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 T_d (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, K_2CO_3, NaCl and $(NH_4)_2SO_4$), 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:

$$n(\lambda) = \sum_i \frac{mf_i}{\rho_i} n_i(\lambda),$$

where mf_i is the mass fraction, ρ_i is the density and $n_i(\lambda)$ is the complex refractive index of the compound i at the wavelength of λ. The density ρ_i and the refrective index n_i 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 volume weighting of the dry refractive index n_{dry} with the refractive index of water n_{H_2O} (see Table Response1-1):

$$n_{wet} = \frac{n_{dry} + n_{H_2O}(g(RH)^3 - 1)}{g(RH)^3},$$

where g(RH) was the hygroscopic growth factor, defined as g(RH)=D_{wet}(RH)/D_{dry}, D_{dry} is the dry particle diameter and D_{wet}(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 >1 μm) 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):

$$g(a_w) = \left(1 + \kappa \frac{a_w}{1 - a_w}\right)^{\frac{1}{3}},$$

where κ was a simple measure of the particle's hygroscopicity and α_w 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 R^2 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 ~15%) of PM_{10} from 2006 to 2012 in LinAn. Secondly, from 1999 to 2013, the sulfate mass concentration decreased from 21.2±11.5 to 8.1±4.1 (mean ± standard deviation), partly because the Chinese government has put a high emphasize on SO_2

control. Thirdly, the SO_2 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 ~4.8 in 1999 to 4.17 in 2009, then increased slightly to ~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) 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 f_b(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), RH_0, 'linear')" to obtain f(RH_0) at the specific relative humidity RH_0, 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(RH_0), and we found linear interpolation was the best and easiest way to get f(RH_0) at a specific RH_0 (e.g. 80%) for our data. Firstly, parameterization were usually used to recalculate f(RH_0) (Fierz-Schmidhauser et al, 2010b; Zieger et al., 2014), however, the fitting curve of f(RH) cannot be perfectly fitted (i.e., R^2 won't be 1). Secondly, another way of getting f(RH_0) was to calculate the mean value of f(RH) values at RH between RH_0-ΔRH and RH_0+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 (~40%) to the highest RH (~90%). This data density allows us to interpolate and get more accurate f(RH_0). 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(RH_0).

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 (~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±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 sulfate, 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 $f(RH)=1+a\ RH^{b}$. 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.

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%), f_b(85%) and f_β(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, NO_3^- plays an important role in determining the magnitude of f(RH) at LinAn.

Humidograms measured at LinAn can be well described by the model $f(RH)=c\cdot(1-RH)^{-d}$ and model $f(RH)=1+a\cdot RH^b$. 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 (RH_{amb}) 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”.

References

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Petters, M., and Kreidenweis, S.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmospheric Chemistry and Physics, 7, 1961-1971, 2007.

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Zieger, P., Fierz-Schmidhauser, R., Poulain, L., Müller, T., 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, doi: 10.3402/tellusb.v66.22716, 2014.

Table Response1-1 Microphysical properties of selected aerosol compounds used for the model predictions. The imaginary part of the complex refractive index n was omitted for all components except for black carbon (BC). All values are in 550nm wavelength.

	$(NH_4)_2SO_4$	NH_4NO_3	OM	BC	H_2O
n	1.530	1.556	1.48	1.75+0.44i	1.333
ρ	1.77	1.72	1.4	1.7	1

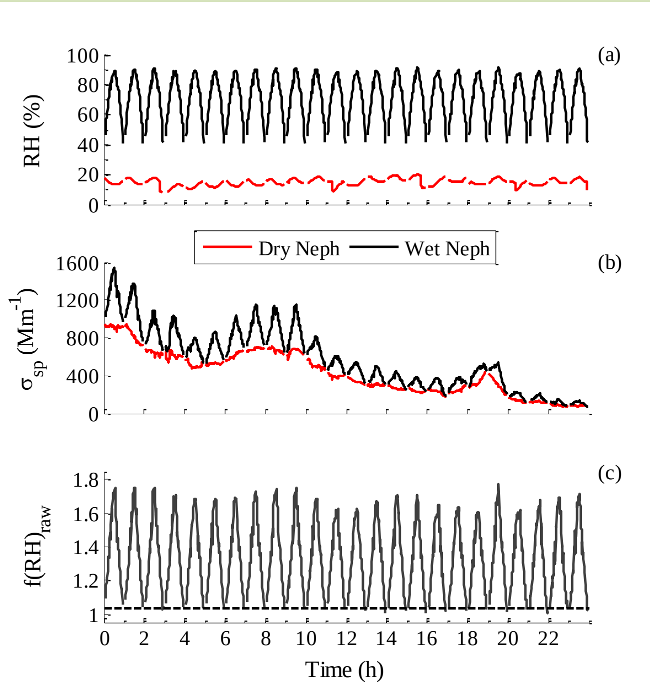

Fig. 1 Example of recorded data on 17 March 2013 (a) Relative humidity inside DryNeph (red line) and WetNeph (black line); (b) Aerosol scattering coefficients measured by DryNeph (red line) and WetNeph (black line) at 550nm wavelength; (c) Raw scattering enhancement factor f(RH, 550nm)$_{raw}$ without normalization, the black dash line was f(RH)$_{raw}$=1.03.

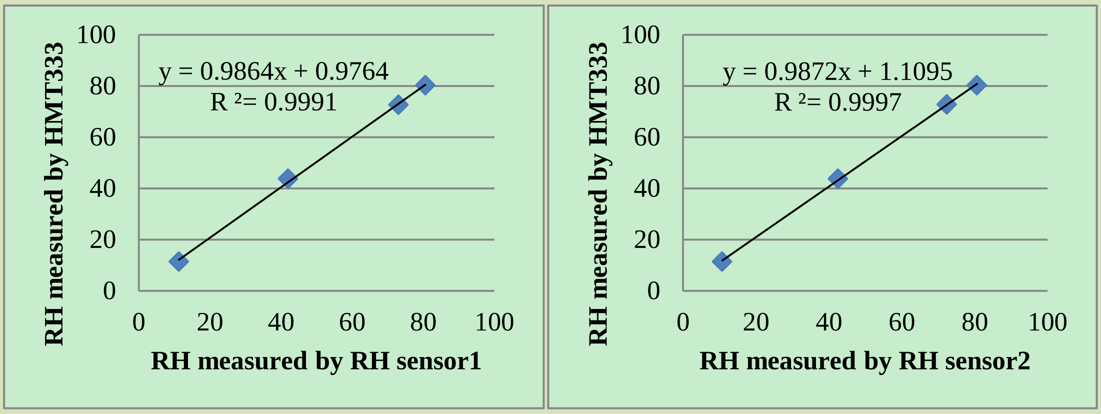

Fig. 2 The calibration of RH sensors. Sensor 1 was the external RH/T sensor (wetout) at the outlet of the WetNeph and sensor 2 was another external RH/T sensor at the inlet of the DryNeph. Sensor 1 and 2 were measured at 22.7 and 22.3 ℃, respectively.

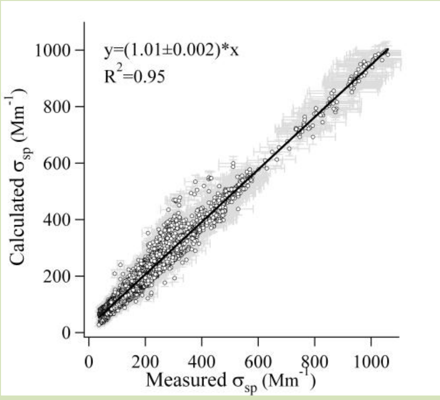

Fig. 3 Comparision of measured scattering cofficients and calculated scattering coefficients at 550nm wavelength (dust-influenced episode excluded). The error bars gave the standard deviation of the calculated σ_{sp} and the uncertainty of the measured σ_{sp}.

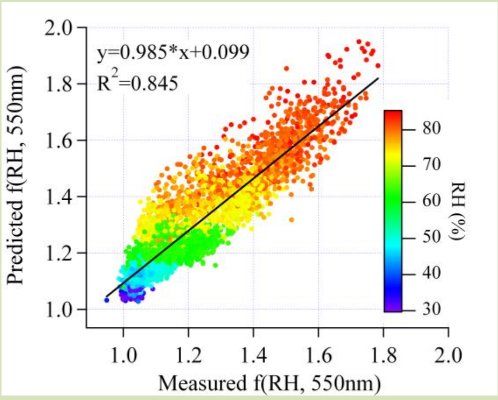

Fig. 4 Predicted and modeled f(RH, 550nm) for various RHs. The black solid line represents the linear least square regression.

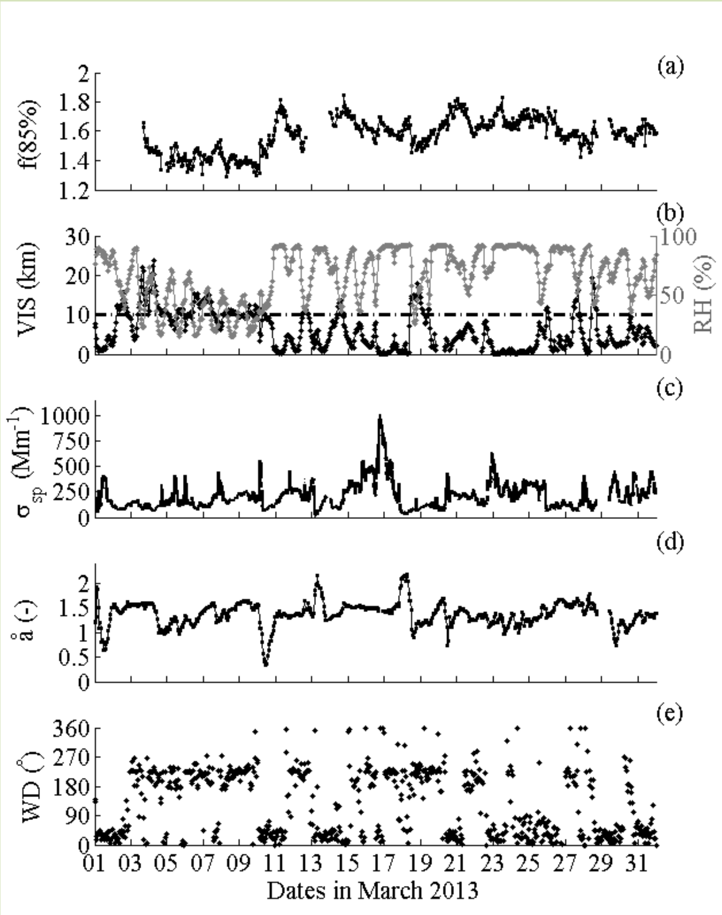

Fig. 5 Time series of measured and derived aerosol variables, as well as the ambient RH and visibility. (a) scattering enhancement factor f(85%) at 550 nm wavelength; (b) visibility (VIS) and relative humidity (RH) at ambient conditions, the dashed line represents VIS=10 km; (c) aerosol scattering coefficient of DryNeph at 550 nm wavelength; (d) Ångström exponent å; (e) wind direction (WD), indicating prevailing wind directions during observation period was mainly northeasterly (NE) and southwesterly (SW).

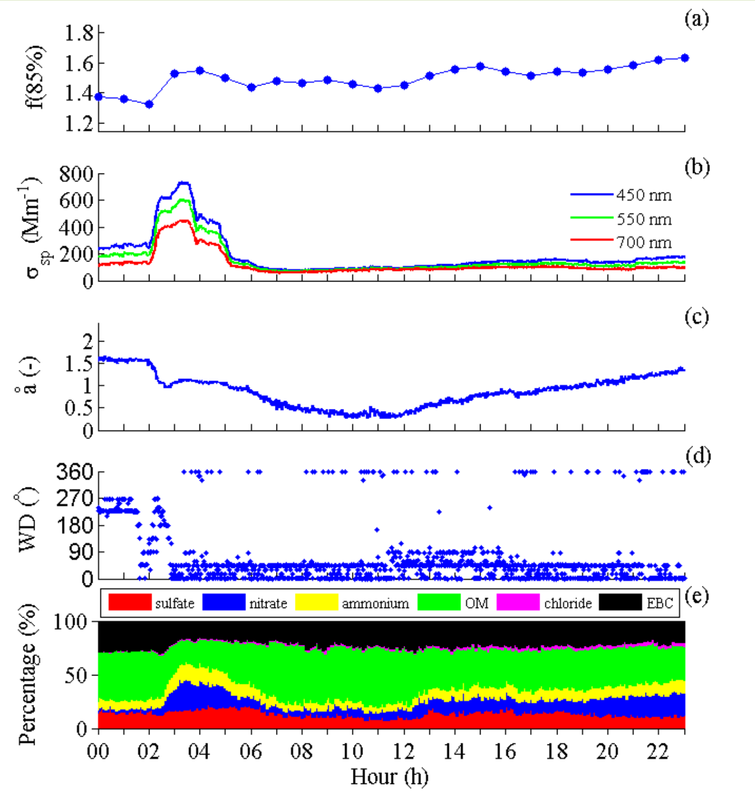

Fig. 6 Parameters in episode influenced by dust on 10 March 2013 at LinAn (a) scattering enhancement factor f(85%) at 550nm; (b) scattering coefficients at 450nm, 550nm and 700nm wavelengths; (c) Ångström exponent å; (d) wind direction; (e) mass percentages of chemical species measured by AMS and MAAP.

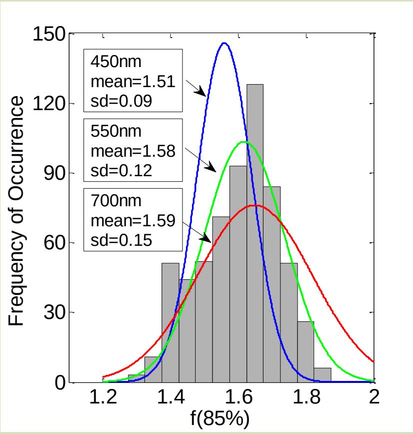

Fig. 7 Histogram of f(85%,550 nm) overlaid with the Gaussian curves based on the statistics for f(85%,450 nm), f(85%,550 nm) and f(85%,700 nm).

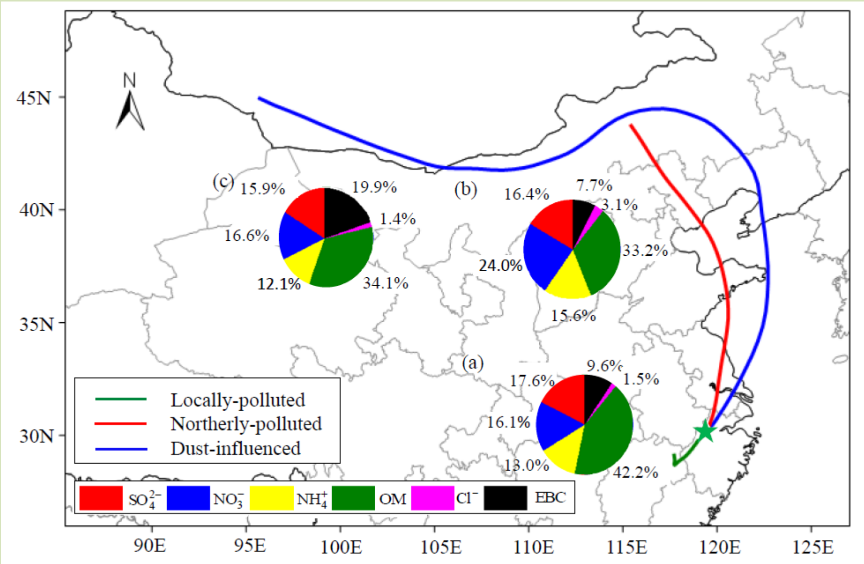

Fig. 8 72h back trajectories of locally-polluted period, northerly-polluted period and dust-influenced period, together with the mean mass fraction of submicron chemical compositions (SO_4^{2-}, NO_3^-, NH_4^+, OM and Cl^-) measured by AMS and EBC in PM_{10} measured by MAAP. The pie chart (a), (b) and (c) were for locally-polluted, northerly-polluted and dust-influenced periods, respectively.

