

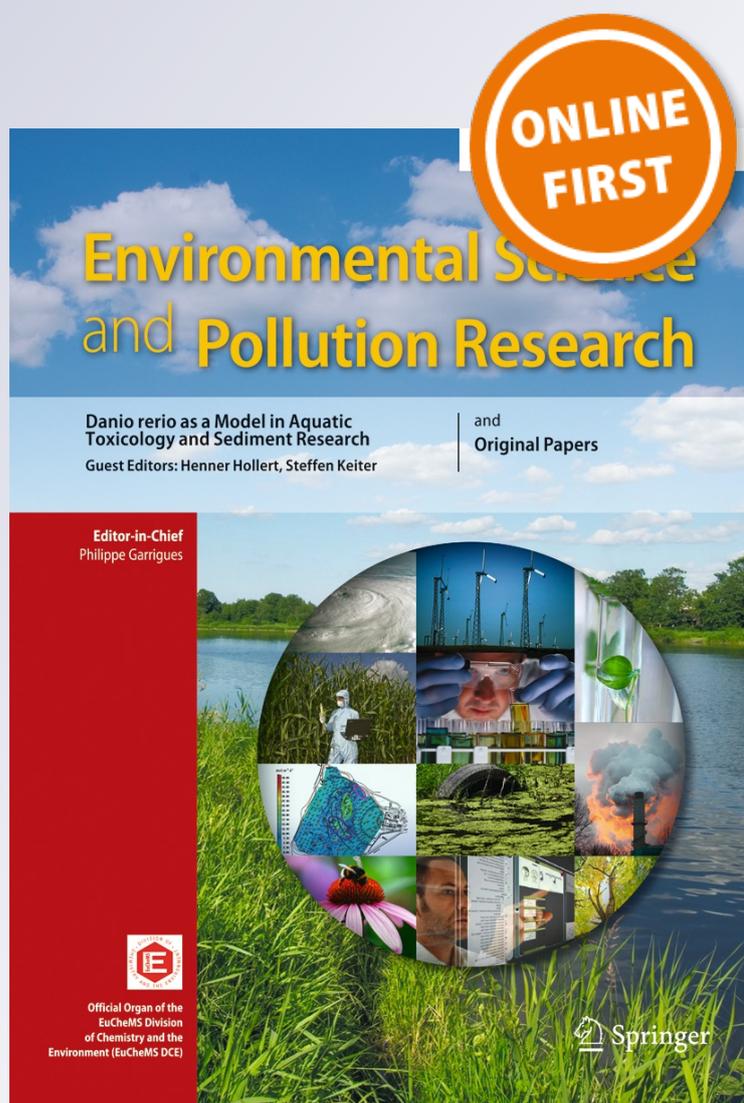
The rural carbonaceous aerosols in coarse, fine, and ultrafine particles during haze pollution in northwestern China

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The rural carbonaceous aerosols in coarse, fine, and ultrafine particles during haze pollution in northwestern China

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Abstract The carbonaceous aerosol concentrations in coarse particle (PM₁₀: Dp≤10 μm, particulate matter with an aerodynamic diameter less than 10 μm), fine particle (PM_{2.5}: Dp≤2.5 μm), and ultrafine particle (PM_{0.133}: Dp≤0.133 μm) carbon fractions in a rural area were investigated during haze events in northwestern China. The results indicated that PM_{2.5} contributed a large fraction in PM₁₀. OC (organic carbon) accounted for 33, 41, and 62 % of PM₁₀, PM_{2.5}, and PM_{0.133}, and those were 2, 2.4, and 0.4 % for EC (elemental carbon) in a rural area, respectively. OC3 was more abundant than other organic carbon fractions in three PMs, and char dominated EC in PM₁₀ and PM_{2.5} while soot dominated EC in PM_{0.133}. The present study inferred that K⁺, OP, and OC3 are good biomass burning tracers for rural PM₁₀ and PM_{2.5}, but not for PM_{0.133} during haze pollution. Our results suggest that biomass burning is likely to be an important contributor to

rural PMs in northwestern China. It is necessary to establish biomass burning control policies for the mitigation of severe haze pollution in a rural area.

