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Chemical Characteristics of $PM_{2.5}$ during 2015 Spring Festival in Beijing, China

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ABSTRACT

Air pollution especially of $PM_{2.5}$ pollution is a serious problem in Beijing. In order to quantify the effect of a festival in which pollution was expected to be reduced, we collected and analyzed $PM_{2.5}$ samples in urban Beijing during the 2015 Spring Festival (from February 9th to March 6th 2015). We divided the sampling period into three phases: non-haze, haze and firework days. The average concentration of $PM_{2.5}$ was highest on firework days at 248.9 μg m^{-3}, followed by haze days (199.9 μg m^{-3}) and non-haze days (90.8 μg m^{-3}). The air quality of non-haze days during the holiday was better than that during non-holiday periods. Secondary inorganic ions (SO_4^{2-}, NO_3^- and NH_4^+) were enriched on the haze days, while those on firework days contained large amounts of Cl^- and K^+, but small amounts of NO_3^- and NH_4^+. Ratios of NO_3^-/SO_4^{2-}, SO_4^{2-}/K^+ and Cl^-/K^+ effectively distinguished the characteristics of $PM_{2.5}$ between firework events and haze days. Ion balance calculations indicated that the acidity of $PM_{2.5}$ from firework days was higher than that from haze and non-haze days. A method using enrichment factors (EF) found that crustal elements (EF < 10 on all three types of days) included Ca, Al, Fe, Na, Co, Ni, P, Ti, and V; firework elements (EF > 10 on firework days, significantly higher than haze days) were made up of Ba, Cr, Cu, Mg, Pb, S, Si, and Zn; common anthropogenic pollution elements (EF > 10 in all three types of days) were As, Cd, Cu, Pb, S, Sb, Zn. Differences in chemical characteristics indicated that holidays such as the Spring Festival can affect air pollution patterns in two ways: a decrease in the population and vehicles but an increase in activities such as firework displays.

Keywords: $PM_{2.5}$; Chemical characteristics; Fireworks; Haze; Spring Festival.

INTRODUCTION

Air pollution caused by fine particles with a diameter of 2.5 μm or less ($PM_{2.5}$) has aroused worldwide attention during the past decades because of its significant negative effects on visibility, human health and global climate (Okada *et al.*, 2001; Kang *et al.*, 2004; Sun *et al.*, 2006; Sun *et al.*, 2014). In China, the average annual number of haze days has fluctuated but generally increased (Gao, 2008). In Beijing, hazy episodes happened more frequently than in others cities (Che *et al.*, 2009; Tang *et al.*, 2013; Li *et al.*, 2016). The annual average number of haze days in Beijing increased from about 50 days yr^{-1} in the 1980s to 72 days yr^{-1} in 2008 (Hu and Zhou, 2009; Zhao *et al.*, 2012; Chen *et al.*, 2014). Pollution in Beijing is generally attributed to industry, road dust, vehicle emissions, coal and biomass burning (Okuda *et al.*, 2004; Wang *et al.*, 2006; Duan *et al.*, 2012; Zhang *et al.*, 2013; Yu *et al.*, 2015). Thus, strategies to improve air quality should take all these factors into account.

Nevertheless, there are still some special anthropogenic factors that lead to dramatic pollution episodes, one of which is the use of fireworks. Burning fireworks releases huge amounts of gaseous pollutants, such as sulfur dioxide (SO_2) (Huang *et al.*, 2012), nitrogen oxides (NO_x), carbon dioxide (CO_2), and carbon monoxide (CO) (Wang *et al.*, 2007; Vecchi *et al.*, 2008; Huang *et al.*, 2012), as well as suspended particles containing water-soluble ions, organic/elemental carbon and trace metals (Drewnick *et al.*, 2006; Sarkar *et al.*, 2010; Jiang *et al.*, 2015; Kong *et al.*, 2015). It has been noted before that firework displays could lead to a sharp increase in $PM_{2.5}$ concentrations (Wang *et al.*, 2007; Feng *et al.*, 2012; Tsai *et al.*, 2012; Han *et al.*, 2014) and generates haze pollution within a short time (Sarkar *et al.*, 2010; Tian *et al.*, 2014b). In China, high-intensity firework displays mainly occur during festivals or special events, especially the Spring Festival. As a mega city, a "Banned Fireworks" policy has been in place in Beijing since 1993, but was replaced by a "Limited Fireworks" policy in 2006 (Jing *et al.*, 2014). In preparation for the Spring Festival, almost half of the population in Beijing (about 9 million in 2013,

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http://news.xinhuanet.com/local/2013-02/18/c_124355887.htm) leave for their hometown. Consequently, factories closed down and the population density and vehicle numbers sharply decrease, reducing the emission intensity of the main pollutants. Meanwhile, fireworks burning becomes an important pollution source during the Spring Festival. There have been some studies on the characteristics of fireworks pollution, including its impacts on fine and coarse particles (Chang *et al.*, 2011; Huang *et al.*, 2012; Li *et al.*, 2013; Jing *et al.*, 2014; Zhao *et al.*, 2014; Jiang *et al.*, 2015; Lin *et al.*, 2016), source apportionment (Feng *et al.*, 2012; Tian *et al.*, 2014b) and human health risk assessment (Zhao *et al.*, 2012; Shi *et al.*, 2014; Yang *et al.*, 2014).

We analyzed the chemical characteristics of $PM_{2.5}$ collected during the 2015 Spring Festival period (February 9th–March 6th 2015). The sampling period was separated into pollution events and non-haze days. The pollution events periods involved firework displays and haze episodes. This allowed us to further study the short-term holiday effects. We hypothesized that the results of this study would provide support for controlling short-term and particular forms of air pollution (such as firework displays) in urban areas.

