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Chemical Composition of Particulate Matter from Traffic Emissions in a Road Tunnel in Xi'an, China

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ABSTRACT

Chemical compositions of particulate matter (PM) from traffic emissions vary by region and with time. Therefore, it is necessary to obtain local mobile source profiles of PM to support regional researches for vehicle emission control policy, source apportionment modeling, etc. In this study, $PM_{2.5}$ and PM_{10} samples were collected from a highway tunnel in Xi'an in northwestern China. The chemical composition, specifically, the OC, EC, water-soluble ions, and elements, was analyzed in detail to (1) provide local PM profiles for a mixed vehicle fleet, (2) identify the origins of different elements in the tunnel environment, and (3) determine the associated factors influencing the profiles. The $PM_{2.5}$ profiles in the tunnel were identified as OC (34.10%), EC (11.96%), water-soluble ions (18.22%), and elements (27.73%), while the PM_{10} profiles included OC (28.48%), EC (8.59%), water-soluble ions (14.17%), and elements (33.36%), respectively. The origins of the elements in the tunnel were classified into three categories by the receptor modeling approach: resuspended road dust and brake wear, vehicle exhaust and tire wear, and tailpipe emissions from diesel vehicles (DV). The mass fractions of OC, EC, and elements originating from resuspended road dust and brake wear were mainly affected by vehicle driving conditions (i.e., uphill/downhill and speed), whereas the mass content of bromine (Br) was influenced by the proportion of DV in the fleet.

Keywords: Traffic emissions; $PM_{2.5}$; PM_{10}; Source profile; Road tunnel.

INTRODUCTION

Particulate matter (PM) is a primary pollutant of atmospheric haze pollution in China, which has an adverse impact on public health, especially on human lungs and hearts (Delfino *et al.*, 2008; Spira-Cohen *et al.*, 2011; Liu *et al.*, 2015a). Motor vehicles are a major source of PM in the atmosphere. According to the official source apportionment results of $PM_{2.5}$ in 15 cities of China (MEPC, 2018), the contribution of motor vehicles in 11 cities was greater than 20%, with an average value of 23.36%. In order to better control PM pollution, it is essential to understand the physical and chemical characteristics of the PM emitted from motor vehicles. Vehicle emissions can be investigated using various testing methods, such as the tunnel test (Pio *et al.*, 2013; Alves *et al.*, 2015b), dynamometer test (Na *et al.*, 2015; Pietikainen *et al.*, 2015), and on-road test (Yao *et al.*, 2015; Zhang *et al.*, 2015a). Compared with other approaches, tunnel test can easily obtain the emission characteristics of PM with mixed vehicle fleet under actual driving conditions (Franco *et al.*, 2013; Pant *et al.*, 2017). Further, PM pollutants collected in the tunnel include not only vehicle exhaust, but also some associated emissions, such as friction emissions (tire, brake, and road surface wear) and road dust (Handler *et al.*, 2008; Thorpe and Harrison, 2008). Therefore, tunnel test can estimate the PM pollution caused by motor vehicles more comprehensively.

Faced with the increasingly serious air pollution problem, several studies have been conducted on the chemical characteristics of PM from vehicle emissions by means of tunnel tests in China. Most of these studies were carried out in the eastern part of China, mainly in Guangzhou (Huang *et al.*, 2006; He *et al.*, 2008; Dai *et al.*, 2015; Zhang *et al.*, 2015b), Hong Kong (Chiang and Huang, 2009; Ho *et al.*, 2009; Cheng *et al.*, 2010), and Shanghai (Zhao *et al.*, 2010; Liu *et al.*, 2015b, c). However, no such studies have

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been conducted in the western part of China, such as Xi'an, which is the capital of Shaanxi Province (Fig. 1). Xi'an is the largest city in northwestern China, with up to 2.42 million licensed vehicles in 2015. Moreover, vehicle emission was the second largest source of $PM_{2.5}$ pollution in Xi'an, with contributions of 14.9% and 12.9% in urban and rural areas, respectively (Wang *et al.*, 2015). Considering that each region has its own characteristic vehicular fleet (vehicle type, fuel quality, etc.) and corresponding conditions (transportation infrastructure, driving conditions, etc.) that influence vehicular emissions, it is important to conduct local studies to obtain local emission profiles (Zhang *et al.*, 2015b; Alves *et al.*, 2016).

This study carried out a tunnel experiment in Xi'an, with the following objectives: (1) investigate the chemical characteristics of PM in a typical highway tunnel, and obtain the local mobile source profiles of PM; (2) identify the main sources of each element in PM, such as vehicle exhaust, tire wear, brake wear, and resuspended road dust; and (3) determine the main factors affecting the chemical composition of PM from road traffic. Results from this study will provide necessary data for subsequent studies in northwestern China, such as vehicle emission control policy, source apportionment modeling, and air quality simulation.

MATERIALS AND METHODOLOGY

Sampling Sites

The sampling experiment was conducted in the Qinling No. 1 Tunnel, which is located in the southwest suburb of Xi'an. There are no significant anthropogenic pollution sources around it, apart from motor vehicles. This highway tunnel has two independent bores (southbound and northbound), each of which has two lanes, as shown in Fig. 1. The southbound bore is 6102 m in length, with an upgrade of +2.58% (uphill); whereas the northbound bore is 6144 m, with a downgrade of –2.58% (downhill). A ventilation shaft was placed at 1657 m from the entrance of the northbound bore, which was ventilated naturally during the test. There were four sampling sites in the southbound bore, with three sampling sites in the northbound bore (Fig. 1). More details of the sampling sites are summarized in Section A1 in the supplementary material. The samplers were placed in the emergency parking areas at a distance of 3 m from the nearside lane. This experiment was performed for several purposes, such as determining gas pollution (e.g., CO, NO_x, SO_2, and O_3) and visibility in the tunnel, while this study only focused on the chemical composition of the PM.

