Characteristics of air quality in Tianjin during the Spring Festival period of 2015

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ABSTRACT
To better understand the characteristics of air quality and the relationship between the chemical composition evolution and source variation, an intensive atmospheric campaign was conducted in Tianjin, a megacity of the North China Plain, from 10 February to 6 March 2015. There were 20 days exceeding the threshold value of secondary Chinese Ambient Air Quality Standards for PM2.5 (75 μg m−3, daily average over 24 h) during the study period. Five air pollution episodes were selected for investigation. During the pre-holiday pollution episode, NH4+ and NO3− were more abundant, indicating that air pollution was caused by motor vehicle exhaust emissions and coal consumption under unfavorable meteorological conditions. During Chinese Lunar New Year’s Eve, widespread use of fireworks resulted in extremely high aerosol concentrations. Firework displays caused increases in K+ and also enrichment of SO42− relative to NO3−. The holiday pollution episode was caused by regional transport, characterized by abundant SO42− and NH4+. In addition, the aging processes of the particles from fireworks discharge played an important role in the formation of NO3− and SO42−. The Lantern Festival episode was characterized by a transition from the enrichment of K+ to secondary inorganic ions (NO3−, SO42−, and NH4+). The results of this study are useful for a detailed understanding of the variation in atmospheric compositions and sources caused by anthropogenic activity, and highlight the importance of controlling intensive fireworks discharge.

1. Introduction
The Spring Festival is the most important festival for the Chinese people. Emissions sources vary greatly during the Spring Festival due to the variety of associated human activity. Many factories and businesses shut down for at least a week, and sometimes as long as 15 days. Numerous migrant residents leave the metropolises and return to their home towns to celebrate the traditional Chinese Spring Festival with their families. The decline in emissions from industrial facilities and vehicles during the Spring Festival improves air quality in the metropolises, while fireworks discharge (FD), which is the people’s favorite traditional custom and practice, causes degradation in air quality (Kong et al. 2015) and health hazards (Wang et al. 2007). As a result, the long holiday during the Spring Festival provides us with a unique opportunity to evaluate the effects of such variation in anthropogenic emissions.

Tianjin has experienced rapid economic growth over the past two decades, leading to a general decline in air quality (Wu et al. 2006). Tianjin faces serious air pollution problems, including high levels of particulate matter (PM) and ozone (Gu et al. 2010). In recent years, haze and fog have begun to occur frequently, causing severely low visibility (Xin et al. 2012). Hence, there is a great need to implement control measures to improve air quality and protect public health.

To better understand the characteristics of air quality and the chemical composition evolution and source variation during the Spring Festival, an intensive field campaign was conducted in Tianjin around the Chinese Spring Festival. In this study, the impact of fireworks on air quality were evaluated, with the aim to help identify possible mitigation practices of concern for short-term air pollution effects, which are key issues for researchers and policymakers alike. Also investigated were the chemical characteristics at different PM2.5 levels and the role of meteorological parameters in the formation and evolution.
of PM$_{2.5}$ episodes, with a focus on the components strongly associated with secondary aerosols and FD.

2. Methodology

2.1. Sampling site

Tianjin (38°33′–40°15′N, 116°42′–118°04′E), located on the North China Plain and on the west Bohai Sea coast, covers an area of 11 919 km$^2$ and has a population over 14.72 million. It is a highly urbanized and industrialized city in China. The vehicle population reached approximately 240 million in 2013. The Gross Domestic Product in Tianjin was worth 2320 billion U.S. Dollars in 2013. The sampling site was located in downtown Tianjin. All the monitoring instruments were installed on the roof of the Laboratory of Air Pollution Monitoring, Tianjin Environment Monitoring Center (39°5′49.30″N, 117°9′4.15″E). There were no industrial sources in the immediate vicinity of the sampling site. The field measurements were collected from 10 February to 6 March 2015.

