Chemical Characteristics and Source Apportionment of PM_{2.5} during Winter in the Southern Part of Urumqi, China

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ABSTRACT

Urumqi, the administrative center of Xinjiang, suffers from severe atmospheric aerosol pollution; however, no study has comprehensively analyzed the local constituents and sources of fine particulate matter (PM_{2.5}). The characteristics of PM_{2.5} in Urumqi were observed the first winter (2012–2013) after natural gas replaced coal as an energy source. Enrichment factors, backward trajectories, the potential source contribution function (PSCF) model, and positive matrix factorization (PMF) were used to identify the source area and categories. The results showed a mean concentration of 197.40 $\mu\text{g m}^{-3}$ for the PM_{2.5}, which significantly decreased after the conversion from coal to natural gas. Although the concentration of NO_3^- increased post-conversion, the SO_4^{2-} and Cl^- decreased by 42.54% and 32.93%, respectively. The water-soluble ions (WSIs) mainly consisted of NH_4HSO_4 , CaSO_4 , MgSO_4 , $\text{Ca}(\text{NO}_3)_2$, $\text{Mg}(\text{NO}_3)_2$, and KCl . Elements such as Pb, Cr, and As decreased following the fuel switch. The organic carbon and elemental carbon were strongly correlated, and the mean concentration of the secondary organic carbon was 18.90 $\mu\text{g m}^{-3}$. Pyr, Chr, BbF, BkF, IcdP, and BghiP were the most prevalent individual polycyclic aromatic hydrocarbons, and BaP exceeded health-based guidelines. The results from trajectory clustering and PSCF modeling suggested that emissions from both the city and its surroundings, as well as the valley-and-basin topography, may be responsible for the heavy PM_{2.5} pollution in southern Urumqi. PMF identified five primary sources: secondary formation, biomass and waste burning, vehicle emissions, crustal minerals, and industrial pollution and coal combustion.

Keyword: Fine particulate matter; Chemical composition; Source apportionment; Urumqi.

INTRODUCTION

Air pollution in China is increasingly severe because of rapid economic growth, urbanization, and industrialization. This problem seriously threatens human health and the environment. Atmospheric particulate matter (PM) originates from human and natural sources and also forms after the conversion of precursor gases into secondary particles through photochemical reactions (Chen *et al.*, 2017a, b). PM varies in shape, size, surface area, solubility, and chemical

composition depending on the source and subsequent atmospheric chemical reactions; therefore, its characteristics differ by region (Hassan and Khoder, 2017). Numerous studies have demonstrated that PM can damage human health directly as well as indirectly through influence on climate and food security. Recently, atmospheric PM_{2.5} (PM with an aerodynamic diameter of $< 2.5 \mu\text{m}$) has attracted worldwide attention because its small size can penetrate the human lungs (Jiang *et al.*, 2019). The sum of major components, including trace elements, water-soluble ions (WSIs), organic carbon (OC), elemental carbon (EC), and polycyclic aromatic hydrocarbons (PAHs), accounts for more than 60% of PM_{2.5} (Lonati *et al.*, 2005).

Aust *et al.* (2002) reported that transition metals are a major toxic component of the toxicity of PM. EC is emitted from a variety of combustion processes and directly

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influences climate and causes atmospheric warming (Ramanathan and Carmichael, 2008). Consistent evidence demonstrates a causal relationship between PAHs and cancer risk, and the International Agency for Research on Cancer has classified benzo[*a*]pyrene (BaP) as a Group 1 carcinogen (Wei *et al.*, 2010). Sun *et al.* (2014) found that WSIs promote haze formation during haze episodes.

PM_{2.5} levels in many Chinese cities frequently exceed the National Ambient Air Quality Standards (NAAQS), especially in winter (Chen *et al.*, 2017a, b). Xinjiang is a developing province in China with the administrative center at Urumqi, which is undergoing rapid economic development, urbanization, population growth, and a critical increase in traffic. In recent years, Urumqi has experienced severe air pollution. In the 2011 World Health Organization (WHO) air quality rankings of 1083 cities, Urumqi ranked 1053, the third worst in China (Li *et al.*, 2016a).

The rapid deterioration of the atmosphere in Urumqi is related to a variety of factors. Most influential are the excessive amount of anthropogenic pollutants and typical topography (is surrounded on three sides by Tianshan Mountains). Under the national energy relocation program and to promote the regional economy (energy industries in China, including oil, natural gas and coal mining industries, petrochemical industries, coal chemical industries, and thermal power generators, have been relocated toward energy-abundant northwestern China), Xinjiang energy industries have been booming in the past decades (Dorian *et al.*, 1999). With the rapid economic development and urbanization, the sources of air pollution in Urumqi have gradually shifted from conventional dust storms to a mixture of dust storms, coal-burning emissions, vehicle emissions, and industrial emissions. The urban area of Urumqi is located on the alluvial plain of the northern foothills of the central Tianshan Mountains (only one mouth facing north exists). The topography of Urumqi causes unique meteorological conditions characterized by a stable atmosphere and calm winds, which are extremely unfavorable to the transmission and diffusion of air pollutants. Moreover, the 6-month heating period (October–March) enhances precursor gas accumulation. Furthermore, being subject to the high pressure over Mongolia in winter, Urumqi frequently experiences foehns, and interactions between foehns and valley winds cause severe air pollution (Li *et al.*, 2015). Daily concentrations of PM_{2.5} reached 263.77 $\mu\text{g m}^{-3}$ in the winter of 2010–2011 in Urumqi, far exceeding the National Ambient Air Quality Standard (NAAQS-II, GB3095) of 35 $\mu\text{g m}^{-3}$ (Limu *et al.*, 2013). In 2012, to rapidly reduce the air pollution caused by coal-fired heating, the municipal government implemented a heating energy structure adjustment strategy that uses natural gas instead of raw coal as fuel. However, Urumqi is still suffering from heavy air pollution and haze during the wintertime. Thus, air pollution has been extensively studied in Urumqi (Li *et al.*, 2008a; Mamtimin and Meixner, 2011; Song *et al.*, 2015; Li *et al.*, 2016b; Ren *et al.*, 2017; Wang *et al.*, 2017). These studies demonstrated high concentrations of particulate matter, SO₂, and NO_x in this city, making it one of the most polluted cities in the world. And Urumqi is

