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Estimation of the Main Factors Influencing Haze, Based on a Long-term Monitoring Campaign in Hangzhou, China

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ABSTRACT

Eight years of data on haze and visibility (2003–2010) and one year of (2010) data on surface meteorological elements (relative humidity, wind speed, air temperature), visibility and the concentrations of air pollutants ($PM_{2.5}$, SO_2, NO_2 and O_3) measured each hour of each day were analyzed using correlation analysis to investigate the main factors influencing haze in Hangzhou, China. The occurrence of hazy weather has become more frequent over the past eight years in Hangzhou, and haze appears about 160 days per year. The occurrence of haze during the day was more frequent in the spring and the winter and less frequent in the summer and the autumn. Low visibility occurred in the morning, and the maximum visibility occurred in the afternoon period. The results of the statistical analysis show that the relative humidity and the concentration of $PM_{2.5}$ played the most important roles in reducing visibility. The correlation coefficients between the concentration of $PM_{2.5}$ and the concentrations of O_3, SO_2 and NO_2 indicate that O_3 and NO_2 are the dominant factors contributing to $PM_{2.5}$ pollution, which, in turn, can lead to haze. To reduce the number of haze days, greater concern and more countermeasures should be taken to decrease the O_3 and NO_2 pollution in Hangzhou, China.

Keywords: Haze; Relative humidity; $PM_{2.5}$; O_3; NO_2.

INTRODUCTION

Haze is defined as the weather phenomenon that leads to an atmospheric visibility of less than 10 km due to the amount of suspended solid or liquid particles, smoke and vapor in the atmosphere. Haze formation is closely related to meteorological conditions and air pollution levels (Lee and Sequeira, 2001; Sun *et al*., 2006; Fu *et al*., 2008). In recent years, haze pollution has attracted a lot of attention due to its significant effects on visibility, cloud formation, public health, and even the global climate (Okada *et al*., 2001; Schichtel *et al*., 2001; Menon *et al*., 2002; Watson, 2002; Chen *et al*., 2003; Yadav *et al*., 2003; Kang *et al*., 2004; Andre, 2005). Fine particulate matter such as water soluble inorganic ions (SO_4^{2-}, NO_3^-, NH_4^+) and carbonaceous species are thought to be the most important contributors to the impairment of visibility (Jacobson, 2001; Brown *et al*., 2002; Chen *et al*., 2003; Kang *et al*., 2004; Sun *et al*., 2006; Alvesa *et al*., 2007; Yang *et al*., 2007; Fu *et al*., 2008; Tan *et al*., 2009a; Tao *et al*., 2009; Tan *et al*., 2009b; Sabbagh-Kupelwieser *et al*., 2010; Vega *et al*., 2010). High concentrations of $PM_{2.5}$ (particulate matter with a diameter less than 2.5 μm), especially high concentrations of secondary species from anthropogenic sources, can have a dominant effect on the formation of haze (Kang *et al*., 2004; Deng *et al*., 2008; Fu *et al*., 2008). Hangzhou is the capital city of the Zhejiang province, located in eastern China in the Yangtze River Delta (YRD). Satellite observations have revealed very high levels of aerosol pollution in eastern China (Tie *et al*., 2006), and previous studies have indicated that there are high concentrations of particulate matter (Chan and Yao, 2008) and extremely low visibilities in the YRD region (Fu *et al*., 2008). Although it is known that hazy weather is becoming more frequent in Hangzhou over the past decade, few studies have analyzed the key factors that affect the formation of haze and their relationships with other factors. There have been several studies in Hangzhou focusing on $PM_{2.5}$ pollution (Huang *et al*., 2006), the correlation between visibility and fine particles (Xu *et al*., 2005) and brief introduction to the cause of haze pollution (Jin *et al*., 2010). In this paper, we identified the factors that directly influence the occurrence of hazy weather, and by studying the correlation between atmospheric pollutants and these factors, the dominant anthropogenic factor in the formation of haze was identified to frame effective measures to alleviate the

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weather phenomenon of haze in Hangzhou, China.

EXPERIMENT AND DATA TREATMENT

Sampling Site Description

Hangzhou is famous for tourism and lies in the east coast of China at 30.3°N and 120.2°E. The climate of Hangzhou is of a semitropical monsoon type, with cold and dry winters and hot and wet summers. The mean annual precipitation in Hangzhou is 1435 mm, which mainly takes place in the summer. Southeast wind is prevalent in the summer, and northwest wind blows frequently in the winter. The sampling site is the Hangzhou environmental monitoring station that is located in the urban area of Hangzhou (in a district with a mixture of residential and business areas). The location of sampling site is shown in Fig. 1.

