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, 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) 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 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.

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<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,λ), enhancement factor for backscattering coefficient f_b(RH,λ) and enhancement factor for hemispheric backscatter fraction f_β(RH,λ). The impact of relative humidity on the aerosol light scattering coefficient is defined as scattering enhancement factor f(RH,λ):

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

where σ_{sp}(dry,λ) and σ_{sp}(RH,λ) 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,λ):

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

where σ_{bsp}(dry,λ) and σ_{bsp}(RH,λ) 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 <40%). Correspondingly, the 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), 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 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

146 performed automatically once per hour. The detailed information of this instrument
147 has been described in many previous studies (Anderson and Ogren, 1998;Charlson et
148 al., 1969;Anderson et al., 1996).

149 The humidifier was built by the aerosol group in Global Monitoring Division,
150 Earth System Research Laboratory, National Ocean & Atmospheric Administration,
151 USA (NOAA/GMD), which was described in Carrico et al. (1998). It consists of 2
152 concentric tubes with a heater and insulation around the outer tube. Sample air flows
153 through the inner tube, while water circulates between the inner and outer tubes. The
154 inner tube is made of porous extruded PTFE (polytetrafluoroethylene) membrane,
155 whose pore size is large enough for water molecules, but too small for larger
156 molecules such as oxygen to cross. The flux of water vapor through the membrane is
157 controlled by regulating the electric current to the humidifier heater until the desired
158 RH is attained. The humidity scan was a one-hour cycle; RH was ramped from ~40 to
159 90% during the first half hour and in the reverse direction during the last half hour.

160 Besides the scattering measurement, particle number size distribution and aerosol
161 chemistry were also measured at the station. Particle number size distributions from 3
162 nm to 10 μm were measured by a twin differential mobility particle sizer (TDMPS)
163 (Birmili et al., 1999) and an aerodynamic particle sizer (APS, model 3321, TSI Inc.).
164 The mass concentrations of sulfate, nitrate, ammonium, organic matter (OM) and
165 chloride (aerodynamic diameter less than 1 μm) were measured by an aerosol mass
166 spectrometer (AMS, Aerodyne Inc.). The equivalent mass concentration of black
167 carbon (EBC) was measured by a multi angle absorption photometer (MAAP, mode
168 5012, Thermo Scientific Inc.) at 637 nm wavelength (Müller et al., 2011), the
169 assumed mass absorption cross-section was 6.6 $m^{2} \cdot g^{-1}$. Visibility was measured using
170 a near-forward scattering sensor (FD12, Vaisala). Meteorological data were provided
171 by the LinAn Regional Atmosphere Background Station.

172 All the instruments were housed in a measurement laboratory where room
173 temperature was controlled at ~25 ℃. All data were reported in Beijing Time
174 (BJT=UTC+8 h) and all the scattering data were referenced at T=0 ℃ and P=1013.25
175 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 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 raw f(40%) value without the normalization. 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 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 is 9 lpm.

2.4 Quality control on scattering measurements

Accurate performance of nephelometers and RH sensors is crucial to retrieve reliable enhancement factors ($f(RH,\lambda)$, $f_b(RH,\lambda)$ and $f_\beta(RH,\lambda)$), 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 <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 RH sensors were calibrated to the external RH sensors with an uncertainty of ≤2%. A good agreement of these RH sensors 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 (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 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 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. 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 2 is an example of our data showing the relative humidity control and corresponding scattering measurements. As can be seen from Fig. 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. 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. 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. 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. 3c) and a sharp decline of Ångström exponent (Fig. 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 ($NH_{4\,\mathrm{predicted}}^+$) (Sun et al., 2010):

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

Figure 4 illustrates the relationship of measured NH_4^+ and predicted NH_4^+. As shown in Fig. 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 for f(85%,550 nm) is shown in Fig. 5. Overlaid on the histogram for f(85%,550 nm) (Fig. 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. 5); while the mean f(85%,450 nm) is ~6% lower than that at 550 nm with a smaller standard deviation (see Fig. 5). 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% 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. 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. 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 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. 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. 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) 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 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. This event was considered to be the 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. 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. 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. 7e). Correspondingly, the scattering enhancement factor f(85%) reached 1.52 at 3 a.m. (see Fig. 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. 7c) and scattering coefficients at 450 nm, 550 nm and 700 nm (Fig. 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. 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. The bivariate linear regressions (Fig. 8) show clearly anti-correlation of f(85%, 550nm) to organics fraction and strongly positive correlation of f(85%, 550nm) to inorganics fraction. This implies that chemical composition plays a vital role in aerosol hygroscopic properties. The absolute values of both slopes (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. 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. 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 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. 9a, Fig. 9b and Fig. 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. 9a) were uncorrelated (R^2=0.14), while γ and Fo ($OM/(OM+NO_3^-)$) (shown in Fig. 9b) and γ and Fo ($OM/(OM+SO_4^{2-}+NO_3^-)$) (shown in Fig. 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 $\mu g \cdot m^{-3}$ 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. 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. 10a) and $\log_{10}(\sigma_{sp})$ (Fig. 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. 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. 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

505 dust episode. It results from the comparably low scattering enhancement factor (e.g.
506 f(80%)=1.44±0.12) of LinAn, which was similar to the value (f(80%)=1.33±0.07)
507 obtained by Gassó et al. (2000) for a dust event. The f(RH) in other studies was much
508 higher than that at LinAn, ranging from 2.04 (polluted marine aerosols in Gassó et al.
509 (2000)) to 3.77 (arctic aerosols in Zieger et al. (2010)), therefore their parameter "g"
510 was much higher.