2.2. Instruments

Ambient NO, NO$_2$, SO$_2$, CO, O$_3$, PM$_{2.5}$, and PM$_{10}$ were measured in situ at Tianjin Environment Monitoring Center from 10 February to 6 March 2015. Detailed descriptions of the calibration and maintenance of the analyzers for NO, NO$_2$, SO$_2$, CO, O$_3$, PM$_{2.5}$, and PM$_{10}$ can be found in Ji et al. (2014). All of the above species were recorded at a resolution of 1 min; however, only the 1-h data are presented in this paper. The organic carbon (OC) and elemental carbon (EC) levels in PM$_{2.5}$ were measured with a thermal optical transmission OC/EC analyzer (RT-4, Sunset Laboratory Inc., Oregon, USA). For charring correction, an He–Ne laser beam monitored the sample transmittance throughout the heating process. When the laser signal returned to its initial value, the split point between OC and EC was determined (Birch and Cary 1996). An ambient ion monitor (Model URG-9000D, URG Co., USA) was deployed to measure the hourly concentrations of water-soluble inorganic ions in PM$_{2.5}$. Multi-point calibrations were performed every four days after changing the eluent solutions. The uncertainties were approximately 10%, and the estimated detection limits ranged from 0.010 to 0.084 μg m$^{-3}$ for SO$_4^{2-}$, NO$_3^-$, NH$_4^+$, Ca$^{2+}$, Cl$^-$, K$^+$, F$^-$, Na$^+$, and Mg$^{2+}$.

3. Results

3.1. Temporal variation of air pollutants

The study period lasted from 10 February to 6 March 2015, covering one of the most important Chinese traditional holidays, the Chinese Spring Festival. People were allowed to set off fireworks between 0600 and 2200 Local Standard Time (LST) from 15 to 20 February, and on 5 March. On New Year’s Eve, the permitted period extended until 0200 LST. Figure 1 shows the times series of hourly PM$_{2.5}$, PM$_{10}$, NO, NO$_2$, SO$_2$, CO, and O$_3$ concentrations for the whole study period.

Figure 1. Variation in PM$_{2.5}$, PM$_{10}$, SO$_2$, CO, NO, NO$_2$, and O$_3$ concentrations for the whole study period.
concentrations. The first pollution episode occurred during 13–15 February. The average PM$_{2.5}$ and PM$_{10}$ concentrations reached 162 ± 76 and 190 ± 83 μg m$^{-3}$, respectively. Correspondingly, the CO concentration occurred with a mean value of 3.0 ± 1.4 mg m$^{-3}$, which was higher than the average concentration of CO during the whole study period (2.1 ± 1.4 mg m$^{-3}$) (Figure 1). As this pollution episode occurred before the Spring Festival, it is referred to as the ‘pre-holiday pollution episode (E1)’. The PM$_{2.5}$ concentrations showed obvious decreases from 0600 LST 15 February, because the increase in wind speed favored the diffusion of pollutants with the passage of a cold front.

Extremely high particle concentrations occurred on Chinese Lunar New Year’s Eve (E2). The PM$_{2.5}$ concentrations began to rise rapidly at 2200 LST 18 February, peaked at 0200 LST 19 February, and then declined and remained at 103 μg m$^{-3}$ until 0700 LST (Figure 1). The hourly PM$_{2.5}$ peak reached 428 μg m$^{-3}$. Correspondingly, the peak value of hourly PM$_{10}$ reached 714 μg m$^{-3}$. The average ratio of PM$_{2.5}$/PM$_{10}$ was 0.52 ± 0.11 during E2, which was lower than the average ratio during the whole study period (0.71 ± 0.23). This suggested that FD contributed more to coarse particles. The average SO$_2$, NO$x$, O$_3$, and CO concentrations reached 144, 44, 41 μg m$^{-3}$ and 2.0 mg m$^{-3}$ at this time, respectively. The intensive and widespread exploding of fireworks was responsible for this burst of heavy pollution, as all the residents were simultaneously celebrating Lunar New Year. The average visibility was 4.9 km, and we defined this period as the ‘firework pollution episode’. Afterwards, PM$_{2.5}$ concentrations dropped considerably due to favorable meteorological conditions.