Measurements

Hourly data for the meteorological conditions and the concentrations of air pollutants, including the concentrations of PM_{10} (particulate matter with a diameter less than 10 µm), $PM_{2.5}$ (particulate matter with a diameter less than 2.5 µm), sulfur dioxide (SO_2), nitrogen dioxide (NO_2), and ozone (O_3), and the relative humidity, wind speed, wind direction, temperature and visibility, were measured in 2010. Meteorological parameters and air pollutants were measured synchronously. The instruments used for the measurements were as follows: (1) PM_{10} and $PM_{2.5}$ mass concentrations were measured with a particle monitor (Trace Oscillation Balance, RP1400, Thermo Fisher Scientific); (2) SO_2, NO_2, and O_3 were determined with the AR500 series analyzer (long optical path, OPSIS) of an environmental air quality automatic monitoring system; (3) Atmospheric visibility was measured with a visibility sensor (Belfort Model 6000); and (4) Relative humidity, wind speed, wind direction and temperature were measured with automatic meteorological instruments (MetOne Instrnments, Inc). The instruments were regularly checked to insure their accuracy. Long-term records of visibility for 2003–2010 were also analyzed to assess the occurrence of hazy weather in Hangzhou.

A haze day is defined by the following conditions: (1) a daily mean visibility of <10 km; (2) no precipitation; and (3) a daily mean relative humidity (RH) of <90% (Wu, 2005). The haze data were screened by looking at the precipitation records for the dataset and excluded when the relative humidity was higher than 90%. The visibility data for 2003 to 2010 were used to analyze the characteristics of haze in Hangzhou, while the hourly data for 2010 were used to analyze the factors influencing visibility and the correlations between the concentrations of $PM_{2.5}$ and other pollutants. The correlations between visibility and wind speed and air temperature, and between visibility and $PM_{2.5}$, $PM_{2.5}$ and O_3, SO_2 and NO_2 concentrations, were performed by using the Statistical Package for Social Sciences 13.0 (SPSS 13.0).

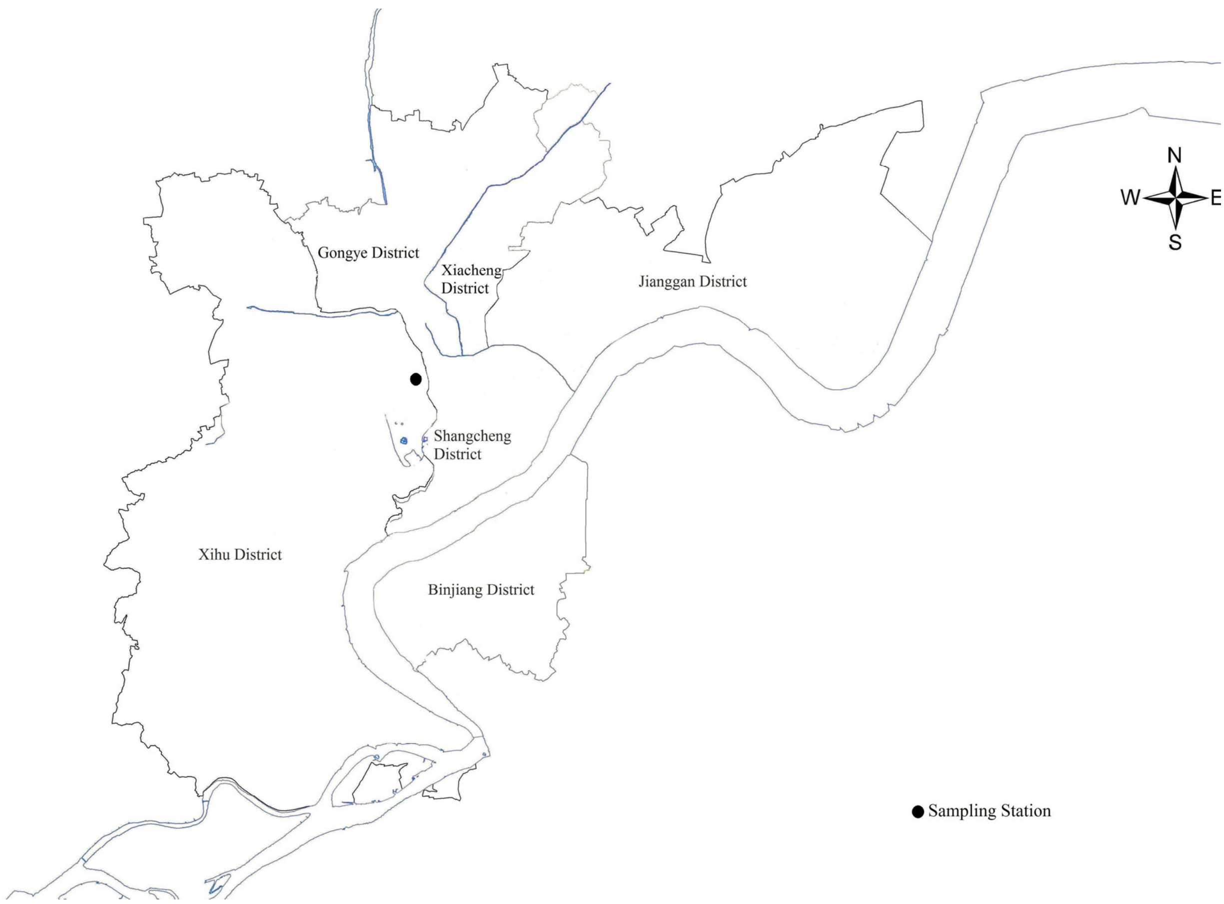

Fig. 1. Location of the sampling station in Hangzhou.