511 **3.6.2 Parameterization with equation $f(RH)=1+a\ RH^b$**

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

514 $$f(RH)=1+a\ RH^b \quad (5)$$

515 where "a" is positive and "b" is greater than 1. This function is convex, and has been
516 used in many previous studies (Pan et al., 2009;Delene and Ogren, 2002;Carrico et al.,
517 2003;Kotchenruther et al., 1999;Kotchenruther and Hobbs, 1998) to describe
518 monotonic growth. Theoretically, parameter "a" determines the largest value f(100%)
519 can reach, and parameter "b" dominates the curvature of the function. The smaller "b"
520 is, the smaller the curvature of humidogram will be; if "b" equals to 1, then
521 f(RH)=1+a RH. The parameters "a" and "b" from our study and previous results for
522 different aerosol types are listed in Table 6. Taking the locally-polluted episode as an
523 example, although parameter "a" is slightly larger (~3%) than in northerly-polluted
524 episode, parameter "b" is ~40% larger, as a result the f(85%) during locally-polluted
525 period is smaller. Parameter "b" is the largest in the locally-polluted episode, and
526 smallest in the northerly-polluted period, i.e. the curvature of RH-f(RH) line is largest
527 during the locally-polluted episode, then is the dust episode, and northerly-polluted
528 period is the smallest, which is consistent with the mass percentages of nitrate (see
529 Fig. 6) (will be discussed later).

530 **3.6.3 Steepness of humidograms**

531 Among all the humidograms measured at LinAn, f(RH) increases continuously
532 and monotonically. However, the curvatures of different humidograms are different
533 (Fig. 11): some increase with a nearly constant rate and the humidogram line is almost
534 straight, while some increase slowly at first and then increase more steeply at

relatively higher RH, thus the curvature of the humidogram is larger. In order to describe the growth pattern quantitatively, a steepness index η is defined based on the fitting curve:

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

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 larger η is, the bigger the curvature. As is shown in Fig. 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. 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. 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. 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 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. 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 $\Delta F_R(RH)/\Delta F_R(dry)$ 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 $\Delta F_R(RH_{amb})/\Delta F_R(dry)$ for locally-polluted, northerly-polluted and dust-influenced aerosols were calculated using the ambient average RH (RH_{amb}=67%) in March at LinAn. The variables $f(RH_{amb})$, $b(RH_{amb})$,

593 $\bar{\beta}(RH_{amb})$ and $\Delta F_R(RH_{amb})/\Delta F_R(dry)$ were the averages of the linear interpolation
594 results of f(RH), b(RH), $\bar{\beta}$ (RH) and $\Delta F_R(RH)/\Delta F_R(dry)$ at 67% RH. The
595 $\Delta F_R(RH_{amb})/\Delta F_R(dry)$ ratios were 1.118, 1.195 and 1.105, respectively (see Table 7).
596 That is to say, averagely, the direct radiative forcing of locally-polluted,
597 northerly-polluted and dust-influenced aerosols increased by 11.8, 19.5 and 10.5% in
598 March at LinAn.
599
600 **4 Conclusions**
601 The influence of aerosol water uptake on particles' light scattering properties and
602 direct radiative forcing have been investigated at LinAn, a regional atmospheric
603 background station of Yangtze River Delta, China, using the scattering enhancement
604 factor measurement system, together with AMS, MAAP and TDMPS providing the
605 chemical composition and size distribution information. The average enhancement
606 factors and mean standard deviations at 85% RH for scattering coefficient,
607 backscattering coefficient and hemispheric backscatter fraction (f(85%), $f_b(85\%)$ and
608 $f_\beta(85\%)$) were 1.58(0.12), 1.25(0.07) and 0.79(0.04), respectively. Slight wavelength
609 dependence of f(85%) was observed at higher f(RH) values.Generally, the highest
610 values of f(RH) corresponded to aged aerosols with a small fraction of OM; while the
611 lowest values corresponded to younger aerosols with a larger fraction of OM. f(RH)
612 of aerosols with relatively low scattering coefficient was usually low with a large
613 variation; while f(RH) of aerosols with high scattering coefficients was relatively high
614 with a small variation. Besides, NO_3^- plays an important role in determining the
615 magnitude of f(RH) at LinAn.
616 Humidograms measured at LinAn can be well described by the model
617 $f(RH)=c\cdot(1-RH)^{-d}$ and model $f(RH)=1+a\cdot RH^b$. Further investigation shows the shape
618 of the humidogram is closely related to the mass percentage of nitrate. A steepness
619 index η has been defined to quantitatively determine the steepness of humidograms.
620 The more nitrate (or less sulfate), the smaller η is and the straighter the curve will be.
621 In March, the average relative humidity (RH_{amb}) was 67%. Consequently, the direct
622 radiative forcing of locally-polluted, northerly-polluted and dust-influenced aerosols

