Aircraft Measurements of the Aerosol Spatial Distribution and Relation with Clouds over Eastern China

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ABSTRACT

To investigate the spectral and spatial distribution characteristics of aerosol particles over eastern China, this study conducted a set of aircraft measurements during August 12–28, 2014, over Anhui province, China. The aerosol number concentration and size distributions from five flights as well as the cloud and meteorological parameters were analyzed. In Anhui province, the average number concentration of aerosol particles in the size range of 0.1–3.0 µm was 481 ± 199 cm⁻³, and accumulation mode particles accounted for more than 95% of the total aerosol particles. Most of the aerosol particles were concentrated in the layer below 1000 m, where the number concentration decreased with the altitude, except in the presence of thermal inversion layers (TILs). The TILs prevented the vertical transport of aerosol particles, and led to a higher number concentration in the boundary layer. A large fraction of aerosol particles was removed when clouds were present, and the removed in-cloud aerosols led to an increase in cloud droplet concentrations for the size range of 3.5–10.0 µm. Our results are valuable for understanding the spatial distribution of aerosol particles and their interactions with clouds.

Keywords: Aircraft observation; Aerosol spatial distribution; Aerosol removal in clouds.

INTRODUCTION

Aerosol particles can reduce atmospheric visibility, impede air quality and harm human health (Cao et al., 2013; Liu et al., 2014; Gupta et al., 2016; Liang et al., 2016; Syu et al., 2016; Khaefi et al., 2017); they can also affect the Earth’s climate directly by scattering and absorbing solar and terrestrial radiation, and indirectly by serving as cloud condensation nuclei (CCN) and ice nuclei, thereby influencing the microphysical and radiative properties of clouds (Mecornick and Ludwig, 1967; Twomey, 1974, 1977; Albrecht, 1989; Charlson et al., 1992; Ramanathan et al., 2001; IPCC, 2013). Aerosols in the boundary layer have been studied extensively during the past decades (e.g., Cao et al., 2003; Hansen et al., 2003; Koulouri et al., 2008; Alam et al., 2012; Xin et al., 2015; Khamkaew et al., 2016; Xu et al., 2017), but their properties in the free atmosphere are still poorly understood; in particular, large uncertainties exist regarding the vertical distribution of aerosols in the lower troposphere (Li et al., 2015b). Aerosol particles appearing at the cloud level can affect cloud properties and precipitation directly, and the complex roles of aerosol vertical distributions in the climate system indicate that additional observations are required to verify climate simulations (Schwartz et al., 2014; Vuolo et al., 2014).

Aircraft observation has a unique advantage in understanding the vertical and horizontal distribution of atmospheric aerosols and clouds because of its high temporal and spatial resolution (Clarke et al., 1998; Bahreini et al., 2003; Schnitzhofer et al., 2009), and it is also an important tool to study aerosol–cloud interactions (Lu et al., 2008; Rosenfeld et al., 2008). Aircraft field observations have been performed over multiple regions including marine, desert, mountain, rural and urban environments (Fouquart et al., 1987; Petzold et al., 2002; Maring et al., 2003; Taubman et al., 2006; Capes et al., 2008; Reid et al., 2008; Perring et al., 2015).

During the past decades, extensive airborne measurements of aerosol properties have been conducted in the northern part of China, including Shenyang, northeast China (e.g., Dickerson et al., 2007), the North China Plain (e.g., Zhang et al., 2006; Liu et al., 2009; Yin et al., 2009; Zhang et al., ...
2009; Sun et al., 2013) and the Loess Plateau (e.g., Li et al., 2015a, b). Using aircraft optical spectrometer probes in addition to Weather Research and Forecasting tracer and Moderate Resolution Imaging Spectroradiometer data, Zhang et al. (2006) provided the origins and spatial distribution of aerosol particles over Beijing, China. Their results demonstrated that a strong vertical gradient of aerosol concentrations existed around the city of Beijing, and particles in the size range of 0.1–1.0 µm dominated the whole particle population.

The number concentration of aerosol particles in the range of 0.01–20 µm, CCN and aerosol optical properties were measured over the Loess Plateau in Shanxi, China (Li et al., 2015a, b); the results revealed that fine particles constituted the majority of the particles below 2 km, and that larger aerosol particles were more abundant above 2 km. Zhang et al. (2011) discussed aerosol–cloud interactions over highly polluted areas of Beijing by using data from seven aircraft flights. They reported that the cloud effective radius was sensitive to the aerosol number concentration and liquid water content.