METHODS

Sampling

$PM_{2.5}$ samples were collected on the rooftop of a residential bungalow (about 4 m above ground level), in Guangning village (39.94°N, 116.15°E), Shijingshan District, Beijing. It is surrounded by a viaduct (city expressway, linear distance of 250 m), a hill and two large thermal power plants (Jingxi and Datang). During the sampling period, the two thermal power plants were shut down. $PM_{2.5}$ particles were collected on Whatman filters (90 mm quartz membrane, diameter 2.2 microns) by a medium volume air sampler (model: TH-150C, flow rate: 100 L min^{-1}). The sampling time was approximately 12 h, normally from 8:00 a.m. to 8:00 p.m., once every three days on non-haze days. If there was an unexpected pollution event, additional samples were collected. Based on the regularity of setting off firework, the samples were collected over three periods during February 18th and 19th, including before the Spring Festival (8:10 a.m.–4:10 p.m.), Spring Festival's eve (4:20 p.m.–0:20 a.m.), and after the Spring Festival (0:30 a.m.–8:30 a.m.). On March 5th (the Lantern Festival), the sample was collected from 6:00 p.m. (the beginning of setting off the firework) to 6:00 a.m. (the next morning). Detailed sampling information is presented in Table 1. The samples were placed in sulfuric acid paper after sampling, wrapped with aluminum foil in the dark, and then stored in a refrigerator (–4°C). The filters were weighed before and after sampling on an analytical balance to an accuracy of 0.0001g (TB-215D, Denver) at a constant temperature (20°C) and relative humidity (40%), then divided by the sampling volumes to obtain $PM_{2.5}$ mass concentrations. Another three blank filters were dealt with simultaneously using the same method as above. All the procedures were strictly quality-controlled to avoid any possible contamination of the samples.

Water-Soluble Ions

One-fourth of each sample was extracted with 10 mL deionized water (resistivity18.2 MΩ cm^{-1}). After a 30 min in a microwave to dissolve the particles, the solution was drawn into a 1 mL syringe, filtered and injected into a 10 mL centrifuge tube with 0.22 mm filter cap. Water-soluble ions (Na^+, $NH4^+$, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, and SO_4^{2-}) were analyzed by ion chromatography (IC, Dionex). Three blank samples were analyzed at the same time to ensure the

Table 1. Sampling schedule, $PM_{2.5}$ mass concentrations and meteorological conditions during the Spring Festival in Shijingshan, Beijing (2015).

Sample type	Date	Start time	End time	Duration (hr)	$PM_{2.5}$ (μg m^{-3})	Temp.[a] (°C)	RH[b] (%)	WS[c] (km h^{-1})	Wind Dir.[d] (°)	Visibility (km)
Non-haze	Feb 12	8:10 am	8:10 pm	12	71.9	9.8	26	6.8	120	16
	Feb 17	8:10 am	8:10 pm	12	154.0	13.2	25	11.8	275	25
	Feb 18-a	8:10 am	4:10 pm	8	67.1	13	22	11.5	291	30
	Feb 27	8:30 am	8:30 pm	12	70.3	6.4	34	7.7	333	18
	Avg.				90.8	10.6	27	9.5	-	22
Haze	Feb 9	8:10 am	8:10 pm	12	149.0	7.5	19	9	84	6
	Feb 14	9:20 am	1:20 pm	4	226.9	11.9	37	5.1	138	3
	Feb 15	9:00 am	9:00 pm	12	194.8	10.6	38	7.2	147	4
	Feb 21	8:40 am	8:40 pm	12	223.1	4.5	67	17.2	296	3
	Feb 24	8:20 am	8:20 pm	12	205.9	9	54	7.4	311	6
	Avg.				199.9	8.7	43	9.2	-	4.4
Fireworks	Feb 18-b	4:20 pm	0:20 am	8	186.1	8.5	26	11	291	14
	Feb 19-a	0:30 am	8:30 am	8	293.7	3.7	55	6.3	296	5
	Feb 19-b	8:30 am	8:30 pm	12	234.5	6.9	56	11.3	296	5
	Mar 5	6:00 pm	6:00 am	12	281.3	6.2	42	6.2	-	7
	Avg.				248.9	6.3	45	8.7	-	7.8

* Feb 18-a: the day before Spring Festival's Eve; Feb 18-b: Spring Festival's Eve (before midnight); Feb 19-a: Spring Festival's Eve (after midnight); Feb 19-b: the day of Spring Festival; Mar 5: Lantern Festival.

[a] Temperature; [b] Relative humidity; [c] Wind speed; [d] Wind direction (North: 0° and 360°, East: 90°, South: 180°, West: 270°).

accuracy of analysis. Anions were analyzed on an ICS-2100 ion chromatograph, which consisted of a separation column (Dionex Ionpac AS11) and guard column (Dionex Ionpac AG11). Cations were analyzed on a DX-600 ion chromatograph, which consisted of a separation column (Dionex CS12A), a guard column (Dionex AG12A), a self-regenerating suppressed conductivity detector (Dionex Ionpac ED50), a gradient pump (Dionex Ionpac GP50), and a chromatographic box (LC20). The details were given elsewhere Yuan *et al.* (2003).

Elements Analysis

Half of each sample was placed in a Teflon vessel and digested in 3 mL HNO_3, 1 mL $HClO_4$, 1 mL HF, at 170°C for 4 h at high-pressure. After cooling, the solutions were dried and then 1 mL concentrated HNO_3 added, and diluted to 10 mL with deionized water (resistivity 18.2 MΩ cm^{-1}) (Wang *et al.*, 2007). 24 trace elements (Al, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, Li, Mg, Mn, Na, Ni, P, Pb, S, Sb, Sc, Se, Sr, Ti, V, Zn) were analyzed using inductively coupled plasma atomic emission spectrometry (ICP-AES, JOBIN-YVON). The detailed analytical procedures are given in Zhuang *et al.* (2001).

