Analysis of a long-lasting haze episode in Nanjing, China

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A B S T R A C T

A long-lasting haze episode occurred in Nanjing and its surrounding areas from October 15 to 31, 2009. Aerosol physical, chemical and optical properties during this pollution event were investigated. During the long-lasting haze, the Yangtze River Delta (YRD) region was under the control of a high-pressure system and surface pressure gradients were extremely small. The transport and diffusion of air pollutants were suppressed by very low surface wind speed, stably stratified atmosphere and lower mixing level depth (MLD). Back trajectory analysis showed that local emission and regional transport were important in this pollution process. The average diurnal variation of Aitken mode and coarse mode particles had bimodal distribution, which was mainly influenced by diurnal variation of atmospheric boundary layer (ABL) and anthropogenic emissions. Accumulation mode particles were mostly influenced by diurnal variation of ABL; while new particle formation process was important for nucleation mode particles. The peak value of aerosol particle number concentrations shifted to larger particle sizes, perhaps due to the fact that the haze was favorable to accumulation mode aerosols through collision and coagulation of Aitken mode particles. The high ratio of NO3−/SO42− indicated that traffic source became more and more important in the YRD region. The average aerosol scattering coefficient was 696.7±445.4 M m−1, mainly due to particle counts within diameter intervals from 0.6 to 1.4 μm that increased remarkably during the pollution process. Higher accumulation mode particle counts and higher relative humidity were the main reasons for atmospheric visibility impairment during the haze.

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1. Introduction

Husar et al. (1997) found that South Asia was often covered with brown aerosol cloud, which was named Asian Brown Cloud (ABC). The brownish haze consists of many kinds of aerosols, such as sulfate, nitrate, black carbon, organic carbon, dust, among others, which may be originated mainly from fossil fuel combustion and biomass burning. Previous studies have revealed that particles reduce visibility through scattering and absorbing visible light (Malm et al., 1994, 1996; Latha and Badarinath, 2003; Kim et al., 2006). Visibility impairment caused by haze pollution can not only increase traffic congestion but also cause respiratory system and cardiovascular diseases (Husar et al., 1979, 1981, 2000; Leavey and Sweeney, 1990; Hornberg et al., 1998; Schichtel et al., 2001; Field et al., 2004; Chang et al., 2009). In addition, haze has an impact on agriculture, climate and ecosystem through scattering and absorbing solar radiation (Sachweh and Koepke, 1995; Davies and Unam, 1999; Ramanathan et al., 2001).

After reforming and opening up its markets, China’s economy, urbanization and energy consumption grew rapidly. Haze pollution occurs frequently, and is one of the most important concerns in cities. At present, there are four severe haze-polluted areas in China, the Bohai Rim, the Yangtze River Delta (YRD) region, the Pearl River Delta region and the Sichuan Basin. The YRD is located on the west coast of the Pacific Ocean, and is the fastest growing economic development region in China. Haze pollution over the YRD was increasing during the past decade (Tie et al., 2006; Fu et al., 2008; Che et al., 2009; Deng, 2011). Some studies (Xu et al., 2002; Qian et al., 2008; Yin et al., 2009; Gao et al., 2009)...
discussed aerosol properties in this region, but studies on long-lasting haze pollution process are limited.

Being the third largest city of the YRD, Nanjing covers an area of over 6500 km² and has a population over eight million. With rapid population growth (partly due to influx of migrant workers) and energy consumption, haze pollution has become one of the most important environmental issues in Nanjing. Tong et al. (2007) found that the occurrence of hazy days had been increasing from 1961 to 2005.

A severe haze episode was experienced from October 15th to 31st, 2009 in Nanjing. This study investigates the meteorological conditions, and aerosol physical, chemical and optical properties during this haze pollution episode.

