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Chemical source profiles of urban fugitive dust $PM_{2.5}$ samples from 21 cities across China

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HIGHLIGHTS

- Chemical profiles of urban fugitive dust $PM_{2.5}$ were investigated in 21 cities across China.
- Soil dust has a higher contribution in Northern China than in Southern China.
- High Ca/Al ratio was a good marker to distinguish urban fugitive dust from Asian dust.
- Low NO_3^-/SO_4^{2-} indicated that fugitive dust was strongly influenced by stationary sources.
- Urban fugitive dust was more influenced by local sources than long-range transport.

GRAPHICAL ABSTRACT

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ABSTRACT

Urban fugitive (road and construction) dust $PM_{2.5}$ samples were collected in 21 cities of seven regions in China. Seven water-soluble ions, eight sub-fractions of carbonaceous components, and 19 elements were determined to investigate the chemical profiles of urban fugitive dust. Among the analyzed chemical compositions and on regional average, the elemental compositions showed the highest proportion (12.5–28.9% in road dust (RD) and 13.1–38.0% in construction dust (CD)), followed by water-soluble ions (5.1–19.0% in RD and 4.2–16.4% in CD) and carbonaceous fractions (5.4–9.6% in RD and 4.9–9.3% in CD). Chemical compositions measured in CD were all slightly lower than those in RD although statistically insignificant ($p > 0.05$). Soil dust, which was estimated from Fe concentration, was proved to be the biggest contributor to urban fugitive dust $PM_{2.5}$ mass. While, it showed a higher contribution in Northern China (71.5%) than in Southern China (52.1%). Higher enrichment factors were found for elemental S, Zn and Pb in RD than CD, reflecting stronger anthropogenic sources (i.e. vehicle exhaust) in RD. Low NO_3^-/SO_4^{2-} and high SO_4^{2-}/K^+ ratios both indicated that fugitive dust was strongly

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influenced by stationary sources (e.g. coal combustion), and this influence was especially strong in Northern China. Coefficients of divergence proved that dust profiles within the same region were more similar than across regions, reflecting that urban fugitive dust was influenced more by local sources than long-range transport.

1. Introduction

Fugitive dust particles from soil (Arimoto et al., 2006; Cao et al., 2008), paved and unpaved roads (Chow et al., 1992; Ho et al., 2006), construction (Chow et al., 2003) and cements (Kong et al., 2011b; Vega et al., 2001) contributed significantly to urban particulate matter (PM) pollution, ranking in importance only after fuel combustion and secondary organic aerosol formation (Chow et al., 2003; Kong et al., 2011a), particularly in arid and semi-arid regions (Shen et al., 2016; Tao et al., 2017). Fugitive dust contributed 12%–34% of winter $PM_{2.5}$ mass and 17%–32% of summer $PM_{2.5}$ mass in 14 Chinese cities (Cao et al., 2012). As a result of rapid urbanization in China, the total length of the road network and total area under construction are expanding at an accelerated pace, which leads to increasing emissions of fugitive dust in cities (Jiang et al., 2017).

Road dust (RD) and construction dust (CD) are the main types of fugitive dust in urban areas. RD is produced from main streets and roads by winds and traffic (Chow et al., 2003; Ho et al., 2003) while CD is originated from construction activities surrounding building sites (Kong et al., 2011a; Vega et al., 2001). The chemical profiles of fugitive dusts generally include carbonaceous components, water-soluble ions (WSI) and various chemical elements. Organic carbon (OC) and elemental carbon (EC) comprise 1%–20% and ~1%, respectively, in dust samples (Kong et al., 2011a), and WSI such as Cl^-, NO_3^-, SO_4^{2-}, Na^+, K^+, Mg^{2+} and Ca^{2+} comprise ~10% in total (Shen et al., 2009b). Elemental components including microelements such as Si, Al, Mg, Ca, and Ti (>1%) and heavy metals such as Ni, Cu, Zn and Pb (<1%) have also been found in fugitive dust (Arimoto et al., 2004).