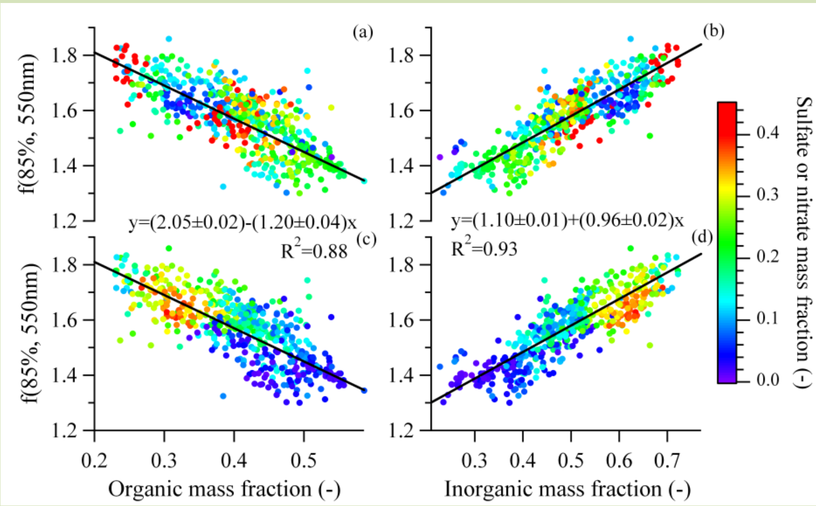

Fig. 9 Scattering enhancement factor f(85%, 550nm) vs. organic mass fraction and inorganic mass fraction determined from AMS and MAAP: (a) (b) f(85%, 550nm) vs. organic mass and inorganic mass fraction colored by sulfate mass fraction, respectively; (c) (d) f(85%, 550nm) vs. organic mass fraction and inorganic mass fraction colored by nitrate mass fraction, respectively. The solid black line represent a bivariate linear regression including the uncertainty of f(85%, 550nm) and the standard deviation of chemical compositions.

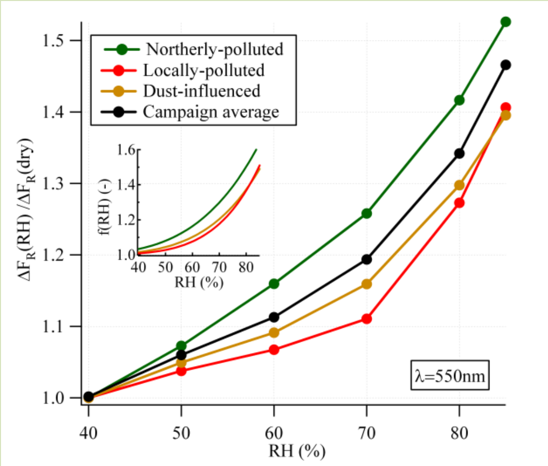

Fig. 10 Influence of relative humidity (RH) on direct radiative forcing for the entire campaign (black line), as well as for the northerly-polluted, locally-polluted and dust-polluted periods, measured by the ratio of radiative forcing at a certain RH to that at dry conditions. The small inlay shows the fitting curves of f(RH) for northerly-polluted, locally-polluted and dust-polluted periods, respectively, using fitting parameters in Table 6. All the parameters were measured at 550nm.

Comments from Referee #2

The contribution contains original measurements, taken in China.
I suggest minor revisions.

Thanks for reviewer's suggestions. We have revised the manuscript accordingly.

1. *Abstract should contain the goal of the paper, the techniques used, and the main results. The techniques applied are missing.*

Following the reviewer's suggestion, we have added the techniques applied in our study and rewritten the second sentence in the abstract as:

"To achieve a better understanding of the effect of aerosol hygroscopic growth on light scattering properties and radiative forcing, the aerosol scattering coefficients at RH varying from 40% to ~90% was measured using the humidification system (consists of a nephelometer and a humidified nephelometer) in the Yangtze River Delta of China in March 2013. In addition, the aerosol size distribution and chemical composition were measured."(see Line 21-25 in the revised marked-up manuscript).

2. *Page 2856, line 20: larger than one, (greater is frequently used in the paper, but larger is appropriate).*

We asked help from native English co-author. The suggestions are: "Greater" and "larger" are two words we should pay attention to. Usually, "greater" is used when referring to values, and "larger" (or "bigger", of course) when referring to the size of entities (whether physical or abstract). So, we revised the manuscript accordingly. For example, there are places we have misused “greater” (Line 26 of p.2871; Line 9 and 17 p.2872), we have changed them to “larger” in the revised marked-up manuscript (Line 544 and 555).

3. Page 2858, line 9: So, you humidify the aerosol from very dry conditions to RH 40% (for neph #1) and then to the elevated RH (for neph #2)? Please clarify!

The aerosols pass through an aerosol dryer, enter the DryNeph, and then WetNeph. The measured RH inside DryNeph was always below 20% RH (see Fig.1 [also shown in Fig. 2 in the revised marked-up manuscript]). There is a humidifier between DryNeph (neph #1) and WetNeph (neph #2), which controls the RH of aerosols entering the WetNeph (neph #2). The humidifier set point is stepped from ~40% to ~90% RH during the first half hour, and back to ~40% RH during the last half hour (see Fig.1 [also shown in Fig. 2 in the revised marked-up manuscript]). For example, at 03min, the RH set point was 40%, the aerosols were humidified directly from <20% to 40% RH; at 30min, the RH set point was 90%, aerosols were humidified directly from <20% to ~90% RH. Each minute correspond to a different RH set point, so that a RH cycle was obtained.

4. Page 2862, line 9: 20 to 1000 Mm-1 corresponds to 4-200 km visibility, and not to 0.1 to 23.7 km as in Fig 4b. Please clarify!

Yes, you are right if it was the ambient scattering coefficient. However, the scattering coefficients were measured were under dry conditions, while the visibility was measured under ambient conditions. There was an automatic regenerating adsorption aerosol dryer at the inlet to provide dry air for all the instruments. The scattering coefficients, number size distribution and the masses of chemical compositions were all measured under dry conditions. We have added “under dry conditions” at Line 263 in the revised marked-up manuscript to make it clear.

5. Page 2863, paragraph, line 3-16: What about the impact (changes)… when the particles are dried and when they get wet again, … Does that not also influence the determination of the enhancement factor? Could be discussed?

Transport losses, thermophoresis, coagulation, evaporation, and irreversible chemical reactions and so on can differentiate the particles from the original ones, and thus impact the f(RH). However, drying and humidifying is necessary. For one thing, measuring aerosol properties at dry conditions can make the data comparable; for another, only if the particles are dried and humidified can we get the scattering enhancement factor f(RH) since it is defined as $f(RH)=\sigma_{sp}(RH)/\ \sigma_{sp}(dry)$. In that case, we have made an effort to retain the aerosol property. For example, the transport path was made as short and straight as possible, particle-free air was diluted to the aerosol stream to reduce coagulation, and lower RH and higher heater temperature was avoided as it will result in semi volatile compounds like weak organic acids and nitrates evaporating from the aerosol.

6. Table 5 According to the old work of Haenel in the 1980ies and subsequent work, c and g is in the range from 0.4-0.9 and g from about 0.6-0.9. But you find c=1.000? and g below 0.3? Can you discuss that?

Furthermore, one can compute gamma from c and g (see Haenel, 1984), and he found *values of gamma around 0.4, but you get rather low gammas below 0.2? How can one explain that?*

Table 5 in this manuscript shows the fitting parameter (c and g) of f(RH) with equation $f(RH)=c(1-RH)^{-g}$. The parameters c and g both determine the magnitude of f(RH). The larger the g and c parameters are, the larger the f(RH) will be. The parameter g determines the curvature of humiograms (f(RH)-RH curve). The larger g is, the bigger the curvature will be. Figure 2 show how the f(RH) varies with the parameter c and g. As is shown, the influence of g on the magnitude of f(RH) is more important than that of c. The parameter c and g in this study was ~0.9 and ~0.3, respectively, since the f(RH) value (e.g. f (80%) = 1.44±0.12) was comparably lower than most of the f(RH) values obtained in Europe, USA, the Arctic and so on. The value of c and g of LinAn was similar to these obtained by Gasso (2000) during a dust event with the f(RH) value f(80%) = 1.33±0.07. Hänel (1984) found the empirical models of f(RH) for background aerosols were $f(RH)=(1-RH)^{-0.4364}$ (RH<70%) and $f(RH)=0.6130*(1-RH)^{-0.6118}$ (70%<RH<99.9%), for urban aerosols were $f(RH)=(1-RH)^{-0.2053}$ (RH<70%) and $f(RH)=0.7008*(1-RH)^{-0.7317}$ (70%<RH<99.9%). The relatively larger value of parameter g at higher RH indicates the rapid increase of f(RH) with RH increase (as shown in Fig. 3), while the humidograms in our study were different, the f(RH) values were not as large as those obtained by Hänel (1984). In conclusion, the parameter c and g are dependent on the humidogram (both its curvature and the magnitude of f(RH)), the relatively low g value obtained in our study indicates the f(RH) is lower in this study, which is determined by the aerosol chemical composition, particle size distribution and so on.

7. *Figure 1, I see a green star!*

We have changed the word “pentagram” to “star”.

8. *Figure 3, a (as very thin green line) and b in one plot may help, but separately plotted, Figure 3a makes no sense, can be skipped.*

We want to show the performance of our humidification measurement system on the RH control, which is a very important part of our experiment. In order to shown more information in this figure, we have added the RH inside the DryNeph to panel (a) and added the ratio of $\sigma_{sp,wet}/\ \sigma_{sp,dry}$ ($f(RH)_{raw}$ value) as panel (c) (see Fig.1 [also shown in Fig. 2 in the revised marked-up manuscript]). Hope this will make this figure more meaningful.

9. *Figure 4 contains the essential results. This figure must be large! Is the information on wind useful? Otherwise leave out. And all periods which are discussed in more detailed should be highlighted in the plot (grey shading or so).*

According to the reviewer’s suggestion, we have redrawn this figure. We have enlarged the size of the figure and used larger font size (see Fig. 4 [also shown in Fig. 3 in the revised marked-up manuscript]).

We have classified the whole month period into three episodes: the locally-polluted period, the northerly-polluted period and the dust-influenced period. The criteria of the classification were the wind direction, back trajectory and weather phenomenon. We have thought about highlighting the three periods, however, there are cases when the northerly-polluted period only last for several hours, and then changed to the locally-polluted period, if we highlight them all, the figure would be a little messy. So we have not highlighted the three periods in the plot.

10. Figure 6: The found histogram (FoO) in (a) has no Gaussian shape, why do you then show, in addition, Gaussian curves?

Yes, the distribution of the occurrence of f(85%) was not an exact Gaussian distribution. But as Pearson once said, ”I can only recognize the occurrence of the normal curve - the Laplacian curve of errors - as a very abnormal phenomenon. It is roughly approximated to in certain distributions; for this reason, and on account for its beautiful simplicity, we may, perhaps, use it as a first approximation, particularly in theoretical investigations”. In our study, Gaussian curves captures the main characteristics of the distribution of the occurrence of f(85%, 450nm), f(85%, 550nm) and f(85%, 700nm) with R^2 of 0.98, 0.94 and 0.92, respectively. Besides, we use the Gaussian fitting curve as a representation of the data so that we could put the three lines together to make the comparison more clearly.

References in the author’s response:

Gasso, S., Hegg, D., Covert, D., Collins, D., Noone, K., Öström, E., Schmid, B., Russell, P., Livingston, J., and Durkee, P.: Influence of humidity on the aerosol scattering coefficient and its effect on the upwelling radiance during ACE‐2, Tellus B, 52, 546-567, 2000.

Hänel, G.: Parameterization of the influence of relative humidity on optical aerosol properties, Ae rosols and Their Climatic Effects, 117-122, 1984.

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, doi:10.5194/acp-10-3875-2010, 2010.

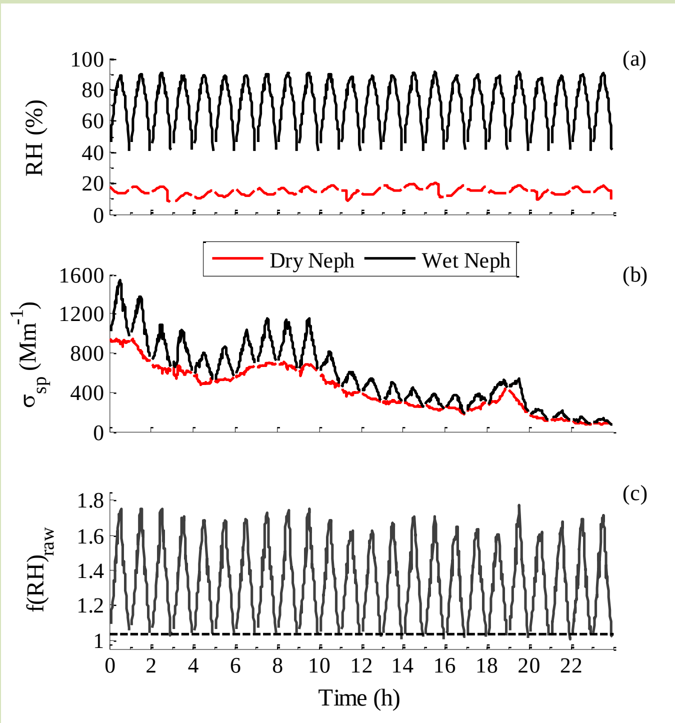

Fig. 1 Example of recorded data on 17 March 2013 (a) Relative humidity inside DryNeph (red line) and WetNeph (black line); (b) Aerosol scattering coefficients measured by DryNeph (red line) and WetNeph (black line) at 550nm wavelength; (c) Raw scattering enhancement factor $f(RH, 550nm)_{raw}$ without normalization, the black dash line was $f(RH)_{raw}$=1.03.

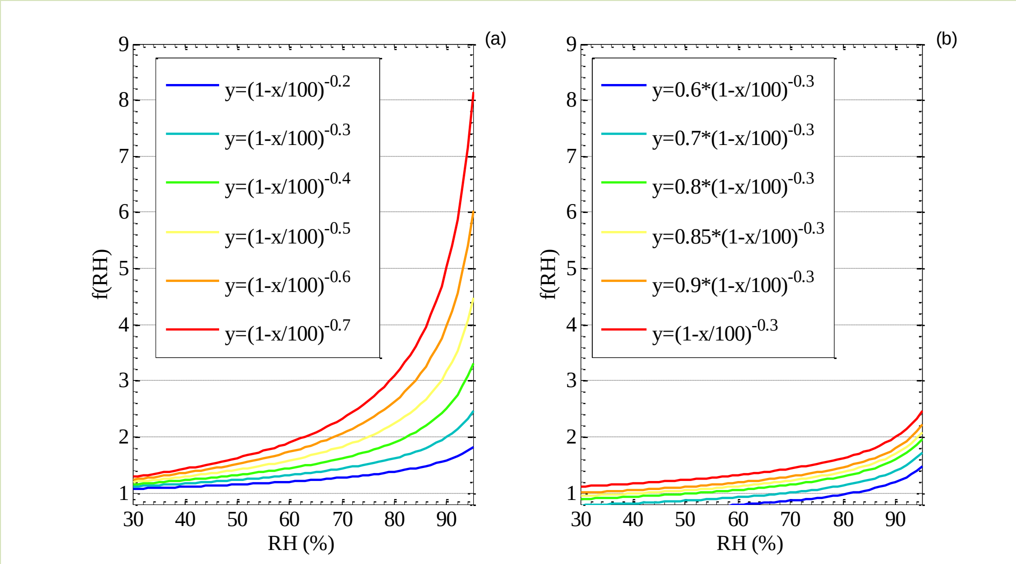

Fig. 2 Sample curves of $f(RH)=c*(1-RH)^{-g}$ with various c and g.

