



Characteristics of Particulate Matter during Haze and Fog (Pollution) Episodes over Northeast China, Autumn 2013

Yang Li^{1*}, Hujia Zhao², Yunfei Wu³

¹ Meteorological Observation Center, CMA, Beijing 100081, China

² Institute of Atmospheric Environment, China Meteorological Administration, Shenyang 110016, China

³ Key Laboratory of Regional Climate-Environment for Temperate East Asia(RCE-TEA), Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China

ABSTRACT

Mass concentrations of particulate matter (PM), including PM_{1.0}, PM_{2.5} and PM₁₀, were measured from October 13th to November 30th 2013 at eight sites in Northeast China to evaluate their variations during pollution periods. Five major pollution periods were identified during the autumn of 2013. The maximum daily average PM_{2.5} concentrations were $437 \pm 85 \mu\text{g}/\text{m}^3$ and $322 \pm 50 \mu\text{g}/\text{m}^3$ in Harbin and Shenyang, respectively. The minimum was $75 \pm 28 \mu\text{g}/\text{m}^3$ in Dandong. The presence of finer particles was significantly related to visibility degradation during pollution periods. Wind speeds had a negative correlation with PM concentrations, while high relative humidity (RH) favored the formation of haze in Northeast China.

Visibility on non-hazy days was approximately 2.5–3.0 times greater than that on hazy days. During hazy days, the PM_{1.0}:PM_{2.5} ratios were 0.89 ± 0.04 , 0.85 ± 0.04 and 0.91 ± 0.04 at Anshan, Shenyang and Dandong, respectively. These results show that PM_{1.0} was the dominant particle pollutant in Northeast China during periods of pollution. High RH and low wind speeds during hazy days may favor the accumulation of atmospheric pollutants. The results of this study provide useful information toward recognizing air pollution episode characteristics in Northeast China.

Keywords: PM; Visibility; Haze; Northeast China.

INTRODUCTION

Particulate matter (PM) pollution exerts an important influence on human health (NRC, 1998; Pope *et al.*, 2009), visibility (Ghim *et al.*, 2005; Che *et al.*, 2007) and climate (Okada *et al.*, 2001; IPCC, 2013; Wang, 2013). The particles suspended in the atmosphere reduce visibility by scattering and absorbing light (Kim *et al.*, 2006; Elias *et al.*, 2009). The reduction of visibility is serious during heavy pollution periods and can be regarded as an indicator of air quality (Watson, 2002).

In China, haze is usually viewed as an atmospheric pollution phenomenon caused by large numbers of aerosol particles in the atmosphere, with visibility < 10 km when ambient relative humidity (RH) is below 90% (CMA, 2003; Sun *et al.*, 2006). The formation of haze is closely related to meteorological factors, as well as to particulate matter in the atmosphere (Hu *et al.*, 2002; Wang *et al.*, 2003), especially

matter contributed by anthropogenic emissions and gas-particle conversions (Dillner *et al.*, 2006).

As the occurrence of haze episodes has become more frequent, the impact of regional haze across East Asia has been more widely studied (Koe *et al.*, 2001; Ramanathan *et al.*, 2001; Franke *et al.*, 2003). In China, haze episodes often occur in developed regions such as the North China Plain (NCP) (Sun *et al.*, 2013; Zhang *et al.*, 2013; Che *et al.*, 2014), the Yangtze River Delta (YRD) (Che *et al.*, 2009; Deng, 2011; Du *et al.*, 2012; Yang *et al.*, 2012; Cheng *et al.*, 2013) and the Pearl River Delta (PRD) (Lü *et al.*, 2009; Yue *et al.*, 2010; Zhang *et al.*, 2012).

Being the largest heavy industry and grain commodity base, the northeastern provinces of China play an important role in China's economic development. Therefore, studying airborne particles in northeastern China is both significant and meaningful. As a result of the rapid increase in population growth, energy consumption and the increasing usage of vehicles in recent decades, particulate pollution, air quality and visibility degradation have become serious environmental issues in Northeast China (Ma *et al.*, 2005; Cheng *et al.*, 2010; Zhao and Ma, 2011). Wang *et al.* (2012) identified central Liaoning Province as one of the key areas for regional air pollution prevention and control because of its growing

* Corresponding author.

Tel.: 86-10-58995140; Fax: 86-10-62176414
E-mail address: liyang@cma.gov.cn

population and economic development. Zhao *et al.* (2012, 2013a), Xia *et al.* (2007) and Wang *et al.* (2010) have analyzed aerosol optical properties and direct radiative forcing in northern China. In addition, Zhao *et al.* (2013b) have analyzed the characteristics of visibility and particulate matter in an urban area of Northeast China.

Several haze & fog episodes happened during October to November of 2013 in the whole northeast China region because of stable weather condition, biomass burning emission and warm-keeping season starting at the same time. These episodes caused closing of high ways, airports, and more than 2000 schools. In this paper, the mass concentrations of PM₁₀, PM_{2.5} and PM_{1.0} (particle matters with sizes smaller than 10 μm , 2.5 μm and 1.0 μm in aerodynamic diameter, respectively) in Northeast China were measured from October 13th to November 30th 2013 to evaluate their characteristics. The potential relations between PM, visibility and other relevant factors, including RH and wind speed, were investigated. The aim of this paper is to better understand pollution in Northeast China, to present the characteristics of PM during pollution episodes, and to evaluate the role of meteorological parameters in visibility deterioration. This study provides useful information toward understanding air pollution in this region, which in turn could be helpful in improving regional air quality.

Site Description, Instruments and Data Measurement

Eight sites in Northeast China were selected for this study. Their geographical information is shown in Fig. 1. Harbin is the capital of Heilongjiang Province, and is

Northeast China's political, economic and cultural center. Changchun is the capital of Jilin Province. Both these cities represent metropolitan areas. Shenyang, Anshan, Fushun and Benxi are important cities in central Liaoning Province, and are located in an area which is not only the core of economic development in Northeast China, but also one of the few urbanized regions in Northeast Asia. Measurements at these four stations reflect the aerosol characteristics in urban, Northeast China. Dandong and Jinzhou are both coastal cities which are far away from sources of anthropogenic emission. The measurements taken there may reflect the aerosol characteristics of coastal areas in Northeast China.

A FD12 Visibility Automatic Observation Instrument and a GRIMM180 Particle Instrument were used to obtain visibility and PM mass concentrations over the eight sites. The measuring time of FD12 instrument is 15 s with an accuracy of $\pm 10\%$ between 0.01 km–10 km and $\pm 20\%$ between 10 km–50 km, and the measuring range is 10–50,000 m. The measuring time of GRIMM180 is 1–60 min which performance with an accuracy of $\pm 2\%$ and the measuring range is 1–1,500 $\mu\text{g}/\text{m}^3$. The daily mean and standard deviation values of visibility, PM and meteorological parameters including RH, temperature, wind speed and precipitation from October 13th to November 30th 2013 were calculated to characterize their variation.

In this article, the pollution episode is classified based on whether the daily PM_{2.5} concentration exceeding the China's national ambient air quality standards (75 $\mu\text{g}/\text{m}^3$) (GB3095-2012: http://kjs.mep.gov.cn/hjbhbz/bzwb/dqjh/bh/dqjhzbz/201203/t20120302_224165.htm). Finally, five heavy

