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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Observations of relative humidity effects on aerosol light scattering in the Yangtze River Delta of China" *by* L. Zhang et al.

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Comments from Referee #1

1 General comments

Zhang et al. investigate in their manuscript the effect of relative humidity on the aerosol light scattering for a rural site in the Yangtze River Delta in China. The results of a one-month campaign are presented, which includes measurements of the scattering enhancement factor f(RH), the particle chemical composition and absorption properties in addition to standard meteorological parameters. The relative contribution of inorganic to organic mass fraction was found to be the main parameter determining the magnitude of the scattering enhancement. The results were further analyzed using a

trajectory analysis and estimating the effect on the direct radiative forcing.

The findings are of general interest to the scientific community since only few measurements of f(RH) exist from that region of the world. The manuscript is plausibly structured and the results are presented in an appropriate way. However, there is still room for improvement by clarifying specific comments, by removing some redundancies, and by additional editorial work (spelling and grammar mistakes). Currently, some needed instrumental and calibration details are missing in the manuscript. An optical closure study using Mie theory would help to put the measurements on a more trustworthy basis. Overall, I recommend the paper to be published in ACP after the following comments have been answered satisfactorily (major revisions).

Thanks for reviewer's suggestions. We have revised the manuscript accordingly. The following are our responses:

2 Specific comments (in arbitrary order)

1. Sect. 2.2 (Measurement system):

(1) Please state the mean and STD of the RH inside the DryNeph. There are also some inconsistencies within the text concerning the RH which has been regarded as dry (30% or 40%?), please precise.

Thanks for reviewer's suggestions. The mean and standard deviation of the RH inside the DryNeph was 12.2±3.4%, we have added it in the revised marked-up manuscript at Line 145, and also shown the RH inside DryNeph in this figure (see Fig.1 [also shown in Fig. 2 in the revised marked-up manuscript]).

RH at 40% is regarded as dry. The inconsistencies at Line140 and 219 in the revised marked-up manuscript were corrected. The potential effect of normalizing f(RH) at 40% to 1 was also discussed in the revised marked-up manuscript at Line 189-198, which will underestimate the hygroscopic growth factor a few percent.

(2) The RH-cycle of 1 hour seems quite fast. Have other (longer) scan times been

tested?

We don't agree with the reviewer's premise that a cycle time of 1-hours is fast, and don't understand what kind of test he/she is suggesting be performed. Numerous studies have used similar, or even faster, cycle times (Carrico et al., 1998; Koloutsou-Vakakis et al., 2001; Sheridan et al., 2001; Fierz-Schmidhauser et al., 2010a). For example, the RH scanned from ∼40% to ∼84% within 15 min in Carrico et al. (1998). Covert's original humidograph paper (1972) used a scan time of 4 minutes.

(3) Where exactly was the RH measured within the humidified nephelometer? Have the authors performed a calibration of the humidified nephelometer using a known hygroscopic substance (e.g. ammonium sulphate)?

The RH was measured by the RH/T sensor inside the WetNeph. However, the sensor usually overestimates RH by up to 15% RH at high RH (Fierz-Schmidhauser et al, 2010a). In that case, we installed an external RH/T sensor at the outlet of the WetNeph and used it to calibrate the RH/T sensor inside the WetNeph with lamp off. The general method was to assume the Td (dew point temperature) of RH/T sensors at the outlet of the WetNeph and inside the WetNeph to be the same (and it should be the same) and the temperature measured by the both sensors was accurate, and then using the Vaisala Humidity Calculator to retrieve the real RH inside the WetNeph. With the real RH and the measured one, the RH of the sensor inside the WetNeph was calibrated.

We have not performed the calibration using a known hygroscopic substance. However, we have calibrated the RH sensors using four well-known salts. The external RH sensors were calibrated with four saturated salt solutions (LiCl, K2CO3, NaCl and (NH4)2SO4), using the Vaisala Humidity Calibrator (HMK15) and RH/T transmitter (HMT333), which was calibrated by the National Center for Meteorological Metrology, China, in November 2012. The results were shown in Fig. 2. Good linear relationships have been achieved. Besides, the consistency of all the RH sensors have been tested, the discrepancy was <3%. Therefore, we trust the RH the RH sensors measured. We

have also done the closure of scattering coefficient and scattering enhancement factor f(RH) to make the measurement more trustworthy. This was described in detail in the following question.

(4) Since there were concurrent particle size distribution measurements at the site, I strongly recommend that the results are being compared to Mie calculations using the chemical composition and size distribution measurements. This will put the results on a more trustworthy basis (see Zieger et al. (2013) for more details).