Another severe PM pollution episode (E3) was recorded during the Chinese Lunar New Year holiday, characterized by an explosive growth in PM$_{2.5}$ concentrations within a few hours. The hourly mean concentrations of PM$_{2.5}$ started to increase at 0000 LST 21 February, reaching peak concentrations at 1700 LST 21 February. Correspondingly, as shown in Figure 1, some pollution gases (i.e. CO, SO$_2$, and NO$_x$) also had similar temporal patterns. The gaseous pollutant concentrations were lower than those during the pre-holiday pollution episode. The explanation is that people had relatively flexible travel schedules during the festival compared with their fixed schedules before the festival. Daily traffic emissions did not concentrate.

A dust storm episode was recorded on 22 February. The maximum hourly PM$_{10}$ concentration reached 645 μg m$^{-3}$. Afterwards, aerosol pollution (E4) started to aggravate on 23 February and peaked on 25 February. We attributed the PM$_{2.5}$ peak to a traffic rush after the holiday, when people from other provinces and cities started returning to Tianjin for work. Daily increasing travel flows were reflected by the increase in NO$_x$, CO, and SO$_2$. Visibility was lower during E4, with mean values of 3.0 ± 1.8 km. High relative humidity (62 ± 5%) was one of the important factors accounting for the low visibility.

The last pollution episode (E5) for investigation was recorded around the Lantern festival. The PM$_{2.5}$ concentrations increased from 1800 LST 5 March and persisted until the end of the study period. Correspondingly, some pollution gases (i.e. CO, SO$_2$, and NO$_x$) also had similar temporal patterns. The intensive and widespread exploding of fireworks was responsible for this burst of heavy pollution. In addition, the relative humidity increased to 46.4% and wind speeds decreased to 1.7 m s$^{-1}$ during this period. This stagnant synoptic meteorology favored the local atmospheric processing and accumulation of pollutants.

### 3.2. Characterization of chemical species in PM$_{2.5}$

The variations of water-soluble ionic species and carbonaceous aerosols during the study periods are shown in Figure 2. The total concentrations of water-soluble ionic species in E1 for PM$_{2.5}$ were 81.4 μg m$^{-3}$, accounting for 50.2% of PM$_{2.5}$ mass. The masses of chemical species for

![Figure 2. Profiles of ionic species and carbonaceous aerosols of PM$_{2.5}$ for the whole study period.](image-url)
PM$_{2.5}$ were in the order NH$_4^+$ > NO$_3^-$ > SO$_4^{2-}$ > Cl$^-$ > F$^-$ > Na$^+$ > K$^+$ > Mg$^{2+}$ > Ca$^{2+}$ (Figure 3); NH$_4^+$, NO$_3^-$, and SO$_4^{2-}$ averaged 21.6, 14.0, and 11.9 $\mu$g m$^{-3}$, respectively. The biggest contributor to PM$_{2.5}$ was OC, which averaged 22.2 $\mu$g m$^{-3}$, accounting for 13.7%. Because low wind speed inhibits the dilution effect, meteorological conditions observed during E1 were favorable for the formation of secondary PM. During E1, the average relative humidity and temperature was 52.6% and 4.8 °C, respectively. The increasing relative humidity and relatively low atmospheric temperatures induced the formation of ammonium nitrate. As previously reported, a ratio of OC/EC larger than 2 implies that secondary OC (SOC) is formed. Castro et al. (1999) suggested an equation to evaluate the production of SOC as follows: OC$_{sec}$ = OC$_{total}$ - EC × (OC/EC)$_{min}$, where OC$_{sec}$ is SOC, OC$_{total}$ is the total OC (TOC), and (OC/EC)$_{min}$ is the minimum ratio of OC to EC concentration monitored. Based on the above equation, the POC and SOC concentrations could be calculated. The POC and SOC concentrations were 11.9 and 10.3 $\mu$g m$^{-3}$ during E1, respectively. Thus, the rapid increase in the OC concentrations was caused by both local emissions and secondary formation during E1. EC showed similar variations as OC. The correlation coefficient between OC and EC in PM$_{2.5}$ was strong ($R^2 = 0.68$, $p < 0.0001$), suggesting that the OC and EC originated from similar emissions sources. Figure 3 shows the linear correlation between OC and CO ($R^2 = 0.56$, $p < 0.0001$). CO in the urban area mainly came from vehicle emissions (Guo et al. 2006). Strong correlation between OC and CO implied they were from the same source. Thus, it can be inferred that among the emissions sources, vehicle emissions played an important role in E1.

