Observations of Cloud Condensation Nuclei in North China*

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ABSTRACT

Using a DMT (Droplet Measurement Technologies) continuous flow streamwise thermal gradient cloud condensation nuclei (CCN) counter mounted on a Cheyenne IIIA aircraft, about 20 flights for aircraft measurements of CCN over North China were conducted in the autumn of 2005 and spring of 2006. According to the design for aircraft observation, the method of spiral ascent or descent in the troposphere was used for the vertical measurement of CCN, and some certain levels were chosen for horizontal measurement. The vertical distributions of CCN concentrations show that most CCN particles are concentrated in the low level of troposphere and CCN concentration decreased with height increasing. It suggests that the main source of CCN is from the surface. This result is consistent with former studies during 1983-1985 in China with a static thermal gradient CCN counter. The comparison of vertical observations between polluted rural area near Shijiazhuang and non-polluted rural area near Zhangjiakou shows that there is about five times difference in CCN concentration. But over two polluted cities, Shijiazhuang and Handan, there is no notable difference in CCN concentration. The horizontal flight measurements for penetrating the cumulus clouds experiment show the apparent decrease of CCN in clouds. It confirms that cloud has a definite consumptive effect on CCN particles because some CCN particles can form cloud droplets.

The surface measurements of CCN in Shijiazhuang City were made during June-August 2005. The statistical CCN data show the great difference in concentration at the same supersaturation (S) in Shijiazhuang summertime. The minimum CCN concentrations were 584, 808, and 2431 cm⁻³, and the maximum concentrations were 9495, 16332, and 21812 cm⁻³ at S=0.1%, 0.3%, and 0.5%, respectively. CCN has a diurnal variation cycle. From 0600 BT, the concentration began to increase and reached the maximum at about noon. Then it generally decreased throughout the afternoon. The reason maybe is related to the onset of emissions from vehicular traffic in the morning followed by the photochemical production of secondary organics that condense on the primary particles. The precipitation has an obvious scavenging effect on CCN particles. With the increase of rainfall rate, the CCN concentrations decrease quickly. The high surface CCN concentrations in Shijiazhuang should be related to the serious air pollution and then influenced by anthropogenic sources.

According to the expression \( N = CS^k \), the CCN spectra can be derived. The fitted spectral parameters of \( C \) (more than 1000) and \( k \) (about 0.7) show that they are classified to the continental CCN in North China.

Key words: North China, cloud condensation nuclei (CCN), observation

1. Introduction

With the development of society, human activities are exerting a great influence on the natural environment. Except for the natural fluctuation of climate, a large number of evidences show that anthropogenic emissions play an important role in the change of weather and climate, such as urban heat island, photochemical smog, etc. The atmospheric aerosols can influence the weather and climate by cloud, precipitation, and radiation.

Cloud Condensation Nuclei (CCN) are the aerosol particles that can form cloud droplets. There is the so-called “indirect effect” arising from the possible influence of anthropogenic CCN. The first is Twomey effect that the increase in cloud droplet number concentration could increase the multiple scattering within clouds thereby increasing cloud-top albedo (Twomey, 1971; Twomey et al., 1984; Platnick and Twomey, 1994). The second is Albrecht effect that the increase in cloud droplet concentration may also inhibit precipitation development, enhancing cloud lifetime, and

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resulting in an increase in planetary shortwave albedo (Albrecht, 1989), and possibly also in the atmospheric absorption of longwave radiation by the resultant increased atmospheric loading of liquid water and water vapor (Schwartz, 1996).

Studies show that the cloud droplets are related to CCN concentrations (Twomey and Squires, 1959). Twomey and Wojciechowski (1969) found CCN are richer in continental air mass than that in marine air mass. Squires pointed out that the high CCN concentration could inhibit the collision-coalescence precipitation process (warm rain) (Hudson, 1993). Many of the cold precipitation processes may also be affected by CCN. Mitchell (1990) found that the snowfall rate can be a linear function of droplet size, which is a function of the CCN concentration. Recent modeling studies suggest that increased CCN concentrations in the tropics change the altitude and mechanism of rain production (Graf et al., 2000). Hygroscopic seeding experimental results show the CCN below cloud are related to the precipitation efficiency. It is important for the prediction of precipitation and global change (WMO, 2000).