We have done closure studies, but we did not put them in this manuscript because we are preparing to draft another manuscript focusing on the f(RH) modeling. We would like to show the closure of scattering coefficient and scattering enhancement factor as follows:

Closure studies

The particle light scattering coefficient at dry and humid conditions were calculated using Mie theory with a computer code based on the work by Bohren and Huffman (2004). We assumed the particles to be spherical and homogeneously and internally mixed. As an input, the measured particle number size distribution and the complex particle refractive index n are needed. The dry particle number size distribution was measured by TDMPS and APS (see Sect. 2.2 in the manuscript); the complex particle refractive index n was derived from the chemical mass concentration measurements of the AMS and the MAAP (see Sect. 2.2 in the manuscript). A time resolved refractive index was determined by a volume fraction averaging:

$n(\lambda)=((mfi)/i*ni(\lambda))$,

where mfi is the mass fraction, i is the density and $ni(\lambda)$ is the complex refractive index of the compound i at the wavelength of λ. The density i and the refrective index ni of each compounds were listed in Table Response1-1 (Fierz-Schmidhauser et al., 2010b). The refractive index of particles at humid conditions was calculated by a vol-

ume weighting of the dry refractive index ndry with the refractive index of water n(H2O) (see Table Response1-1):

nwet=(ndry+n(H2O)(g(RH)^3-1))/g(RH)^3 ,

where g(RH) was the hygroscopic growth factor, defined as g(RH)=Dwet(RH)/Ddry, Ddry is the dry particle diameter and Dwet(RH) is the diameter at a specific RH. The hygroscopic growth factor was determined by the chemical composition measurements measured by AMS and MAAP. The value of g for retrieved salts was taken from Topping et al. (2005), while BC and organics were assumed to be insoluble (g=1). The hygroscopic growth factor g of coarse mode particles (aerodynamic diameter >1μm) was also assumed to be 1 during the f(RH) calculation (Zieger et al., 2014). The mean g was calculated with the Zdanovskii-Stokes-Robinson (ZSR) mixing rule. The one-parameter equation (Petters and Kreidenweis, 2007) was used to determine the RH dependence of g(RH):

g(aw)=(1+κaw/(1-aw))^(1/3),

where κ was a simple measure of the particle's hygroscopicity and αw was the water activity, which was replaced by RH in our study, since the Kelvin effect is small for larger particles, which are relevant to the light scattering at the used wavelengths. For more details on calculation and uncertainty, please refer to Fierz-Schmidhauser et al. (2010b) and Zieger et al. (2013; 2014).

A good agreement of scattering coefficients measured by the DryNeph and calculated with Mie model at 550nm wavelength (dust-influenced episode excluded) was shown in Fig. 3. The standard deviation of 1-hour average was taken for the y-axis error, for y-axis, the uncertainty of the measurements due to the uncertainty of nephelometer was taken (Anderson er al., 1996). Figure 4 shows the predicted and calculated f(RH, 550nm) at various RHs. The regression and R2 shows good agreement of the instruments (humidification system, AMS, MAAP and TDMPS) in this study.

(5) Page 2861, Line 10 and Figure 1: Figure 1 does not contain a real information content and could be removed. The agreement between the two nephelometers can be described within the text (e.g. by stating the result of the linear regression). Has the small difference between the two nephelometers been accounted for when calculating f(RH)?

We think the reviewer was referring to Fig. 2 instead of Fig. 1. Following the reviewer's suggestion, we have removed this figure in the revised marked-up manuscript and described in Sect. 2.4 at Line 232 instead. Yes, the small difference between the two nephelometers have been considered when calculating f(RH).

2 Page 2855, Line 28: Can the authors shortly elaborate further on how the properties of the gas and particulate matter have "changed dramatically" since 1999 at the site?

As far as we know, there are some changes of the properties of the gas and aerosols since 1999. Firstly, according to the measurements of LinAn Regional Atmosphere background station, there is a decreasing tendency (decreased by $\sim$15%) of PM10 from 2006 to 2012 in LinAn. Secondly, from 1999 to 2013, the sulfate mass concentration decreased from 21.2$\pm$11.5 to 8.1$\pm$4.1 (mean $\pm$ standard deviation), partly because the Chinese government has put a high emphasize on SO2 control. Thirdly, the SO2 mass concentration decreased significantly from 50 to 19 μg.m-3 at LinAn from 1999 to 2012 (ZEPB, 1999; 2012). Fourthly, the pH of rainwater of ZheJiang Province decreased from $\sim$4.8 in 1999 to 4.17 in 2009, then increased slightly to $\sim$4.5 in 2012 (ZEPB, 2009; 2012). We have added some words in Line 76-77 in the revised marked-up manuscript.

3 Page 2856, Line 20: Particles could also experience reconstruction at elevated RH (see e.g. Tritscher et al, 2011) and thus f(RH,λ) could in theory also be slightly below 1. I suggest to rephrase, e.g. by stating "usually above 1".

Thanks for reviewer's suggestion. Yes, we have rewritten this sentence as "f(RH,λ) and fb(RH,λ) are always greater than 1 after water uptake (Weingartner et al., 1995)." at

Line 96 in the revised marked-up manuscript.

4 The wavelength of the MAAP is slightly different (637nm instead of 670 nm) than the manufacturer states (see Müller et al., 2011).

Thanks, we have changed it to “637nm” and cited the paper of Müller et al (2011) at Line 176 in the revised manuscript.

5 Page 2862, Line 20: One should not interpolate linearly to calculate f(RH)-values at different RH. The parameterizations discussed in Sect. 3.6.1 and 3.6.2. should be used instead. Generally, I think it would improve the reading if Sect. 3.6.1 and 3.6.2. are moved to the front, where the observations of f(RH) are discussed first.