Distinctive climate conditions exist between Northern and Southern China due to separation of the regions by the Qinling Mountains and Huaihe River and between eastern coast and western plateau because of the unique geographic characteristics (Qian and Lin, 2005). Dry climate in Northern China, especially in fall and winter, is conducive to fugitive dust emissions (Cao et al., 2012). The unbalanced regional developments in China also lead to different regional environmental policies, which in turn can cause different situations for fugitive dust emissions (Ning et al., 1996; Zhang et al., 2010).

The chemical profiles of fugitive dusts have been investigated in several cities in China (Han et al., 2007; Bi et al., 2017). Regional variations in chemical profiles and compositions of fugitive dust have been briefly discussed (Cao et al., 2012; Yao et al., 2002; Zhao et al., 2011), keeping in mind that different sampling methods and analytical equipment were used in different studies, which may cause large uncertainties in statistical comparisons between these studies (Chow et al., 2003; Ho et al., 2003). An integrated research on urban fugitive dusts over the whole China is thus needed to effectively minimize the uncertainties of dust source profiles, which in turn will improve the accuracy of source apportionment analysis results (Cao et al., 2005b; Lee et al., 2008; Watson et al., 2002).

In this study, 21 cities were selected from seven typical regions of China (Northwest, North, Northeast, Central, South, Southwest, and East China) for fugitive (road and construction) dusts experiments. Major chemical components in $PM_{2.5}$ samples including carbonaceous components, water-soluble ions and elemental compositions were analyzed. The objectives of this study are to: 1) build integrated source profiles of urban fugitive dust for the seven selected geographical regions in China, and 2) identify regional characteristics, variations and similarities in fugitive dusts. Knowledge gained in this study is expected to improve future source origins studies on fugitive dusts and to provide scientific evidence for making urban fugitive dust control policies.

2. Methodology

2.1. Study area

The 21 cities selected in this study include three cities (Xi'an, Lanzhou, and Yinchuan) in Northwest China (NWC), six (Beijing, Tianjin, Baoding, Shijiazhuang, Handan and Taiyuan) in North China Plain (NCP), three (Harbin, Changchun, and Shenyang) in Northeast China (NEC), two (Wuhan and Changsha) in Central China (CC), two (Guangzhou and Xiamen) in South China (SC), three (Chongqing, Chengdu and Kunming) in Southwest China (SWC), and two (Nanjing and Shanghai) in East China (EC) (see Fig. S1 in Supplementary materials). The several regions listed above can generally be categorized as in Northern (NWC, NCP and NEC) and Southern (CC, SC, SWC and EC) China for easy discussion below.

2.2. Sample collection and pre-treatment

Two types of fugitive dusts, RD and CD, were collected in every city using consistent method and laboratory analysis. A total of 185 sets of RD and CD samples were collected from the 21 cities on non-rainy days from March 2014 to July 2015. The dust samples were collected by in situ resuspension, that is, the dust on construction ground and road surfaces was manually swept using a broom, which simulated the working condition of mechanical road cleaners (Fig. S2). $PM_{2.5}$ (particulate matter with aerodynamic diameter < 2.5 μm) mini-volume samplers (Airmetrics, Springfield, OR, USA) were employed to collect the dust samples at a flow rate of 5 $L \cdot min^{-1}$. Each sampling period ranged from 30 to 60 min according to different dust loadings at the sampling sites. One set of $PM_{2.5}$ samples was collected on 47-mm diameter polytetrafluoroethylene (Teflon®) membrane filters ($PM_{2.5}$ Air Monitoring PTFE Filters, Whatman Limited, Maidstone, UK) for elemental analysis. Another set was collected on 47-mm quartz microfiber filters (Whatman Limited, Maidstone, UK) for gravimetric, water-soluble ions and carbonaceous composition analyses (Cao et al., 2012; Shen et al., 2009b). The quartz filters were preheated at 800 °C for 3 h and then cooled before use.

2.3. Chemical analysis

Gravimetric analysis of dust mass loadings on Teflon filters was determined using a Sartorius MC5 electronic microbalance (±1 μg sensitivity, Sartorius, Gottingen, DE). The filters were weighed after 24-h equilibration at temperatures between 20 and 23 °C and relative humidity between 35% and 45% following the United States Environmental Protection Agency methodology (Chow and Watson, 1998). Each filter was weighed at least four times before and after sampling following the 24-h equilibration period. The mean net mass for each filter was obtained by subtracting the average pre-deployment weight from the average of the post-sampling weights.