623 increased by 11.8, 19.5 and 10.5%, respectively due to aerosol uptake water in March
624 at LinAn. At 85% RH, the direct radiative forcing increased by as high as 47% due to
625 the aerosol hygroscopicity. In conclusion, water plays an important role in aerosol
626 scattering properties as well as the radiative forcing, and it should be paid high
627 attention when comparing between remote sensing and in-situ measurements and
628 calculating the climate forcing.
629
630
631 **Acknowledgments**: This work was supported by National Basic Research Program of
632 China (2011CB403401), the National Natural Science Foundation of China
633 (41475118，41175113), China International Science and Technology Cooperation
634 Project (2009DFA22800), CAMS Basis Research Project (2013Z007, 2013Y004),
635 and the Meteorological Special Project of China (GYHY-200906038,
636 GYHY201206037). This paper is partially supported by the CMA Innovation Team
637 for Haze-fog Observation and Forecasts. The authors would also like to thank the
638 LinAn observational station staff for their support. The authors would thank Dr. D.
639 Covert of University of Washington Seattle Department of Atmospheric Sciences
640 USA for useful discussions.
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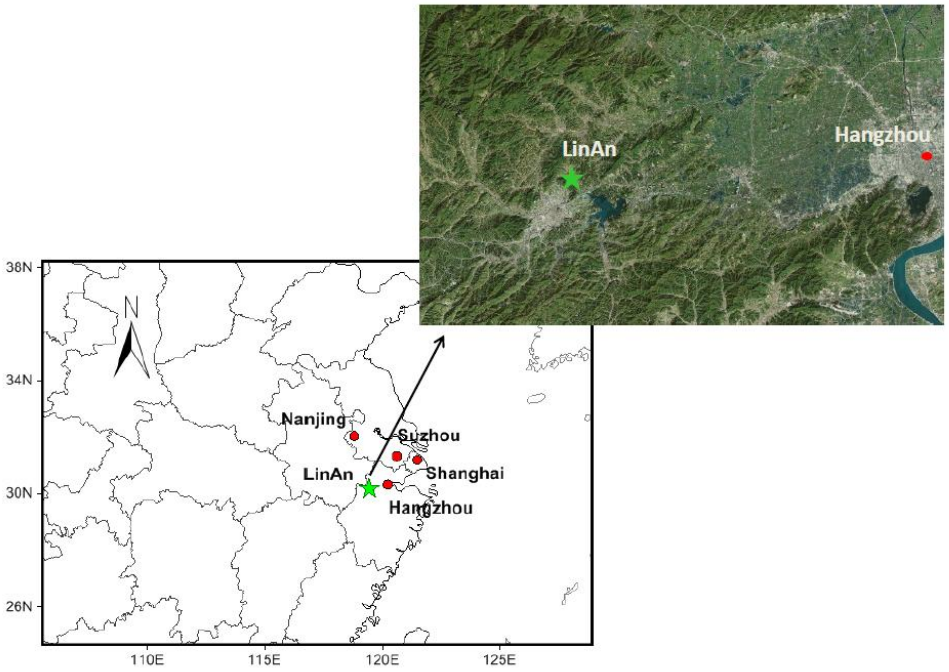
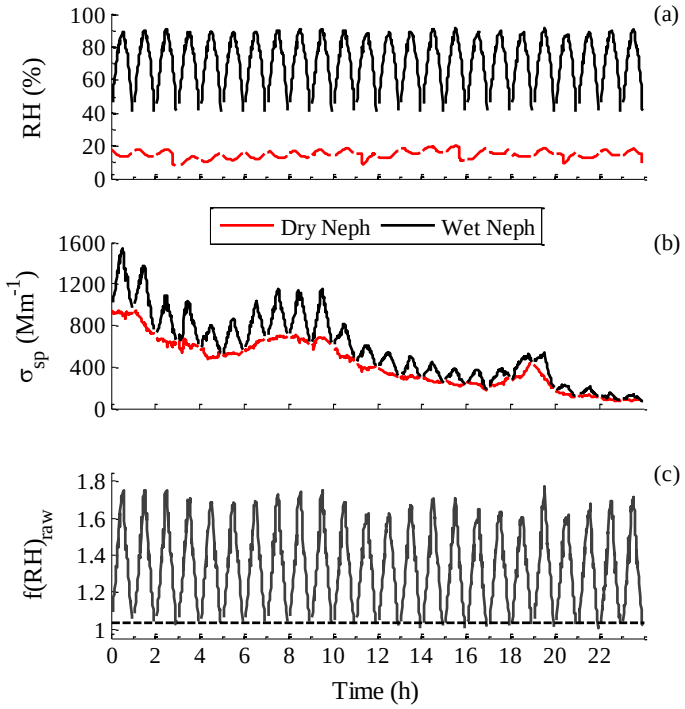
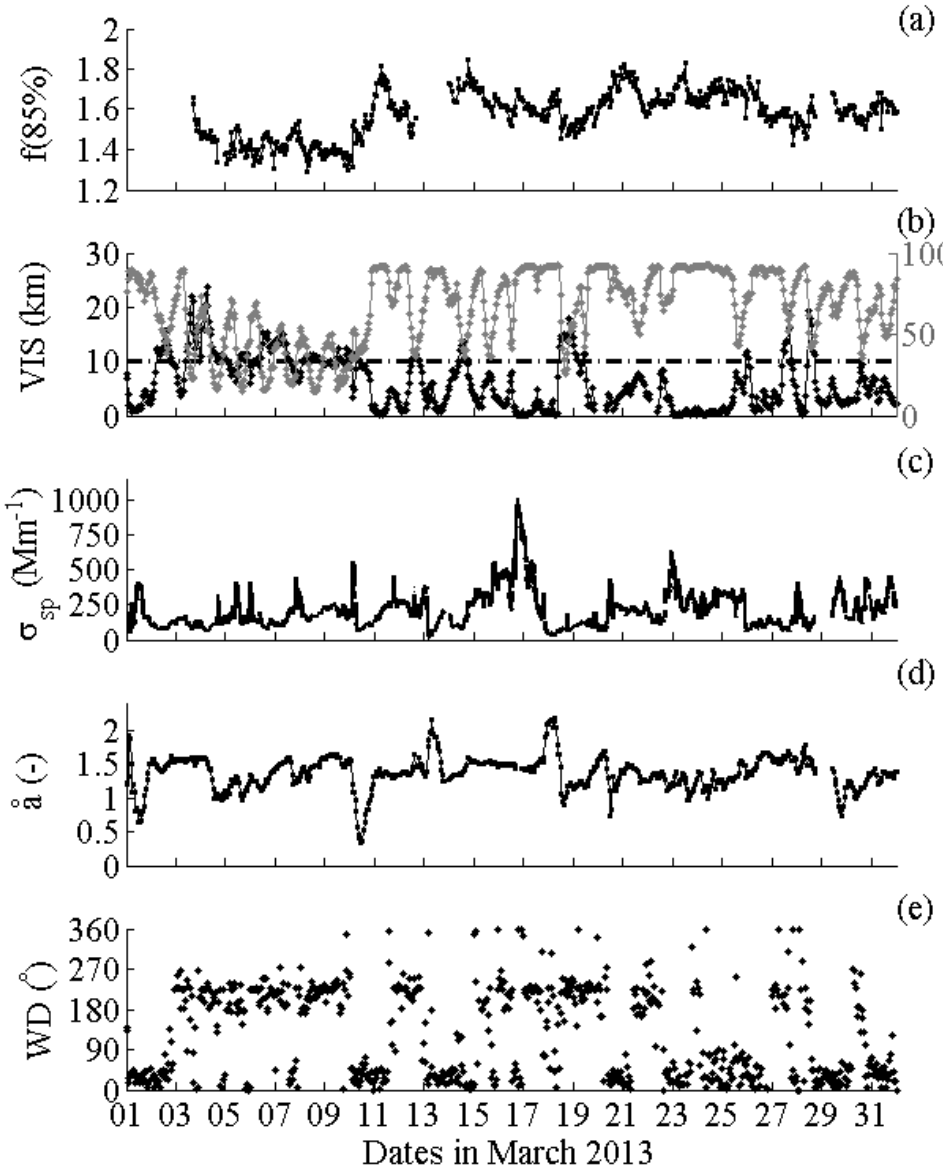