We may not express it clearly. Based on the measured RH-f(RH) curve (humidogram), we used the Matlab command “interp1(RH, f(RH), RH0, ‘linear’)” to obtain f(RH0) at the specific relative humidity RH0, where RH and f(RH) were the RH data array and the corresponding f(RH) values, respectively, “linear” was the interpolation method we used. In fact, there are several ways of achieving f(RH0), and we found linear interpolation was the best and easiest way to get f(RH0) at a specific RH0 (e.g. 80%) for our data. Firstly, parameterization were usually used to recalculate f(RH0) (Fierz-Schmidhauser et al, 2010b; Zieger et al., 2014), however, the fitting curve of f(RH) cannot be perfectly fitted (i.e., R2 won’t be 1). Secondly, another way of getting f(RH0) was to calculate the mean value of f(RH) values at RH between RH0-ΔRH and RH0+ΔRH (Fierz-Schmidhauser et al, 2010a; Fierz-Schmidhauser et al, 2010c; Zieger et al., 2010). This will also bring some error especially at high RH since f(RH) increases quite fast at high RH. Thirdly, as to our Humidograph schedule, the RH increased 1-3% RH every minute and we can get 28 data points from the lowest RH ($\sim$40%) to the highest RH ($\sim$90%). This data density allows us to interpolate and get more accurate f(RH0). Fourthly, among all the ways of interpolation (‘nearest’, ‘spline’, ‘cubic’ and so on), linear interpolation was tested to be the best. So we choose interpolation as the method of obtaining f(RH0).

We prefer not to move Sect. 3.6.1 and Sect. 3.6.2 to the front due to the following reasons. First, the two sections were about the parameterization and we gave the fitting parameters separately for locally-polluted, northerly-polluted and dust-influenced episodes, so this section should behind Sect. 3.4 “Classification of various observation episodes”. Second, the Sect. 3.6.2 was in close relation with the following discussion of Sect. 3.6.3 Steepness of humidograms, which was a further discussion of the relationship of f(RH) and chemical compositions (Sect. 3.5), so we prefer it behind Sect. 3.5 “The relationship of scattering enhancement factor with chemical compositions”. Considering all these factors, we prefer to leave these two sections as where they were (Sect. 3.6) and separate the discussion of the steepness index η as a new section Sect. 3.6.3 (following the reviewer’s suggestion).

6 Figure 3: Why is RH=91% highlighted by a black line? Is this the maximum or set RH? In addition, add the RH inside the DryNeph to panel (a). Please add the wavelength to the graph or the caption.

The black line was intended to show clearly the maximum RH ($\sim$91%). According to the reviewer’s suggestions, we have removed the black line and added the RH inside the DryNeph to panel (a) of Fig. 2 in the revised marked-up manuscript. We have also added the wavelength in the figure caption.

7 Page 2865, Line 26: What is special about the ”3 h”? This information could probably be removed.

According to the reviewer’s suggestion, we have removed “3 h”.

8 Page 2867, Line 16: It is not clear to the reviewer what the difference between the two affected areas are (2.8 vs. 0.27 million square kilometers). Please clarify.

It was a strong dust event. The 2.8 million square kilometers were the total areas affected by the dust event, while the 0.27 million square kilometers only were the areas suffered from dust storms or strong sandstorms. We have rewritten this sentence

as “During a severe cold air outbreak, a strong dust event struck northern China on 8 and 9 March, 2013. The affected area covered about 2.8 million square kilometers, about 0.27 million square kilometers of which suffered from dust storms or strong sandstorms” at Line 412 in the revised marked-up manuscript.

9 Figure 4 and 8: The Ångström exponent has no unit (please remove ”Å” from the figures).

According to the reviewer’s suggestion, we removed ”Å” (see Fig. 5 and 6 [also shown in Fig. 3 and 7 in the revised marked-up manuscript]).

10 Table 5: Factor g from Zieger et al. (2014) is 0.59$\pm$0.08 at 550nm (see Tab. 1 in their publication).

We can not found g in “Influence of water uptake on the aerosol particle light scattering coefficients of the Central European aerosol” (Zieger et al., 2014), Tab. 1 in this paper was f(RH) values for 3 wavelengths.

11 Figure 6: The panels (b) and (c) are repetitive and seen in the first panel. I suggest to just show panel (a).

We have deleted panels (b) and (c) (see Fig. 7 [also shown in Fig. 5 in the revised marked-up manuscript]).

12 Figure 7: Please mark which pie chart belongs to which trajectory.

We have added ‘(a)(b)(c)’ to each pie chart, and added them in the caption (see Fig. 8 [also shown in Fig. 6 in the revised marked-up manuscript]).

13 Figure 9: Please check the fit method, since the slope of inorganic and organic (inorganic mass fraction = 1- organic mass fraction) are not similar (the slope of the organic mass fraction should be -1 times the slope of the inorganic mass fraction). Has an orthogonal or weighted fit been used?