Three main chemical components were analyzed for dust samples, namely carbonaceous fractions (including 8 sub-fractions), seven water-soluble ions and 19 elemental compositions. One-fourth of each quartz filter sample was used to determine major ion concentrations. Three anions (Cl^-, NO_3^- and SO_4^{2-}) and four cations (Na^+, K^+, Mg^{2+}

and Ca^{2+}) in dust samples were determined with ion chromatography (IC, Dionex 500, Dionex Corp., Sunnyvale, CA, USA). Detailed information about the pretreatment procedure for ion measurement was given by Shen et al. (2016). A 0.5 cm^2 patch from each quartz filter was analyzed for eight carbon fractions following the Interagency Monitoring of Protected Visual Environments thermal/optical reflectance protocol (Cao et al., 2003; Chow and Watson, 1998) on a DRI model 2001 carbon analyzer (Atmoslytic Inc., Calabasas, CA, USA). The detailed methodology has been described previously (Cao et al., 2007; Shen et al., 2011). For elemental analysis, each collected Teflon® filter was analyzed for up to 19 elements (Si, Al, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Rb, Sb, Ba, and Pb) by energy dispersive X-ray fluorescence spectrometry (Epsilon 5 ED-XRF, PANalytical B.V., Almelo, Netherlands). Each sample was analyzed for 30 min to obtain a spectrum of X-ray counts versus photon energy, with the individual peak energies matching specific elements, and peak areas corresponding to elemental concentrations (Cao et al., 2012; Xu et al., 2012).

2.4. Data processing

2.4.1. Coefficient of divergence

Coefficient of divergence (COD), a self-normalizing parameter, is used to accurately describe the inter- and intra-regional similarities in the two types of fugitive dust profiles.

The COD was calculated using Eq. (1):

$$COD_{jk} = \sqrt{\frac{1}{p}\sum_{i=1}^{p}\left(\frac{x_{ij}-x_{ik}}{x_{ij}+x_{ik}}\right)^2} \quad (1)$$

where, x_{ij} represents the average concentration for a chemical component i at site j; j and k represent two sampling sites; and p is the number of chemical components (Feng et al., 2007; Zhang et al., 2014a). Generally, a COD value approaching zero meant the two samples were similar for the measured chemical species. Otherwise, the two samples were deemed as different when the COD value approached unity.

2.4.2. Enrichment factor

Enrichment factors (EFs) were calculated to investigate whether the origin of elemental constituents was a natural or anthropogenic source. Fe was used as a reference element, and the compositions of the earth's upper continental crust (UCC) were used, as reported by Taylor and McLennan (1995). EFs were calculated using Eq. (2):

$$EF_x = (X/Fe)_{samples}/(X/Fe)_{crust} \quad (2)$$

where EF_x means EF of element X in dust samples; $(X/Fe)_{samples}$ means the proportions of elements X and Fe in samples and $(X/Fe)_{crust}$ means the proportions of elements X and Fe in UCC. Generally, if the EF of an element was <5, the element was deemed to have originated from UCC (Zhang et al., 2002), and if the EF was >5, the element was considered to be contributed by anthropogenic sources (Cesari et al., 2012).

2.4.3. Soil dust contribution

Abundance of element Fe could be used to estimate the contribution of soil-derived dust in fugitive dust according to Eq. (3):

$$C_{soil\ dust} = C_{Fe}/4\% \quad (3)$$

where $C_{soil\ dust}$ means the contribution from soil dust (%) and C_{Fe} means the elemental Fe fraction in samples (%).

3. Results and discussion

3.1. Water-soluble ions

3.1.1. Road dust

The measured average mass factions and standard deviations of WSI in RD samples from the 21 cities are listed in Table S1a. The proportion of WSI in RD $PM_{2.5}$ in any given cities varied from 5.1% to 19.0%. The highest WSI proportion in RD was observed in Yinchuan (19.0%), a provincial capital city in NWC, while the lowest was in Shijiazhuang (5.1%), a provincial capital city in NCP. Variations in WSI proportions in RD between different cities could be over a factor of 3.0 which is statistically significant ($p < 0.05$). The huge difference may be due to the unique characteristics of local soil, energy structure, climatic conditions and other relative sources (Tian et al., 2017; Yu et al., 2017).