Keywords Particulate matter (PM) · Elemental carbon · Organic carbon · Rural area

Introduction

In recent years, haze pollution is extremely severe in China, accompanied by poor visibility and air quality (Huang et al. 2014). Many studies have reported its chemical composition, source apportionment, and radiative effect (Li et al. 2012; Cheng et al. 2013; Wang et al. 2014; Zhang and Mao 2015). Chemical composition showed that carbonaceous aerosol constitutes a major fraction of the urban particles during haze events, which is essential for controlling particle levels and reducing the impacts on the environment and health (Huang et al. 2011, 2014; Zheng et al. 2011). Source apportionment of particulate matter (PM) indicated that the contribution of biomass burning is higher in a rural area compared to that in an urban area (Wang et al. 2006; Zhu et al. 2012). Control of primary particulate emissions and secondary aerosol precursors from fossil fuel combustion and biomass burning could be an efficient strategy for improving air quality in China (Cao et al. 2005, 2012).

According to the Interagency Monitoring of Protected Visual Environments (IMPROVE) protocol of the thermal/optical reflectance (TOR) method, four organic carbon fractions (OC1–4) and three elemental carbon fractions (EC1–3) can be defined (Chow et al. 1993, 2004) and EC can be divided into char (EC1–OP) and soot (EC2+EC3) (Han et al. 2007). Carbon fractions, OC/EC, and char/soot have been applied to identify the source apportionment of particles

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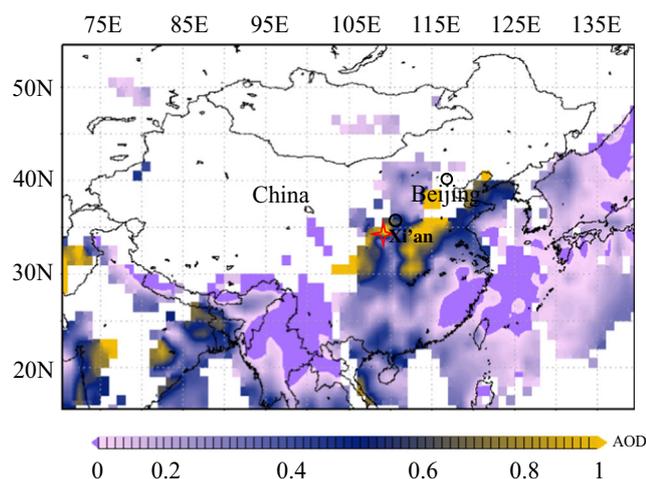


Fig. 1 Geographic location of the rural site (*star*) with haze events of 17–26 January 2014

(Cachier et al. 1989; Cao et al. 2013; Kim et al. 2004; Zhu et al. 2014). Combining the tracer of K^+ , carbon fractions were also inferred as biomass burning tracers for PM (Chuang et al. 2012). The previous studies of carbonaceous aerosols largely focused on the physicochemical characteristics and were mainly performed for urban areas in China (Cao et al. 2012, 2013; Fu et al. 2012; Huang et al. 2012; Zheng et al. 2011). Nevertheless, the investigation is scarce for PM_{10} , $PM_{2.5}$, and $PM_{0.133}$ simultaneously during rural haze events in China. The objective of this study is to reveal the distributions and relationships of carbon fractions and to investigate carbon fractions as biomass burning tracers for PM_{10} ($D_p \leq 10 \mu m$), $PM_{2.5}$ ($D_p \leq 2.5 \mu m$, particulate matter with an aerodynamic diameter less than $2.5 \mu m$), and $PM_{0.133}$ ($D_p \leq 0.133 \mu m$, particulate matter with an aerodynamic diameter less than $0.133 \mu m$) during haze events at a rural site.

Materials and methods

Sample collection

Sampling was conducted at a rural site ($34.12^\circ N$, $108.62^\circ E$) in Huxian County, Shaanxi Province, northwestern China

(Fig. 1). The map presents aerosol optical depth, retrieved from satellite (Terra/Modis) observations during 17–26 January 2014 (<http://www.nasa.gov>). In the monitoring site, it was common for residents to use coal and biomass for cooking and heating in winter. The sampling point was on the rooftop of a private house, which was located at 3 m above the ground level in a village of Huxian County.

