Characterization and Source Analysis of Water-soluble Ions in Atmospheric Particles in Jinzhong, China

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ABSTRACT

Size-segregated samples (< 2.5 , $2.5\text{--}5$, $5\text{--}10$, and $10\text{--}100\text{ }\mu\text{m}$) and $\text{PM}_{2.5}$ samples were collected to analyze the water-soluble inorganic ions (WSIs, including F^- , Cl^- , NO_3^- , SO_4^{2-} , Na^+ , NH_4^+ , K^+ , Mg^{2+} , and Ca^{2+}), through ion chromatography from January to October in 2017 in Jinzhong. The median concentration of the total WSIs in $\text{PM}_{2.5}$ was $37\text{ }\mu\text{g m}^{-3}$, thereby accounting for 31% of the $\text{PM}_{2.5}$, with the lowest level in spring and the highest in autumn. SO_4^{2-} , NO_3^- , and NH_4^+ were the most abundant substances and were primarily on the fine particles ($0\text{--}2.5\text{ }\mu\text{m}$), whereas Ca^{2+} , Mg^{2+} , and F^- were concentrated on the coarse particles ($2.5\text{--}100\text{ }\mu\text{m}$). The results of the correlation analysis led to the conclusions that $(\text{NH}_4)_2\text{SO}_4$, NH_4Cl and K_2SO_4 were the primary compounds on the fine particles, MgSO_4 and CaSO_4 were the major chemical forms of WSIs on the coarse particles, thus indicating that the formation mechanisms of these compounds were different; however, NH_4NO_3 and KNO_3 were present in both the types of particles. The particles that were observed in Jinzhong were alkaline during the study period, and their acidity was negligible. The ratio analysis showed that the highest ratio of Cl^-/K^+ was found in winter in both fine and coarse particles; however, no obvious distinction has been made between $\text{Mg}^{2+}/\text{Ca}^{2+}$ during the four seasons. The $\text{NO}_3^-/\text{SO}_4^{2-}$ ratio in coarse particles was observed to be significantly higher than that in fine particles, particularly in summer, thus indicating that the heterogeneous reaction on particles plays a vital role in the formation of NO_3^- in coarse particles. The PCA analysis showed that the primary factors of WSIs, which were secondary formation, coal combustion, biomass burning, dust particles, and industrial emission. The coal combustion and biomass burning have been considered as the leading emission sources to be controlled for improving air quality in Jinzhong.

Keywords: Fine particles; Coarse particles; WSIs; Size distribution; Source factors.

INTRODUCTION

Over the past few years, China has faced severe air pollution. The 24-hour $\text{PM}_{2.5}$ in many cities has exceeded the limit of National Ambient Air Quality Standards (NAAQS) in China (Zhang *et al.*, 2011; Xu *et al.*, 2014; He *et al.*, 2017). Water-soluble inorganic ions (WSIs) were the primary chemical constituent of PM, accounting for approximately 23%–82% of the mass of the fine particles concentration in Beijing, in which SO_4^{2-} , NO_3^- , and NH_4^+ contributed more than 30% of the fine particles (Particles diameter of 2.1 microns or less was presented as the fine particles) (Liu *et al.*, 2017). WSIs not only affect the atmospheric precipitation and particle pH and increase the frequency of acid rain, but also exacerbate impairment on visibility and human health (Osunsanya *et al.*, 2001). For example, sulfate is the main

species that affects light scattering and visibility and the average contribution to the light scattering coefficient is 40% for sulfate and 16% for nitrate (Tao *et al.*, 2009). The smaller the aerosol particles size, the greater the harm to human health (de Kok *et al.*, 2009) and ultrafine particles easily enter the human body's bronchi to damage the respiratory tract and cause alveolar inflammation, which largely increases the incidence of cardiopulmonary disease and lung cancer (Osunsanya *et al.*, 2001).

Based on the significant hazards of WSIs in the environment and human beings, systematic research about their distribution characteristics and source apportionment is necessary. Studies have shown that the concentration ratio of nitrate and sulfate can indicate the contribution of stationary (e.g., coal) and mobile sources (e.g., motor vehicles) to sulfur and nitrogen in the atmosphere (Huebert *et al.*, 1988). High Cl^- concentrations are often associated with coal combustion (Yao *et al.*, 2003). Many studies have examined the WSIs of atmospheric PM in urban areas in China, e.g., Beijing (Wang *et al.*, 2005; Liu *et al.*, 2017), Guangzhou (Xia *et al.*, 2017), Tianjin (Wang *et al.*, 2016a), Nanjing (Yu *et al.*, 2016).

Shanxi is a heavily industrial region in the northern region

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of China, which partakes in substantial coal consumption for energy production and industrial activities, including electricity, coke, steel, and chemical products. Shanxi ranked third (10%) on the list of cities in China in terms of PM_{2.5} (PM_{2.5} refers to suspended particles with a dynamic diameter of 25 microns or less in ambient air.) concentration peaks (the daily PM_{2.5} maximum concentrations), followed by Hebei (44%) and Beijing (31%) (Zhai *et al.*, 2018). Meng *et al.* (2007) noted that the average daily PM_{2.5} concentration (193 $\mu\text{g m}^{-3}$) in Taiyuan, the provincial capital of Shanxi, exceeded the 24-h US NAAQS (65 $\mu\text{g m}^{-3}$). According to He *et al.* (2017), the average WSIs concentration in Taiyuan is 69 $\mu\text{g m}^{-3}$, thereby accounting for 33% of the PM_{2.5} mass concentration, which is lower than that in Xi'an (39%) (Zhang *et al.*, 2011) but considerably higher than that in Beijing (29%) (Wang *et al.*, 2005) and Chengdu (29%) (Tao *et al.*, 2013). However, few studies have been conducted regarding other cities in Shanxi, with the exception of Taiyuan.

