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# Chemical characteristics of haze particles in Xi'an during Chinese Spring Festival: Impact of fireworks burning

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ABSTRACT

Fireworks burning releases massive fine particles and gaseous pollutants, significantly deteriorating air quality during Chinese Lunar New Year (LNY) period. To investigate the impact of the fireworks burning on the atmospheric aerosol chemistry, 1-hr time resolution of $PM_{2.5}$ samples in Xi'an during the winter of 2016 including the LNY were collected and detected for inorganic ions, acidity and liquid water content (LWC) of the fine aerosols. $PM_{2.5}$ during the LNY was 167 ± 87 μg/m$^3$, two times higher than the China National Ambient Air Quality Standard (75 μg/m$^3$). $K^+$ (28 wt.% of the total ion mass) was the most abundant ion in the LNY period, followed by $SO_4^{2-}$ (25 wt.%) and $Cl^-$ (18 wt.%). In contrast, $NO_3^-$ (34 wt.%) was the most abundant species in the haze periods (hourly $PM_{2.5}$ > 75 μg/m$^3$), followed by $SO_4^{2-}$ (29.2 wt.%) and $NH_4^+$ (16.3 wt.%), while $SO_4^{2-}$ (35 wt.%) was the most abundant species in the clean periods (hourly $PM_{2.5}$ < 75 μg/m$^3$), followed by $NO_3^-$ (23.1 wt.%) and $NH_4^+$ (11 wt.%). Being different from the acidic nature in the non-LNY periods, aerosol in the LNY period presented an alkaline nature with a pH value of 7.8 ± 1.3. LWC during the LNY period showed a robust linear correlation with $K_2SO_4$ and KCl, suggesting that aerosol hygroscopicity was dominated by inorganic salts derived from fireworks burning. Analysis of correlations between the ratios of $NO_3^-/SO_4^{2-}$ and $NH_4^+/SO_4^{2-}$ indicated that heterogeneous reaction of $HNO_3$ with $NH_3$ was an important formation pathway of particulate nitrate and ammonium during the LNY period.

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## Introduction

Air pollution caused by fine particulate matter (particles with an aerodynamic diameter less than 2.5 μm, $PM_{2.5}$) has been a persistent problem in China in the past decades due to fast industrialization and urbanization without efficient emission control. $PM_{2.5}$ could penetrate deeply into the bronchi and lungs, leading to respiratory and cardiovascular diseases (Bauer and Koch, 2005; Chow et al., 2006). Recent studies have found that the high level of $PM_{2.5}$ in China has exerted a significant negative

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impact on human health (Sun et al., 2013; Ulbrich et al., 2009; Zhang et al., 2015). $PM_{2.5}$ can be directly emitted from coal and biomass combustion and traffic exhaust (Feng et al., 2012; Lei et al., 2011; Wang et al., 2016b; Xu et al., 2016) and indirectly formed from photochemical oxidation of gaseous precursors and subsequent gas-to-particle phase partitioning. Fireworks burning, as a common human activity during festival events, is one of important sources of air pollution (Camilleri and Vella, 2010; Drewnick et al., 2006; Jiang et al., 2015; Jing et al., 2014). A number of studies have found that pyrotechnic displays on celebratory dates release massive $SO_2$, NOx (Attri et al., 2001; Huang et al., 2012; Wang et al., 2007) and various fine particulate matters including organic and elemental carbon, ions ($K^+$, $Cl^-$ and $SO_4^{2-}$) and metals (*e.g.*, Mg, K, Sr, Ba, Al, Cu and Pb) (Feng et al., 2016; Li et al., 2013; Moreno et al., 2007; Tsai et al., 2012). Those pollutants can significantly deteriorate air quality and raise health hazards especially for the respiratory-sensitive crowd (Becker et al., 2000; Gouder and Montefort, 2014). For example, it was reported that ten-minute exposure to $SO_2$ at concentrations of 0.5 ppm could cause short-term asthma (Gong et al., 1995). In addition, studies (Godri et al., 2010; Hamad et al., 2016) also observed significant positive correlation between particulate oxidative burden and individual trace metals during fireworks burning periods.