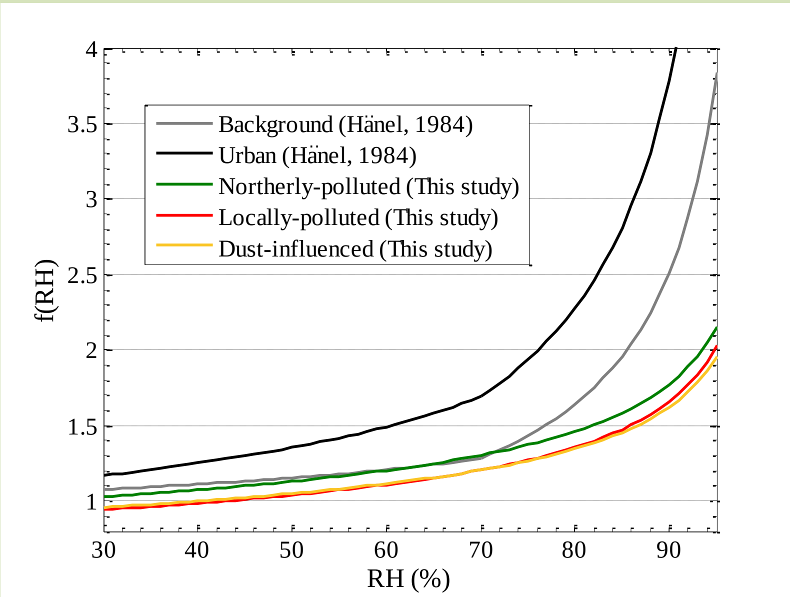

Fig. 3 Fitting curves of humidograms of H änel and this study.

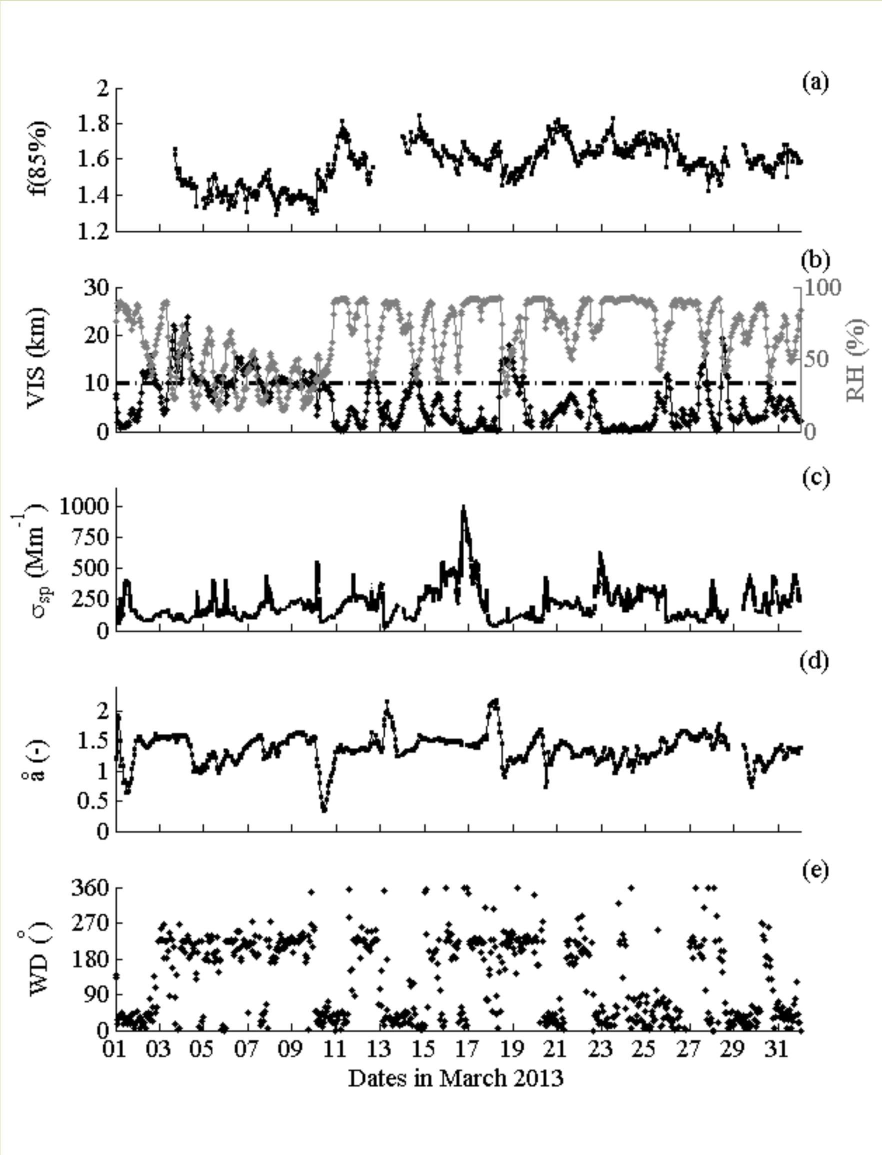

Fig. 4 Time series of measured and derived aerosol variables, as well as the ambient RH and visibility. (a) scattering enhancement factor f(85%) at 550 nm wavelength; (b) visibility (VIS) and relative humidity (RH) at ambient conditions, the dashed line represents VIS=10 km; (c) aerosol scattering coefficient of DryNeph at 550 nm wavelength; (d) Ångström exponent å; (e) wind direction (WD), indicating prevailing wind directions during observation period was mainly northeasterly (NE) and southwesterly (SW).

The marked-up manuscript:

Observations of relative humidity effects on aerosol light scattering in the Yangtze River Delta of China

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Abstract

Scattering of solar radiation by aerosol particles is highly dependent on relative humidity (RH) as hygroscopic particles take up water with increasing RH. To achieve a better understanding of the effect of aerosol hygroscopic growth on light scattering properties and radiative forcing, ~~a field campaign was carried out~~the aerosol scattering coefficients at RH in the range of 40% to ~90% was measured using the humidification system (consists of two nephelometers operating in series with a humidifier in between) in the Yangtze River Delta of China in March 2013. In addition, the aerosol size distribution and chemical composition were measured. During the observation period, the mean and standard deviation (SD) of enhancement factors at RH=85% for the scattering coefficient (f(85%)), backscattering coefficient (f_b(85%)) and hemispheric backscatter fraction (f_β(85%)) were 1.58±0.12, 1.25±0.07 and 0.79±0.04, respectively, i.e. aerosol scattering coefficient and backscattering coefficient increased by 58 and 25% as the RH increased from 40 to 85%. Meanwhile, the aerosol hemispheric backscatter fraction decreased by 21%. The relative amount of organic matter (OM) ~~and~~ or inorganics in PM_1 was found to be a main factor determining the magnitude of f(RH), the highest values of f(RH) corresponded to the

aerosols with a small fraction of OM, and vice versa. The relative amount of NO_3^- in fine particles was strongly correlated to f(85%), which suggests NO_3^- played a vital role in aerosol hygroscopic growth during this study. The mass percentage fraction of nitrate also had a close relation to the curvature of humidograms, namely, the higher the nitrate concentration is, the straighter the humidogram will be. At 85% RH, the aerosol direct radiative forcing increased by 47% compared to that in dry conditions due to the aerosol hygroscopic growth. Air masses that arrived at LinAn in March can be classified into northerly-polluted, locally-polluted and dust-influenced types, the scattering enhancement factors at 85% RH were 1.52±0.10, 1.64±0.09 and 1.48±0.05, respectively. The sensitivity of the aerosol radiative forcing to f(RH) at the measured mean ambient RH 67 % for various aerosol types was also estimated. The direct radiative forcing increased by 11.8, 19.5 and 10.5%, respectively, for locally-polluted, northerly-polluted and dust-influenced aerosols due to aerosol hygroscopic growth at LinAn in March 2013.

1 Introduction

Hygroscopic aerosols take up water as humidity increases (Engelhart et al., 2011;Pilinis et al., 1989;Hänel, 1976;Covert et al., 1972). Aerosol water matters since water can affect both the size and refractive indices of atmospheric aerosols, thereby influencing the mass concentration, size distribution, and corresponding optical properties (e.g., scattering coefficient, backscattering coefficient, single scattering albedo, and asymmetry parameter) (Cheng et al., 2008;Randles et al., 2004;Malm et al., 2003;Carrico et al., 2003). In particular, understanding the effect of relative humidity on aerosol light scattering is important to better estimate the radiative forcing and evaluate visibility impairment (Ackerman et al., 2004;Tang, 1996;Charlson et al., 1992;Covert et al., 1972). Besides, most of the ground-based aerosol measurements are conducted in dry conditions so as to have a consistency within networks. These measurements can differ significantly from the ambient ones. Thus, the determination of enhancement factors for various optical variables are of crucial importance for climate forcing calculations (Quinn et al., 1995;Pilinis et al.,

1995) and the comparison between remote sensing and ground based measurements (Zhang et al., 2012;Wang and Martin, 2007;Zieger et al., 2012).

The Yangtze River Delta, one of the most populated and fastest growing regions in China, has experienced extraordinary economic growth during the last two decades. Amounting to 2.1% of the land area of China, this region contains ~11% of the country's population and produces ~20% of China's Gross Domestic Product (GDP) in 2013 (Wang et al., 2013). Concurrent with population increase and economic growth are the increasing energy consumption and growing number of automobiles, and therefore, the Yangtze River Delta has become a significant source of gas and particulate pollutants and secondary aerosol production. A 5-week field campaign was carried out in the early winter of 1999 at LinAn, a background station in the Yangtze River Delta (Xu et al., 2002). However, since then the physical and chemical properties of gas and particulate pollutants have changed dramatically with the rapidly developing economy and fast growing population, e.g. from 1999 to 2013, the sulfate mass concentration decreased from 21.2 ±11.5 to 8.1 ±4.1 (mean ± SD) (Qi et al., 2012;Xu et al., 2008;ZEPB, 1999;ZEPB, 2013). In order to better understand the aerosol light scattering properties and their dependency on relative humidity in the Yangtze River Delta, both the scattering and backscattering coefficients under dry (RH<~~30~~40%) conditions and controlled relative humidity were measured, along with the chemical composition and particle number size distribution.

The enhancement factors discussed in this work include scattering enhancement factor $f(RH,\lambda)$, enhancement factor for backscattering coefficient $f_b(RH,\lambda)$ and enhancement factor for hemispheric backscatter fraction $f_\beta(RH,\lambda)$. The impact of relative humidity on the aerosol light scattering coefficient is defined as scattering enhancement factor $f(RH,\lambda)$:

$$f(RH,\lambda) = \sigma_{sp}(RH,\lambda)/\sigma_{sp}(dry,\lambda) \qquad (1)$$

where $\sigma_{sp}(dry,\lambda)$ and $\sigma_{sp}(RH,\lambda)$ represent scattering coefficients at wavelength λ in dry conditions and at a defined higher relative humidity, respectively.

Likewise, the impact of relative humidity on aerosol backscattering coefficient can be described as enhancement factor for backscattering coefficient $f_b(RH,\lambda)$:

$$f_b(RH,\lambda) = \sigma_{bsp}(RH,\lambda)/\sigma_{bsp}(dry,\lambda) \quad (2)$$

where $\sigma_{bsp}(dry,\lambda)$ and $\sigma_{bsp}(RH,\lambda)$ represent backscattering coefficients at wavelength λ in dry conditions and at a defined relative humidity, respectively. $f(RH,\lambda)$ and $f_b(RH,\lambda)$ are always greater than 1 if no significant restructuring is taken place after water uptake (Weingartner et al., 1995).

Hemispheric backscatter fraction (b) is closely related to the upscatter fraction ($\bar{\beta}$), the fraction of incident solar radiation scattered into space (Wiscombe and Grams, 1976). The impact of relative humidity on aerosol hemispheric backscatter fraction can be defined as enhancement factor for hemispheric backscatter fraction $f_\beta(RH,\lambda)$ (Adam et al., 2012):

$$f_\beta(RH,\lambda) = b(RH,\lambda)/b(dry,\lambda) \quad (3)$$

where $b(dry,\lambda)$ and $b(RH,\lambda)$ represent hemispheric backscatter fraction at wavelength λ in dry conditions and at the defined relative humidity. b is defined as the ratio of backscattering coefficient to scattering coefficient: $b=\sigma_{bsp}/\sigma_{sp}$. Thus, $f_\beta(RH,\lambda)$ can be rewritten as: $f_\beta(RH,\lambda)=f_b(RH,\lambda)/f(RH,\lambda)$.

The wavelength dependence of scattering enhancement factor $f(RH,\lambda)$ varies with generalized aerosol types. Kotchenruther and Hobbs (1998) and Zieger et al. (2010; 2011) found no pronounced wavelength dependence of $f(RH,\lambda)$ for biomass burning aerosols and arctic aerosols, respectively; Zieger et al. (2013) found small variations (<5%) of $f(RH,\lambda)$ at 450, 550 and 700 nm for several European sites; Kotchenruther et al. (1999) and Magi and Hobbs (2003) reported significant wavelength dependence of $f(RH,\lambda)$ for urban/industrial aerosols off the east coast of the United States. In this study, the wavelength dependence of enhancement factors was also investigated. Except when specially mentioned, all the parameters discussed in this study are based on the measurements at 550 nm wavelength only.

2 Experimental sites and instrumentation

2.1 Site description

This study was carried out during an intensive field sampling period from 1 to 31 March 2013 at LinAn Regional Atmosphere background station, which is a WMO GAW regional station (30.3° N, 119.73° E, 138 m a.s.l.) located in the center of Yangtze River Delta, China (Fang et al., 2013) (as shown in Fig. 1). It is approximately 11 km north of the city of LinAn, with a population of 1.5 million. The site is ~50 km west of Hangzhou (capital of Zhejiang Province with a population of ~8.8 million) and ~210 km southwest of Shanghai (a mega-city with a population of ~20 million). LinAn station is on the top of a small hill, in an area primarily covered by bamboo forests and paddy rice fields, and represents the background conditions of the Yangtze River Delta. North of the station is a small village with ~200 inhabitants. In addition, there is an activated charcoal factory ~1.4 km north of LinAn station that uses bamboo wood as its source material (Qi et al., 2012). During the observation period, the prevailing winds were northeasterly (NE) and southwesterly (SW) with an average wind speed of ~2.5 $m \cdot s^{-1}$ (SD 1.4 $m \cdot s^{-1}$). 72-hour back trajectories showed two contrasting air mass origins: (1) air masses from Northern China through long-distance transport and (2) air masses from southerly/southwesterly directions with a much shorter transport distance.