Meteorological Data and Air Quality Data Collection

During the sampling periods, meteorological factors (hourly average) such as relative humidity (RH), visibility, temperature, wind speed and direction were obtained from Weather Underground (http://www.wunderground.com), which archive meteorological data was generally used by previous researches about air quality (Wang *et al.*, 2009; Guo *et al.*, 2010; Verma *et al.*, 2013)

Air quality data (average of 24 hours, monitored at the end of the $PM_{2.5}$ sampling) were collected from the National Urban Air Quality Real-time Publishing Platform (http://113.108.142.147:20035/emcpublish/). To accurately represent local air quality of the sampling site, we selected the Gucheng site (4.4 km away from Shijingshan $PM_{2.5}$ sampling site). The Data data include NO_2, SO_2 concentrations.

RESULTS AND DISCUSSION

Variation of $PM_{2.5}$ Mass Concentrations

Meteorological data during the sampling period are shown in Table 1. Daily temperatures ranged from 3.7°C to 13.2°C. Wind speed was low (5.1–17.2 km h^{-1}). Relative humidity (RH) was from 19% to 67%. Concentrations of $PM_{2.5}$, SO_2 and NO_2 during the sampling period in Shijingshan are presented in Fig. 1. The $PM_{2.5}$ concentration range was 67–294 μg m^{-3}, which was almost always higher than the limit given by National Ambient Air Quality Standard (NAAQS, GB3095-2012) Grade II (75 μg m^{-3}), exceeding it by times except on Feb.12, Feb.18 (8:10 a.m.–4:10 p.m.) and Feb. 27. In contrast, SO_2 and NO_2 were below the limits (150 μg m^{-3} for SO_2, 80 μg m^{-3} for NO_2).

According to the "Observation and forecasting levels of haze" (Sarkar *et al.*, 2010), when relative humidity is < 80%, haze is an atmospheric phenomenon during which dry particles obscure the clarity of the sky, giving a visibility is less than 10.0 km. Using this definition, the sampling days can be separated into the non-haze days and haze days. There were four samples (Feb. 19, 0:30 a.m.–8:30 a.m., Feb. 19, 8.30 a.m.–20:30 a.m., Mar. 5, 18:00 p.m.–Mar. 6, 6:00 a.m., Feb. 18, 16:20 p.m.–Feb. 19, 0:20 a.m.) collected during

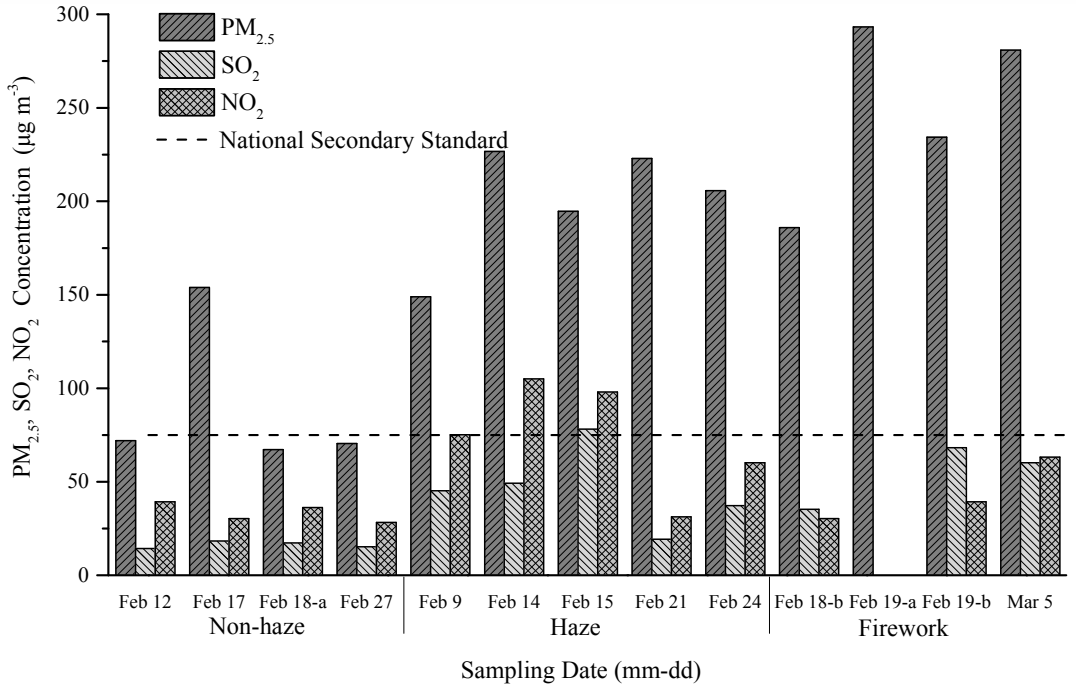

Fig. 1. Time series of $PM_{2.5}$, SO_2 and NO_2 mass concentrations during the sampling period in Shijingshan. The left, middle and right bars in the Figure denote measured values of $PM_{2.5}$ concentration, SO_2 and NO_2 concentrations, respectively, and the dashed line is the national secondary ambient air quality standard for daily $PM_{2.5}$ concentration (75 μg m^{-3}).