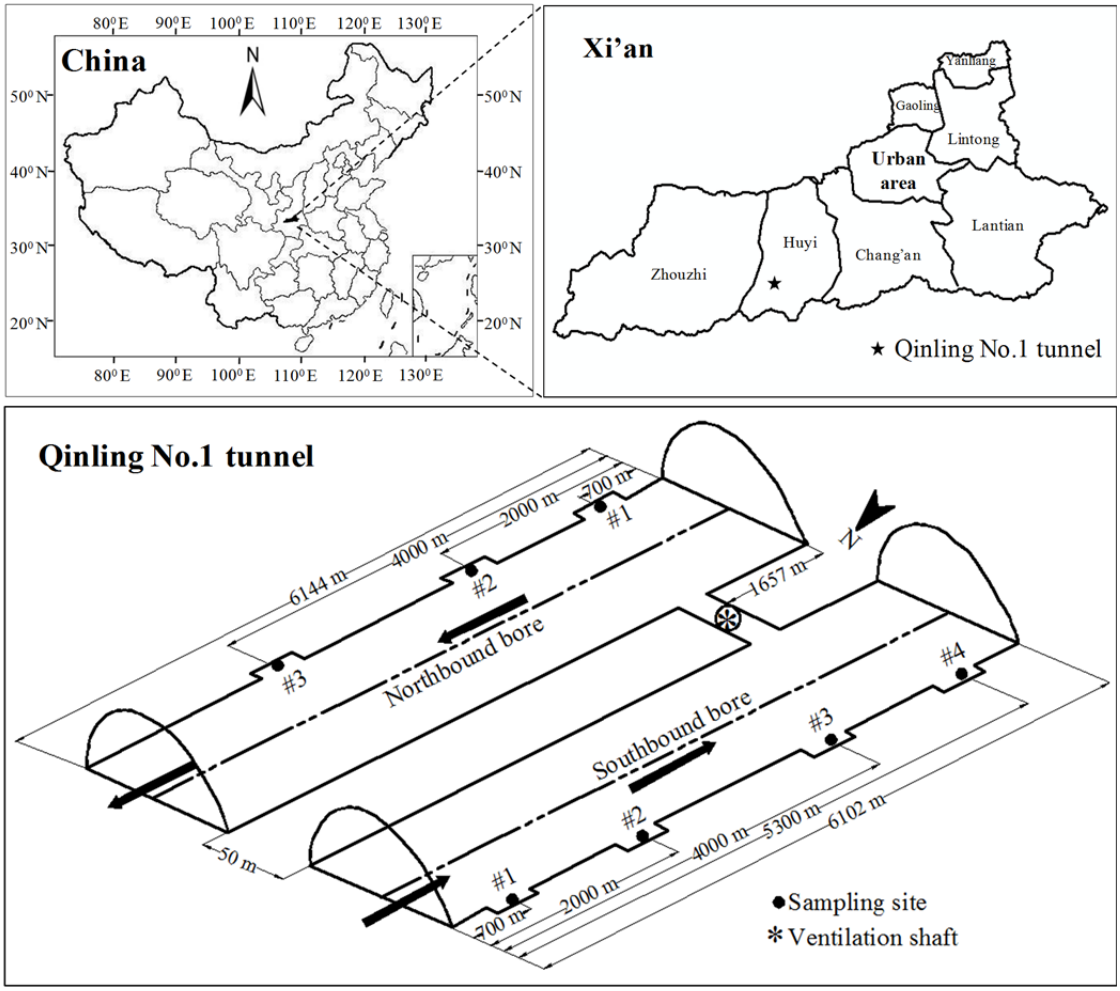

Fig. 1. Location of Qinling No. 1 Tunnel and its sketch map.

Sampling Method

The four-channel PM sampler (TH-16A, Tianhong Corporation, Wuhan, China) was used to sample inhalable particles (PM_{10}) and fine particles ($PM_{2.5}$) simultaneously. Two channels were loaded with 47-mm quartz-fiber filters, while the other two were loaded with 47-mm Teflon filters. The operating flow rate was 16.7 L min^{-1}, with a sampling height of 2 m. During this experiment, two samplers were used to collect PM concurrently, one of which was placed at Site #1, while the other one was moved day by day from Site #2 to Site #3, and then to Site #4. The duration of each sampling was 6–10 h to ensure that sufficient material was collected, with at least two sets of PM samples being collected at each site. The entire experiment lasted for one week (March 14–20, 2016), with 20 sampling sets being collected during two daily periods, one was at daytime between 8:00 to 20:00, and the other one was at nighttime between 20:00 to 8:00 (Table 1).

The meteorological parameters at each sampling site were recorded in parallel using an automatic weather station (GH-BPR, Huayun Puda Corporation, Beijing, China). A roadside laser traffic-survey instrument (AxleLight RLU11, NanoSense Corporation, Beijing, China) was used to investigate the traffic parameters during the entire test, including vehicle volume, speed and vehicle types.

Mass and Chemical Analysis

The PM mass concentrations were determined gravimetrically by weighing the filters before and after sampling (an MC5 Sartorius microbalance was used for weighing). Subsequently, the quartz-fiber filter was cut into two pieces to detect the carbonaceous species and water-soluble ions separately, whereas a Teflon filter was used to analyze the elements.

The contents of organic carbon (OC) and elemental carbon (EC) were determined by a Model-4 Semi-Continuous OC-EC Field Analyzer (Sunset Laboratory Inc.) with the thermal-optical transmission method (TOT; NIOSH, 2003). The water-soluble ions were analyzed by an ICS-5000 ion chromatograph (Thermo Fisher Scientific Inc.), following the method of Zhang *et al.* (2014). Trace elements were analyzed by an Epsilon-5 X-ray fluorescence spectrometer (PANalytical Inc.). Finally, three ionic species (NH_4^+, NO_3^-, and SO_4^{2-}) and seventeen elements (Na, Mg, Al, P, S, K, Ca, Ti, Cr, Mn, Fe, Cu, Zn, Br, Ba, Hg, and Pb) were identified. More details of the analysis procedure are available in Section A2 in the supplementary material. The quality assurance and control of chemical analysis are summarized in Section A3 in the supplementary material.