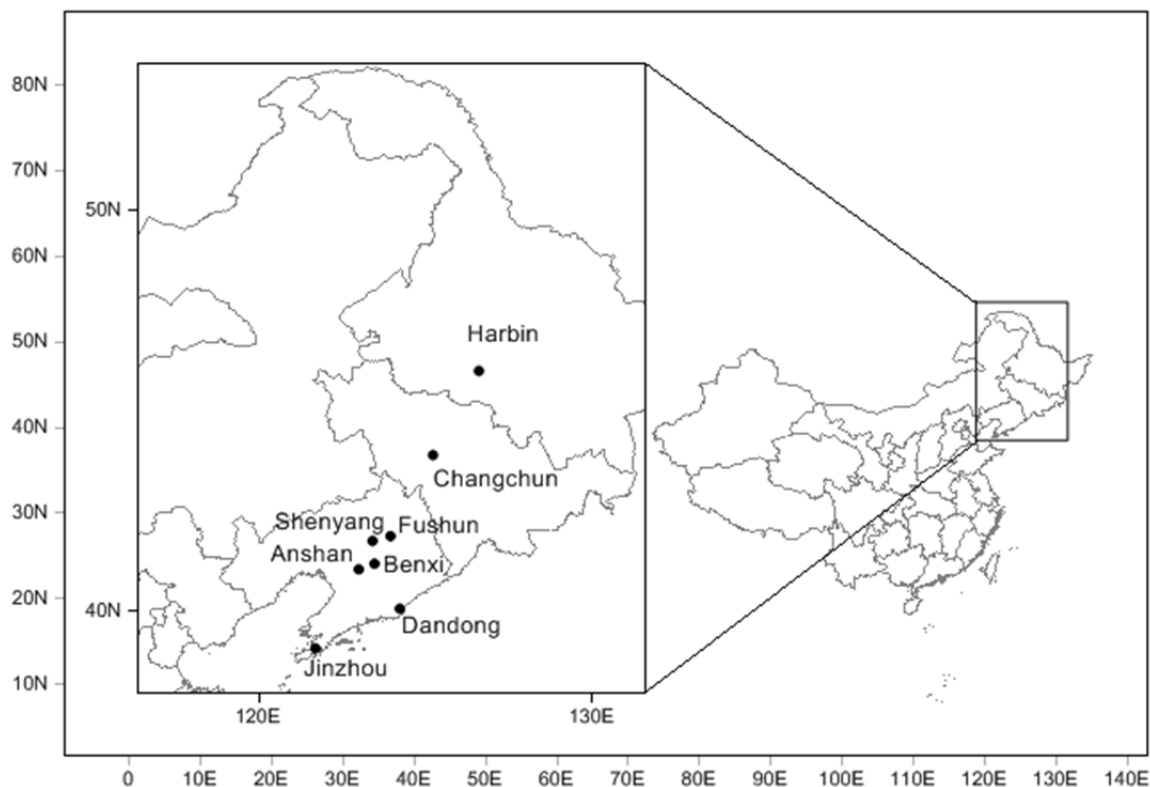


Fig. 1. Distribution of the eight sites in Northeast China.

pollution episodes during the autumn of 2013 are classified. The start times, end times and duration of each pollution episode were as follows: (1) October 16th to 25th; (2) October 26th to 31st; (3) November 1st to 9th; (4) November 10th to 17th; and (5) November 18th to 27th.

RESULTS AND DISCUSSION

Characteristics of PM Concentration during Pollution Episodes

Fig. 2 shows a chronological series of daily average PM concentrations from October 13th to November 30th 2013. The average values of PM concentrations during pollution episodes are presented in Table 1. PM_{2.5} concentrations at two coastal sites of Dandong and Jinzhou were smaller than the others during all pollution episodes. High PM_{2.5} concentrations (> 75 µg/m³) mainly occurred at four sites of Liaoning Province where there were intense industrial and anthropogenic activities.

As shown in Fig. 2, the concentrations of PM₁₀, PM_{2.5} and PM_{1.0} on polluted days were higher than on clean days. The most intense pollution episode occurred between October 16th and 25th. The highest PM concentration occurred between October 20th and 22nd at all eight sites. Looking at PM_{2.5} for Pollution Episode 1 (PE1; for pollution episodes read PE1,

PE2 etc.) at all eight sites, the maximum daily averages of PM_{2.5} were 436.79 ± 85.24 µg/m³ in Harbin and 322.08 ± 50.21 µg/m³ in Shenyang. The lowest value of PM_{2.5} was 74.86 ± 27.59 µg/m³ in Dandong. During the five pollution episodes, most of the daily PM_{2.5} concentrations exceeded China's national ambient air quality standards (75 µg/m³).

PM concentrations in Jinzhou and Dandong were lower among the eight sites. The PM₁₀ concentrations were relatively stable at ca. 150 µg/m³. This may be caused by geographical and meteorological factors; coastal cities have better air quality than inland cities due to the diffusion of pollutants (Yang *et al.*, 2005; Liu and Yang, 2007).

Among the four 'multi-cities' in central Liaoning Province, the average PM concentrations were higher, especially in Shenyang. The maximum PM₁₀ was 803.20 ± 621.24 µg/m³ which is also the highest of the all eight cities. In particular, the PM₁₀ concentration in Shenyang reached 5.33 times China's national ambient air quality standards (150 µg/m³) (GB3095-2012: http://kjs.mep.gov.cn/hjbhbz/bzwb/dqjhjhb/dqjzlbz/201203/t20120302_224165.htm). This result can be attributed to human activities (Zhao *et al.*, 2013b).

PM in Harbin was highest on October 20th 2013, while in Changchun, the maximum PM value occurred on October 22nd 2013, most likely resulting from pollutant accumulation. The maximum value recorded in the central Liaoning

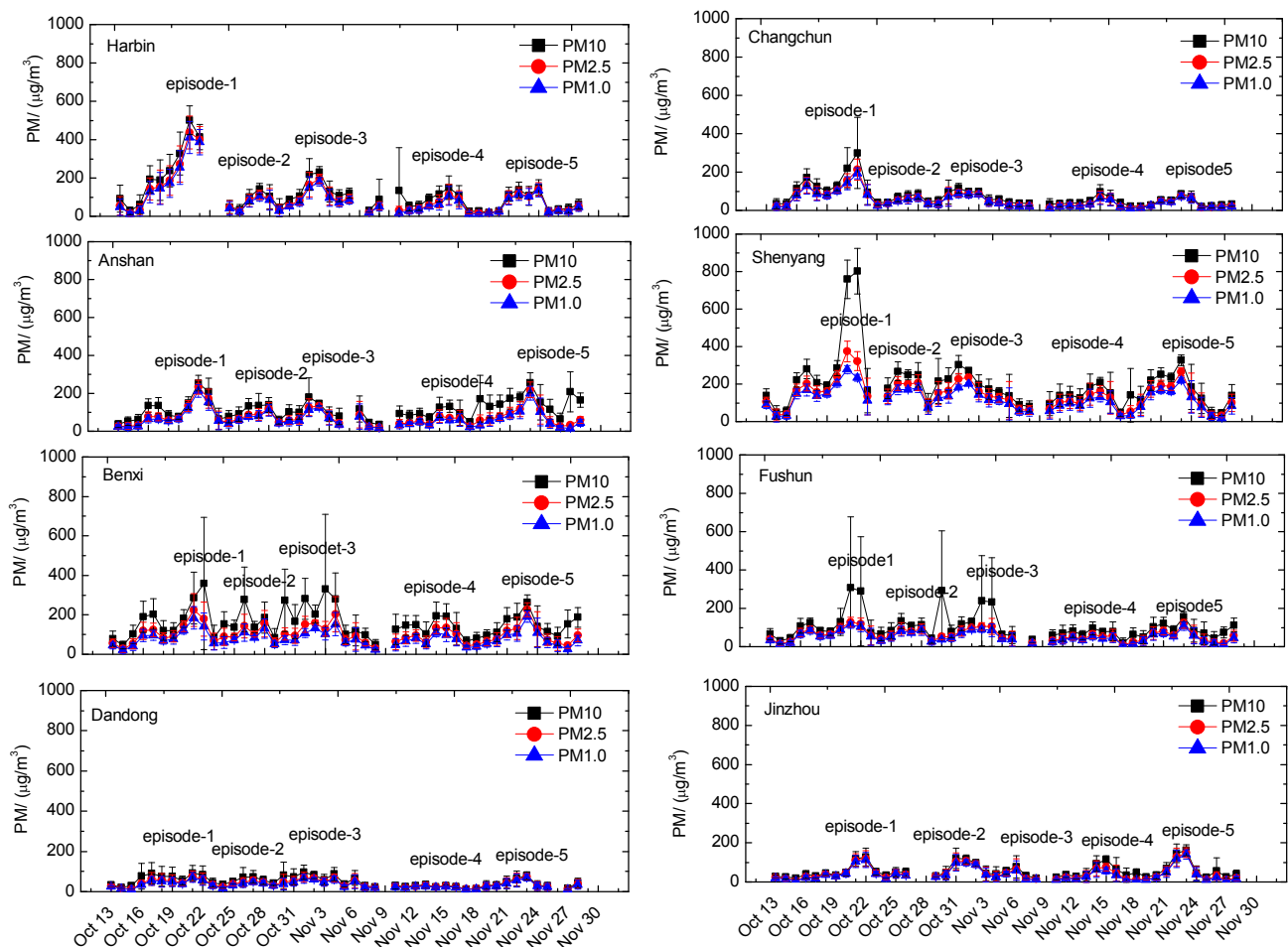


Fig. 2. Daily variations in PM concentrations during pollution episodes.

Table 1. Average PM₁₀, PM_{2.5}, PM₁ concentrations for all pollution episodes.

