

Aircraft measurements of aerosol distribution, warm cloud microphysical properties, and their relationship over the Eastern Loess Plateau in China

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ABSTRACT

In situ aircraft measurements of aerosols and clouds were performed over the eastern Loess Plateau in Shanxi Province, China. The microphysical properties of both aerosols and warm clouds, including aerosol number concentration (N_a), particle effective diameter (ED), number concentration of cloud droplets (N_c), cloud droplet diameter (D_c), and liquid water content (LWC) of clouds, determined through six flight observations performed in May 2013 were obtained and analysed. The mean magnitude of measured N_a over the six flights was 10^3 cm^{-3} , and accumulation mode particles dominated the majority. Most EDs of aerosol particles were less than $1 \mu\text{m}$. Large amounts of fine aerosol particles were constrained to the lower layer, with ED smaller than $0.5 \mu\text{m}$, and N_a decreased with height. The base heights of warm clouds ranged from 1000 to 2800 m. The maximum and average values of the measured N_c ranged from 147 to 311 cm^{-3} and 51 to 157 cm^{-3} , respectively. The maximum and average D_c ranged from 13.5 to 28.9 and 5.8 to $13.1 \mu\text{m}$, respectively. The average LWC of warm clouds was $0.05 \text{ g}\cdot\text{m}^{-3}$. N_a was negatively correlated with N_c either in the vertical or horizontal direction. N_c was higher with a smaller size of cloud droplets under high aerosol loading conditions. A small number of cloud droplets with larger size were formed under low aerosol loading conditions. At a certain range of LWC, N_c and D_c showed a negative correlation. The increase in LWC was related to an increase in the size of cloud droplets rather than the number of cloud droplets. The cloud droplet size distribution showed that small droplets dominated the total cloud droplet concentration. A bimodal lognormal distribution function can be efficiently used to describe the average spectrum of warm cloud droplets.

Keywords: aerosol, warm clouds, microphysical properties, aerosol–cloud interaction, aircraft measurements

1. Introduction

Atmospheric aerosols exert a significant effect on Earth's radiative forcing by scattering and absorbing solar radiation directly (Yang et al., 2016a, 2016b). In addition, atmospheric aerosols affect the microphysical properties of clouds by acting as cloud condensation nuclei (CCN) or ice nuclei (IN) during cloud formation (Twomey, 1977; Garrett et al., 2004; Garrett and Zhao, 2006; Zhao et al., 2012; Qiu et al., 2017; Zhao et al., 2018a); this, in turn, modifies the formation regime of rain, snow, and

other forms of precipitation and further affects the climate system (Haywood and Boucher, 2000; Haywood et al., 2003; Lohmann and Feichter, 2005; Andreae and Rosenfeld, 2008; Li et al., 2011a; Tao et al., 2012; Zhao et al., 2018b). The spatial distribution of aerosol particles, including the number concentration (N_a) and effective diameter (ED), exerts strong effects on the estimates of direct and indirect radiative forcing by aerosols (Welton et al., 2002; Han et al., 2003; Garrett and Zhao, 2006; IPCC, 2007; Campbell et al., 2013; Ma and Yu, 2014; Zhao and Garrett, 2015; Li et al., 2015a). Changes in the concentration, particle size, source, and type of aerosols

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also affect the optical properties, cover, and lifetime of clouds (Haywood and Boucher, 2000; Hudson et al., 2011; Hudson and Noble, 2011, 2014a, 2014b). Aerosol particles appearing at the cloud level can affect the microphysical properties of clouds, including cloud droplet number and size distribution and liquid water content (LWC), which are crucial factors in the formation process of precipitation (Deng et al., 2009; Zhang et al., 2011). Many studies have been conducted to understand the physical, chemical, and optical properties of aerosol particles and their effects on cloud, precipitation, and other weather phenomena (Yin et al., 2000, 2002; Zhao et al., 2006; Grützun et al., 2008; Li et al., 2011b; Calvo et al., 2013; Junkermann and Hacker, 2015; Zhao et al., 2015, 2017; Yuan et al., 2016). However, an in-depth understanding of the properties of atmospheric aerosol particles is still lacking, and large uncertainties remain in the study of aerosols, especially in terms of interactions between aerosols and clouds. In particular, Fan et al. (2018) indicated that the representation of the aerosol–cloud interaction in climate models is an important factor contributing to the poor simulation of cloud cover. Therefore, the spatial distribution of aerosols and the relationship between aerosols and clouds should be explored by performing extensive measurements and simulations in different places (Yin et al., 2000, 2002; Fan et al., 2012; Hudson and Noble, 2014a, 2014b).

In different observation methods, data on aerosols and clouds are mainly retrieved from ground-based instruments such as a ground-based LIDAR or cloud radar, and remote sensing measurements such as satellite data products; however, detailed microphysical parameters of aerosols and clouds remain poorly observed and analysed. Aircraft observations can afford the principal advantage of yielding detailed information on aerosol and cloud properties with height, albeit within short time periods over geographic areas of limited extent, thereby enabling the question of aerosol and clouds properties in the vertical direction to be addressed. So, aircraft measurements enable us to directly determine the spatial distribution characteristics of aerosols and the microphysical properties of cloud droplets with high temporal and spatial resolution in the target area (Junkermann and Hacker, 2015; Pace et al., 2015; Li et al., 2015a, 2015b). Thus, aircraft measurements are important for studying aerosol–cloud interactions (Bahreini et al., 2003; Andreae and Rosenfeld, 2008; Zhang et al., 2011; Hao et al., 2017; Zhao et al., 2018).

In recent decades, extensive airborne measurements of aerosol properties have been performed in China, such as in the North China Plain (Zhang et al., 2006, 2009; Liu et al., 2009; Yin et al., 2009; Sun et al., 2013), the Loess Plateau (Li et al., 2011, 2012, 2015a, 2015b), and eastern

China (Hao et al., 2017). A few aircraft measurements of aerosol–cloud interactions have also been carried out, and some valuable results have been reported. For example, Deng et al. (2009) used data from 75 flights around the Beijing area between 2005 and 2006 to analyse the statistical microphysical properties and parameterization of the effective radius of warm cloud droplets; they suggested that LWC plays an important role in controlling the cloud droplet number concentration and affects the droplet size. Zhang et al. (2011) analysed aerosol–cloud interactions over highly polluted areas of Beijing by using data from seven aircraft flights and reported that the cloud droplet size is considerably affected by the aerosol number concentration and LWC. Zhao et al. (2018) used six *in situ* flight observations to study the relationship between aerosol loading and cloud microphysical properties in September 2015 over Hebei, China, and they reported that the aerosol first indirect effect was approximately 0.10–0.19. However, because of high operating costs, strict control of airspace, and other reasons, aircraft observations of the spatial distribution and microphysical properties of aerosols and clouds over different areas are still lacking. Therefore, this study evaluated uncertainties in measured aerosol and cloud properties including the measured characteristics of aerosols and cloud droplets. The relationship between aerosols and cloud droplets in different LWCs, and the effect of high and low aerosol concentrations on cloud formation were also discussed.

An airborne campaign focused on comprehensive observation of atmospheric aerosol and cloud characteristics was conducted in May 2013 in Shanxi Province over the eastern Loess Plateau of China. In this study, data from six flights on five cloudy days were selected to analyse the spatial distribution of aerosols and microphysical properties of clouds during spring.

This article is organised as follows. Section 2 describes the observation area, aircraft campaign details, and data sets. Section 3 presents and provides a detailed discussion of measured results. Section 4 presents the conclusion.

2. Description of the field campaign

2.1. Observation area

The observation area is located in Shanxi Province. As shown in Fig. 1, Shanxi Province is located on the eastern Loess Plateau of northern China, and the Loess Plateau covers most of the province. The average surface altitude of the entire province is approximately 1000 m (height above sea level [ASL]). As one of the most important energy and heavy chemical bases of China, coal burning, steel, and chemical industries are the main industries here