PM_{10} , $PM_{2.5}$, and $PM_{0.133}$ samples were collected simultaneously during the high-pollution event of 17–26 January 2014 by using two MiniVol Portable Air Samplers (Airmetrics Inc., 2095 Garden Ave. Suite 102 Eugene, OR 97403, USA) operating at 5 L/min and a personal active nanoparticle sampler (PENS) with 1.5 L/min, respectively (Cao et al. 2005; Tsai et al. 2012). The samples were collected on 47-mm (PM_{10} and $PM_{2.5}$) and 37-mm ($PM_{0.133}$) Whatman quartz microfiber filters (QM/A). All quartz filters were pre-heated at $900^\circ C$ for 3 h and then stored in aluminum foils before sampling. The filters were stored in a refrigerator at about $-20^\circ C$ immediately after sampling to prevent the evaporation of volatile components. The mass concentrations of PM_{10} , $PM_{2.5}$, and $PM_{0.133}$ were determined gravimetrically using a Sartorius MC5 electronic microbalance (Sartorius, Gottingen, Germany) with a $\pm 1\text{-}\mu g$ sensitivity. The filters were analyzed gravimetrically after a 24-h equilibration at a temperature between 20 and $23^\circ C$ and a relative humidity (RH) between 35 and 45 %. The net mass was obtained by subtracting the average of pre-sampling weights from the average of post-sampling weights. Differences among replicate weighings were $<10 \mu g$ for pre-sampling quartz filters and $<20 \mu g$ for post-sampling quartz filters. Field blanks were collected to correct the mass concentrations.

Chemical analysis

All the filters were analyzed for carbon fractions using a DRI Model 2001 Thermal/Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, CA, USA). Carbon fractions were analyzed following the Interagency Monitoring of Protected Visual Environments (IMPROVE-A) thermal/optical reflectance (TOR) protocol (Chow et al. 2007). The method produced data for four OC fractions (OC1, OC2, OC3, and OC4 in a helium atmosphere at 140, 280, 480, and $580^\circ C$, respectively), a

Table 1 The concentrations of mass and carbon fractions for PM_{10} , $PM_{2.5}$, and $PM_{0.133}$

Mean conc. and stdev. ($\mu g m^{-3}$)		OC1	OC2	OC3	OC4	OP	EC1–OP	EC2	EC3	OC	EC	PM conc.
PM_{10}	Ave.	14.2	16.0	23.6	18.9	29.4	4.8	1.0	0.1	102.3	6.0	318.2
	Stdev.	4.6	4.4	5.5	4.8	12.9	1.3	0.2	0.1	24.6	1.5	84.7
$PM_{2.5}$	Ave.	11.9	13.0	19.1	15.0	23.8	4.1	0.5	0.0	82.9	4.6	208.1
	Stdev.	3.5	3.5	4.6	4.9	11.9	1.1	0.1	0.0	20.9	1.1	73.2
$PM_{0.133}$	Ave.	8.9	7.8	15.4	8.5	8.3	0.1	0.3	0.0	48.9	0.4	79.0
	Stdev.	2.3	2.6	6.4	3.7	3.4	0.2	0.1	0.0	18.1	0.3	28.1

pyrolyzed carbon fraction (OP, determined when reflected laser light attained its original intensity after oxygen was added to the combustion atmosphere), and three EC fractions (EC1, EC2, and EC3 in a 2 % oxygen/98 % helium atmosphere at 580, 740, and 840 °C, respectively). The IMPROVE protocol defined OC as OC1+OC2+OC3+OC4+OP and EC as EC1+EC2+EC3-OP. The EC fraction was also divided into char and soot. Char is defined as EC1 minus OP, and soot is defined as the sum of EC2 and EC3 (Han et al. 2009). The analyzer was calibrated with known quantities of CH₄ each day. Replicate analyses were performed once every ten samples. A blank sample was also analyzed in order to correct the sample results. The detailed quality assurance/quality control (QA/QC) procedures have been described elsewhere (Cao et al. 2003; Chow et al. 2011).

A Dionex-600 Ion Chromatograph (Dionex Inc., Sunnyvale, CA, USA) was used to determine the concentration of K⁺. The instrument was equipped with an IonPacCS12A column (20 mM methanesulfonic acid as the eluent) to analyze the cations. The minimum detection limit was 0.001 µg/mL for K⁺. Standard reference materials produced by the National Research Center for Certified Reference Materials (Beijing, China) were analyzed for QA/QC purposes. All of the reported K⁺ concentrations were corrected using the field blanks. The experimental uncertainty was ±0.01 for K⁺. The overall uncertainty in sampling and analysis is reasonable.