RESULTS AND DISCUSSION

Characterization of Haze in Hangzhou

Annual Variation of Haze Days

Fig. 2 shows the haze day status of Hangzhou from the years 2003 to 2010. Haze days occur about 160 days per year, meaning that Hangzhou is under a low-visibility condition for about 40% of the time. The number of haze days per year in Hangzhou was comparable to the situation in Guangzhou, where there were around 150 haze days per year from 1980 to 2006 (Deng *et al*., 2008), and was slightly lower than for Beijing, where the number of days with visibility <10 km was around 180 days per year for the past 30 years (Chang *et al*., 2009)

Seasonal Variation of Haze Days

Fig. 3 depicts the frequency of haze days in each month from 2003 to 2010. It shows that the frequency of haze days in each month presents a bi-modal distribution; the haze days were more frequent in spring (from March to May) and winter (from December to February) and less frequent in summer (from June to August) and autumn (from September to November). The minimum number of haze days was in the summer. This condition is similar to Guangzhou and Shanghai, where visibility is best in summer, but it differs from Beijing and Shengyang, where visibility is best in spring (Chang *et al*., 2009). These different seasonal patterns may result from the distinct meteorological conditions as well as the differences in air pollution in these different

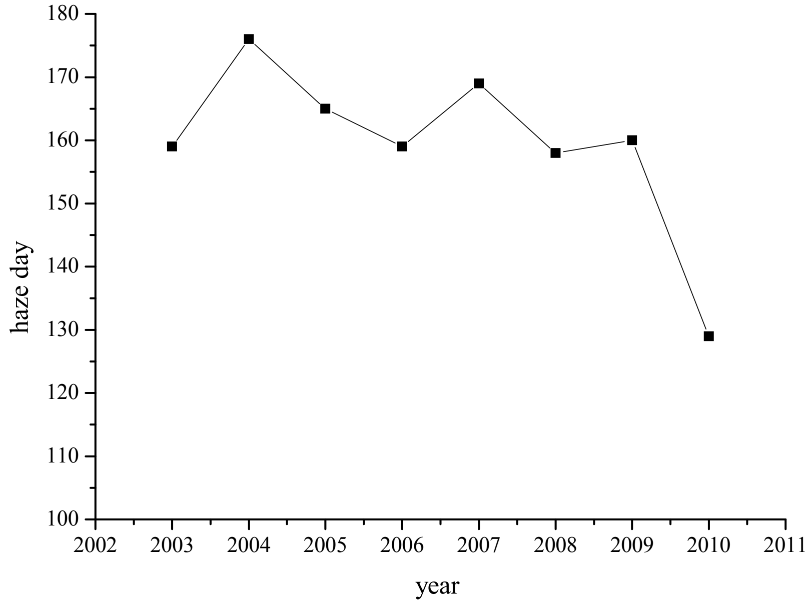

Fig. 2. Haze days observed in Huangzhou from 2003 to 2010.

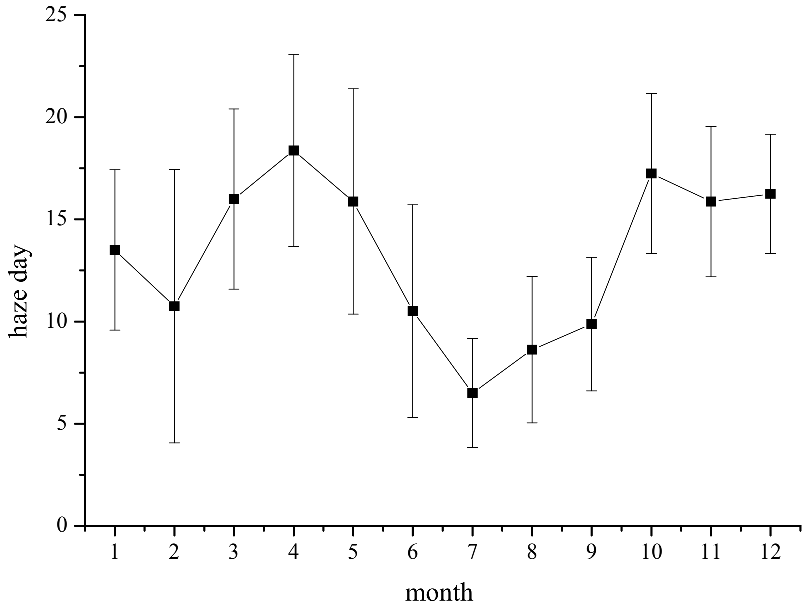

Fig. 3. Occurrence frequency of haze days in each month from 2003 to 2010.

regions. In summer, the less frequency of haze days was probably caused by the low concentration of particulate matter. Generally, southeast wind prevailing in the summer can bring clear air from the ocean, which can dilute the concentration of particulate matter; therefore, the level of visibility was higher in the summer. The greater frequency of haze days in the spring and the winter can probably be ascribed to poor conditions for the dispersion of air pollution and the influence of dust storm from inland.