The total mass concentration was calculated as the sum of mass concentrations of sul-

fate, nitrate, ammonium, chloride and organic measured by AMS and EBC measured by MAAP. The organic and inorganic mass fractions were calculated by dividing the mass concentration of organics (measured by AMS) and inorganic ions (the sum of sulfate, nitrate, ammonium and chloride measured by AMS) by the total mass concentration, respectively. Therefore, the sum of the organic fraction and inorganic fraction is not 1, thus the absolute value of the two slopes were not equal. We have used the orthogonal linear fit instead of linear least square fit and drawn a new figure (see Fig. 9 [also shown in Fig. 8 in the revised marked-up manuscript]). The error of the measurements was discussed at Line 460-465 in the revised marked-up manuscript.

14 The discussion of the steepness of the humidograms should be a separate section (following 3.6.3.). It is not really clear, if real deliquescence behaviour (so real and obvious jumps at a sudden transition from solid particle to liquid droplet) has been observed or if just the steepness increased with increasing inorganic mass fraction. Please clarify. As shown in Fig. 12b, the normalization or calculation of f(RH) using the scattering coefficient at RH=40% could increase a bias in the results, since the particles could still change their water content below 40%. As mentioned above, an optical closure study using Mie theory will help to give more confidence in the measurement results.

No deliquescence behavior was clearly observed in our study although ammonium sulfate concentration was high at sometimes. The steepness index η proposed in this study aims to provide a way of quantitatively describing the steepness of humidograms well fitted into equation f(RH)=1+aRHˆb. The steepness decreases with the increasing of nitrate to be precise. We have separated the discussion of steepness as a new Sect. 3.6.3. Please see answers to Question 1(4) for the information on the closure study.

15 The sensitivity on the direct aerosol radiative forcing is a useful exercise. However, the chosen RH of 67% as the campaign average is a bit arbitrary since the effect will be much larger at increased RH. The authors could add a figure showing the difference in radiative forcing for the entire RH range for the four cases (see e.g. Figure 8 in

Fierz-Schmidhauser et al, 2010).

The chosen RH of 67% is the mean ambient RH during the entire campaign. We used it to see the sensitivity of the direct radiative forcing of different aerosols to f(RH). Following the reviewer's suggestions, we have added a figure (see Fig. 10 [also shown in Fig. 14 in the revised marked-up manuscript]) showing the influence of RH on direct forcing for the northerly-polluted period, locally-polluted period, dust-polluted period and entire campaign.

16 The conclusions should be rewritten to really focus on the main findings. Currently, it is a repetition of sentences from the main discussion. Comparison to other findings with a literature discussion (e.g. sentence on Page 2875, Line 14- 15) should be moved to the discussion of the results.

According to the reviewer's suggestion, we have rewritten our conclusion focusing on the main findings (see Line 630-682 in the revised manuscript). The new conclusion was follows:

"The influence of aerosol water uptake on particles' light scattering properties and direct radiative forcing have been investigated at LinAn, a regional atmospheric background station of Yangtze River Delta, China, using the scattering enhancement factor measurement system, together with AMS, MAAP and TDMPS providing the chemical composition and size distribution information. The average enhancement factors and mean standard deviations at 85% RH for scattering coefficient, backscattering coefficient and hemispheric backscatter fraction (f(85%), fb(85%) and fβ(85%)) were 1.58(0.12), 1.25(0.07) and 0.79(0.04), respectively. Slight wavelength dependence of f(85%) was observed at higher f(RH) values. Generally, the highest values of f(RH) corresponded to aged aerosols with a small fraction of OM; while the lowest values corresponded to younger aerosols with a larger fraction of OM. f(RH) of aerosols with relatively low scattering coefficient was usually low with a large variation; while f(RH) of aerosols with high scattering coefficients was relatively high with a small variation.

Besides, NO3- plays an important role in determining the magnitude of f(RH) at LinAn.

Humidograms measured at LinAn can be well described by the model f(RH)=c.(1-RH)ˆ(-d) and model f(RH)=1+a.RHˆb. Further investigation shows the shape of the humidogram is closely related to the mass percentage of nitrate. A steepness index η has been defined to quantitatively determine the steepness of humidograms. The more nitrate (or less sulfate), the smaller η is and the straighter the curve will be. In March, the average relative humidity (RHamb) was 67%. Consequently, the direct radiative forcing of locally-polluted, northerly-polluted and dust-influenced aerosols increased by 11.8, 19.5 and 10.5%, respectively due to aerosol uptake water in March at LinAn. At 85% RH, the direct radiative forcing increased by as high as 47% due to the aerosol hygroscopicity. In conclusion, water plays an important role in aerosol scattering properties as well as the radiative forcing, and it should be paid high attention when comparing between remote sensing and in-situ measurements and calculating the climate forcing."

3 Technical corrections I strongly recommend a second proof-reading regarding the English grammar. The reviewer is unable to correct all the typos, missing articles and grammar mistakes.

(1)Page 2858, Line 4: Define "SD" at its first appearance.

SD is the abbreviation for standard deviation, we have defined "SD" at its first appearance at Line 26 in the revised marked-up manuscript.

(2)Page 2857, Line 27: Replace "activated" by "active".

