Aerosol and Air Quality Research, 17: 888–895, 2017

ISSN: 1680-8584 print / 2071-1409 online
doi: 10.4209/aaqr.2016.09.0407

Pollution Characteristics of $PM_{2.5}$ Aerosol during Haze Periods in Changchun, China

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ABSTRACT

To study the pollution characteristics of $PM_{2.5}$ during the haze period in October 2014, the hourly automatic monitoring data and 22-h atmospheric $PM_{2.5}$ samples were collected in Changchun city. According to the statistical results of the hourly average $PM_{2.5}$ concentration, heavy pollution occurred from October 13 to November 1 in 2014 in Changchun city. The daily concentration of $PM_{2.5}$ during this period was 53.18 µg m^{-3} to 450.69 µg m^{-3}. In addition, the $PM_{2.5}$ concentration was higher at night and lower during the daytime on haze days, which is affected by pollutant emission and meteorological conditions. The Pearson correlation coefficient between $PM_{2.5}$ and CO was high (r = 0.745), which implies that the concentration suddenly increased, probably because of the combustion of fossil fuels and other organic matter. The meteorological condition investigation shows that the wind speed, temperature and pressure are low and that the RH value is relatively high on haze days. In addition, stable weather during haze days makes the pollution heavier. The analysis of water-soluble inorganic ions (WSIIs) suggests that WSIIs constituted up to 21–56% of $PM_{2.5}$, and the total ions, anions and cations showed a significant correlation. During the haze days, the NO_3^-/SO_4^{2-} ratio was substantial (3.7–4.6), except at the CP and JZP stations, which suggests that the pollutants in the atmosphere are from a mixture of stationary emission and mobile emission. The NOR and SOR values were more than 0.1 at most sampling stations, so serious secondary pollution occurs in the atmosphere on haze days, and secondary ion formation is enhanced by the atmospheric conditions and emissions of gaseous SO_2 and NO_2.

Keywords: $PM_{2.5}$; Meteorological condition; Water soluble inorganic ions; Changchun.

INTRODUCTION

Haze weather has increased in recent years; fine particulate matter ($PM_{2.5}$) and inhalable particulate matter (PM_{10}) have received increasingly more attention. Particulate matter (PM) significantly affects visibility, human health, climate change and the ecosystem. In particular, fine particulate matter can penetrate people's respiratory systems and cause many fatal effects on the human body.

$PM_{2.5}$ observations and research have been performed in many countries worldwide. $PM_{2.5}$ is composed of various chemical components such as carbonaceous components (organic carbon and elemental carbon), water-soluble ions, metal elements and organic matter (Kulkarni and Venkataraman, 2000; Park *et al.*, 2002). Many scholars have analyzed the composition of $PM_{2.5}$ to perform source apportionments such as biomass burning, traffic emission, coal combustion, soil dust, and industrial emissions.

Changchun is an industrial city in the northeastern plain of China, and it has a temperate continental monsoon climate. The temperature increases and precipitation decreases from east to west. The climate is dry and windy in spring, hot and humid in summer, invigorating in autumn, and cold in winter. The annual average temperature in Changchun is 4.8°C, the highest temperature is 39.5°C, and the lowest temperature is –39.8°C. The annual average precipitation ranges from 522 mm to 615 mm.

Water-soluble inorganic ions (WSIIs) account for at least one-third of the fine particulate matter. The WSII concentrations distinctly vary in different seasons. Typically, SO_4^{2-}, NO_3^- and NH_4^+ are the major WSII species in $PM_{2.5}$ and the most abundant component in secondary ion species (Yin *et al.*, 2012). The particulate compositions during hazy weather and normal weather were analyzed in 2009 (Tan *et al.*, 2009). The SO_4^{2-}, NH_4^+ and NO_3^- concentrations significantly increased on haze days.

Some studies have investigated the sources of $PM_{2.5}$ in many cities of China such as the Beijing-Tianjin-Hebei

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region, the Yangtze River delta and the Pearl River Delta (Wang *et al.*, 2008; Gu *et al.*, 2011; Wang *et al.*, 2012; Cheng *et al.*, 2013; Dai *et al.*, 2013; Huang *et al.*, 2013) However, fewer studies on $PM_{2.5}$ in northeast China have been performed. To better understand the temporal and spatial distribution and the component characteristics of $PM_{2.5}$, the hourly automatic monitoring data in 2014 and 22-h atmospheric $PM_{2.5}$ samples were collected in Changchun during the haze period in October 2014. The $PM_{2.5}$ concentration and its WSII components during haze days were analyzed in this paper.

MATERIALS AND METHODS

Sites and Sampling

Changchun is an industrial city in the northeastern plain of China. There are 10 automatic atmospheric monitoring stations in Changchun: Jingyue Park (JYP), Jun Zilan Park (JZP), Bus Factory Hospital (BFH), Labor Park (LP), Daishan Park (DP), High-tech Zone Management Committee (HZMC), Economic development zone Environment Sanitary Administration (EESA), Children's Park (CP), Institute of Posts and Telecommunications (IPT), and Shuai Wan Zi (SWZ).