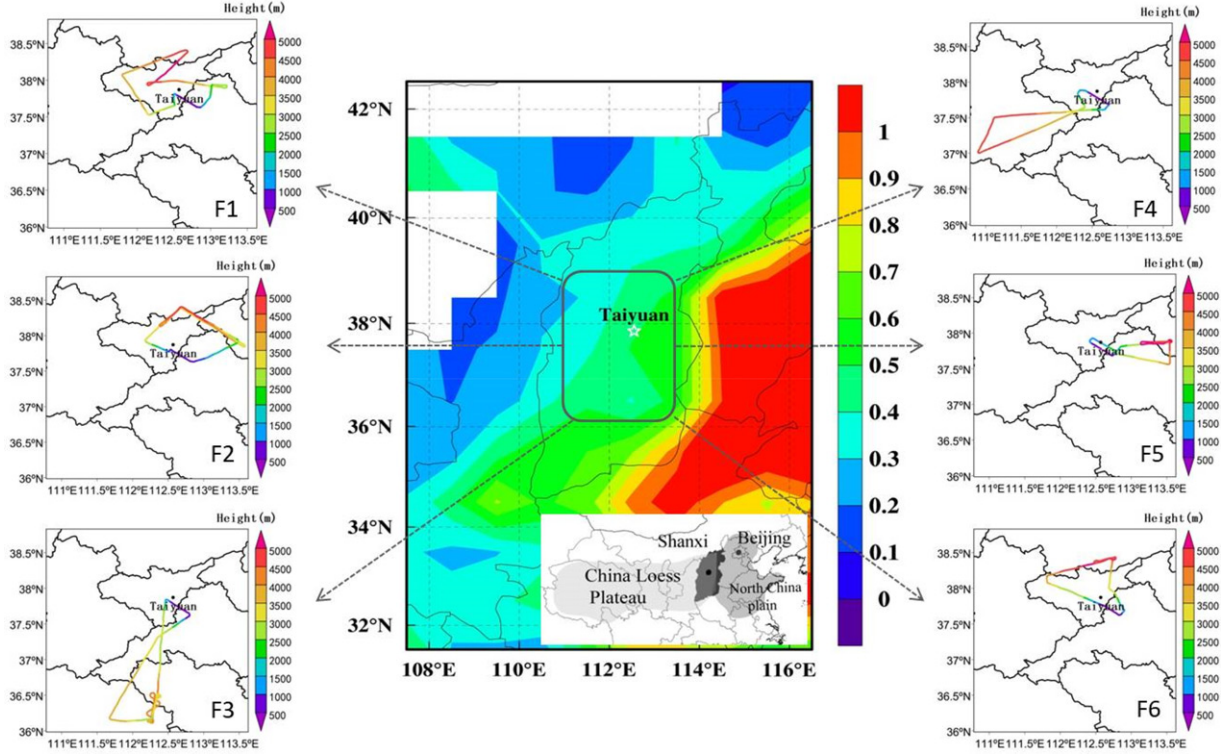


Fig. 1. Map of Shanxi Province in North China with the six flight tracks. The shaded contour represents the mean distribution of aerosol optical depth during 121–151 days (May 1–May 30) in 2013 obtained from moderate resolution imaging spectroradiometer data, and Taiyuan City is marked with a white star. The inset map shows the location of the observation area in China, and the dark gray-shaded area shows Shanxi Province.

(Li et al., 2011, 2012). An increase in the concentration of anthropogenic aerosol particles from industrial emissions may exert a strong effect on the regional atmospheric environment and climate. From the year 2011, a few observational experiments concerning aerosols and clouds have been performed in this area, and some results have been reported (Li et al., 2011, 2012, 2015a, 2015b, 2017a; Wang et al., 2012), which attracted considerable attention from researchers and the public. This aircraft observation campaign was conducted at Wusu International Airport located in Taiyuan (112.55°E, 37.867°N and altitude: 778 m ASL), the capital of Shanxi Province, and the observation area was mainly in the central area of Shanxi (Fig. 1).

2.2. Instruments in the aircraft

A Y-12 turboprop airplane was used as the main observation platform. The typical flight speed is $60\text{--}70\text{ ms}^{-1}$, and the rate of ascent and descent is $2\text{--}5\text{ ms}^{-1}$. This airplane is equipped with multiple aerosol and cloud instruments. Each instrument was calibrated and tested rigorously before the airborne campaign. A passive cavity

aerosol spectrometer probe (PCASP-100X, DMT) was used to observe aerosol particles with diameters of $0.09\text{--}3.0\text{ }\mu\text{m}$ within 30 variable size bins at a frequency of 1 Hz. The particle number concentration range recorded by the PCASP is $0\text{--}2.0 \times 10^4\text{ cm}^{-3}$. And the estimated uncertainty of the aerosol number concentration by PCASP is approximately 10–15% (Duan et al., 2012; Cai et al., 2013). A cloud droplet probe (CDP, DMT, USA) was used to observe cloud droplets with diameters of $2\text{--}50\text{ }\mu\text{m}$ within 30 size bins at a frequency of 1 Hz. The width of the first 12 channels of the CDP is $1\text{ }\mu\text{m}$, and the width of bins ranged from 13 to 30 is $2\text{ }\mu\text{m}$. The measurement resolution of the CDP is $1\text{ }\mu\text{m}$, and the observed cloud droplet number concentration range of CDP is $0\text{--}10^4\text{ cm}^{-3}$. The estimated uncertainty of the cloud droplet number concentration by CDP probe is about 20% (Lance, 2012). The PCASP and CDP were calibrated using polystyrene latex spheres manufactured by Particle Metrics Inc. (PMI, USA) before conducting measurements. The meteorological parameters including ambient temperature (T), relative humidity (RH), and location were measured by using a meteorological package named Aircraft Integrated Meteorological

Table 1. Flight summary (date: yy-mm-dd, time: hh:mm (CST), height: m).

Flight	Date	Time range	Maximum height of flight	Height of 0 °C layer	Height of cloud base	Cloud type
F1	2013-05-08	10:25–12:50	5464	2980	1040	As
F2	2013-05-08	14:30–17:10	5540	2890	1325	As
F3	2013-05-23	9:45–13:10	4310	3040	2825	As
F4	2013-05-25	15:20–17:25	4640	3540	2010	Cu
F5	2013-05-26	9:20–11:25	5610	3250	1210	As
F6	2013-05-28	10:45–12:30	5440	2940	978	Sc

Table 2. Statistical values of warm clouds and aerosols below and in clouds (height: m, N_a : cm^{-3} , N_c : cm^{-3} , D_c : μm , LWC: $\text{g}\cdot\text{m}^{-3}$, STP).

Flight	Cloud height	Sample number	N_a under cloud	N_a in cloud base	Aerosol activation ratio	N_c			D_c			LWC		
						Max.	Ave.	Sd.	Max.	Ave.	Sd.	Max.	Ave.	Sd.
F1	1088–3022	609	2733	1123	0.59	201	63	41	26.3	13.1	5.7	0.23	0.05	0.04
F2	1325–3275	648	3944	348	0.91	147	51	25	28.9	13	5.4	0.29	0.05	0.05
F3	2825–3184	406	982	847	0.14	311	118	74	15.9	9.9	3.1	0.53	0.07	0.09
F4	2011–3022	233	4573	4017	0.12	305	157	92	13.5	8.6	2.5	0.24	0.05	0.06
F5	1202–3007	705	3683	2850	0.23	273	130	70	18.4	10.1	4.2	0.45	0.06	0.08
F6	978–2973	270	1087	240	0.78	286	110	74	17.3	5.8	1.7	0.06	0.01	0.01
Ave.			2834	1571	0.46	254	105	63	20.1	10.1	3.8	0.30	0.05	0.05

Measurement System (AIMMS-20, Advantech Research Inc.) at a frequency of 1 Hz. Temperature measured by AIMMS-20 ranges from -20°C to 40°C with a detection accuracy of 0.3°C , and the relative humidity detection accuracy is 2.0% with a resolution of 0.1%. Cloud LWC was measured by a LWC King probe (DMT, USA). The estimated uncertainty of LWC data is approximately 15%. A global positioning system (GPS) device was used to record the airplane location during the flights, too. Data in the first bins of the PCASP ($0.09\text{--}0.1\mu\text{m}$) and CDP ($2\text{--}3\mu\text{m}$) were inaccurate due to detection limits; thus, they were eliminated from the analysis. In this study, all the concentrations of aerosol particles and cloud droplets are presented in terms of standard temperature and pressure conditions (STP, 273.15 K and 101.3 kPa.). The time indicated in this study refers to the China Standard Time (CST), and heights indicated in this paper refer to heights above ground level (AGL) of Taiyuan.

2.3. Data sets

Data from the six flights were used in this study. To obtain complete observational data, all equipments were turned on before the aircraft took off. After takeoff, the aircraft always slowly climbed to the designated observation area and performed observation. The sampling interval was 1 s during all flights. The aircraft flight altitudes were normally from surface to 6000 m height. If the

aircraft penetrated clouds, horizontal observations were performed at different heights in the cloud region. Flight details are listed in Table 1, and flight tracks are shown in Fig. 1.

To define the appearance of clouds, several criteria were used according to previous studies. The number concentration of the CDP is often used as a criterion; for example, $N_c > 10\text{cm}^{-3}$ (Rangno and Hobbs, 2005; Stith et al., 2006), and $\text{LWC} > 0.01\text{g}\cdot\text{m}^{-3}$ (Gultepe et al., 1996; Gultepe and Isaac, 2004) or $\text{LWC} > 0.001\text{g}\cdot\text{m}^{-3}$ (Deng et al., 2009; Zhang et al., 2011) is another criterion. In this study, valid records of cloud measurements were determined using the following criterions. First, records with a total cloud droplet number concentration of $>10\text{cm}^{-3}$ and an integrated LWC of $>0.001\text{g}\cdot\text{m}^{-3}$ were selected for analysis. Second, only records with ambient temperature warmer than 0°C were chosen to avoid the influence of ice particles. Third, a record was removed if it was not in 5-s or longer consecutive serials. Cloud bases and tops were determined as the lowest and highest heights, respectively, for a continuous layer. Thus, a total of 18 cloud profile detections were performed through the six flights during the observation period, and the total number of warm cloud data records was 2871 (Table 2), including 2368, 233, and 270 records for altostratus (As), small cumulus (Cu), and stratocumulus (Sc) clouds, respectively. The measurements of aerosol particles in and out of clouds can affect cloud formation (Zhang et al., 2011; Hao et al., 2017). To discuss