2. Methodology

2.1. Data and site information

Aerosol properties, visibility and meteorological factors were monitored simultaneously at the campus of Nanjing University of Information Science & Technology located at the northern suburban of Nanjing, about 15 km away from the city center. A steel factory is situated 2 km to the east of the campus, a chemical industry park about 10 km to the northeast, and there are residential areas and farm lands to the west and north of the campus. It is an epitome of the fast developing YRD suburban, and most of the aerosol and fog monitors were placed here (Kang et al., 2009; Liu et al., 2010, 2011; Lu et al., 2010). The field study took place from October 19 to November 1, 2009 and did not cover the whole haze pollution episode (from October 15th to 31st, 2009). The daily air pollution index (API) of Nanjing in October 2009 was obtained from the Ministry of Environmental Protection (MEP) of People’s Republic of China (http://datacenter.mep.gov.cn/TestRunQian/air_dairy.jsp), NCEP GDAS 1°×1° meteorological data (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1/) was used to initialize the Hybrid Single Particle Lagrangian Integrated Trajectory Model 4.8 (HYSPLIT-4).

2.2. Instruments

A wide-range particle spectrometer (WPS; MSP Corporation model 1000XP) was used to measure aerosol size distributions in the range of 0.01–10 μm. The instrument combines the principles of differential mobility analysis (DMA), condensation particle counting (CPC) and laser particle spectrometry (LPS). The DMA classifies particles in the size range from 10 to 500 nm with 24, 48 or 96 channels. In this study, we chose the sample mode with 48 channels. The CPC measures the number concentration of particles coming from the aerosol exit of the DMA. The LPS is a single-particle, wide-angle optical sensor used for measuring particle size from 0.35 to 10 μm with fixed 24 channels. In this study, one sampling period was set to 5 min. An FA-3 aerosol size distribution sampler was used to collect particles smaller than 10 μm with Teflon filter for mass and ion analyses. Filter masses were determined using a Mettler Toledo MX5 electronic. The ion compositions of particles were analyzed by Dionex ICS-2000 Chromatography System. Aerosol light scattering coefficients were monitored by Model 3563 Integrating nephelometer (TSI Incorporated), which has three band-pass filters centered at wavelengths of 450 nm (blue), 550 nm (green), and 700 nm (red).

3. Results and discussion

3.1. Depiction of the long-lasting haze pollution episode

The API is a simple and generalized way to describe the air quality in China. It is calculated from concentrations of sulfur dioxide (SO2), nitrogen dioxide (NO2) and suspended particulates (PM10). An individual score is assigned to the level of each pollutant and the final API is the highest of those three scores. The API limit of each pollution level and its corresponding mass concentration of each component are shown in Table 1. A long-lasting air pollution process occurred in Nanjing and its surrounding areas from October 15th to 31st, 2009. The API in Nanjing and its nearby cities in October 2009 is also shown in Fig. 1. The API values at these cities increased from October 15th to 28th, reaching or exceeding the limit of China’s air quality standards Grade II (100). When the API is higher than 100, we define it as a haze pollution process. The API in Nanjing remained higher than 100, from October 16th to 30th, with a mean value of 125, which was the highest value in all the cities in the YRD region. When a cold front passed through the YRD region on November 1st, air pollutants were reduced due to stronger wind and precipitation, and the API of these cities decreased to about 50. Since the primary pollutant was aerosol, we named this process “a long-lasting haze episode.”

3.2. Meteorological factors

Meteorological conditions often play an important role in the transport, diffusion and deposition of air pollutants. Stable continental high-pressure and low-pressure systems are the major weather systems that make it favorable for meso-scale and large-scale heavy pollutions to form (Chen et al., 2007; Zhu et al., 2010). Fig. 2a is the surface weather pattern over the eastern Asia at 0800 LST on October 20th, 2009. From this map we can see that the YRD region was under the control of a high-pressure system, and the large-scale stagnation was indicated by the large spacing between the isobars. The surface pattern throughout this long-lasting pollution process remained similar to what is shown in Fig. 2a. This weather system would lead to a lower surface wind speed and stably stratified atmosphere, favorable for the accumulation of air pollutants. In order to know the transport pathway of the air parcels that passed through