The $PM_{2.5}$ samples in this study were collected at nine Changchun environmental monitoring stations, excluding SWZ. A medium-volume TSP/PM_{10}/$PM_{2.5}$ sampler with Teflon filters (KC-120 Particulate Sampler, China) was used to collect particulate samples for the WSII analysis on October 21, 2014. There were 9 samples with a 22-h sampling duration. Before and after sampling, the filters were conditioned at a temperature of (25 ± 1)°C and a relative humidity of (35% ± 2%) for 24 h and weighed using an electronic balance with a detection limit of 0.01 mg. The $PM_{2.5}$ mass concentrations for each sampling were calculated from the weight and corresponding air volume.

Water-soluble Inorganic Ions Analysis

The $PM_{2.5}$ sample filters were submerged in a 50-mL colorimetric tube with 15 mL of pure water; the WSIIs from the samples were extracted by ultrasound for 10 min (KQ-700E Ultrasonic, China). The extract solutions were filtered with 0.45-µm filters and analyzed using an ion chromatograph. According to the peak retention time and peak height (or peak area) of ions in the mixed standard solution, the WSIIs in the samples were identified and quantified. Cations (Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}) in the extract solution were analyzed using a DX-120 Ion Chromatograph (Dionex Corporation, America); the anions (F^-, Cl^-, NO_3^-, and SO_4^{2-}) were analyzed using Metrohm-MIC-7 (Metrohm AG, Switzerland).

RESULT AND DISCUSSION

Mass Concentration of $PM_{2.5}$

The contour map produced using Surfer software shows the hourly variation in $PM_{2.5}$ concentrations during each week in 2014. The average $PM_{2.5}$ concentrations of 10 environmental monitoring stations are presented in Fig. 1. The horizontal coordinate is time, and the vertical coordinate represents 52 weeks. In 2014, each data point indicates the weekly average $PM_{2.5}$ concentration and is denoted with different colors.

The result in Fig. 1 shows that heavy pollution occurred

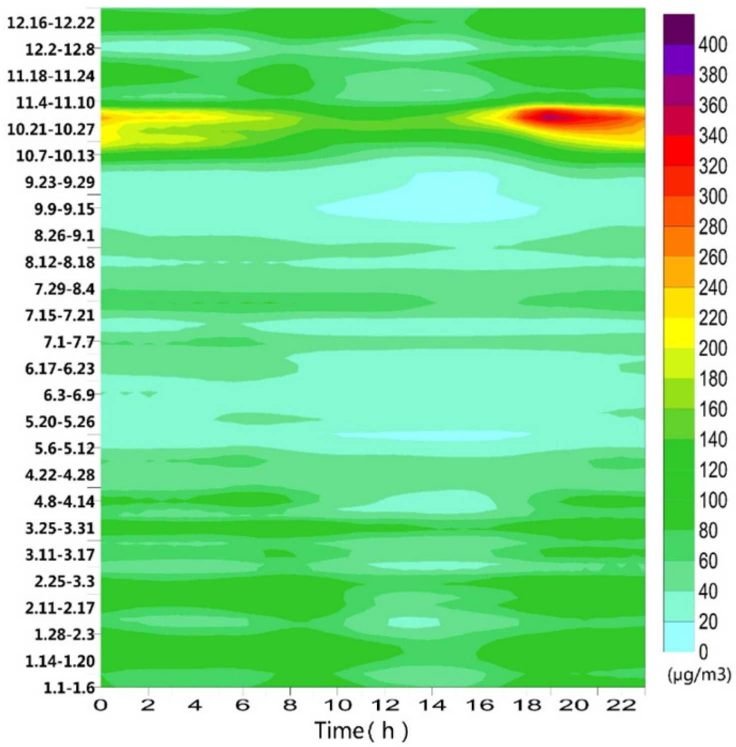

Fig. 1. Hourly variation of $PM_{2.5}$ concentrations in each week in 2014.

from October 13 to November 1 in 2014 in Changchun city. The $PM_{2.5}$ concentration was high from 16:00 to 8:00 and had a peak value at 19:00. The air quality in May, June and September was relatively low. The haze period and normal-weather period are compared in Fig. 2. The haze period was from October 13 to November 1, and the normal-weather period was from November 2 to November 10. The $PM_{2.5}$ concentrations on October 14, 18, 22, 25, 28, 29, 30, and 31 and November 1 were obviously higher than those on the other days and show an increasing trend. Hazy weather initially occurred every 3 days; then, a high-$PM_{2.5}$ pollution occurred for up to 5 days because of the concentration of pollutant; the $PM_{2.5}$ concentration reached a peak on October 30. The daily average concentration of $PM_{2.5}$ in this period ranged from 53.18 µg m^{-3} to 450.69 µg m^{-3}. According to the statistical results, 11 daily average values exceeded 150 µg m^{-3}, and 6 exceeded 250 µg m^{-3}. The $PM_{2.5}$-to-PM_{10} ratio shows that $PM_{2.5}$ is the main component of PM_{10}, and the changing trend of the $PM_{2.5}$ concentration is similar to that of the $PM_{2.5}/PM_{10}$ ratios. The $PM_{2.5}$ concentration and $PM_{2.5}/PM_{10}$ decreased from November 2 to November 10, whereas $PM_{2.5-10}$ had no obvious variation, which indicates that the increase in the particulate matter concentration in air is directly related to $PM_{2.5}$ pollution, and a high $PM_{2.5}$ concentration can aggravate the haze.

