0.”.

Reply: Revised.

- Line 406: Zeiger->Zieger

Reply: Corrected.

- Line 412: Add “partially” before “affected”

Reply: It is added now. Thanks.

- Line 448: The reference of Meier et al, 2009 is not correct here since this work is not about the particle light scattering enhancement  $f(\text{RH})$ . In my precious work, I have modelled (and observed) the effect of particle size on  $f(\text{RH})$ , see e.g. our paper in ACP from 2013: <https://acp.copernicus.org/articles/13/10609/2013/> Please also have a look at the recommendation section for humidified nephelometer measurements.

Reply: We have added the citation of this paper which is indeed very useful.

- Line 528: Is the strong correlation with or without the green points? Is R now the coefficient of

determination or the Pearson correlation coefficient? Please clarify here and throughout the manuscript (incl. the figures with regression lines) which r-value you are showing.

Reply: The correlation without the green points ( $R=0.69$ ,  $R$ : the Pearson's correlation coefficient) is stronger than that with the green points ( $R=0.62$ ). They are clarified in the manuscript.

- Line 549: What threshold or exact criteria was applied to identify those points as “cases with large systematic errors”? Please clarify.

Reply: The threshold applied to identify those points as “cases with large systematic errors” was that the light-scattering coefficient ( $\sigma_{\text{sp},525\text{nm}}$ ) was lower than  $20 \text{ Mm}^{-1}$ .

- Line 552: Ny-Alesunf -> Ny-Alesund

Reply: Revised.

- Eq 13: The second part is not defined. What is the parameter “n” doing here? It is also not used later on. I guess this is an empirical equation that you introduce here? Or is there a reference for the “steepness index”?

Reply: Yes,  $n$  is an empirical parameter introduced in Eq. (10), while  $\tau$  is the ratio of the derivatives of  $f'$  (80%) over  $f'$  (60%) to characterize the change of curvature of  $f(\text{RH})$  with RH (L. Zhang et al., 2015). From Eq. 10, we can derive the two derivatives and their ratio:

$$f' (80\%) = m \times n \times \left(\frac{4}{5}\right)^{(n-1)}$$

$$f' (60\%) = m \times n \times \left(\frac{3}{5}\right)^{(n-1)}$$

$$\tau = \frac{f' (80\%)}{f' (60\%)} = \left(\frac{4}{3}\right)^{(n-1)}$$

The parameter  $n$ , as defined in Eq. (10), dominates the magnitude of the scattering enhancement, which dictates the change in the curvature of the humidogram as described by  $\tau$ .

L. Zhang et al. (2015) firstly introduced the steepness index  $\eta$  to quantitatively describe the growth pattern in their study. Because we have already used  $\eta$  to represent the hysteresis index,  $\tau$  is used to describe the curvature of  $f(\text{RH})$  humidogram in this paper.

- Line 679: What do you mean by “The compensating effect of aerosols may be one of the main reasons for this phenomenon?” Which compensating effect?

Reply: We found that the positive correlation between  $f(\text{RH}=85\%,525\text{nm})$  and the inorganic matter mass fraction was very strong when inorganic matter mass fractions were smaller than 40%. However, when inorganic matter mass fractions were larger than 40%, the increasing tendency in  $f(\text{RH}=85\%,525\text{nm})$  as the proportion of inorganic matter increased slowly decreased. A similar phenomenon was also found for the negative correlation between organic matter and  $f(\text{RH}=85\%,525\text{nm})$ .

The compensating effect between the size and chemical composition of aerosol may be one of the main reasons for this phenomenon. Many previous studies have found that for particles with strong hygroscopicity (e.g. NaCl), if their particle number size distribution tends towards large particle sizes, their hygroscopic growth ability may be similar to that of smaller particles with weak hygroscopicity (e.g.  $\text{NH}_4\text{HSO}_4$ ; Zieger

et al., 2010, 2013; Y. Wang et al., 2017, 2018). In this study, it may be that when the proportion of inorganic matter was high, the inorganic matter of aerosol is mainly composed of relatively large particles.