Statistical Analysis

Data analysis was performed using SPSS (Version 17). Species' correlation analysis was conducted based on Pearson correlation. Multivariate analysis of variance (multivariate-ANOVA) was carried out to test for significant variations in chemical composition between $PM_{2.5}$ and PM_{10} (Haase and Ellis, 1987). Moreover, the receptor modeling approach, including enrichment factor (EF) and principal component analysis (PCA), were adopted to identify the main sources of elements in the PM. Enrichment factor (EF) is a good indicator of the contributions of anthropogenic sources (Cevik *et al.*, 2009). It is calculated by the following equation:

$$EF_i = \frac{(C_i/C_r)_{Tunnel}}{(C_i/C_r)_{Background}} \quad (1)$$

where EF_i is the enrichment factor of element i, C_i is the concentration of element i, C_r is the concentration of the reference element, tunnel and background represent the sampling sources of the elements. Aluminum (Al) was selected as the reference element in this study (Sakan *et al.*, 2014), while the average background concentrations of elements in the soil of A Layer in Shaanxi Province were considered as the background values in this calculation (NEMCC, 1990).

PCA is a factor analysis method, which is a useful tool to apportion the emission sources and quantify their contributions (Larsen and Baker, 2003). In PCA, all factors with eigenvalues over 1 were extracted and rotated using the varimax method. Each of these factors can be identified as either an emission source or a chemical interaction. In order to determine the suitability of the dataset for PCA, a Kaiser-Meyer-Olkin (KMO) value of greater than 0.6 was required (Lawrence *et al.*, 2013). High values (close to 1) usually indicate that the PCA is useful with the selected dataset. Finally, the multifactorial analysis of variance (multifactorial-ANOVA) was applied to analyze the main influential factors on PM composition in the tunnel (Wolny and Kedzia, 2008).

RESULTS AND DISCUSSION

Chemical Composition of PM

The chemical compositions of $PM_{2.5}$ and PM_{10} were analyzed separately in this study. The results are summarized in Table 2. Moreover, the results of multivariate-ANOVA

Table 1. Summary of the samples in the tunnel.

Site	Bore	Sampling number	Sampling period
#1	Southbound	6	
#2		2	
#3		2	
#4		2	Daytime or Nighttime
#1	Northbound	4	
#2		2	
#3		2	

Table 2. Chemical compositions of PM in several studies (%).

City	Xi'an		Xi'an	Xining	Hong Kong	Guangzhou	Yantai	
Year	2016		2010	2010	2003	2013	2014	
Source	Qinling No.1 Tunnel		Ambient air	Road dust	Shing Mun tunnel	Zhujiang tunnel	Wuzushan tunnel	Kuixinglou tunnel
References	This study		Wang *et al.* (2015)	Han *et al.* (2014)	Cheng *et al.* (2010)	Dai *et al.* (2015)	Cui *et al.* (2016)	
PM	$PM_{2.5}$	PM_{10}	$PM_{2.5}$[a]	PM_{10}	$PM_{2.5}$[b]	$PM_{2.5}$[b]	$PM_{2.5}$[b]	$PM_{2.5}$[b]
OC	34.10 ± 6.43	28.48 ± 16.02	13.04	5.98 ± 1.86	27.25	18.07	27.67	17.70
EC	11.96 ± 4.30	8.59 ± 3.81	4.70	0.60 ± 0.13	50.23	17.75	32.10	10.36
NH_4^+	3.19 ± 2.04	2.14 ± 1.54	5.82		2.14	0.18	0.87	0.14
NO_3^-	6.05 ± 4.42	4.30 ± 2.97	12.48	0.06 ± 0.06	0.84	0.11	3.28	3.65
SO_4^{2-}	8.98 ± 4.82	7.73 ± 4.40	16.20	1.38 ± 0.83	5.42	0.66	3.71	3.02
Na	1.48 ± 1.20	3.19 ± 2.67		0.84 ± 0.24	0.76	3.82	0.37	0.32
Mg	2.29 ± 1.27	4.17 ± 3.91		1.71 ± 0.39	0.27	0.54	2.01	1.13
Al	2.77 ± 1.57	3.67 ± 1.83		5.16 ± 0.62	0.17	3.41		
P	0.24 ± 0.14	0.26 ± 0.11		0.11 ± 0.03	0.05			
S	16.00 ± 7.64	12.89 ± 4.77			2.06			
K	0.48 ± 0.27	0.59 ± 0.24		1.55 ± 0.17	0.22	0.37	0.97	0.59
Ca	1.85 ± 1.27	4.27 ± 2.40		9.84 ± 2.33	0.42	2.09	5.42	2.12
Ti	0.04 ± 0.03	0.08 ± 0.04	0.07	0.26 ± 0.05	0.06		0.21	0.09
Cr	0.01 ± 0.01	0.02 ± 0.02		0.02 ± 0.01	0.01	0.01	0.01	0.14
Mn	0.03 ± 0.03	0.06 ± 0.04	0.07	0.08 ± 0.01	0.02	0.09	0.09	0.09
Fe	2.14 ± 1.86	3.56 ± 2.87	0.98	2.74 ± 0.49	0.73	4.23	4.59	4.19
Cu	0.02 ± 0.02	0.04 ± 0.03		0.03 ± 0.04	0.04	0.10	0.04	0.11
Zn	0.19 ± 0.04	0.26 ± 0.05	0.98	0.10 ± 0.08	0.15	0.17	0.17	0.11
Br	0.004 ± 0.004	0.003 ± 0.002			0.004			
Ba	0.14 ± 0.13	0.27 ± 0.23			0.06			
Hg	0.008 ± 0.005	0.011 ± 0.009						
Pb	0.03 ± 0.02	0.04 ± 0.02	0.21	0.03 ± 0.01	0.02	0.01	0.01	0.03

[a] Mass fractions of each species were calculated based on the mass concentrations of $PM_{2.5}$ and its corresponding species.
[b] Mass fractions of each species were calculated based on the emission factors of $PM_{2.5}$ and its corresponding species.

showed that the chemical compositions between $PM_{2.5}$ and PM_{10} were statistically distinct (the *p*-values from all the four tests, i.e., Pillai's trace, Wilk's lambda, Hotelling's trace, and Roy's largest root, were 0.001 less than 0.05). Moreover, EC and the elements of Na, Mg, Ca, Ti, Cu, Zn, and Ba were significantly influenced by particle size (Table 3). The mass fraction of EC in $PM_{2.5}$ was higher than that in PM_{10}, whereas the fractions of Na, Mg, Ca, Ti, Cu, Zn, and Ba in $PM_{2.5}$ were lower than those in PM_{10}.