located in the source area of Asian dust, and the mineral dust mixing with anthropogenic aerosol has a great impact on the regional environment and global climate change. Moreover, the chemical characteristics and formation mechanisms of air pollution are different from other cities in China. Thus, intensive environmental surveys in Urumqi would allow for better assessment of the impact of human activities on the environment in Urumqi as well as other Chinese cities.

Therefore, the present study collected PM_{2.5} data during the winter of 2012–2013 (the first winter after the conversion from coal to gas) in Urumqi. The objectives were (1) to determine the level of PM_{2.5} after the switch to natural gas, (2) to reveal the chemical profile of PM_{2.5} during winter in Urumqi, and (3) to analyze the various combinations of PM_{2.5} chemical components to obtain potential sources. The results provide valuable information for more effective PM_{2.5} control in Urumqi or other cities in northwestern China during winter.

METHODOLOGY

PM_{2.5} Sampling

The sampling site was located on the roof (15 m above ground level) of the building of the Chemistry and Chemical Engineering College at Xinjiang University (43°77'N, 87°61'E) in Tianshan District. The sampling site was under the influence of residential, traffic, and construction emissions and representative of urban Urumqi. Daily PM_{2.5} samples were collected from 10:00 a.m. to the following day at 8:00 a.m. by using a high-volume air sampler (TH-1000; Wuhan Tianhong Instruments Co., Ltd.) at a flow rate of 1.05 m³ min⁻¹. A total of 38 aerosol samples, including 34 daily samples and 4 field blank samples, were collected on 47-mm quartz microfiber (QM-A; Whatman, Mainstone, UK) filters from October 16, 2012, to May 3, 2013. The filters were prebaked for 4 h at 480°C to remove organic and inorganic material. Before and after sampling, the quartz filters were equilibrated for 48 h in a desiccator at a constant temperature of 25°C and relative humidity of 45%. Each filter was weighed at least three times. After sampling, the filters were wrapped in aluminum foil and stored at -20°C to prevent evaporation of volatilized components.

Daily meteorological data including wind speed, temperature, relative humidity, and visibility during the sampling period were obtained from a website (<https://www.wunderground.com>).

Chemical Analysis

A punch was extracted from each filter and submerged in HNO₃ (10 mL, 69%) and HClO₄ (4 mL, 70%) in acid-cleaned glass test tubes. The mixtures were left overnight and heated progressively to 170°C in a heating digester for 4 h to near dryness. After the test tubes were cooled, the solutions were filtered into 50-mL volumetric flasks using 1% HNO₃ and then decanted into acid-cleaned polyethylene tubes. Cr, Co, Ni, Cu, Zn, Pb, and Mn metals were analyzed using an atomic absorption spectrometer; Hg and As were detected using a dual-channel atomic fluorescence

photometer. The standard deviations of each element were less than 5%. Field and laboratory blanks were extracted and analyzed in the same manner as the field samples.

A punched-out section of the quartz filter was placed into a centrifuge tube and submerged in 10 mL of ultrapure water (resistivity = 18.25 MΩ cm). To completely dissolve all the WSIs, the tubes were shaken twice through ultrasound at a low temperature (< 10°C) for 20 min each time and subsequently centrifuged at 11,000 rev min⁻¹ for 12 min. The solution was filtered through a polytetrafluoroethylene (PTFE) membrane (Whatman, Middlesex, UK) with a 0.22-μm pore size, transferred to clean plastic bottles, and stored at 4°C in a refrigerator for analysis. Field and laboratory blanks were also analyzed in the same manner as the samples.

Five cations (Ca²⁺, NH₄⁺, Mg²⁺, K⁺, and Na⁺) and four anions (NO₃⁻, SO₄²⁻, Cl⁻, and F⁻) were analyzed through ion chromatography (IC; 883 Basic IC plus; Metrohm AG, Switzerland). The MagIC Net 2.0 workplace was used to calculate the concentration of WSIs. In this study, the detection limits of the IC system were 0.0018, 0.006, 0.007, 0.019, 0.006, 0.013, 0.007, 0.009, and 0.0105 mg L⁻¹ for Ca²⁺, NH₄⁺, Mg²⁺, K⁺, Na⁺, NO₃⁻, SO₄²⁻, Cl⁻, and F⁻, respectively. The recovery of each ion was 90–105%.

A punch of each filter (0.5 cm²) was analyzed for OC and EC using an Optical Carbon Analyzer (Model 2015; Desert Research Institute, Sunset Laboratory Inc., USA). OC fractions were produced in a helium atmosphere, and EC fractions were produced in a 2%-oxygen/8%-helium atmosphere. The detection limits for OC and EC were 0.05 μg.