Further study is currently being performed to observe the hourly variation in $PM_{2.5}$ concentrations from October 13 to November 1. In Fig. 3, the horizontal coordinate is time, which ranges from 0:00 to 23:00, and each column represents the 10–90% quantile of concentration values in 20 days. The dots represent the 50% quantile values, and the dots with line represent the average values. Obviously, the $PM_{2.5}$ concentration remained high from 0:00 to 5:00 and decreased from 5:00 to 13:00. It had the lowest value at 13:00, increased from 13:00 to 20:00 and remained high from 20:00 to 24:00. The $PM_{2.5}$ concentration exhibited a U-shaped trend and was higher at night and lower in the day time during haze days, which testifies that straw burning and coal combustion at night make an enormous contribution to pollution in Changchun city. This result also proves that the meteorological condition affects the $PM_{2.5}$ concentration during haze days. At night, low temperatures and an unobvious pressure gradient are not conducive to the spread of pollutants to high altitudes. In addition, the inversion layer, which often occurs in early winter in northeastern China, promotes the accumulation of pollutants. During the day time, the pollutants diffuse into the upper atmosphere because of the high temperature.

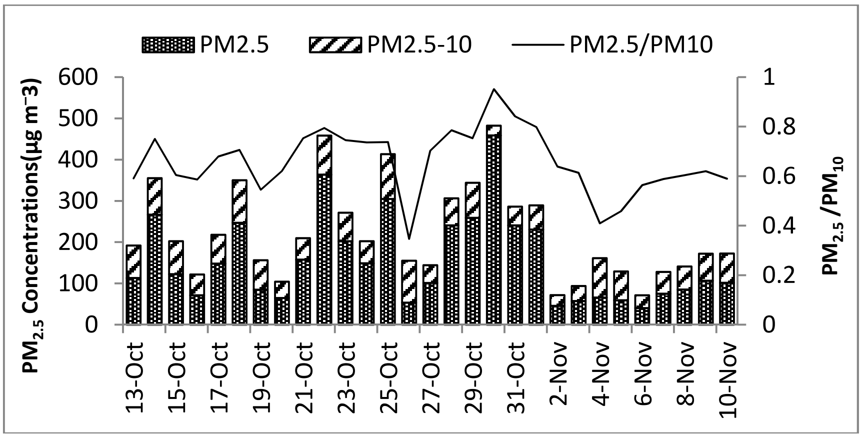

Fig. 2. Daily average concentration from October 13 to November 10.

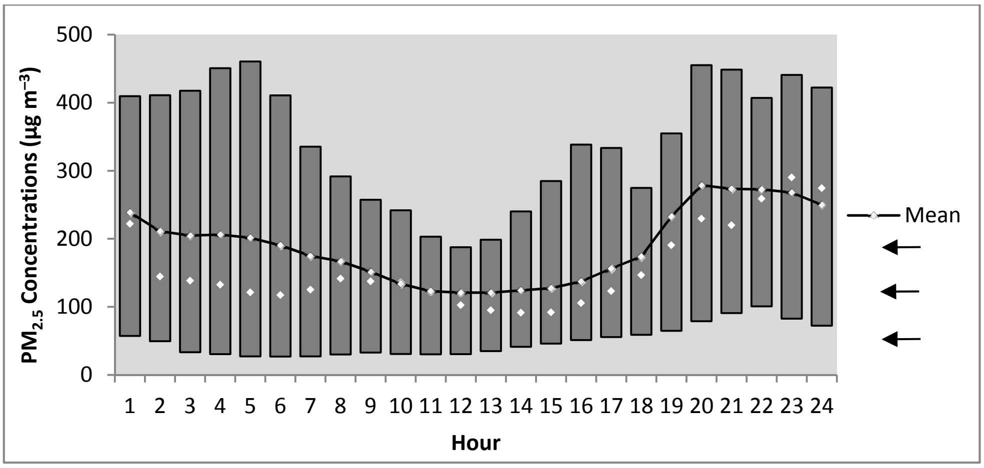

Fig. 3. Boxplot of the hourly $PM_{2.5}$ concentration.