OC and EC

The mass fractions of OC and EC in $PM_{2.5}$ were 34.10 ± 6.43% (mean ± SD, n = 20) and 11.96 ± 4.30%, while their counterparts in PM_{10} were 28.48 ± 16.02% and 8.59 ± 3.81%. The mass ratios of OC/EC in $PM_{2.5}$ and PM_{10} were 2.99 ± 1.11 and 3.30 ± 1.28, respectively, which showed that the contents of OC were noticeably higher than those of EC. EC is formed due to the incomplete combustion of fuel, and it can be emitted from motor vehicles directly. However, sources of OC are relatively complex, including the primary organic carbon (POC) directly emitted by motor vehicles and secondary organic carbon (SOC) produced from the gaseous precursors (Shen *et al.*, 2011). SOC generation is usually considered to occur when the mass ratio of OC/EC is greater than 2 (Deng *et al.*, 2013; Yu *et al.*, 2004). On the other hand, significant correlations were observed between OC and EC ($r_{PM_{2.5}}$ = 0.91, $r_{PM_{10}}$ = 0.90), suggesting a similar origin (i.e., direct emissions from motor vehicles) for both.

Water-Soluble Ions

The mass fractions of NH_4^+, NO_3^-, and SO_4^{2-} in $PM_{2.5}$ were 3.19 ± 2.04% (n = 20), 6.05 ± 4.42%, and 8.98 ± 4.82%, while those in PM_{10} were 2.14 ± 1.54%, 4.30 ± 2.97%, and 7.73 ± 4.40%. The order of the three ions in terms of their contents from highest to lowest was $SO_4^{2-} > NO_3^- > NH_4^+$. Usually, NH_4^+, NO_3^-, and SO_4^{2-} can be called secondary ions, which are mainly derived from the gas-to-particle conversion of gaseous precursors (NH_3, NO_x, and SO_2), subsequently being coagulated in the PM in the form of $(NH_4)_2SO_4$, NH_4HSO_4, and NH_4NO_3 (Seinfeld and Pandis, 2016). In this study, NH_4^+ was more strongly correlated with SO_4^{2-} ($r_{PM_{2.5}}$ = 0.95, $r_{PM_{10}}$ = 0.91) than NO_3^- ($r_{PM_{2.5}}$ = 0.72, $r_{PM_{10}}$ = 0.70). It is likely because the NH_4NO_3 is more easily volatized into gas phase again due to its strong volatility (Chow, 1995). Moreover, it also indicated that the NH_4^+ in the PM sampled in this tunnel was primarily in the form of $(NH_4)_2SO_4$ or NH_4HSO_4.

Elements

The elements can be classified into five groups (> 5%, 1–5%, 0.1–1%, 0.01–0.1%, and < 0.01%), in terms of their mass fractions (Table 4). It can be seen that the mass fraction of each element varied significantly. S was the most abundant element (16.00 ± 7.64% for $PM_{2.5}$ and 12.89 ± 4.77% for PM_{10}, n = 20), followed by Al, Mg, Fe, Ca, and Na. These six elements accounted for 95.68% and 95.15% of the total mass of the 17 elements in $PM_{2.5}$ and PM_{10}, respectively.

Elements can be categorized into metallic and non-metallic (Fig. 2). The non-metallic elements accounted for 58.59% and 39.42% of the total mass of the 17 elements in $PM_{2.5}$ and PM_{10}, respectively, with S being the most abundant element. Meanwhile, the mass fractions of metallic elements were 41.41% and 60.58%, with Al, Mg, and Fe being the most abundant elements. Moreover, metallic elements can be further categorized into heavy and other metallic elements (Fig. 3). The heavy metallic element content was low, with contributions of 21.32% and 19.69% to the total mass of metallic elements in $PM_{2.5}$ and PM_{10}, respectively. Fe was the most abundant element. Meanwhile, the mass fractions of other metallic elements were 78.68% and 80.31%, with Al, Mg, and Ca being the most abundant elements.

Source Identification

PM samples collected in the tunnel are a mixture derived from, but not limited to, vehicle exhaust, tire wear, brake wear, and resuspended road dust (Lawrence *et al.*, 2013). The origins of OC, EC, and secondary ions in the tunnel are known, based on past studies (Shen *et al.*, 2011; Seinfeld and Pandis, 2016). Therefore, this study emphasized the analysis of origins of the elements in the tunnel.

Application of Enrichment Factor (EF) Method

Fig. 4 shows the enrichment factors of 14 elements, except for Al, P, and S (P and S were not detected in the soil). The value of EF being equal to 10 is usually regarded as the criterion to determine whether an element is

Table 3. Analysis of the influence of particle size on PM composition.

Species	*F*-value	*p*-value	Species	*F*-value	*p*-value
OC	1.198	0.281	Ca	16.942	**0.000**
EC	6.904	**0.012**	Ti	16.160	**0.000**
NH_4^+	2.872	0.098	Cr	3.157	0.084
NO_3^-	0.036	0.851	Mn	3.896	0.056
SO_4^{2-}	1.535	0.223	Fe	3.919	0.055
Na	10.012	**0.003**	Cu	5.481	**0.025**
Mg	5.724	**0.022**	Zn	18.967	**0.000**
Al	2.925	0.095	Br	0.271	0.606
P	0.019	0.890	Ba	4.420	**0.042**
S	2.393	0.130	Hg	2.189	0.147
K	2.598	0.115	Pb	1.381	0.247

Table 4. Mass fraction distribution of elements.