	PM	Harbin		Changchun		Anshan		Shenyang		Benxi		Fushun		Dandong		Jinzhou	
		PM ₁₀ (μg/m ³)	PM _{2.5} (μg/m ³)	PM ₁₀ (μg/m ³)	PM _{2.5} (μg/m ³)	PM ₁₀ (μg/m ³)	PM _{2.5} (μg/m ³)	PM ₁₀ (μg/m ³)	PM _{2.5} (μg/m ³)	PM ₁₀ (μg/m ³)	PM _{2.5} (μg/m ³)	PM ₁₀ (μg/m ³)	PM _{2.5} (μg/m ³)	PM ₁₀ (μg/m ³)	PM _{2.5} (μg/m ³)	PM ₁₀ (μg/m ³)	PM _{2.5} (μg/m ³)
PE1	PM ₁₀ (μg/m ³)	243.65 ± 71.88	133.69 ± 54.82	127.66 ± 38.59	344.05 ± 176.14	183.71 ± 92.20	134.67 ± 96.36	68.18 ± 36.89	57.86 ± 21.64								
	PM _{2.5} (μg/m ³)	207.98 ± 65.05	100.86 ± 31.18	99.18 ± 27.91	211.12 ± 43.31	120.36 ± 43.32	78.00 ± 24.75	48.72 ± 21.57	50.19 ± 18.71								
	PM _{1.0} (μg/m ³)	195.63 ± 62.64	94.57 ± 27.74	89.00 ± 24.72	172.28 ± 29.74	97.10 ± 32.17	69.86 ± 20.76	42.78 ± 17.47	46.06 ± 17.25								
PE2	PM ₁₀ (μg/m ³)	95.85 ± 37.46	69.83 ± 29.39	111.22 ± 39.88	218.41 ± 62.24	187.37 ± 93.64	127.42 ± 77.42	66.04 ± 35.88	61.72 ± 28.34								
	PM _{2.5} (μg/m ³)	76.06 ± 28.81	54.09 ± 23.97	76.97 ± 23.87	168.10 ± 40.30	109.38 ± 38.56	66.27 ± 20.76	46.29 ± 21.71	51.46 ± 23.07								
	PM _{1.0} (μg/m ³)	69.52 ± 26.04	51.18 ± 22.94	69.49 ± 21.53	142.81 ± 29.98	86.49 ± 27.81	57.75 ± 18.21	40.26 ± 17.37	47.54 ± 21.35								
PE3	PM ₁₀ (μg/m ³)	132.40 ± 48.00	67.61 ± 17.80	99.81 ± 42.39	176.51 ± 45.69	182.84 ± 105.83	126.03 ± 85.47	60.15 ± 24.16	59.92 ± 26.07								
	PM _{2.5} (μg/m ³)	102.09 ± 29.98	52.63 ± 14.75	73.05 ± 28.22	140.97 ± 36.05	111.59 ± 37.87	71.42 ± 21.66	47.79 ± 19.08	50.86 ± 22.51								
	PM _{1.0} (μg/m ³)	93.09 ± 27.70	50.21 ± 14.47	65.62 ± 25.16	119.16 ± 29.91	87.65 ± 27.68	61.49 ± 18.38	43.23 ± 17.53	46.68 ± 20.95								
PE4	PM ₁₀ (μg/m ³)	91.83 ± 57.68	50.05 ± 22.74	93.40 ± 36.01	137.15 ± 55.64	138.28 ± 59.63	68.04 ± 31.50	25.95 ± 10.56	53.89 ± 28.37								
	PM _{2.5} (μg/m ³)	57.92 ± 25.93	33.63 ± 16.58	53.41 ± 20.20	105.97 ± 44.64	89.70 ± 34.35	42.83 ± 18.82	23.14 ± 9.56	35.58 ± 20.48								
	PM _{1.0} (μg/m ³)	49.32 ± 22.99	30.89 ± 16.21	45.31 ± 17.71	88.58 ± 36.66	70.82 ± 25.29	36.28 ± 15.75	21.90 ± 9.18	29.15 ± 17.86								
PE5	PM ₁₀ (μg/m ³)	69.04 ± 25.22	41.12 ± 13.74	159.49 ± 64.79	169.75 ± 60.03	142.28 ± 53.32	85.58 ± 38.18	38.35 ± 20.24	63.56 ± 27.38								
	PM _{2.5} (μg/m ³)	60.82 ± 21.57	33.79 ± 11.17	85.83 ± 31.99	127.24 ± 39.99	96.31 ± 36.07	52.81 ± 22.67	34.30 ± 18.29	47.58 ± 18.85								
	PM _{1.0} (μg/m ³)	57.25 ± 20.11	32.32 ± 10.92	72.04 ± 26.79	107.02 ± 32.73	79.22 ± 29.59	45.00 ± 19.57	32.49 ± 17.60	42.49 ± 16.48								

Province ‘multi-cities’ occurred on October 21st and 22nd 2013. PM in Jinzhou was highest on October 22nd 2013. The above results show that a possible pollution transition process may exist. It is noted that the daily averaged PM is a more accurate tool for detecting and tracking pollution episodes.

Fig. 3 and Table 2 show PM_{2.5}/PM₁₀, PM_{1.0}/PM₁₀ and PM_{1.0}/PM_{2.5} ratios for days experiencing pollution. The accumulation of pollutants and the formation of secondary pollutants were the main factors causing pollution episodes. The effects of these two factors are better identified by comparing the concentration ratios of PM with different diameters.

The ratios of PM_{2.5}/PM₁₀ for Harbin and Jinzhou were 0.83 ± 0.07 and 0.85 ± 0.09 during PE1, much higher than those at the other sites. The PM_{2.5}/PM₁₀ ratios in Benxi and Fushun were low during all five pollution episodes, with means of 0.68 ± 0.10 and 0.66 ± 0.14, respectively.

PM_{1.0}/PM_{2.5} ratios in Harbin and Changchun during PE1 reached ca. 0.93 ± 0.03 and 0.95 ± 0.06, respectively. This result shows that PM_{1.0} were dominant in Northeast China during the pollution episodes.

Coarse atmospheric particles are usually considered to come mainly from natural sources such as dust; fine particles are mostly attributed to anthropogenic activities, including both primary emission and the secondary formation of particles from gaseous pollutants (Sun *et al.*, 2013). The high PM_{1.0}/PM_{2.5} ratio on pollution days highlights the increased contribution of anthropogenic pollutants, as observed by many field measurements (Tan *et al.*, 2009; Du *et al.*, 2011; Yin *et al.*, 2012).

Analysis of PM and Meteorological Data

Fig. 4 shows that in Anshan 37.50% of the daily visibility averages are < 10.0 km, and only 14.58% are > 20.0 km. At Shenyang site, >50% of the daily visibility averages are < 10.0 km, indicating poor air quality in this region. The daily visibility average in Dandong is similar to that of Anshan; 40.82% of the daily visibility averages are < 10.0 km and only 20.41% are > 20.0 km.

It has been demonstrated by previous studies that atmospheric visibility is greatly influenced by particulate pollutants in Northeast China (Liu *et al.*, 2010; Zhao *et al.*, 2013c). The relation between visibility and PM mass concentrations has been plotted in Fig. 5, and a fitting nonlinear equation is also given.

There is a better non-linear relation between visibility and PM mass concentrations over Shenyang, and the results are similar to the graph produced by Jing *et al.* (2014). This relation shows that visibility decreased exponentially as PM concentrations increased. The correlation coefficients (R²) between PM_{2.5} and visibility are 0.71, 0.86 and 0.48 for Anshan, Shenyang, and Dandong, respectively and the correlation coefficients (R²) between PM_{1.0} and visibility are 0.73, 0.89 and 0.50, respectively. The results suggested that there is a better correlation between PM and visibility in Shenyang, following by Anshan and Dandong. The high correlation between PM_{2.5}/PM_{1.0} concentrations and visibility indicates the seriousness of fine particle pollution in Shenyang. These observations suggest that higher