Fig. 1 Location of LinAn station (green star) and the main cities in the Yangtze River Delta (red dots) in the lower left panel. The upper right panel is the topography of the surrounding area.



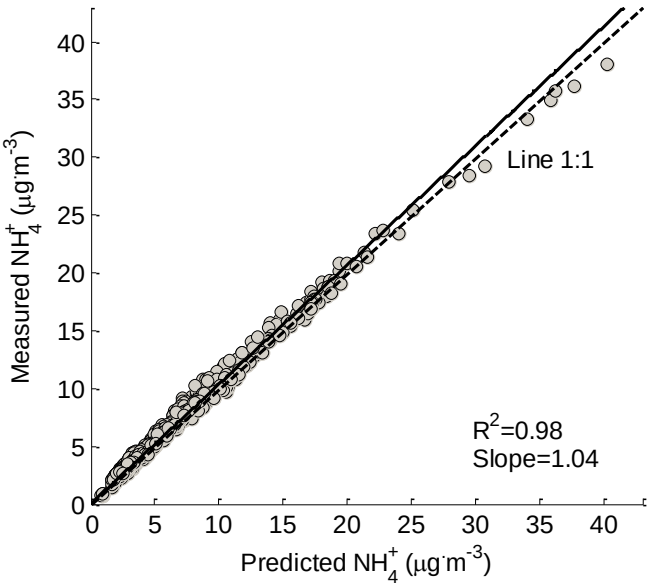
855

856 Fig. 2 Example of recorded data on 17 March 2013 (a) Relative humidity inside
857 DryNeph (red line) and WetNeph (black line); (b) Aerosol scattering coefficients
858 measured by DryNeph (red line) and WetNeph (black line) at 550nm wavelength; (c)
859 Raw scattering enhancement factor $f(RH, 550nm)_{raw}$ without normalization, the black
860 dash line was $f(RH)_{raw}=1.03$.