Analysis of PAHs followed the method described by Gao *et al.* (2012) and Yu *et al.* (2016). Before solvent extraction, a mixture of three isotopes of labeled PAH compounds (phenanthrene-d10, chrysene-d12, and perylene-d12), tetracosane-d50, and levoglucosan-¹³C₆ were added as surrogates prior to extraction. The quartz was ultrasonically extracted twice for 20 min at a low temperature (< 10°C) with a mixed solvent of dichloride methane (DCM)/hexane (1:1, vol/vol) and then extracted twice with a mixed solvent of DCM/methanol (1:1, vol/vol). The extracts were purified by an anhydrous sodium sulfate column to remove potentially interfering compounds. The extracts of each sample were combined. The samples were concentrated to a volume of approximately 2 mL with a rotary evaporator and further concentrated to a volume of 0.5 mL through nitrogen blow-down. Subsequently, 200 μL of DCM, 10 μL of methanol, and 300 μL of freshly made diazomethane were added and maintained at room temperature for 1 h. The extracts were then analyzed for PAHs using a gas chromatography-mass spectrometry system (6890/5973N; Agilent) equipped with a DB-5ms column (50 m, 0.32 mm, 0.17 μm). Concentrations of 17 PAHs were quantified in this study, with the following elution order: naphthalene (Nap), acenaphthylene (Ace), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), pyrene (Pyr), benzo[*a*]anthracene (BaA), chrysene (Chr), benzo[*b*]fluoranthene (BbF), benzo[*k*]fluoranthene (BkF), BaP, indeno[1,2,3-*cd*]pyrene (IcdP), benzo[*g,h,i*]perylene

(BghiP), dibenz[*a,h*]anthracene (DahA), and coronene (Cor).

The field and laboratory blanks were analyzed using the same method, and none of the target compounds were detected. Recoveries for all compounds were higher than 90%. The aforementioned measurement procedures underwent strict quality control and quality assurance to avoid possible contamination of the samples.

Pollution Pathways and source Area

Transport pathways and source regions can influence local air quality (Zhang *et al.*, 2013; Tan *et al.*, 2017). To address this, both trajectory clustering and the potential source contribution function (PSCF) method were used. The 48-h air mass back trajectories were investigated using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model. The meteorological field was obtained from National Oceanic and Atmospheric Administration's Air Resources Laboratory (ARL) archives, and the model was run at a height of 100 m above ground level with a time interval of 1 h. The total trajectories were used for cluster analysis in the geographic information system (GIS)-based software TrajStat.

The PSCF model has been widely used to identify potential source areas of pollutants. In the current study, this model was implemented using TrajStat based on the results of HYSPLIT. The study domain was 20.00–60.00°N and 65.00–100.00°E with a horizontal resolution of 0.5° × 0.5°. The weighting function W_{ij} was adopted to accurately reflect the uncertainty in cells with a small number of endpoints that fall within cell *ij*. In this study, the daily average PM_{2.5} concentration standard (75 μg m⁻³) was treated as the pollution threshold (Zhang *et al.*, 2013; Zhao *et al.*, 2015).

$$W_{ij} = \begin{cases} 1.00, & 80 < n_{ij} \\ 0.70, & 20 < n_{ij} \leq 80 \\ 0.42, & 10 < n_{ij} \leq 20 \\ 0.05, & n_{ij} \leq 10 \end{cases} \quad (1)$$

Source Apportionment Analysis

Source apportionment analysis is vital to developing effective control strategies for PM_{2.5}. Positive matrix factorization (PMF) is an effective mathematical receptor model recommended by the U.S. Environmental Protection Agency (EPA) and has been used worldwide for source apportionment for PM_{2.5}. The principle of PMF is minimizing the objective function Q , which was determined as follows:

$$Q = \sum_{i=1}^n \sum_{j=1}^m \left[\frac{x_{ij} - \sum_{k=1}^p g_{ik} f_{kj}}{u_{ij}} \right] \quad (2)$$

In this function, x_{ij} and u_{ij} are the concentration (*Con*) and uncertainty (*Unc*) of the j^{th} species in the i^{th} sample, f_{kj} is the fraction of the j^{th} species from the k^{th} source, g_{ik}

indicates the contribution of the k^{th} source to the i^{th} sample, p is the number of sources, and m and n are the total number of species and samples, respectively. A detailed description can be found in other studies (Paatero and Tapper, 1994; Hopke, 2016).

This model required *Con* and *Unc* input data. Values below the method detection limit (MDL) were substituted with half the MDL, and missing data were substituted with the median *Con*. *Unc* was calculated by the following equation when the concentration was higher than the MDL:

$$Unc = \sqrt{(Error\ Fraction \times Con)^2 + (0.5 \times MDL)^2} \quad (3)$$

If the concentration was less than or equal to the MDL:

$$Unc = \frac{5}{6} MDL \quad (4)$$

In this study, PMF5.0 was employed for source apportionment. The input data included PM_{2.5}, OC, EC, Ca²⁺, NH₄⁺, Mg²⁺, K⁺, Na⁺, NO₃⁻, SO₄²⁻, Cl⁻, F⁻, Cr, Co, Ni, Cu, Zn, Pb, Mn, and As. PM_{2.5} was set as the total variable. In this section, the extra modeling uncertainty of PMF was 10%. To determine the proper number of source factors, three to eight source factors were separately run with different *Fpeak* values (-2, -1.5, -1, -0.5, 0, 0.5, 1, 1.5, and 2) for optimum results. When the changes in *Q* values become smaller and the *Q* (Robust) and *Q* (True) values are closed, it can suggest that it could be an optimal

solution.