Correlations between $PM_{2.5}$ and Gaseous Pollutants (SO_2, NO_X, CO and O_3)

The Pearson correlation coefficient (SPSS, bivariate correlation) was used to investigate the relationship among SO_2, NO_x, CO, O_3 and $PM_{2.5}$. This analysis was based on the hourly data of ten automatic monitoring sites in Changchun city from October 28 to November 1. We also performed two-sided tests, as shown in Table 1.

The analysis results show a strong correlation between $PM_{2.5}$ and CO (r = 0.745), which implies that the CO emission process is accompanied by the emission of fine particles. The sudden increase in concentration is probably because of the combustion of fossil fuels and other organic matter. However, the correlations among SO_2, NO_x, O_3 and $PM_{2.5}$ are slightly lower ($PM_{2.5}$ and SO_2: r = –0.190; $PM_{2.5}$ and NO_x: r = 0.214; $PM_{2.5}$ and O_3: r = –0.280). Previous studies have shown that the oxidation of SO_2 and NO_x forms sulfuric acid, nitric acid, and sulfate and nitrate aerosols. These species chemically react with one another in the presence of sunlight and produce toxic secondary pollutants such as O_3 and aerosols, which are major concerns for urban air quality (Xie *et al.*, 2015).

Table 1. Pearson correlation coefficients between $PM_{2.5}$ and gaseous pollutants (N = 120).

	SO_2	NO_x	CO	O_3
$PM_{2.5}$	-0.190^{*}	0.214^{*}	0.745^{**}	-0.280^{**}

* and **: Correlation is significant at the 0.01 and 0.05 levels (2-tailed).

Analysis of Meteorological Conditions

The mass concentration of $PM_{2.5}$ is closely related to the metrological conditions in the atmosphere during the heavy haze period. Investigating the meteorological factors can provide insight into the variations in the $PM_{2.5}$ mass concentrations. The major meteorological conditions in the haze period are shown in Fig. 4.

In Fig. 4, the variations in temperature and wind speed are similar. On October 13, 17, 21, 25, 28, and 31, the temperature and wind speed were notably low; on October 15, 19, 24 and 26, they were high. According to the $PM_{2.5}$ variation in Fig. 2, haze pollution occurred on October 14, 18, 22 and 25. Thus, low wind speed and temperature are not conducive to pollutant dispersion. Moreover, the high RH and low pressure on October 18, 25 and 31 indicate that high humidity and low air pressure can accelerate the development of hazy weather conditions.

To study the effect of atmospheric activity on the formation and development of hazy weather in Changchun, the 72-h backward and forward trajectories that began from Changchun (43.84N, 125.29E) at the arrival level of 500 m and mixed layer depth (Oct 29–31) were generated with the NOAA HYSPLIT model (http://ready.arl.noaa.gov/HYSPLIT_traj.php) in Figs. 5 and 6. The trajectories were calculated at the starting time of 00:00 on October 30.