861

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

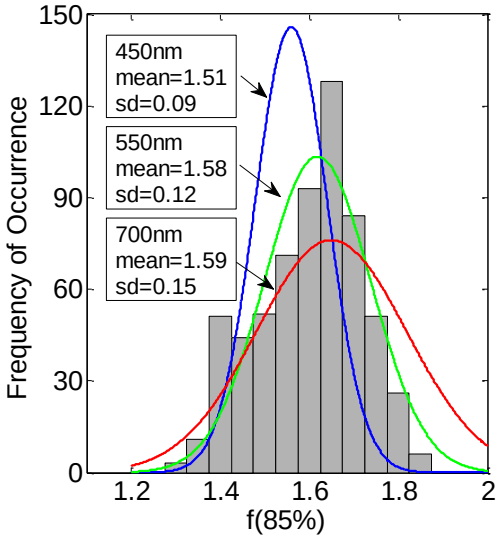


869

870 Fig. 4 Measured and predicted mass concentration of ammonium. The predicted mass

871 concentration of ammonium ($NH_{4\,\mathrm{predicted}}^{+}$) is calculated by Eq. (4). The solid black

872 line represents the linear least square regression.



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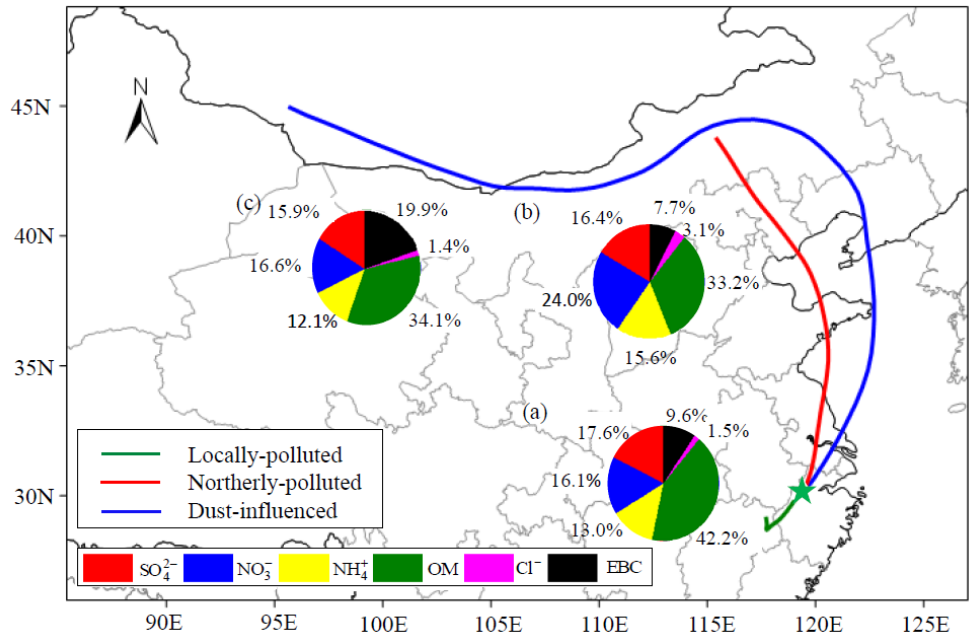
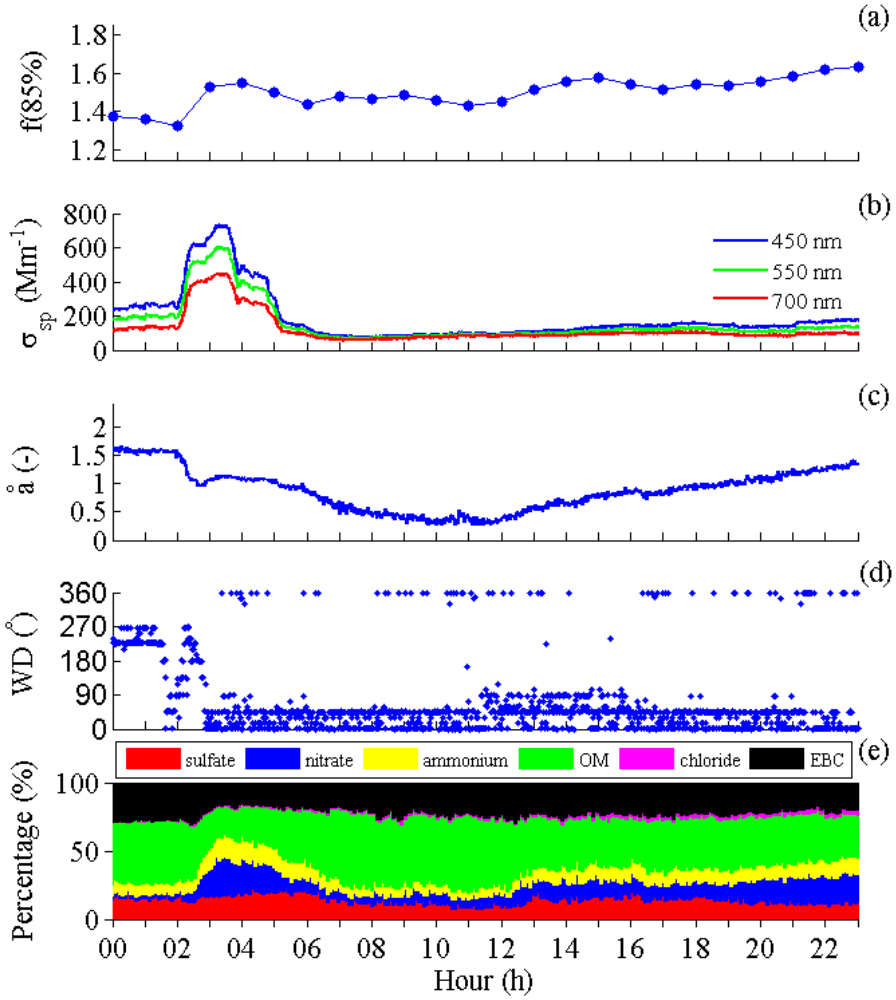
874 Fig. 5 Histograms of f(85%,550 nm) overlaid with the Gaussian curves based on the