As shown in Figures 2 and 4, the PM$_{2.5}$ samples provided some insights into their composition from fireworks. The mass concentrations of the PM$_{2.5}$ ions followed the order K$^+$ > Cl$^-$ > SO$_4^{2-}$ > NH$_4^+$ > NO$_3^-$ > Mg$^{2+}$ > Na$^+$ > F$^-$ > Ca$^{2+}$. The K$^+$, Cl$^-$, SO$_4^{2-}$, NH$_4^+$, and NO$_3^-$ concentrations were 65.8, 42.9, 38.9, 8.7, and 1.9 $\mu$g m$^{-3}$, respectively. The K$^+$ and Cl$^-$ concentrations during the peak period were 23.5 and 15.5 times higher than those at 1300 LST on Chinese Lunar New Year, respectively. This is because KNO$_3$, KClO$_3$, KClO$_4$, K$_2$CrO$_4$, and K$_2$Cr$_2$O$_7$ were used as oxidizers in the fireworks (Azhagurajan et al. 2011). Because potassium compounds in black powder (commonly in the form perchlorate or chlorate) act as the main oxidizers, the corresponding chemical equations (2KClO$_3$ = 2KCl + 3O$_2$ and KClO$_4$ = KCl + 2O$_2$) led to high concentrations of Cl$^-$ during burning. In addition, the Cl$^-$/Na$^+$ ratio was approximately 2.9 during the FD period, as Cl$^-$ came from the chlorine content of the fireworks powder. Moreover, the mass ratio of OC to EC was approximately 4.1 in Tianjin during the whole study period, but increased to 4.4 during fireworks display periods. Therefore, the Cl$^-$/Na$^+$ and OC/EC ratio of atmospheric fine particles can be used as an indicator of FD. Fireworks contain charcoal (Tsai, Chien, and Yuan 2012) and organic materials are used as adhesives, such as polyvinyl alcohol, polyoxyethylene, phenol formaldehyde resin, and shell-lac (Kong et al. 2015). They are responsible for elevated OC and EC concentrations during such events (Tsai, Chien, and Yuan 2012). The slopes between the equivalent [K$^+$ + NH$_4^+$] concentration and the sum of SO$_4^{2-}$ and NO$_3^-$ equivalent concentrations were slightly higher than 1, indicating complete neutralization. The NO$_3^-$/SO$_4^{2-}$ ratio decreased with an average value of 0.1, which was lower than the previously reported value of 0.5–0.6 (Wang et al. 2007). The NO$_3^-$ and NO$_2$ concentrations were lower, suggesting that FD contributed little to NO$_3^-$ and NO$_2$. The SO$_4^{2-}$ and SO$_2$ correlation was stronger ($R^2 = 0.55$, $p < 0.0001$), suggesting SO$_4^{2-}$ and SO$_2$ shared the same sources.