Quality Assurance/Quality Control and Statistical Analyses

All analytical procedures were monitored with strict quality assurance (QA) and quality control (QC) measures. Both pre- and post-sampling and chemical analysis strictly guaranteed that the filter was intact and undamaged, and the cracked filters were excluded. To reduce the uncertainties in chemical and statistical analyses created by sample size, we made large efforts (e.g., combining principal component analysis (PCA) and PMF model for source apportionment) to offset the defect.

RESULTS AND DISCUSSION

PM_{2.5} Mass Concentration

The time series of the meteorological parameters during the sampling period is shown in Fig. 1. Affected by the temperate continental climate, the first sampling period experienced high temperatures, low humidity, high visibility, and sensitive wind speed. However, from November, the temperature, visibility, and wind speed decreased slightly, and the humidity rose. The average temperature, relative humidity, and wind speed were -8.4°C, 70.7%, and 1.5 m s⁻¹.

From October 2012 through March 2013, the mean concentration of PM_{2.5} in Urumqi was 197.40 µg m⁻³; the highest value was recorded on January 8 (481.43 µg m⁻³), and the lowest value was recorded on February 14 (55.78 µg m⁻³). All samples far exceeded NAAQS-II (35 µg m⁻³)

Fig. 1. Temporal variations of PM_{2.5}, major components, and meteorological parameters during winter from 2012 to 2013 in Urumqi.

during the sampling period. The mass concentrations in urban Urumqi were higher than in most coastal cities in China, such as Guangzhou ($89.30 \mu\text{g m}^{-3}$; Tao *et al.*, 2014) and Shanghai ($62.88 \mu\text{g m}^{-3}$; Tao *et al.*, 2017), and other inland cities, such as Lanzhou ($120.5 \mu\text{g m}^{-3}$ in winter), illustrating the severity of $\text{PM}_{2.5}$ pollution in winter in Urumqi. The low temperature and high humidity might favor the growth of aerosol particles and $\text{PM}_{2.5}$ formation, and the low wind speed might be unfavorable to the diffusion and transport of pollutants. Moreover, the typical valley and basin topography of Urumqi, which is surrounded by hills and mountains on three sides, blocks the flow of air and suppresses the transport of pollutants.

However, the concentrations of $\text{PM}_{2.5}$ found in this study were lower than those measured in Urumqi before substituting natural gas for coal, such as the values observed by Limu *et al.* (2013) from September 2010 through March 2011 (mean: $263.77 \mu\text{g m}^{-3}$). The concentration of $\text{PM}_{2.5}$ has obviously decreased since the switch to natural gas, indicating that this project (shifting coal to natural gas) has been successful, which is a crucial finding for the control of air pollution.

Chemical Composition of $\text{PM}_{2.5}$

The components measured in this study (WSIs, metallic elements, OC, EC, and PAHs) accounted for 66.21% of the $\text{PM}_{2.5}$ mass concentration on average (Fig. 1), and a large unidentified mass accounted for 33.79% (on average) in Urumqi in winter. The measured masses explained the majority of the components of $\text{PM}_{2.5}$ in Urumqi.

WSIs

Table 1 lists the main WSIs, including Na^+ , NH_4^+ , K^+ , Mg^{2+} , Ca^{2+} , F^- , Cl^- , NO_3^- , and SO_4^{2-} , and their concentrations in the $\text{PM}_{2.5}$ in Urumqi during the sampling period. On average, the total WSI concentration was $58.08 \mu\text{g m}^{-3}$, accounting for 33.72% of the $\text{PM}_{2.5}$ mass, which is approximately 20% lower than that observed in Urumqi before the switch from coal to gas (Li *et al.*, 2008a). The concentrations of the WSIs in the $\text{PM}_{2.5}$ in Urumqi followed the order of $\text{SO}_4^{2-} > \text{NH}_4^+ > \text{NO}_3^- > \text{Cl}^- > \text{Ca}^{2+} > \text{Na}^+ > \text{K}^+ > \text{F}^- > \text{Mg}^{2+}$, with mean concentrations of 26.66, 8.99, 8.52, 5.09, 4.05, 3.25, 0.81, 0.22, and $0.21 \mu\text{g m}^{-3}$, respectively. SO_4^{2-} , NH_4^+ , and NO_3^- were the three most prevalent WSIs, and the sum of their concentrations was $44.45 \pm 20.64 \mu\text{g m}^{-3}$, or approximately 76.52% of the total WSIs; this finding is consistent with those in Taiyuan (75.21%) (He *et al.*, 2017) and Lanzhou (73%) (Tan *et al.*, 2017) but lower than those in megacities, such as Beijing (88%) (Zhang *et al.*, 2013). It is well known that SO_4^{2-} and

Cl^- originate from coal combustion, and coal was the sole energy source for domestic heating, power, and industries in Urumqi before 2012. However, considerable efforts have been made by the local government to alter the energy structure so as to improve the air quality in Urumqi, such as changing coal to natural gas in 2012 (Wang *et al.*, 2017). Therefore, in this study, the concentrations of SO_4^{2-} and Cl^- decreased by approximately 42.54% and 32.93%, respectively, compared with the concentrations measured in 2007 (before the switch to gas) in Urumqi by Li *et al.* (2008a).