Monthly Variations of Visibility at Different Times of Day

Fig. 4 depicts the monthly variation of visibility at 02:00, 08:00, 14:00 and 20:00 from 2003 to 2010. As shown in Fig. 4, there are clear patterns in the variation of visibility at these different times. The daily patterns of visibility were similar in different months, with low visibility in the morning period (08:00 hours), which increased and reached a maximum in the afternoon period (14:00 hours). The best visibility occurred in the summer, and the worst visibility happened in the winter, which is consistent with the frequency of haze days in each month. These temporal patterns are typical for urban areas (Wang *et al*., 2003; Shen *et al*., 2007) and are likely caused by variations in emission sources and meteorological conditions. The low values of visibility during the early morning could be attributed to the increased emissions associated with the traffic during the morning rush hour as well as the high relative humidity and the low mixing height, which lead to the accumulation of aerosols and high aerosol concentrations (traffic density was about 3800 per rush hour on the main roads, and the mixing height was about 490 m in the morning). The steady increase in visibility that follows could be associated with the increases in the ambient temperature and the vertical convective activity, which increases the vertical dispersion of aerosols and is also associated with a decrease in vehicle density. The best visibility occurred at 14:00 h and was probably associated with the highest temperatures and mixing heights and lower relative humidities and traffic densities (in the afternoon, the traffic density was about 2300 per hour on the main roads, and the mixing height was about 1200 m).

Analysis of Factors Influencing Visibility

Influence of the Main Meteorological Factors on Visibility

Humidity is an important factor that can influence visibility (Malm and Day, 2001). For atmospheric aerosols, during the process of humidification, numerous small particles combine into a more effective mid-visible light scattering range in sizes and increase the amount of particle scattering in the atmosphere. The resultant increase in the scattering coefficient leads to reduced visibility (Tang, 1997; Lee and Tsai, 1998). According to the criteria for warning signals of haze weather of Wu (2005), the occurrence frequencies of different visibilities at different relative humidities were analyzed. As shown in Fig. 5, a higher level of visibility was associated with a lower relative humidity. For example, when the relative humidity was below 60%, the level of visibility was most frequent >15 km, and the frequency of a visibility <2 km was 0%. When relative humidity varied from 60% to 90%, the frequency of the visibilities <2 km, 2–5 km, 5–10 km, 10–15 km, and >15 km ranged from 0% to 0.96%, 3.97% to14.35%, 18.25% to 22.3%, 16.23% to 18.25%, and 61.54% to 44.22%, respectively, indicating that lower levels of visibility usually happened when the relative humidity was higher. A similar situation was also observed in the Yangtze River Delta (Fu *et al*., 2008).

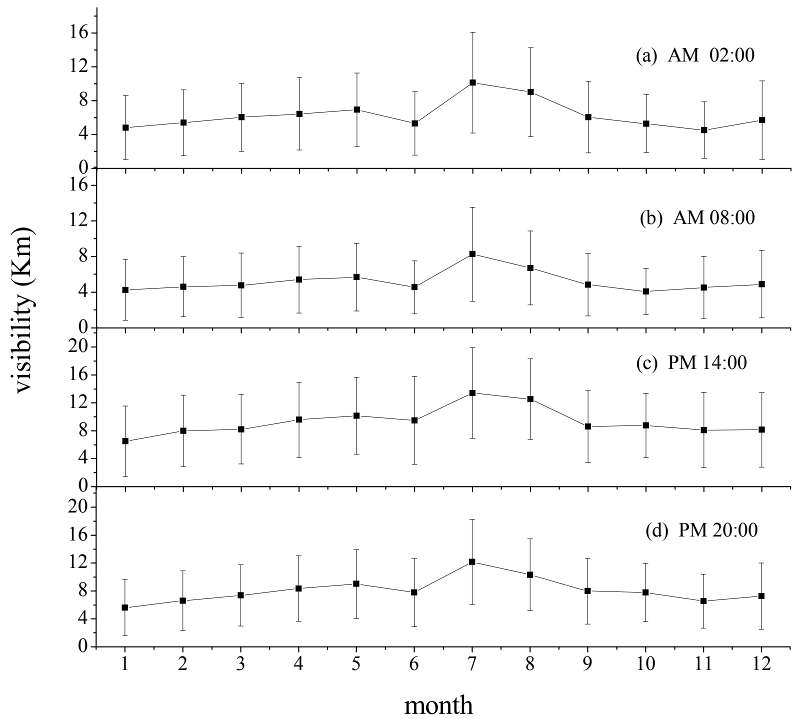

Fig. 4. Monthly variation of visibility at different times of day from 2003 to 2010.