875 statistics for f(85%,450 nm), f(85%,550 nm) and f(85%,700 nm).



876

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



882

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

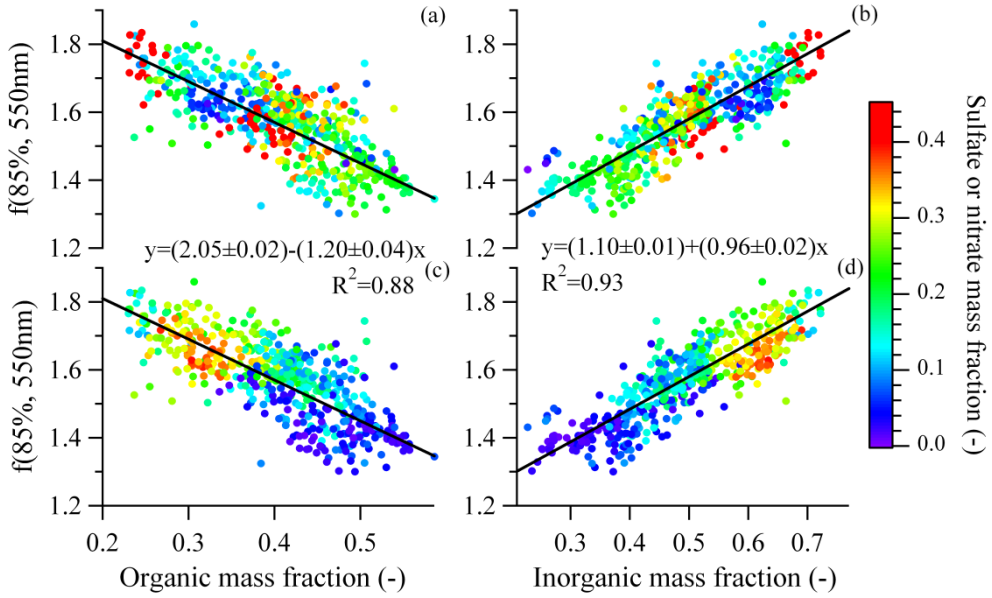


Fig. 8 Scattering enhancement factor f(85%, 550nm) vs. organic mass fraction and inorganic mass fraction determined from AMS and MAAP: (a) (b) f(85%, 550nm) vs. organic mass and inorganic mass fraction colored by sulfate mass fraction, respectively; (c) (d) f(85%, 550nm) vs. organic mass fraction and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

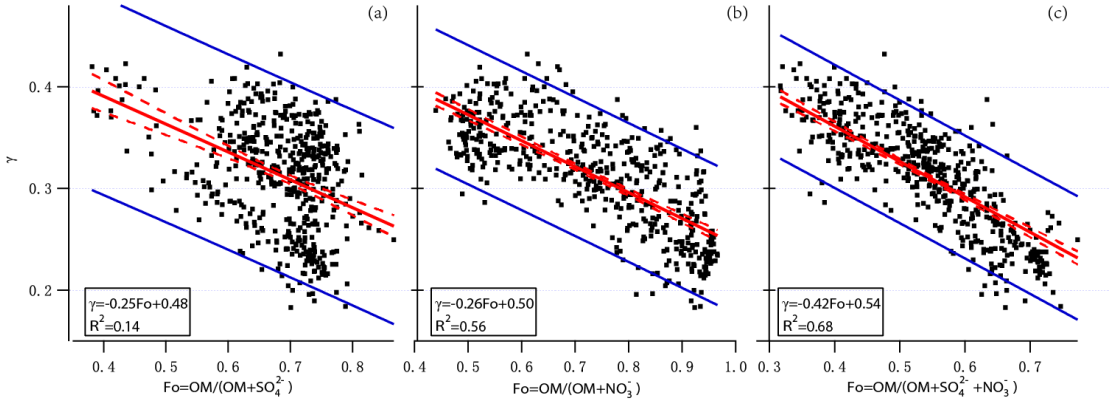


Fig. 9 scatter plots of γ versus Fo (a) Fo=OM/(OM+SO_4^{2-}), (b) Fo=OM/(OM+NO_3^-) and (c) Fo=OM/(OM+SO_4^{2-}+NO_3^-). Solid red lines represent the linear fit, dashed red lines show the 95% confidence level for the fit, and solid blue lines show the 95% prediction bands.