Generally, sulfuric acid and nitric acid react with ammonia to form salts (Hewitt, 2001). NH_3 preferentially reacts with H_2SO_4 to form NH_4HSO_4 when NH_3 is inadequate and transforms into $(\text{NH}_4)_2\text{SO}_4$ when NH_3 is abundant (Hassan and Khoder, 2017; Zhou *et al.*, 2018). Therefore, the aforementioned mechanisms as well as the correlation (0.779) and molar ratio (0.091) of NH_4^+ to SO_4^{2-} (Fig. 2(a)) indicate that NH_4HSO_4 , rather than $(\text{NH}_4)_2\text{SO}_4$, was the dominant chemical species in Urumqi in winter. Fig. 2(b) illustrates that the ratios of NH_4^+ to $\text{SO}_4^{2-} + \text{NO}_3^-$ ranged from 0.01 to 0.96 (all lower than unity), suggesting that some SO_4^{2-} and NO_3^- may be present in chemical forms other than NH_4HSO_4 , $(\text{NH}_4)_2\text{SO}_4$, and NH_4NO_3 . Li and Shao (2008) demonstrated that heterogeneous reactions between NO_x , SO_2 (and its products, such as N_2O_5 , HNO_3 , and H_2SO_4), and dust carbonate often occur in northern China. Accordingly, we examined the correlation between $\text{NH}_4^+ + \text{Ca}^{2+} + \text{Mg}^{2+}$ and $\text{SO}_4^{2-} + \text{NO}_3^-$ (Fig. 2(c)), given that a strong correlation between Ca^{2+} and Mg^{2+} (Fig. S1) suggests the dominant source of Mg^{2+} is mineral (mainly carbonate) dust. These ions were strongly correlated ($R^2 = 0.827$), and the mean molar ratio was 0.98 (the regression lines were near unity), indicating that SO_4^{2-} and NO_3^- were present partly in CaSO_4 , MgSO_4 , $\text{Ca}(\text{NO}_3)_2$, and $\text{Mg}(\text{NO}_3)_2$. Fig. 2(d) shows that strong correlations between K^+ and Cl^- were observed, whereas K^+ and SO_4^{2-} were not significantly correlated (Fig. S2), indicating that KCl was the main form of potassium, consistent with earlier studies (Zhang *et al.*, 2013; Zhou *et al.*, 2018).

Previous studies (Arimoto *et al.*, 1996; Hassan and Khoder, 2017) reported that ion mass concentration ratios, such as $\text{NO}_3^-/\text{SO}_4^{2-}$, could be used as an indicator of the relative importance of WSI sources (i.e., stationary vs. mobile sources). In the present study, the value of $\text{NO}_3^-/\text{SO}_4^{2-}$ ranged from 0.06 to 1.60, with an average of 0.41, theoretically indicating that the WSIs originated from a stationary source. However, the ratio has a wide range. Li *et al.* (2008) reported that surface soil from the Junggar Basin (an agricultural area, where chemical fertilizer is

Table 1. Concentrations of WSIs in the $\text{PM}_{2.5}$ during the sampling period ($\mu\text{g m}^{-3}$).

	$\text{PM}_{2.5}$	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	F^-	Cl^-	NO_3^-	SO_4^{2-}
Mean	197.40	3.25	8.99	0.81	0.21	4.05	0.22	5.09	8.52	26.66
SD	81.79	1.64	6.99	0.74	0.16	3.22	0.19	2.80	4.78	15.05
Min	55.78	0.23	0.21	0.11	0.05	0.04	0.01	0.60	1.90	8.87
Max	481.43	6.60	25.30	4.00	0.60	23.45	0.85	11.76	22.51	63.21

SD: standard deviation.

Fig. 2. Scatter plots of certain cations and anions: (a) NH_4^+ vs. SO_4^{2-} , (b) $\text{NO}_3^- + \text{SO}_4^{2-}$ vs. NH_4^+ , (c) $\text{NO}_3^- + \text{SO}_4^{2-}$ vs. $\text{NH}_4^+ + \text{Ca}^{2+} + \text{Mg}^{2+}$, and (d) Cl^- vs. K^+ .

used widely) and salt flats (saltwater lakes) contained high concentrations of sulfates, and they could be transported into the atmosphere by wind and human activity. Therefore, transported soil dust and anthropogenic aerosols were the major sources of WSIs in Urumqi in winter.

Trace Elements

Table 2 shows a statistical description of the concentrations of trace elements obtained from Urumqi and other cities around the world. The total concentration of trace elements was $9.12 \pm 8.21 \mu\text{g m}^{-3}$, accounting for 4.5% of the $\text{PM}_{2.5}$ mass concentration. The mean concentrations of atmospheric Pb, Cr, and As (elements linked to coal combustion) were 87.60, 83.48, and 34.99 ng m^{-3} in Urumqi in 2012 and respectively decreased by 27.72%, 62.82%, and 14.71% compared with winter 2009, which demonstrated that substituting natural gas for coal was useful in controlling these elements. However, the elements of Ni, Cu, and Mn respectively increased by 6.24, 2.40, and 1.75 times after the natural gas substitution. Previous studies observed that

particles from traffic emissions are rich in Cu, Mo, Ca, and Mn; these metals are linked to non-exhaust sources, such as brake and tire wear debris, and traffic-related dust, and Ni can be obtained from diesel exhaust (Hsu *et al.*, 2016). Thus, the rising trend of some $\text{PM}_{2.5}$ -bound elements suggests that the increase of emissions and vehicles might outweigh the urban pollution control efforts. The concentration of atmospheric Cd was 22.43 ng m^{-3} , nearly four times higher than the NAAQS of China (GB3095-2012) and the WHO limit of 5 ng m^{-3} . The concentration of Zn was much higher than in other cities, indicating severe atmospheric Zn pollution in Urumqi. Studies have shown that Cd and Zn are carcinogenic (Duan and Tan, 2013; Huang *et al.*, 2015); therefore, the municipal government should strengthen the control of metal pollution.