Jinzhong is a city wherein coal combustion occurs; it is located in the northern region of the Taiyuan basin and is connected with Taiyuan in the northwest region. The terrain expanding from northern to southern Taiyuan is known to be similar to a dustpan, thus resulting in limited diffusion channels for air pollution (Meng *et al.*, 2007). The unique geographical location and meteorological conditions (such as wind speed and wind direction) affect the pollution distribution of Jinzhong. Jinzhong is located at the downwind direction and northwest wind prevails in winter. Therefore, it is likely to be polluted by the pollutants from Taiyuan. Information regarding the characterization of WSIs in Jinzhong is considered to be crucial for understanding the formation mechanism and emission sources of air pollutants in this heavily polluted region.

As a vital parameter of PM, particle size significantly alters the nature of the particles and influences the residence time in the atmospheric environment. Particles with small particle size have a longer suspension time in the air and are prone to be adsorbed into the lung, which poses a great hazards to the respiratory system of human. WSIs are the main components of atmospheric aerosols. By studying the size distribution of WSIs, it is helpful to understand the transformation and transport of aerosols, and to explore the formation pathway. Some studies have examined the size distribution of WSIs in the northern cities in China, such as Beijing (Wang *et al.*, 2005; Yang *et al.*, 2015), Jing-Jin-Ji (Li *et al.*, 2013) and Taiyuan (He *et al.*, 2017). However, no relevant studies have investigated the size distribution characteristics of WSIs in Jinzhong.

In this study, PM_{2.5} and size-segregated samples with varying particle sizes, including < 2.5 μm (PM_{2.5}), 2.5–5 μm (PM_{2.5-5}), 5–10 μm (PM₅₋₁₀), and 10–100 μm (PM₁₀₋₁₀₀), were collected from Jinzhong. This study has the following research objectives: (1) to obtain the size distribution characteristics, seasonal variations, and chemical form of WSIs and (2) to identify the sources of WSIs in Jinzhong. The results will be beneficial for an improved understanding about PM pollution and for establishing related pollution control measures in China.

EXPERIMENTAL METHODS

Aerosol Sampling

The sampling instruments were mounted on the roof of an office building on the campus of the Taiyuan University of Technology, Mingxiang, approximately 12 m above the ground (Fig. 1). A total of 109 samples, including 64 graded samples, 41 PM_{2.5} samples, and 4 parallel samples were obtained over the course of four seasons. The sampling was conducted at 24 h intervals for 10 days during each season. The four periods were April 26–May 6 during spring, July 9–18 during summer, October 11–21 during autumn, and January 3 to 13 during winter. Due to the weather conditions, intermittent rain occurred during the sampling period. The collected samples during the rainy days account for 15% of the whole samples in this study (none in winter, 20% in spring, 30% in summer and 10% in autumn during individual season, respectively).

Daily samples were collected on quartz fiber filters (75 and 90 mm) with two medium-flow air samplers (MH1200-A) at a flow rate of 100 L min⁻¹. One of the samplers collected the PM_{2.5} samples, and the other sampler was used to collect size-segregated samples, including PM_{2.5}, PM_{2.5-5}, PM₅₋₁₀, and PM₁₀₋₁₀₀. The samplers were calibrated and checked using the flow calibrator before each sampling. Sampling basic information and statistics of the meteorological data during the sampling time are shown in Table 1. After being balanced for 72 h in a dryer, the filters were weighed before and after sampling by using an electronic balance with a reading precision of 0.1 mg. Field blank filters were also collected in the sampler during the sampling period, without drawing air. The filters were placed in a filter cartridge after the sampling and were stored at –20°C before the analysis to prevent the evaporation of any volatile components. During the sampling period, meteorological data, including ambient temperature, relative humidity (RH), wind direction, and speed, were obtained for the first 24 hours at 9:00 every day from the China Weather website online (<http://www.weather.com.cn/>). The supplementary material of meteorological data is shown as Table S1. In this study, PM_{2.5} was considered to be fine particles; particles ranging from 2.5 to 100 μm were presented as coarse particles.

Chemical Analysis

The samples were analyzed for WSIs species. Using 10 mL ultra-pure deionized water in a centrifuge tube for 30 min, 1/8 of the filter was extracted and was then filtered with a 0.45 μm Teflon filter to remove any insoluble species. The extracted solutions were transferred to clean plastic bottles and were then stored in the refrigerator. Blank filters were also analyzed using the same method. Four anions (F⁻, Cl⁻, NO₃⁻, and SO₄²⁻) and five cations (Na⁺, NH₄⁺, K⁺, Mg²⁺, and Ca²⁺) were analyzed using two ion chromatographs (Dionex ICS-90, USA and 861 Advanced Compact IC). The results of the samples for the WSIs were corrected using filter blanks. In order to select more accurate samples for analysis, the samples of four days (sampling for the whole 24 h) per season were chosen for the analysis on the coarse particles.

Fig. 1. Sketch of a combined overview map of the study site and DEM (ASTER DEM V2; Resolution: 30 meters).