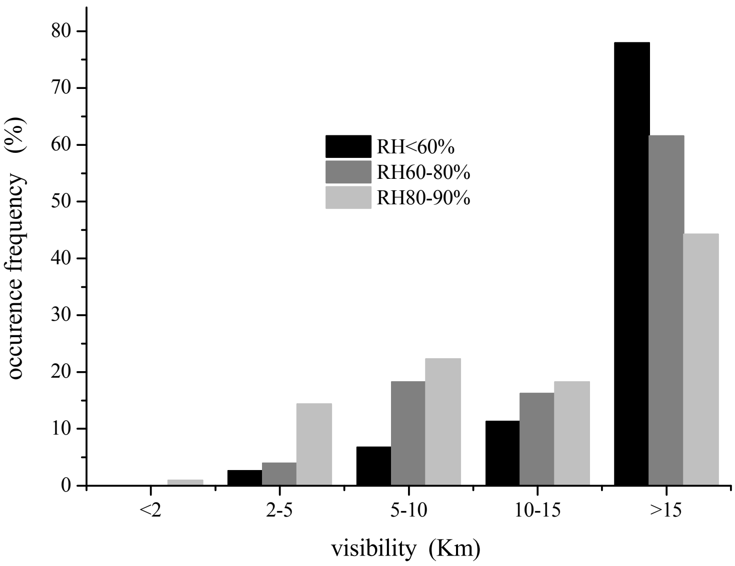

Fig. 5. Occurrence frequency of different visibilities at different relative humidities.

On the other hand, when the correlations between the visibility and the wind speed and the air temperature were analyzed, the coefficients were 0.31 and 0.36 at the 99% confidence level, respectively (the sample number was 5460, $p = 0.000 < 0.01$, and the critical coefficient was 0.081). The wind speed and temperature did not influence visibility directly. In fact, the wind speed was the most important factor affecting the dispersion of particulate matter, and the temperature was the important factor affecting the emergence of secondary aerosols. Therefore, these parameters are the factors indirectly influencing the variation in visibility.

Influence of Fine Particulate Matter on Visibility

A high concentration of fine particulate matter was closely related to the impairment of visibility (Kang *et al*., 2004; Sun *et al*., 2006; Fu *et al*., 2008; Tan *et al*., 2009a; Tan *et al*., 2009b). According to the Koschmeider equation, the extinction coefficient has a directly proportional relationship with the concentration of particulate matter, but has an inverse relationship with visibility. Hence, the concentration of particulate matter has an inverse relationship with visibility; therefore, correlation of $PM_{2.5}$ and the inverse of visibility was analyzed. The results are shown in Fig. 6. From Fig. 6, their correlation coefficient was 0.32 at the 99% confidence level (the sample number is 5460, $p = 0.000 < 0.01$, and the critical coefficient is 0.081), and the result was significant at this confidence level. The results show that the concentration of fine particles also plays an important role in reducing visibility in Hangzhou. This phenomenon has also been observed in Beijing (Song *et al*., 2003; Wang and Liu, 2006), Guangzhou (Tan *et al*., 2009b), Shanghai (Fu *et al*., 2008) and Jinan (Yang *et al*., 2007), Seoul (Kang *et al*., 2004) and the mid-Atlantic region (Chen *et al*., 2003).

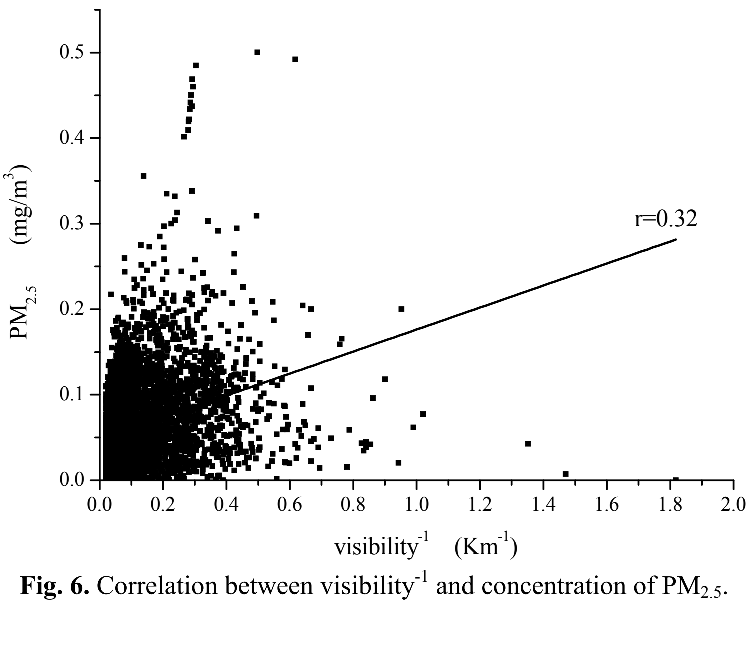

Fig. 6. Correlation between visibility^{-1} and concentration of $PM_{2.5}$.