Results and discussion

PM concentration and carbonaceous contributions

As shown in Table 1, daily average concentrations of PM₁₀, PM_{2.5}, and PM_{0.133} were 318.2±84.7, 208.1±73.1, and 79.0±28.1 µg m⁻³ during the haze events, respectively. Similar variations of mass and OC were obtained for PM₁₀, PM_{2.5}, and PM_{0.133}; nevertheless, this was not seen for EC. During the sampling period, PM_{0.133} accounted for 17 % to 30 % with an average of 24 % of PM₁₀ and 28 % to 52 % with the average of 38 % of PM_{2.5}. PM_{2.5} accounted for 54 % to 74 % with an average of 64 % of PM₁₀. The results indicated that PM_{2.5} comprised a large fraction in PM₁₀ during the sampling period. It is worthy to note that measurements of filter samples are influenced by both positive and negative artifacts (Watson and Chow 2011). The previous results showed that the evaporation loss of collected particles during a single filter sampling process is severe, accounting for 5.8–36.0 % of PM_{2.5} (Liu et al. 2014, 2015).

The average OC and EC concentrations for PM₁₀ were 102.3±24.6 and 6.0±1.5 µg m⁻³, while those for PM_{2.5} were 82.9±20.9 and 4.6±1.1 µg m⁻³, respectively. The lowest OC and EC concentrations were obtained for PM_{0.133} with 48.9±18.1 and 0.4±0.3 µg m⁻³, respectively (Table 1). The ratios of

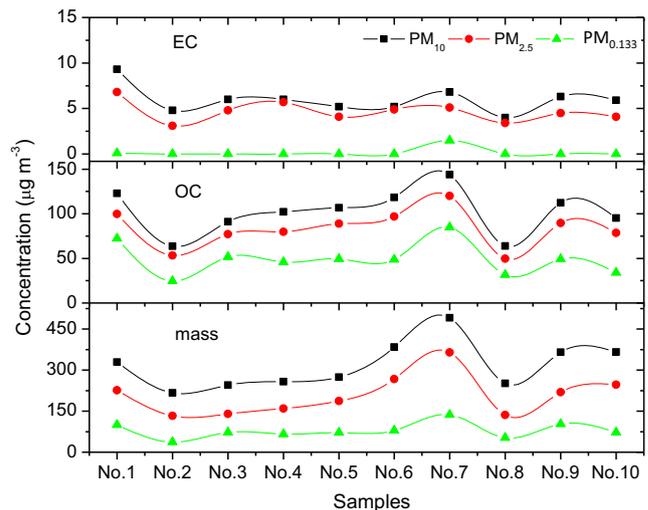


Fig. 2 Variations of mass, OC, and EC for PM₁₀, PM_{2.5}, and PM_{0.133}

maximum OC to minimum OC for PM₁₀, PM_{2.5}, and PM_{0.133} were 2.3, 2.3, and 3.7, respectively, while those for EC were 2.1, 2.2, and 3.5, respectively (Fig. 2). The results indicated the variability for OC and EC were comparable among PM₁₀, PM_{2.5}, and PM_{0.133}.

As shown in Fig. 3, total carbonaceous aerosol (TCA=OC×1.2+EC) contributed 41, 52, and 75 % of PM₁₀, PM_{2.5}, and PM_{0.133}, respectively (White and Roberts 1977; Turpin and Lim 2001). OC accounted for 33, 41, and 62 % of PM₁₀, PM_{2.5}, and PM_{0.133}, whereas EC accounted for 2, 2.4, and 0.4 % of PM₁₀, PM_{2.5}, and PM_{0.133}, respectively. Less than 2.5 % of char and soot resided in PMs was observed. The percentage of TCA in PM₁₀ was lowest among three PM sizes due to higher contributions of geological matter in coarse particles.