Regional differences in WSI proportions in RD samples could be over a factor of 2.0 between the seven regions, with a descending order in WSI proportion of CC > NWC > SWC > EC > SC > NCP > NEC (Fig. 1). However, due to the large variations between the cities in the same region, the regional differences were not statistically significant ($p > 0.05$). Another obvious phenomenon identified in Fig. 1 was that the abundance of cations were dramatically higher than those of anions, implying that the RD samples were mostly alkaline.

Among the seven measured ion species, Ca^{2+} was the most abundant ion and accounted for 59.5% of the total WSI if averaging all the samples together, followed by Na^+ (17.9%) and SO_4^{2-} (10.4%). The usage of calcium-rich building materials (i.e., cement) on paved roads was likely one of the major causes for the high contents of Ca^{2+} (Zhang et al., 2014a). As a crucial natural source of Na^+, sea salts are not negligible in Southern Chinese cities (Lai et al., 2007). As a typical

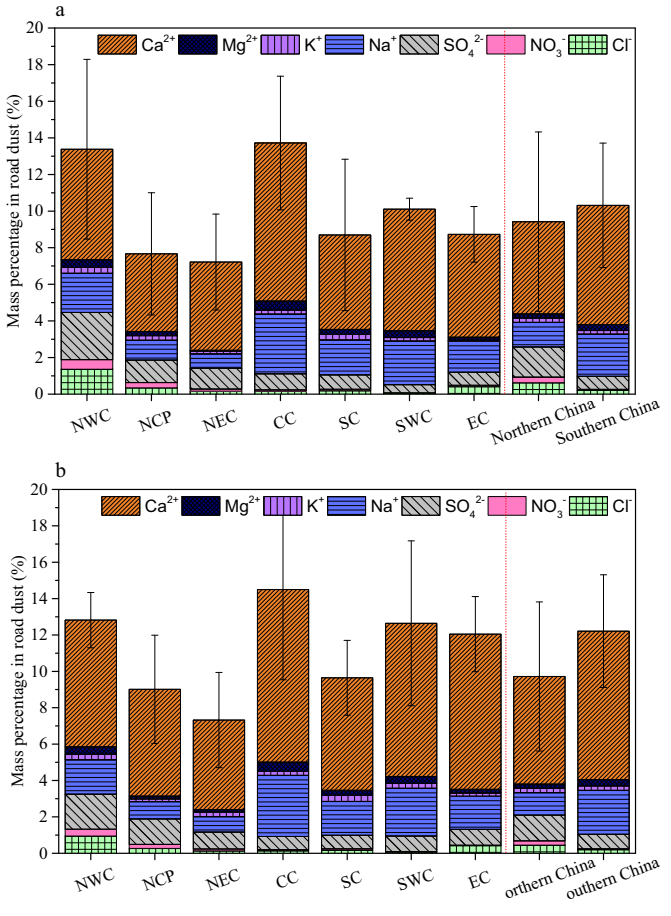

Fig. 1. Water-soluble ions distribution in road dust (a) and construction dust (b) samples.

ion from anthropogenic sources (Shen et al., 2009b; Wang et al., 2006; Yao et al., 2002; Yatkin and Bayram, 2008), SO_4^{2-} showed twice higher contribution in samples from Northern China (17.5% in total WSI) than in those from Southern China (6.9%), mainly due to the high coal consumption in Northern China. It is not surprised that samples from Kunming (a famous tourist city in SWC) had the lowest SO_4^{2-} content in total WSI (3.2%) among the 21 selected cities. The proportion of Cl^- in RD samples also showed a significant difference among individual cities, especially the extremely high Cl^- content in samples from NWC (Xi'an, Yinchuan and Lanzhou). Besides contributions from coal combustion and biomass burning sources, the high Cl^- content in NWC dust can also be ascribed to the saline-alkaline soils in NWC. Wind-eroded soil dust with high Cl^- concentration in NWC can be transported easily to urban areas and lead to relatively high Cl^- proportions in fugitive dust (Suhayda et al., 1997).