2.2 Measurement system and data processing

The scattering enhancement factor f(RH) is defined as the ratio of aerosol scattering coefficient at a given, elevated RH to that at a low RH (usually <~~3~~40%). Correspondingly, the ~~measurement~~ humidification system included two nephelometers operating in series with a humidifier between them. Sample air entered the first nephelometer (reference nephelometer or DryNeph) through an aerosol dryer (Shen et al., 2011;Tuch et al., 2009) to ensure the aerosol was at dry conditions (RH inside DryNeph was 12.2±3.4% (mean±SD) for the whole field campaign)~~, where the sample RH was controlled to <30%~~, then passed through the humidifier, where the sample RH was regulated to a higher RH that was ramped from ~40 to 90%, and finally entered the second nephelometer (humidified nephelometer or WetNeph) where the scattering coefficient of humidified aerosols was measured.

Aerosol total scattering (between 7 and 170 degrees) and backscattering coefficients (between 90 and 170 degrees) were measured by an ~~Integrating~~ integrating ~~Nephelometer~~ nephelometer (TSI Inc., Model 3563) at three wavelengths: blue (450 nm), green (550 nm) and red (700 nm). Data were recorded as 1-minute average and a zero check was performed automatically once per hour. The detailed information of this instrument has been described in many previous studies (Anderson and Ogren, 1998;Charlson et al., 1969;Anderson et al., 1996).

The humidifier was built by the aerosol group in Global Monitoring Division, Earth System Research Laboratory, National Ocean & Atmospheric Administration, USA (NOAA/GMD), which was described in Carrico et al. (1998). It consists of 2 concentric tubes with a heater and insulation around the outer tube. Sample air flows through the inner tube, while water circulates between the inner and outer tubes. The inner tube is made of porous extruded PTFE (polytetrafluoroethylene) membrane, whose pore size is large enough for water molecules, but too small for larger molecules such as oxygen to cross. The flux of water vapor through the membrane is controlled by regulating the electric current to the humidifier heater until the desired RH is attained. The humidity scan was a one-hour cycle; RH was ramped from ~40 to 90% during the first half hour and in the reverse direction during the last half hour.

Besides the scattering measurement, particle number size distribution and aerosol chemistry were also measured at the station. Particle number size distributions from 3 nm to 10 μm were measured by a twin differential mobility particle sizer (TDMPS) (Birmili et al., 1999) and an aerodynamic particle sizer (APS, model 3321, TSI Inc.). The mass concentrations of sulfate, nitrate, ammonium, organic matter (OM) and chloride (aerodynamic diameter less than 1 μm) were measured by an aerosol mass spectrometer (AMS, Aerodyne Inc.). The equivalent mass concentration of black carbon (EBC) was measured by a multi angle absorption photometer (MAAP, mode 5012, Thermo Scientific Inc.) at ~~670~~ 637 nm wavelength (Müller et al., 2011), the assumed mass absorption cross-section was 6.6 $m^2 \cdot g^{-1}$. Visibility was measured using a near-forward scattering sensor (FD12, Vaisala). Meteorological data were provided by the LinAn Regional Atmosphere Background Station.

All the instruments were housed in a measurement laboratory where room temperature was controlled at ~25 ℃. All data were reported in Beijing Time (BJT=UTC+8 h) and all the scattering data were referenced at T=0 ℃ and P=1013.25 hPa. Truncation error correction, proposed by Anderson and Ogren in 1998 (Anderson and Ogren, 1998), was applied to retrieve the final scattering and backscattering coefficients. ~~The hemispheric backscatter fraction (b) was derived from equation $b=\sigma_{bsp}/\sigma_{sp}$ as mentioned above.~~ The Ångström exponent å was defined as å=-log[$\sigma_{sp}(\lambda_1)/\sigma_{sp}(\lambda_2)$]/log[$\lambda_1/\lambda_2$]. It represented the wavelength dependence of light scattering assuming a power law relationship of σ_{sp} and σ_{bsp}. In this study, scattering coefficients at 450 nm and 700 nm were used to derive å. Normalization of f(RH) (Day and Malm, 2001) has been carried out to get the final f(RH) scan values, i.e. f(40%) (the lowest RH in one cycle) is set to 1 and used to normalize other f(RH) values in this cycle. It's worth mentioning that the normalization of f(RH) (see Sect. 2.2) may underestimate f(RH) to some extent, since some organics (e.g. humic acid sodium) take up water even when RH <40% (Sjogren et al., 2007;Dick et al., 2000). To evaluate its impact, we calculated the ~~unnormalized~~ raw f(40%) value without the normalization ~~the raw data~~. The average and standard deviation were 1.03 and 0.03 with a maximum of 1.08, which means this normalization may cause an underestimate of 5% (an error of 3% was caused by the inconsistency~~t~~ of DryNeph and WetNeph, see Sect. 2.4) at most. Figure 2c shows the un-normalized f(RH) value, the lowest value of each cycle was around 1.03, considering the inconsistency of DryNeph and WetNeph, f(RH) is close to unity at the lowest RH (~40%).

2.3 Inlet system

An automatic regenerating adsorption aerosol dryer (Tuch et al., 2009) was used to provide low RH sample air to DryNeph, TDMPS, APS, AMS and MAAP to ensure comparability of measurements. The aerosol dryer was housed in a separate shelter which was located on the rooftop (~ 5 m a.g.l.) of the measurement laboratory. Aerosols entered the shelter through a commercially available PM_{10} impactor (PM_{10} inlet, URG Corporation). Then these particles went through the adsorption aerosol dryer (Tuch et al., 2009) to ensure the RH less than 30%. The dried aerosols passed through a splitter via 3/4" stainless steel tubes, and then reached different instruments.

The total sample flow through this dryer inlet was kept at 16.7 lpm to ensure a 50% collection efficiency at 10 μm aerodynamic diameter (Berner et al., 1979). Since a lot of instruments share the total flow, the sample flow for the ~~Nephelometer~~ nephelometer is 9 lpm.

2.4 Quality control on scattering measurements

Accurate performance of nephelometers and RH sensors is crucial to retrieve reliable enhancement factors (f(RH,λ), f_b(RH,λ) and f_β(RH,λ)), since they are defined as the ratio of aerosol scattering coefficient/ backscattering coefficient/ hemispheric backscatter fraction at a higher RH to those at a low RH (usually <~~3~~40%). In addition, the RH control in the WetNeph sensing volume is also critical to f(RH) measurement. Therefore, several comparisons and calibrations have been carried out before and during the experiment. Three external RH sensors (Vaisala, model HMP60) were calibrated in the RH range of 11% to 80% using Vaisala Humidity Calibrator (HMK15) with four saturated salt solutions (LiCl, K_2CO_3, NaCl, $(NH_4)_2SO_4$), and humidity/temperature transmitter (Vaisala, model HMT333), which was calibrated by the National Center for Meteorological Metrology, China. Two internal ~~Nephelometer~~ nephelometer RH sensors were calibrated to the external RH sensors with an uncertainty of ≤2%. ~~Good~~ A good agreement of these RH sensors ~~were~~ was achieved with the discrepancy <3%. Both nephelometers were calibrated with CO_2 (purity 99.999%) and filtered air. Filtered air measurements were made automatically every hour to track the instrument stability. Comparison of scattering and backscattering coefficients of the two nephelometers under low RH (~~<30%~~9.6 ±3.2%) was performed during 1 to 3 March, 2013 (as shown in Fig. 2). The total scattering coefficient and backscattering coefficient measured by WetNeph were~~, on average, ~~~ constantly 3% (y=1.03x+1.60, R^2=1.000) and 4% (y=1.04x+0.09, R^2=0.997) higher than those obtained by DryNeph at 550 nm (similarly for other wavelengths), the high consistency ~~which~~ demonstrates that the two nephelometers were operating quite steadily and the scattering/backscattering coefficients measured by DryNeph can be corrected in order to make them comparable to the measurements of WetNeph~~within their normal accuracy range~~. The uncertainty of nephelometer measurements was ~10%

(Anderson et al, 1996), combining the uncertainty of the measurements of the internal RH sensor, the uncertainty of f(85%) was ~20% at large, which may decrease for less hygroscopic particles or smaller RHs.

The RH at the outlet of WetNeph was regulated via a feedback system between the Vaisala RH signal, a PID controller and a heater. The humidifier set point was stepped from low to high RH and back to low RH every hour with the set point changing every one or two minutes. Figure ~~3~~ 2 is an example of our data showing the relative humidity control and corresponding scattering measurements. As can be seen from Fig. ~~3~~2, good relative humidity control was achieved no matter whether the scattering/backscattering coefficient was high or low.

The nephelometers were operated at a constant flow of 20 lpm, comprised of 9 lpm sample air and 11 lpm particle-free air (dilution flow). The total flowrate through the nephelometer was controlled by a mass flow controller. The dilution flow was regulated by a needle valve and measured by a mass flowmeter. The sample and dilution flow have been calibrated with a Gilibrator bubble flowmeter before the experiment. Filtered air tests have also been conducted to make sure that all the instruments were in good condition and that there were no leaks in the system.

3 Results and discussion

3.1 Overview

Figure 3 shows the time series of the measured and derived aerosol variables in March 2013, as well as the ambient RH and visibility. The scattering enhancement factor f(85%) ranged from 1.29 to 1.86 (as shown in Fig. ~~4a~~3a) with an average of 1.58 (Table 1) for the whole campaign. During 4-9 March, f(85%) stayed at a low value of 1.42 (±0.05) when LinAn was dominated by air masses from the south under clear sky. In March, the hourly averaged aerosol scattering coefficient under dry conditions (shown in Fig. ~~4c~~3c) varied from 21 to 1067 Mm^{-1} and the maximum occurred on 16 March, when a severe haze occurred. The mean value and standard deviation of the hourly averaged aerosol scattering coefficient was 223 Mm^{-1} (140 Mm^{-1}). Visibility (Fig. ~~4b~~3b) varied from 0.1 km to 23.7 km with a mean value of 6.2

km. It was quite low on 23 and 24 March because the station was in cloud. From 15 to 16 March, visibility declined to 4.4 km with the accumulation of pollutants in the atmosphere, which was a severe haze episode (as mentioned above). An air mass from Northwest China with high dust levels arrived at LinAn on 10 March, with an abrupt increase of the aerosol scattering coefficient (Fig. ~~4c~~3c) and a sharp decline of Ångström exponent (Fig. ~~4d~~3d).

Based on nephelometer measurements, the enhancement factors for scattering coefficient f(RH), backscattering coefficient f_b(RH) and hemispheric backscatter fraction f_β(RH) were determined by Eq. (1), (2) and (3), respectively. As can be seen from Table 1, their values at different RHs (50, 60, 70, 80 and 85%) were obtained using linear interpolation from the half-hourly humidogram data. The enhancement factors f(RH) and f_b(RH) increased as the RH increased, but f_b(RH) increased much more slowly than f(RH). The f(85%) and f_b(85%) were 1.58 and 1.25, respectively, suggesting that the scattering coefficient and backscattering coefficient at 85% RH were 58 and 25% higher than those in dry conditions due to aerosol water uptake. The f_β(RH) decreased with increasing RH, i.e. hemispheric backscatter fraction becomes smaller with the increase of RH and the fraction of radiation that would be backscattered into space was reduced. The f_β(RH) decreased ~21% as the RH increased from 40 to 85%. All these parameters are of crucial importance in evaluating the aerosol radiative forcing.

Generally, the scattering enhancement factor (f(80%)=1.44) is much lower than the result (f(80%)=1.7-2.1) obtained by Xu et al. (2002) for LinAn in 1999. This value is also lower than the results obtained by Carrico during ACE-1 (Carrico et al., 1998) and ACE-Asia (Carrico et al., 2003), the values obtained by Zieger et al. (2013) in several European sites and the Arctic, as well as the values achieved by Malm and Day (Malm et al., 2005;Malm et al., 2003;Malm and Day, 2001;Day and Malm, 2001;Malm and Day, 2000) in America. However, the difference between measured f(RH) in this study and previous studies performed in China (Yan et al., 2009;Pan et al., 2009;Liu et al., 2009;Delene and Ogren, 2002;Cheng et al., 2008) are much smaller. The enhancement factor for backscattering coefficient and hemispheric backscatter fraction (f_b(85%) and f_β(85%)) is 1.25(0.07) and 0.79(0.04), respectively, similar to the results (f_b(82%)=1.22±0.06 and f_β(82%) =0.83) obtained by Carrico at Sagres, Portugal during ACE-2 (Carrico et al., 2000) and the results (f_b(82%)=1.27 and f_β(82%) =0.75) obtained by Carrico et al. (2003) during the dust-dominant period in ACE-Asia.

3.2 Aerosol chemical properties

The submicron mass concentration of sulfate, nitrate, ammonium, chloride and organic matter (OM) measured by AMS and EBC in PM_{10} measured by MAAP are summarized in Table 2. The mass concentration of OM is the largest, while the mass concentration of chloride is the smallest, in accord with previous studies in LinAn (Meng et al., 2012;Yan et al., 2005). The mean mass concentration of nitrate and sulfate were 9.8±12.1 $\mu g \cdot m^{-3}$ and 8.1±4.1 $\mu g \cdot m^{-3}$ in this study, similar to the values (9.4±7.1 $\mu g \cdot m^{-3}$ for nitrate and 8.6±3.7 $\mu g \cdot m^{-3}$ for sulfate in $PM_{2.5}$) at LinAn in 2010 summer (Meng et al., 2012).

Aerosol acidity is a key parameter affecting aerosol hygroscopic growth. It is usually examined by comparing the NH_4^+ mass concentration measured by AMS and the amount needed to fully neutralize sulfate, nitrate and chloride ions ($NH_4^+{}_{predicted}$) (Sun et al., 2010):

$$NH_4^+{}_{predicted} = 18 \times (2 * SO_4^{2-}/96 + NO_3^-/62 + Cl^-/35.5) \quad (4)$$

Figure ~~5~~ 4 illustrates the relationship of measured NH_4^+ and predicted NH_4^+. As shown in Fig. ~~5~~4, the regression slope is close to 1, which implies that there was sufficient NH_3 in the atmosphere to neutralize H_2SO_4, HNO_3 and HCl, and that the PM_1 aerosol at LinAn was bulk neutralized during the measurement period. Therefore, the dominant chemical form of sulfate aerosol is ammonium sulfate (AS) rather than acidic sulfate (H_2SO_4 or NH_4HSO_4) and the nitrate existed in the form of NH_4NO_3 (AN). By calculating Pearson's correlation coefficient among 5 different chemical compositions, it could be found that NH_4^+ and NO_3^- are strongly correlated with r=0.93; NH_4^+ and SO_4^{2-}, Cl^- are highly related with r equal to 0.77 and 0.74 respectively, which also implies the main form of inorganics would be NH_4NO_3,

$(NH_4)_2SO_4$ and NH_4Cl. However, because the average mass concentration of chloride was very low (see Table 2) at LinAn, it suggests that NH_4NO_3 and $(NH_4)_2SO_4$ are the dominant water-soluble ionic species, which are consistent with previous results at LinAn based on filter chemical measurements (Meng et al., 2012).