Table 1

<table>
<thead>
<tr>
<th>API limit</th>
<th>Air pollution level</th>
<th>Mass concentration (μg/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>SO2</td>
</tr>
<tr>
<td>50</td>
<td>Excellent</td>
<td>50</td>
</tr>
<tr>
<td>100</td>
<td>Good</td>
<td>150</td>
</tr>
<tr>
<td>150</td>
<td>Slight polluted</td>
<td>475</td>
</tr>
<tr>
<td>200</td>
<td>Lightly polluted</td>
<td>800</td>
</tr>
<tr>
<td>250</td>
<td>Moderately polluted</td>
<td>1200</td>
</tr>
<tr>
<td>300</td>
<td>Heavily polluted</td>
<td>1600</td>
</tr>
<tr>
<td>300+</td>
<td>–</td>
<td>–</td>
</tr>
</tbody>
</table>
Nanjing, backward trajectory analysis by HYSPLIT-4 was carried out. Fig. 3 gives the result of cluster analysis of 72-hour back trajectory and MODIS fire distributions from October 20th to 30th, 2009. It clearly shows the three main pathways from the cluster analysis. The first pathway represented the long-range transport process from the northwest, which only accounted for 8% of the air mass transported. The second pathway was the most important, about 66% of the air mass transported. The third pathway indicated that about 26% of the air mass came from the southwest direction. There were plenty of fire spots along the third pathway, implying this haze pollution episode was partly influenced by biomass burning. A cold front passed through the YRD region on November 1st (Fig. 2b), which ended the long-lasting air pollution episode.

Fig. 4 shows the local meteorological conditions in Nanjing from October 20th to 31st, 2009. The surface wind, visibility, relative humidity, rainfall and daily maximum MLD are presented as a function of time. During the pollution period, the dominating wind directions were easterly and southerly, and hourly-mean wind speeds were less than 5 m/s, which was unfavorable for dispersing air pollutants horizontally. Atmospheric visibility was less than 10 km, with a mean value of only 2 km in the heavily polluted days from October 26th to 30th; moreover, the relative humidity during those days was 72.5%, higher than 51.7% on other pollution days. Atmospheric extinction was strengthened by higher aerosol concentration and relative humidity, resulting in lower visibility during the whole pollution process. The daily maximum MLD was lower than 1000 m from October 22nd to November 1st, and was only 248 m on October 27th. One of the most important reasons for this long-lasting haze pollution process was the accumulation of air pollutants near the surface caused by low MLD. Although the pollution process ended on November 1st, the maximum MLD was still lower than 1000 m, which means the MLD was not the major contributor to the ending of this episode. A cold front passed through Nanjing in the afternoon of October 31st. Wind direction changed to northerly, wind speed and visibility increased, and precipitation occurred in the evening, all of which helped to end this event.

3.3. Aerosol physical, chemical and optical properties

3.3.1. Aerosol physical property

Fig. 5 shows the time series of particle number size distributions in 0.01–1 μm. In this long-lasting haze pollution period, aerosol number concentrations mostly fell between 0.02 and 0.2 μm, with a spike higher than 70,000 cm$^{-3}$. Aerosol number concentrations decreased remarkably when the cold front passed through Nanjing in the afternoon of October 31st. Diurnal variation of nucleation mode (0.01–0.02 μm), Aitken mode (0.02–0.1 μm), accumulation mode (0.1–1 μm) and coarse mode (1–10 μm) aerosol number concentrations is shown in Fig. 6. Aitken mode, coarse mode and total aerosol show similar diurnal variation, with a bimodal distribution. The first peak appeared at 0800 LST, mostly affected by increased anthropogenic emissions and low ABL. Then particle number concentrations decreased with intensifying solar radiation and increasing boundary layer; they reached the minimum at 1500 LST. The second peak value appeared at 2100 LST with a decrease of the boundary layer height. Number concentrations of accumulation mode particles were less affected by emissions than by atmospheric stability. They reached the minimum at 1500 LST. Nucleation mode particles reached their peak at 1300 LST, possibly affected by new particle formation process. However, the number concentration of nucleation mode particles was significantly lower than that of either Aitken mode or accumulation mode, which implies that the new particle formation process was not obvious during the episode, because of high relative humidity and high concentrations of larger particles.