WSI proportion in RD from Southern China was ~1.0% higher than that from Northern China but the difference was not statistically significant ($p > 0.05$). As for the individual ions in WSI, an obvious difference was observed in that SO_4^{2-} proportion was doubled while Na^+ proportion was only half in Northern China than in Southern China, indicating a higher alkalinity in RD from Southern China.

3.1.2. Construction dust

The proportion of WSI in CD $PM_{2.5}$ in any given cities varied from 4.2% to 16.4%, which was slightly lower than those in RD (Table S1b). The highest WSI proportion in CD was also observed in Yinchuan (16.4%) as was for RD, but the lowest was in Shenyang (4.2%), differing from that for RD in Shijiazhuang. As shown in Fig. 1b, regional differences in the WSI proportion in CD were similar to those in RD. Large but statistically insignificant ($p > 0.05$) regional differences were identified. The high proportions of cations in CD than RD ionic compositions may imply higher alkalinity in CD. WSI proportions were ~2% higher in Southern than Northern China although the differences were not statistically significant ($p > 0.05$). The differences in ionic composition between RD and CD in the same region were not as obvious as those between Northern and Southern China, suggesting large-scale urban dust research should focus on interregional differences of WSI components.

Similarly to RD ionic compositions, Ca^{2+} was absolutely the dominant ion species in CD with a mean fraction of 64.6%, which was even higher than that in RD, possibly due to the usage of calcium-rich building materials (i.e., cement and gravels) at construction sites (Ho et al., 2003; Zhang et al., 2014a, 2014b). SO_4^{2-} was the dominant anion in CD (10.2% in WSI) and its higher abundance in Northern cities reflecting the large consumption of coal (Zhang et al., 2017). CD from the three NWC cities also showed dramatical high Cl^-, reflecting that CD would also be affected by local sources such as saline-alkali soil and biomass burning emissions (Shen et al., 2009b; Shen et al., 2011; Tian et al., 2011).

3.2. Carbonaceous fractions

3.2.1. Road dust

The means and deviations of mass fractions for total carbon (TC), OC, EC and eight sub-fractions in RD are listed in Table S2a. Generally, Abound TC was found in RD with its mass fraction ranging from 5.4% to 9.6%, somewhat lower than that of the total WSI. The highest and lowest TC fraction in RD were observed in Yinchuan and Changchun (a provincial capital city in NEC), respectively. Within TC, OC showed a much higher proportion than EC in RD.

On regional scale (Table 1), TC fraction in RD followed the sequence of NWC > CC > NCP > EC > SWC > SC > NEC. The interregional differences in OC and EC proportions were also obvious for urban RD with the highest OC fraction (5.1%) in NWC and the lowest OC fraction (7.1%) in NEC. Overall, OC fractions in urban RD samples were higher in Northern than Southern China while the opposite trend was found for EC fractions, although the difference is not significant ($p > 0.05$). EC fractions were much lower than those of OC and OC/EC ratios also varied a lot (2.55–6.85), both indicating that OC and EC in urban dust may be derived from different sources (Xie et al., 2012).

To characterize the distribution of carbonaceous fractions in the dust samples, the carbonaceous fractions in regional fugitive dust samples, as well as their diagnostic ratios, were compared with those described in relevant literature as shown in Table 1. The TC levels in urban dust in the present study were one order of magnitude lower than those from roadside samples, and markedly lower than US country RD (Watson et al., 2001). OC and EC fractions were consist with those reported in Fushun (Kong et al., 2011a). In most cases, TC was dominated by OC, as indicated by the high average OC/TC ratio (0.72–0.83). It was widely reported that carbonate carbon was abundance in fugitive dust samples (Cao et al., 2005a; Shen et al., 2007; Shen et al., 2009a). OC/EC ratios in soil dust and cement were all dramatically high due to the low content of EC, while the relative high EC abundance in urban fugitive dust samples indicated that there should be additional EC-rich sources contributing to urban dust. A similar situation was also found for OC because the OC fractions in soil dust and cement were both lower than those in urban dust samples in this study (Watson et al., 2001; Ho et al., 2003).