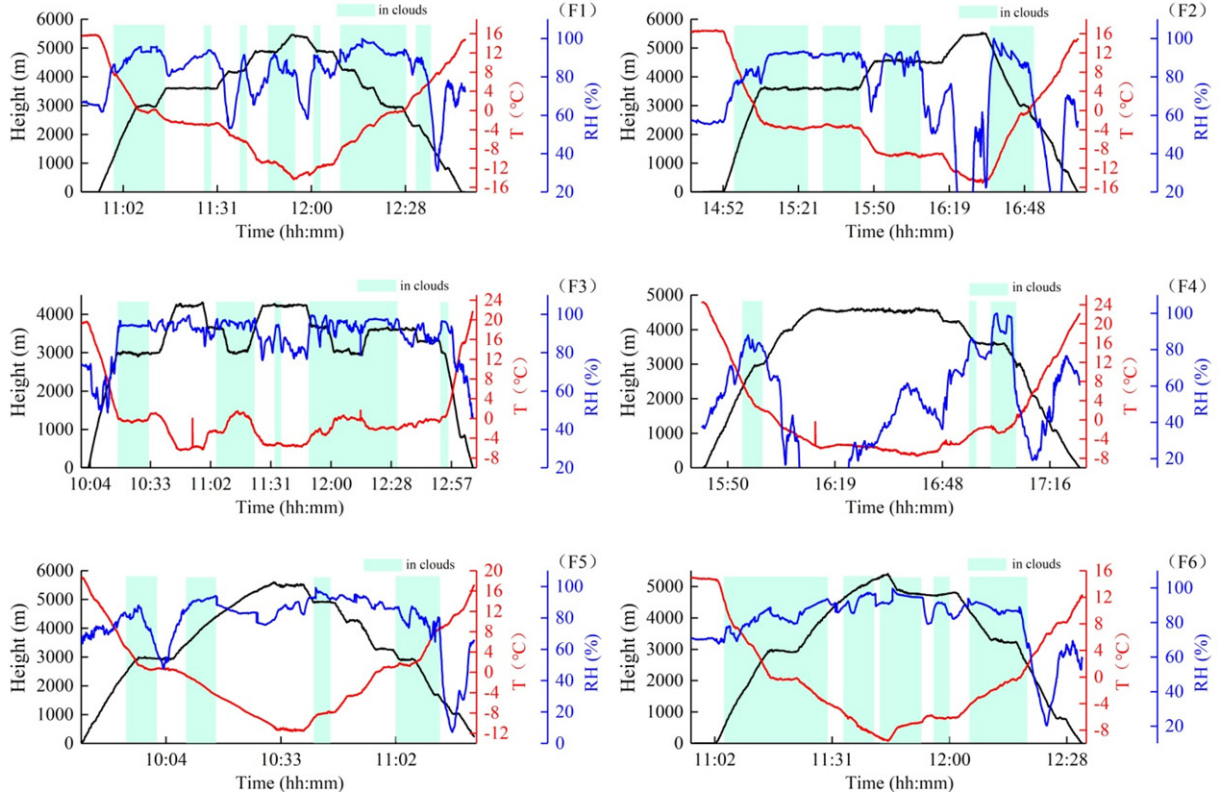


Fig. 2. Time series of flight height (black line), T (red line), and RH (blue line) from the six aircraft measurements. (The light green shades indicate in clouds data.)

aerosol–cloud interactions, some studies have determined the aerosol–cloud relationship by using column aerosol properties (Yuan et al., 2008) or surface aerosol properties (Garrett et al., 2004; Garrett and Zhao, 2006). In this study, based on aircraft observations, aerosol properties at heights approximately 300 m below the cloud base were taken as the aerosol background, and aerosol properties in clouds were calculated as the average value at 200 m above the cloud base, which is more objective because most cloud properties are more affected by aerosol particles near cloud bases than by those at the surface (Zhao et al., 2018).

Flights were conducted across 5 days. Four flights were conducted in the morning, and two flights were conducted in the afternoon. All flights lasted for approximately 2–3 h. The six flights are labelled as F1 (20130508-1), F2 (20130508-2), F3 (20130523), F4 (20130525), F5 (20130526), and F6 (20130528), respectively. Most of the maximum flight heights were above 5000 m, except F3 and F4. The height of the 0 °C layer of the six flights ranged from 2890 to 3540 m. Three types of clouds were observed, including As in four flights, Cu in one flight, and Sc in one flight. Clouds with two or three layers were encountered in most

flights, and cloud bases ranged from 978 to 2825 m. During the six flights, the maximum temperature (approximately 15 °C–25 °C) was observed near the ground. The temperature normally decreased with height, and the lowest temperature was below 0 °C. RH was normally relatively low when the aircraft was outside clouds, with the minimum value ranging from 16 to 66%, and the mean value ranging from 62 to 82%. When the aircraft entered clouds during the flight, RH was always higher with values greater than 90% due to the saturation of water vapour in clouds. Figure 2 shows the time series of the meteorological data (T and RH) with flight height from the six aircraft measurements, and the light green shades indicate in clouds data. The profiles of both aerosol and cloud properties were obtained from these aircraft observations.

The aerosol number concentration (N_a), cloud droplet number concentration (N_c), aerosol ED, and LWC are defined as follows:

$$N_a = \sum N_{i_a} \quad (1)$$

$$N_c = \sum N_{i_c} \quad (2)$$

$$ED = \frac{\sum D_i^3 N(D_i)}{\sum D_i^2 N(D_i)} \quad (3)$$

Table 3. Statistical characteristics of aerosol particles (STP).

Flight	N_a (cm^{-3})					N_{acc} (cm^{-3})					N_{acc} ratio (%)		ED (μm)				
	Max.	Ave.	Md.	25 Pct.	75 Pct.	Max.	Ave.	Md.	25 Pct.	75 Pct.	Ave.	Md.	Max.	Ave.	Md.	25 Pct.	75 Pct.
F1	5840	709	158	85	465	5829	701	153	79	460	94.7	99.8	2.83	0.78	0.38	0.12	1.44
F2	9025	1223	404	150	1549	8970	1205	362	136	1547	94.1	99.8	2.84	0.79	0.43	0.11	1.52
F3	9745	781	636	415	890	9686	755	620	396	862	95.7	99.7	2.86	0.82	0.45	0.26	1.11
F4	7179	1749	1243	488	2751	7116	1714	1226	485	2712	98.5	99.7	2.62	0.61	0.48	0.34	0.66
F5	7461	993	463	121	1100	7426	969	450	117	1080	96	99.4	2.84	0.95	0.62	0.28	1.66
F6	2518	609	483	153	999	2517	596	470	140	985	94.7	99.4	2.85	1.09	0.9	0.21	1.87
Ave.	6962	1011	564	235	1292	6924	990	547	226	1274	95.6	99.6	2.8	0.8	0.5	0.20	1.40

$$\text{LWC} = \sum n_i r_i^3 \rho_w 4\pi/3 \quad (4)$$

where N_{ia} is the aerosol number concentration of the i th bin in Equation (1), and N_{ic} is the cloud droplet number concentration of the i th bin in Equation (2). D_i is the diameter of the i th bin in Equation (3). The r_i is the radius of the i th bin, and ρ_w is the water density in Equation (4).

3. Results and discussion

3.1. Statistical characteristics of aerosol particles and warm clouds

Aerosol particles are usually divided into groups or modes according to the particle size. Given that aerosol measurements were obtained from the PCASP, only particles with diameters of 0.1–3 μm could be detected. Thus, in this study, we discuss the total number concentration measured in the size range of 0.1 to 3 μm (N_a), the number concentrations of accumulation mode particles (N_{acc} , 0.1–1 μm) and coarse mode particles (1–3 μm). Statistical properties, including the maximum and minimum values (marked as Max. and Min.), median values (Md.), and 25 and 75 percentiles (25 Pct. and 75 Pct.) of aerosol particles in the six flights, are shown in Table 3, and Fig. 3 shows the probability distribution function of N_a .

The highest number concentration of aerosol particles (N_a) was observed on 23 May (F3), and the maximum N_a reached 9745 cm^{-3} . Although the maximum mean value of N_a occurred on 25 May (F4), the mean N_a reached 1749 cm^{-3} over the 2-h flight. The lowest value of N_a was observed on 28 May (F6). The maximum and average N_a values were 2518 cm^{-3} and 609 cm^{-3} , respectively, during the flight. It rained slightly before the flight; thus, the precipitation wet removal effect on aerosol particles resulted in a relatively low N_a on that day. The mean value of the maximum N_a , the average value of N_a , and the median value of N_a over the six flights were 6962 cm^{-3} , 1011 cm^{-3} , and 564 cm^{-3} , respectively. The mean and median values of N_a of each flight showed

relatively large differences, because only few large values of N_a increased average N_a values (Fig. 3). Nearly 80% of N_a values were below 1500 cm^{-3} , and the probability of obtaining high values of N_a was small. Of the total aerosol particles measured from the six flights, accumulation mode particles dominated the majority (Table 3). The average value of maximum N_{acc} was 6924 cm^{-3} , and the ratio of N_{acc} to N_a ranged from 94.1% to 98.5% with an average value of 95.6%. Coarse mode particles were very few. Similar results were reported by Hao et al. (2017) who reported that N_{acc} (ranging from 0.1 to 1.0 μm) accounted for more than 95% of N_a (0.1–3 μm) during August 2014 over Anhui Province, east China. As shown in Table 3, most EDs of aerosol particles were less than 1 μm . In summary, the mean magnitude of N_a over the observation area was 10^3 cm^{-3} , the mean ED of aerosol particles over the six observation flights was 0.8 μm , and the average value of median ED was 0.5 μm . We compared these results with other aircraft measurements conducted in China, such as in Beijing (Zhang et al., 2006, 2009, 2011; Liu et al., 2009), Hebei (Sun et al., 2013), and Shanxi (Li et al., 2015a, 2015b), and found that the aerosol particle number concentration and particle size were comparable to the previous studies, but specific values differed due to different weather backgrounds, observation time periods, and locations.