intensive firework displays, a typical custom of the Spring Festival. These samples were therefore defined as 'fireworks days'. Thus, the sampling period was divided into non-haze, haze and fireworks phases (see Table 1). The $PM_{2.5}$ average concentration on non-haze days was 90.8 μg m^{-3}. Compared with results from other studies in Beijing, the mass concentration of non-haze day in the 2015 Spring Festival was lower than that on typical clear days (Wang *et al.*, 2006; Shen *et al.*, 2009; Sun *et al.*, 2013c). This suggests that $PM_{2.5}$ concentrations on non-haze days during the Spring Festival were strongly affected by the sharp decrease in population and human activity. The $PM_{2.5}$ average concentrations were 199.9 and 248.9 μg m^{-3} on haze and fireworks days respectively, approximately 2.2 and 2.7 times higher than this on non-haze days. The $PM_{2.5}$ concentration on firework days was approximate 1.2 times higher than on haze days. Compared with other studies, the average $PM_{2.5}$ concentration on firework days was similar to that of 255.3 μg m^{-3} measured in Shanghai (Feng *et al.*, 2012), but higher than the 184.0 μg m^{-3} and 116.5 μg m^{-3} measured in 2006 and 2012 in Beijing (Wang *et al.*, 2007; Cheng *et al.*, 2014), which may be explained by the sampling location of the above two studies being part of the area covered by the"Limited Fireworks" policy, while the sampling site in this study was not. However, the $PM_{2.5}$ concentration was much lower than that measured in other firework display studies in Lucknow (352 μg m^{-3} during Diwali festival) (Barman *et al.*, 2009), Delhi (588 and 389 μg m^{-3} during Deepawali festivals in 2007 and 2008, respectively) (Tiwari *et al.*, 2012), and the YanShuei Area of Southern Taiwan (437 μg m^{-3} during the beehive fireworks display) (Lin *et al.*, 2014). For the former two studies, high relative humidity, low wind speeds in November (the Diwali and Deepawali festivals month) contributed to the high $PM_{2.5}$ concentrations. Furthermore, there is uncertainty in the scale and intensity of the firework displays, which could strongly effect air pollution levels, such as that during "the beehive fireworks display" at one site in Taiwan which seemed to seriously aggravate air quality much more than civil firework burning. It is acknowledged that firework displays release large amounts of gaseous pollutants (Ravindra *et al.*, 2003) and particulate matter (Vecchi *et al.*, 2008), which results in elevated levels of $PM_{2.5}$. A study of in Chengdu in 2009–2010 also observed that the average $PM_{2.5}$ mass concentration during a firework display (212.7 μg m^{-3}) was higher than that on haze days (187.3 μg m^{-3}) (Wang *et al.*, 2013). Thus, much attention has been paid to this serious short-term air pollution. The average concentrations of SO_2 on firework days was 57.8 μg m^{-3}, higher than on haze days and non-haze days. However, the average concentration of SO_2 on firework days was less than that measured in a study in 2006 in Beijing (Wang *et al.*, 2007). This was probably because that 2015 was the first year in which a sulphur-free propellant technology was used in civil fireworks, the same type as those used in the APEC (2014) fireworks display (http://report.qianlong.com/33378/2014/11/11/8785@9991966.htm). In addition, (Hooper and Johnson, 1999) fireworks sales in 2015 decreased by 41% compared with 2014 since people are gradually aware of the seriousness of pollution (http://www.askci.com/chanye/2015/02/24/123461ywk.shtml).

Previous studies have shown that the relatively stagnant meteorological factors and low mixed boundary layer can prevent pollutants diffusion and result in the accumulation of pollution concentrations and species in atmosphere (Sun *et al.*, 2013a, b; Sun *et al.*, 2013c). Low wind speed and high relative humidity as external meteorological factors promote high $PM_{2.5}$ concentration, especially in winter (Sun *et al.*, 2006; Sun *et al.*, 2013c; Tian *et al.*, 2014a). In our study, low wind speed and low temperatures (shown in Table 1) covered all three periods (i.e., haze, non-haze and firework days). Although the emission density of common anthropogenic pollutants declined during the sampling period, meteorological factors might play a primary role in the emergence of haze during the Spring Festival. The average ambient visibilities during the firework days averaged 7.8 km. On Feb. 19, 0:30 a.m.–8:30 a.m., when the average $PM_{2.5}$ concentrations reached the maximum (293.7 μg m^{-3}), the mean visibility reached its lowest value of 5 km. This is likely due to the firework displays generally happening at midnight (12:00 a.m.) and dawn (about 5:00 a.m.) as is the traditional Chinese custom. Qin *et al.* (2013) reported similar results, noticing that the visibility was only 1.2 km when the PM_{10}, $PM_{2.5}$ and PM_1 hourly concentrations reached a maximum value on the eve of the Spring Festival. Aerosol loadings and visibility degradation are negatively correlated (Watson, 2002; Lee *et al.*, 2005; Shen *et al.*, 2009; Shen *et al.*, 2014).

Water-Soluble Inorganic Ion in $PM_{2.5}$

Water-soluble inorganic ions (WSIIs) are important components of atmospheric particles and play a significant role in the ambient air quality and visibility. Fig. 2 illustrated the variations of WSIIs during the sampling periods. A high proportion of WSIIs in particulates occurred on haze and firework days, at 44.7% (76.0 μg m^{-3}) and 50.8% (126.3 μg m^{-3}) respectively, whereas during non-haze days it was 21.6% (19.6 μg m^{-3}).