Correlation between Visibility and the Concentration of $PM_{2.5}$ at Different Relative Humidities on Haze Days

The correlations between the visibilities and the concentrations of $PM_{2.5}$ at different relative humidities during haze days were analyzed in order to understand how the concentration of fine particulate matter influenced the visibility. It can be seen from Fig. 7 (from (a) to (d)) that there were large differences in the correlation coefficients between the visibility and the concentration of $PM_{2.5}$ at different relative humidities. When the relative humidity was below 50%, a higher correlation was found between the visibility and the concentration of $PM_{2.5}$, with a coefficient of 0.95. When the relative humidity was in the range of 50–60%, the correlation coefficient between the visibility and the concentration of $PM_{2.5}$ was 0.76. When the relative humidity was greater than 60%, the correlation between the visibility and the concentration of $PM_{2.5}$ decreased; the coefficients were 0.42 and 0.37. The variation in this correlation indicates that, when the relative humidity was below 60%, the visibility was highly influenced by the concentration of $PM_{2.5}$ in Hangzhou; however, when the relative humidity increased to greater than 60%, the visibility was not only influenced by the particulate matter concentration but also by the relative humidity. Under dry conditions, the aerosol concentration (especially that of fine particles) is the most important factor affecting visibility (Covert *et al*., 1972), but many aerosol components are hygroscopic and take up water as function of relative humidity. At relative humidities greater than 60%, the humidity itself becomes more and more important because water generally comprises more than 50% of the fine particle mass at relative humidities exceeding 70–80% (Zhang and McMurry 1993; McMurry 2000), and the other indicated variables are humidity-dependent (Covert *et al*., 1972; Clarke *et al*., 2004). Thus, with an increase in relative humidity, the correlation between the visibility and the concentration of $PM_{2.5}$ becomes weaker. A similar situation was found for the summer in Beijing (Wang and Liu, 2006).

Correlation between $PM_{2.5}$ and other Pollutants on Haze Days

The relative humidity, wind speed, air temperature and concentration of $PM_{2.5}$ are important factors influencing the formation of hazy weather in Hangzhou. It is well know that the concentration of $PM_{2.5}$ is largely affected by human activity. To create effective measures to reduce haze, the correlation between $PM_{2.5}$ and other atmospheric pollutants from anthropogenic sources on haze days should be analyzed to identify the main factors that can enhance $PM_{2.5}$ concentrations.

Diurnal Variations of Different Pollutants on Haze Days

The data for haze days in 2010 were averaged into each hour of the day to investigate the diurnal variability of $PM_{2.5}$ and other pollutants. The diurnal patterns of different pollutants are depicted in Fig. 8. As shown in Fig. 8(a), the first high value of $PM_{2.5}$ occurred in the morning period, and after that, the values decreased. Then, at about 14:00–15:00, it reached a second high value, and a third high value occurred in the evening. These diurnal cycles are likely due to the high traffic emissions during the morning and evening rush hours and the photochemical reactions

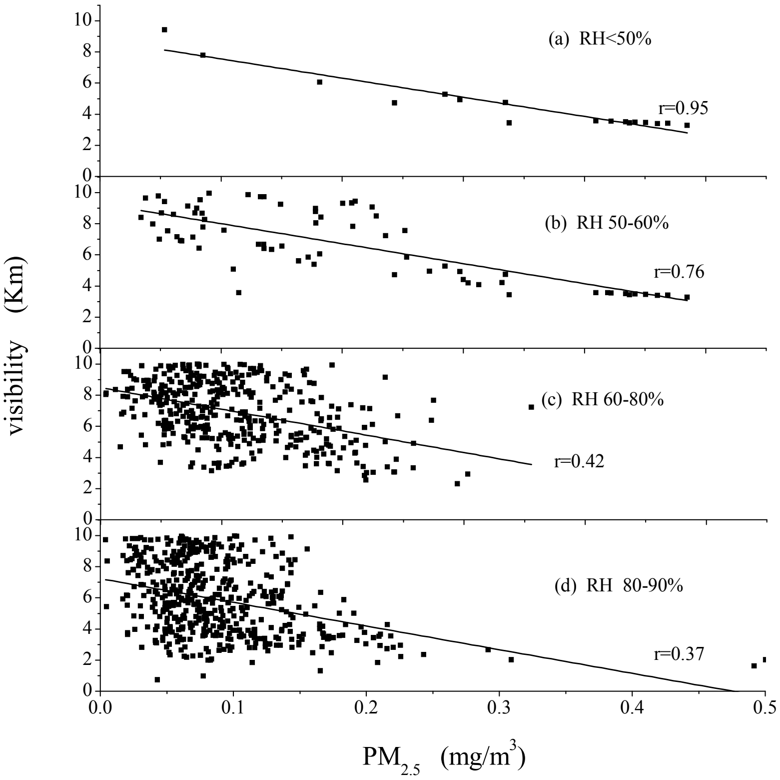

Fig. 7. Correlations between visibility and concentration of $PM_{2.5}$ at different relative humidities on haze days.