It is well known that most of the ambient particles in urban atmosphere are in the ultrafine size range (<0.1 μm) (Woo et al., 2001; Gao et al., 2007). In this study, 62% of the total particle count was found in the size range of 0.01–0.1 μm. This is comparable to the 61% reported in Atlanta (Woo et al., 2001), but smaller than the 72% in Eastern Germany (Tuch et al., 1997) and 94% in Taicang (Gao et al., 2009). This percentage will be extremely high when the new particle formation process is significant. Comparisons of particle number concentrations between this study and
Fig. 2. Surface weather patterns over the eastern Asia at 0800 LST on a) October 20, 2009 and b) November 1, 2009.
other studies are shown in Table 2. Aitken mode particle number concentration in this long-lasting pollution period was lower than those in other studies; however, the accumulation mode particle count was much higher than those in other studies except for Beijing (Wu et al., 2008). The peak value of aerosol particle number concentrations shifted to a larger particle size, perhaps due to the fact that the haze pollution process was favorable to accumulation mode aerosols through collision and coagulation of Aitken mode particles. Similar phenomena were also found in Beijing (Wu et al., 2008) and Guangzhou (Tan et al., 2009a). Eighty-one percent of the total surface area concentration fell between 0.1 and 1 μm diameter range. Accumulation mode and coarse mode were the two major components in volume concentration, which accounted for 99% of the total volume concentration. The relationship of the total aerosol number concentration and the surface concentration was calculated but no apparent correlation was found between the two (R = 0.28); neither in the relationship between the number concentration and the volume concentration (R = 0.21). This is consistent with the results observed in Pittsburg (Stanier et al., 2004) and Jinan (Gao et al., 2007). Correlation coefficients between visibility and total particle number concentration, surface area concentration, and volume concentration are −0.04, −0.82 and −0.83, respectively. Aerosol surface area concentration and volume concentration may be good indicators for visibility impairment. Fig. 7 shows the average size distributions of particle number concentrations, surface area and volume concentrations during the pollution episode. The peak value of particle number concentration appeared at 0.1 μm. This is different from the other field experiments at the same site; e.g., Qian et al. (2008) and Kang et al. (2009) found that the peak value usually appeared within the diameter interval between 0.02 and 0.05 μm. The size distribution of particle surface area and volume concentration were bimodal, with the peaks of each distribution at about 0.2 μm and 3 μm, and 0.6 μm and 3 μm.

3.3.2. Aerosol chemical property

Water-soluble inorganic ions are important components of atmospheric particles and thought to be a significant contributor to the visibility impairment. Fig. 8 shows the mass concentration of PM$_{10}$ and its main anion components. The average mass concentration of PM$_{10}$ from October 19th to 31st, 2009 was 296 μg/m$^3$, and its maximum value reached 380 μg/m$^3$ on October 26th. Main anion components’ mass concentrations were in the sequence of NO$_3^-$ > SO$_4^{2-}$ > Cl$^-$ > F$^-$ > NO$_2^-$; the
mass contributions of these anions to PM10 were in the sequence order of 7.9%, 7.5%, 1.1%, 1.0% and 0.8%. These percentage values are comparable with those found in Guangzhou haze pollution process (Tan et al., 2009a). The mass contribution of total anions to PM10 during this long-lasting episode was significantly lower than the biomass burning and clean process but was higher than the dust process observed in the same area (Zhang et al., 2012).

As the most important water soluble ions, \( \text{SO}_4^{2-} \) and \( \text{NO}_3^- \) mass concentrations depended on \( \text{SO}_2 \) and \( \text{NO}_2 \) concentrations and their oxidation rates in the atmosphere. Previous studies have indicated that nitrogen oxidation ratio (\( \text{NOR} = \frac{n\text{NO}_3^-}{n\text{NO}_3^- + n\text{NO}_2} \)) (\( n \) refers to molar concentration) and sulfur oxidation ratio (\( \text{SOR} = \frac{n\text{SO}_2^-}{n\text{SO}_2^- + n\text{SO}_2} \)) can be used to estimate the degree of transformation of nitrogen and sulfur (Sun et al., 2006). The larger SOR and NOR values are, the greater oxidation of gaseous species will occur and more aerosols will be formed. Ohta and Okita (1990) proposed that when oxidation ratio is larger than 0.1, the photolysis reaction of \( \text{SO}_2 \) will take place. The average SOR and NOR in this pollution period were 0.13 and 0.16 respectively, which means sulfate and nitrate produced from \( \text{SO}_2 \) and \( \text{NO}_2 \) oxidations were not significant. These ratios are lower than those in Guangzhou (Tan et al., 2009b), where the SOR and NOR in haze days were 0.29 and 0.24, respectively.