Table 1. Sampling basic information and statistics of meteorological data during the sampling period.

Season	Sampling period	Number of size-segregated samples	Number of PM _{2.5} samples	T °C	RH %	Air pressure kPa	PM _{2.5} µg m ⁻³
Winter	3 Jan.–2 Mar.	16	12	7 (–12 to 12)	52 (27–82)	101	105 (25–290)
Spring	26 Apr.–6 May	24	11	16 (4–29)	12 (5–27)	102	41 (18–300)
Summer	9 Jul.–18 Jul.	16	11	28 (19–38)	43 (35–66)	100	37 (19–62)
Autumn	11 Oct.–21 Oct.	16	11	11 (3–19)	88 (75–99)	103	74 (42–120)

Backward Trajectory Analysis

The model called Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT), which was developed by the National Oceanic and Atmospheric Administration, was used to preliminarily distinguish the pollution sources of the atmospheric particles (https://ready.arl.noaa.gov/HYSPLIT_traj.php). The Global Data Assimilation System database was used as meteorological inputs in this model, and the university town, Shanxi (37.75°N, 112.72°E), was selected as the starting point. Considering the strong movement of dust aerosols below a height of 1000 m (Han, 2007), three height levels (100 m, 500 m, 1000 m) were chosen to investigate the primary transport path and potential source areas in Jinzhong in this study. For each sampling season, a 10-day back trajectory was computed at 16:00 h UTC.

RESULTS AND DISCUSSION

Size Distribution of PM and WSIs

PM Distribution

During the sampling process, the TSP concentrations ranged from 180 to 293 $\mu\text{g m}^{-3}$, with a median of 219 $\mu\text{g m}^{-3}$, which was slightly lower than that of Xi'an (265 $\mu\text{g m}^{-3}$) (Shen *et al.*, 2009), whereas it was significantly higher than the first-level value of NAAQS (120 $\mu\text{g m}^{-3}$). Fig. 2 shows the size distribution of PM and WSIs. The findings show that PM_{2.5} was the primary component of the atmospheric particulates, which accounted for 41% of the TSP, followed by PM₁₀₋₁₀₀,

which accounted for 22%. The seasonal variations in PM_{2.5} were winter (57%) > autumn (44%) > spring (39%) > summer (33%), and the highest of PM_{2.5} concentrations in winter may be related to the increased consumption of coal and the decreased temperature and rainfall compared with the summer (Zhou *et al.*, 1992).

WSIs Distribution

The mass concentrations of the total WSIs in the TSP ranged from 15 to 139 $\mu\text{g m}^{-3}$, with a median of 39 $\mu\text{g m}^{-3}$, which accounted for 21% of the TSP, which was lower than that of Xi'an (33%). The order of the annual WSIs concentration was $\text{SO}_4^{2-} > \text{NO}_3^- > \text{NH}_4^+ > \text{Ca}^{2+} > \text{Cl}^- > \text{K}^+ > \text{Na}^+ > \text{Mg}^{2+} > \text{F}^-$. The secondary water-soluble inorganic ions (SNAs), including SO_4^{2-} , NO_3^- , and NH_4^+ , were the most abundantly observed substances, thus accounting for 81% of the total WSIs. Research has shown that WSOC is one of the main water-solution species of TSP and accounts for about 4% in Guiyang (Chen, 2010) and 6% in Nanjing (Wu *et al.*, 2017). Based on this, we can estimate the mass concentration of WSOC to TSP in Jinzhong, ranged from 1 to 8 $\mu\text{g m}^{-3}$.

Fig. 2 shows that the proportion of ions that are distributed in PM_{2.5} was the highest, thus accounting for 75% of the total ions in the TSP. The contribution of Cl^- , NO_3^- , SO_4^{2-} , Na^+ , NH_4^+ , and K^+ in PM_{2.5} is considerably higher than that of the coarse particles. Kumar *et al.* (2017) examined the content of WSIs in different sized particles in New Delhi and concluded

Fig. 2. Size distribution of WSIs and particles.

Fig. 2. (continued).

that when the particle size decreases, the contribution of WSIs in the particles increase. Therefore, the results of this study are consistent with those of Kumar *et al.* (2017).

Compared with other ions, the contribution of Ca^{2+} , Mg^{2+} , and F^- in the coarse particles increased, thereby accounting for 76%, 68%, and 69% of the total WSIs, respectively. Li *et al.* examined the size distribution of WSIs in Huangshan and their findings also showed that the concentration of Ca^{2+} increased along with the increase of particle size in PM_{100} (Li *et al.*, 2014). Re-suspended road dust, soil dust, and construction dust are the primary sources of Mg^{2+} and Ca^{2+}

(Liu *et al.*, 2017). The increased content of Ca^{2+} and Mg^{2+} in the coarse particles obtained in this study may be related to the road and construction dust around the sampling point. The high F^- concentration in the coarse particles may be attributed to the soil dust and decomposition of living organisms (Xiu *et al.*, 2004).