3.3 Wavelength dependence of the scattering enhancement factor f(85%)

The wavelength dependence of scattering enhancement factor is needed to estimate the aerosol radiative forcing since solar radiation at Earth's surface depends on wavelength. The histogram~~s~~ for f(85%,550 nm)~~, f(85%,550 nm)-f(85%,450 nm) and f(85%,550 nm)-f(85%,700 nm) are~~ is shown in Fig. ~~6~~5. Overlaid on the histogram for f(85%,550 nm) (Fig. ~~6a~~5) are Gaussian curves based on the statistics for f(85%) at each wavelength. No apparent shift of mean f(85%) is seen for the 550 nm and 700 nm wavelength pair (see Fig. ~~6a a~~5~~nd Fig. 6c~~); while the mean f(85%,450 nm) is ~6% lower than that at 550 nm with a smaller standard deviation (see Fig. ~~6a a~~5~~nd Fig. 6b~~). For higher values (90th and 70th percentile values in Table 3), slightly wavelength dependence of f(RH) can be observed, i.e. the f(RH) increases with the increase of wavelength. However, the differences are mostly under 10% ~~(see Fig. 6b and Fig. 6c)~~ and therefore the discussion is focused on 550 nm wavelength in this study. Similar results were obtained by Zieger at a regional continental research site at Melpitz, Germany (Zieger et al., 2014).

3.4 Classification of various observation episodes

Based on wind direction, back trajectory analysis and weather phenomenon, observation periods can be classified into three main sectors: a northerly-polluted period (influenced by long-distance transport from northern China), a locally-polluted period, and a dust-influenced episode. Air mass back trajectories over 72 hours at 300m a.g.l. arrival height were calculated using the Trajectory Statistics (TrajStat) model (Wang et al., 2009) with 6-hourly archived meteorological data provided by the US National Centers for Environmental Prediction (NCEP). The characteristics of these three periods are as follows:

1. Periods when the wind direction is between 120 and 270° are chosen as "locally-polluted periods". During this period, pollutants mostly came from Anhui

province, Jiangxi province and the southern region of Zhejiang province as well as LinAn (green line in Fig. ~~7~~6). Economy in these areas is mainly made up of manufacturing, tourism and agriculture.

2. Periods when the wind direction is greater than 270 or less than 120 ° are described as "northerly-polluted periods". Back trajectories indicate that most of the air masses came from northern China and passed over heavily polluted areas such as the Beijing-Tianjin-Tangshan economic region and the Yangtze River Delta during long-distance transport (red line in Fig. ~~7~~6).
3. A heavy dust event occurred at LinAn on 10 March (approximately from 02:00 BJT) according to satellite information (https://earthdata.nasa.gov/labs/worldview/) and ~~3 h~~ meteorology information (provided by China Meteorological Administration, CMA). The 72 h back trajectory shows the air masses tracked from Mongolia and passed over Inner Mongolia (blue line in Fig. ~~7~~6).

3.4.1 Locally-polluted period

In the periods of 4-9, 15-20 and 26-30 March, 2013 aerosols were mainly from locally mixed pollutants from Zhejiang and/or nearby provinces. The mean f(80%) and f(85%) were 1.36 and 1.52 (as shown in Table 4), ~10 and 8% lower than those in northerly-polluted periods.

The enhancement factor for scattering coefficient and backscattering coefficients at 80% during locally-polluted period is 1.36 and 1.15, respectively, similar to the values (f(82.5%)=1.4-1.5) and (f_β(82.5%)=1.1-1.2) obtained by Koloutsou-Vakakis et al. (2001) at a northern hemisphere, continental site (Bondville, Illinois, US). The measured dry scattering coefficient is 217 Mm^{-1}, ~15% lower than that of the northerly-polluted period (251 Mm^{-1}). The averaged mass percentage of sulfate, nitrate, ammonium, OM, chloride and EBC are 17.6, 16.1, 13.0, 42.2, 1.5 and 9.6%, respectively (shown in Fig. ~~7a~~6a). Compared to the northerly-polluted period, the mass percentage of OM is ~27% higher during the locally-polluted period, while the mass percentage of nitrate is ~33% lower. Although the OM/(OM+SO_4^{2-}) ratios during locally-polluted period (~0.70) and northerly-polluted period (~0.67) are similar, the OM/(OM+NO_3^-+SO_4^{2-}) ratio during locally-polluted period (~0.56) is 24% higher than that during northerly-polluted period (~0.45), which may partly explain the lower f(RH) during locally-polluted episode (as discussed later in Sect. 3.5).

3.4.2 Northerly-polluted period

The air masses reaching LinAn during the period March 1-3, 11-15, 20-26 and 30-31 (dust episode excluded) mainly came from northern China through long-distance transport. The mean f(80%) and f(85%) were 1.50 and 1.64, respectively (as shown in Table 4).

The value (f(80%)=1.50) is similar to the previous results (f(80%)=1.48) obtained by Yan et al. (2009) for periods under the downwind of urban plume from Beijing, (f(80%)=1.46±0.10) reported by Carrico et al. (2000) for anthropogenic aerosols in Europe during the 2nd Aerosol Characterization Experiment (ACE-2) campaign, and (f(80%)=1.55-1.59) indicated by Pan et al. (2009) for a rural site (Xin'an) near Beijing city during pollution periods. However, the measured f(80%) was much lower than (f(80%)=2.0-2.43) during a pollution episode reported by Kim et al. (2006) at the Gosan regional background site, 720 km northeast of LinAn and results (f(82%)=2.24±0.20) obtained by Carrico et al. (2003) in ACE-Asia for polluted air masses measured over the ocean. The f(RH) of continental air masses transported over the ocean was higher than that over the continent, and the possible mechanisms for that increase might include coagulation with sea-salt particles and the oxidation of SO_2 and VOCs (volatile organic compounds) ~~to produce hygroscopic compounds~~leading to an increase in the particle's hygroscopicity.

3.4.3 Dust-influenced episode

During a severe cold air outbreak, a strong dust event struck northern China ~~experienced a strong dust event~~ 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 ~~the~~ dust storms or strong sandstorms~~ area covered about 0.27 million square kilometers, and~~. This event was ~~is~~ considered to be the ~~widest~~ largest and strongest dust event to hit China in 2013. During this process, suspended dust appeared in most of northwestern China, northern China, north and west Huanghuai region and west Liaoning province, the west-central Inner Mongolia, west Gansu, northern Shanxi, and several parts of Xinjiang experienced a sandstorm. Along with the extreme dust event, there was a dramatic increase in PM_{10}, for example, the PM_{10} in Yulin, Shanxi even reached 10,000 $\mu g \cdot m^{-3}$ (Wang et al., 2013;Zhang and Sun, 2013).

At 2 a.m. on March 10, wind direction changed abruptly to northerly (see Fig. 8d). The scattering coefficient increased abruptly from ~200 Mm^{-1} to > 600 Mm^{-1} (see Fig. ~~8b~~7b). PM_{10} mass concentrations at LinAn increased rapidly from 100 $\mu g \cdot m^{-3}$ to 637 $\mu g \cdot m^{-3}$, while the $PM_{2.5}$ mass concentration was only 190 $\mu g \cdot m^{-3}$, accounting for 30% of PM_{10}. The Ångström exponent decreased from 1.2 to 0.8 (see Fig. ~~8c~~7c). All these phenomena implied the arrival of cold front from northern China enriched in coarse mode particles. The mass percentage of nitrate increased significantly and reached its peak (~26%) at 3 a.m.; meanwhile, the mass percentage of OM decreased sharply from 2 a.m. to 3 a.m. (see Fig. ~~8e~~7e). Correspondingly, the scattering enhancement factor f(85%) reached 1.52 at 3 a.m. (see Fig. ~~8a~~7a), an increase of ~16% compared with that before the dust arrival. The most dust-dominated period, from 7 a.m. to 1 p.m., when the Ångström exponent was below 0.5 (Fig. ~~8c~~7c) and scattering coefficients at 450 nm, 550 nm and 700 nm (Fig. ~~8b~~7b) were almost the same, the scattering enhancement factor f(85%) was ~1.46. This value is much higher than the results (f(80%)=1.20) reported by Pan et al. (2009) in rural Beijing, (f(82.5%)=1.18) obtained by Carrico et al. (2003) in East Asia (ACE-Asia) during a dust episode, (f(80%)=1.20) reported by Fierz-Schmidhauser et al. (2010) at a high alpine site (Jungfraujoch, 3580m a.s.l.) in Switzerland during a strong Saharan dust event, and (f(80%)=1.0-1.1) measured by Li-Jones et al. (1998) in South America during an investigation of long-range transported Saharan dust. Meanwhile it is much lower than the value (f(85%)=1.73-2.20) obtained by Kim et al. (2006) in Gosan (South Korea) during a dust-dominated period. According to Tobo et al. (2010), Ca-rich particles can react with gaseous HNO_3 to form $Ca(NO_3)_2$, thus the liquid cloud-nucleating ability would be enhanced. Similar results have also reported that aerosol hygroscopicity would be largely enhanced if coarse mode Ca-rich particles combined with nitrate (Shi et al., 2008;Sullivan et al., 2009). Thus, it is speculated that the relatively high f(RH) may have resulted from the reactions of coarse mode particles with inorganics (very likely to be nitrate) during long-range transport.

3.5 The relationship of scattering enhancement factor with chemical compositions

Scattering enhancement factor f(85%) versus organic mass fraction and inorganic mass fraction were shown in Fig. ~~9~~8. The total mass concentration was calculated as the sum of mass concentrations of sulfate, 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 inorganics (the sum of sulfate, nitrate, ammonium and chloride measured by AMS) by the total mass concentration, respectively. The bivariate linear regression was applied with the uncertainty of f(85%, 550nm) which was discussed in Sect. 2.4 and the standard deviation of chemical compositions.~~As~~ The bivariate linear regressions ~~shown in~~ (Fig. ~~9~~8) show clearly anti-correlation of f(85%, 550nm) to organics fraction and strongly positive correlation of f(85%, 550nm) to inorganics fraction~~, organics were clearly anti-correlated to f(85%), while inorganics showed strongly positive correlation with f(85%) (R^2=0.86)~~. This implies that chemical composition plays a vital role in aerosol hygroscopic properties. The absolute values of both slopes (~~1.1~~1.2 for f(85%) vs. organic mass fraction and 0.96 for f(85%) vs. inorganic mass fraction) were much lower than those (3.1 and 2.2, respectively) measured at Melpitz, Germany (Zieger et al., 2014). This may partly account to the higher organic (or lower inorganic) contents at LinAn. Comparing Fig. ~~9~~ 8 (a)(b) and (c)(d), a more clearly trend of increasing nitrate with increasing f(85%) was observed. The role nitrate plays in aerosol hygroscopic properties will be discussed in the following paragraph.

f(RH) in Fig. ~~10~~ 9 was expressed in terms of γ so as to be applied to a broader RH range (Doherty et al., 2005;Quinn et al., 2005): $\gamma = \ln f(RH)/\ln((100-RH_{ref})/(100-RH))$. Here γ was based on RH_{ref}=40% and RH=85%. The relative amount of OM and inorganics can be expressed as $Fo=Cc/(Cc+C_i)$, where Cc and C_i are the mass concentrations of OM and inorganics, respectively. Figure ~~10~~ 9 shows γ versus Fo where C_i was the mass concentrations of SO_4^{2-}, NO_3^- and NO_3^-+SO_4^{2-} in Fig. ~~10a~~9a, Fig. ~~10b~~ 9b and Fig. ~~10c~~9c, respectively. For all the three scatter plots, there is a trend of decreasing γ with increasing Fo. However, unlike the results of Quinn et al. (2005), Malm et al. (2005), Pan et al. (2009) and Yan et al. (2009), γ and Fo (OM/(OM+SO_4^{2-})) (shown in Fig. ~~10a~~9a) were uncorrelated (R^2=0.14), while γ and Fo (OM/(OM+NO_3^-)) (shown in Fig. ~~10b~~9b) and γ and Fo (OM/(OM+SO_4^{2-}+NO_3^-)) (shown in Fig. ~~10c~~9c) were more strongly correlated (R^2 of 0.56 and 0.68, respectively). This result implies that NO_3^- (rather than SO_4^{2-}) plays an important role in aerosol hygroscopic growth during this study.. This increasingly importance of nitrate corresponds to many recent studies in Shanghai (a mega city in Yangtze River Delta) (Shi et al., 2014) and Beijing (Sun et al., 2012). This may partly result from increasing availability of NH_3 to form NH_4NO_3 (Morgan et al., 2010) due to the decrease of SO_2. Chinese government has put an emphasis on the control of SO_2 emission in recent years. The desulfurization technology were installed at coal-fired power units as well as certain steel and cement production facilities, as a result, the annual average concentration of SO_2 decreased significantly from 56 to 19 μg·m^{-3} at LinAn from 2006 to 2012 (ZEPB, 2012; 2006).

The molar ratio of particulate SO_4^{2-} to total sulfur (SO_4^{2-}+gas phase SO_2) was used as an indicator of the relative age of aerosols (Quinn et al., 2005). For relatively younger aerosols, there is insufficient time for the conversion of SO_2 to SO_4^{2-} via gas and aqueous phase oxidation process and therefore the SO_4^{2-}/(SO_4^{2-}+SO_2) molar ratio is low. As aerosol ages, more SO_2 is converted to SO_4^{2-} and thus the ratio increases. To illustrate the effects of this ratio and scattering coefficient on γ, Fig. ~~11~~ 10 shows γ versus Fo=OM/(OM+SO_4^{2-}+NO_3^-) colored by the SO_4^{2-}/(SO_4^{2-}+SO_2) molar ratio (Fig. ~~11a~~10a) and $\log_{10}(\sigma_{sp})$ (Fig. ~~11b~~10b). The highest values of γ (or f(RH)) corresponded to more aged aerosols with a low OM content ; while the lowest values corresponded to younger aerosols with a higher OM content, consistent with the result of Quinn et al. (2005). For aerosols with relatively low scattering coefficient, the value of f(RH) was usually low with a large variation (dots with cooler colors in Fig. ~~11b~~10b); while aerosols with high scattering coefficients, the value of f(RH) was relatively high with a small variation(dots with warm colors in Fig. ~~11b~~10b).

3.6 Parameterization of scattering enhancement factor f(RH)

Scattering enhancement factor f(RH) can be parameterized using empirical equations (Kotchenruther and Hobbs, 1998; Kotchenruther et al., 1999; Gassó et al., 2000; Carrico et al., 2003; Liu et al., 2008; Pan et al., 2009; Zieger et al., 2010; Zieger et al., 2014). Humidograms of LinAn were fitted into two empirical equations and the fitting results were shown below.

3.6.1 Parameterization with equation $f(RH)=c\,(1-RH)^{-g}$

Kasten (1969) proposed an empirical equation $f(RH)=c\,(1-RH)^{-g}$ to describe how f(RH) varies with RH, which has been used in previous reports e.g. by Kotchenruther and Hobbs (1998), Gassó et al. (2000), Carrico et al. (2003) and Zieger et al. (2010, 2014). Table 5 shows the fitting results of this work and other previous studies. The larger "c" and "g", the larger f(RH). In this work, "g" was much lower than that in most of the other studies, while was similar to the result of Gassó et al. (2000) during dust episode. It results from the comparably low scattering enhancement factor (e.g. f(80%)=1.44±0.12) of LinAn, which was similar to the value (f(80%)=1.33±0.07) obtained by Gassó et al. (2000) for a dust event. The f(RH) in other studies was much higher than that at LinAn, ranging from 2.04 (polluted marine aerosols in Gassó et al. (2000)) to 3.77 (arctic aerosols in Zieger et al. (2010)), therefore their parameter "g" was much higher.