During E3, the main water soluble ions in PM$_{2.5}$ samples followed this order: SO$_4^{2-}$ > NH$_4^+$ > NO$_3^-$ > K$^+$ > Cl$^-$ > F$^-$ > Na$^+$ > Mg$^{2+}$ > Ca$^{2+}$. The average total ion concentration in PM$_{2.5}$ during E3 was 59.5 $\mu$g m$^{-3}$, which was equal to 38.2% of the PM$_{2.5}$ mass. SO$_4^{2-}$, NO$_3^-$, Mg$^{2+}$, and Cl$^-$ were enriched in fine particles during E3. To the best of our knowledge, contributions from other human sources were low, and so the contribution form FD and chemical processes related
to FD led to the degradation in air quality. In particular, non-local residents in Tianjin usually return to their home towns to celebrate Chinese Lunar New Year with their families, which can significantly reduce the traffic flow (vehicle emissions obviously declined). In fact, the CO and NO\textsubscript{x} concentrations decreased during the Chinese New Year period, leading to the lower NO\textsubscript{3}\textsuperscript{−} concentrations. However, the NH\textsubscript{4}\textsuperscript{+} and SO\textsubscript{4}\textsuperscript{2−} concentrations still increased. The increased NH\textsubscript{4}\textsuperscript{+} was attributed to anthropogenic activities such as human excrement and mariculture (Streets et al. 2007). The equivalent NH\textsubscript{4}\textsuperscript{+} concentrations had a perfect linear correlation ($R^2 = 0.98$, $p < 0.0001$) with the sum of equivalent SO\textsubscript{4}\textsuperscript{2−} and NO\textsubscript{3}\textsuperscript{−} concentrations in PM\textsubscript{2.5}. Different from E1, air pollution aggravated with the increase in wind speed. The wind speed increased from 1.0 to 3.7 m s\textsuperscript{−1} and the average relative humidity remained 85% during E3, while the average wind speed and relative humidity were 1.8 m s\textsuperscript{−1} and 54%, respectively, during E1. This meant regional transport played an important role in E3. As already reported, intensive FD can influence air quality for about four days (Barman et al. 2009). Southerly wind carried air pollutants emitted from FD on Chinese New Year’s Eve from southern areas of the North China Plain to Tianjin, leading to deterioration in air quality. As shown in Figure 3, the strong correlation between sulfate and K\textsuperscript{+} also demonstrated sulfate and K\textsuperscript{+} came from the same source (i.e. FD). Note that the aging of fresh FD particles existed via heterogeneous reactions during long-range transport. Secondary transformation occurred through condensation onto pre-existing aerosols from intensive FD emissions. During aging, the direct emissions of KCl from fireworks can react with H\textsubscript{2}SO\textsubscript{4} and HNO\textsubscript{3} to form SO\textsubscript{4}\textsuperscript{2−} and NO\textsubscript{3}\textsuperscript{−}, resulting in a decline in the Cl\textsuperscript{−}/K\textsuperscript{+} ratio and increase in the SO\textsubscript{4}\textsuperscript{2−}/K\textsuperscript{+} ratio. In addition, the increase in the OC/EC ratio indicated the gradual formation of secondary (aged) organic aerosols. The slope between the equivalent NH\textsubscript{4}\textsuperscript{+} concentration and the equivalent [SO\textsubscript{4}\textsuperscript{2−} + NO\textsubscript{3}\textsuperscript{−}] concentration was approximately 1.5, which indicated FD particles were alkaline. Further SO\textsubscript{2} and NO\textsubscript{x} emissions might lead to deteriorating air quality through condensation onto pre-existing aerosols.

**Figure 4.** Percentages of the main chemical components in PM\textsubscript{2.5} during the pollution episodes.
With the end of the holiday, the traffic flows gradually increased. Correspondingly, the SO$_2$, NO$_x$, and CO concentrations increased. The secondary inorganic species were at moderate levels. NH$_4^+$, SO$_4^{2-}$, and NO$_3^-$ averaged 14.9, 9.1, and 4.4 μg m$^{-3}$, respectively. The mass ratio of NO$_3^-$ to SO$_4^{2-}$ (0.59) increased compared with the value over the holiday, which indicates that the contribution of mobile emissions became more significant. Another significantly enhanced group of aerosols was OC and EC. OC and EC in PM$_{2.5}$ averaged 15.5 and 4.7 μg m$^{-3}$ from 1600 LST 24 February to 0900 LST 25 February, and they were enhanced by approximately 86.7 and 161.1%, respectively, compared with the average values observed during 1600–1900 LST 23 February. OC and EC were reasonably well correlated, indicating that both were mostly from common major emissions sources in the urban area. In addition, the meteorological conditions also played an important role in E4. The average wind speed was 2.1 m s$^{-1}$ during E4, which was lower than that observed during 1600–1900 23 February (2.5 m s$^{-1}$), while the average relative humidity was 62% during E4, which was greater than that observed during 1600–1900 LST 23 February (25%).