Concentrations and Seasonal Variations in $\text{PM}_{2.5}$ and WSIs $\text{PM}_{2.5}$

Fig. 3 shows the daily $\text{PM}_{2.5}$ concentrations in Jinzhong during the sampling period. The $\text{PM}_{2.5}$ concentrations ranged

from 45 to 326 $\mu\text{g m}^{-3}$, with a median of 133 $\mu\text{g m}^{-3}$, thereby exceeding the 24-h $\text{PM}_{2.5}$ concentration limitation value of 35 $\mu\text{g m}^{-3}$ in the United States Environmental Protection Agency (US EPA) and 75 $\mu\text{g m}^{-3}$ in Chinese NAAQS. The $\text{PM}_{2.5}$ concentration obtained in this study was higher than that of many domestic cities, such as Tianjin (69 $\mu\text{g m}^{-3}$) (Xing *et al.*, 2017), Lanzhou (77 $\mu\text{g m}^{-3}$) (Tan *et al.*, 2017), Beijing (107 $\mu\text{g m}^{-3}$) (Xu *et al.*, 2017b) and some foreign countries, such as Kathmandu (Nepal) (76 $\mu\text{g m}^{-3}$) (Shakya *et al.*, 2017), Verneuil (Central France) (10 $\mu\text{g m}^{-3}$) (He *et al.*, 2018), and Veneto Region (Italy) (24 $\mu\text{g m}^{-3}$) (Khan *et al.*, 2016). These findings may be related to the increased coal combustion in the Shanxi province. Moreover, the sampling point was located in the Taiyuan basin, thereby hindering the diffusion of contaminants.

The $\text{PM}_{2.5}$ concentration was higher in winter (159 $\mu\text{g m}^{-3}$) and spring (187 $\mu\text{g m}^{-3}$) and was lower in autumn (119 $\mu\text{g m}^{-3}$) and summer (91 $\mu\text{g m}^{-3}$). During winter, coal combustion increases because it is used for heating; moreover, the relatively low temperature and wind speed and relatively stable atmospheric conditions in autumn and winter are not

suitable for the diffusion of pollutants. By contrast, frequent rainfall was observed to be favorable for the removal of $\text{PM}_{2.5}$ in summer and autumn. A surprising finding is that the $\text{PM}_{2.5}$ concentration in spring was slightly higher than in winter. Zhao *et al.* (2016) noted that cities in the desert regions of northwestern and west-central China experienced increased pollution in spring compared with winter because of dust storms. During the sampling period from May 3 to May 6, 2017, strong sandstorms occurred in most northern regions in China; specific information regarding the sandstorm can be obtained through the Weather China website (<http://www.weather.com.cn/zt/tqzt/2699655.shtml>). Therefore, the higher $\text{PM}_{2.5}$ concentration in spring observed in this study may be related to the severe dust storm in spring of 2017.

Fig. 4 shows that the backward trajectory during the collection period arrived at an altitude of 100 m, 500 m and 1000 m (above ground level) during different seasons. Except for the fall, the air mass trajectories are basically consistent in different heights. Fig. 4(a) reproduce well the typical wind direction during winter in Jinzhong. In summer, the air mass came from southern and eastern cities to the

Fig. 3. Distribution characteristics of $\text{PM}_{2.5}$ and WSIs.

Fig. 4. Seasonal backward trajectory at an altitude of 100 m, 500 m and 1000 m (above ground level) during the sampling period of 2017.

sampling site. Fig. 4(b) shows that the air mass in the study area during spring primarily originated from Mongolia and Inner Mongolia, which further demonstrates that the increased $PM_{2.5}$ concentrations during spring is primarily related to long-distance transportation in Mongolia and Inner Mongolia. Koracin *et al.* (2011) showed that HYSPLIT backward trajectories have a tendency to underestimate sources that are close to the receptor site and to overestimate the effects of the more distant sources. Based on this limitation, it may lead to an overestimation the effects of long-distance transport

from Mongolian and Inner Mongolia and an underestimation of the impact of local anthropogenic pollution sources for $PM_{2.5}$ in Jinzhong in this study. In autumn, air masses under 500 m in height mainly originate from eastern areas. However, air masses in the height of 1000 m primarily occur in the northwest (Fig. 4(d)). This was mainly caused by the geographical location of Jinzhong (west, north and east surrounded by mountains) and the dominant wind direction in autumn (westerly and easterly). Thus, the migration of air masses below the height of 500 m was blocked by mountains,

then entered Jinzhong from the east to the southwest. The air masses of 1000 m height passed through the northwestern region and crossed the mountain into Jinzhong.

WSIs

Fig. 3 shows the mass WSIs concentrations in $PM_{2.5}$. The concentrations of total ions ranged from 13 to $117 \mu\text{g m}^{-3}$, with a median of $37 \mu\text{g m}^{-3}$, which accounted for 31% of the $PM_{2.5}$ concentrations. The ratio was basically consistent with those observed in studies conducted in Lanzhou (31%) (Tan *et al.*, 2017); the ratio was lower than that in Handan (45%) (Meng *et al.*, 2016) and Taiyuan (33%) (He *et al.*, 2017) and higher than Kunming (26%) (Shi *et al.*, 2016). SNAs were the primary components of WSIs, which accounted for 79% of the total ions and 25% of the $PM_{2.5}$ concentrations. SO_4^{2-} and NH_4^+ were the highest in the anions and cations and accounted for 37% and 18% of the total WSIs, which align with the observations derived by He *et al.* in Taiyuan (He *et al.*, 2017).