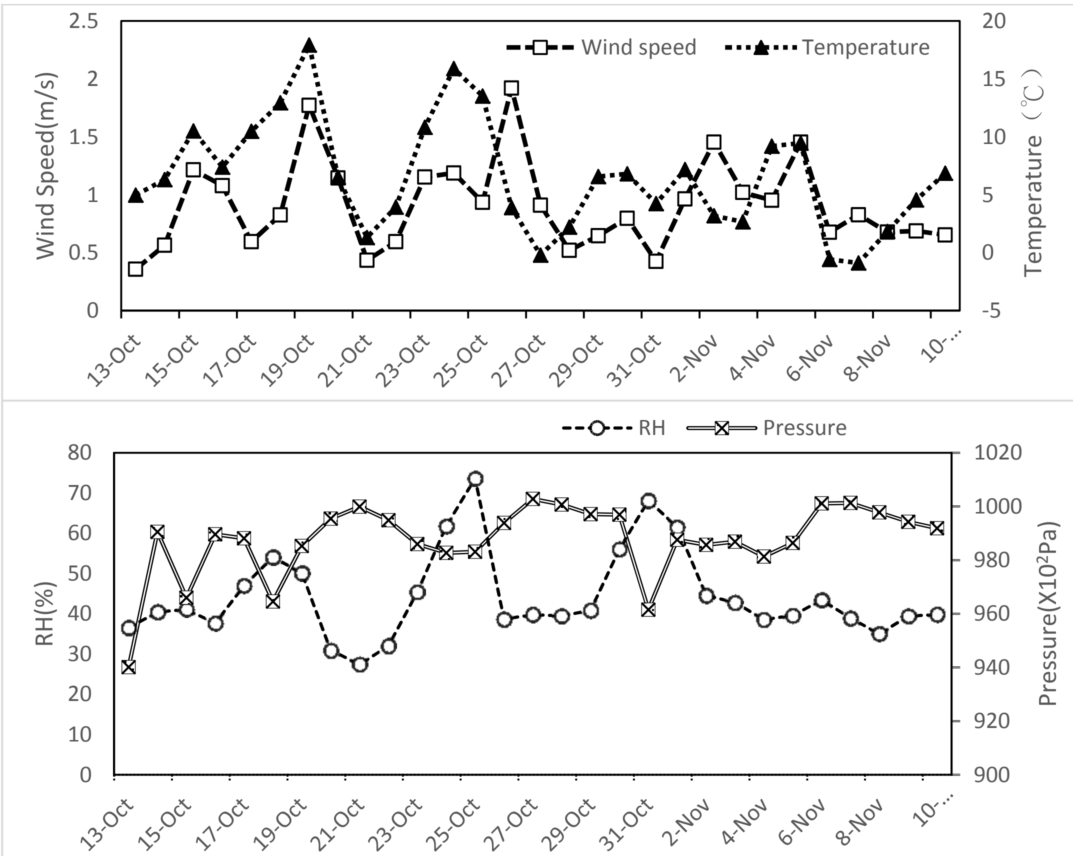

Fig. 4. Variation in wind speed, temperature, RH, pressure from October 13 to November 10 in 2014 in Changchun.

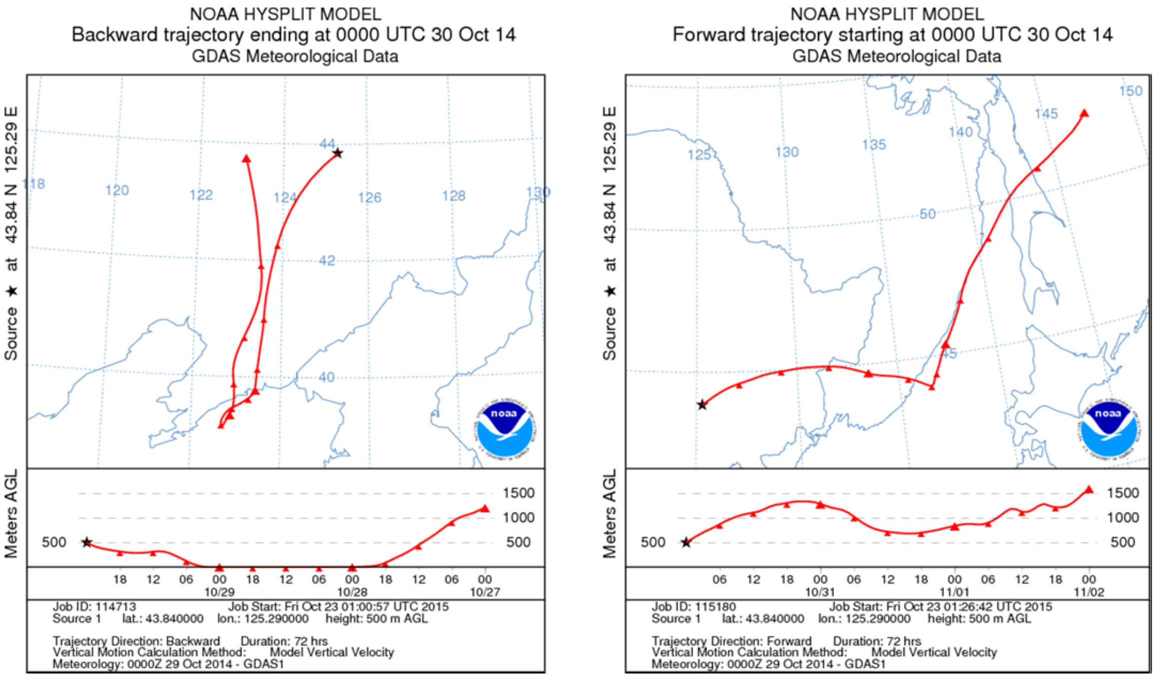

Fig. 5. 72-h backward and forward trajectories at the starting time of 00:00 on October 30 from Changchun.

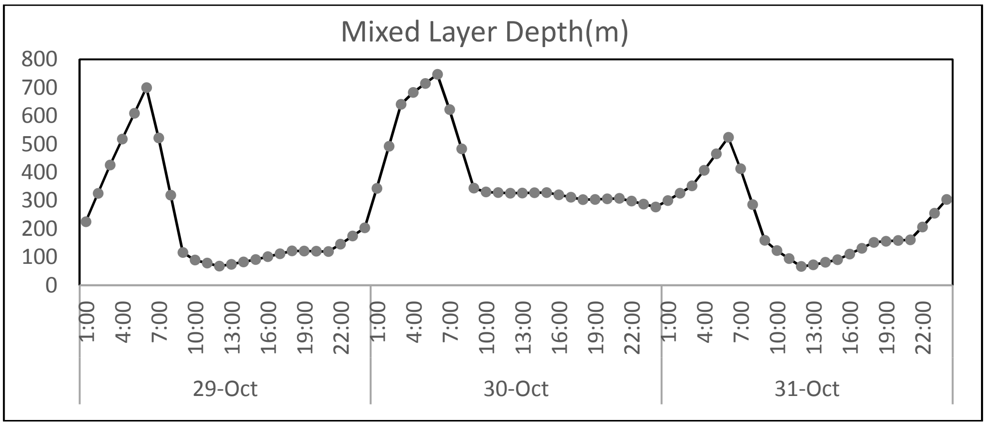

Fig. 6. Mixed Layer Depth on October 29–31, 2014, in Changchun.