Many observations based on filter samples have been conducted to investigate the impact of fireworks burning on air quality. However, such measurements were often performed with a time resolution from 12 to 24 hr, which is much longer than the duration time of fireworks burning activity (Cheng et al., 2014; Kong et al., 2015; Kumar et al., 2016), and thus could result in significant bias on understanding the physical–chemical properties of fine particles derived from fireworks burning due to the interferences of non-firework aerosols collected. Furthermore, those studies have mainly focused on chemical compositions of gaseous and particulate matters from fireworks burning (Huang et al., 2012; Lin et al., 2014; Moreno et al., 2007), and none of them has investigated the acidity and liquid water content (LWC) of aerosols derived from fireworks burning. Acidity is an important property of the aerosol, which could affect the formation of sulfate (Wang et al., 2016a), nitrate (Guo et al., 2016) and secondary organic aerosols (SOA) (Han et al., 2016; Jang et al., 2002). Aerosol LWC is also important for heterogeneous reactions and gas-to-particle phase partitioning of water-soluble compounds (Carlton and Turpin, 2013; Cheng et al., 2016; Wong et al., 2015). High LWC could significantly enhance secondary aerosol formation and increase aerosol light scattering efficiency (Chen et al., 2012; Malm and Day, 2001). Therefore, characterization of physical and chemical properties of aerosols derived from fireworks burning including acidity and LWC with a high time resolution is necessary for a comprehensive understanding of their impact on the environment and human health.

In this study, atmospheric $PM_{2.5}$ aerosols in Xi'an were collected during the Chinese Lunar New Year (LNY) of 2016 with a 1-hr time resolution and measured for inorganic ions, acidity and LWC. We first explored the difference in ion chemistry of the airborne fine particles in the city during the haze, clean and LNY periods, then investigated acidity and LWC of the aerosols in the three periods and finally discussed the formation mechanism of nitrate during the LNY episode.

## 1. Experimental section

### 1.1. Semi-continuous measurement of gas and aerosol phase pollutants

From 29 January to 16 February 2016, the field campaign was conducted at the site of the Institute of Earth Environment, Chinese Academy of Science (34.22°N, 108.88°E) (Fig. 1), in the northwest of Xi'an, inland China. During the sampling period the wind speeds were generally less than 2.5 m/sec with the lowest occurring in the LNY period (Fig. 2b). The concentrations of inorganic ions and associated gaseous precursors were measured with a 1-hr time resolution using *in-situ* Gas and Aerosol Compositions Monitor (IGAC). The IGAC configuration includes three units: (1) wet annular denuder (WAD) for capturing the precursor gas (HCl, $HNO_2$, $HNO_3$ and $SO_2$), (2) scrub and impact aerosol collector (SIC) to trap particle of fine fraction ($PM_{2.5}$), of which operating principle is similar to Steam Jet Aerosol Collector, (3) an ion chromatography (IC) (Dionex, ICS-1100) for the analysis of anions and cations ($SO_4^{2-}$, $NO_3^-$, $Cl^-$, $NO_2^-$, $F^-$, $NH_4^+$, $Na^+$, $K^+$, $Mg^{2+}$ and $Ca^{2+}$). Ambient aerosols from the inlet would flow through the WAD coated with absorption water solution containing $H_2O_2$ (4.2 nmol/L in water), which acts as the biocide and oxidant to promote the gaseous species to diffuse into the aqueous solution. After the WAD the particles enter the SIC, which creates a supersaturated environment for the growth of particles by deliquescence, and the grown particles are finally collected by inertial separation. Two pairs of syringe pumps separately transport the collected gas and particle samples to the IC system, which simultaneously analyze the cations and anions of the sample. More details of a field measurement using IGAC were described elsewhere (Liu et al., 2017; Young et al., 2016).

To evaluate the measurement accuracy of IGAC, $PM_{2.5}$ samples were also simultaneously collected using a high-volume filter sampler (1.13 $m^3$/min) at the same site with a 3-hr time resolution. $PM_{2.5}$ aerosols were collected onto pre-baked (450°C for 6 hr) quartz fiber filter, extracted with pure water and analyzed by the same ion chromatography (Wang et al., 2017b). As shown in Fig. S1, concentrations of major species such as $SO_4^{2-}$, $NO_3^-$ and $NH_4^+$ measured by the IGAC were in a good agreement with those measured by the offline filter method ($y = 1.09x$ for $SO_4^{2-}$, $y = 1.05x$ for $NO_3^-$ and $y = 0.65x$ for $NH_4^+$, respectively). Such a consistence was also reported by Liu et al. (2017), demonstrating the accuracy of the *in-situ* measurement using the IGAC monitor.

### 1.2. $PM_{2.5}$ and meteorological data

The concentration of $PM_{2.5}$ on the sampling days was downloaded from the website of Chinese air quality online monitoring analysis platform (http:// www.aqistudy.cn). Meteorological data of Xi'an, including ambient temperature, relative humidity, wind speed and visibility, were obtained from the Shaanxi Meteorological Bureau and shown in Fig. 2a, b.

### 1.3. Acidity and liquid water content (LWC) of $PM_{2.5}$

#### 1.3.1. Total hydronium ion

Total hydronium ions include *in-situ* $H^+$, bisulfate ions ($HSO_4^-$) and any other $H^+$ in the solid phase of aerosol at nitrate

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