On non-haze days, the dominant ions were the secondary inorganic ions (SNA: SO_4^{2-}, NO_3^- and NH_4^+), which accounted for approximately 73.6% of the total WSIIs. The proportion of WSIIs on typical winter days approaching 90% (Gao *et al.*, 2015; Chen *et al.*, 2017), was higher than the proportion on non-haze days during the 2015 Spring Festival. The short-term holiday effect was very clear. On haze days, ion concentrations in $PM_{2.5}$ were in order NO_3^- > SO_4^{2-} > NH_4^+ > Cl^- > K^+ > Ca^{2+} > Na^+ > F^- >Mg^{2+}. Approximately 83.9% of the total measured WSIIs were attributed to SNA, and the concentrations of SNA on haze days was 63.7 μg m^{-3}, 5.5 times higher than that on non-haze days. Thus, secondary aerosol species were relatively abundant during haze events. In particularly, the concentration of NH_4^+ on haze days was 6.2 times that on non-haze days. SO_4^{2-} and NO_3^- are formed by the conversion of gaseous precursors (SO_2 and NO_2), which on haze days in our study were higher than on non-haze days (shown in Fig. 1). Higher SO_2 emissions and adverse weather conditions (e.g., low temperature, low wind speed and high RH) during haze days could accelerate aqueous reactions and increase conversion

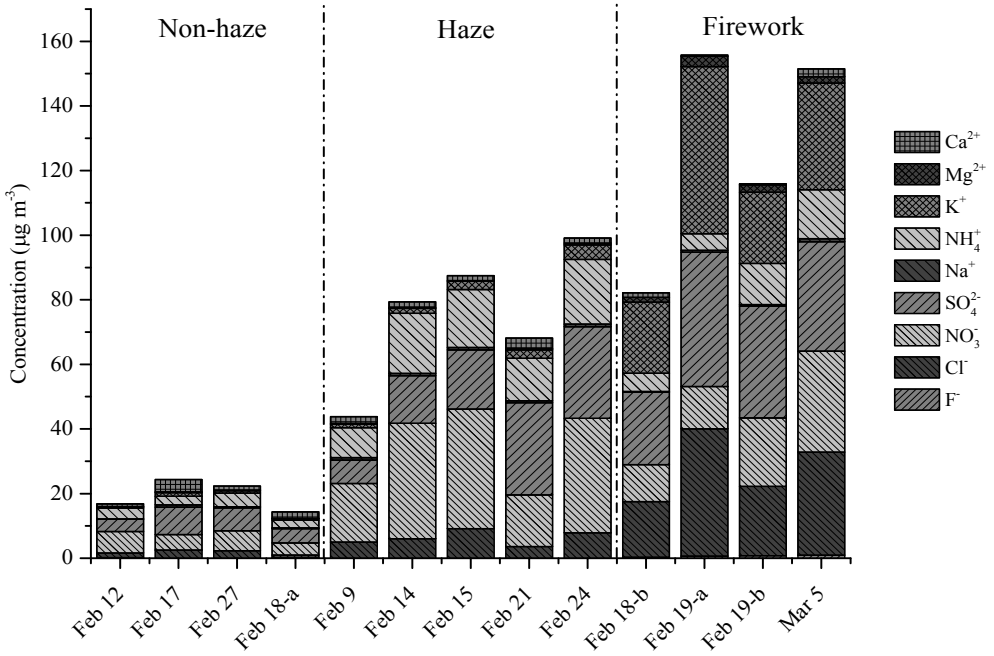

Fig. 2. The water-soluble inorganic ionic composition of $PM_{2.5}$ in Shijingshan.

efficiency of the accumulating secondary particles (He *et al.*, 2001, 2002; Sun *et al.*, 2013b, 2016; Zhang *et al.*, 2016).

However, compared with the data during the haze episode, the order of the ion concentrations on fireworks days was very different, with $SO_4^{2-} > K^+ > Cl^- > NO_3^- > NH_4^+ > Mg^{2+} > Ca^{2+} > F^- > Na^+$. The concentration of SNA was 62 μg m^{-3} accounting for 49.6% of the total WSIIs on firework days. The concentrations of NH_4^+ and NO_3^- were both clearly lower than on haze days. NO_3^- is generally produced by fossil fuel combustion and vehicle emissions, and traffic may also be an important source of ammonia in Beijing (Ianniello *et al.*, 2010; Wang *et al.*, 2014). Therefore, the decreases of NH_4^+ and NO_3^- concentrations were largely related to people leaving Beijing for their hometowns for the Spring Festival, leading to a decline in the population and vehicle. These results are similar to those of other studies of the impacts of firework displays conducted in Jinan by Yang *et al.* (2014). The chemical compositions of particulates during firework days was strongly enriched in Cl^- and K^+, which made up 21.5% and 25.2% of total WSIIs. The concentration of K^+ in particulates during firework days was approximately 13 and 49 times that of haze days and non-haze days, respectively. Prior studies also have shown that K^+ can be enriched in firework aerosols and may be a tracer of PM from fireworks (Drewnick *et al.*, 2006; Vecchi *et al.*, 2008; Cheng *et al.*, 2014). Wang *et al.* (2007) reported that firecrackers contain 75% KNO_3. Notably, KNO_3, $KClO_3$ and $KClO_4$ are usually used as oxidizer, and converted into KNO_2 and KCl if there is sustained burning of fireworks (Yang *et al.*, 2014).

To further distinguish the characteristics of $PM_{2.5}$ between fireworks days and haze days, several ion ratios were calculated and compared (Fig. 3). Previous research has recognized the NO_3^-/SO_4^{2-} ratio as an indicator of the influence of mobile versus stationary pollution sources (Arimoto *et al.*, 1996; Hu *et al.*, 2002; Ye *et al.*, 2003). However, the ratio of NO_3^-/SO_4^{2-} not only indicates the intensity of primary emissions, but also depends on chemical reactions. In our data, the ratio of NO_3^-/SO_4^{2-} decreased from 1.5 on haze days to 0.9 on non-haze days and 0.6 on firework days. In Xi'an, the ratio was 0.4 on non-haze and 0.7 on haze days (Shen *et al.*, 2009). During a heavy haze pollution episode in 2013, the ratio was 0.73 in Beijing (Wang *et al.*, 2014). High mass ratio in this study suggests more contribution of mobile sources in the observation episode. A low NO_3^-/SO_4^{2-} ratio on firework days was attributed to increased SO_4^{2-} caused by larger precursor SO_2 emissions from fireworks. Compared with firework days, the high ratio of NO_3^-/SO_4^{2-} on haze days was mainly due to the increase of nitrate above that of sulfate. K^+ and Cl^- are mainly produced by primary emissions. The ratio of Cl^-/K^+ (0.9) was lowest on firework days because the concentrations of K^+ were enriched. The abundance of K^+ on firework days also affected the ratio of SO_4^{2-}/K^+, which varied considerably when compared with that on non-haze days and haze days. Examination of these three ratios would be useful to help distinguish firework days from other events.