Over the past three decades, the increase in industrial emissions resulting from the rapid economic development of megacities in eastern China, such as Shanghai and Nanjing, has impeded the local and regional air quality. Extensive ground-based studies of atmospheric pollutants have been conducted in this region, whereas little is known about the vertical distribution of aerosol properties. In this study, aircraft observations were conducted during August 12–28, 2014, over Anhui Province in Eastern China to measure the aerosol spatial distributions and relation with clouds. Data obtained from five flights were selected to analyze the spatial distribution of aerosol properties and the relationship between aerosols and cloud droplets during summer.

The remainder of manuscript is organized as follows. Section 2 introduces the instruments and methodology. The measured results and discussions for the vertical properties of aerosols and the relations with clouds are presented in Section 3. Section 4 provides the conclusions.

INSTRUMENTATION AND METHODOLOGY

**Instrumentation**

The aircraft used in the experiment was the Cheyenne-III from the Weather Modification Office of Hebei Province, its maximum flight height with equipped instruments was 8 km, and cruising speed was 360–400 km h⁻¹. The aircraft was equipped with several instruments including a passive cavity aerosol spectrometer probe (PCASP-100X), a forward scattering spectrometer probe (FSSP-ER), and a two-dimensional cloud (2D-C) probe, all produced by DMT Co., USA. The PCASP-100X probe, along with 15 variable-sized bins, was used for measuring the number concentration and size distribution of aerosol particles ranging from 0.1 to 3.0 µm. The frequency and sample flow volume were set to 1 Hz and 1 cm³ s⁻¹, respectively. To minimize aerosol losses, the sample inlet was placed at the front of the probe. The intake tube cone provides a flow deceleration ratio of 10:1 to kinetically match the inlet needle flow of 8–10 m s⁻¹. A needle with an inner diameter of 0.5 mm is placed in the sample inlet of the sample cavity. The total transit time in the inlet section is a few hundreds of a second (Sun et al., 2013). The FSSP-ER, with 15 equal-sized bins (3.0-µm interval), is used to measure cloud particles with diameters ranging from 2.0 µm to 47.0 µm, and the 2D-C probe is used for two-dimensional images of droplets and raindrops. The ambient meteorological parameters, cloud liquid water content (LWC), and position of the aircraft were measured using a meteorological package, King LWC probe, and global positioning system device, respectively. Before the measurements were performed, the aerosol and cloud probes were calibrated using polystyrene latex spheres. In this paper, all the concentrations of aerosols and clouds are shown in terms of standard temperature and pressure conditions.

**Flight Description**

The observations were conducted over Anhui Province located in Eastern China (Fig. 1). The main industries in Anhui are coal mines, power plants, petrochemical industries, metallurgy plants and mechanical industries, leading to increased industrial emissions. In addition, a population of 69.49 million and transport sector can also increase pollutants in Anhui Province. Five flights were conducted during August 12–28, 2014, and the flight trajectories are depicted in Fig. 1. In these flights, the aircraft took off from Bengbu airport (117.34°E, 32.93°N), which is in the north of the province, and then climbed southward to detect aerosol vertical profiles. To measure the horizontal variation of aerosols in clear-sky areas and in clouds, the aircraft flew at several altitudes around Anhui and continually descended back to Bengbu.

The flights were conducted across 4 days, and the flight details are listed in Table 1. All the flights were carried out in the afternoons and lasted for 1–2.5 h. The clouds were measured in each flight; in flight 1–4 (f1–f4), only one type of cloud was encountered, and two types of clouds were observed in flight 5 (f5). According to the synoptic analysis, these 4 days were influenced by different weather systems.

**Methodology**

The microphysical parameters of aerosols and clouds are statistically calculated using the following formulas:

Number concentration \(N = \sum n_i\) (1)

Aerosol mean diameter \(MD = \sum (d_i/n_i)N\) (2)

where \(n_i\) is the particle number concentration of the \(i\)th bin, and \(d_i\) is the geometrical mean diameter (MD) at the \(i\)th bin. Eq. (1) is used for both aerosol and cloud particles.

To distinguish cloud areas from clear-sky areas, the criteria of \(N_c > 10 \text{ cm}^{-3}\) and \(LWC > 0.001 \text{ gm}^{-3}\) were used, where \(N_c\) represents the cloud droplet number concentration and LWC represents the cloud LWC. The LWC and cloud effective radius are calculated using Eqs. (3) and (4), respectively.

Liquid water content \(LWC = \sum n_i r_i^3 \rho_A 4\pi/3\) (3)

Cloud effective diameter \(R_e = \sum n_i r_i^3 \sum n_i r_i^2\) (4)
where \( n_i \) is the cloud droplet number of the \( i \)th bin, \( r_i \) is radius of the \( i \)th bin, and \( \rho_w \) is the water density.