Table 2 shows the microphysical properties of warm clouds and aerosol particles below and in clouds. The cloud base height (AGL) from the six aircraft measurements ranged from 1000 to 2800 m, and the top of clouds ranged from 3000 to 3300 m. The total number of warm cloud data records was 2871 according to the cloud definition in this study. The value of N_a in the cloud was always lower than that below the cloud, possibly reflecting the process of CCN activation. The aerosol activation ratio ($[N_a \text{ under cloud} - N_a \text{ in cloud}]/N_a \text{ under cloud}$) ranged from 0.14 to 0.91 with an average value of 0.46, suggesting that a high amount of aerosol particles were converted to cloud droplets under high humidity conditions in clouds. The maximum and average values of measured N_c ranged from 147 to 311 cm^{-3} and 51

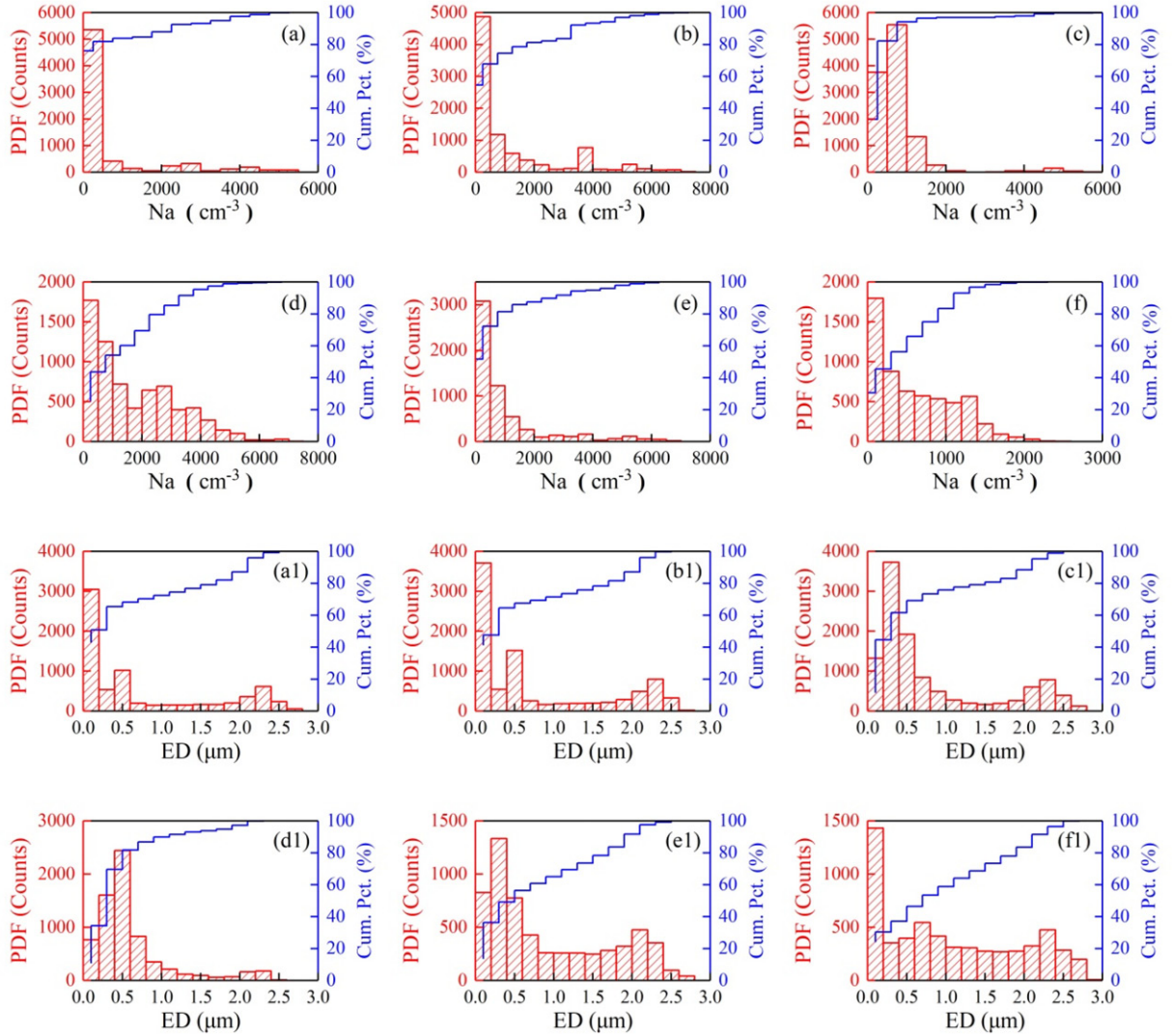


Fig. 3. The probability distribution function of N_a and ED of aerosol particles from the six aircraft measurements.

to 157 cm^{-3} , respectively. The maximum cloud droplet size ranged from 13.5 to $28.9\text{ }\mu\text{m}$, and the average value ranged from 5.8 to $13.1\text{ }\mu\text{m}$ based on CDP measurements. The maximum LWC of warm clouds ranged from 0.06 to $0.53\text{ g}\cdot\text{m}^{-3}$, and the average value of LWC was $0.05\text{ g}\cdot\text{m}^{-3}$.

3.2. Vertical profiles of N_a and ED of aerosol particles

Figure 4 shows the mean vertical profiles of N_a (a) and ED (b) and vertical profiles of N_a (c) and ED (d) under relatively clean and polluted weather conditions, respectively, measured using the PCASP during the six flights. As shown in Fig. 4(a), large amounts of aerosol particles were constrained to the lower layer of the atmosphere (below 1000 m), and this amount decreased sharply with height, which might be

attributed to the effects of the atmospheric boundary layer. The atmospheric boundary layer often hinders the vertical transmission and dispersion of aerosol and other pollutants. Large amounts of aerosol particles might be constrained below the boundary layer and formed an accumulation area. The maximum and average values of N_a below 1500 m were approximately 3500 and 3000 cm^{-3} with an average standard deviation of 1700 cm^{-3} . These results are considerably lower than the vertical profiles of aerosol particles obtained through aircraft observations in other regions, such as in Beijing from July to September 2008 (Zhang et al., 2011) (the average N_a below 1 km was approximately 6600 cm^{-3}). Furthermore, the results of this study are a little lower than those reported for September 2015 over Hebei Province (Zhao et al., 2018) and comparable to those reported for July and August 2013 in Shanxi (Li et al., 2015a, 2015b). In