Ion balance calculations are commonly used to study the acid–base balance of aerosol particles. We converted the ions' mass concentrations into microequivalents to evaluate the cation/anion balance of $PM_{2.5}$. The cation and anion microequivalents in the three types of $PM_{2.5}$ samples were calculated as follows:

$$C \text{ (cation microequivalents)} = Na^+/23 + NH_4^+/18 + K^+/39 + Mg^{2+}/12 + Ca^{2+}/20 \quad (1)$$

$$A \text{ (anion microequivalents)} = F^-/19 + Cl^-/35.5 + NO_3^-/62 + SO_4^{2-}/48 \quad (2)$$

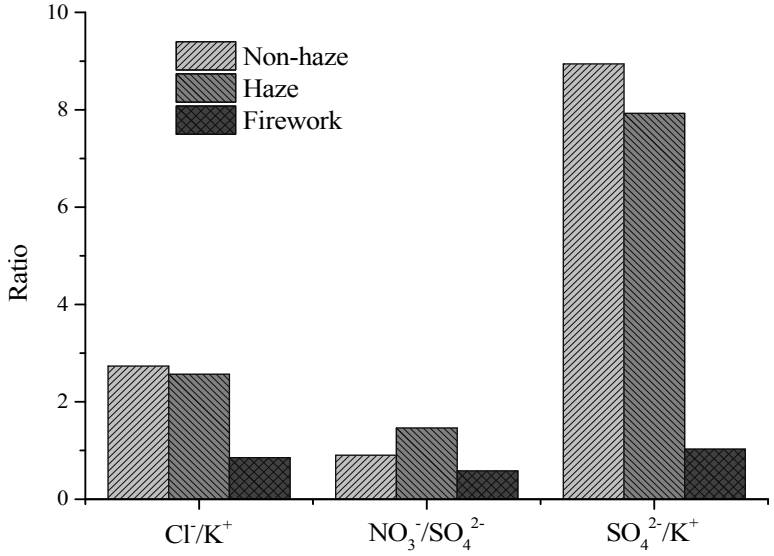

Fig. 3. Ratios of Cl^-/K^+, NO_3^-/SO_4^{2-}, SO_4^{2-}/K^+ in $PM_{2.5}$ on non-haze, haze and firework days.

The mean equivalent ratios of A/C during non-haze, haze, and firework episodes were 0.84, 0.95, and 1.10 for $PM_{2.5}$ respectively. Correlations between cation and anion equivalents for the three types of $PM_{2.5}$ samples were observed (Fig. 4). Most of which were close to unity, indicating that the eight ion species in these samples were the major ions comprising $PM_{2.5}$ and they were basically neutral for this study even though carbonate and bicarbonate were not measured in the samples. The mean equivalent ratios of A/C suggested that particles on fireworks days were more acidic than those on haze and non-haze days. The relatively higher A/C ratios during fireworks days could be due to the higher concentrations of SO_4^{2-}.

Element Species in $PM_{2.5}$

Total mass concentrations of 24 elements accounted for about 12.3%, 8.0% and 12.7% of $PM_{2.5}$ on non-haze, haze and fireworks days, respectively. Al, Ca, Fe, and S were the dominant elements on non-haze days, accounting for 82.3% of all measured elements (Fig. 5). The concentrations of As, Ba, Cd, Cr, Cu, Pb, S, Se, and Zn during haze days were 2.1–10.4 times higher than on non-haze days. Concentration of the other measured elements on haze days were all < 1 μg m^{-3}, together accounting for 87.9% of all measured elements.

The concentrations of typical fireworks elements Al (8.0 μg m^{-3}), Mg (5.7 μg m^{-3}), S (8.8 μg m^{-3}), and Ba (2.4 μg m^{-3}) were higher during firework days than non-haze and haze days, and the sum of these accounted for 79.0% of all measured elements. The levels of some pollution elements (e.g., Al, Ba, Cu, Mg, Pb, S, Sr, and Zn) on firework days were 1.6–18.6 times higher than those on haze days, which indicated that the high concentrations of these elements was at least practically due to the fireworks. A similar result was found in India, where metal concentrations during Diwali fireworks were 5–80 times greater than those on non-haze days (Chatterjee *et al.*, 2013). However, Do *et al.* (2012) found that high levels of Na, K, Si, Al, Mg, Ba, Ca, and Fe were present in the atmosphere prior to the fireworks display. Enhanced concentrations of Pb, Sr, Cu, and Ba during fireworks days had also been observed in Beijing (Wang *et al.*, 2007) and Chengdu (Wang *et al.*, 2013).