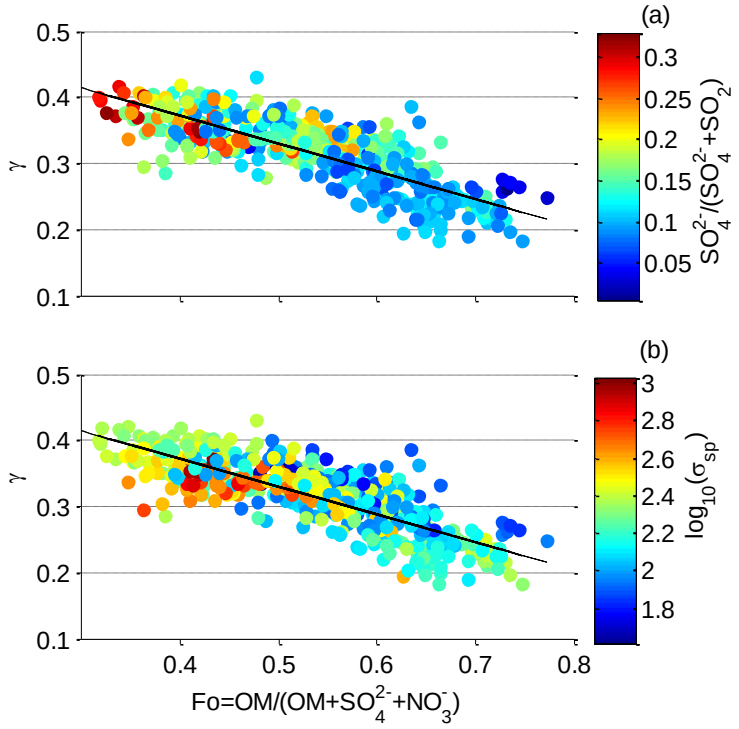


Fig. 10 γ versus Fo=OM/(OM+SO_4^{2-}+NO_3^-) colored by (a) SO_4^{2-}/(SO_4^{2-}+SO_2) molar ratio and (b) $\log_{10}(\sigma_{sp})$.

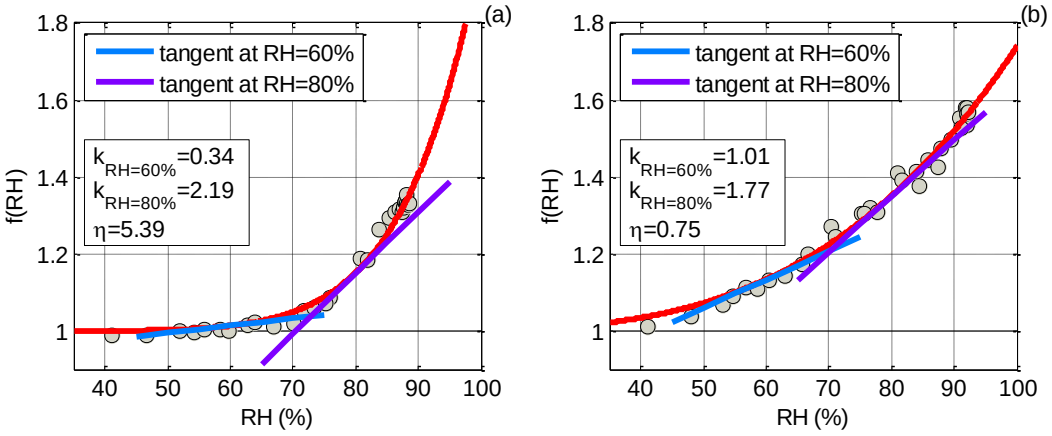
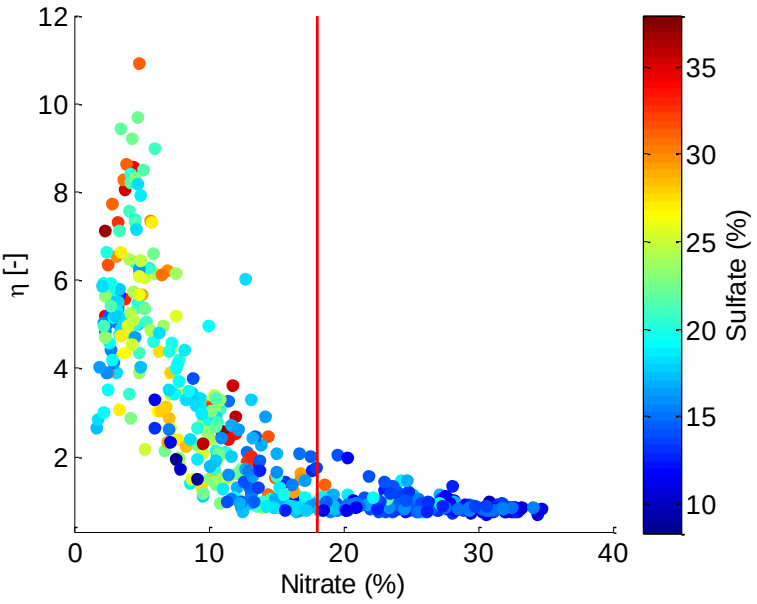