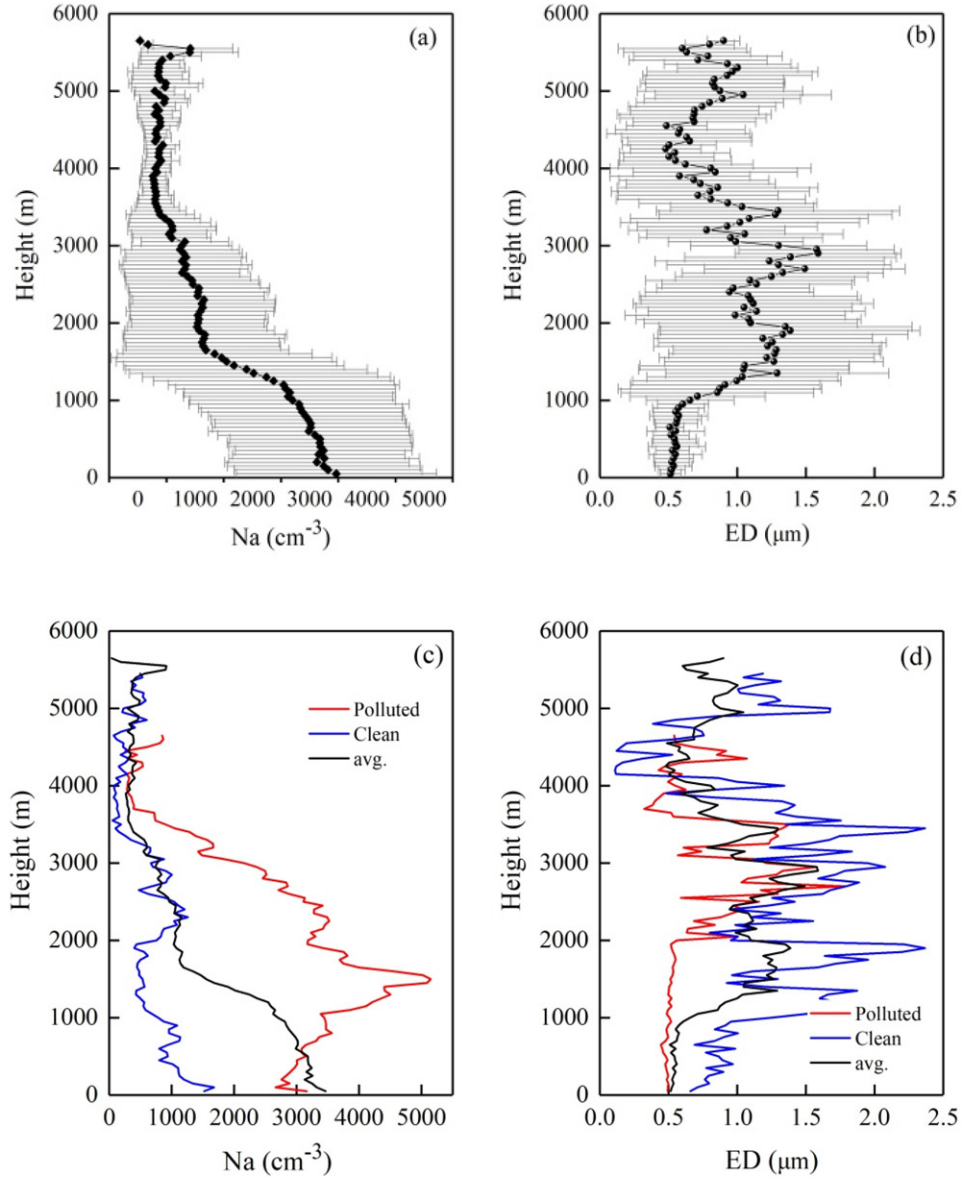


Fig. 4. Vertical profiles of averaged N_a (a) and ED (b) with error bars (standard deviation), and the vertical profiles of N_a (c), and ED (d) in relatively clean and polluted weather conditions.

addition, the concentration of aerosol particles with sizes ranging from 10 nm to 0.1 μm could reach 10^4 cm^{-3} near the surface under clear sky conditions in the North China Plain (Wang et al., 2012; Zhang et al., 2014; Li et al., 2015a, 2017a). Above 1000 m, there was a sharp decrease in N_a , and the average N_a decreased to 1100 cm^{-3} at heights ranging from 1000 m to 3500 m. The average N_a further decreased to approximately 400 cm^{-3} above the height of 3500 m. The vertical distribution of the particle size showed different features (Fig. 4(b)). The EDs of particles were very small with an average value of 0.55 μm in the lower layer of the atmosphere, particularly below 1000 m, and the values

gradually increased at heights ranging from 1000 to 3500 m and then decreased above the height of 3500 m. For example, the averaged ED was 1.0 and 0.7 μm at heights ranging from 1000 to 3500 m and from 3500 to 5500 m, respectively. The results of ED and N_a vertical profiles suggested that the majority of aerosol particles within the lower layer (below 1000 m) were fine particles with ED less than 0.5 μm , indicating that aerosol particles mainly originated from local and regional surface emissions, which might include both direct emissions of particles and particles originate through gas-to-particle formation. Particles with ED larger than 0.5 μm were distributed at higher altitudes,

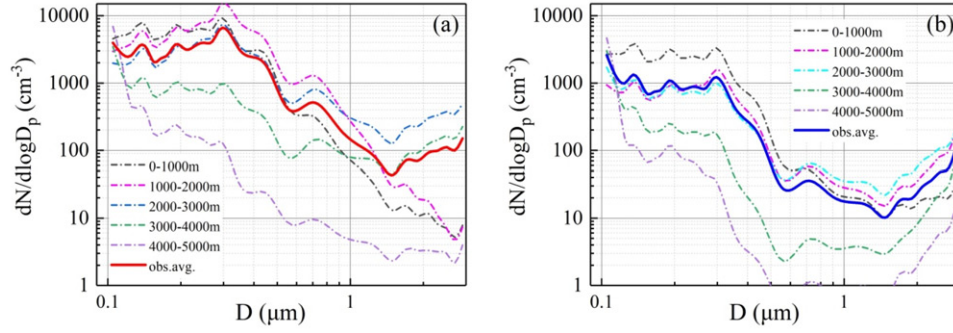


Fig. 5. Size distributions of aerosol particles on relatively polluted (a) and clean (b) weather conditions. The different coloured dashed lines are the average particle size distributions in different height ranges (0–1000 m, 1000–2000 m, 2000–3000 m, 3000–4000 m, 4000–5000 m), and the thick solid red and blue lines show the average particle size distributions from the whole flights of F4 (a) and F6 (b), relatively.

suggesting that these particles were transported to the region from afar or because of the aging of aerosol particles. Furthermore, clouds present in the high layer may affect the particle size because of aerosol hygroscopicity under high humidity conditions. Figure 4(c and d) shows the vertical profiles of N_a and ED under a relatively low and high aerosol loading conditions. Under polluted weather conditions (F4), N_a was very high with a maximum value of 5000 cm^{-3} at a height of 1500 m. Under a relatively clean weather conditions (F6), N_a was considerably lower, and even the maximum value of N_a was less than 1700 cm^{-3} . However, the vertical profiles of the particle size showed opposite properties. The ED of particles was considerably small under polluted weather conditions (F4) and relatively large under clean weather conditions (F6), indicating that fine particles contributed greatly to air pollution.

Figure 5 shows the size distributions of aerosol particles on relatively polluted (a) and clean (b) weather conditions. There were a lot of aerosol particles especially small particles on relatively polluted condition (Fig. 5(a)). For example, the magnitude of N_a with particle size less than $0.5\text{ }\mu\text{m}$ was above 10^3 cm^{-3} , and the magnitude of N_a with size from 0.5 to $1\text{ }\mu\text{m}$ is 10^2 cm^{-3} . In addition, the particle number concentration was particularly large at low layer (below 3000 m height), and the peak value appeared at the particle size of $0.3\text{ }\mu\text{m}$. In contrast, the N_a was much lower on the relatively clean condition (Fig. 5(b)), and most of particle concentration values were lower than 10^3 cm^{-3} . The size distribution of aerosol particles also suggested that the large amount fine particles at low layer had a great impact on air pollution.

3.3. Vertical and horizontal distribution of N_a , N_c , and LWC

Data with cloud penetration during the aircraft ascent and descent periods of the six flights were selected to analyse the

vertical structural characteristics of aerosols and clouds (Fig. 6). As shown in Fig. 6, when the aircraft passed through clouds, N_a generally decreased significantly, and N_a was negatively correlated with the number concentration of cloud particles (N_c), suggesting the process of aerosol activation in clouds. The LWC of clouds and N_c showed the same variation characteristics, and the large value area of LWC was generally located in the middle layer of clouds. Furthermore, the vertical distribution of aerosols and clouds indicated that aerosol concentrations were fairly well-mixed up to cloud levels from 1000 to 4000 m. Similarly, the ED of aerosols was fairly constant up to 1000 m and then increased in cloudy layers presumably due to aerosol hydration and hygroscopic growth (Fig. 4(b)). The high aerosol number concentration could significantly affect cloud properties within the atmospheric boundary layer (Fig. 6(a, b, e, and f)). However, clouds with bases above the atmospheric boundary layer (approximately 900–1200 m over the time periods of the six flights) were less influenced by high aerosol concentrations near the ground surface (Fig. 6(c and d)). Thus, it is better to choose aerosol data near the cloud base to study aerosol–cloud interactions (Zhao et al., 2018), and this is one of the advantages of the aircraft observation.

3.4. Horizontal distribution of N_a , N_c , and LWC

Figure 7 presents the N_a , N_c , and LWC in two short time series, and the airplane was at a stable height in these two time segments. The N_a , N_c , and LWC showed non-uniform characteristics in the horizontal direction. Similarly, when the aircraft started going into the clouds, N_a normally decreased. When the aircraft flew through the cloud layer and out of clouds, N_a usually showed an increased trend. During the period of cloud penetration, different features were observed at different locations of a cloud. Overall, N_a and N_c showed a negative correlation through the whole time segments (Fig. 7).