Mass fraction	> 5%	1–5%	0.1–1%	0.01–0.1%	< 0.01%
$PM_{2.5}$	S	Al, Mg, Fe, Ca, Na	K, P, Zn, Ba	Ti, Pb, Mn, Cu, Cr	Hg, Br
PM_{10}	S	Ca, Mg, Al, Fe, Na	K, Ba, P, Zn	Ti, Mn, Pb, Cu, Cr	Hg, Br

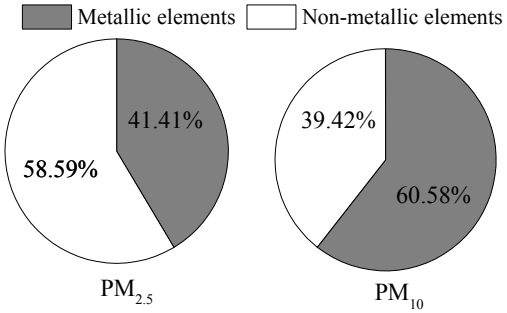

Fig. 2. Mass fractions of metallic and non-metallic elements (Metallic elements: Na, Mg, Al, K, Ca, Ti, Cr, Mn, Fe, Cu, Zn, Ba, Hg, and Pb; Non-metallic elements: P, S, and Br).

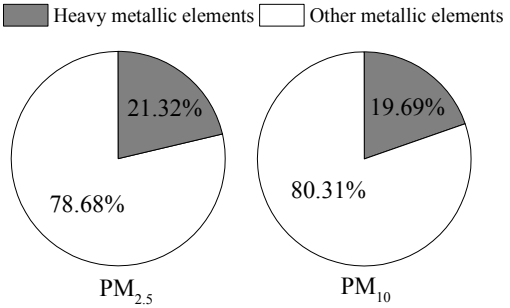

Fig. 3. Mass fractions of heavy metallic and other metallic elements (Heavy metallic elements: Cr, Mn, Fe, Cu, Zn, Hg, and Pb; Other metallic elements: Na, Mg, Al, K, Ca, Ti, and Ba).

influenced by anthropogenic sources (Voutsa and Samara, 2002). EF<10 indicates that the element is not enriched, mainly originating from natural sources (soil, crust, etc.), whereas EF>10 implies that the element is significantly enriched, being greatly influenced by anthropogenic sources (i.e., motor vehicles in this study). It can be seen from Fig. 4 that Cu, Zn, Br, and Pb were enriched (EF values were between 10 and 100), while Hg was significantly enriched (EF > 1000). Therefore, these elements were mainly derived from traffic-related emissions in the tunnel. The EF values of Na, K, Ca, Ti, Mn, and Fe were less than 5, while the EF values of Mg, Cr, and Ba were between 5 and 10. Hence, the influence of anthropogenic sources was very weak on them. They mainly originated from natural sources in the tunnel, such as resuspended road dust.

Application of Principal Component Analysis (PCA)

The mass concentration of elements from $PM_{2.5}$ and PM_{10} were used for the PCA. The dataset provided KMO values of 0.652 for $PM_{2.5}$and 0.675 for PM_{10}, ensuring their suitability for PCA. In Table 5, three principal components with eigenvalues over 1 were extracted, which reflected specific elemental sources. All components explained 83.43% and 89.15% of the total variance in the dataset of $PM_{2.5}$and PM_{10}, respectively.

Component One showed the high loadings of Na, Mg, K, Ca, Ti, Cr, Mn, Fe, Cu, and Ba, with medium loading of Al. Usually, Mg, Al, and Ca are regarded as typical crustal elements (Tran *et al.*, 2012). Mg and Ca can also be derived from road surface wear (Kupiainen *et al.*, 2003). Nevertheless, the EF values of all these elements were lower than 10 (Fig. 4), which suggested that they mainly originated from resuspended road dust. Fe originates from crust, as well as brake wear (Sanders *et al.*, 2003). However, its EF values were lower than 10 (Fig. 4). Hence, its source was more likely to be resuspended road dust. Cu and Ba are the two main additives in brake pads (Sanders *et al.*, 2003; Birmili *et al.*, 2006). Furthermore, the EF values of Cu were greater than 10, which indicated that it was significantly influenced by the traffic flow in the tunnel. Therefore, Component One was strongly associated with a mixed source of resuspended road dust and brake wear.

Component Two was extracted based on the high loadings of P, S, and Zn, and medium loading of Hg. In addition, a high loading of Pb was observed in $PM_{2.5}$ in Component Two, while medium loadings were observed in PM_{10} in both Component One and Two. On the other hand, the correlation analysis results (Table 6) showed that only five elements had good correlations with EC emissions, including P, S, Zn, Hg, and Pb. Moreover, the EF values of

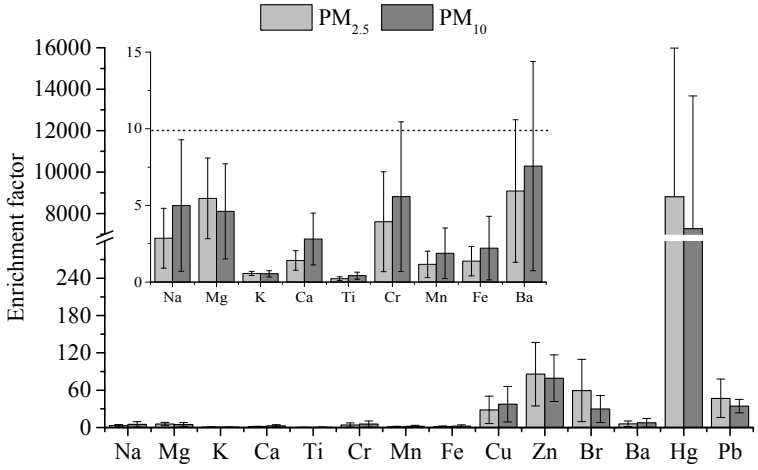

Fig. 4. Enrichment factors of elements in PM.

Table 5. Factor loadings of elements in the PCA (only loadings > 0.5 are shown).

Element	$PM_{2.5}$			PM_{10}		
	Component 1	Component 2	Component 3	Component 1	Component 2	Component 3
Na	0.844			0.897		
Mg	0.772			0.941		
Al	0.516			0.599		
P		0.889			0.926	
S		0.919			0.970	
K	0.770			0.926		
Ca	0.908			0.944		
Ti	0.922			0.947		
Cr	0.971			0.986		
Mn	0.950			0.948		
Fe	0.982			0.985		
Cu	0.737			0.753		
Zn		0.910			0.841	
Br			0.884			0.951
Ba	0.917			0.978		
Hg		0.646			0.700	
Pb		0.836		0.660	0.533	
Variance (%)	46.77	26.62	10.04	56.61	24.20	8.34

Table 6. Correlation coefficients between elements and EC.