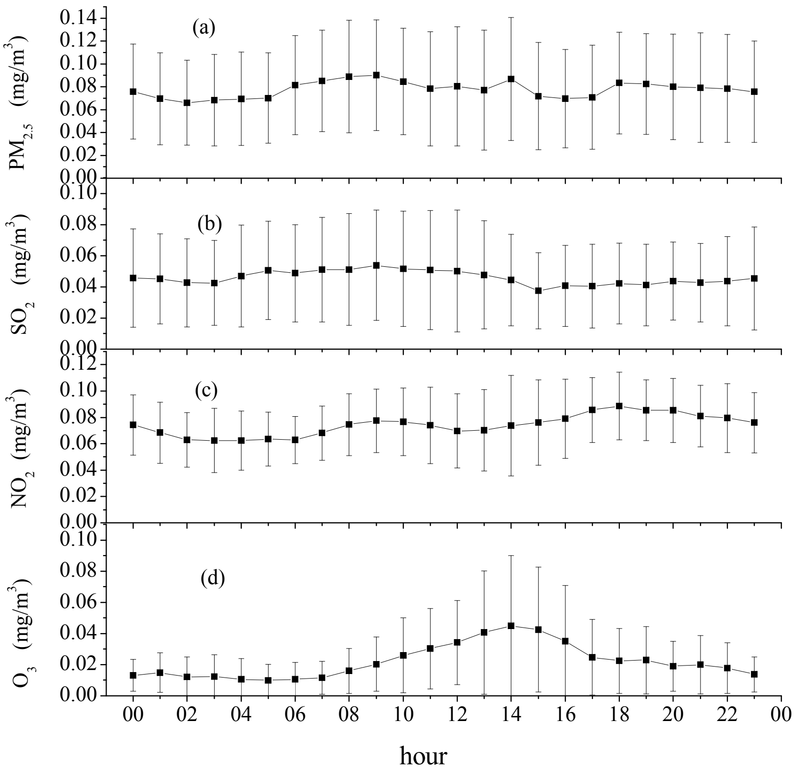

Fig. 8. Diurnal variation of different pollutants on haze days in 2010.

occurring at noon because the diurnal patterns of ozone concentrations had increasing trends at about 08:00 and reached peak values at around 15:00 (Fig. 8(d)), which indicates the enhancement of oxidation in the atmosphere. The second peak in $PM_{2.5}$ concentration might be partially attributed to photochemical transformations, and a similar result was also found in Beijing (Wang, *et al*., 2005). The diurnal patterns of NO_2 appeared to be bimodal, which were associated with the morning and afternoon rush hours.

The Correlations between $PM_{2.5}$ and O_3, SO_2, and NO_2 on Haze Days

The correlations between $PM_{2.5}$ and O_3, SO_2 and NO_2 on haze days were analyzed by using the SPSS software. The results are shown in Fig. 9. The correlation coefficients between $PM_{2.5}$ and O_3, SO_2 and NO_2 were 0.24, 0.17 and 0.44, respectively, and the results were significant at the confidence level of 99% (the sample number is 4520, $p = 0.000 < 0.01$, and the critical coefficient is 0.081). The Hangzhou environmental monitoring station is a district with a mixture of residential and business areas. The study site is located near the road, and there are a lot of vehicle emissions. The fact that the correlation coefficient between $PM_{2.5}$ and NO_2 was larger than the others indicates that mobile source emissions had an important effect on the formation of $PM_{2.5}$. This result is also consistent with the findings of Bao *et al*. (2010). The relationship between ozone and the concentration of fine particulate matter was complex. It is known that ozone, as an oxidant, can change the concentration of free radicals in the atmosphere and can thus impact the formation of secondary aerosols and the concentration of $PM_{2.5}$. Therefore, O_3 and NO_2 were important contributing factors to $PM_{2.5}$ pollution, which can lead to the formation of haze in Hangzhou.