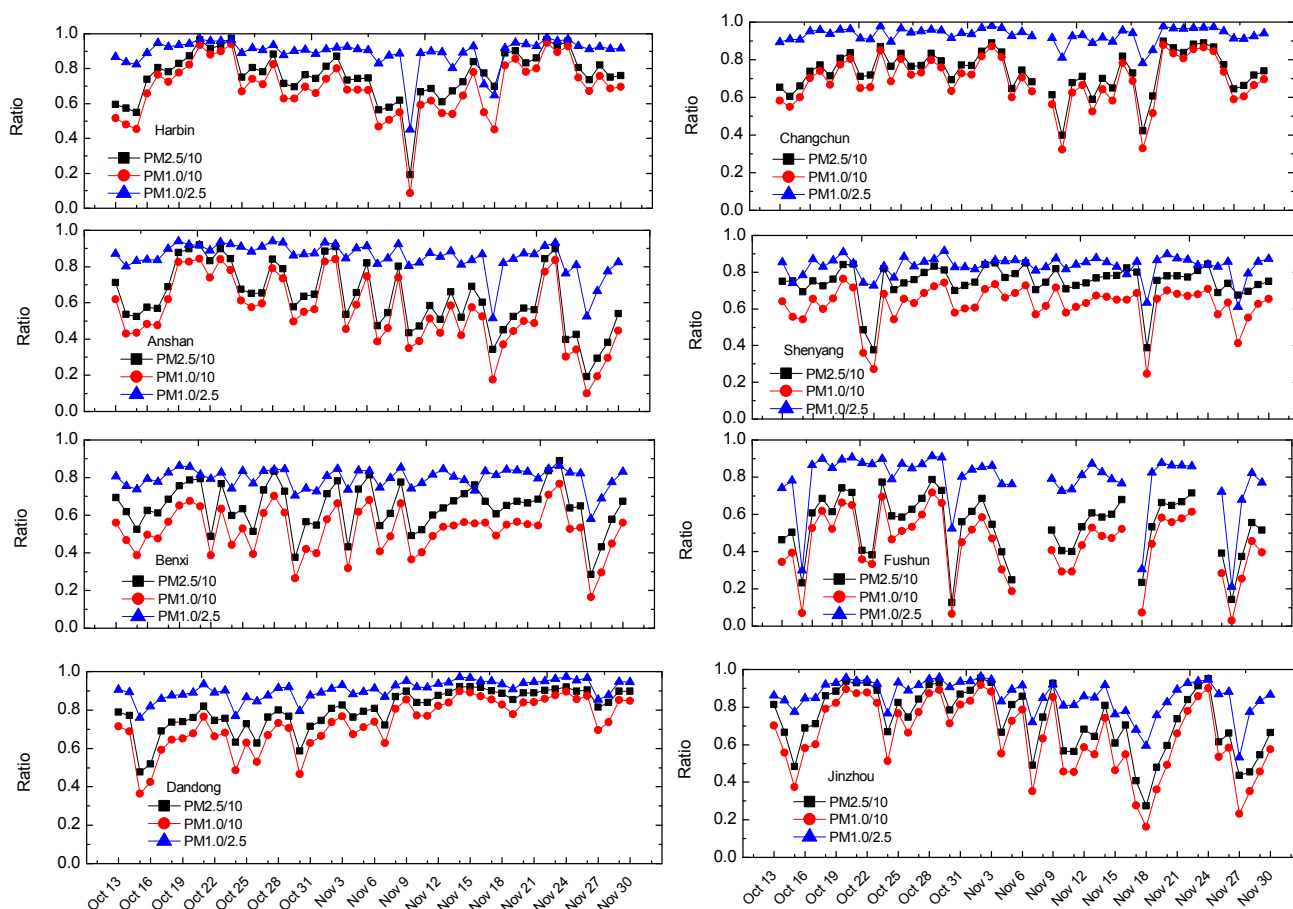


Fig. 3. Daily variations in PM ratios during pollution episodes.

Table 2. $PM_{2.5}/PM_{10}$, $PM_{1.0}/PM_{10}$, $PM_{1.0}/PM_{2.5}$ ratios for all pollution episodes.

		Harbin	Changchun	Anshan	Shenyang	Benxi	Fushun	Dandong	Jinzhou
PE1	$PM_{2.5}/PM_{10}$	0.83 ± 0.07	0.77 ± 0.16	0.76 ± 0.14	0.71 ± 0.16	0.68 ± 0.10	0.66 ± 0.14	0.71 ± 0.09	0.85 ± 0.09
	$PM_{1.0}/PM_{10}$	0.77 ± 0.09	0.74 ± 0.16	0.68 ± 0.15	0.60 ± 0.16	0.56 ± 0.10	0.60 ± 0.14	0.63 ± 0.10	0.78 ± 0.11
	$PM_{1.0}/PM_{2.5}$	0.93 ± 0.03	0.95 ± 0.06	0.89 ± 0.04	0.84 ± 0.06	0.82 ± 0.03	0.90 ± 0.01	0.88 ± 0.03	0.91 ± 0.04
PE2	$PM_{2.5}/PM_{10}$	0.73 ± 0.07	0.75 ± 0.05	0.69 ± 0.10	0.77 ± 0.05	0.63 ± 0.17	0.64 ± 0.23	0.67 ± 0.09	0.83 ± 0.06
	$PM_{1.0}/PM_{10}$	0.66 ± 0.08	0.70 ± 0.07	0.63 ± 0.11	0.66 ± 0.07	0.50 ± 0.17	0.57 ± 0.23	0.57 ± 0.13	0.76 ± 0.08
	$PM_{1.0}/PM_{2.5}$	0.90 ± 0.02	0.94 ± 0.03	0.90 ± 0.03	0.85 ± 0.03	0.79 ± 0.06	0.86 ± 0.09	0.86 ± 0.05	0.92 ± 0.02
PE3	$PM_{2.5}/PM_{10}$	0.75 ± 0.09	0.79 ± 0.05	0.69 ± 0.15	0.79 ± 0.05	0.63 ± 0.14	0.60 ± 0.14	0.81 ± 0.05	0.85 ± 0.10
	$PM_{1.0}/PM_{10}$	0.69 ± 0.08	0.75 ± 0.06	0.61 ± 0.16	0.66 ± 0.06	0.50 ± 0.12	0.52 ± 0.14	0.74 ± 0.06	0.77 ± 0.12
	$PM_{1.0}/PM_{2.5}$	0.91 ± 0.01	0.95 ± 0.03	0.88 ± 0.04	0.84 ± 0.03	0.79 ± 0.04	0.85 ± 0.04	0.91 ± 0.02	0.91 ± 0.05
PE4	$PM_{2.5}/PM_{10}$	0.65 ± 0.19	0.65 ± 0.04	0.57 ± 0.08	0.77 ± 0.04	0.64 ± 0.09	0.61 ± 0.09	0.89 ± 0.03	0.64 ± 0.13
	$PM_{1.0}/PM_{10}$	0.54 ± 0.19	0.59 ± 0.03	0.48 ± 0.08	0.64 ± 0.03	0.51 ± 0.07	0.51 ± 0.09	0.84 ± 0.04	0.52 ± 0.14
	$PM_{1.0}/PM_{2.5}$	0.81 ± 0.14	0.90 ± 0.03	0.85 ± 0.03	0.84 ± 0.03	0.79 ± 0.04	0.84 ± 0.04	0.95 ± 0.02	0.81 ± 0.08
PE5	$PM_{2.5}/PM_{10}$	0.86 ± 0.09	0.78 ± 0.13	0.52 ± 0.20	0.72 ± 0.13	0.66 ± 0.16	0.58 ± 0.18	0.88 ± 0.04	0.65 ± 0.23
	$PM_{1.0}/PM_{10}$	0.79 ± 0.14	0.74 ± 0.16	0.43 ± 0.22	0.59 ± 0.16	0.54 ± 0.16	0.49 ± 0.19	0.83 ± 0.08	0.56 ± 0.26
	$PM_{1.0}/PM_{2.5}$	0.92 ± 0.09	0.94 ± 0.11	0.79 ± 0.16	0.81 ± 0.11	0.81 ± 0.08	0.81 ± 0.12	0.93 ± 0.05	0.82 ± 0.12

concentrations of fine particles are significantly associated with visibility degradation.

The overall averages of RH, wind speed (V), pressure (P), temperature (T) and total water precipitation (TWP) over Northeast China during the five episodes are presented in Table 3.

Meteorological conditions were quite stable during PE1

and PE2. The average V was almost < 2.0 m/s and the average RH was ~ 71 – 72% . The average precipitation during the two episodes was 2.78 ± 1.93 mm and 3.82 ± 2.95 mm, respectively. During PE3, PE4 and PE5, wind speed increased, while RH decreased slightly. The average V was 2.31 m/s, 2.80 m/s and 2.38 m/s during these three episodes and the average RH was 68.82%, 62.09% and 62.05%,