Based on the HYSPLIT model, the air flow ran at a notably low height from 18 pm October 28 to a relatively high height on October 31. The mixed layer depth in Changchun remained notably low (even less than 80 m) and caused the high $PM_{2.5}$ concentration on the 29, 30 and 31. Thus, stable weather on these days made the $PM_{2.5}$ pollution heavier.

Water-soluble Inorganic Ion (WSII) Compositions in $PM_{2.5}$

WSIIs account for 21–56% of $PM_{2.5}$. The order is NO_3^- (39%) > Cl^- (28%) > K^+ (11%) > SO_4^{2-} (10%) > NH_4^+ (8%) > Ca^{2+} (3%) > Na^+ (1%) > F^- > Mg^{2+}.

The correlation analysis results of WSIIs are shown in Table 2. Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+} are highly homologous with a significant correlation; F^-, SO_4^{2-} and Cl^- also have a high homology. The total ions, anions and cations show significant correlations and homology. The good correlation between K^+ and Cl^- indicates that straw burning contributes to pollution in Changchun during haze days.

Ratio of NO_3^-/SO_4^{2-}

The mass concentrations of NH_4^+, SO_4^{2-} and NO_3^-, which were collected at nine sampling stations on haze days, and the NO_3^-/SO_4^{2-} ratios are shown in Fig. 7.

Previous studies show that the major WSIIs in $PM_{2.5}$ are NH_4^+, SO_4^{2-} and NO_3^-, and these are also the most abundant components in the secondary ion species (Yue *et al.*, 2015). The estimated NO_x-to-SO_x ratios from the emission of gasoline and diesel fuel burning are 13:1 and 8:1, respectively. The sulfur content in coal is approximately 1%, and the estimated emission NO_x-to-SO_x ratio is 1:2 from coal burning (Wang *et al.*, 2005). Based on the characteristics of these two ratios, the values of NO_3^-/SO_4^{2-} and the stationary/mobile emission ratio were calculated and are shown in Table 3.

As shown in Fig. 4, the NO_3^-/SO_4^{2-} ratio in Changchun

Table 2. Correlation analysis of the WSIIs.

	F^-	Cl^-	NO_3^-	SO_4^{2-}	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	total ion	total anion	Total cation
F^-	1	0.943^{**}	0.829^{**}	0.424	0.970^{**}	0.960^{**}	0.971^{**}	0.921^{**}	0.880^{**}	0.934^{**}	0.890^{**}	0.985^{**}
Cl^-	0.943^{**}	1	0.951^{**}	0.412	0.860^{**}	0.994^{**}	0.942^{**}	0.790^{*}	0.722^{*}	0.991^{**}	0.976^{**}	0.955^{**}
NO_3^-	0.829^{**}	0.951^{**}	1	0.485	0.714^{*}	0.918^{**}	0.801^{**}	0.663	0.591	0.966^{**}	0.989^{**}	0.831^{**}
SO_4^{2-}	0.424	0.412	0.485	1	0.488	0.381	0.394	0.608	0.619	0.526	0.544	0.438
Na^+	0.970^{**}	0.860^{**}	0.714^{*}	0.488	1	0.892^{**}	0.949^{**}	0.976^{**}	0.953^{**}	0.861^{**}	0.801^{**}	0.963^{**}
NH_4^+	0.960^{**}	0.994^{**}	0.918^{**}	0.381	0.892^{**}	1	0.967^{**}	0.818^{**}	0.751^{*}	0.979^{**}	0.953^{**}	0.975^{**}
K^+	0.971^{**}	0.942^{**}	0.801^{**}	0.394	0.949^{**}	0.967^{**}	1	0.884^{**}	0.830^{**}	0.923^{**}	0.871^{**}	0.995^{**}
Mg^{2+}	0.921^{**}	0.790^{*}	0.663	0.608	0.976^{**}	0.818^{**}	0.884^{**}	1	0.991^{**}	0.814^{**}	0.757^{*}	0.909^{**}
Ca^{2+}	0.880^{**}	0.722^{*}	0.591	0.619	0.953^{**}	0.751^{*}	0.830^{**}	0.991^{**}	1	0.751^{*}	0.691^{*}	0.859^{**}
Total ion	0.934^{**}	0.991^{**}	0.966^{**}	0.526	0.861^{**}	0.979^{**}	0.923^{**}	0.814^{**}	0.751^{*}	1	0.993^{**}	0.943^{**}
Total anion	0.890^{**}	0.976^{**}	0.989^{**}	0.544	0.801^{**}	0.953^{**}	0.871^{**}	0.757^{*}	0.691^{*}	0.993^{**}	1	0.897^{**}
Total cation	0.985^{**}	0.955^{**}	0.831^{**}	0.438	0.963^{**}	0.975^{**}	0.995^{**}	0.909^{**}	0.859^{**}	0.943^{**}	0.897^{**}	1

* and **: Correlation is significant at the 0.01 and 0.05 levels (2-tailed).