We think it should be "activated" charcoal, please see WIKIPEDIA via http://en.wikipedia.org/wiki/Activated_carbon.

(3)Please be consistent on how to capitalize instrument names (sometimes it is Nephelometer, sometimes nephelometer).

We have changed all the "Nephelometer" to "nephelometer".

(4)As a symbol for the Ångström exponent, one usually uses α as a symbol. I suggest to replace by α.

Both symbols are used in the literature, but we prefer using "å" rather than "α " for Ångström exponent, because "α " is used to denote a mass scattering efficiency in this manuscript.

(5)Page 2859, Line 26: Add the Ångström-exponent symbol at the end of the sentence.

We have added the symbol å at the end of the sentence at Line 186 in the revised marked-up manuscript.

(6)Page 2859, Line 23: The definition of the hemispheric backscatter fraction is a repetition and can be removed.

We have deleted this sentence.

(7)Page 2863, Line 12: Verb missing.

We have changed it, Line 301 in the revised marked-up manuscript.

(8)Page 2867, Line 13: Suggest to replace "to produce hygroscopic compounds." by "leading to an increase in the particle's hygroscopicity."

Following the reviewer's suggestions, we have changed the sentence accordingly at Line 410 in the revised marked-up manuscript.

(9)Page 2868, Line 6 and 7: Please add a "the" before "dust" and "Ångström".

We have added "the" at Line 434 and 435 in the revised marked-up manuscript.

(10)Please add the wavelength to the captions in Tab. 1, 4, 5, 6, and 7 and as well to all figures were optical parameters are shown.

We have added the wavelength to related Figures and Tables (Fig. 2, 3, 7, 8, 11, 14 and Table 1, 4, 5, 6 in the revised marked-up manuscript).

(11)Figure 1: Replace "pentagram" by "star".

We have changed it to "star".

(We are sorry that there are some equations and tables cannont be properly displayed here, we have attached a pdf version of this response in the supplement, together with a revised marked-up manuscript and a revised manuscript.)

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of combustion aerosols at high relative humidity, Environ. Sci. Tech., 29, 2982-2986, 1995.

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Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C2282/2015/acpd-15-C2282-2015-supplement.zip

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 2853, 2015.