The mass ratio of nitrate to sulfate can tell us whether a stationary source or traffic source dominates. If this ratio is greater than 1.0, it means \( \text{SO}_2 \) and \( \text{NO}_x \) in the atmosphere mainly come from the traffic source; otherwise, \( \text{SO}_2 \) and \( \text{NO}_x \) are mainly from a stationary source (Huebert et al., 1988). The average ratio of \( \text{NO}_3^- / \text{SO}_4^{2-} \) during this pollution process was 1.05, which is comparable with the result in Guangzhou (Tan et al., 2009b), where the ratio in haze days was 1.02. During this haze process, the concentration of \( \text{NO}_x \) greatly surpassed that of \( \text{SO}_2 \). Under high \( \text{NO}_x \) condition, the concentrations of OH and \( \text{H}_2\text{O}_2 \) were reduced (Poppe et al., 1993), and further suppressed the \( \text{SO}_4^{2-} \) formation. However, we found that the ratio of \( \text{NO}_3^- / \text{SO}_4^{2-} \) was higher than that found in Nanjing (0.84) during a heavy haze pollution process in 2001 (Wang et al., 2003). This increasing trend
3.3. Aerosol optical property

Aerosol extinction in visible wavelength refers mainly to the particles within the diameter interval from 0.6 to 1.4 μm (Bullrich, 1964). Typically, aerosol scattering effect is greater than its absorption effect. Fig. 9 shows the aerosol scattering coefficient \( \sigma_{sp} \) at 550 nm (green light) and aerosol counts within the diameter range from 0.6 to 1.4 μm. The correlation coefficient of the two is 0.94. As Fig. 9 shows, the scattering coefficients and their amplitudes were relatively small before October 25th; after that, \( \sigma_{sp} \) increased and fluctuated remarkably between the minimum and maximum values of 395 M m\(^{-1}\) and 2650 M m\(^{-1}\). This fluctuation was caused by aerosol number concentrations in the size range of 0.6–1.4 μm, which may be driven by aerosol collision and coagulation process and by many meteorological variables, including air-parcel trajectory, level of stagnation, relative humidity and aerosol transport. The diurnal variation of \( \sigma_{sp} \) was bimodal with the two peaks appeared around 0900 LST and 2100 LST; the minimum value was found at about 1400 LST. The diurnal variation of \( \sigma_{sp} \) was similar to aerosol number concentration, mostly influenced by diurnal variation of ABL. We divided aerosols into three bins (0.01–0.6 μm, 0.6–1.4 μm and 1.4–10 μm) by diameter and compared the aerosol number concentrations with visibility during this pollution period in a scatter plot (Fig. 10). Only those aerosols within the diameter interval from 0.6 to 1.4 μm show the expected reciprocal relationship, while the aerosols in other bins have no significant correlations with visibility. There is also an apparent reciprocal relationship between scattering coefficient and visibility (Fig. 10).

The mean values and standard deviations of total scattering coefficient \( \sigma_{sp} \), hemispheric backscatter coefficient \( \sigma_{bsp} \) and hemispheric backscatter fraction \( b = \sigma_{bsp}/\sigma_{sp} \) of this pollution process at the green wavelength are 696.7 ± 445.4 M m\(^{-1}\), 77.1 ± 48.2 M m\(^{-1}\) and 0.110 ± 0.012, respectively. Table 3 summarizes the scattering coefficient in
this study and compares it with those from other campaigns in different parts of China (Bergin et al., 2001; Xu et al., 2002, 2004; Li et al., 2007; Pan, 2007; Andreae et al., 2008; Fan et al., 2010). The measured $\sigma_{sp}$ in this study is higher than the values found in other places in China, mainly due to the aerosol counts in the diameter range of 0.6–1.4 $\mu$m that increased dramatically in this long-lasting haze pollution process.