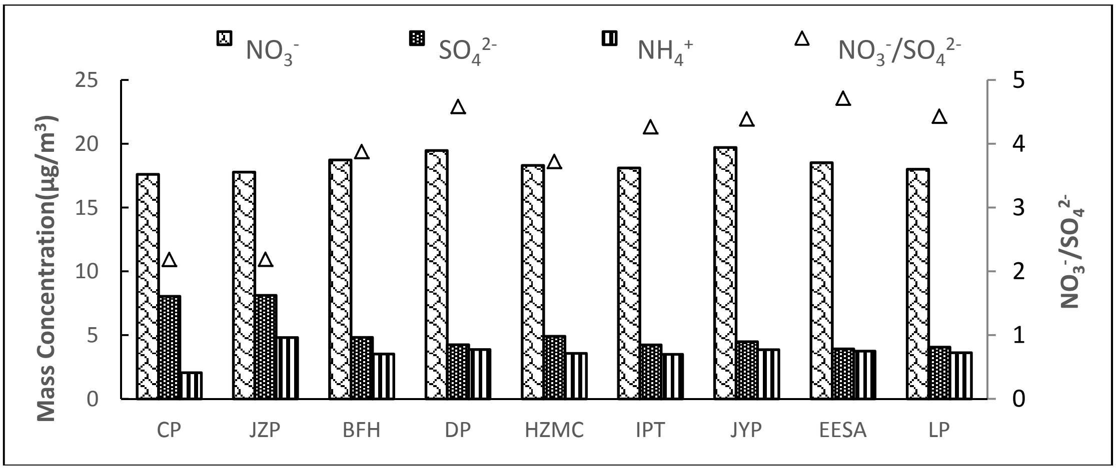

Fig. 7. Mass concentrations of NH_4^+, SO_4^{2-}, and NO_3^- and the NO_3^-/SO_4^{2-} ratio.

Table 3. Diagnostic values for NO_3^-/SO_4^{2-} with different ratios of stationary and mobile emission.

stationary/mobile	10:0	8:2	6:4	4:6	2:8	0:10
NO_3^-/SO_4^{2-}	0.5	2–3	3.5–5.5	5–8	6.5–10.5	8–13

was 3.7–4.6 except at stations CP and JZP, which implies that pollutants are from the mixture of stationary emission and mobile emission, and stationary emission is the main pollution source; the ratio of stationary to mobile is approximately 6:4. This result occurred partly because of the beginning of coal combustion heating activities during October 2014 in Changchun.

Values of NOR and SOR

The nitrogen oxidation ratio (NOR) and sulfur oxidation ratio (SOR) values can reflect the degree of atmospheric conversion of NO_2 and SO_2 to NO_3^- and SO_4^{2-}, respectively. Higher SOR and NOR values indicate that more secondary aerosol particulates are converted from SO_2 and NO_2, which are generated in gas or liquid phase reactions. In the primary pollutant, SOR is typically below 0.1, so it can be considered a cutoff value for the SO_2 secondary conversion. In the atmosphere, SO_2 is photochemically oxidized when SOR is above 0.1 (Ohta and Okita, 1990; Kaneyasu *et al.*, 1995).

The SOR and NOR are defined as follows:

$$\mathrm{SOR} = \frac{\left[\mathrm{SO_4^{2-}}\right]}{\left\{\left[\mathrm{SO_4^{2-}}\right]+\left[\mathrm{SO_2}\right]\right\}} \tag{1}$$

$$\mathrm{NOR} = \frac{\left[\mathrm{NO_3^{-}}\right]}{\left\{\left[\mathrm{NO_3^{-}}\right]+\left[\mathrm{NO_2}\right]\right\}} \tag{2}$$

The NOR and SOR in each sampling station are shown in Fig. 8. All SOR and NOR values were more than 0.1. The JYP sampling station had the highest SOR and NOR of 0.30 and 0.36, respectively. Thus, there is serious secondary pollution in the atmosphere during hazy weather. Moreover, the secondary ion formation progress is enhanced by the atmospheric condition and gaseous SO_2 and NO_2 emissions.