Carbon fractions for PM₁₀, PM_{2.5}, and PM_{0.133}

As shown in Table 1, for PM₁₀ and PM_{2.5}, OC3 was the most abundant carbon fraction, followed by OC4, OC2, and OC1 in OC fractions. EC1-OP is dominant in EC fractions. The

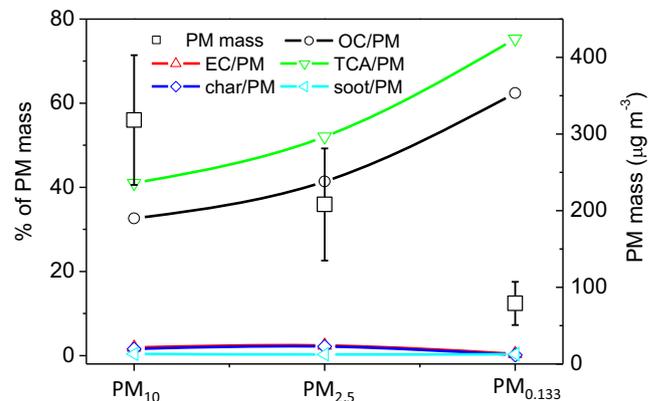


Fig. 3 Contributions of carbon fractions to PMs

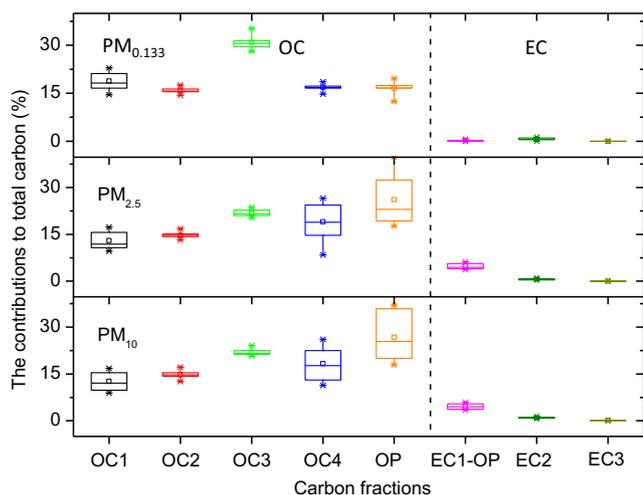


Fig. 4 Percentage contributions of carbon fraction to total carbon

ranking order for $PM_{0.133}$ was $OC3 > OC1 > OC4 > OC2$ and $EC2 > EC1-OP > EC3$, respectively. OP was the most abundant carbonaceous fraction in both PM_{10} and $PM_{2.5}$, indicating a large amount of low-volatility species in OC particles of PM_{10} and $PM_{2.5}$, e.g., humic-like substances as stated by Schmidl et al. (2008). The concentration of EC1-OP was higher than any other elemental carbon fractions in PM_{10} and $PM_{2.5}$, while EC2 was the highest in $PM_{0.133}$. EC3 was almost under the method detection limit in PM_{10} , $PM_{2.5}$, and $PM_{0.133}$. The results showed that char dominated EC in PM_{10} and $PM_{2.5}$ and soot dominated EC in $PM_{0.133}$, which were consistent with the previous study (Zhu et al. 2010). $PM_{0.133}$ OC accounted for 59 % of $PM_{2.5}$ OC, whereas $PM_{0.133}$ EC accounted for 9 % of $PM_{2.5}$ EC. $PM_{2.5}$ OC accounted for 81 % of PM_{10} OC, and that for $PM_{2.5}$ EC to PM_{10} EC was 77 %, which reflected the presence of richer OC and EC content for $PM_{2.5}$. Less contribution of EC was observed in $PM_{0.133}$.

There were distinct differences among PM_{10} , $PM_{2.5}$, and $PM_{0.133}$ samples for carbon fraction contributions (Fig. 4). The contributions of four organic carbon fractions (OC1, OC2, OC3, and OC4) to total carbon (TC=OC+EC) in PM_{10} , $PM_{2.5}$, and $PM_{0.133}$ were comparable, which ranged from 12.7 % to 22 % in PM_{10} , 13 % to 21.8 % in $PM_{2.5}$,