Element	EC		Element	EC	
	$PM_{2.5}$	PM_{10}		$PM_{2.5}$	PM_{10}
Na	−0.123	0.237	Mn	−0.129	−0.016
Mg	−0.150	0.022	Fe	−0.024	0.106
Al	0.237	0.425	Cu	0.378	0.473
P	**0.733**	**0.902**	Zn	**0.723**	**0.866**
S	**0.555**	**0.897**	Br	0.227	0.271
K	−0.210	0.310	Ba	0.198	0.069
Ca	0.151	0.310	Hg	**0.555**	**0.506**
Ti	0.155	0.274	Pb	**0.562**	**0.637**
Cr	−0.059	0.062			

Zn, Hg, and Pb were greater than 10 (Fig. 4). Hence, Pb was grouped into Component Two in this study. P is usually used as an additive in the lubricant oil (Alves *et al.*, 2015a). S is mainly generated by the combustion of sulfur-bearing fuel (Lowenthal *et al.*, 1994). Zn is mostly present in rubber tires. Therefore, it is regarded as a product of tire wear (Degaffe and Turner, 2011). In addition, tailpipe can also emit a certain amount of Zn (Huang *et al.*, 1994).

Considering that EC is mainly derived from vehicle exhaust, with all the elements in Component Two being correlated with it, Component Two was attributed to a mixed source of vehicle exhaust and tire wear.

Component Three was interpreted as the exhaust emissions of diesel vehicles (DV), as it had the highest loading of Br. Br has been used as a marker for PM emissions from DV (Chow, 1995). Its EF values were greater than 10 (Fig. 4), which implied that it was clearly affected by the vehicles traveling through the tunnel.

Factors Influencing PM Profiles

Mobile source profiles of PM can be affected by a variety of factors, especially traffic conditions (Gillies *et al.*, 2001; Grieshop *et al.*, 2006; Nelson *et al.*, 2008). The traffic conditions in this study are illustrated in Fig. 5, including uphill/downhill, vehicle volume, speed, and fuel composition. Vehicle volumes at nighttime (227 veh h^{-1}) were significantly less than those at daytime (600 veh h^{-1}), as were the vehicle speeds (48.53 km h^{-1} at nighttime and 55.60 km h^{-1} at daytime). The reason was the difference in the fuel composition of vehicles during the two sampling periods. Vehicles were classified into two categories according to their fuel type, including diesel vehicles (DV: buses and trucks) and gasoline vehicles (GV: other vehicles such as passenger cars). There were more DV in the tunnel at nighttime (69.29% of DV at nighttime and 35.94% of DV at daytime), most of them being heavy-duty trucks, which moved slower than the GV under the same road conditions. Moreover, vehicle speeds during the test were around 50 km h^{-1}, which indicated that the traffic was free-flowing in the tunnel. Based on these traffic parameters collected in the tunnel, the running direction (i.e., southbound and northbound) reflected the differences in vehicle driving conditions (e.g., uphill/downhill and speed) synthetically, while the sampling period (i.e., daytime and nighttime) represented the discrepancies in vehicle fleet composition (e.g., vehicle volume and fuel type). Therefore, these two indexes were chosen as the influential variables. Their impacts on the contents of PM species were analyzed by the multifactorial-ANOVA. Meanwhile, according to the previous PCA results (Table 5), the 17 elements were combined into three components for this analysis. Furthermore, three secondary ions (NH_4^+, NO_3^-, and SO_4^{2-}) were combined into one component as well.

It can be seen from Table 7 that the PM species of OC, EC, and elements in Component One were strongly influenced by the running direction in the tunnel. The fractions of OC and EC in the southbound bore were both higher than those in the northbound bore (Figs. 6–7) due to the reverse slopes in two bores. The speed was reduced, while the engine torque was increased, when vehicles ascended through the southbound bore, which has a positive slope (+2.58%). Meanwhile, the fuel supply was increased to guarantee the engine's power, which induced poor fuel combustion and higher emissions of OC and EC. Nevertheless, the integrated contents of elements in Component One in the northbound bore were greater than those in the southbound bore (Fig. 8). Considering that these elements mainly originated from resuspended road dust and brake wear in the tunnel, it can be interpreted that more braking occurred when the vehicles descended through the northbound bore, which has a negative slope (–2.58%). Moreover, the vehicle speeds were slightly higher in the northbound bore during the same sampling period (Fig. 5). Therefore, more particles might be produced due to a stronger disturbance of the road dust.

Significant influence of the sampling period was observed on the contents of the elements in Component Three in $PM_{2.5}$, whereas its influence was not obvious in PM_{10}. The *p*-value (0.061) in PM_{10} was very close to the significance level (0.05). Hence, this variable had some influence on the elements in Component Three. Only Br was grouped in Component Three, with the fractions of Br sampled at nighttime being higher than those collected at daytime (Fig. 9). Nevertheless, vehicle volumes at nighttime were significantly less than those at daytime (Fig. 5), which presented a reverse trend. Therefore, the influence of vehicle

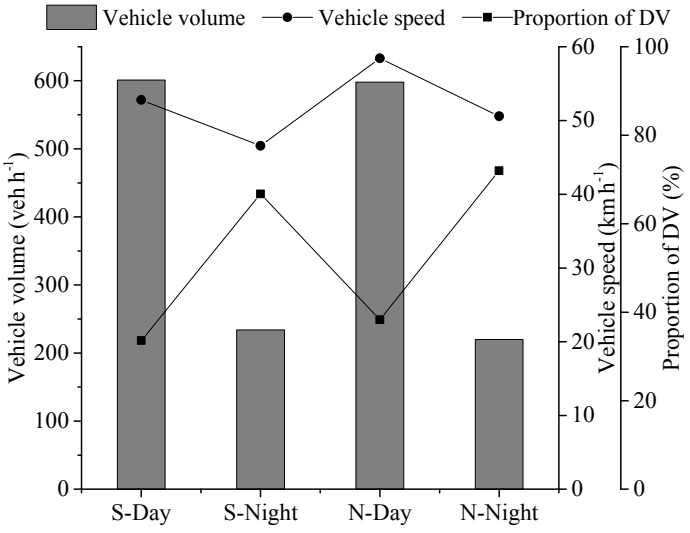

Fig. 5. Traffic conditions during the tunnel test (S: southbound, N: northbound, Day: daytime, Night: nighttime).