In recent years, the emphasis of cloud physics has been shifted from precipitation to radiative properties as the increasing of global climate studies (Hudson, 1993). CCN are one of the most important factors which are related to cloud radiative properties. Much of the uncertainty in estimates of both the direct and indirect aerosol forcing arises from the present lack of ability to describe the processes governing the loading and geographical and vertical distributions of anthropogenic aerosols. This uncertainty is mainly attributed to deficient understanding of the cloud droplet nucleating properties of aerosols that govern not only the indirect effect but also the removal of aerosol particles from the atmosphere in precipitation (Schwartz, 1996). CCN measurements are very important to the study of weather and climate change.

Many CCN observation projects have been conducted all over the world recently. CCN and cloud droplet measurements were made during ACE-2 in Spain (Snider and Brenguier, 2000). Hitzenberger et al. (2000) measured CCN concentrations in the European Alpine aerosol using a newly developed static thermal diffusion counter in 1997-2000. CRYSTAL-FACE project conducted aerosol and CCN aircraft observations in 2002 (Conant et al., 2004). The APEX (1999-2005) project also included the aircraft observations of CCN and aerosol. But there is a paucity of ground CCN observations in heavily polluted cities with the population more than a million.

In China, the observation of CCN was mainly carried out in the study of Northern Stratiform Cloud Rain Enhancement Experiment during 1983-1985 using a MEE-130 CCN counter (Wang et al., 1989; You et al., 2002). CCN observation was conducted in Helan Mountains of Ningxia during 1994-1995 (Niu et al., 1998). Now the MEE-130 CCN counter has been disused. With the rapid development of the industry and agriculture in China, human activities have obvious effects on the environment (Chen et al., 2004; Wang et al., 2004; Ren et al., 2005; Wu et al., 2006). Because of the absence of CCN instruments in the New Age in China, the observations on the temporal and spatial distributions of CCN in different areas, seasons, and weathers, and the study on the cloud physical effect of CCN remain blank.

North China is one of the most important economic centers in China. Shijiazhuang is one of the most polluted cities in 113 important cities in China (SEPA, 2004; SEPA, 2005). The CCN observation in North China especially in Shijiazhuang is very important to study the effect of anthropogenic emissions on the environment.

2. Instrumentations and observations

2.1 Instrumentation

CCN were first measured with a chemical gradient diffusion chamber (Twomey and Squires, 1959). Since that time, nearly all measurements have used the thermal gradient diffusion chamber. All early measurements were made with static photographic instruments that used batch processing of the samples. Continuous flow chambers using optical particle counters
were introduced in the 1970s. At present, some of the atmospheric and cloud physical research aircrafts in developed countries have mounted CCN instruments. Weather Modification Office of Hebei Province bought airborne cloud physical instruments from USA and re-fitted a Cheyenne IIIA aircraft in 2004. The instrument for CCN measurement is DMT (Droplet Measurement Technologies, USA) CCN counter.

The main structure schematic diagram of DMT CCN counter is shown in Fig.1. It uses a cylindrical column continuous-flow streamwise thermal gradient chamber with 50-cm height and 2.3-cm inner diameter. The temperature gradient is maintained with three sets of thermal electric coolers (TECs) at the top, mid, and bottom along the length of the column. A positive temperature gradient is applied to the CCN column in the streamwise direction. The inner wall of the CCN column is wetted continuously by water flow. Water vapor diffuses from the chamber walls inward more quickly than heat, and thus supersaturation \((S)\) is generated with the maximum \(S\) at the centerline of the column (Roberts and Nenes, 2005). An ambient sample air is introduced into the instrument, split into “sheath” and “sample” flows. The sheath flow is filtered, humidified, and heated, and provides a particle-free “blanket” of air within the CCN column that constrains the sample particles to the centerline of the flow chamber. Activated droplet counting is done with an Optical Particle Counter (OPC) at the exit of the CCN column. The OPC utilizes side scattering technology for the particle size and number measurements. A diode laser at a wavelength of 660 nm is used as the light source. The lowest detectable particle size is 0.75 \(\mu m\) in diameter.

The DMT CCN counter has a loaded computer system and a touch screen which can record, process, and show all the data. The operating software written in LabView runs in embedded Windows XP. It can easily set \(S\) and other variables, and can monitor the operating status. The real data can be transformed to other computers by RS-232 serial port. USB port can link to other data storage devices.