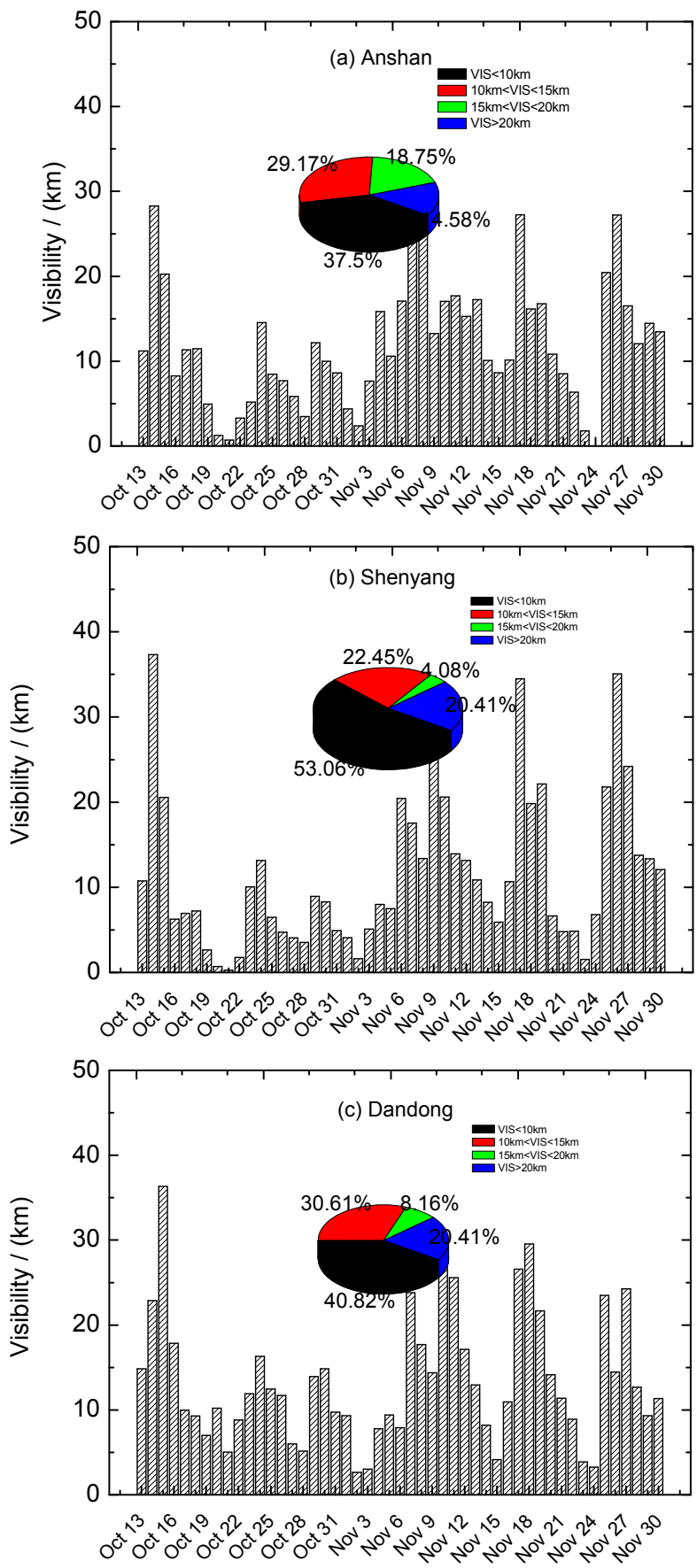


Fig. 4. Daily variations in visibility during pollution episodes at (a) Anshan, (b) Shenyang, and (c) Dandong.

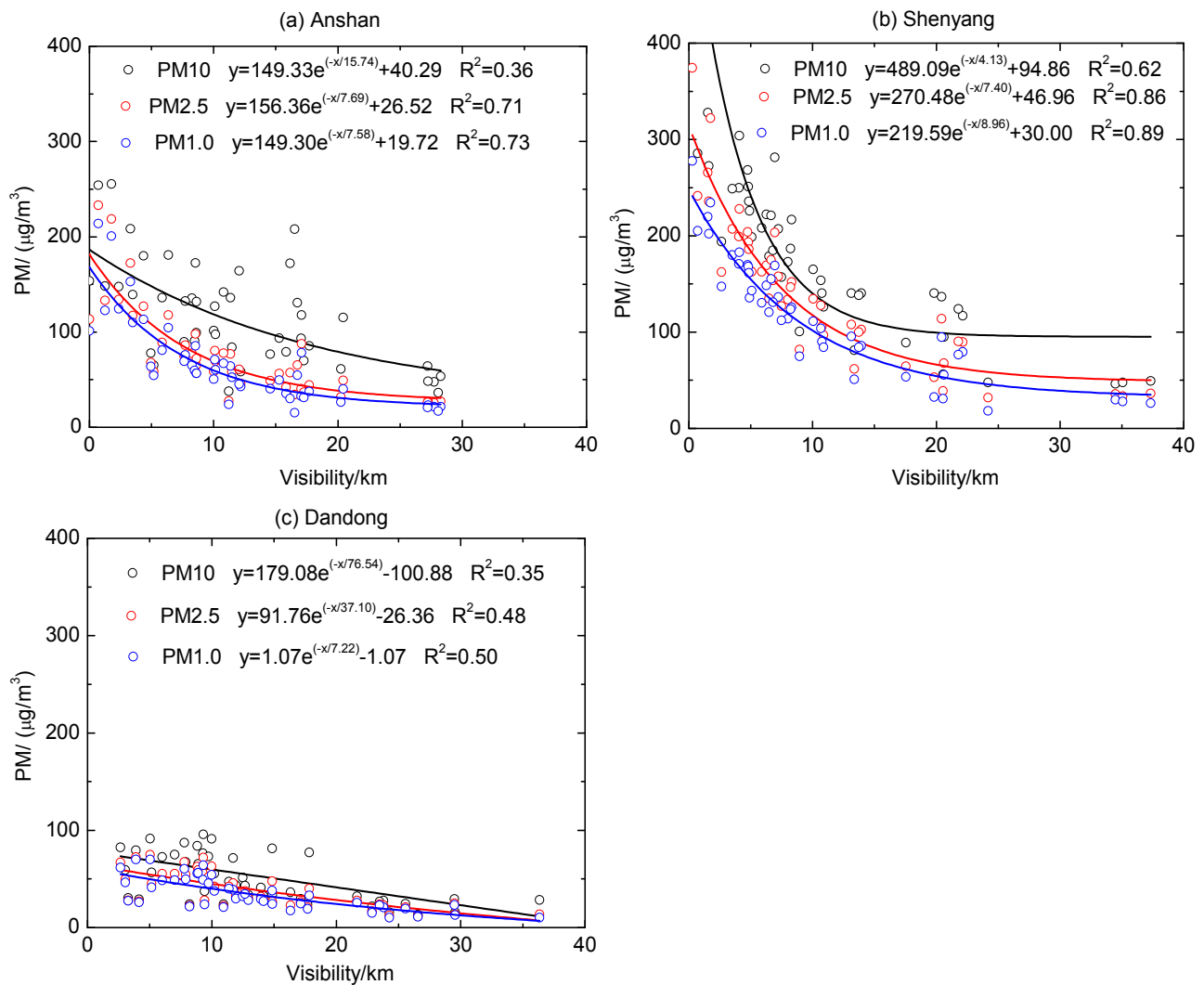


Fig. 5. Correlation between visibility and PM at (a) Anshan, (b) Shenyang, and (c) Dandong.

Table 3. Meteorological parameters for all pollution episodes.

	Pressure (Pa)	Temperature (°C)	RH (%)	V (m/s)	Precipitation (mm)
PE1	1014.47 ± 7.46	9.21 ± 1.93	72.92 ± 6.93	1.92 ± 0.34	2.78 ± 1.93
PE2	1013.96 ± 7.51	7.72 ± 2.39	71.17 ± 9.21	1.95 ± 0.38	3.82 ± 2.95
PE3	1012.13 ± 7.82	8.91 ± 1.86	68.82 ± 6.03	2.31 ± 0.32	2.05 ± 1.16
PE4	1008.98 ± 7.91	3.21 ± 2.16	62.09 ± 9.28	2.80 ± 0.36	1.29 ± 1.39
PE5	1008.95 ± 7.83	-0.29 ± 2.59	62.05 ± 9.52	2.38 ± 0.35	2.15 ± 1.15

respectively. Compared to PE1 and PE2, precipitation was less in the later three episodes, with averages of 2.05 mm, 1.29 mm and 2.15 mm, respectively. Lower temperatures, with an average below 0°C, were observed in PE5.

In general, the temporal variation of V showed a negative correlation with PM concentrations (Table 4). Both low and high V affected the accumulation and diffusion of aerosol particles during pollution days (Che *et al.*, 2014). RH also correlated with PM concentrations. It was found that high RH was conducive to haze formation in Northeast China. Hennigan *et al.* (2008) identified that the formation of secondary aerosol species could be enhanced in high-humidity conditions. However, the washout effect of rainfall

may result another negative correlation between PM and RH in some cases.

Variations in PM and Relative Meteorological Factors during Pollution Episodes

Based on the classification of haze in China (visibility < 10 km, RH < 90%) (CMA, 2003), during the entire study period there were 16, 22, and 18 hazy days in Anshan, Shenyang, and Dandong, respectively. The average daily visibility at these three sites is shown in Fig. 6 together with V and RH. The gray bars represent hazy days. Generally, lower V and higher RH were observed on hazy days. Table 5 shows the average variations in PM concentrations and

Table 4. Correlation between PM and meteorological parameters at Harbin, Shenyang, Benxi, Fushun, Dandong and Jinzhou.