Table 7. Influence analysis of traffic conditions on PM profiles.

PM	Influential variables	OC		EC		Secondary ions	
		F-value	*p*-value	*F*-value	*p*-value	*F*-value	*p*-value
$PM_{2.5}$	Running direction	3.695	**0.045**	33.079	**0.000**	0.492	0.495
	Sampling period	0.001	0.973	0.036	0.853	0.009	0.927
	Running direction × Sampling period	0.012	0.913	0.673	0.426	0.493	0.494
PM_{10}	Running direction	5.005	**0.042**	14.726	**0.002**	0.933	0.350
	Sampling period	0.156	0.699	1.001	0.334	0.011	0.919
	Running direction × Sampling period	0.290	0.599	1.971	0.182	0.542	0.474
PM	Influential variables	Elements					
		Component 1		Component 2		Component 3	
		F-value	*p*-value	*F*-value	*p*-value	*F*-value	*p*-value
$PM_{2.5}$	Running direction	73.103	**0.000**	1.063	0.320	0.180	0.678
	Sampling period	0.854	0.371	0.200	0.662	4.658	**0.049**
	Running direction × Sampling period	1.102	0.312	0.063	0.805	3.621	0.078
PM_{10}	Running direction	37.034	**0.000**	0.429	0.523	0.225	0.642
	Sampling period	1.725	0.210	0.182	0.676	4.143	0.061
	Running direction × Sampling period	0.014	0.908	0.074	0.790	0.764	0.397

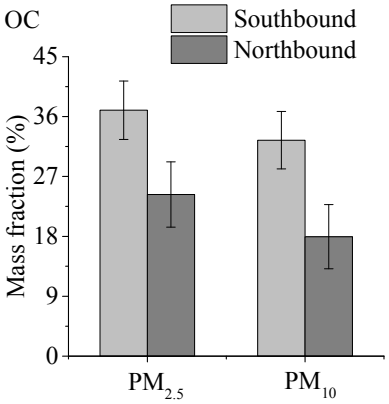

Fig. 6. Mass fractions of OC in the two directions.

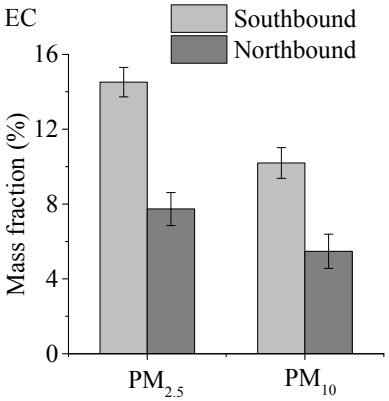

Fig. 7. Mass fractions of EC in the two directions.

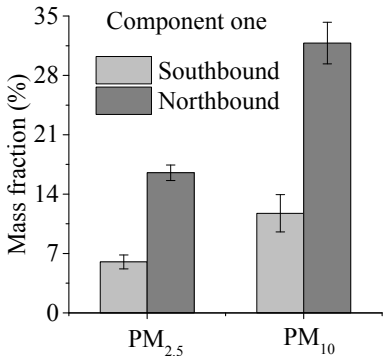

Fig. 8. Mass fractions of elements in Component One in the two directions. (Component One include elements of Na, Mg, Al, K, Ca, Ti, Cr, Mn, Fe, Cu, and Ba).

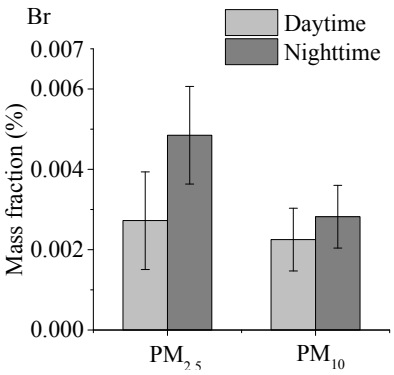

Fig. 9. Mass fractions of Br during the two sampling periods.

volume on the Br content was very weak. Moreover, Br is primarily derived from tailpipe emissions of DV, and the proportions of DV at nighttime were significantly higher than those at daytime (Fig. 5). It can be concluded that the fuel composition in the tunnel was a major factor affecting the content of Br in the PM. In addition, the running direction and the sampling period did not have obvious influences on the contents of secondary ions and elements in Component Two during this test (Table 7).

Comparison of PM Profiles

The mobile source profiles of PM were obtained with a mixed vehicle fleet in this study. The total mass of all the

detected constituents accounted for 92.00% and 84.60% of the total mass of $PM_{2.5}$ and PM_{10}, respectively (Fig. 10). Carbonaceous species were the dominant constituent in the PM (46.06% for $PM_{2.5}$ and 37.07% for PM_{10}), while the contents of OC were higher than those of EC. The mass fractions of elements were 27.73% for $PM_{2.5}$ and 33.36% for PM_{10}. The non-metallic elements were the highest elements in $PM_{2.5}$, while the other metallic elements were the most abundant elements in PM_{10}. Moreover, the mass contributions of secondary ions were the lowest (18.22% for $PM_{2.5}$ and 14.17% for PM_{10}).

Comparison with Other PM Sources

The source profiles of PM obtained in this study were compared with those from the other sources collected in northwestern China (Table 2), including ambient air ($PM_{2.5}$ profile) and road dust (PM_{10} profile; Han *et al.*, 2014; Wang *et al.*, 2015). The contents of OC and EC were significantly higher (2–14 times) than those from ambient air and road dust, which indicated that carbonaceous species were the primary markers of PM from vehicle emissions (Ancelet *et al.*, 2011; Pant *et al.*, 2017). Further, the mass fractions of three secondary ions were approximately 2 times lower than those from ambient air. A possible explanation for this difference was the weak illumination intensity in the tunnel, which partly limited the gas-to-particle conversion (Kang *et al.*, 2002; Lin *et al.*, 2010). The content of Fe was approximately 2 times higher than that from ambient air, which was similar to that from road dust. This implied that Fe primarily originated from the resuspended road dust in this tunnel. In addition, the fractions of P and Zn were up to 2 times higher than those from road dust, while the fractions of Cu and Pb were also slightly higher, indicating that these elements were influenced by passing vehicles, being highly enriched in the tunnel.