Enrichment factors of elements in $PM_{2.5}$ during the non-haze, haze, and fireworks are shown in Table 2. According to Lantzy and Mackenzie (1979), crust (surface soil) was primary source of elements in the atmosphere when that element's enrichment factor (EF) is less than 10, which means that the element from soil or rock weathering. If the EF is between 10 and 10^4, it means that the element enrichment was the result of crust and human activities (Tang *et al.*, 2006). Following this approach, we classified 24 elements into three groups:

a. Crustal elements (EF < 10 on all three types of days), including Ca, Al, Fe, Na, Co, Ni, P, Ti, and V, which could be mainly produced by high-intensity of crustal dust. The EF of crustal elements remained stable during the sampling period, which is due to these elements being largely from natural sources (e.g., soil and construction materials) and rarely transported into the atmosphere, hence, their contribution to PM on haze days is limited. Stable weather conditions and high RH are also important factors (Tian *et al.*, 2014b).
b. Common anthropogenic pollution elements (EF > 10 in all three types of days), such as As, Cd, Cu, Pb, S, Sb and Zn, which are mainly attributed to anthropogenic sources. Studies have indicated that the elements (e.g., As, Cd, Cr, Cu, Ba, Pb, Se, and Zn) would mainly arise from anthropogenic sources (e.g., industrial activities, fossil fuel combustion and vehicle emissions) (Monaci *et al.*, 2000; Marcazzan *et al.*, 2001; Gao *et al.*, 2015).
c. Fireworks elements (EF > 10 on firework days, and significantly higher than on haze days), made up of Ba, Cr, Cu, Mg, Pb, S, Si, and Zn, since they are generally used as fuels, oxidizers and coloring agents in fireworks. Tsai *et al.* (2012) found firework powders burn and release a large amount of metal-related aerosol particles into the atmosphere. Cu, Sr, and Ba are used to create fireworks

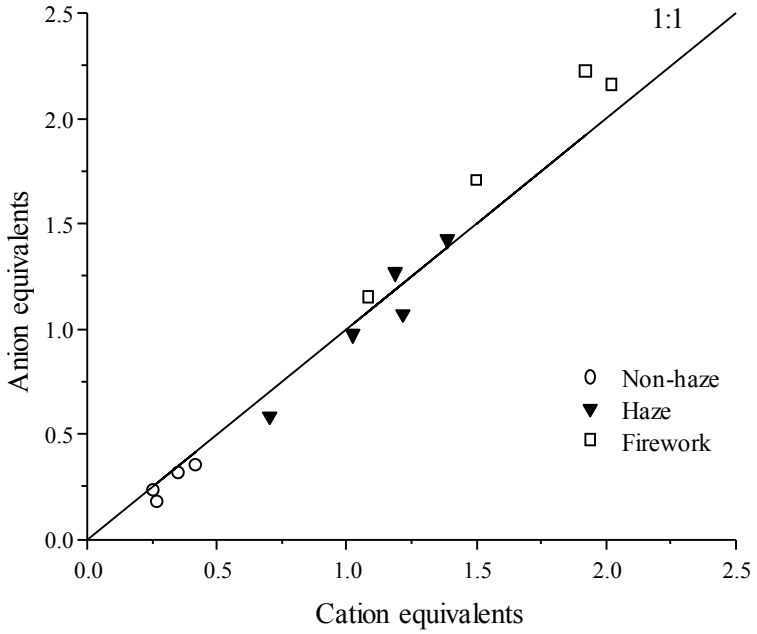

Fig. 4. Total anion microquivalents versus total cation microequivalents for the non-haze, haze, and firework $PM_{2.5}$.

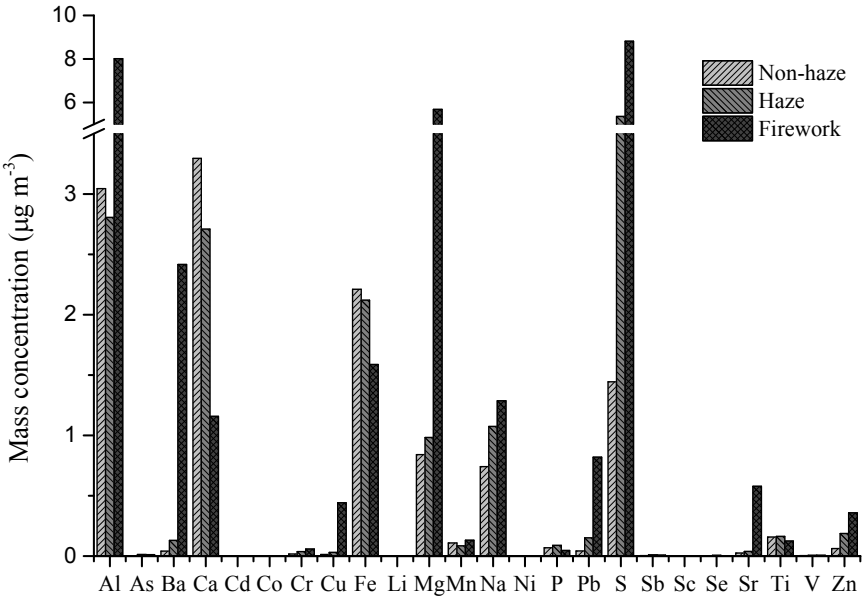

Fig. 5. Mass concentrations of elements in $PM_{2.5}$ during the sampling period.

coloring effects (blue, red, and green, respectively) (Do *et al.*, 2012). Mg is used as a fuel and a producer of sparks (Lancaster and Shimizu, 1998). $CaCl_2$ and $CaSO_4$ are used to produce orange sparks (Moreno *et al.*, 2007).

CONCLUSION

This study provides insights into the chemical compositions of $PM_{2.5}$ during the 2015 Spring Festival Period and explores the differences in chemical characteristics of $PM_{2.5}$ between non-haze days, haze days and firework days as well as the overall effects of short-term holidays such as the Spring Festival.