![Fig.1. The main structure schematic diagram of DMT CCN counter.](image-url)
Calibrated by known polydisperse dry aerosol distributions of \((\text{NH}_4)_2\text{SO}_4\), under conditions of flow rate of 500 cm\(^3\) min\(^{-1}\) with a measured temperature difference of 5 K \((S=0.26\%)\), there is a strong 1:1 relationship between the expected and measured CCN concentrations \((r^2=0.81; n=26)\) (Roberts and Nenes, 2005). Roberts et al. (2006) compared DMT CCN counter with Wyoming University static thermal gradient CCN counter mounted on the same King Air aircraft during level flight \((S=0.2\%)\). The result shows they are all consistent with the slope of the best fit 1.055 \((r^2=0.673)\).

DMT CCN counter is designed for both ground and airborne operation with \(S\) range of 0.1%-2%. It can be set for a single \(S\) or as large as five different \(S\) continuous observations at a sampling rate of 1 Hz. The main parameters are shown in Table 1.

<table>
<thead>
<tr>
<th>(S) setting range</th>
<th>Total flow rate</th>
<th>Best flow ratio of sheath air to sample air</th>
<th>Sampling frequency</th>
<th>Range of measured particles</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1%-2.0%</td>
<td>500 cm(^3) min(^{-1})</td>
<td>10:1</td>
<td>1 Hz</td>
<td>0.75-10 (\mu)m</td>
</tr>
</tbody>
</table>

2.2 Aircraft observation

A Piper Cheyenne IIIA twin turbo-prop aircraft was used for the observations. About 20 flights were taken over North China during September and October 2005 and March 2006. The method of spiral ascent or descent in the troposphere was used for the vertical measurement of CCN, and some certain levels were chosen for horizontal measurement. The observation height was from 500 to 8000 m. \(S\) during aircraft measurements was mainly set for 0.3%.

2.3 Ground observation

The in-situ ground observations were made in Shijiazhuang City during June-August 2005. The measurement site is located in the southeast of Shijiazhuang downtown. It is on the flat of No.16 floor of Hebei Meteorological Bureau Building. There are resident areas near the building. The diurnal variation of CCN and the effect of precipitation on CCN were observed with cycling different \(S\) (0.1%, 0.3%, and 0.5%) and single \(S\) (0.2% or 0.3%).

3. The characteristics of CCN over North China

Figure 2 shows the vertical distribution of CCN over Shijiazhuang City on 21 October 2005 \((S=0.3\%)\). On the 0800 BT 500-hPa weather map (figure omitted), North China was governed by northwest flows, and Shijiazhuang was in the easterly flow of surface high pressure. In lower level there was stratocumulus. It can be seen from Fig.2 that CCN were mainly concentrated on the levels from near surface to 3500 m. The CCN concentrations were very low above 3500 m. It suggests that the main source of CCN is from the surface. It is consistent with the early result obtained in China that CCN mainly came from the ground and CCN concentration was higher in the low level of troposphere, and concentration decreased with height increasing (Wang et al., 1989). At 1900-2500 m in Fig.2, CCN concentrations decreased obviously, and the aircraft observation showed there was stratocumulus layer.

In order to study the CCN distributions over polluted and non-polluted areas, a flight for vertical measurements over polluted rural area near Shijiazhuang (38\(^\circ\)19’-38\(^\circ\)37’N, 114\(^\circ\)40’-114\(^\circ\)34’E) and non-polluted rural area near Zhangjiakou (41\(^\circ\)04’-41\(^\circ\)11’N, 114\(^\circ\)41’-114\(^\circ\)46’E) was taken on 7 October 2005. From weather

Fig.2. The vertical distribution of CCN over Shijiazhuang City (21 October 2005, \(S=0.3\%\)).
situations, North China was in the northwest air flows behind the 500-hPa trough. The surface was controlled by the stable high pressure whose center was in middle Hetao area. The observation areas were clear. Figure 3 shows that the CCN concentrations over polluted rural area near Shijiazhuang are notably higher than that over non-polluted rural area near Zhangjiakou. There is about five times difference in the CCN concentration.

Handan is also one of the most polluted cities in 113 important cities in China (SEPA, 2004; SEPA, 2005). A flight for measuring vertical CCN distributions over Shijiazhuang City and Handan City was taken on 17 October 2005. On 0800 BT 500-hPa weather map, Hebei was governed by weak northwest air flows, and there was a stable high pressure formed in Zhangjiakou at the surface. The observation areas were clear but with serious air pollution in lower levels. From Fig.4 it can be seen that the CCN concentrations decreased with height. Except for that the concentration over Shijiazhuang is slightly higher than that over Handan near surface, there is no notable difference in the CCN concentration over the two polluted cities.