	PE1	PE2	PE3	PE4	PE5	
PM ₁₀ /PM _{2.5} /PM _{1.0} & RH	Harbin	0.65/0.71/0.72	-0.77/-0.66/-0.65	0.64/0.68/0.69	-0.55/-0.46/-0.48	0.62/0.68/0.69
	Shenyang	0.38/0.46/0.45	-0.25/-0.10/-0.04	0.53/0.59/0.65	0.03/-0.02/-0.11	0.83/0.85/0.85
	Benxi	0.03/0.22/0.26	-0.09/-0.36/-0.34	0.59/0.56/0.60	-0.05/0.09/0.01	0.12/0.50/0.56
	Fushun	0.23/0.20/0.20	-0.15/0.08/0.13	0.90/0.87/0.85	-0.44/-0.49/-0.57	0.41/0.67/0.69
	Dandong	0.04/0.37/0.42	-0.51/0.02/0.26	0.21/0.27/0.29	0.04/0.10/0.14	0.50/0.53/0.56
PM ₁₀ /PM _{2.5} /PM _{1.0} & V	Jinzhou	0.61/0.66/0.67	0.28/0.36/0.38	0.64/0.72/0.74	0.23/0.46/0.49	0.50/0.56/0.58
	Harbin	0.17/0.15/0.14	0.29/0.34/0.33	-0.48/-0.43/-0.43	-0.43/-0.33/-0.34	-0.52/-0.50/-0.49
	Shenyang	-0.35/-0.41/-0.40	-0.66/-0.53/-0.48	-0.70/-0.66/-0.66	-0.69/-0.83/-0.88	-0.85/-0.85/-0.87
	Benxi	-0.39/-0.64/-0.67	-0.48/0.16/0.31	-0.86/-0.70/-0.65	-0.69/-0.71/-0.71	-0.32/-0.67/-0.71
	Fushun	-0.51/-0.68/-0.71	-0.68/-0.50/-0.36	-0.83/-0.86/-0.84	-0.75/-0.88/-0.93	-0.59/-0.70/-0.72
Dandong		-0.74/-0.62/-0.58	-0.66/-0.81/-0.75	-0.30/-0.33/-0.34	-0.88/-0.88/-0.88	-0.66/-0.66/-0.66
	Jinzhou	-0.12/-0.11/-0.12	-0.67/-0.61/-0.59	-0.88/-0.88/-0.88	-0.31/-0.56/-0.60	-0.60/-0.59/-0.57

meteorological factors for clear days, hazy days, and the entire study period. On hazy days, average visibility was 5.7 ± 2.7 km, 5.5 ± 2.2 km and 7.4 ± 2.2 km at Anshan, Shenyang and Dandong, respectively. Visibility on the non-hazy days was ~ 2.5 – 3.0 times higher than that on hazy days. In contrast, the mean fine particle (PM_{2.5} and PM_{1.0}) concentrations on hazy days were ~ 1.6 – 2.2 times higher than those on non-hazy days. Therefore, it is evident that the extremely high concentrations of airborne particles during hazy days were the most significant cause of bad air quality.

The PM_{1.0}/PM_{2.5} ratios during hazy days were 0.89 ± 0.04 , 0.85 ± 0.04 and 0.91 ± 0.04 in Anshan, Shenyang and Dandong, respectively. This suggests that PM_{1.0} are dominant during hazy days.

The means of RH during hazy days were $61.7 \pm 15.2\%$, $77.2 \pm 8.8\%$ and $76.1 \pm 8.4\%$ for Anshan, Shenyang and Dandong, respectively, much higher than the values on non-hazy days. The average V for the aforementioned three cities during hazy days was lower compared to non-hazy days, with mean values of 1.8 ± 0.6 , 1.5 ± 0.6 , and 2.4 ± 0.6 m/s. Weather conditions with high RH and low V during hazy days facilitate the accumulation of atmospheric pollutants, resulting in increased PM concentrations and a deterioration in air quality (Sun *et al.*, 2006; Fan *et al.*, 2009; Li and Shao, 2010; Quan *et al.*, 2013). Moreover, hygroscopic growth under favorable weather conditions enhances the effect that particles have on air quality.

SUMMARY

PM characteristics in Northeast China during one and a half months of intense pollution between October 13th and November 30th 2013 are summarized below.

The eight sites in Northeast China experienced five major pollution episodes during the autumn of 2013. The maximum daily PM_{2.5} averages were 436.79 ± 85.24 $\mu\text{g}/\text{m}^3$ in Harbin and 322.08 ± 50.21 $\mu\text{g}/\text{m}^3$ in Shenyang. The lowest recorded value of daily PM_{2.5} was 74.86 ± 27.59 $\mu\text{g}/\text{m}^3$, in Dandong. These results shows a possible pollution transition process which firstly occurred in Harbin with the highest aerosol loading on October 20th 2013, then happened in Changchun and the central Liaoning Province ‘multi-cities’ with a higher PM level occurred on October 21st and 22nd 2013, while in Jinzhou the highest PM concentration appeared on October 22nd 2013.

PM_{1.0}/PM_{2.5} ratios at Changchun and Harbin are higher than those results at the majority of the other sites. The results indicate that there is a greater proportion of PM_{1.0} during pollution periods in Northeast China urban areas.

The values of R² between PM_{2.5} and visibility were 0.71, 0.86 and 0.48 in Anshan, Shenyang, and Dandong, respectively. Those between PM_{1.0} and visibility were 0.73, 0.89 and 0.50, respectively. These observations suggest that higher concentrations of fine particles are significantly related to visibility degradation.

Visibility on non-hazy days is nearly 2.5–3.0 times that of hazy days. Observations of relative meteorological conditions show that high RH and lower V during hazy days may facilitate the accumulation of atmospheric pollutants.

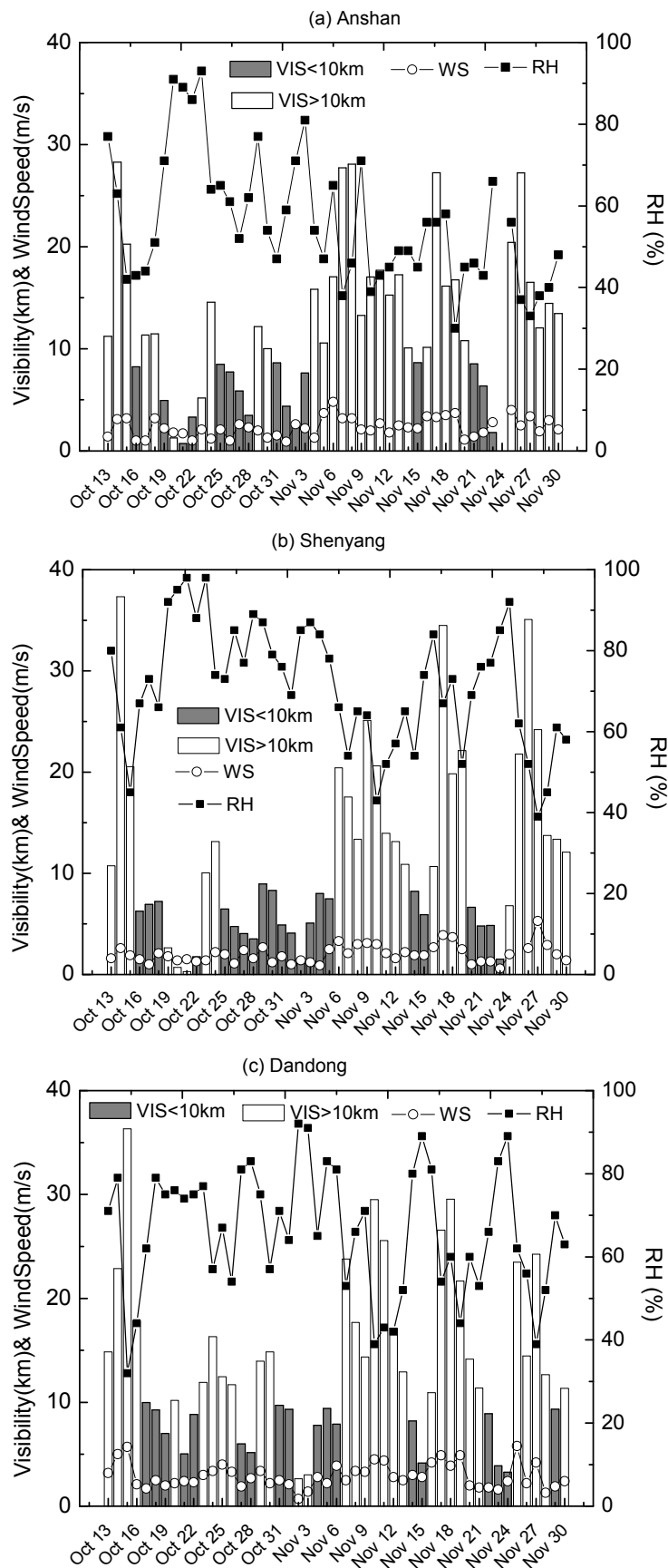


Fig. 6. Daily variations in visibility and meteorological elements during pollution periods at (a) Anshan, (b) Shenyang, (c) Dandong.