The concentration of Pb was higher than those in Seoul and New York but lower than those in New Delhi and other cities in China. The Cd concentration was much higher than those in Lanzhou, New York, and Korea; similar to that in New Delhi; and lower than that in Nanjing. Cr, Zn, Ni, Cu,

Table 2. Concentrations of elements in the PM_{2.5} in Urumqi and other cities (ng m⁻³).

Site	Pb	Cd	Cr	Zn	Ni	Cu	Fe	Mn	Hg	As
Urumqi ^a (2012–2013, winter)	87.60	22.43	83.48	1018.21	310.89	125.47	7156.26	276.69	1.17	34.99
Urumqi ^b (2009–2010, winter)	121.2		224.46		49.78	52.29		158.2	1.31	41.02
Lanzhou ^c (2012–2013, winter)	1056	8.47	30.56	497	14.19	69.15		244		34.16
Beijing ^d (2010–2011, winter)	112.7		25.8	332.2	28.9	38.3	1051.8	74.6		41.1
Nanjing ^e (2013, winter)	393	39.9	35	764	24	121	979	111		24
New Delhi ^f (2013, winter)	600	20	10	640	10	70	1150	640	1.03	40
Korea ^g (2012–2013, winter)	29.2	12.8	0.529	42.8	1.19	4.11	134	9.04		5.06
New York ^h	5	3	3	24	5	6	130	3	1	
NAAQS ⁱ (GB3095-2012)	500	5	0.025						50	6
WHO ⁱ	500	5			25			150	1000	6.6

Note: ^a This study; ^b ABuDaLi-mu *et al.*, 2012; ^c Tan *et al.*, 2017; ^d Yu, 2013; ^e Li *et al.*, 2016a; ^f Pant *et al.*, 2015; ^g Han *et al.*, 2015; ^h Qin and Hopke, 2006; ⁱ Duan and Tan, 2013.

and Fe concentrations were much higher than those in other cities and the NAAQS of China and the WHO limit. The concentration of arsenic was similar to those in other Chinese cities and New Delhi but higher than those in Korea and the NAAQS and WHO limits.

To examine the preliminary sources of elements, the EF method was employed to distinguish man-made from natural sources (Han *et al.*, 2015; Hsu *et al.*, 2016). The EF values of ten elements were calculated based on Fe as a crustal reference (Han *et al.*, 2015). The EF value for Mn was < 10 (Fig. S3), indicating that the element was present mostly from natural processes. By contrast, the EF values of As, Cr, Hg, Pb, Cu, Zn, Ni, and Cd were > 10, suggesting that they were mostly from anthropogenic sources.

Carbonaceous Species

Fig. 3 illustrates the daily variation in carbonaceous species in PM_{2.5} during winter in Urumqi. OC and EC had similar daily variation patterns during the sampling period. The concentrations of OC and EC ranged from 7.66 to 106.24 $\mu\text{g m}^{-3}$ and from 2.89 to 31.19 $\mu\text{g m}^{-3}$, with mean

values of 33.98 and 10.84 $\mu\text{g m}^{-3}$, respectively. The total carbonaceous aerosols (TCA = 1.6 $\times$ OC + EC) accounted for 34.02% of the PM_{2.5}, suggesting that carbonaceous species were a substantial fraction of the PM_{2.5} in Urumqi in winter but were lower than the proportions (> 40%) found in megacities such as Beijing, Guangzhou, Zhengzhou, and Lanzhou (Zhang *et al.*, 2013; Tan *et al.*, 2017). Generally, the OC/EC ratio is a useful tool for identifying sources of carbonaceous aerosols (Li *et al.*, 2009). The OC/EC ratio fell mostly within 2.0–5.5, with a mean ratio of 3.21, in Urumqi in winter (Figs. 3 and S4). Generally, the OC/EC ratio has a great distinction between coal combustion (0.3–7.6), vehicle emission (0.7–2.4), and biomass burning (4.1–14.5) (Watson *et al.*, 2001). In this study, the high OC/EC ratio and strong correlation ($R^2 = 0.748$) indicated that OC and EC originated from similar sources, such as fossil fuel combustion and biomass burning.

OC originates either directly as primary OC or from photochemical conversion as secondary OC (SOC). Additionally, the stable atmosphere, low temperatures, and high relative humidity in winter accelerate the condensation

Fig. 3. Time series of carbonaceous species in the PM_{2.5} in Urumqi.

Fig. 4. PAH profiles of the Urumqi aerosol in winter.

of volatile organic compounds (VOCs) on PM (Sheehan and Bowman, 2001). Moreover, the OC/EC was higher than 2.0 in all sampling periods (Fig. S4). Therefore, in this work, SOC was estimated using the EC tracer method [$\text{SOC} = \text{OC} - \text{EC} \times (\text{OC}/\text{EC})_{\min}$] (Cabada *et al.*, 2004). The time series of SOC is illustrated in Fig. 4. The estimated SOC in this study was $0\text{--}62.85 \mu\text{g m}^{-3}$, with a mean concentration of $18.90 \pm 12.92 \mu\text{g m}^{-3}$, and the SOC/OC ratio was 0.53 ± 0.13 . The high levels of SOC and SOC/OC can be attributed to domestic heating in winter enhancing emissions of SOC precursor gases. Additionally, a low planetary boundary layer and thermal inversion height frequently occur in winter in Urumqi and facilitate the accumulation of atmospheric pollutants. Furthermore, the high relative humidity and low temperature in winter (Fig. 1) also accelerate the adsorption or condensation of VOCs on PM.