CONCLUSIONS

A long-term record of haze days in Hangzhou shows that haze was a common phenomenon during the past eight years, and there were about 160 haze days every year from 2003 to 2010. The occurrence of haze days was more frequent in the spring and the winter and less frequent in the summer and the autumn. The minimum number of haze days occurred in the summer. There were clear patterns in the variation in visibility at different times of day, with low visibility in the morning period (08:00 hours), which increased and reached a maximum in the afternoon period (14:00 hours). This situation was likely due to the variations in emission sources and meteorological conditions. A high level of relative humidity has an adverse affect on visibility; the relationship between visibility and relative humidity showed that, as the relative humidity increased, lower visibilities occurred more frequently. When the relative humidity varied from 60% to 90%, the frequencies of visibilities <2 km, 2–5 km, 5–10 km, 10–15 km, and >15 km were in the range of 0% to 0.96%, 3.97% to 14.35%, 18.25% to 22.3%, 16.23% to 18.25%, and 61.54% to 44.22%, respectively. The correlations between the level of visibility and the concentration of $PM_{2.5}$, wind speed and air temperature indicate that the concentration of fine particles, wind speed and air temperature all play important roles in reducing visibility, and their correlation coefficients

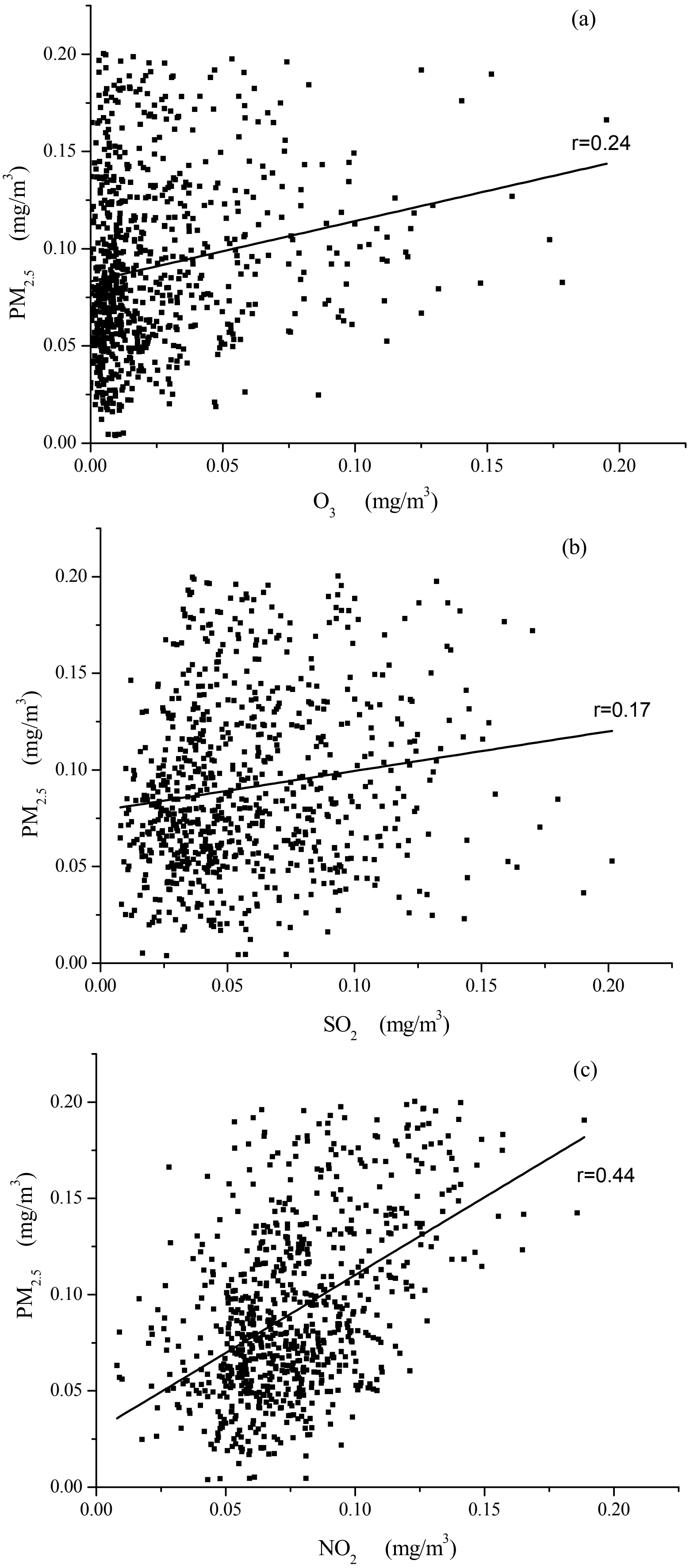

Fig. 9. Correlations between $PM_{2.5}$ and O_3, SO_2 and NO_2 on haze days (from (a) to (c)).

were 0.32, 0.31 and 0.36, respectively, at a 99% confidence level. When the relative humidity was below 60%, the impairment of visibility was mostly highly influenced by the concentration of $PM_{2.5}$. When the relative humidity was greater than 60%, the correlation between the level of visibility and the concentration of $PM_{2.5}$ became smaller. The results of SPSS software indicate that O_3 and NO_2 are important precursors to $PM_{2.5}$ pollution, which can lead to the formation of haze.

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