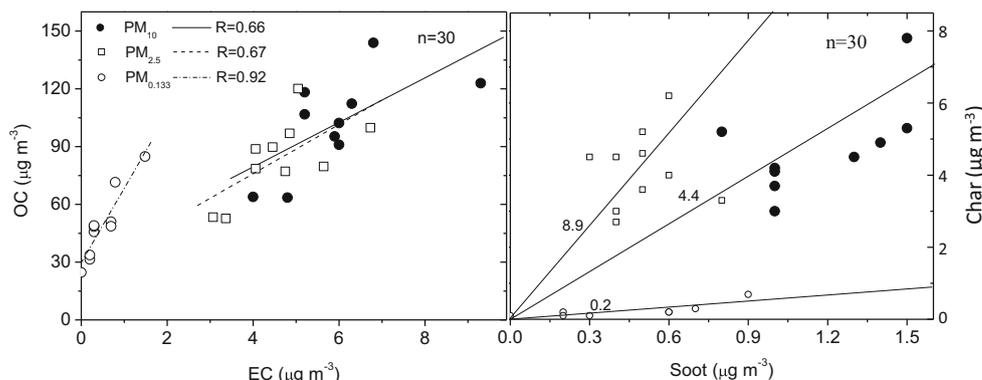
and 16 % to 31 % in $PM_{0.133}$, respectively. EC1-OP accounted for 4.5 % and 4.8 % of TC in PM_{10} and $PM_{2.5}$ samples, higher than that in $PM_{0.133}$ samples (0.2 %). EC2 contributed 1 % for TC in PM_{10} samples, while lower contributions of 0.6 % and 0.7 % were found in $PM_{2.5}$ and $PM_{0.133}$, respectively. The results were consistent with the previous study which showed EC1-OP dominated EC in coarse and fine particulate matter (Zhu et al. 2010).

Correlations of carbon fractions for PM_{10} , $PM_{2.5}$, and $PM_{0.133}$

Figure 5 shows that OC and EC are moderately correlated in PM_{10} ($R=0.66, p=0.03925$) and $PM_{2.5}$ ($R=0.67, p=0.03445$), respectively, indicating that OC and EC for PM_{10} and $PM_{2.5}$ are produced from complicated contributors. The result is different from the carbonaceous content in $PM_{0.133}$, as OC and EC are significantly correlated ($R=0.92, p<0.0001$). Correlations among carbon fractions of PM_{10} , $PM_{2.5}$, and $PM_{0.133}$ are shown as follows. Strong correlations were observed among OC1, OC2, and OC3 for PM_{10} ($R, 0.84-0.99$) and $PM_{2.5}$ ($R, 0.85-0.99$), which attributed to the common contributors of OC1-3. OC4 was not correlated evidently with other fractions for PM_{10} and $PM_{2.5}$, except that between OC4 and EC1-OP for PM_{10} . There was a low correlation between EC2 and EC3, and EC3 and EC1-OP for PM_{10} ($R \approx 0.70$). Low correlations were obtained among other carbon fractions for $PM_{2.5}$. Significant correlations among carbon fractions for $PM_{0.133}$ showed impacts from a combination of common source contributions.

The average ratios of OC to EC were 17.3 and 18.1 for PM_{10} and $PM_{2.5}$, respectively, while those for $PM_{0.133}$ were much higher. The ratios are much higher than those from previous researches (Cao et al. 2007; Chow et al. 2004). The higher OC to EC ratio for $PM_{0.133}$ is due to the lower amount of EC contained in $PM_{0.133}$ and a certain amount of volatile organic compounds, which may have been condensed to become OC after leaving the burning sites (Robinson et al. 2007). These results indicate that OC concentrations at the rural site would be heavily influenced by secondary OC

Fig. 5 Relationships among carbonaceous species



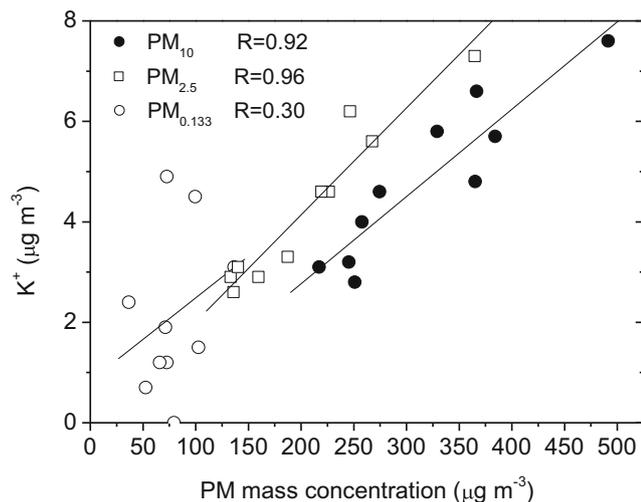


Fig. 6 Correlations among K^+ and PMs

compounds or biomass burning emissions, which can affect the OC/EC ratio significantly.