- Data availability: Please use a proper data repository with a given DOI (e.g. zenodo.org or other data bases) with proper commented uploaded data. The current link leads to a website in Chinese which is difficult to read and use.

Reply: Thank you for your suggestion. We submitted the data to a data repository called the Science Data Bank with proper descriptions. The DOI of the dataset is : 10.11922/sciencedb.00785. Please notice that DOI does not work as the data has not been published yet. It will be valid automatically as soon as ScienceDB publishes the data. Before that you can view the data by the dataset private access link: <http://www.scidb.cn/en/s/pzUjMjm>.

Data description:

Sheet name	Column ID	Explanation
Sheet 1	Column 1	Year
	Column 2	Month
	Column 3	Day
	Column 4	Hour
	Column 5	Minute
	Column 6	Second
	Column 7	The corrected scattering coefficients measured by the dry nephelometer at 635nm wavelength
	Column 8	The corrected scattering coefficients measured by the dry nephelometer at 525nm wavelength
	Column 9	The corrected scattering coefficients measured by the dry nephelometer at 450nm wavelength
	Column 10	Original scattering coefficients measured by the dry nephelometer at 635nm wavelength
	Column 11	Original scattering coefficients measured by the dry nephelometer at 525nm wavelength
	Column 12	Original scattering coefficients measured by the dry nephelometer at 450nm wavelength
	Column 13	The temperature in the optical chamber of the dry nephelometer
	Column 14	The relative humidity in the optical chamber of the dry nephelometer
	Column 15	The pressure in the optical chamber of the dry nephelometer
	Column 16	The corrected scattering coefficients measured by the wet nephelometer at 635nm wavelength
	Column 17	The corrected scattering coefficients measured by the wet nephelometer at 525nm wavelength
	Column 18	The corrected scattering coefficients measured by the wet nephelometer at 450nm wavelength
	Column 19	Original scattering coefficients measured by the wet nephelometer at 635nm

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		wavelength
	Column 20	Original scattering coefficients measured by the wet nephelometer at 525nm wavelength
	Column 21	Original scattering coefficients measured by the wet nephelometer at 450nm wavelength
	Column 22	The temperature in the optical chamber of the wet nephelometer
	Column 23	The relative humidity in the optical chamber of the wet nephelometer
	Column 24	The pressure in the optical chamber of the wet nephelometer
	Column 25	The temperature measured by the sensor located at the inlet of the wet nephelometer
	Column 26	The relative humidity measured by the sensor located at the inlet of the wet nephelometer
	Column 27	The temperature measured by the sensor located at the outlet of the wet nephelometer
	Column 28	The relative humidity measured by the sensor located at the outlet of the wet nephelometer
	Column 29	The corrected relative humidity in the optical chamber of the wet nephelometer
Sheet 2	Column 1	Time information
	Column 2	Wind speed
	Column 3	Wind direction
	Column 4	Temperature
	Column 5	Relative humidity
	Column 6	The corrected scattering coefficients measured by the dry nephelometer at 525nm wavelength
	Column 7	The absorbing coefficients at 525nm wavelength retrieved from the a seven-wavelength aethalometer (AE33)
	Column 8	The single scattering albedo at 525nm
	Column 9	The scattering Ångström exponent calculated by the corrected scattering coefficients at 635nm and 525nm wavelengths
Sheet 3	Column 1	Time information
	Column 2	The mass concentration of organic matter
	Column 3	The mass concentration of nitrate ions
	Column 4	The mass concentration of sulfate ions
	Column 5	The mass concentration of ammonium ions
	Column 6	The mass concentration of chloride ions
	Column 7	The mass concentration of black carbon

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Sheet 1 is the data measured by a dual-nephelometer system.

Sheet 2 is the hourly average meteorological data and optical properties data.

Sheet 3 is the data of the chemical constitution of aerosol.

- Line 944 (caption of Fig 2): Replace “processes” with “occurrence”

Reply: Done.