3.2.2. Construction dust

TC fractions in CD samples ranged from 4.9% to 9.3%, with the highest and lowest values observed in Changsha (a provincial capital city in CC) and Handan (a prefecture level city in NCP), respectively. These cities with the highest and lowest values were different from those found in RD samples, reflecting different sources of carbonaceous fractions between RD and CD samples. Moreover, the variations of TC fractions in CD among the different cities were lower than those in RD, which may be due to lower influence from anthropogenic sources on CD than RD (Bi et al., 2007; Shen et al., 2016).

The regional patterns in TC fractions in CD resembled those in RD, although the mean TC fraction value was ~1.0% lower in CD than RD due to lower OC and EC. Specifically, OC fractions were 5.6% and 5.4% in CD compared to those of 6.4% and 5.8% in RD in Northern and Southern China, respectively, while the corresponding numbers for EC were 1.1% and 1.4% in CD versus 1.5% and 1.8% in RD. Watson et al. (2001) also found that OC fractions followed a descending order of urban RD > country RD > urban CD. One explanation was that OC-rich particles emitted from vehicle exhaust could enhance OC contents in RD (Watson et al., 2001). The slightly lower EC fractions in CD than RD suggest that CD suffered less influence from vehicle emissions than RD (Ho et al., 2003; Kong et al., 2011a). OC/EC ratios were on average higher in CD than RD due to less EC abundance or enriched cements with high OC/EC ratio in CD (Chow et al., 1994; Ho et al., 2003; Kong et al., 2011a). In addition, the OC/EC ratios showed an obvious difference between samples from Northern and Southern China for both RD and CD, a phenomenon that is potentially useful for identifying sources contributing to regional fugitive dusts and associated long-range transport passways.

3.3. Elemental composition

3.3.1. Road dust

Table S3 shows the elemental chemical profiles of the two types of dust samples from the 21 cities. The average proportion of the total measured elements in RD was 19.1% and ranged from 12.5% to 28.9% for individual cities with the highest one in Xi'an and the lowest one in Chongqing. Ca, Si, Fe, and Al were the most abundant elements in most samples. Si and Ca were comparable and dominant, reflecting combined influence by building materials and soil dust (Yatkin and Bayram, 2008; Zhang et al., 2014a).

Fe accounted for 2.0% of RD mass and 12.5% of the total elements. The fraction of soil-derived dust in fugitive dust was estimated from that of Fe according to Eq. (3) (Table 2). Overall, soil dust contributed more than half of the total mass in RD. RD Samples from Northern

Table 1
Carbonaceous fractions and ratios in road dust (RD), construction dust (CD) and similar studies in literature ($\mu g \cdot g^{-1}$).