The WSIs concentration was higher in autumn ($78 \mu\text{g m}^{-3}$) and winter ($44 \mu\text{g m}^{-3}$) and was lower in summer ($30 \mu\text{g m}^{-3}$) and spring ($21 \mu\text{g m}^{-3}$), which accounted for 55% (autumn), 27% (winter), 40% (summer), and 10% (spring) of $PM_{2.5}$ concentrations, respectively (Table 2). Many studies have shown that the concentration of ions was the lowest in summer and highest in winter (Zhang *et al.*, 2015; Deng *et al.*, 2016; Xu *et al.*, 2017a). However, SO_4^{2-} , NO_3^- , NH_4^+ , and K^+ increased rapidly during autumn and even exceeded the content reported during winter in the study period. SO_4^{2-} , NO_3^- , and NH_4^+ were the main SNAs, whereas K^+ is the indicator of biomass burning in the fine particles. SO_2 is primarily produced through fossil fuels combustion and can be oxidized into SO_4^{2-} on the fine particles by heterogeneous or homogeneous reactions (Cheng *et al.*, 2000). The fine mode NO_3^- was primarily attributed to the photo-oxidation reaction of N_2O_5 that was derived through fossil fuel combustion and was generated by the gas-to-particles conversion (Li *et al.*, 2014). Particulate ammonium is formed by the gas-phase reactions of acid precursors (H_2SO_4 , HNO_3 , and HCl , etc.) with ammonia vapor (NH_3) (Ho *et al.*, 2003; Zhang *et al.*, 2011). The formation process of these reaction products is affected by the air temperature and humidity (Zhang *et al.*, 2011). Ammonium nitrate can be

preferentially formed in areas with rich ammonia (NH_3) and HNO_3 in lower ambient temperatures (Li *et al.*, 2014). The atmospheric oxidation rates are considerably affected by physical parameters, such as high temperature, high solar radiation, and high levels of atmospheric oxidant (O_3 , O_2 , H_2O_2 , OH radical, etc.). Zong *et al.* (2016) demonstrated that the atmosphere oxidation processes during autumn are stronger than those observed during the other three seasons, which can serve to accelerate the related secondary aerosol formation processes). During autumn, the temperature and solar radiation are higher than those during winter and compared with that of summer, rainfall is decreased (Fig. 3), whereas the pollution precursor increases. Besides, the local stagnant weather, higher relative humidity and lower wind speed, can be conducive to the accumulation of pollutants in autumn (Fig. 3). Combined with the backward trajectory during autumn (Fig. 4(d)), the possibility of long-distance transport was excluded. Therefore, the highest concentration during fall may be related to pollution in local areas, such as secondary aerosols and biomass combustion sources (Contini *et al.*, 2010).

Chemical forms of WSIs

Considering the chemical valence and concentrations of WSIs, Cl^- , NO_3^- , SO_4^{2-} , Na^+ , NH_4^+ , K^+ , Mg^{2+} , and Ca^{2+} were used to calculate their chemical forms. The correlation coefficients of WSIs in fine and coarse particles are shown in Table 3.

In fine particles, strong correlations were noted among SO_4^{2-} , NO_3^- , Cl^- , and NH_4^+ during winter and among SO_4^{2-} , NO_3^- , and K^+ during autumn, thus indicating that NH_4NO_3 ($R^2 = 0.71$), $(NH_4)_2SO_4$ ($R^2 = 0.97$), and NH_4Cl ($R^2 = 0.81$) were the main compounds found during winter, and KNO_3 ($R^2 = 0.69$) and K_2SO_4 ($R^2 = 0.69$) were noted during autumn. The linear fits of the data were described as $[NH_4^+] = 1.55 [SO_4^{2-}] + 0.16$, $[NH_4^+] = 0.95[SO_4^{2-} + NO_3^-] + 0.09$, $[NH_4^+] = 0.87 [SO_4^{2-} + NO_3^- + Cl^-] - 0.06$ (Fig. S1), which imply that $(NH_4)_2SO_4$ is the main chemical form. The equivalent ratio of NH_4^+ and SO_4^{2-} in the compound of NH_4HSO_4 and $(NH_4)_2SO_4$ were 0.5 and 1, respectively. The slope of the regression equation between NH_4^+ and $[SO_4^{2-} + NO_3^-]$ was 0.95; however, the ratio was reduced to 0.87 when Cl^- was included, thus indicating that the NH_3 concentration

Table 2. Mass concentrations of water-soluble ions in $PM_{2.5}$ in different seasons ($\mu\text{g m}^{-3}$).

Component	Winter	Spring	Summer	Autumn	Annual
F^-	0.21 ± 0.07	0.18 ± 0.08	0.14 ± 0.09	0.07 ± 0.02	0.15 ± 0.09
Cl^-	5 ± 2	0.96 ± 0.68	0.16 ± 0.09	2 ± 1	2 ± 2
NO_3^-	15 ± 11	5 ± 3	3 ± 2	19 ± 10	11 ± 10
SO_4^{2-}	19 ± 12	8 ± 5	17 ± 6	25 ± 6	17 ± 10
Na^+	0.84 ± 0.24	0.55 ± 1.1	0.11 ± 0.13	0.27 ± 0.16	0.45 ± 0.61
NH_4^+	9 ± 5	2 ± 2	5 ± 2.5	16 ± 6	8 ± 7
K^+	0.64 ± 0.22	0.27 ± 0.3	0.31 ± 0.21	0.69 ± 0.15	0.48 ± 0.29
Mg^{2+}	0.2 ± 0.07	0.25 ± 0.16	0.19 ± 0.06	0.16 ± 0.03	0.2 ± 0.09
Ca^{2+}	4 ± 2	6 ± 3	5 ± 2	3 ± 1	4 ± 2
Sum (Ions)	44	21	30	78	37
Percent ¹⁾	27%	10%	40%	55%	31%

Sum of ions divided by concentration of $PM_{2.5}$.