In order to study the change in and out clouds, the level flight measurement by penetrating the cumulus clouds was made on 7 October 2005. The cloud droplet concentration was measured by PMS (Particle Measuring Systems, PMI Inc., USA) FSSP-100ER probe. Figure 5 shows the apparent decrease of CCN in clouds. It indicates that some CCN particles in clouds can form cloud droplets and result in the difference between in and out clouds.
CCN spectra can be fitted by the expression 
\[ N = CS^k \] (Twomey, 1959), where \( N \) is the number of CCN activated at supersaturation \( S \), variable \( C \) is the number activated at \( S=1\% \), and \( k \) is a constant.

Figure 6 gives the horizontal measurements of CCN over Handan area at 3000 m ASL during the flight on 17 October 2005 (\( S = 0.1\%, 0.3\%, \) and \( 0.5\% \)). At 500 hPa on 26 March 2006, Hebei was governed by westerly flows. In the afternoon Hebei was behind the weak high pressure and before the low pressure at the surface. The weather was fine to partly cloudy. The fitted CCN spectrum (Fig.7) from the horizontal measurements over middle and southern Hebei at 3000 m ASL (\( S=0.1\%, 0.3\%, \) and \( 0.5\% \)) can be written as 
\[ N = 1830S^{0.84}. \]

Figure 7 also shows some spectra obtained at 3000 m ASL of Arctic Ocean (Hegg et al., 1995) and United Arab Emirates (Salazar et al., 2003), and the boundary layer over Florida of America (Yum and Hudson, 2001). Because the air mass was less affected by anthropogenic emissions, \( C \) and \( k \) are very low over the Arctic Ocean. The observation in United Arab Emirates of the Sea Gulf area was made over industrial areas, \( k \) is not high but \( C \) is higher. \( C \) and \( k \) are higher from the measurements of continental air mass over Florida. Over North China, \( C \) and \( k \) are also higher because of serious pollution.

4. The characteristics of surface CCN during summertime in Shijiazhuang City

4.1 Variation range of CCN concentrations

Based on the ground measurement statistical data obtained during June-August in 2005, the CCN concentrations differ greatly at the same \( S \). The minimum concentrations were 584, 808, and 2431 cm\(^{-3}\), and the maximum concentrations were 9495, 16332, and 21812 cm\(^{-3}\) at \( S=0.1\%, 0.3\%, \) and \( 0.5\% \), respectively (Table 2).

Table 2. The extreme values of surface CCN concentration at different \( S \) during summertime in Shijiazhuang

<table>
<thead>
<tr>
<th>( S )</th>
<th>Min. concentration (cm(^{-3}))</th>
<th>Max. concentration (cm(^{-3}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1%</td>
<td>584</td>
<td>9495</td>
</tr>
<tr>
<td>0.3%</td>
<td>808</td>
<td>16332</td>
</tr>
<tr>
<td>0.5%</td>
<td>2431</td>
<td>21812</td>
</tr>
</tbody>
</table>

4.2 Diurnal variation of CCN

CCN has an obvious diurnal variation. Figures 8 and 9 show the CCN diurnal variation with \( S=0.3\% \) on 11 July and \( S=0.1\% \) on 6-7 July, respectively. On 0800 BT 500-hPa weather map on 11 July, Shijiazhuang was governed by northwest flows behind a weak cold vortex, and it was behind a low pressure at the surface. The weather was clear. At 0800 BT 7 July, Shijiazhuang was governed by westerly flows at 500 hPa, and the surface was governed by southerly flows before Hexi low pressure. Because of the stability of the low pressure, the weather was clear to partly cloudy. It can be seen from Figs.8 and 9 that CCN concentration
began to increase from 0600 BT and reached the maximum at about noon. Then it generally decreased throughout the afternoon. It is similar to the result observed in Mexico City which also has the large population more than a million and serious pollution (Baumgardner, 2004). This maybe is related to the onset of emissions from vehicular traffic in the morning followed by the photochemical production of secondary organics that condense on the primary particles.