RESULTS AND DISCUSSION

Statistical Characteristics of Aerosol and Cloud Particles

The statistical properties of aerosols in clear-sky areas and in clouds, and those of clouds observed in f1–f3 and f5 are shown in Table 2. The aerosol number concentration measured by the PCASP-100X in clear-sky areas \((N_a)\), from the surface to approximately 6 km over Anhui was 338–771 cm\(^{-3}\), which is lower than that observed around Shijiazhuang, Northern China (Sun et al., 2013). The number concentration \((N_{acc})\) of the accumulation mode particles (ranging from 0.1 to 1.0 µm) accounted for more than 95% of the total concentration. The MD of the aerosol particles ranged from 0.19 to 0.43 µm, and small particles constituted the majority. The number concentration of the in-cloud aerosol particles \((N_{incloud})\) was 71–400 cm\(^{-3}\), which is lower than the \( N_a \) value, indicating that some of the aerosol particles are removed by cloud processes in the clouds. The altitudes of the cloud base observed in f1, f2 and f3 were approximately 5.0, 1.0, and 4.0 km, respectively, and \( N_{incloud} \) decreased with cloud base altitude. The number concentration of the accumulation mode particles in the clouds \((N_{acc-incloud})\) accounted for 60%–85% of the total \( N_{incloud} \), which was observed to be 10%–35% lower than the ratio of \( N_{acc} \) in clear-sky areas. This was most likely caused by two factors: (1) the accumulation mode aerosol particles grew larger into coarse mode aerosol particles through hygroscopic growth in the clouds, where the relative humidity is higher than that of the ambient air; (2) simultaneously, tiny cloud particles smaller than 3 µm may have wrongly been counted as coarse aerosol particles by the instruments. Meanwhile, the MD of in-cloud aerosols \((\text{In-MD})\) was 0.53–1.06 µm; this value is much larger than the MD of aerosol particles in the clear sky, which was also affected by the aforementioned two factors.

The number concentration of cloud droplets \((N_c)\) ranged from 29 to 297 cm\(^{-3}\), but this value is dependent on the cloud type. Generally, low clouds contain a greater number of cloud droplets of smaller sizes \((R_e)\), which are controlled

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**Fig. 1.** Map of flight ranges and flight trajectories.

**Table 1.** Summary of the flights.

<table>
<thead>
<tr>
<th>Flight</th>
<th>Date</th>
<th>Time range (CST)</th>
<th>Maximum altitude (m)</th>
<th>Cloud type</th>
<th>Weather system</th>
</tr>
</thead>
<tbody>
<tr>
<td>f1</td>
<td>2014-08-12</td>
<td>15:34-17:10</td>
<td>6090</td>
<td>As</td>
<td>Surface inverted trough</td>
</tr>
<tr>
<td>f2</td>
<td>2014-08-14</td>
<td>14:39-17:04</td>
<td>6022</td>
<td>Sc</td>
<td>Surface cold front</td>
</tr>
<tr>
<td>f3</td>
<td>2014-08-16</td>
<td>16:07-17:56</td>
<td>6552</td>
<td>As</td>
<td>Changjiang-Huaihe cyclone</td>
</tr>
<tr>
<td>f4</td>
<td>2014-08-28a</td>
<td>12:28-13:22</td>
<td>1254</td>
<td>Ns</td>
<td>Surface low pressure</td>
</tr>
<tr>
<td>f5</td>
<td>2014-08-28b</td>
<td>15:46-17:07</td>
<td>5750</td>
<td>As</td>
<td>Surface low pressure</td>
</tr>
</tbody>
</table>
by the air dynamics. The LWC values ranged from 0.051 to 0.127 g m\(^{-3}\). Both \(N_a\) and LWC decreased with the cloud altitude, whereas \(R_e\) increased with the cloud altitude; this finding is consistent with previous cloud observations at other locations (e.g., Lu et al., 2008; Deng et al., 2009).

### Spatial Distribution of Aerosol Particles

#### Vertical Distribution

Fig. 2 depicts the vertical \(N_a\), temperature, MD, and LWC profiles measured on August 12, 14, 16 and 28 (f5). The black and gray lines represent profiles of the ascent and descent flights, respectively. Data of \(N_a\) and MD were averaged over a vertical distance of 100 meters, and the in-cloud aerosols were also included.