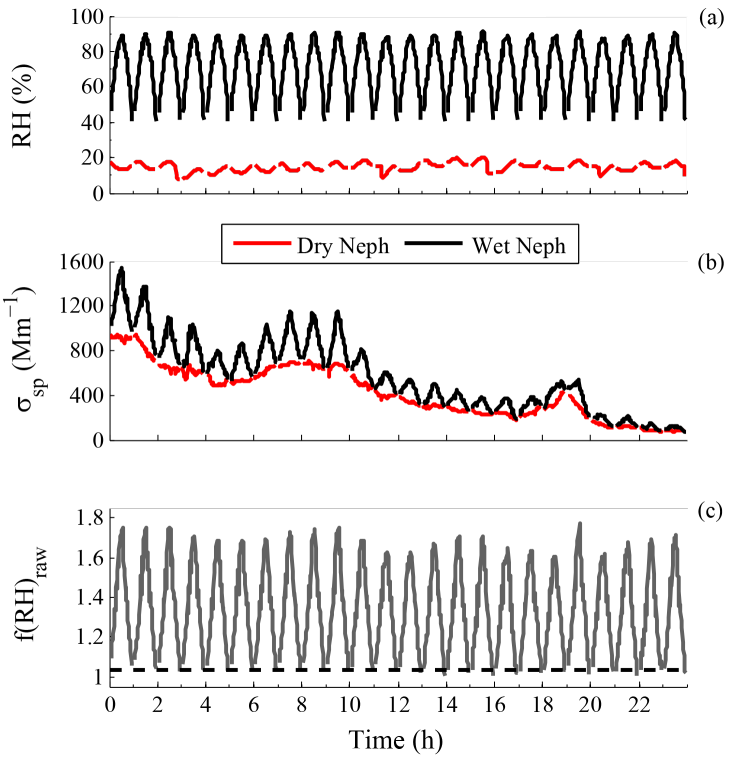

Fig. 1. Example of recorded data on 17 March 2013

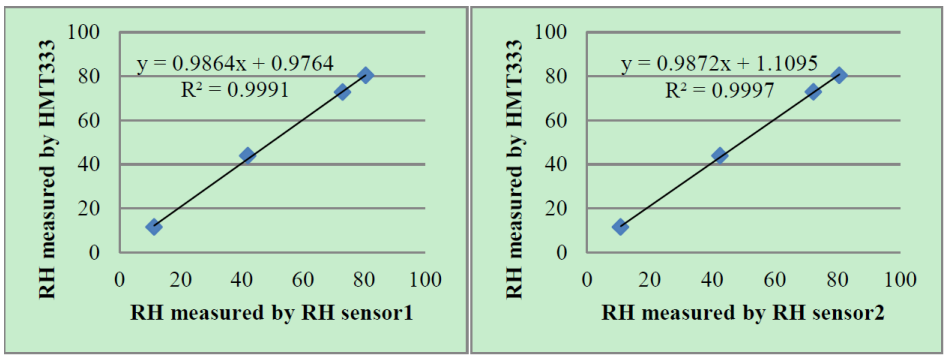

Fig. 2. The calibration of RH sensors

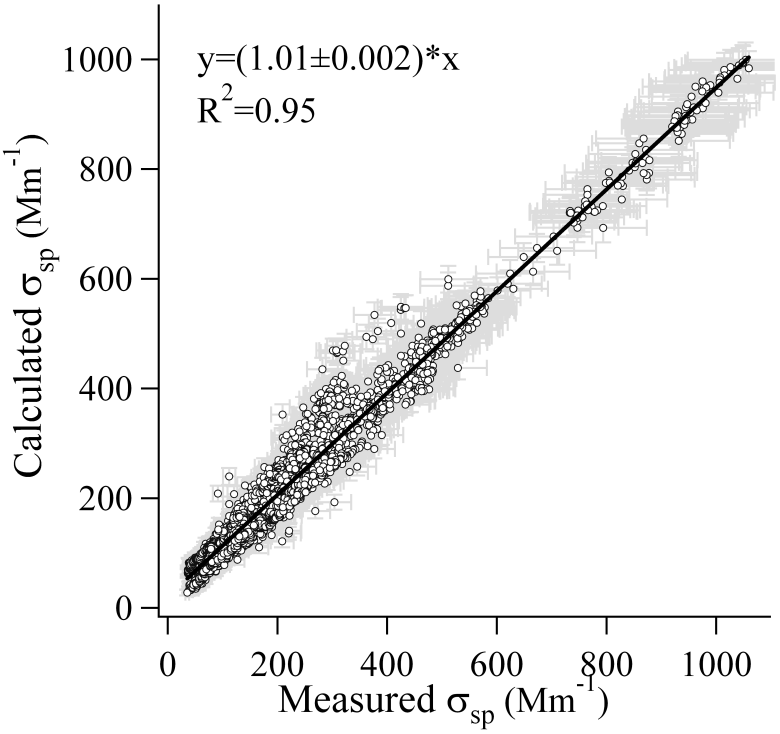

Fig. 3. Comparision of measured scattering cofficients and calculated scattering coefficients at 550nm wavelength (dust-influenced episode excluded).

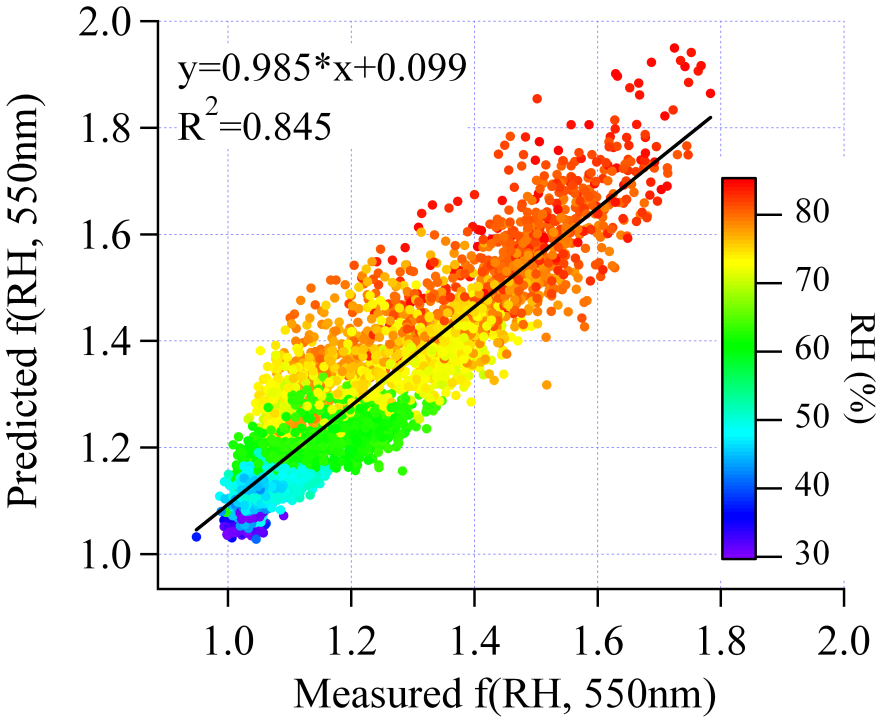

Fig. 4. Predicted and modeled f(RH, 550nm) for various RHs. The black solid line represents the linear least square regression.