Table 5. PM and meteorological parameters during pollution and non-pollution episodes at Anshan, Shenyang and Dandong.

	Whole Period			Pollution days			Non-pollution days		
	Anshan	Shenyang	Dandong	Anshan	Shenyang	Dandong	Anshan	Shenyang	Dandong
PM ₁₀ (µg/m ³)	115.4 ± 50.5	172.0 ± 78.2	48.1 ± 23.9	152.9 ± 52.7	225.6 ± 52.1	64.6 ± 20.3	96.7 ± 37.9	128.3 ± 68.5	38.6 ± 20.6
PM _{2.5} (µg/m ³)	73.0 ± 45.8	138.5 ± 76.4	37.5 ± 17.4	112.6 ± 52.9	180.5 ± 52.0	51.2 ± 14.2	53.2 ± 24.7	104.3 ± 76.8	29.5 ± 13.8
PM _{1.0} (µg/m ³)	63.6 ± 43.1	115.7 ± 61.6	33.9 ± 15.8	101.2 ± 49.3	152.1 ± 38.1	46.6 ± 12.7	44.8 ± 23.5	85.9 ± 61.5	26.6 ± 12.4
Visibility (km)	12.3 ± 7.3	11.5 ± 9.0	13.5 ± 7.8	5.7 ± 2.7	5.5 ± 2.2	7.4 ± 2.2	15.7 ± 6.6	16.5 ± 9.5	17.1 ± 7.6
RH (%)	56.0 ± 15.8	71.3 ± 15.1	66.0 ± 15.2	61.7 ± 15.2	77.2 ± 8.8	76.1 ± 8.4	53.1 ± 15.6	66.4 ± 17.5	60.1 ± 15.3
Wind speed (m/s)	2.3 ± 0.9	2.0 ± 0.9	2.9 ± 1.2	1.8 ± 0.6	1.5 ± 0.6	2.4 ± 0.6	2.6 ± 1.0	2.5 ± 0.9	3.2 ± 1.3
PM _{1.0} /PM _{2.5}	0.85 ± 0.09	0.83 ± 0.06	0.91 ± 0.05	0.89 ± 0.04	0.85 ± 0.04	0.91 ± 0.04	0.83 ± 0.10	0.82 ± 0.07	0.90 ± 0.05

ACKNOWLEDGMENTS

This work is financially supported by grants from the National Key Project of Basic Research (2014CB441201), the National Natural Science Foundation of China (41275167, 41475124, 41305128), and the project of Institute of Atmospheric Environment programme (2014IAE-cma05).

REFERENCES

- Che, H.Z., Zhang, X.Y., Li, Y., Zhou, Z.J., Qu, J.J. and Hao, X.J. (2009). Haze Trends over the Capital Cities of 31 Provinces in China, 1981–2005. *Theor. Appl. Climatol.* 97: 235–242.
- Che, H., Xia, X., Zhu, J., Li, Z., Dubovic, O., Holben, B., Goloub, P., Chen, H., Estelles, V., Cuevas-Agulló, E., Blarel, L., Wang, H., Zhao, H., Zhang, X., Wang, Y., Sun, J., Tao, R. and Shi, G. (2014). Column Aerosol Optical Properties and Aerosol Radiative Forcing during a Serious Haze-fog Month over North China Plain in 2013 Based on Ground-based Sunphotometer Measurements. *Atmos. Chem. Phys.* 14: 2125–2138.
- Cheng, T., Han, Z., Zhang, R., Du, H., Wang, J. and Yao, J. (2010). Black Carbon in a Continental Semi-arid Area of Northeast China and Its Possible Sources of Fire Emission. *J. Geophys. Res.* 115, doi: 10.1029/2009JD013523.
- Cheng, Z., Wang, S.X., Jiang, J.K., Fu, Q., Chen, C., Xu, B., Yu, J., Fu, X. and Hao, J. (2013). Long-term Trend of Haze Pollution and Impact of Particulate Matter in the Yangtze River Delta, China. *Environ. Pollut.* 182: 101–110.
- CMA (2003). Specifications for the Surface Meteorological Observations. Meteorological Press, Beijing, China (in Chinese).
- Deng, J.J., Wang, T.J., Jiang, Z.Q., Min, X., Zhang, R., Huang, X. and Zhu, J. (2011). Characterization of Visibility and Its Affecting Factors over Nanjing, China. *Atmos. Res.* 101: 681–691.
- Dillner, A.M., Schauer, J.J., Zhang, Y., Zeng, L. and Cass, G.R. (2006). Size-resolved Particulate Matter Composition in Beijing during Pollution and Dust Events. *J. Geophys. Res.* 111, doi: 10.1029/2005JD006400.
- Du, J., Cheng, T., Zhang, M., Chen, J., He, Q., Wang, X., Zhang, R., Tao, J., Huang, G., Li, X. and Zha, S. (2012). Aerosol Size Spectra and Particle Formation Events at Urban Shanghai in Eastern China. *Aerosol Air Qual. Res.* 12: 1362–1372.
- Du, H., Kong, L., Cheng, T., Chen, J., Du, J., Li, L., Xia, X., Leng, C. and Hang, G. (2011). Insights into Summertime Haze Pollution Events over Shanghai Based on Online Water-soluble Ionic Composition of Aerosols. *Atmos. Environ.* 45: 5131–5137.
- Elias, T., Haefelin, M., Drobinski, P., Gomes, L., Rangognio, J., Bergot, T., Chazette, P., Raut, J.C. and Colomb, M. (2009). Particulate Contribution to Extinction of Visible Radiation: Pollution, Haze and Fog. *Atmos. Res.* 92: 443–454.
- Fan, X.Q. and Sun, Z.B. (2009). Analysis on Features of Haze Weather in Xiamen City during 1953-2008. *Trans.*