As shown in Figs. 2(a1)–(d1), most aerosol particles were concentrated (in the order of \(10^3\) cm\(^{-3}\)) below 1 km, and the maximum value of 3500 cm\(^{-3}\) was measured on August 12. The particle concentration (\(N_a\)) decreased sharply to \(10^2\) cm\(^{-3}\) at the altitude of 1 km and remained at a constant of \(10^1\) cm\(^{-3}\) from 2 to 6 km on August 12 and 16. Figs. 2(a2)–2(d2) illustrate the vertical profiles of temperature. Several temperature inversion layers (TILs) existed during all the observed flights. TILs can prevent the vertical transport of aerosol particles; hence, aerosols can accumulate at the level under a TIL and contribute to the increment of \(N_a\). For example, in the descent leg on August 12 (Fig. 2(o)), a shallow TIL was observed between 1300 and 1400 m, and of \(N_a\) profile exhibited a peak below the TIL layer at an altitude of 1200–1300 m. On August 14 (Fig. 2(b)), two TIL layers were observed during both the ascent and descent periods (between 1900 and 2100 m and between 2100 and 2500 m), which resulted in two \(N_a\) peaks (between 900 and 1800 m and between 1800 and 2100 m).

The vertical MD profiles are depicted in Figs. 2(a3)–2(d3). The MD changed in the range of 0.14–0.24 \(\mu\)m, and it was maintained at approximately 0.2 \(\mu\)m at the lower layer and then decreased gradually to approximately 0.16 \(\mu\)m at higher altitudes in clear-sky areas. The aerosol MD has a mean value of 0.1 \(\mu\)m over Beijing (Zhang et al., 2009) and 0.10–0.17 \(\mu\)m over Shijiazhuang (Yin et al., 2009), and the MD over eastern China is 0.09–0.18 \(\mu\)m under dry conditions, with the average value being 0.14 \(\mu\)m (the MD in dry conditions can be calculated by assuming a realistic hygroscopic growth factor); the differences in MD over separate locations may be caused by different chemical compositions. The increment of MD at 3.0–5.6 km in the subcloud area on August 12 was possibly affected by drying cloud droplets, which left large residue particles due to a cloud process. Furthermore, the sudden increments in the MD inside the cloud area at 0.8–1.2 km on August 14, 3.0–4.7 km on August 16, and 0.2–1.0 km and 4.5–6.0 km on August 28 were likely to be contaminated by tiny cloud particles smaller than 3 \(\mu\)m, which can be considered coarse mode aerosols. However, these particles had little effect on the vertical distribution of the aerosol number concentration, because the accumulation mode aerosols accounted for a majority of the total \(N_a\) and the tiny cloud particles were merely regarded as coarse mode aerosols.

Although the vertical profiles of ascent and descent depicted in Fig. 2 are quite similar, a small difference is still discernible, and the changes in the boundary layer height between the ascent and descent flights could be responsible for this difference. Liu et al. (2009) also revealed that the planetary boundary layer (PBL) structure is crucial for determining the vertical distribution of aerosol in the lower troposphere. Accordingly, the different aerosol vertical profiles here are also related to the different meteorological conditions (Table 1) and structures of PBL. For example, the aerosol vertical distribution on August 12 exhibited a linear declining tendency, whereas the profile on August 16 exhibited an exponential declining tendency, and these 2 days were controlled by different synoptic systems, as explained in more detail as follows.

On August 12, the region was dominated by westerlies at high altitudes with a weak inverted trough on the surface. Light fog was observed at forenoon in this region, indicating that the atmosphere had a stable structure, and the turbulence and convection were restrained. Thereafter, with the vanishing of the fog, the top of the PBL increased to approximately 3 km (Fig. 2(a2)) in the afternoon, and weak turbulence transport in the PBL could lift some aerosol particles from the surface. The vertical velocity (w) profiles (Fig. 3) at 14:00 (CST) indicate that on August 12, an updraft could be observed below 5500 m; the updraft at approximately 1 km was the strongest and was associated with convergence motion in the whole atmosphere (strongest at 1 km), facilitating development of an aerosol vertical profile with a linear declining tendency. On August 16, the observed area was dominated by warm airflow with high humidity at high altitude, but the surface pressure was high. A downdraft was observed from the surface to approximately 5 km (Fig. 3), and the atmosphere stratification was stable, which prevented the vertical transportation of aerosols from the surface and contributed to an exponential declining tendency (Sun et al., 2013). During the aircraft ascent period, the top of the PBL was at approximately 2 km, and the corresponding aerosol layer top was at 1.5 km. However, in the descent period, the PBL top and aerosol layer top increased to 3 km and 2 km, respectively, indicating that PBL development can lift aerosol particles to higher altitudes through turbulence.