The ratios of char to soot were 4.4, 8.9, and 0.2 for PM_{10} , $PM_{2.5}$, and $PM_{0.133}$, respectively (Fig. 5). The ratio for $PM_{2.5}$ was similar to the value from biomass burning sources (Cao et al. 2005). For $PM_{0.133}$, the char to soot values vary low because the amounts of char and soot in $PM_{0.133}$ are very low and invariable (char, 0~0.6 $\mu\text{g m}^{-3}$; soot, 0~0.9 $\mu\text{g m}^{-3}$). OC to EC and char to soot ratios among

PM_{10} , $PM_{2.5}$, and $PM_{0.133}$ varied largely, which indicated the effect of PM size distribution on source identification is worth investigating.

Assessment of biomass burning tracers for PM_{10} , $PM_{2.5}$, and $PM_{0.133}$

Aerosol K^+ is the recognized biomass burning tracer (Andreae and Merlet 2001). Therefore, K^+ can be used to assess the capability of carbon fractions as biomass burning tracers (Chuang et al. 2012). The investigations of carbon fractions as biomass burning tracers among PM_{10} , $PM_{2.5}$, and $PM_{0.133}$ were also conducted in the present study. K^+ had a significant correlation with PM_{10} ($R=0.92$, $p<0.0001$) and $PM_{2.5}$ ($R=0.96$, $p<0.0001$), while a poor correlation ($R=0.30$, $p=0.41547$) was found between K^+ and $PM_{0.133}$ (Fig. 6). The results indicated biomass burning emissions were the important contributors to PM_{10} and $PM_{2.5}$ at the rural site. K^+ is a primary product from biomass burning emissions, while the secondary condensation of gaseous precursors contributes largely to $PM_{0.133}$ when biomass burning smoke is cooled as well as other factors (Hata et al. 2014), which contributed to the poor correlation between K^+ and $PM_{0.133}$.

Among the resolved compositions, K^+ is highly correlated with OP ($R=0.91$, $p=0.00026$) and moderately with EC2 ($R=0.75$, $p=0.01$), OC3 ($R=0.74$, $p=0.014$), and OC2 ($R=0.68$,