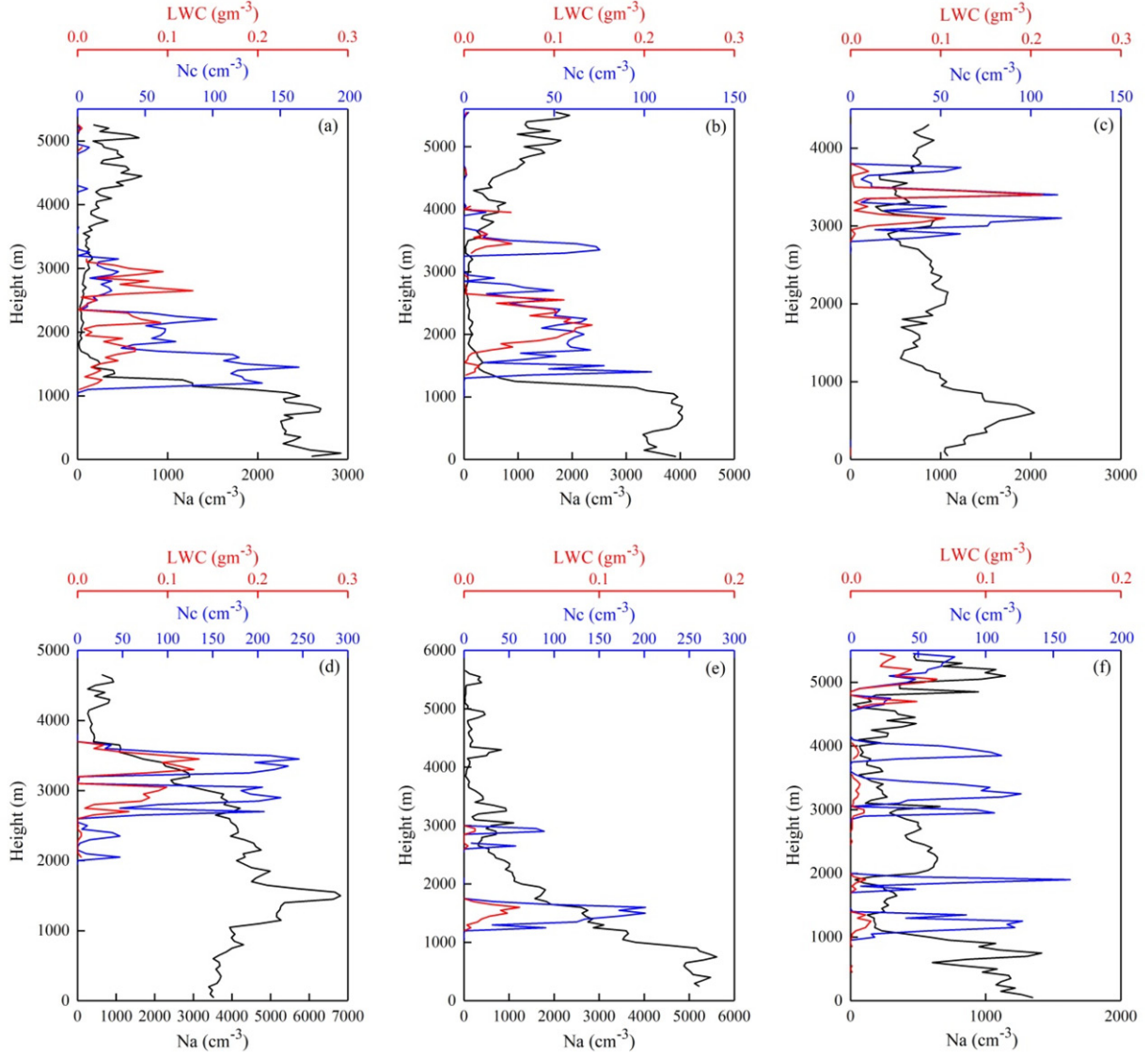


Fig. 6. Vertical distribution of N_a , N_c , and LWC (a, b, and f were from the airplane ascent period and c, d, and e were from the descent period).

This phenomenon indicates that favourable humidity conditions in clouds are beneficial for the conversion process of aerosol particles to cloud droplets. The LWC of clouds and N_c showed the same variation in characteristics and a positive correlation.

3.5. Cloud formation under different aerosol conditions

To analyse the cloud formation process under different weather conditions, two cases were selected in this study. One case was the flight observation on May 23, which was a less polluted case with a relatively low aerosol concentration (Case 1, F3, Fig. 8). In this case, the average

N_a during the whole flight was 781 cm^{-3} , and N_a below the cloud base was 982 cm^{-3} . The second case was the flight observation on May 25, which was a more polluted case with a high aerosol concentration (Case 2, F4, Fig. 9). In this case, the average N_a during the whole flight was approximately 1750 cm^{-3} , and N_a below the cloud base was 4573 cm^{-3} .

Figures 8 and 9 show the relationship between N_c and D_c under different LWC conditions for the aforementioned two cases. The cloud droplet concentration (N_c) and the diameter of the cloud droplet increased with LWC, especially when LWC was higher than $0.1\text{ g}\cdot\text{m}^{-3}$, based on these two cases. When the aerosol concentration was relatively low (Case 1), the magnitude of N_c

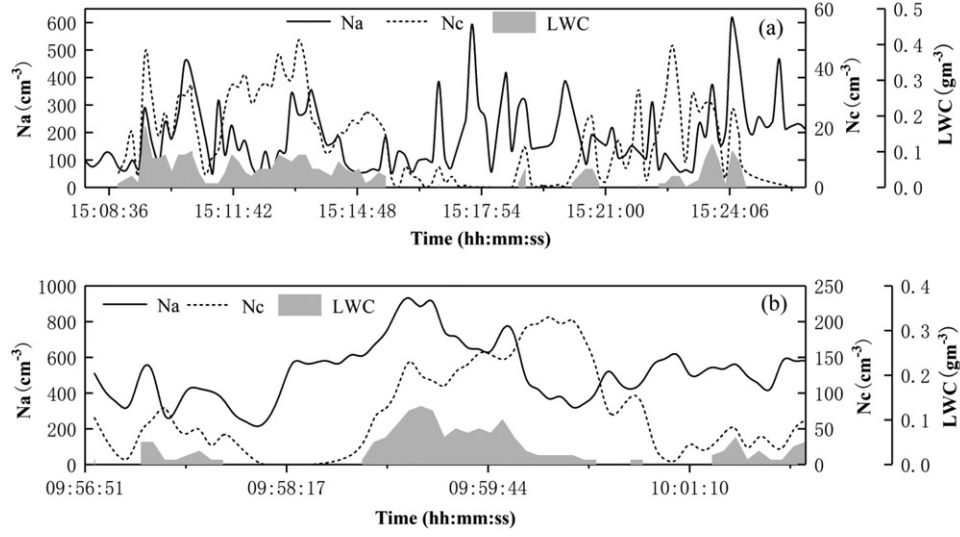


Fig. 7. Horizontal distribution of N_a , N_c , and LWC.

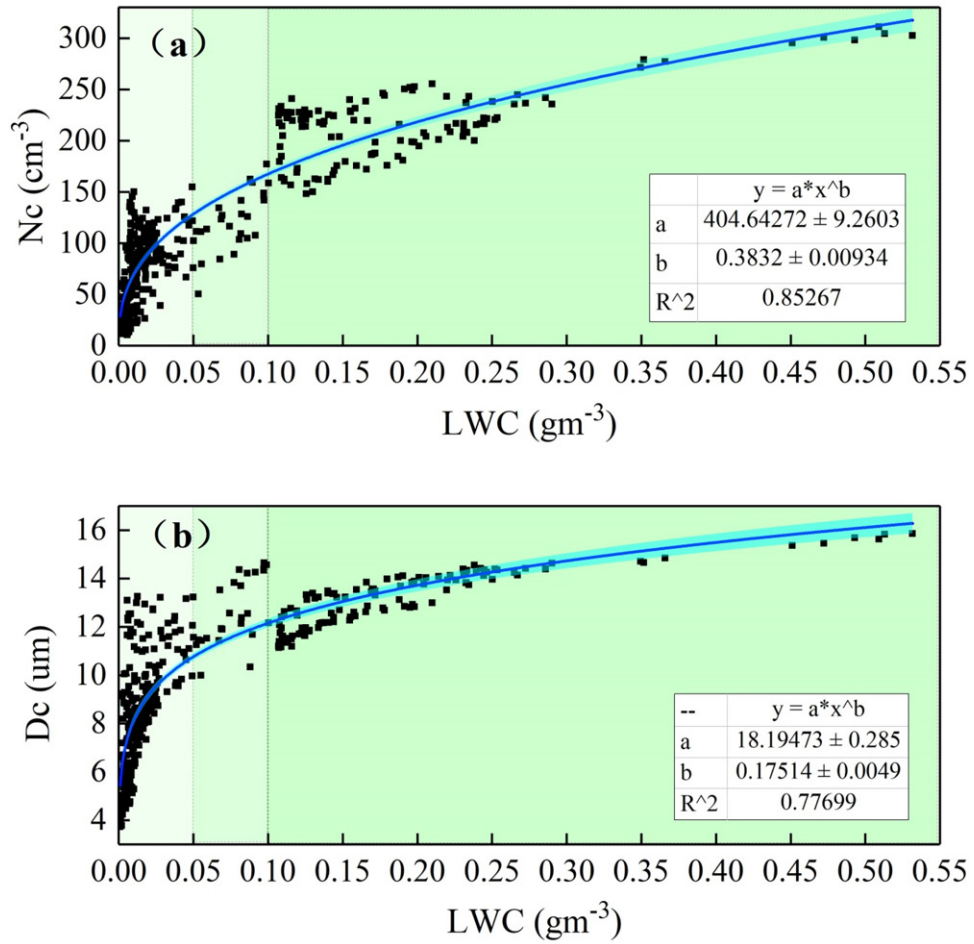


Fig. 8. Relationships between N_c (a) and D_c (b) at different LWC ranges under low aerosol loading conditions.