- Atmos. Sci.* 5: 604–609 (in Chinese).
- Franke, K., Ansmann, A. and Müller, D. (2003). Optical Properties of Indo-Asian Haze Layer over the Tropical Indian Ocean. *J. Geophys. Res.* 108, doi: 10.1029/2002JD002473.
- Hennigan, C.J., Bergin, M.H., Dibb, J.E. and Weber, R.J. (2008). Enhanced Secondary Organic Aerosol Formation Due to Water Uptake by Fine Particles. *Geophys. Res. Lett.* 35, doi: 10.1029/2008GL035046.
- Hu, M., He, L., Zhang, Y., Wang, M., Kim, Y.P. and Moon, K.C. (2002). Seasonal Variation of Ionic Species in Fine Particles at Qingdao, China. *Atmos. Environ.* 36: 5853–5859.
- Hyslop, N.P. (2009). Impaired Visibility: The Air Pollution People See. *Atmos. Environ.* 43:182–195.
- IPCC (Intergovernmental Panel on Climate Change) (2001). *Climate Change 2001*. Cambridge University Press, New York, 2001.
- Kang, C.M., Lee, H.S., Kang, B.W., Lee, S.K. and Sunwoo, Y. (2004). Chemical Characteristics of Acidic Gas Pollutants and PM_{2.5} Species during Hazy Episodes in Seoul, South Korea. *Atmos. Environ.* 38:4749–4760.
- Kim, Y.J., Kim, K.W., Kim, S.D., Lee, B.K. and Han, J.S. (2006). Fine Particulate Matter Characteristics and Its Impact on Visibility Impairment at Two Urban Sites in Korea: Seoul and Incheon. *Atmos. Environ.* 40: S593–S605.
- Lawrence, C., Arellano, A. and McGregor, J. (2001). Investigating the Haze Transport from 1997 Biomass Burning in Southeast Asia Its Impact Upon Singapore. *Atmos. Environ.* 35: 2723–2734.
- Li, W. and Shao, L. (2010). Mixing and Water-soluble Characteristics of Particulate Organic Compounds in Individual Urban Aerosol Particles. *J. Geophys. Res.* 115, doi: 10.1029/2009JD012575.
- Liu, N.W., Ma, Y.J., Wang, Y.F. and Liu, X.M. (2010). Mass Concentration Variation of Atmospheric Particles and Relationship with Visibility in Dandong. *Res. Environ. Sci.* 23: 642–646 (in Chinese).
- Lü, H.X., Cai, Q.Y., Wen, S., Chi, Y., Guo, S., Sheng, G., Fu, J. and Blanca, A.L. (2009). Carbonyl Compounds in the Ambient Air of Hazy Days and Clear Days in Guangzhou, China. *Atmos. Res.* 94: 363–372.
- Ma, Y.J., Zuo, H.C., Zhang, Y.H. and Hui, X.Y. (2005). Analyses on Variation Trends of Atmospheric Visibility and Its Effect Factor in Multi-cities in Central Liaoning. *Plateau Meteor.* 24: 623–628 (in Chinese).
- Menon, S., Hansen, J., Nazarenko, L. and Luo, Y. (2002). Climate Effects of Black Carbon Aerosols in China and India. *Science* 297: 2250–2253.
- NRC (1998). *Research Priorities for Airborne Particulate Matter*. National Academy of Sciences/National Research Council, Washington DC.
- Okada, K., Ikegami, M., Zaizen, Y., Makino, Y., Jensen, J.B. and Gras, J.L. (2001). The Mixture State of Individual Aerosol Particles in the 1997 Indonesian Haze Episode. *J. Aerosol Sci.* 32: 1269–1279.
- Pope, C.A., Ezzati, M. and Dockery, D.W. (2009). Fine-Particulate Air Pollution and Life Expectancy in the United States. *New Engl. J. Med.* 360: 376–386.
- Quan, J., Gao, Y., Zhang, Q., Tie, X., Cao, J., Han, S., Meng, J., Chen, P. and Zhao, D. (2013). Evolution of Planetary Boundary Layer under Different Weather Conditions, and Its Impact on Aerosol Concentrations. *Particuology* 11: 34–40.
- Ramanathan, V., Crutzen, P.J., Lelieveld, J., Mitra, A.P., Althausen, D., Anderson, J., Andreae, M.O., Cantrell, W., Cass, G.R., Chung, C.E., Clarke, A.D., Coakley, J.A., Collins, W.D., Conant, W.C., Dulac, F., Heintzenberg, J., Heymsfield, A.J., Holben, B., Howell, S., Hudson, J., Jayaraman, A., Kiehl, J.T., Krishnamurti, T.N., Lubin, D., McFarquhar, G., Novakov, T., Ogren, J.A., Podgorny, I.A., Prather, K., Priestley, K., Prospero, J.M., Quinn, P.K., Rajeev, K., Rasch, P., Rupert, S., Sadourny, R., Satheesh, S.K., Shaw, G.E., Sheridan, P. and Valero, F.P.J. (2001). Indian Ocean Experiment: An Integrated Analysis of the Climate Forcing and Effects of the great Indo-Asia Haze. *J. Geophys. Res.* 106: 28371–28398. doi: 10.1029/2001JD900133.
- Schichtel, B.A., Husar, R.B., Falke, S.R. and Wilson, W.E. (2001). Haze Trends over the United States, 1980–1995. *Atmos. Environ.* 35: 5205–5210.
- Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K.B., Tignor, M. and Miller, H.L. (2007). *Climate Change 2007: The Physical Science Basis. In Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.
- Sun, Y., Zhuang, G., Tang, A., Wang, Y. and An, Z. (2006). Chemical Characteristics of PM_{2.5} and PM₁₀ in Haze-Fog Episodes in Beijing. *Crit. Rev. Env. Sci. Technol.* 40: 3148–3155.
- Sun, Y., Zhou, X., Wai, K., Yuan, Q., Xu, Z., Zhou, S., Qi, Q. and Wang, W. (2013). Simultaneous Measurement of Particulate and Gaseous Pollutants in an Urban City in North China Plain during the Heating Period: Implication of Source Contribution. *Atmos. Res.* 134: 24–34.
- Tie, X., Wu, D. and Brasseur, G. (2009). Lung Cancer Mortality and Exposure to Atmospheric Aerosol Particles in Guangzhou, China. *Atmos. Environ.* 43: 2375–2377.
- US EPA (US Environmental Protection Agency) (1997). *National Ambient Air Quality Standards for Particulate Matters, Final Rule, Federal Register Vol. 62. (Pt. II), EPA, 40CFR (Pt.50), 18 July.*
- Wang, C. (2013). Impact of Anthropogenic Absorbing Aerosols on Clouds and Precipitation: A Review of Recent Progresses. *Atmos. Res.* 122: 237–249.
- Wang, P., Che, H., Zhang, X., Song, Q., Wang, Y., Zhang, Z., Dai, X. and Yu, D. (2010). Aerosol Optical Properties of Regional Background Atmosphere in Northeast China. *Atmos. Environ.* 44:4404–4412.
- Wang, S., Zhang, X. and Xu, X. (2003). Analysis of Variation Features of Visibility and Its Effect Factors in Beijing. *Meteor. Sci. Technol.* 31: 109–114 (in Chinese).
- Wang, S. and Hao, J. (2012). Air Quality Management in China: Issues, Challenges, and Options. *J. Environ. Sci.-China* 24: 2–13.

- Watson, J.G. (2002). Visibility: Science and Regulation. *J. Air Waste Manage. Assoc.* 52: 628–713.
- Xia, X., Chen, H., Goloub, P., Zhang, W., Chatenet, B. and Wang, P. (2007). A Compilation of Aerosol Optical Properties and Calculation of Direct Radiative Forcing over an Urban Region in Northern China. *J. Geophys. Res.* 112, doi: 10.1029/2006JD008119.
- Yang, F., Chen, H., Du, J., Yang, X., Gao, S., Chen, J. and Geng, F. (2012). Evolution of the Mixing State of Fine Aerosols during Haze Events in Shanghai. *Atmos. Res.* 104–105: 193–201.
- Yin, L., Niu, Z., Chen, X., Xu, L. and Zhang, F. (2012). Chemical Compositions of PM_{2.5} Aerosol during Haze Periods in the Mountainous City of Yong'an, China. *J. Environ. Sci.* 24: 1225–1233.
- Yue, D., Hu, M., Wu, Z., Guo, S., Wen, M., Nowak, A., Wehner, B., Wiedensohler, A., Takegawa, N., Kondo, Y., Wang, X., Li, Y., Zeng, L. and Zhang, Y. (2010). Variation of Particle Number Size Distributions and Chemical Compositions at the Urban and Downwind Regional Sites in the Pearl River Delta during Summertime Pollution Episodes. *Atmos. Chem. Phys.* 10: 9431–9439.
- Zhang, R., Jing, J., Tao, J., Hsu, S.C., Wang, G., Cao, J., Lee, C.S.L., Zhu, L., Chen, Z., Zhao, Y. and Shen, Z. (2013). Chemical Characterization and Source Apportionment of PM_{2.5} in Beijing: Seasonal Perspective. *Atmos. Chem. Phys.* 13: 9953–10007, doi: 10.5194/acp-13-7053-2013.
- Zhang, X., Wang, Y., Niu, T., Zhang, X., Gong, S., Zhang, Y. and Sun, J. (2012). Atmospheric Aerosol Compositions in China: Spatial/Temporal Variability, Chemical Signature, Regional Haze Distribution and Comparisons with Global Aerosols. *Atmos. Chem. Phys.* 12: 779–799.
- Zhao, H. and Ma, Y. (2011). Comparison between Instrumental and Visual Measurements of Visibility in Urban Area of Central Liaoning Province. *Meteor. Sci. Technol.* 39: 468–472 (in Chinese).
- Zhao, H., Ma, Y., Wang, X., Su, H. and Zhu, Y. (2012). Optical Characteristics of Aerosol in Autumn of 2010 over Anshan. *J. Meteor. Environ.* 28: 55–62 (in Chinese).
- Zhao, H., Che, H., Zhang, X., Ma, Y., Wang, Y., Wang, X., Liu, C., Hou, B. and Che, H. (2013a). Aerosol Optical Properties over Urban and Industrial Region of Northeast China by Using Ground-based Sun-photometer Measurement. *Atmos. Environ.* 75: 270–278.
- Zhao, H., Che, H., Zhang, X., Ma, Y., Wang, Y., Wang, H. and Wang, Y. (2013b). Characteristics of Visibility and Particulate Matter (PM) in an Urban Area of Northeast China. *Atmos. Pollut. Res.* 4: 427–434.
- Zhao, H., Ma, Y., Zhu, Y. and Gao, Q. (2013c). Characteristics of Visibility over Anshan from 2010 to 2011. *J. Meteor. Environ.* 29: 18–22 (in Chinese).

Received for review, August 5, 2014

Revised, November 7, 2014

Accepted, November 10, 2014