In most days during Spring Festival, the $PM_{2.5}$ concentration exceeded the NAAQS (GB3095-2012) Grade II standard (75 μg m^{-3}) and ranged from 67 to 294 μg m^{-3}. High $PM_{2.5}$ average concentrations were observed during firework days, approximately 2.7 and 1.2 times higher than that on non-haze days and haze days, respectively. Meanwhile, the concentration of SO_2 and NO_2 were both below the national allowable concentrations (150 μg m^{-3} for SO_2, 80 μg m^{-3} for NO_2) during the sampling period. The SO_2 concentration during firework days was less than that measured in previous research into the effects of fireworks because of the introduction of sulphur-free propellant technology in civil fireworks.

Table 2. Enrichment factors of elements in $PM_{2.5}$ during the sampling period.

Elements	Abundance in crust (%)[a]	Non-haze				Haze					Firework			
		Feb 12	Feb 17	Feb 27	Feb 18-a	Feb 9	Feb 14	Feb 15	Feb 21	Feb 24	Feb 18-b	Feb 19-a	Feb 19-b	Mar 5
Al	7.96	0.71	1.08	0.85	0.98	0.73	0.68	0.77	1.01	1.11	3.72	12.28	6.62	4.15
As	0.00017	113.57	37.38	89.53	/	185.40	447.33	480.04	51.65	250.18	195.46	622.16	1133.77	284.52
Ba	0.0584	/	1.56	N/A	/	0.00	0.00	1.46	3.50	14.65	123.83	548.43	246.41	176.58
Ca	3.85	2.58	2.54	1.88	1.70	2.85	2.54	2.31	1.38	1.31	1.78	1.02	1.47	2.01
Cd	0.00001	411.08	96.68	355.57	85.87	587.58	910.19	1625.55	102.83	558.77	474.83	1297.09	1463.19	938.78
Co	0.0024	2.90	1.60	1.95	1.35	2.17	3.38	2.22	1.33	1.65	2.80	10.64	4.95	3.57
Cr	0.0126	18.79	1.78	3.59	3.77	10.26	33.32	6.54	1.58	2.50	23.06	49.94	25.40	21.75
Cu	0.0025	26.11	16.18	16.74	10.68	31.16	41.84	46.08	13.62	74.73	646.09	2853.95	947.36	539.08
Fe	4.32	1.83	1.37	1.43	0.87	1.28	1.17	2.01	1.03	1.09	1.35	5.09	0.86	1.52
Li	0.0018	4.50	4.04	4.40	2.49	5.16	6.32	7.53	3.30	5.32	5.87	11.72	10.30	7.25
Mg	2.2	1.04	1.18	1.00	0.67	1.16	1.12	1.19	1.06	1.61	8.50	37.22	14.64	10.17
Mn	0.0716	6.01	3.21	7.16	3.73	4.08	4.14	6.16	1.95	2.74	7.98	17.44	8.81	10.45
Na	2.36	0.98	0.86	1.02	0.54	1.38	1.41	1.80	0.77	1.41	2.03	5.49	4.70	2.20
Ni	0.0056	0.81	1.50	0.08	/	1.39	/	3.75	0.96	0.72	0.87	4.42	3.12	2.94
P	0.0757	1.51	2.44	0.05	0.07	2.88	0.46	3.59	1.64	1.70	0.12	0.00	1.45	2.41
Pb	0.00148	133.35	58.15	207.85	36.32	167.45	304.79	843.93	62.89	421.71	1514.37	9387.49	3606.50	1621.13
S	0.0697	109.35	42.37	108.69	30.51	111.03	231.17	314.95	136.74	322.73	423.19	1438.39	1410.03	369.47
Sb	0.00003	982.01	144.01	290.77	350.51	2288.96	2071.35	1096.18	96.36	521.89	600.86	2936.97	3014.81	945.37
Sc	0.0016	1	1	1	1	1	1	1	1	1	1	1	1	1
Se	0.000012	/	/	1071.09	/	658.91	1933.08	4578.42	79.66	2415.25	1990.01	/	3279.91	2049.68
Sr	0.0333	2.42	2.29	1.78	1.71	2.28	2.83	2.35	1.84	7.61	57.73	280.22	108.07	51.12
Ti	0.401	1.14	1.15	1.02	0.72	1.12	1.13	1.11	1.04	0.96	1.36	3.42	1.55	1.43
V	0.0098	1.09	1.40	1.82	0.78	1.21	1.53	3.90	1.25	2.21	2.71	8.18	4.60	2.63
Zn	0.0065	41.30	25.39	39.80	10.48	92.91	136.87	197.88	14.21	70.36	168.60	619.43	422.56	250.46

[a] Adapted from: The composition of the continental crust* (Wedepohl, 1995).

SO_4^{2-}, NO_3^- and NH_4^+ were the dominant ions during haze days, while the concentrations of NH_4^+ and NO_3^- significantly decreased during firework days. The $PM_{2.5}$ chemical compositions on firework days were dominated by Cl^- and K^+, largely exceeding those on non-haze days and haze days. Particulates on firework days were more acidic than those on haze and non-haze days, mainly due to the high concentrations of SO_4^{2-}.

According to enrichment factors of elements in $PM_{2.5}$ during the sampling period, we classified 24 elements into crustal elements (Ca, Al, Fe, Na, Co, Ni, P, Ti, and V), common anthropogenic pollution elements (As, Cd, Cu, Pb, S, Sb and Zn) and fireworks elements (Ba, Cr, Cu, Mg, Pb, S, Si, and Zn). This allowed for further investigation of the differences in chemical characteristics between common air pollution and short-term special air pollution (e.g., from fireworks).

Our results demonstrate that the air quality of normal days in holidays was better than that in non-holidays. But burning of fireworks contributes to sudden and dramatic air pollution, which is much more severe than haze pollution in this study. Differences in chemical characteristics indicated that holidays such as the Spring Festival can affect air pollution patterns in two ways: a decrease in the population and vehicles but an increase in activities such as firework displays. Thus, special human activities during holiday periods should be seriously considered when we take measures to improve air quality.

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