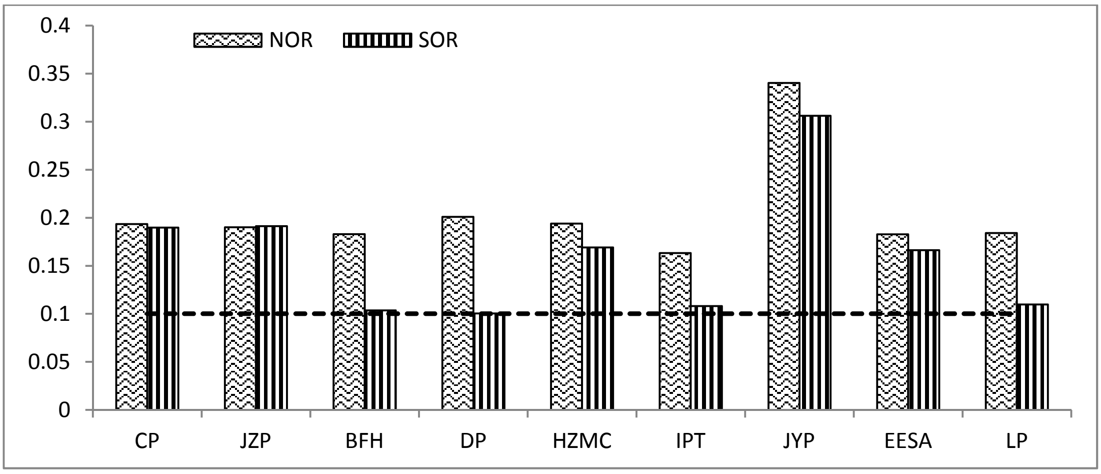

Fig. 8. Values of NOR and SOR at nine sampling stations.

CONCLUSIONS

In this paper, the hourly automatic monitoring data and the 22-h atmospheric receptor $PM_{2.5}$ samples were collected in Changchun city to investigate the pollutant characteristics of $PM_{2.5}$ during the haze period in October 2014.

According to the statistical results of the hourly average PM concentration, heavy pollution occurred from October 13 to November 1 in 2014 in Changchun city. The daily average concentration of $PM_{2.5}$ during this period was 53.18 µg m^{-3} to 450.69 µg m^{-3}, and the $PM_{2.5}$ concentration was higher on October 14, 18, 22, 25, 28, 29, 30, and 31 and November 1 than on other days. The $PM_{2.5}$-to-PM_{10} ratio shows that the change in $PM_{2.5}$ concentration is similar to the change in $PM_{2.5}/PM_{10}$, which indicates that the increase in the mass concentration of particulate matter in air is directly related to the $PM_{2.5}$ pollution. The hourly variation in $PM_{2.5}$ concentration from October 13 to November 1 has a U-shaped trend and is higher at night and lower in the day time during haze days, which indicates that straw burning and coal combustion at night make an enormous contribution to pollution in Changchun city. In addition, the meteorological conditions are an additional factor that affects the $PM_{2.5}$ concentration during haze days. At night, the low temperature and unobvious pressure gradient are not conducive to the spread of pollutants to high altitudes. In addition, the inversion layer promotes the accumulation of pollutants.

The Pearson correlation coefficient is used to investigate the relationship among SO_2, NO_x, CO, O_3 and $PM_{2.5}$. The analysis results show a strong correlation between $PM_{2.5}$ and CO (r = 0.745), which implies that the sudden increase in concentration is probably because of the combustion of fossil fuels and other organic matter. However, the correlations among SO_2, NO_x, O_3 and $PM_{2.5}$ are slightly lower ($PM_{2.5}$ and SO_2: r = –0.190; $PM_{2.5}$ and NO_x: r = 0.214; $PM_{2.5}$ and O_3: r = –0.280).

Meteorological conditions were investigated from October 13 to November 10 in 2014 in Changchun. The results show that the low wind speed, temperature, and pressure are not conducive to pollutant dispersion. Meanwhile, a high-humidity environment promotes haze. The low mixed layer depth in Changchun caused the high $PM_{2.5}$ concentration on October 29, 30 and 31. Thus, stable weather during these days made the pollution heavier.

According to the WSII analysis of $PM_{2.5}$, WSIIs constituted up to 21–56% of the $PM_{2.5}$. The correlation analysis results of the nine WSIIs show that the total ions, anions and cations have significant correlations. Meanwhile, the NO_3^-/SO_4^{2-} ratio was substantial (3.7–4.6), except at stations CP and JZP. Pollutants are from the mixture of stationary emission and mobile emission, and the stationary emission is the main pollution source. NOR and SOR at each sampling station were more than 0.1. The JYP sampling station had high SOR and NOR values of 0.30 and 0.36, respectively, which indicates that there was serious secondary pollution in the atmosphere during the haze.

ACKONWLEDGMENTS

This study is supported by the Jilin Provincial Science & Technology Department (No. 20130204051SF) and the Jilin Provincial Environment Protection Department (No. 2013EP01-03). We thank the Changchun environmental monitoring center for assistance in data sourcing and sampling.

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Received for review, October 30, 2016
Revised, February 16, 2017
Accepted, February 20, 2017