The last pollution episode for investigation was characterized by complicated pollution. A rapid increase in NO$_3^-$, NH$_4^+$, SO$_4^{2-}$, and OC was observed after high K$^+$ and SO$_4^{2-}$ concentrations were caused by FD. FD led to the rapid increase in K$^+$ and SO$_4^{2-}$ during the lantern festival. A strong positive correlation ($R^2 = 0.49$, $p < 0.0001$) between K$^+$ and SO$_4^{2-}$ was found during the FD period. After FD, the K$^+$ and SO$_4^{2-}$ concentrations decreased due to diffusion. Afterwards, a rapid increase in NO$_3^-$, NH$_4^+$, SO$_4^{2-}$, and OC was found with the decrease in wind speed and increase in RH. The lower wind speed and mixing layer heights were favorable for capturing pollution gases on the surface and their secondary transformation (R1), and following condensation onto preexisting aerosols after FD. The high correlation coefficients between the equivalent NH$_4^+$ concentration and the equivalent [SO$_4^{2-}$ + NO$_3^-$] concentration revealed that (NH$_4$)$_2$SO$_4$, K$_2$SO$_4$, NH$_4$NO$_3$, and KNO$_3$ were major sulfate and nitrate salts during the stage of the rapid increase in water-soluble ions. Since NH$_4$Cl and NH$_4$NO$_3$ are unstable, particulate Cl$^-$ or NO$_3^-$ favored being converted into gaseous HCl and HNO$_3$ (R2) when the atmospheric temperature increased. These conversion processes occurred and consequently some SO$_4^{2-}$ and NO$_3^-$ were presumably formed by the condensation of gaseous SO$_2$ and NO$_2$ compounds on the surface of pre-existing KCl particles, as follows:

$$\text{SO}_2(\text{NO}_2) + \text{KCl} + \text{H}_2\text{O} + \text{NH}_3 \rightarrow \text{K}_2\text{SO}_4(\text{KNO}_3) + \text{NH}_4\text{Cl}$$ \hspace{1cm} (R1)

$$\text{NH}_4\text{Cl} (s) \rightarrow \text{NH}_3(g) + \text{HCl}(g)$$ \hspace{1cm} (R2)

Thus, to some extent, FD could cause or influence the complicated pollution. These chemical characteristics of aerosols hinted that emissions from FD combined with local emissions led to a significant alteration of FD aerosols through adsorption of polluted air gases and sequential chemical reactions on their surfaces.

### 4. Conclusions

An intensive atmospheric campaign was conducted in Tianjin, a megacity of the North China Plain, from 10 February to 6 March 2015. Hourly concentrations of major water-soluble inorganic ions (NH$_4^+$, SO$_4^{2-}$, NO$_3^-$, Na$^+$, K$^+$, Ca$^{2+}$, Mg$^{2+}$, and Cl$^-$) and carbonaceous aerosols (OC and EC) in PM$_{2.5}$ and trace gases (SO$_2$, NO$_x$, CO, and O$_3$) in the atmosphere, were measured. Five pollution episodes were identified during the 2015 Spring Festival period. The main conclusions can be summarized as follows:

1. Because low wind speed inhibits the dilution effect, meteorological conditions observed during E1 were favorable for the formation of secondary PM. Based on the OC/EC ratios and the correlation between OC and CO, it was inferred that vehicular emissions played an important role in E1.

2. During Chinese Lunar New Year’s Eve, widespread use of fireworks caused heavy pollution with extremely high aerosol concentrations. SO$_2$, K$^+$, Cl$^-$, and SO$_4^{2-}$ rapidly increased. The fireworks particles were alkaline with comparatively smaller amounts of secondary components. Air pollutants emitted by FD could have been transported to downwind areas where they aggravated air pollution.

3. The conversion of SO$_2$ and NO$_2$ into particle phases, taking place on the surfaces of particles from FD, was an important mechanism of secondary transformation.

This study enhances our understanding of the mechanisms involved in issues of high PM$_{2.5}$ concentrations caused by variations in source emissions. It is evident that stricter control measures on SO$_2$, NO$_x$, and NH$_3$ should be issued and executed to improve air quality. Under unfavorable meteorological conditions, FD should be forbidden and guidance on alternative types of celebration provided.

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