3.6.2 Parameterization with equation $f(RH)=1+a\,RH^{b}$

The f(RH) obtained at LinAn station can be well described by the following equation, which was proposed by Kotchenruther and Hobbs (1998):

$$f(RH)=1+a\,RH^{b} \quad (5)$$

where "a" is positive and "b" is greater than 1. This function is convex, and has been used in many previous studies (Pan et al., 2009;Delene and Ogren, 2002;Carrico et al., 2003;Kotchenruther et al., 1999;Kotchenruther and Hobbs, 1998) to describe monotonic growth. Theoretically, parameter "a" determines the largest value f(100%) can reach, and parameter "b" dominates the curvature of the function. The smaller "b" is, the smaller the curvature of humidogram will be; if "b" equals to 1, then

$f(RH)=1+a\cdot RH$. The parameters "a" and "b" from our study and previous results for different aerosol types are listed in Table 6. Taking the locally-polluted episode as an example, although parameter "a" is slightly larger (~3%) than in northerly-polluted episode, parameter "b" is ~40% ~~greater~~larger, as a result the f(85%) during locally-polluted period is smaller. Parameter "b" is the largest in the locally-polluted episode, and smallest in the northerly-polluted period, i.e. the curvature of RH-f(RH) line is largest during the locally-polluted episode, then is the dust episode, and northerly-polluted period is the smallest, which is consistent with the mass percentages of nitrate (see Fig. ~~7~~6) (will be discussed later).

3.6.3 Steepness of humidograms

Among all the humidograms measured at LinAn, f(RH) increases continuously and monotonically. However, the curvatures of ~~the~~ different humidograms are different (Fig. ~~12~~11): some increase with a nearly constant rate and the humidogram line is almost straight, while some increase slowly at first and then increase more steeply at relatively higher RH, thus the curvature of the humidogram is ~~greater~~larger. In order to describe the growth pattern quantitatively, a steepness index η is defined based on the fitting curve:

$$\eta=f'(80\%)/f'(60\%)-1=(4/3)^{b-1}-1 \qquad (6)$$

where f'(60%) and f'(80%) represent the derivatives of the fitting curve at 60% and 80% RH, respectively. η is a nonnegative number. Zieger et al. (2010) has defined an index describing the magnitude of deliquescence transitions based on fitting equation $f(RH)=(1-RH)^{-g}$ (see Sect. 3.6.1), while the steepness index η proposed in this study provided a way of quantitatively describing the steepness of humidograms well fitted into equation $f(RH)=1+a\ RH^{b}$. The ~~bigger~~ larger η is, the ~~greater~~ bigger the curvature. As is shown in Fig. ~~12a~~11a, for a large η, the f'(60%) is very small, meaning that aerosol scattering coefficient barely increases (f(RH)≈1) under low RH (usually <70%). Once reaching larger RH (~70%), f(RH) begins to increase. However, for a small η (Fig. ~~12b~~11b), the difference of the derivatives at 60% and 80% RH was small, meaning the curvature of humidogram is much smaller.

A scatter plot of η and the mass percentage of nitrate is shown in Fig. ~~13~~12, colored by the mass percentage of sulfate. As can be seen, η is negatively correlated with the mass percentage of nitrate. When the mass percentage of nitrate is below ~18%, the more nitrate, the smaller η is, which means the humidogram line becomes straighter and the difference of the derivatives at lower and higher RHs becomes smaller. For a mass percentage of nitrate higher than 18% (correspondingly, a lower sulfate mass percentage), η is ~1.1, meaning the humidogram line is almost straight (as shown in Fig. ~~12b~~11b) and aerosol scattering coefficient experiences a continuous and smooth growth at almost the same rate with RH.

3.7 Sensitivity of the direct radiative forcing of different aerosols to f(RH)

Direct radiative forcing of aerosols is quite sensitive to changes of relative humidity. The impact of relative humidity on globally-averaged, direct radiative forcing can be obtained by the following expression (Chylek and Wong, 1995):

$$\Delta F_R(RH) = -[S_0/4][T_a^2(1-A_c)][2(1-R_s)^2\bar{\beta}(RH)M\alpha_s f(RH) - 4R_s M\alpha_a] \quad (7)$$

where S_0 is the solar constant, T_a is the transmittance of the atmosphere above the aerosol layer, A_c is the fractional cloud amount, R_s is the albedo of the underlying surface, $\bar{\beta}(RH)$ is the solar radiation scattered back to space at defined RH, f(RH) is the scattering enhancement factor, M is the column burden of aerosol (in $g \cdot m^{-2}$), α_s is the mass scattering efficiency, and α_a is the mass absorption efficiency.

In order to estimate the sensitivity of the forcing to ~~the observed values of f(RH)~~different RHs for various aerosol types (locally-polluted, northerly-polluted and dust-influenced aerosols), the ratio of direct aerosol radiative forcing ΔF_R at a defined RH to that at dry condition was calculated:

$$\frac{\Delta F_R(RH)}{\Delta F_R(dry)} = \frac{(1-R_s)^2\bar{\beta}(RH)\alpha_s f(RH) - 2R_s\alpha_\alpha}{(1-R_s)^2\bar{\beta}(dry)\alpha_s f(dry) - 2R_s\alpha_\alpha} \quad (8)$$

Parameters used in Eq. (8) were R_s=0.15, and α_a=0.3 $m^2 \cdot g^{-1}$ (Wang et al., 2012;Hand and Malm, 2007). The mass scattering efficiency α_s is 2.76 $m^2 \cdot g^{-1}$, which is derived from the slope of a linear regression of the measured scattering coefficients and the calculated PM_{10} mass concentrations based on TDMPS and APS measurement (see Fig. ~~14~~13); the high mass scattering efficiency is explained by the high ratio of PM_1 to PM_{10} mass at this site (average 0.81). The average upscatter fraction $\bar{\beta}$ was calculated as $\bar{\beta}$=0.0817+1.8495b-2.9682b^2 (Delene and Ogren, 2002). The sensitivity of direct radiative forcing to RH for various aerosol types were shown in Fig. 14. As is shown in the figure, the variation of ΔF_R(RH)/ΔF_R(dry) with RH was in accordance with the variation of humidograms. The f(RH) was the largest during the northerly-polluted period, correspondingly, the effects of RH on aerosol radiative forcing during this period was the largest. The same was true for the locally-polluted period and the dust-influenced period. Since b decreases with increasing RH, this correspondence also demonstrated the vital role f(RH) played in direct forcing enhancement. At 85% RH, the average ratio was 1.47, i.e. the direct radiative forcing increased by 47% owing to the aerosol hygroscopicity.

Table 7 shows the mean influence of aerosol hygroscopicity on direct radiative forcing in March at LinAn. The ratios ΔF_R(RH$_{amb}$)/ΔF_R(dry) for locally-polluted, northerly-polluted and dust-influenced aerosols ~~are~~were calculated ~~presented in Table 7, where ΔF_R(RH$_{amb}$) was calculated at~~ using the ambient average ~~realtive humidity~~RH (RH$_{amb}$=67%) in March at LinAn. The variables f(RH$_{amb}$), b(RH$_{amb}$), $\bar{\beta}$(RH$_{amb}$) and ΔF_R(RH$_{amb}$)/ΔF_R(dry) were the averages of the linear interpolation results of f(RH), b(RH), $\bar{\beta}$(RH) and ΔF_R(RH)/ΔF_R(dry) at 67% RH.

~~As shown in Eq. (8), f(RH) has a direct influence on the ratios ΔF_R(RH)/ΔF_R(dry). At the ambient average RH of 67%, the average f(RH$_{amb}$)s for locally-polluted, northerly-polluted and dust-influenced aerosols were 1.17, 1.26 and 1.15, respectively, thus the~~ The ~~ratios~~ ΔF_R(RH$_{amb}$)/ΔF_R(dry) ratios were 1.118, 1.195 and 1.105, respectively (see Table 7). That is to say, averagely, the direct radiative forcing of locally-polluted, northerly-polluted and dust-influenced aerosols increased by 11.8, 19.5 and 10.5% in March at LinAn. ~~due to the aerosol hygroscopic growth at RH=67%. We could also see that the f(RH) and the ratio ΔF_R(RH)/ΔF_R(dry) for northerly-polluted aerosols were the largest, which suggests the effect of f(RH) on direct radiative forcing is larger than that of the upscatter fraction $\bar{\beta}$(RH), since $\bar{\beta}$(RH) shows negative relationship with f(RH).~~

4 Conclusions

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%), f_b(85%) and f_β(85%)) were 1.58(0.12), 1.25(0.07) and 0.79(0.04), respectively~~, which means that at 85% RH, the aerosol scattering coefficient and backscattering coefficient increased by 58 and 25%. The mean values of f(85%) at 550nm and 700nm were similar; while the mean f(85%, 450nm) was approx. 6% lower~~. Slight wavelength dependence of f(85%) was observed at higher f(RH) values.

~~Our experimental data from LinAn station can be categorized into 3 episodes (northerly polluted, locally polluted and dust episodes) according to the wind direction, back trajectory and weather phenomenon. (1) During the locally polluted period, air masses mainly came from relatively clean districts, and the corresponding average and mean standard deviation of f(85%), f_b(85%) and f_β(85%) were 1.52(0.10), 1.21(0.06) and 0.80(0.02), respectively; (2) During the northerly polluted period, air masses came from northern China and passed through heavily polluted areas, and the average and mean standard deviation of f(85%), f_b(85%) and f_β(85%) were 1.64(0.09), 1.28(0.06) and 0.78(0.02), respectively; (3) The average and mean standard deviation of f(85%) during dust episode was 1.48(0.05), much higher than the values immediately before dust arrival.~~

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. ~~Unlike the results of many previous reports like Quinn et al. (2005), Malm et al. (2005), Pan et al. (2009) and~~

~~Yan et al. (2009), f(85%) and OM/(OM+SO_4^{2-}) were unrelated in this study, while~~ Besides, NO_3^- plays an important role in determining the magnitude of f(RH) at LinAn.

~~The h~~Humidograms measured at LinAn can be well described by the model f(RH)=c·(1-RH)$^{-d}$ and model f(RH)=1+a·RH^b~~, and the “a” and “b” for each episode are: 1.24±0.29 and 5.46±1.90 for locally-polluted period, 1.20±0.21 and 3.90±1.27 for northerly-polluted period, and 1.02±0.19 and 4.51±0.80 for dust episode~~. Further investigation shows the shape of the humidogram is closely related to the mass percentage of nitrate. A ~~variable~~ steepness index η has been defined to quantitatively determine the ~~degree of the curvature~~steepness of humidograms. The more nitrate (or less sulfate), the smaller η is and the straighter the curve will be. ~~When mass percentage of nitrate is larger than 18%, most of η is approx. 1.1, which means the aerosol scattering increases continuously and monotonically with a nearly constant speed.~~

In March, the average relative humidity (RH_{amb}) was 67%.~~and the averaged f(RH_{amb})s for locally-polluted, northerly-polluted and dust-influenced aerosols were 1.17, 1.26 and 1.15, respectively. Correspondingly,~~ 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.

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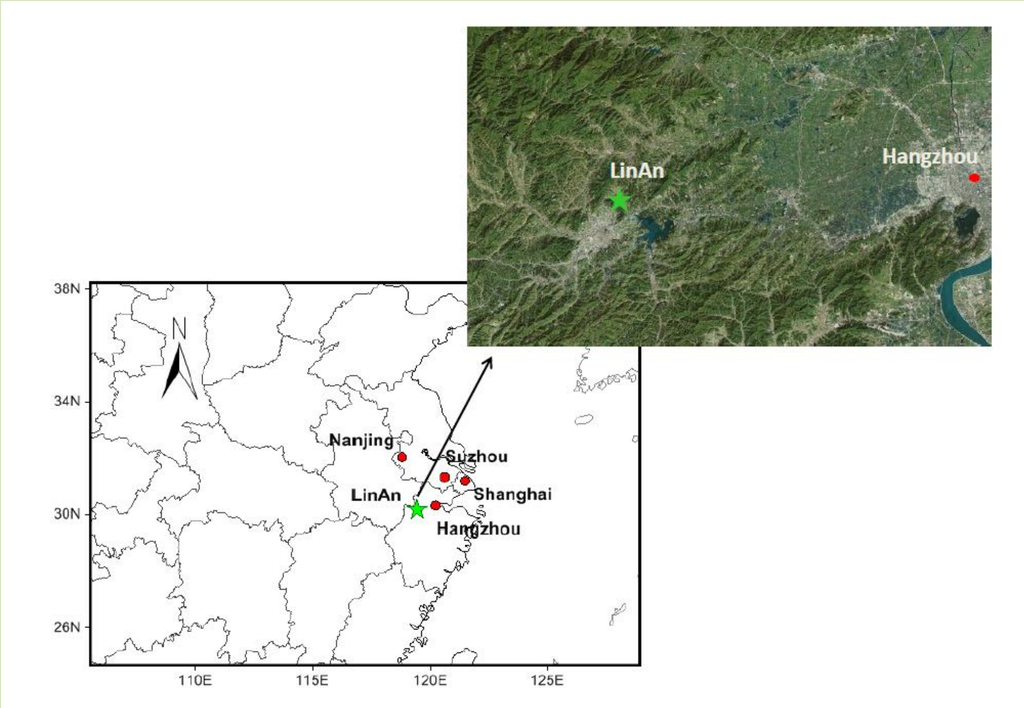

Fig. 1 Location of LinAn station (green pentagramstar) and the main cities in the Yangtze River Delta (red dots) in the lower left panel. The upper right panel is the topography of the surrounding area.

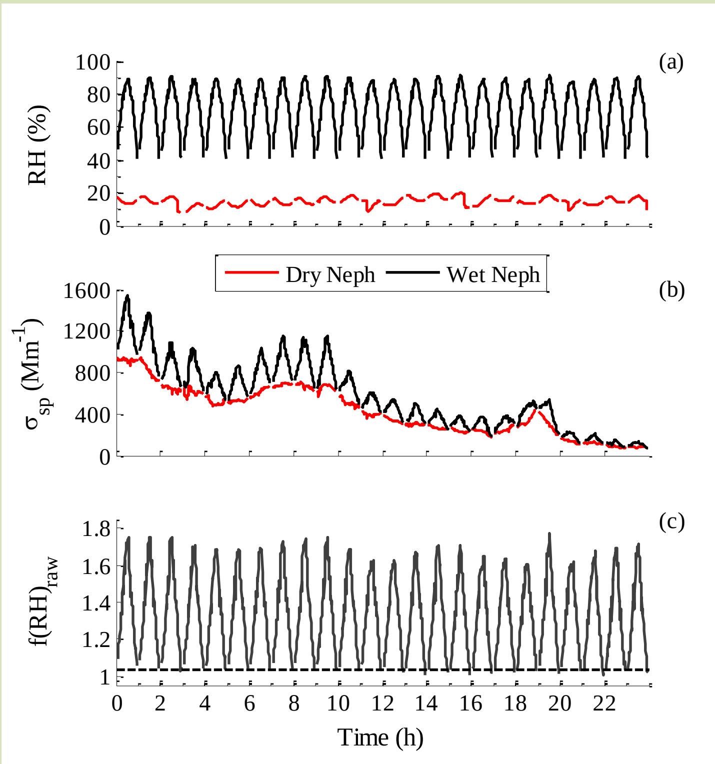

Fig. 3 2 Example of recorded data on 17 March 2013 (a) Relative humidity of inside DryNeph (red line) and WetNeph (black line), the black line represents RH=91%; (b)

Aerosol scattering coefficients measured by DryNeph (red line) and WetNeph (black line) at 550nm wavelength; (c) Raw scattering enhancement factor f(RH, 550nm)$_{raw}$ without normalization, the black dash line was f(RH)$_{raw}$=1.03.