Comparison with Other Tunnel Studies

The $PM_{2.5}$ profiles presented in this study were also compared with those from other tunnels in China (Table 2; Cheng *et al.*, 2010; Dai *et al.*, 2015; Cui *et al.*, 2016). For the same species, the mass fractions presented in these tunnel tests were of the same order of magnitude. Meanwhile, considerable differences were also observed, depending on the differences in vehicle type, fuel type or quality, driving conditions, dust in the tunnel environment, etc. Therefore, local mobile source profiles need to be established.

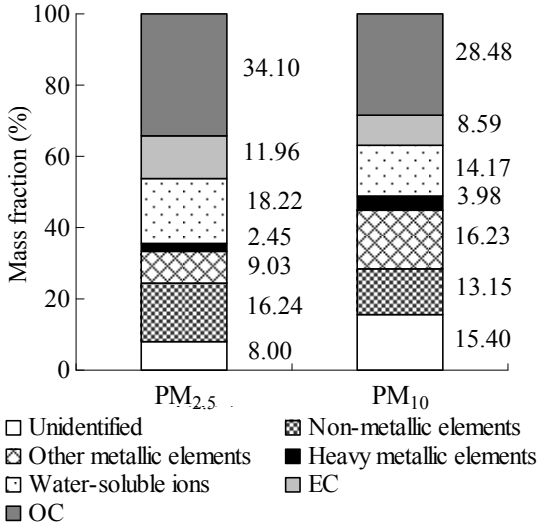

Fig. 10. Mass fractions of PM species in the tunnel.

The content of OC was higher (1.2–2.8 times) than those in the other tunnels, whereas the fraction of EC was similar to the values in the other tunnels, and very close to the value in the Kuixinglou tunnel. Moreover, the contents of the three secondary ions were also higher than those in the other tunnels. It can be attributed to the difference in sampling depths. Most of the sampling sites in this experiment were as deep as 2000 m inside the tunnel in the running direction (Fig. 1), whereas the maximum sampling depths were approximately 1000 m in the other tunnels. Hence, the secondary conversions of OC and ions in this experiment were significantly greater.

For the elements in Component One, the contents of Na, K, and Cr were within the range reported in the other tunnel studies. The fraction of Mg was close to the value in the Wuzushan tunnel. However, the elements of Ti and Cu were less than those in the other tunnels, and the elements of Al, Ca, Mn, and Fe were less than those in all other tunnels, except for the Shing Mun tunnel. These elements primarily originated from resuspended road dust and brake wear according to the previous PCA results. A probable reason was that, the vehicle volume during this tunnel test was less (411 veh h^{-1} on average), with higher vehicle speed (51.84 km h^{-1} on average), whereas a volume of more than 1000 veh h^{-1} was recorded in the other tunnels, with an average speed of 33.40 km h^{-1} in the Zhujiang tunnel. Heavy traffic flow and lower speed would cause frequent braking, leading to a stronger disturbance of the dust on the road surface.

With regard to the elements in Component Two, the contribution of Zn agreed well with the value reported in the other tunnel tests. The content of Pb was similar to the result in the Kuixinglou tunnel. While the fractions of P and S were higher (4–8 times) than the results from the Shing Mun tunnel, it was potentially due to the abundant phosphorus content in the lubricant oil and higher abundance of sulfur in the vehicle fuel during this tunnel test (Lowenthal *et al.*, 1994; Alves *et al.*, 2015a). In addition, the element of Br in Component Three was consistent with the result from Shing Mun tunnel.

CONCLUSIONS

$PM_{2.5}$ and PM_{10} samples were collected in the Qinling No. 1 Tunnel in Xi'an, China. A comprehensive snapshot of the chemical characteristics of the traffic-related PM emissions was generated that included OC, EC, three secondary ions (NH_4^+, NO_3^-, and SO_4^{2-}), and 17 elements (Na, Mg, Al, P, S, K, Ca, Ti, Cr, Mn, Fe, Cu, Zn, Br, Ba, Hg, and Pb). On average, carbonaceous species were the most dominant constituents in the PM, forming mass

contributions of 46.06% to the $PM_{2.5}$ and 37.07% to the PM_{10}, with a higher content of OC than EC. The total mass fractions of the secondary ions were 18.22% for the $PM_{2.5}$ and 14.17% for the PM_{10}, with SO_4^{2-} being the most abundant ion. Further, the summed mass of the 17 elements constituted 27.73% and 33.36% of the total mass of the $PM_{2.5}$ and PM_{10}, respectively, with S being the most abundant element.

The sources of the different elements (i.e., exhaust and non-exhaust) in the PM were identified, and the 17 elements were grouped into 3 components by using PCA combined with EF. Component One comprised the elements Na, Mg, Al, K, Ca, Ti, Cr, Mn, Fe, Cu, and Ba, which were attributed to resuspended road dust and brake wear, whereas Component Two comprised the elements P, S, Zn, Hg, and Pb, which were attributed to vehicle exhaust and tire wear. Finally, Component Three contained only Br, which was mainly derived from the exhaust emissions of DV. Additionally, the emissions of OC, EC, and the elements in Component One were strongly influenced by vehicle driving conditions: The fractions of OC and EC increased with higher engine torque and poorer fuel combustion (when vehicles were ascending and moving at lower speeds), and the fraction of elements increased with frequent braking and the disturbance of road dust (when vehicles were descending and moving at higher speeds). Br, on the other hand, was mainly affected by the vehicle fleet composition, especially the fuel type, resulting in a higher fraction at nighttime because of the higher proportion of DV. However, the secondary ions and elements in Component Two were less influenced by these traffic parameters. Finally, based on a comparison between this study and other tunnel tests, the amount of measured OC and secondary ions may be affected by the sampling depth.

This is the first study to quantify PM profiles from mobile sources by using tunnel methodology in Xi'an in northwestern China. These results complement other studies conducted in various parts of China, improving our comprehension of PM pollution due to road traffic in this country.

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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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