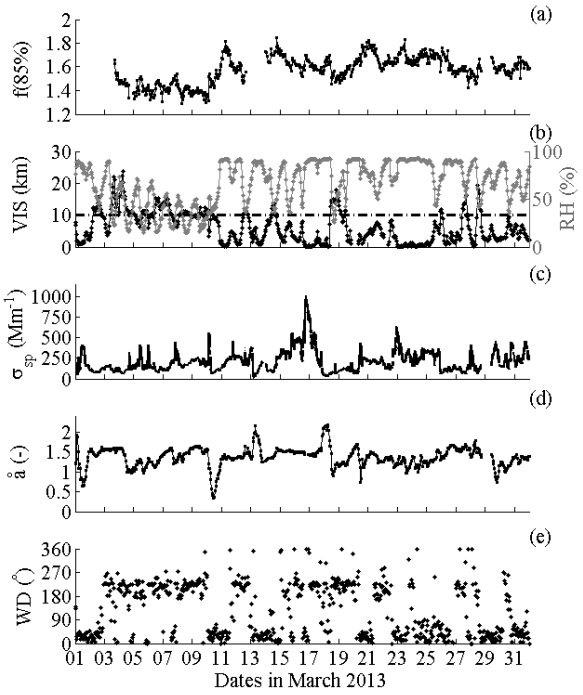

Fig. 5. Time series of measured and derived aerosol variables, as well as the ambient RH and visibility.

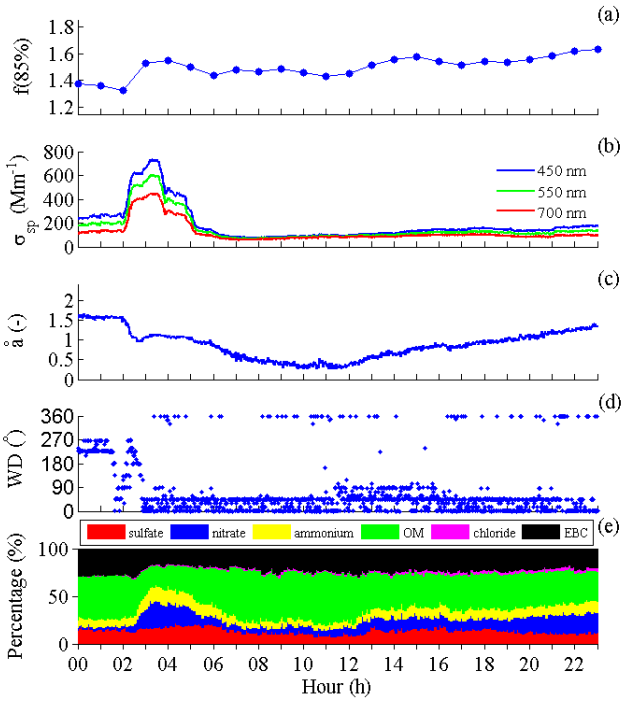

Fig. 6. Parameters in episode influenced by dust on 10 March 2013 at LinAn

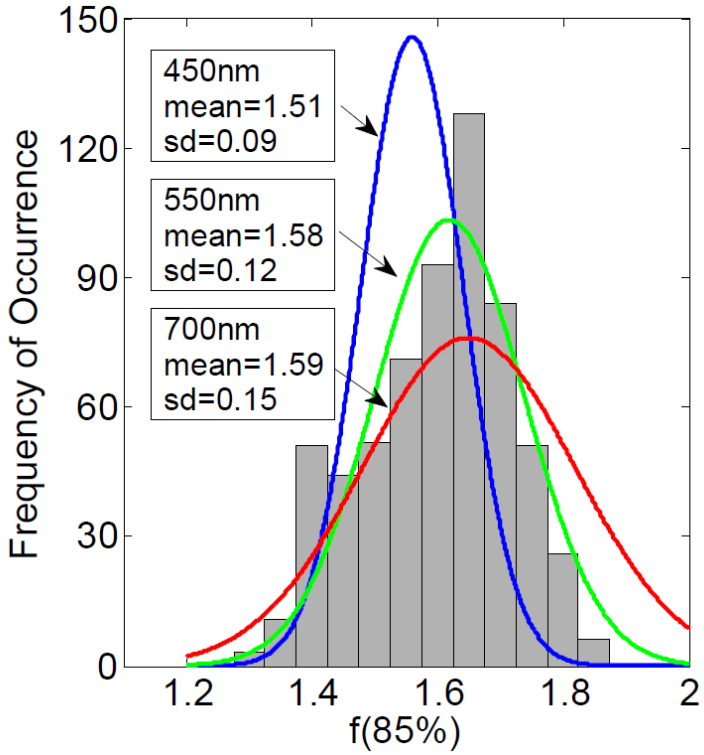

Fig. 7. Histogram of f(85%,550 nm) overlaid with the Gaussian curves based on the statistics for f(85%,450 nm), f(85%,550 nm) and f(85%,700 nm).