### Table 2. Statistical values of aerosol and cloud particles.

<table>
<thead>
<tr>
<th>Flight</th>
<th>(N_a) (cm(^{-3}))</th>
<th>(N_{acc}) ratio</th>
<th>MD (µm)</th>
<th>(N_{cloud}) (cm(^{-3}))</th>
<th>(N_{acc-incloud}) ratio</th>
<th>In-MD (µm)</th>
<th>(N_c) (cm(^{-3}))</th>
<th>(R_e) (µm)</th>
<th>LWC (g m(^{-3}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>f1</td>
<td>771</td>
<td>99%</td>
<td>0.30</td>
<td>136</td>
<td>60%</td>
<td>1.00</td>
<td>29</td>
<td>9.38</td>
<td>0.044</td>
</tr>
<tr>
<td>f2</td>
<td>438</td>
<td>99%</td>
<td>0.19</td>
<td>400</td>
<td>82%</td>
<td>0.53</td>
<td>297</td>
<td>5.15</td>
<td>0.127</td>
</tr>
<tr>
<td>f3</td>
<td>338</td>
<td>95%</td>
<td>0.43</td>
<td>71</td>
<td>67%</td>
<td>1.06</td>
<td>48</td>
<td>8.73</td>
<td>0.051</td>
</tr>
<tr>
<td>f5</td>
<td>372</td>
<td>99%</td>
<td>0.20</td>
<td>344</td>
<td>85%</td>
<td>0.70</td>
<td>209</td>
<td>6.30</td>
<td>0.112</td>
</tr>
</tbody>
</table>
Fig. 2. Vertical distributions of aerosol number concentration ($N_a$, cm$^{-3}$), temperature (°C), mean diameter (µm) and liquid water content (g m$^{-3}$).
Moreover, these two cases constituted the dominant factors of the synoptic system and PBL structure.

**Horizontal Variations**

To study the horizontal variations in aerosol particles over Eastern China, several horizontal flights at different altitudes were conducted. Six flight legs at three different altitudes on August 14 and 28 were chosen for the following analyses and discussions.

Fig. 4 depicts the horizontal variations in \(N_a\) (black lines and symbols) and MD (gray lines and symbols) on August 14 and 28 at three altitudes (< 1, 3, and 5 km). Compared with the vertical distribution, the horizontal changes in the aerosol number concentration and MD were much smaller, and the value at every altitude was almost a constant. Table 3 shows the calculated average values and standard deviations (SDs) of \(N_a\) and MD during the six legs, revealing that every SD of \(N_a\) and MD has a low value, indicating the uniformity of the aerosol horizontal distribution. The average \(N_a\) sharply decreased from 600 to 900 m, and the magnitude decreased from \(10^3\) to \(10^2\) cm\(^{-3}\). The \(N_a\) values at the altitudes of 3 and 5 km were similar (in the order of \(10^2\) cm\(^{-3}\)), indicating a sharp decrease in aerosol particle concentration with altitudes below 1 km and uniformity at altitudes above 3 km. The MD value decreased from 0.22 to 0.17 µm from < 1 to 3 km and then increased to 0.18 µm on August 14 and decreased to 0.16 µm on August 28; however, the difference was tiny.

Fig. 4 (except for Fig. 4(b)) shows a negative correlation between \(N_a\) and MD; that is, a higher \(N_a\) value always corresponded to a lower MD value. Table 3 shows high ratios of accumulation mode particles in these five legs except for leg 2. Fig. 4(b) depicts a positive correlation between \(N_a\) and MD; that is, a higher number concentration corresponded to a larger MD, signifying that the particles observed in leg 2 might have experienced hygroscopic growth.

**Spatial Variations of Aerosol Size Distributions**

The number size distributions of particles can reveal the source of aerosol and may change under the influence of different synoptic conditions. It is also a key factor to determine the life cycle and optical properties of aerosols. Fig. 5 depicts the vertical variation in the aerosol number size distribution observed on August 14, 2014. The data were averaged over a vertical distance of 1 km, with only the clear-sky aerosol data being included. The average spectral distributions are shown for six altitudes: < 1, 1–2, 2–3, 3–4, 4–5, and 5–6 km. The following multi-lognormal distribution function was used to fit the aerosol particle size distributions,

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

where \(n\) is the number of modes per size distribution for the best fit, \(D_p\) is the particle diameter in µm, \(N_i\) is the particle number in mode \(i\), \(D_{g,i}\) is the geometric MD of mode \(i\) (in µm) and \(\sigma_{g,i}\) is the SD of mode \(i\) (Seinfeld and Pandis, 1998; Sun et al., 2013; Li et al., 2015b).