Cor and 16 priority PAHs recommended by the U.S. EPA were measured in this study. The mean concentration of the total PAHs was $829.81 \pm 185.30 \text{ ng m}^{-3}$ (Fig. 4 and Table S1), which was higher than that observed in Urumqi in 2010 (54.11 ng m^{-3}) by Limu *et al.* (2013). Pyr, Chr, BbF, BkF, IcdP, and BghiP were the most prevalent individual PAH compounds, accounting for 54.94% of the total PAHs. In the present study, BaP ranged from 0.40 to 16.67 ng m^{-3} , and the mean concentration was $6.15 \pm 4.3 \text{ ng m}^{-3}$, which exceeded the health-based guideline of the NAAQS (GB 3095-2012) and indicated a high contribution to the cancer risk in winter in Urumqi.

To facilitate further analysis, PAHs were divided into three types: low molecular weight (LMW; two- and three-ringed PAHs), middle molecular weight (MMW; four-ringed PAHs), and high molecular weight (HMW; PAHs with five or more rings). HMW PAHs were the predominant compounds in the present study, accounting for $49.78 \pm 6.60\%$ of the total PAHs, whereas MMW and LMW PAHs

only accounted for $32.65 \pm 4.50\%$ and $17.57 \pm 4.31\%$, respectively (ternary plots in Fig. 4). The high contribution of HMW PAHs was caused by the many conversions of volatile compounds from the gas phase into particles in winter (Yu *et al.*, 2016). The ratios of IcdP/(BghiP + IcdP), Flu/(Flu + Pyr), and BaA/(Chr + BaA) were 0.42–0.64, 0.34–0.61, and 0.21–0.52, respectively (Fig. S5), which suggested that combustion (e.g., grass, wood, coal, petroleum, or natural gas combustion) was the major source of PAHs in Urumqi in winter.

Pollution Pathways, Source Area Distribution, and Source Apportionment

Pollution Pathways and Source Area Distribution

All the transport trajectories for the $\text{PM}_{2.5}$ were clustered into three clusters (different colors representing different origins of air masses), and the concentrations of $\text{PM}_{2.5}$ and chemical species of different clusters are shown in Fig. 5(a). Affected by the cold Siberian current, west winds prevail in Urumqi in winter (Li *et al.*, 2008b). Therefore, airflow trajectories from the west are dominant. Moreover, the valley topography facilitates the enrichment of pollutants. Clusters 1 (C1), 2 (C2), and 3 (C3) accounted for 67.55%, 21.18%, and 11.27% of the total trajectories, respectively. C1 represented the airflow from western Xinjiang (the surrounding western area of Urumqi) and exhibited higher load values for $\text{PM}_{2.5}$, OC, SO_4^{2-} , EC, and metals. C2 and C3 were the long-range-transported emissions from eastern Kazakhstan, which then passed through Karamay, Kuitun, and Dushanzi. These areas include numerous petrochemical facilities. Therefore, the $\text{PM}_{2.5}$ and relative components maintained a high concentration despite the long-distance transport and high-wind-speed dilution of contaminants.

Earlier studies on $\text{PM}_{2.5}$ in Urumqi reported that the typical topography (surrounded on three sides by Tianshan Mountains, with only one open mouth facing north, where the

Fig. 5. Analysis of wintertime Urumqi aerosol based on (a) clustering the 48-h air particle backward trajectories and (b) PSCF.

pollution source area lies) may accelerate the accumulation of gas-phase pollutants such as SO_2 , NO_2 , and VOCs; these gas-phase components then undergo heterogeneous reactions with mineral dust particles in the atmosphere and enhance haze formation (Li *et al.*, 2008a, b; Ma *et al.*, 2012; Wang *et al.*, 2017). Moreover, in order to find the sources of highly enriched elements in $\text{PM}_{2.5}$, Li *et al.* (2008a) collected surface soil samples on the ground in Urumqi and the surrounding areas of Xinjiang, which could transport aerosols to the city, and found that the surface soil samples of Urumqi contain concentrations of Pb, As, Cd, and S as high as 6.6, 7.6, 303.8, and 7.8 times their crustal abundance, which could be from the deposition of these elements in the early years. Thus, in this study, using backward trajectories, PSCF was applied to explore the likely regional sources and transport pathways of $\text{PM}_{2.5}$ in southern Urumqi. Fig. 5(b) shows that areas to the north (the Midong-Wujiaqu industrial zone) and the west (the Junggar Basin and Gobi and Taklamakan Deserts) of the city might be vital source areas of air pollution for southern Urumqi. Therefore, anthropogenic aerosols (from inside the city) and transported mineral dust (from outside the city) are the main sources of PM in Urumqi.

Source Identification and Apportionment

Using the PMF model with the obtained full data set as input data, five main sources were identified: crustal minerals, vehicle emissions, biomass and waste incinerator burning, industrial pollution and coal combustion, and secondary formation. The concentrations and source profiles modeled for each source are shown in Fig. 6, and the relative contributions from each source to the $\text{PM}_{2.5}$ are illustrated in Fig. 7.

Crustal material was characterized by high levels of Mg^{2+} , Ca^{2+} , Fe, and Mn. Ca^{2+} content was higher than that of other components, indicating Ca-rich dust (Zhang *et al.*, 2013). This source also contained nearly 20% trace elements, such as Cd, Cr, Zn, and Cu, suggesting that some trace elements in the $\text{PM}_{2.5}$ in Urumqi may originate not only from anthropogenic sources but also from resuspended road dust and transported soil from the Gobi Desert and Junggar Basin; this finding agrees with that of Li *et al.*

(2008). Crustal minerals accounted for 18.94% of the $\text{PM}_{2.5}$, which was higher than the proportion for other cities in China (Zhang *et al.*, 2013; Tan *et al.*, 2017).