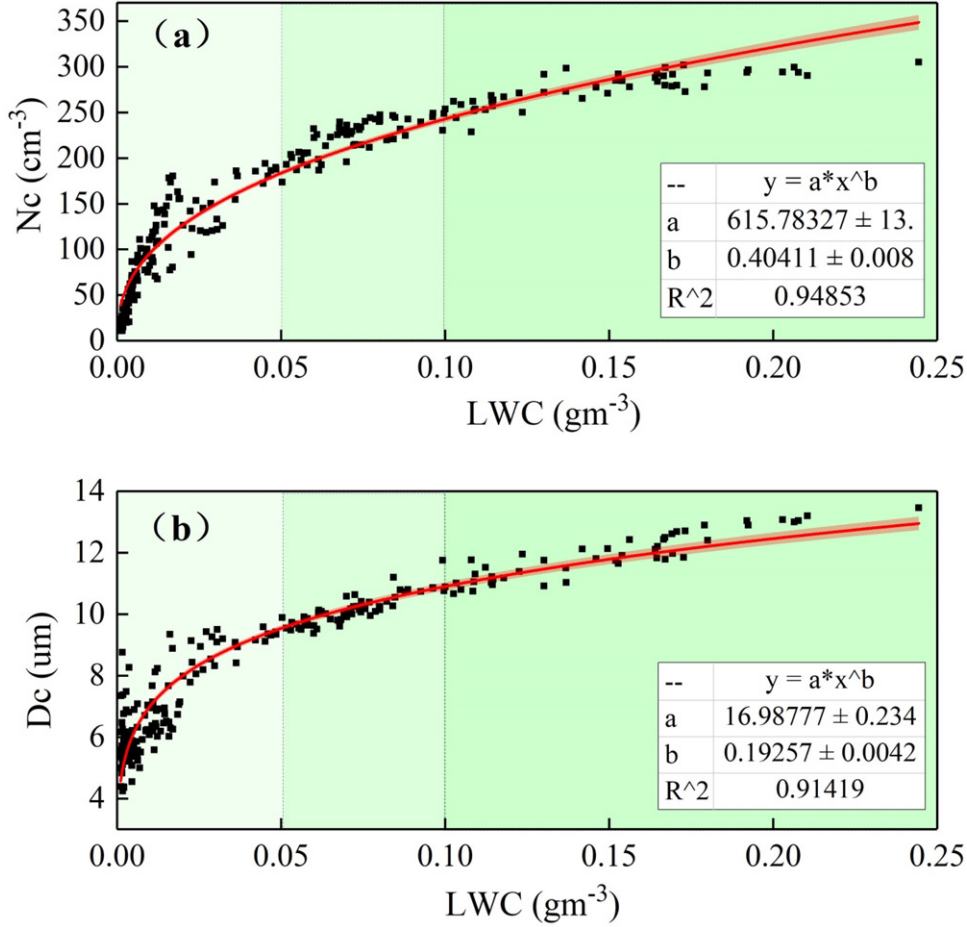


Fig. 9. Relationships between N_c (a) and D_c (b) at different LWC ranges under high aerosol loading conditions.

increased from 10^1 cm^{-3} to 10^2 cm^{-3} , and the minimum and maximum value of N_c were 11.4 cm^{-3} and 154 cm^{-3} , respectively with LWC values ranging from 0 to $0.05 \text{ g} \cdot \text{m}^{-3}$. When LWC ranged from 0.1 to $0.5 \text{ g} \cdot \text{m}^{-3}$, the maximum value of N_c reached approximately 180 cm^{-3} . When LWC was $>0.1 \text{ g} \cdot \text{m}^{-3}$, the maximum N_c increased to 300 cm^{-3} , but the majority were concentrated within 240 cm^{-3} . LWC and N_c showed a good correlation with the correlation coefficient (r^2) reaching 0.85 (Fig. 8(a)). Meanwhile, with a certain number of cloud droplets, the increase in LWC led to an increase in the size of cloud droplets rather than in the number of cloud droplets. The size of cloud droplets increased rapidly as the value of LWC increased; D_c increased from 3.7 to $16 \mu\text{m}$ (the average D_c was $10.1 \mu\text{m}$), and both LWC and D_c showed a good correlation, especially at a high LWC, with the correlation coefficient (r^2) reaching 0.78 (Fig. 8(b)).

When the amount of aerosol particles was considerably higher (Case 2), more particles could potentially be activated to become cloud droplets, and N_c in the three LWC ranges was larger than that in Case 1. When LWC

ranged from 0 to $0.05 \text{ g} \cdot \text{m}^{-3}$, N_c increased to 200 cm^{-3} . When LWC ranged from 0.05 to $0.1 \text{ g} \cdot \text{m}^{-3}$, the maximum value of N_c reached approximately 260 cm^{-3} . The maximum N_c increased to more than 300 cm^{-3} when LWC was more than $0.1 \text{ g} \cdot \text{m}^{-3}$, and most of the N_c values were higher than 250 cm^{-3} . Furthermore, N_c was strongly correlated to the abundance of LWC. For example, the correlation coefficient (r^2) was 0.95 between LWC and N_c (Fig. 9(a)), which is higher than the value obtained when the aerosol concentration was relatively low (Case 1). However, because a certain amount of liquid water was shared by many particles, the size of cloud droplets was limited. Therefore, many small cloud droplets were formed in clouds. For example, the size of cloud droplets in Case 2 increased slowly because of a high aerosol concentration with certain LWC (LWC ranges of <0.05 , $0.05\text{--}0.1$, and $>0.1 \text{ g} \cdot \text{m}^{-3}$); D_c increased from 4.2 to $13 \mu\text{m}$ (the average D_c was 8.5) when the value of LWC increased. The D_c and LWC also showed a good correlation with the correlation coefficient (r^2) being 0.91 (Fig. 9(b)).

The results of both these cases indicate that background aerosol concentration can affect the formation of cloud droplets. The larger the number of aerosol particles, the more cloud droplets formed, and the cloud droplets sizes were smaller at a certain LWC range, which is consistent with Twomey theory (Twomey, 1977).

3.6. N_c and D_c under different LWC conditions

To investigate the relationship between N_c and D_c under different LWC conditions in detail, we calculated the mean values of N_c and D_c in three LWC ranges (<0.05 , $0.05\text{--}0.1$, and $>0.1\text{ g}\cdot\text{m}^{-3}$) of each flight; the results are shown in Table 4 and Fig. 10. At a certain range of LWC, if the cloud droplet number concentration was high, the droplet size was relatively small. Both of the two parameters showed negative correlations. We selected two cases from F1 and F2 on a same day, May 8 (data

Table 4. Mean values of N_c and D_c under different LWCs for each flight (N_c : cm^{-3} , D_c : μm , LWC: $\text{g}\cdot\text{m}^{-3}$, STP).

Flight	<0.05		$0.05\text{--}0.1$		>0.1	
	N_c	D_c	N_c	D_c	N_c	D_c
F1	72	9.91	50.6	16.67	38.6	21
F2	43.3	9.53	53.8	16.43	62.8	17.68
F3	70.1	7.99	116.3	12.56	215.6	13.12
F4	83.6	6.68	219.8	10.13	276.4	11.9
F5	122.4	7.9	116.3	13.55	164.2	15.37
F6	99.8	5.61	205.9	9.66		

of each case was from one individual cloud), for analysis (Fig. 10).

Figure 10 shows that the average cloud droplet diameters were strongly related to the values of LWC and strongly varied with the number of aerosol particles in the same cloud. D_c and N_c presented a negative correlation. Under certain LWC conditions, especially at a high LWC, D_c showed a negative exponential growth with a decline in N_c . Their correlation coefficient was approximately 0.8 at LWCs ranging from 0.05 to $0.1\text{ g}\cdot\text{m}^{-3}$ and approximately 0.7 at LWCs larger than $>0.1\text{ g}\cdot\text{m}^{-3}$ in the two cases, respectively.

3.7. Number size distribution of cloud droplets in warm clouds

Cloud droplets have a variety of particle sizes. The distribution of the cloud droplet number concentration with different droplet sizes, called as the droplet spectral distribution, is an important physical quantity used to describe cloud droplet distribution features. Clouds in different atmospheric environments, different heights, and different stages of development have different spectral distributions. Figure 11 shows the cloud droplet size distributions of all detected clouds from the six flights (different coloured dashed lines), and the thick deep purple line shows the mean state of the cloud droplet size distribution. In this study, considering that many small cloud droplets were observed in the experiment, a multi-lognormal distribution function (Equation 5) was used to fit the mean