The shapes of the particle spectra on August 14 were similar for each layer, and the concentration decreased with the altitude in almost each bin (Fig. 5). The results derived for the lognormal fits to the particle spectra are presented in Fig. 6. All the particle spectra at each altitude could be fitted with four lognormal distribution functions, and Table 4 shows the parameters describing the number size distributions of four modes in six different layers. The \(N_a\) value of each mode generally decreased with the altitude; in the main modes (mode I and mode II), the geometric MD (\(D_g\)) was...
0.10–0.29 µm, indicating that small accumulation mode particles dominated the whole atmosphere on sunny days. Analysis of fire map and air mass backward trajectories confirmed that aerosol emission over Anhui were mainly from biomass burning.

Relation between Clouds and Aerosols

Removal of Aerosols in Clouds

Fig. 7 presents the cloud droplet number concentration \( N_c \), LWC, aerosol particle concentration \( N_a \) and altitude time series. As mentioned in section Methodology, the

Table 3. Average, SD of \( N_a \) and MD, and accumulation mode aerosol concentration in six horizontal legs.

<table>
<thead>
<tr>
<th>Date</th>
<th>Height (m)</th>
<th>( N_a ) (cm(^{-3}))</th>
<th>MD (µm)</th>
<th>( N_{acc} ) ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>2014-08-14</td>
<td>600</td>
<td>1338</td>
<td>140</td>
<td>0.22</td>
</tr>
<tr>
<td>2014-08-28</td>
<td>900</td>
<td>335</td>
<td>36</td>
<td>0.22</td>
</tr>
<tr>
<td>2014-08-14</td>
<td>3700</td>
<td>33</td>
<td>8</td>
<td>0.17</td>
</tr>
<tr>
<td>2014-08-28</td>
<td>3800</td>
<td>44</td>
<td>19</td>
<td>0.17</td>
</tr>
<tr>
<td>2014-08-14</td>
<td>5600</td>
<td>34</td>
<td>7</td>
<td>0.18</td>
</tr>
<tr>
<td>2014-08-28</td>
<td>5700</td>
<td>21</td>
<td>5</td>
<td>0.16</td>
</tr>
</tbody>
</table>
clouds were determined using the $N_c$ and LWC criteria of 10 cm$^{-3}$ and 0.001 g m$^{-3}$, respectively; therefore, the areas surrounded by the blue and green lines in Fig. 7 represent clouds, and the pink shadow areas indicate clear-sky areas.

Fig. 7(a) depicts the ascending legs of flights on August 14, from 14:49 to 14:53. The aircraft climbed to 1536 m from takeoff in 4 min. Clouds were detected at altitudes of 845–1197 m, with a vertical extent of 352 m. The clear sky over this cloud layer was at an altitude of 1197–1228 m, and another cloud layer with a vertical extent of 60 m was at a higher level. The aerosol number concentration decreased gradually in the clear sky below the clouds. After the aircraft flew into the clouds, $N_a$ decreased sharply, with the average $N_a$ value of 431 cm$^{-3}$. Subsequently, the aircraft flew through the first cloud layer into a clear sky with increasing $N_a$ and the value of $N_{cloud}$ in the second cloud layer was lower than $N_a$ in the clear sky (marked by the pink shadow). As discussed, $N_a$ decreased with the altitude except in the presence of TILs. In this ascending process, the temperature decreased with the altitude in the absence of TILs; accordingly, $N_a$ decreased with the altitude. However, $N_a$ in the clear sky over the clouds was higher than $N_{cloud}$; this phenomenon illustrates that the clouds may remove the aerosol particles.

Fig. 7(b) depicts the results of the horizontal flight on August 28 at an altitude of approximately 1200 m; five
clouds which are numbered I–V in the figure were detected during 12 min, and the clear-sky areas are marked by pink shadows in this figure. It is obvious that, the aerosol number concentration was lower in clouds than in the clear sky, even in clouds as thin as cloud IV. The number concentrations of aerosol particles in the five clouds and clear sky around the clouds are shown in Table 5. The aerosol concentration in cloud I ($N_{\text{incloud}}$) was $181 \pm 121 \, \text{cm}^{-3}$, and in surrounding clear sky, the $N_a$ values for the left and right sides were $481 \pm 93$ and $654 \pm 67 \, \text{cm}^{-3}$, respectively. The $N_{\text{incloud}}$ value did not reach half the value of $N_a$ in the clear sky, and more than 50% aerosol particles were removed in cloud I. Cloud II was much larger than cloud I, but $N_c$ was lower (Fig. 7(b)). The $N_{\text{incloud}}$ value of cloud II was $557 \pm 55 \, \text{cm}^{-3}$ and in its surrounding clear sky, the $N_a$ values for the left and right sides were $654 \pm 67$ and $761 \pm 33 \, \text{cm}^{-3}$, respectively; approximately 100 cm$^{-3}$ of aerosol particles were removed. Similar results were noted for clouds III–V. In short, in the horizontal flights, the number concentration of the aerosol particles removed by clouds rangd from 100 to 300 cm$^{-3}$.