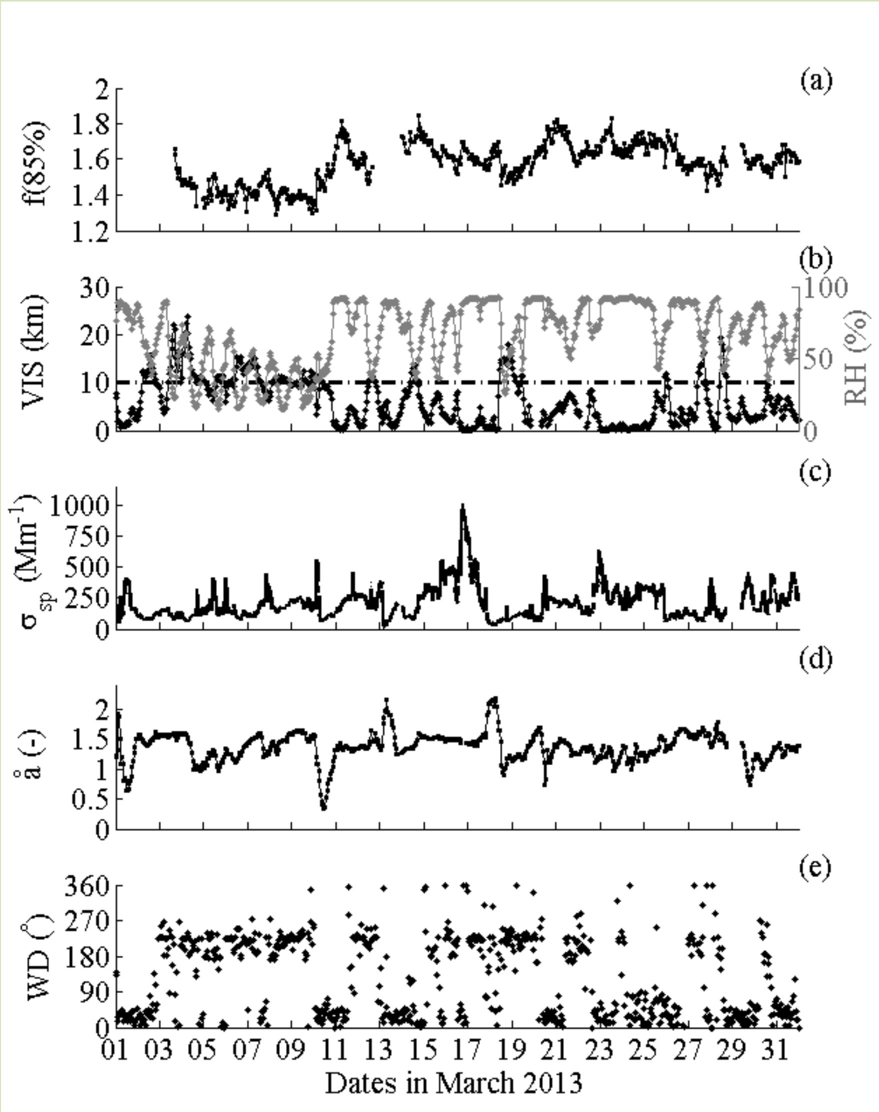

Fig. ~~4~~ 3 Time series of measured and derived aerosol variables, as well as the ambient RH and visibility. (a) scattering enhancement factor ~~at RH~~=f(85%) at 550 nm wavelength; (b) visibility (VIS) and relative humidity (RH) at ambient conditions, the dashed line represents VIS=10 km; (c) aerosol scattering coefficient of DryNeph at 550 nm wavelength; (d) Ångström exponent å; (e) wind direction (WD), indicating prevailing wind directions during observation period was mainly northeasterly (NE) and southwesterly (SW).

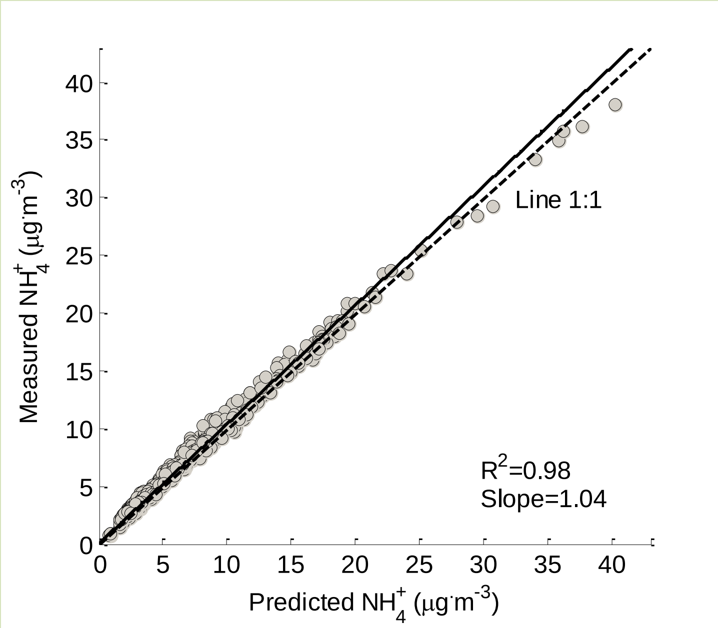

Fig. ~~5~~ 4 Measured and predicted mass concentration of ammonium. The predicted mass concentration of ammonium ($NH_4^+{}_{predicted}$) is calculated by Eq. (4). The solid black line represents the linear least square regression.

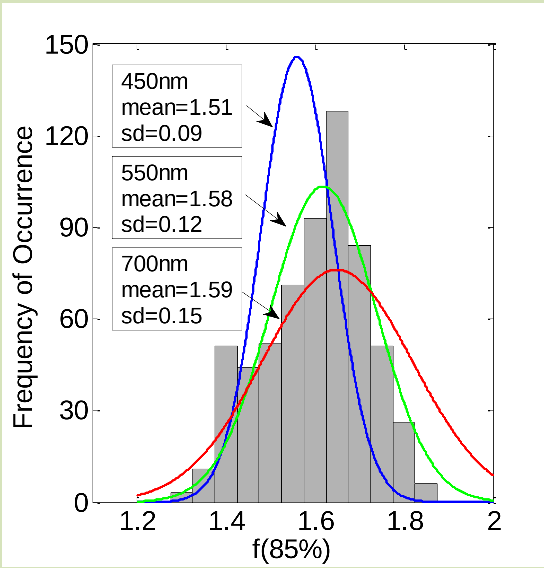

Fig. ~~6~~ 5 Histograms of ~~(a)~~ f(85%,550 nm) overlaid with the Gaussian curves based on the statistics for f(85%,450 nm), f(85%,550 nm) and f(85%,700 nm)~~; (b) Frequency of occurrence of f(85%,550 nm)-f(85%,450 nm) and (c) f(85%,550 nm)-f(85%,700 nm)~~.

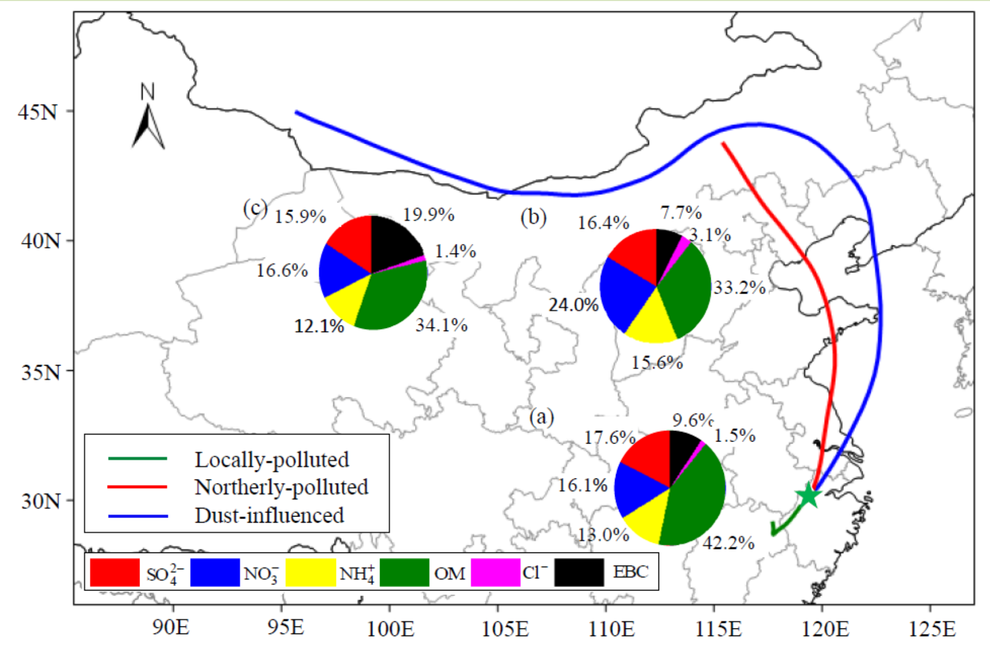

Fig. ~~7~~ 6 72h back trajectories of locally-polluted period, northerly-polluted period and dust-influenced period, together with the mean mass fraction of submicron chemical compositions (SO_4^{2-}, NO_3^-, NH_4^+, OM and Cl^-) measured by AMS and EBC in PM_{10} measured by MAAP. The pie chart (a), (b) and (c) were for locally-polluted, northerly-polluted and dust-influenced periods, respectively.

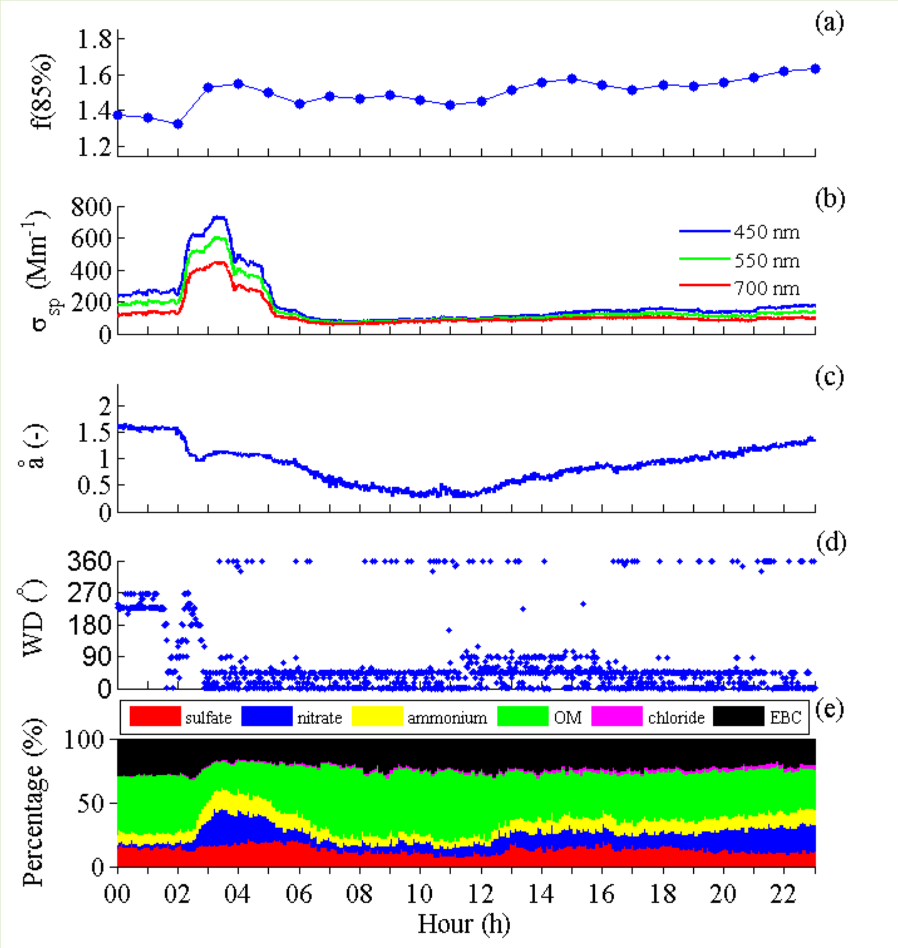

Fig. ~~8~~ 7 Parameters in episode influenced by dust on 10 March 2013 at LinAn (a) scattering enhancement factor ~~at~~ f(85%) at 550nm wavelength ~~RH~~; (b) scattering coefficients at ~~3~~ 450nm, 550nm and 700nm wavelengths; (c) Ångström exponent å; (d) wind direction; (e) mass percentages of chemical species measured by AMS and MAAP.

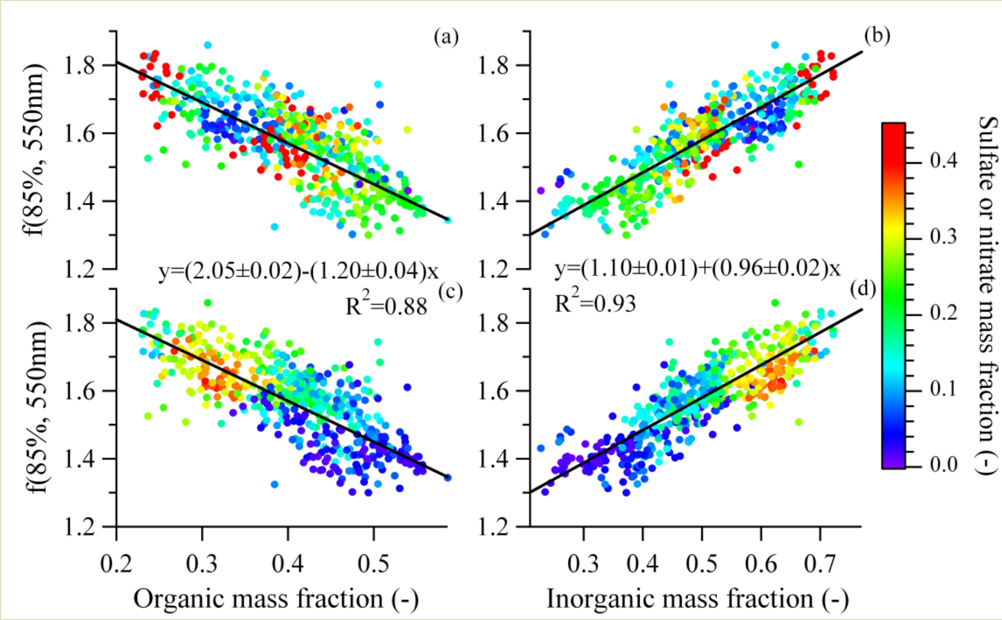

Fig. 9 8 Scattering enhancement factor f(85%, 550nm) vs. organic mass fraction and inorganic mass fraction determined from AMS and MAAP: (a) (b) f(85%, 550nm) vs. organic mass and inorganic mass fraction colored by sulfate mass fraction, respectively; (c) (d) f(85%, 550nm) vs. organic mass fraction and inorganic mass fraction colored by nitrate mass fraction, respectively. The Solid solid black lines represent the linear least square regressiona bivariate linear regression including the uncertainty of f(85%, 550nm) and the standard deviation of chemical compositions.