- Figure 11: The font size (especially of the text within the graphs) is too small. As for all figures, you can always move less important graphs to the SI to save space.

Reply: Thank you for your suggestions. The font size within the figure 9 (the original figure is Fig. 11) has been revised as big as possible. And original figure 1, figure 4 and figure 12 were moved to the supplementary information already.

### Reference:

- Anderson, T. L. and Ogren, J. A.: Determining aerosol radiative properties using the TSI 3563 integrating nephelometer, *Aerosol Sci. Tech.*, 29, 57–69, <https://doi.org/10.1080/02786829808965551>, 1998.
- Cheung, H. H. Y., Yeung, M. C., Li, Y. J., Lee, B. P., and Chan, C. K.: Relative humidity-dependent TDMA measurements of ambient aerosols at the HKUST supersite in Hong Kong, China, *Aerosol Sci. Tech.*, 49, 643–654, <https://doi.org/10.1080/02786826.2015.1058482>, 2015.
- James, D. J., Brianna, M. M., Dennis, E., Scott, C. C., and Scott, A. B.: Comparison of Single-Point and Continuous Sampling Methods for Estimating Residential Indoor Temperature and Humidity, *J. Occup. Environ. Hyg.*, 12, 785–794, <https://doi.org/10.1080/15459624.2015.1047024>, 2015.
- Wang, Y., Zhang, F., Li, Z., Tan, H., Xu, H., Ren, J., Zhao, J., Du, W., and Sun, Y.: Enhanced hydrophobicity and volatility of submicron aerosols under severe emission control conditions in Beijing, *Atmos. Chem. Phys.*, 17, 5239–5251, <https://doi.org/10.5194/acp-17-5239-2017>, 2017.
- Wang, Y., Li, Z., Zhang, Y., Du, W., Zhang, F., Tan, H., Xu, H., Fan, T., Jin, X., Fan, X., Dong, Z., Wang, Q., and Sun, Y.: Characterization of aerosol hygroscopicity, mixing state, and CCN activity at a suburban site in the central North China Plain, *Atmos. Chem. Phys.*, 18, 11739–11752, <https://doi.org/10.5194/acp-18-11739-2018>, 2018.
- Wanielista, M., Kersten, R., and Eaglin, R.: *Hydrology: Water Quantity and Quality Control*, 2nd edition, John Wiley & Sons, New York, the United States, 1997.
- 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.
- Zieger, P., Aalto, P. P., Aaltonen, V., Äijälä, M., Backman, J., Hong, J., Komppula, M., Krejci, R., Laborde, M., Lampilahti, J., de Leeuw, G., Pfüller, A., Rosati, B., Tesche, M., Tunved, P., Väänänen, R., Petäjä, T.: Low hygroscopic scattering enhancement of boreal aerosol and the implications for a columnar optical closure study, *Atmos. Chem. Phys.*, 15(13), 7247–7267, <https://doi.org/10.5194/acp-15-7247-2015>, 2015.
- Zieger, P., Fierz-Schmidhauser, R., Gysel, M., Ström, J., Henne, S., Yttri, K. E., Baltensperger, U., and Weingartner, E.: Effects of relative humidity on aerosol light scattering in the Arctic, *Atmos. Chem. Phys.*, 10, 3875–3890, <https://doi.org/10.5194/acp-10-3875-2010>, 2010.
- Zieger, P., Fierz-Schmidhauser, R., Weingartner, E., Baltensperger, U.: Effects of relative humidity on aerosol light scattering: results from different European sites, *Atmos. Chem. Phys.*, 13, 10609–10631,

<https://doi.org/10.5194/acp-13-10609-2013>, 2013.

Zieger, P., Fierz-Schmidhauser, R., Poulain, L., Müller, T. J., Birmili, W., Spindler, G., Wiedensohler, A., Baltensperger, U., and Weingartner, E.: Influence of water uptake on the aerosol particle light-scattering coefficients of the Central European aerosol, *Tellus B.*, 66, 22716, <https://doi.org/10.3402/tellusb.v66.22716>, 2014.