City	Type	TC	OC	EC	OC/EC	OC/TC	Reference
NWC	RD	93,089 ± 3456	76,766 ± 4518	16,324 ± 7022	4.70	0.82	This study
NCP	RD	78,035 ± 10,143	64,975 ± 7360	13,060 ± 3796	4.98	0.83	
NEC	RD	65,928 ± 18,229	50,835 ± 16,843	15,093 ± 4830	3.37	0.77	
CC	RD	92,279 ± 10,136	71,417 ± 7987	20,862 ± 2149	3.42	0.77	
SC	RD	67,100 ± 12,517	55,041 ± 343	12,058 ± 12,861	4.56	0.82	
SWC	RD	68,172 ± 3249	51,719 ± 2254	16,453 ± 5375	3.14	0.76	
EC	RD	76,229 ± 905	54,731 ± 7789	21,498 ± 8693	2.55	0.72	
Northern China	RD	79,018 ± 13,607	64,192 ± 12,983	14,825 ± 1648	4.33	0.81	
Southern China	RD	75,945 ± 11,627	58,227 ± 8920	17,718 ± 4390	3.29	0.77	
NWC	CD	81,980 ± 1909	71,532 ± 1429	10,448 ± 3277	6.85	0.87	
NCP	CD	60,919 ± 10,694	51,611 ± 6223	9308 ± 5507	5.54	0.85	
NEC	CD	59,409 ± 6750	44,745 ± 6596	14,664 ± 334	3.05	0.75	
CC	CD	79,569 ± 18,960	62,963 ± 11,446	16,606 ± 7514	3.79	0.79	
SC	CD	61,237 ± 10,476	49,924 ± 2241	11,313 ± 12,717	4.41	0.82	
SWC	CD	63,603 ± 4623	50,979 ± 10,271	12,624 ± 11,375	4.04	0.80	
EC	CD	68,420 ± 10,915	51,704 ± 2241	16,716 ± 8675	3.09	0.76	
Northern China	CD	67,436 ± 12,618	55,962 ± 13,914	11,473 ± 2821	4.88	0.83	
Southern China	CD	68,207 ± 8143	53,892 ± 6091	14,315 ± 2762	3.76	0.79	
US[1]	Roadside motor vehicle exhaust	722,810 ± 174,650	598,750 ± 124,060	208,950 ± 55,800	2.86	0.83	Watson et al. (2001)
US[1]	Country road dust	117,370 ± 44,360	113,540 ± 42,260	3830 ± 2100	29.64	0.97	
US[1]	Soil dust	36,290 ± 17,280	36,180 ± 16,600	110 ± 680	328.91	0.99	
Fushun[2]	CD	54,352 ± 19,191	29,500 ± 9316	24,852 ± 9785	1.19	0.54	Kong et al. (2011a)
Fushun[2]	RD	72,786 ± 56,252	34,664 ± 35,815	21,268 ± 20,152	0.90	0.47	
Hong Kong[3]	Cement	14,376 ± 4683	13,232 ± 4035	1154 ± 955	11.47	0.92	Ho et al. (2003)

China showed ~20% higher soil dust contribution than those from Southern China, indicating that Northern cities were more influenced by soil dust, which could be explained by the differences in climatic conditions, e.g., exposed soil was easily suspended by wind erosion in late fall, winter and early spring in Northern China (Zhao et al., 2006). NWC was reported to be frequently affected by desert dust and soil dust (Alfaro et al., 2003; Cao et al., 2008; Zhang et al., 2014b), and in the present study RD from NWC was most influenced by soil dust (84.1%).

The average fractions of heavy metal elements in RD were very low, e.g., below 0.1% for Pb and Zn and below 0.01% for Ni, Cu, Co, Cr and V. Some of the heavy metals could be deemed as tracers of anthropogenic sources, e.g., Pb as a marker for motor vehicle exhaust (Suhayda et al., 1997; Zhang et al., 2012) or coal combustion (Xu et al., 2012). EFs were frequently used to distinguish natural and anthropogenic sources. EFs were calculated according to Eq. (2) and the results are shown in Fig. 2. In RD, EFs of Si, Al, K, Ti, V, Cr, Mn, Fe, Ni, and Cu were < 5, indicating that these elements originated from the UCC. In contrast, EFs for S, Cl, Zn and Pb were much >5, indicating their anthropogenic sources such as coal burning, vehicle exhaust and residues from tire brakes (Ho et al., 2003; Xu et al., 2012; Zhang et al., 2012). Element Ca showed an EF value around the threshold, indicating its mixed natural and anthropogenic sources. Elemental Cl showed a markedly higher EF in NWC than in the other regions, which could be partly due to the huge amount of saline and alkaline lands in this region because coal consumption and biomass burning in this region were similar to other northern regions. Cl^- is abundant in saline-alkaline soils with its concentration up to 179 meq/L in soil in NWC (Suhayda et al., 1997). Wind-eroded soil dust in NWC with high Cl^- concentration can be easily transported to urban areas and lead to relatively high EFs of elemental Cl in fugitive dust (Shen et al., 2016).

Table 2
Soil dust contribution to road dust and construction dust in different regions in China.