Relation between $N_c$ and $N_{\text{incloud}}$

As illustrated in Fig. 7(b), when the cloud droplet number concentration was high, the in-cloud aerosol number concentration was low, as observed in cloud I, and when $N_c$ decreased, $N_{\text{incloud}}$ simultaneously increased. Table 6 shows the statistical properties of the five clouds (I–V), regarding the cloud aerosol and clear sky aerosol particles detected in the horizontal flights during 12:46:30–12:58:30 (Fig. 7(b)).

The observed $N_c$ was $55–199 \, \text{cm}^{-3}$ and the LWC changed from $0.003$ to $0.06 \, \text{g m}^{-3}$; no direct correlation between the LWC and $N_c$. An obvious negative correlation was observed between $N_c$ and $N_{\text{incloud}}$. The correlation between the LWC and in-cloud aerosol number concentration is described in Table 6. The aerosol particle concentrations for the particle sizes of 1–3 μm ($N_{\text{coa-incloud}}$) were positively correlated with the LWC, indicating that under higher LWC conditions, a higher number of aerosol particles may grow to larger sizes through hygroscopic growth in the clouds. The $N_a$ values for the surrounding clear sky showed little variation, but $N_{\text{incloud}}$ values were quite different (Table 6).

To identify the removal ability of the clouds, 497 sets of cloud droplet number concentrations and corresponding in-cloud aerosol number concentrations were used to conduct a linear correlation analysis. A good negative relation (Fig. 8) was observed between $N_c$ and $N_{\text{incloud}}$ with a Pearson’s $r$ of $-0.76$, and the equation $y = 716.80 – 1.65x$ can express the linear relation between $N_c$ and $N_{\text{incloud}}$. Fig. 8 illustrates that the relation between the clouds and in-cloud aerosols can be proven when the cloud contains a greater number of cloud droplets; therefore, the removal of in-cloud-aerosols can be more effective under such conditions.

Fig. 9 depicts the number size distributions of the five clouds. In the size range of 3.5–10.0 μm, the sequence of the number concentration in the clouds was as follows: cloud I > V > II = III > IV; moreover, the sequence of the
Table 5. Aerosol number concentration in clouds and clear sky on both the left and right sides of the cloud.

<table>
<thead>
<tr>
<th>Cloud number</th>
<th>Left side $N_a$ in clear sky (cm$^{-3}$)</th>
<th>$N_{inclou}(cm^{-3})$</th>
<th>Right side $N_a$ in clear sky (cm$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>481(±93)</td>
<td>181(±121)</td>
<td>654(±67)</td>
</tr>
<tr>
<td>II</td>
<td>654(±67)</td>
<td>557(±55)</td>
<td>761(±33)</td>
</tr>
<tr>
<td>III</td>
<td>761(±33)</td>
<td>592(±36)</td>
<td>781(±37)</td>
</tr>
<tr>
<td>IV</td>
<td>781(±37)</td>
<td>671(±56)</td>
<td>861(±39)</td>
</tr>
<tr>
<td>V</td>
<td>861(±39)</td>
<td>503(±87)</td>
<td>684(±35)</td>
</tr>
</tbody>
</table>

Table 6. Statistical characteristics of clouds and aerosol particles.

<table>
<thead>
<tr>
<th>Cloud number</th>
<th>$N_c$ (cm$^{-3}$)</th>
<th>LWC (g m$^{-3}$)</th>
<th>$N_{inclou}(cm^{-3})$</th>
<th>$N_{coa-inclou}$ Ratio</th>
<th>$N_a$ (cm$^{-3}$)</th>
<th>Aerosol removal ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>199 (±96)</td>
<td>0.034(±0.01)</td>
<td>181(±121)</td>
<td>16%</td>
<td>491(±106)</td>
<td>63%</td>
</tr>
<tr>
<td>II</td>
<td>107(±42)</td>
<td>0.060(±0.04)</td>
<td>557(±55)</td>
<td>12%</td>
<td>725(±70)</td>
<td>23%</td>
</tr>
<tr>
<td>III</td>
<td>99(±35)</td>
<td>0.032(±0.026)</td>
<td>592(±36)</td>
<td>5%</td>
<td>768(±35)</td>
<td>23%</td>
</tr>
<tr>
<td>IV</td>
<td>56(±34)</td>
<td>0.003(±0.003)</td>
<td>671(±56)</td>
<td>2%</td>
<td>851(±47)</td>
<td>21%</td>
</tr>
<tr>
<td>V</td>
<td>145 (±59)</td>
<td>0.026(±0.018)</td>
<td>503(±87)</td>
<td>6%</td>
<td>803(±92)</td>
<td>37%</td>
</tr>
</tbody>
</table>

Fig. 8. Linear relationship between in-cloud aerosol number concentration and cloud droplet number concentration.