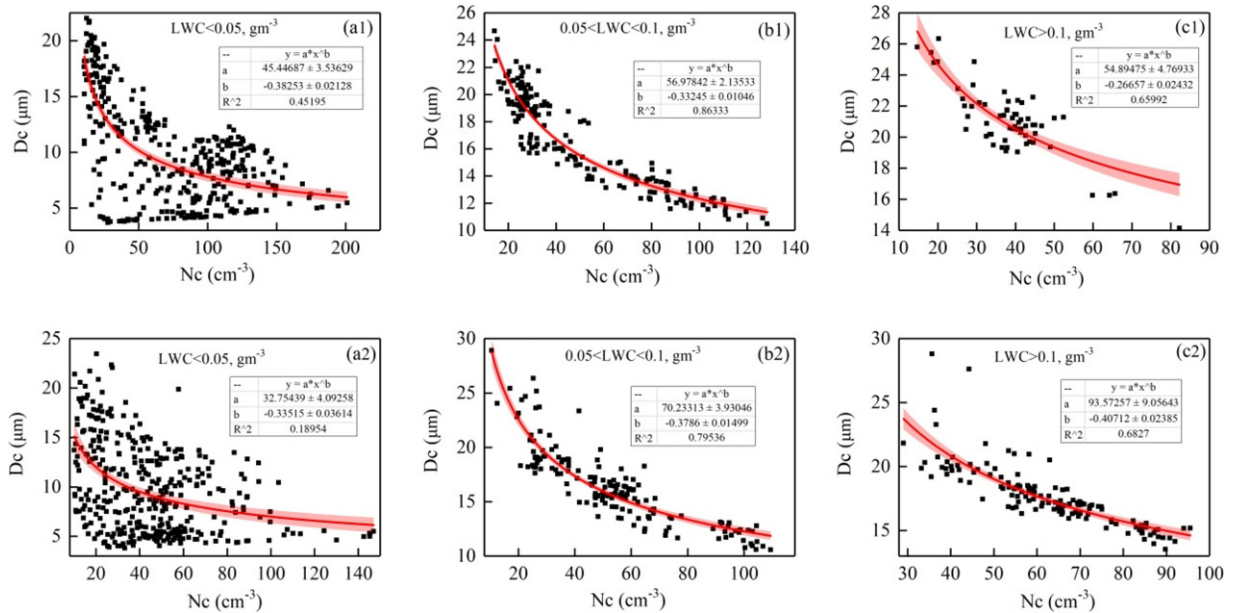


Fig. 10. Two cases of N_c and D_c under three LWC ranges of <0.05 , $0.05\text{--}0.1$, and $>0.1\text{ g}\cdot\text{m}^{-3}$ in the same cloud system. (Case 1: a1, b1, and c1; Case 2: a2, b2, and c2).

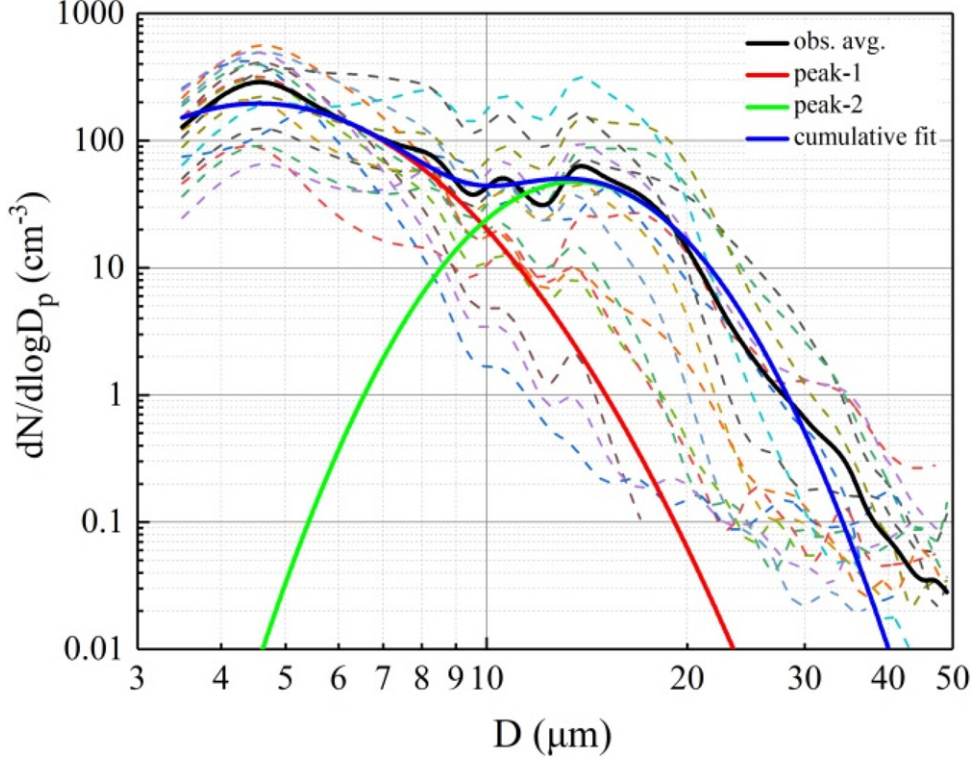


Fig. 11. The cloud droplet size distributions of all detected clouds from six flights (the different coloured dashed lines are droplet size distributions from each cloud system, the thick deep purple line shows the mean state of the cloud droplet size distribution, and red and green lines show the lognormal fits of two modes to the mean cloud droplet spectrum).

number size distribution of all measured cloud droplets of warm clouds.

$$\frac{dN(D)}{d\log(D_p)} = \sum_{i=1}^n \frac{N_i}{\sqrt{2\pi} \log(\sigma_{g,i})} \exp \left[-\frac{(\log(D_p) - \log(D_{g,i}))^2}{2(\log \sigma_{g,i})^2} \right] \quad (5)$$

where n is the number of modes per size distribution for the best fit, D_p is the peak droplet diameter, N_i is the droplet number in mode i , $D_{g,i}$ is the geometric mean diameter of the mode i , and $\sigma_{g,i}$ is the standard deviation of mode i . The red and green lines in Fig. 9 show the lognormal fits of the two modes to the mean cloud droplet spectrum, and parameters that characterise the number size distributions of the two modes are listed in Table 5. Normally, the cloud droplet spectrum showed one or two peaks. The first N_c peak was observed at the size of $4.56 \mu\text{m}$ in diameters for Mode I, and the second peak was observed at the size of $13.6 \mu\text{m}$ in diameters for Mode II. According to the two modal lognormal distribution fits, small droplets (Mode I) dominated the total cloud droplet number concentration, and a bimodal distribution best described the cloud droplet spectrum of warm clouds (the blue fit line in Fig. 11). The fitting

Table 5. Parameters characterising the number size distributions of warm cloud droplets (N_c : cm^{-3} , D_g : μm).

	Parameter	Value	SD	Related coefficient
Mode I	N_c	78.6	17.7	0.79
	D_g	4.56	0.34	0.46
	$\log \sigma$	0.16	0.05	0.83
Mode II	N_c	13.6	8.4	0.71
	D_g	13.6	2.41	0.48
	$\log \sigma$	0.11	0.07	0.65

parameters can provide a scientific reference for the establishment and improvement of a cloud parametric numerical model.

4. Conclusions

Based on six flights measurements of aerosol and clouds during a spring aircraft campaign over the eastern Loess Plateau of China, this study analysed the aerosol and cloud droplet distribution characteristics and the relationship between aerosol loading and microphysical properties of warm clouds.

The mean magnitude of the measured aerosol number concentration was 10^3 cm^{-3} (1011 cm^{-3}), and accumulation mode particles dominated. Most ED of aerosol particles was less than $1 \mu\text{m}$, with a mean value of $0.8 \mu\text{m}$. The vertical profiles of N_a and ED showed that large amounts of fine aerosol particles were constrained to the lower layer (below 1000 m) with particle EDs less than $0.5 \mu\text{m}$. Clear differences were demonstrated through a detailed analysis on the N_a and ED vertical profiles and the aerosol particles size distribution on relatively clean and polluted weather conditions. The aerosol particle size distributions further showed that more particles concentrated within smaller size ranges under polluted weather condition. The results indicated that fine particles at the lower layer contributed greatly to air pollution.

Eighteen cloud detections with 2871 warm cloud data records during the observation period were analysed, and aerosol–cloud interactions in different weather backgrounds were discussed. The statistical properties of cloud droplets number concentration and effective radius were showed in this study, which were two critical parameters in calculating cloud albedo and estimating the direct effects of clouds. The N_a values in the cloud base were always smaller than that below the clouds, and the average aerosol activation ratio was 0.46. The vertical and horizontal distribution of N_a , N_c , and LWC indicated N_a was normally negatively correlated with N_c either in the vertical or horizontal direction. The LWC of clouds and N_c showed a positive correlation, and the large value distribution area of LWC was generally located in the middle part of clouds in the vertical direction.

Two cases were selected to analyse the cloud formation process under different aerosol concentration conditions. Under high aerosol loading conditions, a large number of small cloud droplets were formed in clouds. By contrast, under low aerosol loading conditions, a relatively small number of cloud droplets were formed with larger size. The relationship between LWC and aerosol particles/cloud droplets was studied. The increase in LWC was related to an increase in the size of cloud droplets rather than the number of cloud droplets. At a certain range of LWC, D_c presented a negative exponential growth with a decline in N_c . The results indicate that the number of aerosol particles has important impacts on microphysical characters of clouds. And the LWC in clouds is also an important factor when discussing the aerosol–cloud interaction, which closely correlated to the average size of cloud droplets.

The cloud droplet size distributions of all detected warm clouds from the six flights were obtained, and a bimodal lognormal distribution function was used to fit the average cloud droplet spectrum. Furthermore, the characteristic parameters of warm clouds were reported

in this study. The parameters are required in studying the radiation transfer and the microphysical processes, and also can provide real observation results for the cloud parameterization in numerical simulation.

Disclosure statement

No potential conflict of interest was reported by the authors.

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