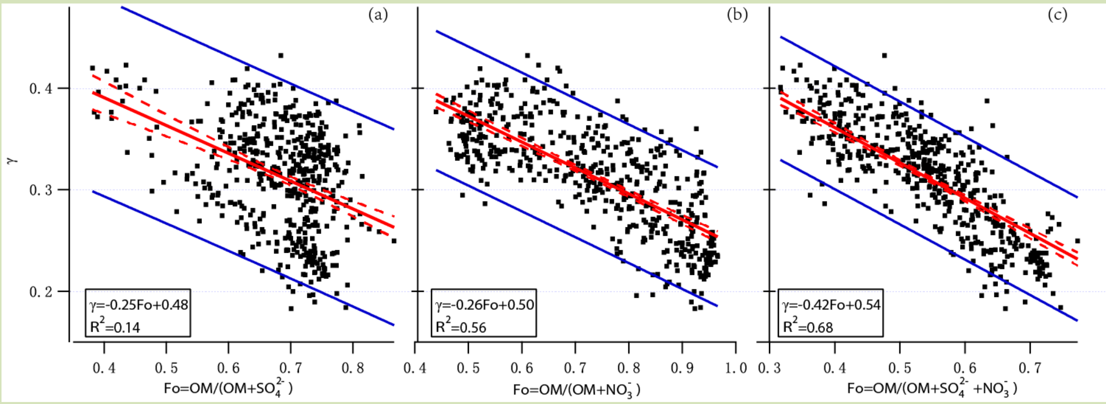

Fig. 10 9 scatter plots of γ versus Fo (a) Fo=OM/(OM+SO_4^{2-}), (b) Fo=OM/(OM+NO_3^-) and (c) Fo=OM/(OM+SO_4^{2-}+NO_3^-). Solid red lines represent the linear fit, dashed red lines show the 95% confidence level for the fit, and solid blue lines show the 95% prediction bands.

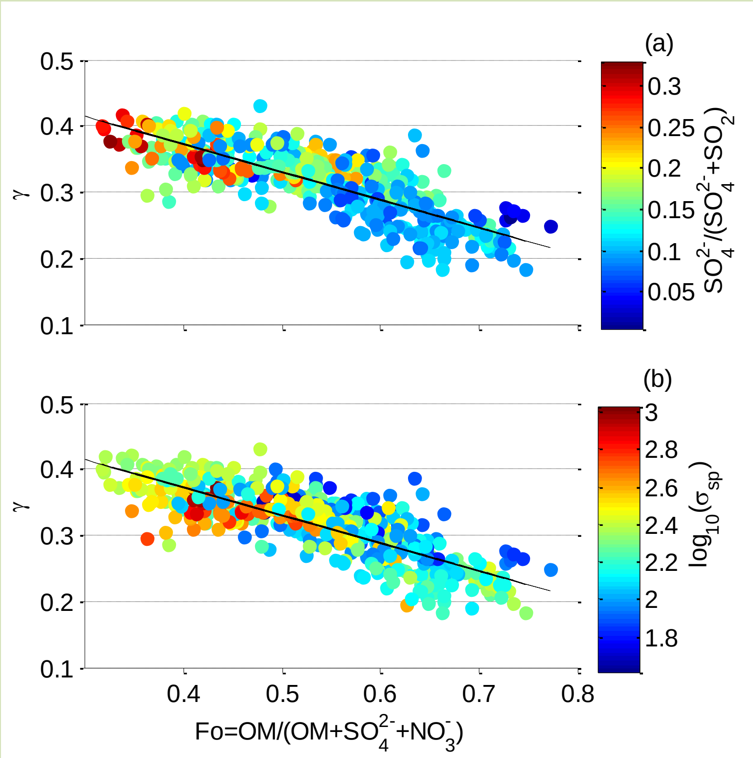

Fig. ~~11~~ 10 γ versus $Fo=OM/(OM+SO_4^{2-}+NO_3^-)$ colored by (a) $SO_4^{2-}/(SO_4^{2-}+SO_2)$ molar ratio and (b) $\log_{10}(\sigma_{sp})$.

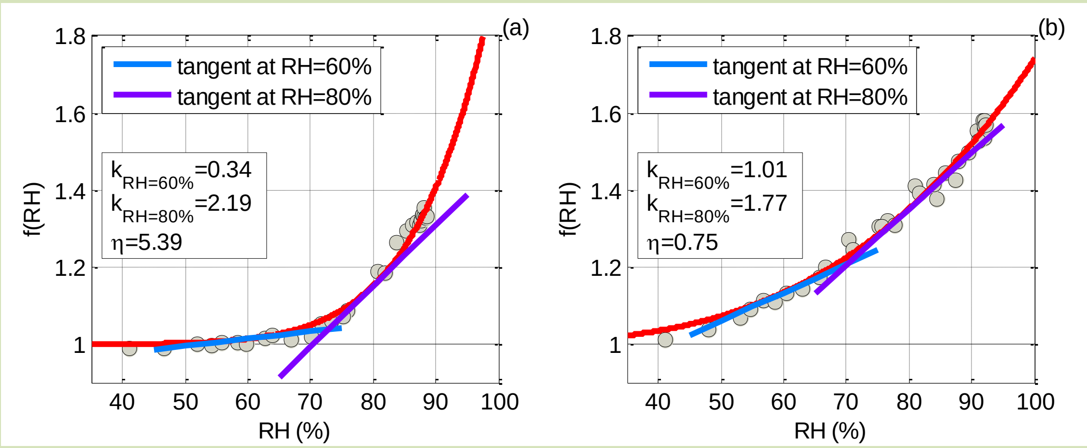

Fig. ~~12~~ 11 Two distinct examples showing different growth patterns and the corresponding η (a) 2013.03.08 18h f(RH) increased slowly at low RH (usually <70%) and then increase more steeply, thus η is big; (b) 2013.03.10 21h f(RH) increased with nearly constant speed and η is small. $k_{RH=60\%}$ and $k_{RH=80\%}$ represent the derivatives at 60% and 80% RH, respectively. f(RH) were at 550nm wavelength.

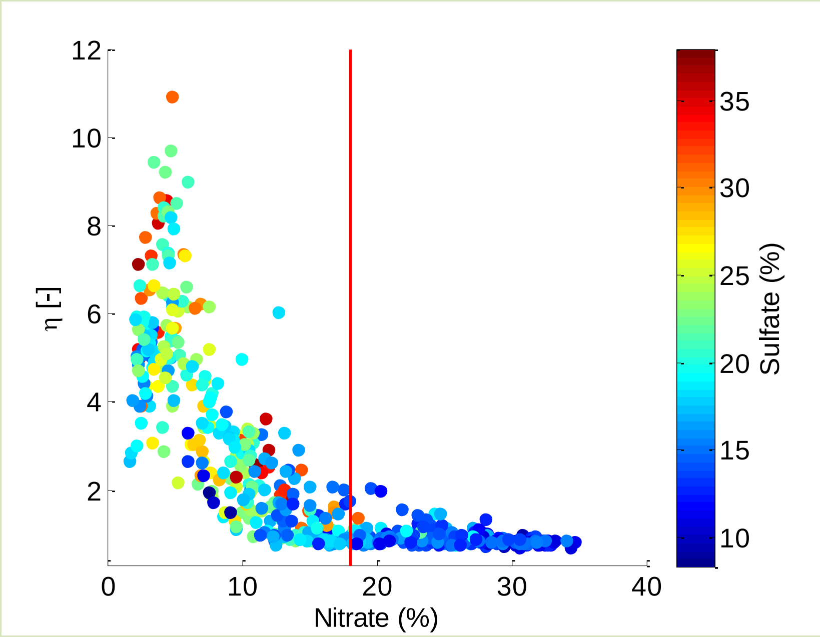

Fig. ~~13~~ 12 Scatter plot of η and the mass percentage of nitrate, colored by the mass percentage of sulfate.

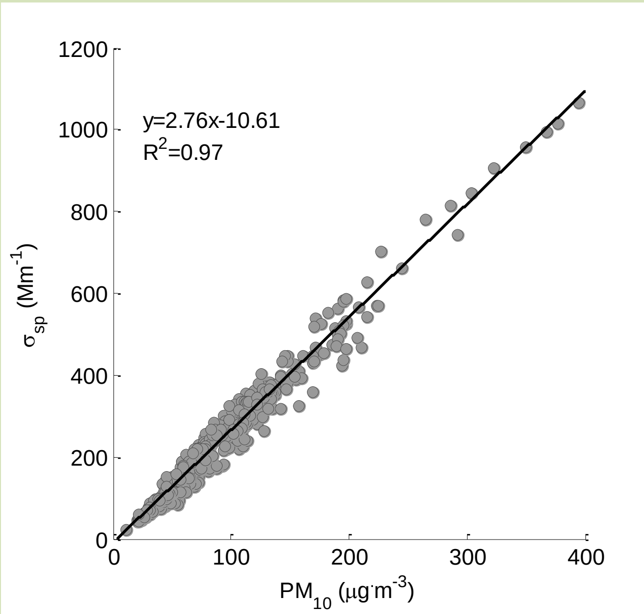

Fig. ~~14~~ 13 Linear regression of scattering coefficients (σ_{sp}) at 550nm wavelength and PM_{10} mass concentration.

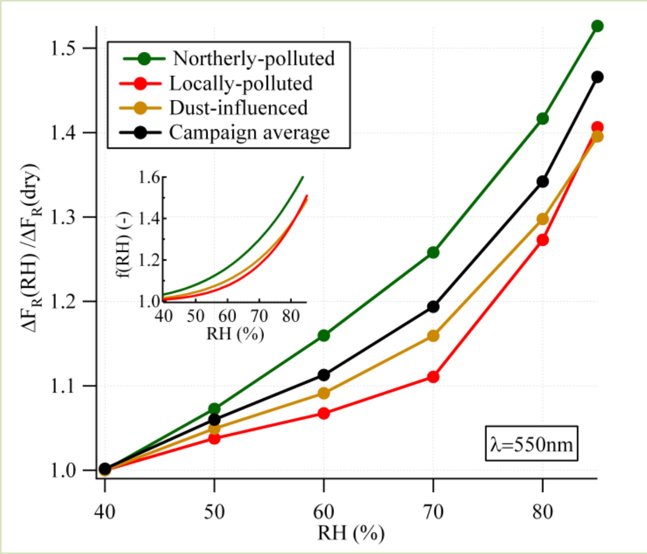

Fig. 14 Influence of relative humidity (RH) on direct radiative forcing for the entire campaign (black line), as well as for the northerly-polluted, locally-polluted and dust-polluted periods, measured by the ratio of radiative forcing at a certain RH to that at dry conditions. The small inlay shows the fitting curves of f(RH) for northerly-polluted, locally-polluted and dust-polluted periods, respectively, using fitting parameters in Table 6. All the parameters were measured at 550nm wavelength.

Table 1 Averaged enhancement factors and mean standard deviations for scattering coefficient, backscattering coefficient and hemispheric backscatter fraction at different RHs (550nm wavelength).

RH(%)	f(RH)	f_b(RH)	f_β(RH)
50	1.07(0.04)	1.04(0.02)	0.96(0.02)
60	1.14(0.08)	1.06(0.04)	0.93(0.04)
70	1.24(0.11)	1.10(0.05)	0.89(0.05)
80	1.43(0.12)	1.18(0.07)	0.83(0.05)
85	1.58(0.12)	1.25(0.07)	0.79(0.04)

Table 2 Summary of mass concentrations ($\mu g\cdot m^{-3}$) of aerosol species measured by AMS as well as MAAP(*) (SD: standard deviation)

	Mean	SD	Minimum	Maximum

Sulfate	8.1	4.1	0.1	26.1
Nitrate	9.8	12.1	0.2	79.2
Ammonium	6.9	5.5	0.5	42.8
Chloride	1.1	2.0	0.002	22.9
OM	17.7	11.1	2.8	93.9
EBC*	4.1	2.8	0.7	25.3

* EBC was meausured by MAAP in PM_{10}.

Table 3 Statistical values of f(85%) at 450 nm, 550 nm and 700 nm wavelengths (SD: standard deviation; prctl: percentile)

λ	mean	SD	90th prctl.	75th prctl.	median	25th prctl.	10th prctl.
450 nm	1.51	0.09	1.63	1.58	1.53	1.47	1.39
550 nm	1.58	0.12	1.72	1.65	1.59	1.49	1.40
700 nm	1.59	0.15	1.77	1.70	1.62	1.46	1.36

Table 4 Average enhancement factors and mean standard deviations for scattering coefficient, backscattering coefficient and hemispheric backscatter fraction in various observation episodes (550nm wavelength).

	Locally-polluted	Northerly-polluted	Dust-influenced
f(80%)	1.36(0.11)	1.50(0.09)	1.37(0.05)
f_b(80%)	1.15(0.06)	1.21(0.06)	1.15(0.03)
f_β(80%)	0.85(0.04)	0.81(0.03)	0.84(0.03)
f(85%)	1.52(0.10)	1.64(0.09)	1.48(0.05)
f_b(85%)	1.21(0.06)	1.28(0.06)	1.19(0.04)
f_β(85%)	0.80(0.02)	0.78(0.02)	0.81(0.03)
N	295	303	14

Table 5 Curve-fitting parameters of f(RH) at 550nm wavelength for various aerosol types in terms of equation $f(RH)=c\,(1-RH)^{-g}$.

	c	g	Reference

Locally-polluted	0.85±0.08	0.29±0.04	This work
Northerly-polluted	0.93±0.07	0.28±0.03	
Dust-influenced	0.87±0.05	0.27±0.02	
Continental	0.9±0.1	-	Zieger et al. (2014)
Arctic[a]	1	0.58±0.09	Zieger et al. (2010)
Marine	0.99	0.54	Carrico et al. (2003)
Polluted	0.59	0.77	
Dust	0.60	0.61	
Polluted Marine	1	0.57±0.06	Gassó et al. (2000)
Dust	1	0.23±0.05	
Clean Marine1[b]	1	0.69±0.06	
Clean Marine2[c]	1	0.73±0.07	

a fitting results for aerosol samples with RH>75%

b fitting results for aerosol samples with RH>60%

c fitting results for aerosol samples with RH>80%

Table 6 Curve-fitting parameters of f(RH) at 550nm wavelength for various aerosol types in terms of Eq. (5).

	a	b	Reference
Locally-polluted	1.24±0.29	5.46±1.90	This work
Northerly-polluted	1.20±0.21	3.90±1.27	
Dust-influenced	1.02±0.19	4.51±0.80	
Clean	1.20±0.06	6.07±0.27	Pan et al. (2009)
Polluted	2.30±0.03	6.27±0.10	
Dust	0.64±0.04	5.17±0.40	
Urban	2.06	3.60	Liu et al. (2007)
Mixed	3.26	3.85	
Marine	4.92	5.04	

Table 7 Estimated effects of aerosol hygroscopic growth on direct radiative forcing by locally-polluted, northerly-polluted and dust-influenced aerosols at LinAn, measured by the ratio ($\Delta F_R(RH_{amb})/\Delta F_R(dry)$) of direct aerosol radiative forcing at the ambient average relative humidity (RH_{amb}=67%) for the entire campaign to that in dry condition. All the parameters were measured at 550nm wavelength.

	$f(RH_{amb})$	b(dry)	$\bar{\beta}$(dry)	$b(RH_{amb})$	$\bar{\beta}(RH_{amb})$	$\Delta F_R(RH_{amb})/\Delta F_R(dry)$
Entire campaign	1.21	0.126	0.268	0.115	0.255	1.157
Locally-polluted	1.17	0.131	0.274	0.123	0.263	1.118
Northerly-polluted	1.26	0.121	0.262	0.106	0.243	1.195
Dust-influenced	1.15	0.146	0.289	0.132	0.274	1.105