Table 3. Correlation coefficients between selected ions.

		Fine particles					Coarse particles				
		NH ₄ ⁺	K ⁺	Mg ²⁺	Ca ²⁺	Na ⁺	NH ₄ ⁺	K ⁺	Mg ²⁺	Ca ²⁺	Na ⁺
Winter	Cl [−]	0.81	0.49	−0.10	0.00	−0.01	−0.12	0.58	0.33	0.43	0.77
	NO ₃ [−]	0.71	0.30	0.19	0.03	−0.08	0.91	0.61	0.27	0.66	−0.42
	SO ₄ ^{2−}	0.97	0.47	0.00	−0.11	0.03	0.79	0.60	0.93	0.96	−0.24
Spring	Cl [−]	−0.02	−0.12	−0.05	−0.07	−0.05	0.11	−0.41	0.53	−0.49	0.88
	NO ₃ [−]	0.27	−0.11	−0.06	0.01	0.02	−0.26	0.92	−0.27	−0.25	−0.27
	SO ₄ ^{2−}	−0.07	−0.11	0.02	0.33	0.53	0.28	−0.50	−0.34	−0.15	0.05
Summer	Cl [−]	−0.12	0.03	−0.01	0.10	0.32	−0.31	−0.23	0.62	−0.50	---
	NO ₃ [−]	−0.13	−0.12	0.35	−0.08	−0.12	0.27	−0.14	0.81	−0.37	---
	SO ₄ ^{2−}	0.28	0.03	0.13	0.17	−0.13	−0.09	−0.06	0.83	−0.47	---
Autumn	Cl [−]	−0.13	−0.11	−0.11	0.40	−0.01	−0.21	−0.01	0.15	−0.50	0.01
	NO ₃ [−]	0.57	0.69	−0.08	−0.07	−0.06	0.78	−0.20	−0.40	−0.43	0.75
	SO ₄ ^{2−}	0.38	0.69	−0.07	−0.00	−0.04	0.14	−0.27	−0.45	0.16	0.01

was inadequate for fully neutralizing the SO₄^{2−} and NO₃[−], and the excess of SO₄^{2−} and NO₃[−] may be combined with K⁺ in the form of KNO₃ and K₂SO₄ because moderate correlations were noted among these ions (Table 3). Moreover, the slope of [NH₄⁺]/[SO₄^{2−} + NO₃[−] + Cl[−]] was 0.87, thus indicating that part of the NH₄⁺ can also be associated with NO₃[−] and Cl[−] in the form of NH₄NO₃ and NH₄Cl. Wang *et al.* (2015) demonstrated that NH₄⁺ is primarily derived from NH₃ in the air, through gas-phase and aqueous-phase reactions with acidic species (H₂SO₄, HNO₃, and HCl, etc.) to form (NH₄)₂SO₄, NH₄NO₃, and NH₄Cl, which are primarily present in the fine particles. (NH₄)₂SO₄ is a preferentially formed ion and is the most stable, followed by NH₄NO₃; NH₄Cl is the most volatile ion and was formed last (Zhang *et al.*, 2008). These findings were consistent with the chemical composition of WSIs that was observed in Beijing (Wang *et al.*, 2005).

Compared with the correlations in the fine particles, strong correlations were noted between NO₃[−] and NH₄⁺, SO₄^{2−} and Mg²⁺ and between SO₄^{2−} and Ca²⁺ during winter and between K⁺ and NO₃[−] during spring, thereby suggesting that NH₄NO₃ (R² = 0.91), MgSO₄ (R² = 0.93), CaSO₄ (R² = 0.96), and KNO₃ (R² = 0.92) primarily existed in the coarse particles. The linear regression equation is described as [NH₄⁺] = 0.38 [SO₄^{2−} + NO₃[−]] + 0.03, [NH₄⁺ + Mg²⁺ + Ca²⁺] = 1.04 [SO₄^{2−} + NO₃[−]] + 0.13 (Fig. S2), thereby suggesting that NH₃ was insufficient for completely neutralizing the SO₄^{2−} and NO₃[−]. The excess of SO₄^{2−} and NO₃[−] may be associated with Mg²⁺ and Ca²⁺, which occurred as a form of MgSO₄ and CaSO₄, and part of NO₃[−] can also be associated with K⁺ to form KNO₃ because higher correlations exist on these ions. The sulfuric acid and nitric acid on mineral dust could be through the heterogeneous reaction to form the coarse sulfate and nitrate (Liu *et al.*, 2017).

Acidity-alkalinity Analysis of Particles

The acidity-alkalinity analysis of the fine and coarse particles was evaluated using the R_{C/A} (cation and anion equivalence ratio) in this study (Tian *et al.*, 2017). The mass concentrations (μg m^{−3}) of the anions and cations were converted to mole concentrations (μmol m^{−3}), and the calculation method is depicted in Eq. (1). The time series of R_{C/A} during the sampling period are shown in Fig. 5.

$$R_{C/A} = \frac{Na^+ / 23 + NH_4^+ / 18 + K^+ / 39 + Mg^{2+} / 12 + Ca^{2+} / 20}{F^- / 19 + Cl^- / 35.5 + NO_3^- / 62 + SO_4^{2-} / 48} \quad (1)$$

During the sampling period, the median value of R_{C/A} in coarse particles was 1.93, and the R_{C/A} values of more than 94% samples were greater than 1.00. The median value of R_{C/A} in PM_{2.5} concentrations was 1.17, and the R_{C/A} values of more than 83% samples were greater than 1. These values indicated that the fine and coarse particles in Jinzhong were alkaline or neutralized if other anions (e.g., NO₂[−], CO₃^{2−}, HCO₃[−]) and organic acids were included, and their acidity was negligible, which is consistent with the findings of Shen *et al.* (2009).