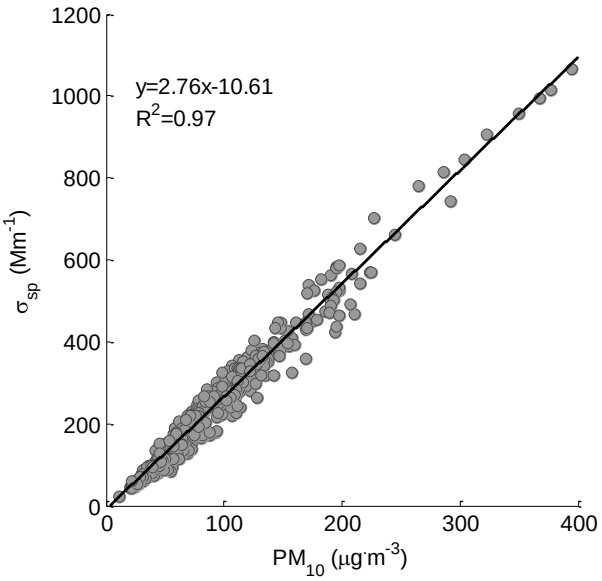
Fig. 11 Two distinct examples showing different growth patterns and the corresponding η (a) 2013.03.08 18h f(RH) increased slowly at low RH (usually <70%) and then increase more steeply, thus η is big; (b) 2013.03.10 21h f(RH) increased with nearly constant speed and η is small. $k_{RH=60\%}$ and $k_{RH=80\%}$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



910

911 Fig. 12 Scatter plot of η and the mass percentage of nitrate, colored by the mass

912 percentage of sulfate.



913

914 Fig. 13 Linear regression of scattering coefficients (σ_{sp}) at 550nm wavelength and

915 PM_{10} mass concentration.

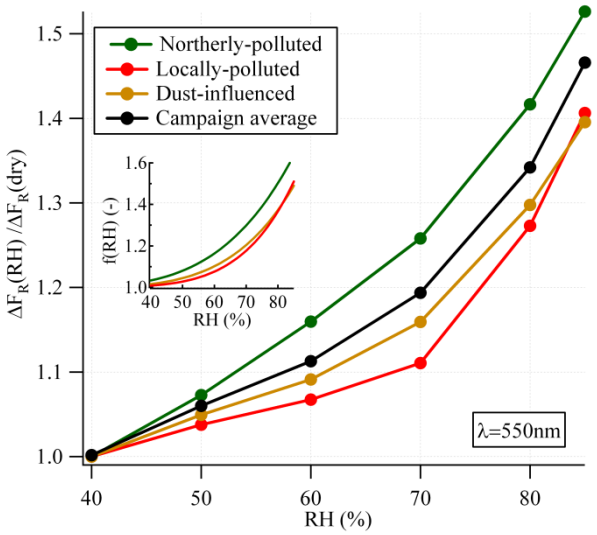


Fig. 14 Influence of relative humidity (RH) on direct radiative forcing for the entire campaign (black line), as well as for the northerly-polluted, locally-polluted and dust-polluted periods, measured by the ratio of radiative forcing at a certain RH to that at dry conditions. The small inlay shows 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

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

931

932 Table 3 Statistical values of f(85%) at 450 nm, 550 nm and 700 nm wavelengths (SD:
933 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

934

935 Table 4 Average enhancement factors and mean standard deviations for scattering
936 coefficient, backscattering coefficient and hemispheric backscatter fraction in various
937 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

938

939 Table 5 Curve-fitting parameters of f(RH) at 550nm wavelength for various aerosol
940 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	

941 a fitting results for aerosol samples with RH>75%

942 b fitting results for aerosol samples with RH>60%

943 c fitting results for aerosol samples with RH>80%

944

945 Table 6 Curve-fitting parameters of f(RH) at 550nm wavelength for various aerosol
946 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	

947

948 Table 7 Estimated effects of aerosol hygroscopic growth on direct radiative forcing by
949 locally-polluted, northerly-polluted and dust-influenced aerosols at LinAn, measured
950 by the ratio ($\Delta F_R(RH_{amb})/\Delta F_R(dry)$) of direct aerosol radiative forcing at the ambient
951 average relative humidity (RH_{amb}=67%) for the entire campaign to that in dry
952 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

953