4. Conclusion

1) During the haze pollution process, the YRD region was under a high-pressure system and large-scale stagnation, as indicated by the large spacing between the isobars (Fig. 2). Smaller surface wind speed, a stably stratified atmosphere and lower mixing level depth in the pollution period were unfavorable for the transport and diffusion of air pollutants. Back trajectory analysis showed that the haze pollution in Nanjing originated from local emission and regional transportation.

2) The average diurnal variation of Aitken mode, coarse mode and total aerosol counts showed a bimodal distribution, which was mainly influenced by diurnal variation of ABL and human activities. Accumulation mode particles were mostly influenced by diurnal variation of ABL. Nucleation mode particles reached their peak at 1300 LST, possibly affected by new particle formation process. The peak value of aerosol particle number concentrations shifted to larger particle sizes, perhaps due to the haze pollution process that was favorable to accumulation mode aerosols through collision and coagulation of Aitken mode particles.

3) The amounts of sulfate and nitrate formed through SO$_2$ and NO$_2$ oxidations were not significant during this pollution process. The ratio of NO$_3^-$/SO$_4^{2-}$ was 1.05, larger than before, probably due to the SO$_4^{2-}$ formation which was suppressed by high NOX concentration and the rapid growth of traffic in the YRD region.

4) The average aerosol scattering coefficient and standard deviation were 696.7 ± 445.4 M m$^{-1}$. A significant reciprocal relationship between visibility and particle concentration within the diameter interval from 0.6 to 1.4 $\mu$m was found. Aerosol counts in the diameter range of 0.6 to 1.4 $\mu$m increased dramatically in this long-
lasting haze pollution process, which led to increased $\sigma_{sp}$ and remarkably decreased atmospheric visibility. The diurnal variation of $\sigma_{sp}$ showed a bimodal distribution, mostly influenced by the height of ABL.

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**References**


**Table 3**

Comparison of aerosol scattering coefficients between this study and other studies.

<table>
<thead>
<tr>
<th>Location</th>
<th>Observation time</th>
<th>$\lambda$ (nm)</th>
<th>$\sigma_{sp}$ (M m$^{-1}$)</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nanjing</td>
<td>2009.10.20–31</td>
<td>550</td>
<td>696.7 ± 445.4</td>
<td>This study</td>
</tr>
<tr>
<td>Xianghe</td>
<td>2005.05</td>
<td>550</td>
<td>468 ± 472</td>
<td>Li et al. (2007)</td>
</tr>
<tr>
<td>Guangzhou</td>
<td>2004.10.04–11.05</td>
<td>530</td>
<td>463 ± 178</td>
<td>Andreae et al. (2008)</td>
</tr>
<tr>
<td>Yulin</td>
<td>2001.03.30–05.01</td>
<td>530</td>
<td>158 ± 193</td>
<td>Xu et al. (2004)</td>
</tr>
<tr>
<td>Linan</td>
<td>1999.10.28–11.30</td>
<td>530</td>
<td>353 ± 202</td>
<td>Xu et al. (2002)</td>
</tr>
<tr>
<td>Beijing</td>
<td>1999.06.11–16</td>
<td>530</td>
<td>488 ± 370</td>
<td>Bergin et al. (2001)</td>
</tr>
<tr>
<td>Beijing</td>
<td>2006.03.18–04.19</td>
<td>525</td>
<td>278.9 ± 248.1</td>
<td>Pan (2007)</td>
</tr>
<tr>
<td>Tianjin</td>
<td>2006.04.24–05.16</td>
<td>525</td>
<td>302.1 ± 171.8</td>
<td>Pan (2007)</td>
</tr>
</tbody>
</table>

Fig. 10. Scatter plot of aerosol number concentrations, aerosol scattering coefficient ($\lambda = 550$ nm) versus visibility.