During spring, R_{C/A} reached 8.45, and the alkalinity was the strongest. It was primarily connected with the sandstorm weather that occurred in spring. Dust is the primary source of Mg²⁺ and Ca²⁺, which indicates that alkaline ions were significantly increasing. During winter and autumn, the R_{C/A} value for PM_{2.5} remained relatively stable, with a median of 1.05 and 1.12, exceeding 1, which indicates that the particles are approximately similar to the weakly alkaline. This is primarily due to the low alkaline ion (e.g., Mg²⁺, Ca²⁺) caused by the freezing of the soil and the high acid ion Cl[−] content caused by coal combustion (Liu *et al.*, 2017). The median R_{C/A} value during spring reached the maximum, and the alkalinity was stronger than those of other seasons, which was primarily contributed by the significant increase in alkaline ions from sandstorms. During summer, 90% of the PM_{2.5} samples had an R_{C/A} of more than 1.00, with a median of 1.39, which can be attributed to the higher alkaline ion concentration and indicate that particles were alkaline because of the higher concentration of NH₃ and mineral dust emissions (Liu *et al.*, 2017).

Ratio Analysis

Fig. 6 shows the ratio distribution of WSIs in fine and coarse particles. Coal combustion during winter releases increased pollutants and causes the increase of coal-related ions, such as Cl[−], thus resulting in the highest ratio of Cl[−]/K⁺ found during winter in both fine and coarse particles. Many

Fig. 5. $R_{C/A}$ values in fine and coarse particles during different seasons.

Fig. 6. Time series of $\text{NO}_3^-/\text{SO}_4^{2-}$, $\text{Mg}^{2+}/\text{Ca}^{2+}$, $\text{SO}_4^{2-}/\text{K}^+$, and Cl^-/K^+ ratios in (a) fine particles and (b) coarse particles.

studies have suggested that K^+ in fine particles can often be used as an indicator of biomass burning (Fourtziou *et al.*, 2016; Rajput *et al.*, 2017). Unlike the seasonal characteristics of the Cl^-/K^+ ratio, there was no obvious distinction for Mg^{2+}/Ca^{2+} among the four seasons, and the median ratio in coarse particles was higher than that of fine particles.

The NO_3^-/SO_4^{2-} mass ratio has been used as an indicator to evaluate the contribution to sulfate and nitrate from mobile versus stationary pollution sources (Xu *et al.*, 2012; Wang *et al.*, 2015b). The estimated ratios of NO_x to SO_x from vehicle exhausts are 8:1–13:1 compared with the estimated ratio of NO_x to SO_x , which is 1:2, from coal combustion (Shen *et al.*, 2008). The annual median of the NO_3^-/SO_4^{2-} ratio in fine particles obtained in this study was 0.63, which was lower than that of Beijing (1.12) (Wang *et al.*, 2016b) and Hefei (1.10) (Deng *et al.*, 2016), thus indicating that the contribution of stationary sources (e.g., coal) to fine particles in Jinzhong was increased in comparison to that of mobile sources (e.g., motor vehicles).

The seasonal median of the NO_3^-/SO_4^{2-} ratio followed the order of autumn (0.82) > spring (0.75) > winter (0.73) > summer (0.17) in fine particles, which is consistent with the findings of Wang *et al.* (2005). Significantly increased NO_3^-/SO_4^{2-} ratios were observed during autumn, winter, and spring, compared with that during summer. High temperature could cause the decomposition of NH_4NO_3 into gaseous HNO_3 and NH_3 , thereby reducing the NO_3^- concentration in fine particles (Li *et al.*, 2014). Different from fine particles, the NO_3^-/SO_4^{2-} ratio in coarse particles followed the order of summer (11.56) > spring (4.37) > autumn (1.17) > winter (0.85), which is consistent with the findings of Shen *et al.* (2008). The increased concentrations of the coarse-mode NO_3^- may be explained by the heterogeneous reaction of the gas precursor on coarse particles that contain increased calcium and magnesium during summer (Li *et al.*, 2014), thereby causing the increased NO_3^-/SO_4^{2-} ratio in the coarse particles.

Source Analysis of WSIs in Fine and Coarse Particles

A principal component analysis (PCA) was conducted to

further apportion the potential sources of WSIs in fine and coarse particles in this study. All the principal factors were extracted with the initial eigenvalue > 1.0. The Kaiser-Meyer-Olkin values in fine and coarse particles were above 0.6, and the PCA result for WSIs is shown in Tables 4 and 5.