4.3 The effect of precipitation on CCN

The precipitation has an obvious scavenging effect on CCN particles. Figures 10 and 11 show the CCN concentration variations ($S=0.2\%$) at the same time in the afternoon of 1, 3, and 4 August, respectively. It was clear and hot at 1500 BT 1 August when CCN concentration was about 8000 cm$^{-3}$. At 1640 BT, the sky was covered with cumulonimbus when CCN concentration was about 7500 cm$^{-3}$. At 1827 BT, the thunderstorm started. From 1832 to 1844 BT, the rainfall was larger. At 1850 BT, the rain stopped. It can be seen from Fig.10 that the CCN concentrations were higher during the initial stage of rain. As the increasing of rain intensity, the CCN concentrations decreased sharply. It dropped from 7000 to about 1800 cm$^{-3}$. When the rain stopped, the CCN concentrations had slight increase. It was clear on 3 August morning and there was a thunderstorm from 1410 to 1513 BT. At 1730 BT, it was overcast and became clear from 1850 BT. It was wholly clear at 1923 BT. From Fig.11 it can be seen that the CCN concentrations were about 2000-3000 cm$^{-3}$ after precipitation. On 4 August, it was clear all the day. At the same time, CCN concentrations were about 3000-3500 cm$^{-3}$ and higher than that on 3 August.

4.4 CCN spectra

Surface CCN spectra were fitted by $N = CS^k$. The surface CCN parameters are listed in Table 3.
Figure 12 shows the CCN spectrum from measurements on 8 August. The CCN spectra at Yinchuan Airport (Niu et al., 1998), Urumqi (Chen and Yan, 1989), and Immokalee, an American southern city (Sax and Hudson, 1981) are also shown in Fig.12. There was almost no anthropogenic emission at measurement site of Yinchuan Airport, and and values are not high there. There was almost no traffic in South Florida Immokalee. The measurements were affected by continental air mass, is not high and is high. Because of the pollution in Urumqi, is high. Shijiazhuang has serious air pollution, is high and is higher than others.

**Table 3.** Surface CCN parameters during summertime in Shijiazhuang

<table>
<thead>
<tr>
<th>Date</th>
<th>Weather</th>
<th>Min. concentration (cm$^{-3}$)</th>
<th>Max. concentration (cm$^{-3}$)</th>
<th>C</th>
<th>k</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>0.1%</td>
<td>0.2%</td>
<td>0.3%</td>
<td>0.5%</td>
</tr>
<tr>
<td>25-26 June</td>
<td>Light rain</td>
<td>890</td>
<td>2168</td>
<td>2984</td>
<td>2620</td>
</tr>
<tr>
<td>6 July</td>
<td>Clear, high temperature</td>
<td>3080</td>
<td>7641</td>
<td>10606</td>
<td>9495</td>
</tr>
<tr>
<td>22 July</td>
<td>Thunderstorm rain</td>
<td>949</td>
<td>2403</td>
<td>3308</td>
<td>3787</td>
</tr>
<tr>
<td>25-26 July</td>
<td>Clear</td>
<td>2463</td>
<td>5159</td>
<td>7373</td>
<td>10846</td>
</tr>
<tr>
<td>8 August</td>
<td>Clear, outside typhoon</td>
<td>1860</td>
<td>3931</td>
<td>5401</td>
<td>7294</td>
</tr>
</tbody>
</table>

5. Conclusions

From the above discussions and analyses, the following conclusions may be drawn.

1. CCN observations were carried out during 1983-1985 and 1994-1995 in China by MEE-130 CCN counter. This instrument has long sampling time (more than 30 seconds) and calibration problem. DMT CCN counter has a sampling frequency of 1 Hz which is especially suitable for aircraft observation.

2. The distribution of CCN over North China is related to surface sources. The CCN concentrations over polluted areas are obviously higher than that over non-polluted areas. Vertical and horizontal aircraft observations show that there is a notable decrease of CCN in cloud. It illustrates that cloud has a definite consumptive effect on CCN.

3. Twomey and Wojciechowski (1969) studied CCN spectra over different areas. They found that the typical $k$ is 0.5 over ocean and 0.7 over land. $C$ is much higher over land than that over ocean. The fitted spectra parameters of $C$ (more than 1000) and $k$ (about 0.7) in North China show the characters of continental CCN.

4. The CCN concentrations during summertime in Shijiazhuang are higher which maybe is related to the serious pollution. It is necessary to do longtime work to study the influence.

5. Because CCN are related to the composition and size of aerosols, the study on atmospheric chemistry of pollutants is very important in the future.

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