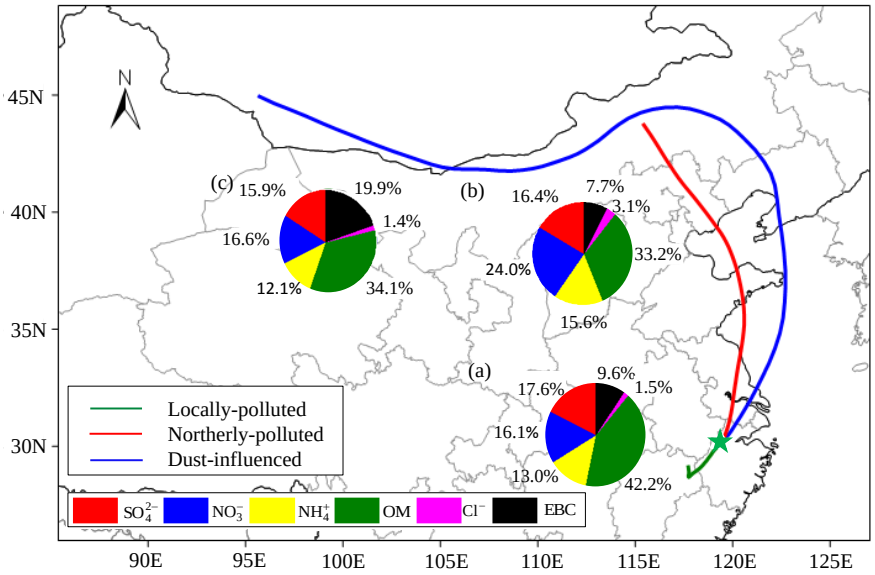

Fig. 8. 72h back trajectories of locally-polluted period, northerly-polluted period and dust-influenced period, together with the mean mass fraction of submicron chemical compositions and EBC

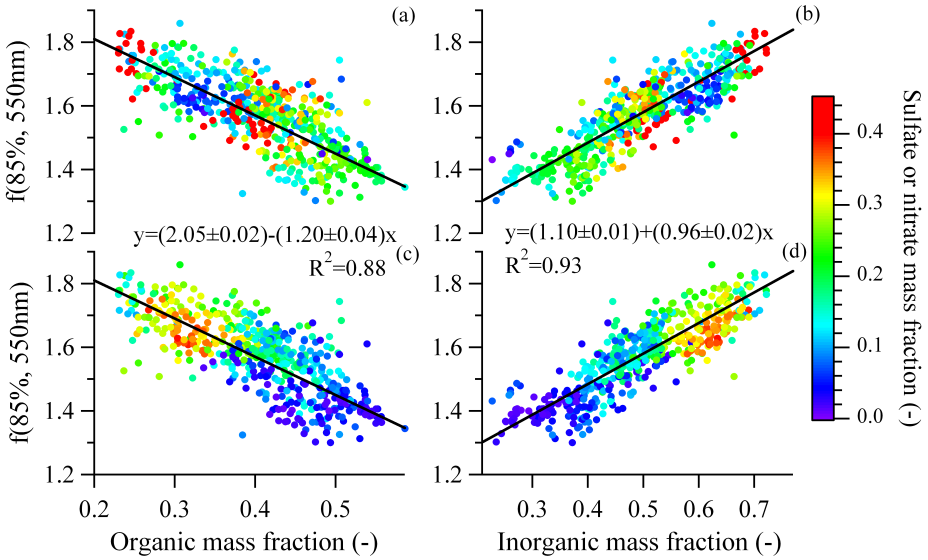

Fig. 9. Scattering enhancement factor f(85%, 550nm) vs. organic mass fraction and inorganic mass fraction determined from AMS and MAAP

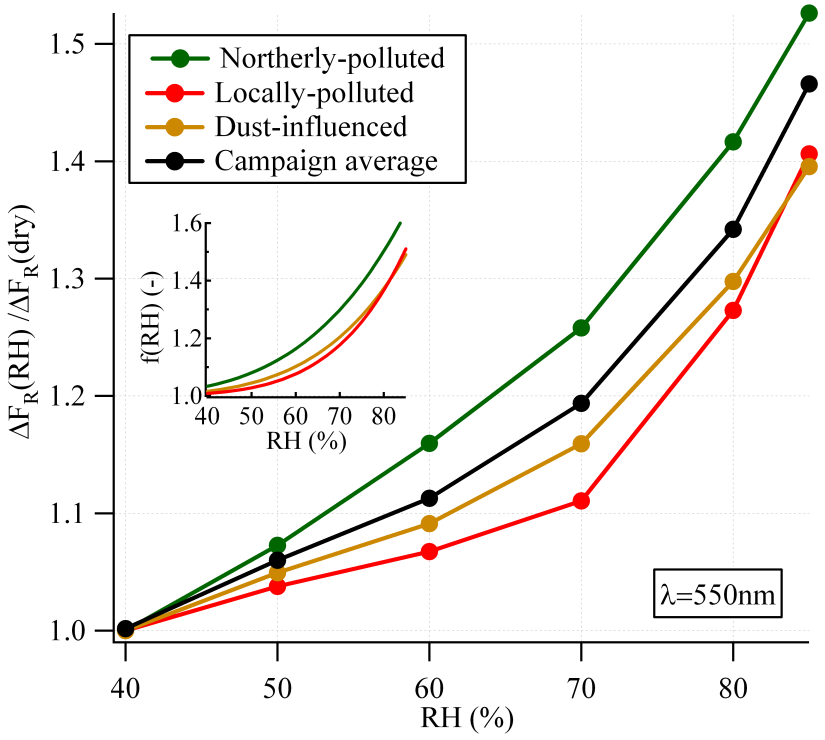

Fig. 10. Influence of relative humidity (RH) on direct radiative forcing for the entire campaign (black line), as well as for the northerly-polluted, locally-polluted and dust-polluted periods