Vehicle emissions were characterized by high Ni (88.78%), Zn (43.05%), Pb (42.10%), Cu (32.63%), OC (33.01%), and EC (27.64%) content. Ni, OC, and EC are major pollutants originating from diesel and gasoline combustion (Du *et al.*, 2017; Hsu *et al.*, 2016; Liu *et al.*, 2017). Cheng *et al.* (2010) reported that zinc is a common additive in lubricating oil for engines, and Cu and Pb are linked to brake wear. The contribution of this source to the $\text{PM}_{2.5}$ was 20.87%. It is likely to be an influential source of $\text{PM}_{2.5}$ in Urumqi because the number of automobiles has been increasing in recent years.

Another source was characterized by high levels of K^+ (52.71%), F^- (39.68%), and Cl^- (36.36%). K^+ is an excellent tracer of biomass burning (Zhang *et al.*, 2013; Yu *et al.*, 2016), and Wang *et al.* (2016) demonstrated that F^- and Cl^- are released from not only coal but also waste incineration. Thus, this source was identified as biomass and waste burning, and the contribution of this source to the $\text{PM}_{2.5}$ was 21.42%, constituting the second largest source of $\text{PM}_{2.5}$. Xinjiang is a large agricultural area, and the high contribution of biomass and waste burning is probably caused by the open burning of straw during the harvest season in addition to the domestic usage of straw in winter, which releases atmospheric particles that are then transported to Urumqi.

Another source was represented by high loadings of As (83.76%), Cd (43.27%), Cr (36.44%), SO_4^{2-} (31.95%), and Cl^- (32.95%), which are closely related to industrial pollution and coal combustion (Hsu *et al.*, 2016; Liu *et al.*, 2017). Thus, this source was identified as industrial pollution and coal combustion. The contribution of this source to the $\text{PM}_{2.5}$ was 10.46%. Despite Urumqi replacing coal with natural gas, air from the surrounding areas, such as the Midong-Wujiaqu industrial zone, Karamay, Kuitun, and Dushanzi, which are characterized by large-scale petrochemical and oil-refining facilities and coal-fired power plants, could enter Urumqi (as shown in Fig. 5). This indicates that comprehensive improvement of the atmosphere requires regional cooperation.

Fig. 6. Factor profiles (% of the total species) obtained from the PMF analysis.

The remaining source was dominated by NH₄⁺ (91.84%), SO₄²⁻ (45.64%), NO₃⁻ (37.08%), and Na⁺ (65.13%), which are related to secondary formation (Tao *et al.*, 2014; Liu *et al.*, 2017). Studies have suggested that NO₃⁻, SO₄²⁻, and NH₄⁺ are formed through reactions of SO₂, NO_x, and NH₃ and transformed from a gaseous state into particles (Zhang *et al.*, 2013; Tao *et al.*, 2014; Liu *et al.*, 2017). This source was identified as secondary formation, and the contribution of this source to the PM_{2.5} was 28.31%. Secondary formation was the largest contributor to the PM_{2.5} in Urumqi; this result is probably attributable to the common secondary reactions in Urumqi caused by its high concentrations of precursor gases, high humidity, and low wind speed, which are favorable to photochemical reactions.

Overall, secondary formation was the source contributing the most to PM_{2.5} in southern Urumqi in winter, followed by biomass and waste burning, vehicle emissions, crustal minerals, and industrial pollution and coal combustion; their mean contributions to PM_{2.5} were 28.31%, 21.42%, 20.87%, 18.94%, and 10.46%, respectively (Fig. 7). Other sources were not identified because sources of PM are diverse and complicated, especially open sources, such as agricultural production and catering oil fumes. Regional transportation of atmospheric pollution may contribute substantially to the atmospheric PM in Urumqi (Fig. 5). According to the source apportionment, biomass burning, vehicle exhaust, and soil dust should be managed and controlled effectively. Additionally, regional cooperation should be implemented to reduce the PM_{2.5} pollution in

Urumqi

CONCLUSIONS

This study investigated the chemical composition, emission sources, and source areas of PM in Urumqi during winter after the energy switch from coal to natural

Fig. 7. Source apportionment of the PM_{2.5} in winter in Urumqi.

gas. Despite the atmospheric pollution becoming more severe, the $\text{PM}_{2.5}$ mass concentration decreased after the conversion; however, the average mass concentration, at $197.40 \mu\text{g m}^{-3}$, remained above five times the annual limit specified by the NAAQS ($35 \mu\text{g m}^{-3}$). WSIs, metallic elements, and carbonaceous species were the major components, accounting for 66.21% of the total $\text{PM}_{2.5}$ mass, with the WSIs and carbonaceous aerosols forming the largest fractions (33.72% and 34.02%, respectively). Furthermore, hazardous levels of carcinogenic substances, such as Pb, Cd, Cr, Zn, Ni, As, and BaP, were observed in this study. The five primary sources of $\text{PM}_{2.5}$ were identified as secondary formation, biomass and waste burning, vehicle emissions, crustal minerals, and industrial pollution and coal combustion. Finally, soil dust and anthropogenic aerosols transported from neighboring areas were major factors; therefore, regional cooperation is recommended for reducing the $\text{PM}_{2.5}$ pollution in Urumqi.

ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China (No. 41465007) and the State Key Laboratory of Organic Geochemistry, GIGCAS (Grant No. SKLOG-2016201624).

SUPPLEMENTARY MATERIAL

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

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Received for review, January 4, 2019

Revised, April 16, 2019

Accepted, April 25, 2019