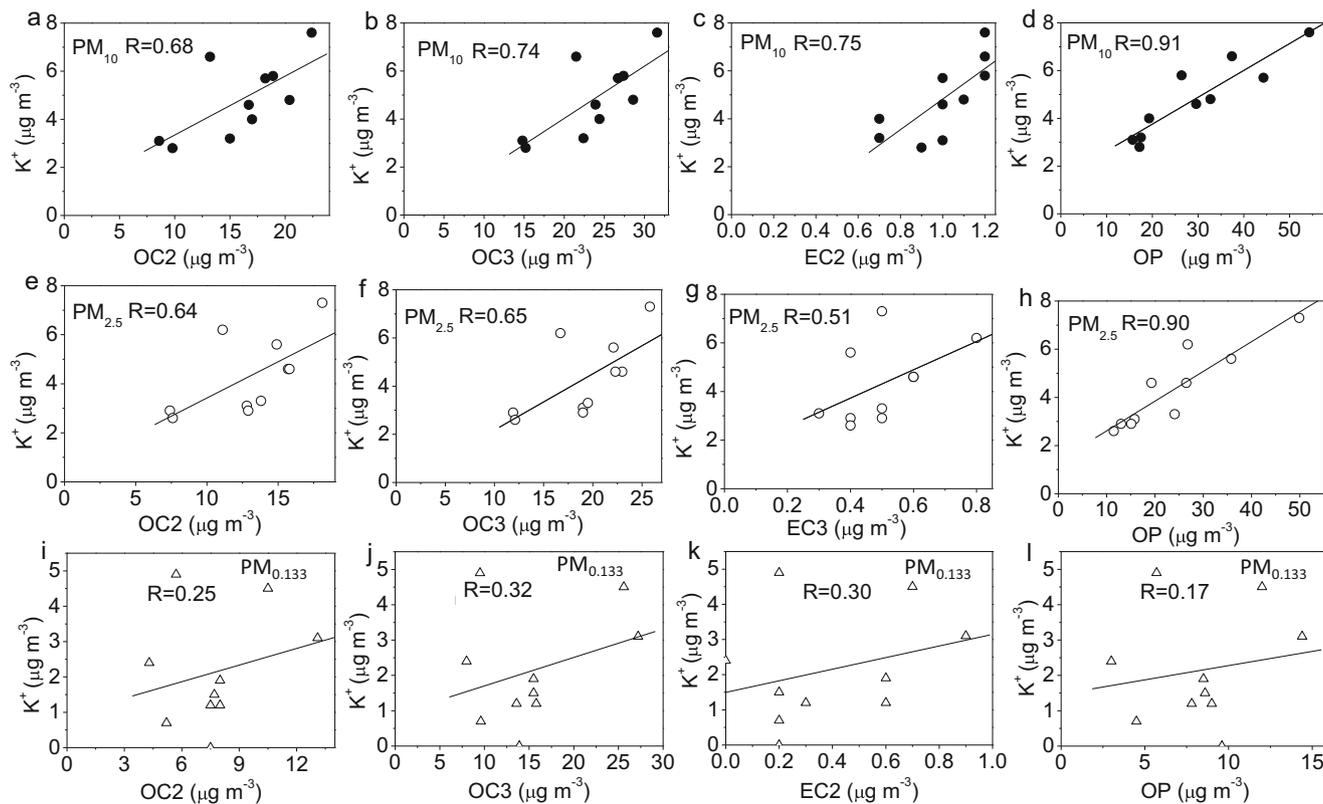


Fig. 7 Correlations among K^+ and carbon fractions

$p=0.03$) in PM_{10} . K^+ is also highly correlated with OP ($R=0.90$, $p=0.00035$) and moderately with OC2 ($R=0.64$, $p=0.04827$) and OC3 ($R=0.65$, $p=0.04057$) in $PM_{2.5}$. Poor correlations among K^+ and $PM_{0.133}$ carbon fractions were observed at the rural site (R , 0.17–0.32), which was consistent with the suggestion discussed in the previous section that secondary condensation of gaseous precursors from biomass burning was an important source of $PM_{0.133}$ in the rural area (Fig. 7).

Based on this assessment, OP, OC3, and EC2 for PM_{10} can be biomass burning tracers; those for $PM_{2.5}$ are OP, OC2, and OC3, but no carbon fraction can be a biomass burning tracer for $PM_{0.133}$. Certainly, there are also some limitations in the study, such as different samplers using, and the uncertainty of the thermal optical analysis.

Conclusions

In this study, the carbon fractions of PM_{10} , $PM_{2.5}$, and $PM_{0.133}$ at a rural site during haze events were determined. The main findings are as follows:

- (1) The concentrations of PM_{10} , $PM_{2.5}$, and $PM_{0.133}$ during the haze events were 318.2 ± 84.7 , 208.1 ± 73.1 , and $79.0 \pm 28.1 \mu\text{g m}^{-3}$, respectively. Given the correlations among K^+ and PMs, the contributions of biomass burning were higher for PM_{10} and $PM_{2.5}$ compared to that for $PM_{0.133}$.
- (2) Higher correlations among carbon fractions were obtained in $PM_{0.133}$ than those in PM_{10} and $PM_{2.5}$, which showed the impacts from a combination of common source contributions for $PM_{0.133}$.
- (3) OC3 and OP were more abundant than other organic carbon fractions in PMs. The present study inferred that K^+ , OP, OC3, and EC2 are good biomass burning tracers for PM_{10} , and those for $PM_{2.5}$ are K^+ , OP, OC2, and OC3. A similar tracer was not observed for $PM_{0.133}$.

This study provides new insights regarding the rural carbonaceous aerosol for coarse, fine, and ultrafine particles during severe haze pollution, and suggests that stringent controls on rural biomass burning activities could be efficient measures to reduce haze pollution in northwestern China.

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