Table 4 shows that the WSIs in $PM_{2.5}$ aerosol samples comprises three PCA factors, thus accounting for approximately 80% of the total variance in the concentration data. Factor 1 accounts for 44% of the total variance and is strongly loaded with NH_4^+ (0.92), SO_4^{2-} (0.89), NO_3^- (0.88), and K^+ (0.75), thereby indicating that it potentially originated from the secondary source aerosol and anthropogenic sources, particularly coal combustion and biomass burning (Wang *et al.*, 2015a). The formation of SO_4^{2-} and NO_3^- in the fine particles were primarily associated with the secondary pollutants transformed from SO_2 and NO_3 by using photochemical oxidation (Zhao *et al.*, 2011). Factor 2, which accounts for 20% of the variance, is primarily affected by Mg^{2+} and Ca^{2+} , with an obvious loading of 0.85 and 0.66, respectively. Mg^{2+} and Ca^{2+} were considered as traces of dust particles from construction sites and roads (Zhang *et al.*, 2011; Meng *et al.*, 2016). Factor 3 is loaded with Na^+ (0.69) and F^- (0.65), and it accounts for 16% of the total variance, thereby suggesting that the potential source is industrial emissions (Wang *et al.*, 2015a).

Table 5 presents three PCA factors of WSIs in coarse particles, thereby accounting for approximately 79% of the total variance, and Al as a tracer for soil dust particles was also included as input data. Factor 1, which accounts for 31% of the variance, is heavily loaded by K^+ (0.91) and Na^+ (0.82), which can indicate the potential source from coal combustion and biomass burning. Factor 2 is loaded with Ca^{2+} (0.76), Mg^{2+} (0.64), and Al (0.63) and accounts for 26% of the total variance, thereby suggesting that the dust originates from soil particles and falling dust. Factor 3 accounts for 22% of the total variance and is dominated by SO_4^{2-} (0.92), NH_4^+ (0.65), and NO_3^- (0.52), thereby indicating that it potentially originates from a secondary reaction. The PCA source analysis of components on the coarse particles was consistent with the findings of Wang *et al.* (2015a). The coarse sulfate

Table 4. Result of principal component analysis for water-soluble ions in the fine particles.

Ion	PCA source		
	PC1	PC2	PC3
F^-	−0.33	0.45	0.65
Cl^-	0.59	0.37	0.57
NO_3^-	0.88	0.20	−0.06
SO_4^{2-}	0.89	−0.03	0.15
Na^+	0.26	−0.09	0.69
NH_4^+	0.92	0.07	−0.23
K^+	0.75	0.49	−0.14
Ca^{2+}	−0.26	0.85	−0.24
Mg^{2+}	−0.62	0.66	−0.22
Variance	44	20	16
Cumulative	44	64	80
Source	Secondary source Coal combustion Biomass burning	Dust source	Industrial emission

Table 5. Result of principal component analysis for water-soluble ions in the coarse particles.

Ion	PCA source		
	PC1	PC2	PC3
F ⁻	-0.58	-0.30	-0.45
Cl ⁻	0.53	-0.78	-0.07
NO ₃ ⁻	-0.28	-0.07	0.52
SO ₄ ²⁻	-0.28	-0.14	0.92
Na ⁺	0.82	-0.43	-0.08
NH ₄ ⁺	0.51	0.51	0.65
K ⁺	0.91	0.15	-0.23
Ca ²⁺	0.07	0.76	0.18
Mg ²⁺	-0.45	0.64	-0.53
Al	0.59	0.63	-0.24
Variance	31	26	22
Cumulative	31	57	79
Source	Biomass burning Coal combustion	Dust source	Secondary reaction

and nitrate were primarily attributed to the reaction with crustal species in the coarse particles or through the heterogeneous reaction of precursor gases with crustal particles (Anlauf *et al.*, 2006).

CONCLUSION

The PM_{2.5} concentrations ranged from 45 to 326 $\mu\text{g m}^{-3}$, with a median of 133 $\mu\text{g m}^{-3}$, which significantly exceeded the daily concentration limitation value of 35 $\mu\text{g m}^{-3}$ that was issued by the US EPA and 75 $\mu\text{g m}^{-3}$ that was issued by Chinese NAAQS and MEP (Ministry of Environmental Protection of the People's Republic of China). The proportion of ions distributed in fine particles was the highest, thereby accounting for 75% of the total ions in TSP. The seasonal variations of WSIs followed the order of spring < summer < winter < autumn, thereby indicating that the different characteristics of the four seasons was a crucial factor that affected the WSIs distributions. The ions (NH₄⁺, K⁺, NO₃⁻, and SO₄²⁻) primarily existed in the form of NH₄NO₃, (NH₄)₂SO₄ and K₂SO₄ in fine particles, and Ca²⁺ and Mg²⁺ were primarily concentrated in the form of MgSO₄ and CaSO₄ in coarse particles, thereby suggesting that the fine and coarse particles in Jinzhong were alkaline during the study period. The result of the ratio analysis showed that the highest ratio of Cl⁻/K⁺ was observed during winter in both fine and coarse particles; however, there was no obvious distinction for Mg²⁺/Ca²⁺ among the four seasons. The NO₃⁻/SO₄²⁻ ratio in coarse particles was significantly higher than that in fine particles, particularly in summer, thus indicating that the heterogeneous reaction on particles plays a vital role in the formation of NO₃⁻ in coarse particles. The result of the PCA showed that the source of WSIs was in Jinzhong secondary source aerosol, dust particles, coal combustion, biomass burning and industrial emission. The coal combustion and biomass burning have been considered as the leading emission sources to be controlled for improving air quality. The government can promote to use clean energy and accelerate the achievement of a high economic value of biomass, which will significantly improve air quality in Jinzhong.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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