Fig. 9. Cloud particle number size distribution.
Aerosol removal ratio (from high to low) was identical to that of the cloud number concentration for the size range of 3.5–10.0 µm. This finding indicates that the greater is the number of aerosol particles removed, the higher is the number concentration of cloud particles in the size range of 3.5–10.0 µm.

Discussion of In-Cloud Removal Mechanisms

Fig. 10 depicts time series of the number size distribution and LWC of five clouds (I–V), the cloud number size distributions are filled with different colors, color bar of each cloud is different; LWC is represented by black lines and dots. Cloud I, cloud II-A (first part of cloud II from 12:53:19 to 12:54:19) and cloud IV were in evolution period, mature stage and dissipation stage, respectively.

Most of the cloud particles in cloud I were smaller than 10 µm (Fig. 10(a)), the size distribution was unimodal with peak mode ranged from 5 to 7 µm; LWC value was approximately 0.05 g m\(^{-3}\) with the average value of 0.034 g m\(^{-3}\). High concentration of small cloud particles and low

![Fig. 10. Time series of cloud particle size distribution (filled with colors) and liquid water content (black lines and dots) of five clouds (I–V).](image-url)
LWC could indicate that cloud I was in evolution period. The number concentration in cloud II-A was much lower than cloud I (Fig. 10(b)), cloud number size distribution was bimodal distribution with peak modes in 4–8 µm and approximately 20 µm, the maximum LWC was 0.15 g m⁻³, two times of the value in cloud I. High value of LWC and bimodal distribution of cloud particles were the evidences that cloud II-A was in mature stage. Both number concentration and LWC in cloud IV were very low (Fig. 10(e)), most cloud particles were smaller than 9 µm with unimodal distribution, and peak mode size was smaller than 6 µm, horizontal range of this cloud was also small. LWC in cloud IV was approximately 0.005 g m⁻³, indicating it was in dissipation period.

Aerosol removal ratio in these three clouds was 64%, 23%, and 21%, respectively (Table 6), the clouds in developing period can remove aerosols more effectivity, but lower in mature stage clouds and lowest in dissipation stage clouds. Therefore, we infer that the in-cloud aerosol removal process in our study was possibly affected by the condensation of CCN by cloud activation.

**CONCLUSIONS**

This paper describes aircraft observations (August 12–28, 2014) of aerosol spatial properties and the relation between aerosol particles and clouds over Anhui provence, eastern China. In this paper, the spectral and spatial distributions of aerosol particles are presented, and the removal process of in cloud aerosols is also discussed.

The mean aerosol number concentration from the surface to an altitude of 6 km was 338–771 cm⁻³, and more than 95% of aerosol particles were accumulation mode particles. The in-cloud aerosol number concentration, with values of 71–400 cm⁻³, was lower than that under clear sky conditions, and the in-cloud accumulation mode aerosols accounted for 60%–85% of the particles. Nc decreased from 29 to 297 cm⁻³ with an increasing cloud base from 200–5000 m, whereas Re increased from 5.15 µm to 9.38 µm with altitude of the cloud base. The low clouds contained more and smaller droplets than the middle clouds.

Most aerosol particles were concentrated below 1 km, and the concentration decreased with the altitude. TILs can prevent the vertical transport of aerosol particles; hence, the aerosols can accumulate at levels under the TILs. The MD was maintained at a constant of approximately 0.2 µm under 1–2 km and, then decreased at higher levels. The horizontal variations in the aerosol number concentration and MD were small compared with the vertical distributions, and the values at three altitudes were almost constants. The aerosol number concentrations and MDs were inversely correlated in most of the cases. The Aerosol number size distributions changed with the altitude, and all aerosol spectra could be fitted with four lognormal distributions functions.

Clouds with a cloud base lower than 2 km could remove aerosols effectively, with a removal rate of 21%–63%. The higher the cloud droplet number concentration was, the more aerosol particles were removed, and a negative correlation was observed between Nc and Ncloud with a Pearson’s r of −0.76. Meanwhile, the in-cloud removal process of aerosol particles resulted in the increment of cloud droplets in the sizes of 3.5–10.0 µm, and the mechanism of the aerosol removal is condensation of CCN by cloud activation probably.

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