Area	Road dust		Construction dust	
	Soli contribution (%)		Soli contribution (%)	
	AVG	SD	AVG	SD
NWC	84.1	17.5	80.8	12.5
NCP	64.5	3.9	68.6	10.5
NEC	65.5	14.1	75.6	11.5
CC	45.7	14.1	46.5	2.4
SC	52.1	16.4	58.0	20.2
SWC	49.7	14.7	71.8	18.6
EC	54.5	10.4	29.3	6.6
Northern China	69.6	13.2	73.4	11.5
Southern China	50.4	11.7	53.7	20.8

3.3.2. Construction dust

The mean proportion of the total elements was obviously higher in CD (23.3%) than RD (19.1%) (Table S3b). The fraction of elements in CD ranged from 13.1% to 38.0% for individual cities with the highest value in Xi'an and the lowest in Chongqing. The regional patterns of the elements fractions in CD followed the descending order of NWC > NEC > NCP > EC > SC > CC > SWC. Different from that in RD, Ca was the most abundant element in CD in nearly all the cities with mass fractions ranged from 5.3% to 16.6%. The results were actually slightly lower than previously reported (Chow et al., 1992; Yatkin and Bayram, 2008), likely caused by the specific characteristics of local CD. In fact, CD samples were collected from different construction sites at different phases of the study, which may have led to various ratios of cement, soil and grit in the aggregate of CD samples.

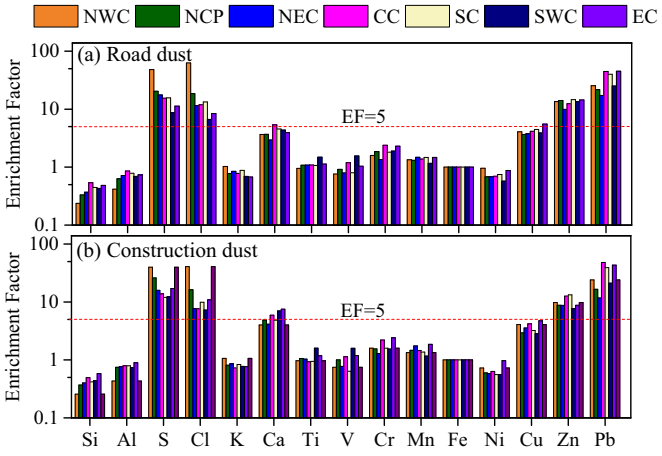

Fig. 2. Enrichment factors of abundant elements in seven regions in China.

The estimated contributions from soil dust to CD in different regions resembled those to RD (Table 2). Soil contribution to CD was ~20% higher in Northern than Southern China, similar to the case of RD, but with larger variations than those of RD, which may be due to the different construction materials used in different phases.

Markedly lower average fractions of some heavy metal elements (especially Pb and Zn) were found in CD than RD, providing additional evidence that RD suffered more from anthropogenic emissions such as vehicle exhaust and tire brake. For elements from natural sources, EFs values in CD samples were similar to those in RD. However, for anthropogenic dominant elements such as S, Cl, Zn and Pb, their EFs were relatively lower in CD than RD. In addition, element Ca had higher EFs in CD than RD and was also around the threshold value, reflecting the massive usage of calcium-rich materials at construction sites.

3.4. Comparability of chemical profiles of fugitive dust among cities and regions in China

Similarities in chemical profiles between RD and CD samples were obvious because more than half of the RD and CD masses were originated from soil dust. For instance, Ca^{2+} was the dominant species in all dust samples and comprised ~50% of WSI. Due to the rich existence of carbonate, all dust samples were calculated to be alkaline. Si, Fe, and Al were the most abundant elements besides Ca in all the samples. For the majority of RD and CD samples, EFs of Si, Al, K, Ti, V, Cr, Mn, Fe, Ni, and Cu were <5, indicating that these elements were from the UCC, while S, Cl, Zn, and Pb were identified to be originated from anthropogenic sources.

However, there were some differences in the chemical profiles of RD and CD between different cities and regions. For example, WSI fractions in RD or CD ranged from 4.2% to 19.0% between the seven regions, a 4-fold difference. The most dominant WSI chemical species were also different between Northern and Southern China, so was the case for elements. Soil dust contributions to both RD and CD were ~20% higher in Northern than Southern China. To accurately describe the inter- and intra-regional similarities and differences in the two types of fugitive dust profiles, COD, a self-normalizing parameter was calculated between cities (Tables S4a) and regions (Table 3a,b,c).

COD values of RD were approximately 0.2 among cities within the same region (NWC, NCP, NEC and CC) in northern China and ranged from 0.3 to 0.4 within the same region (SC, SWC and EC) in southern China, which were all obviously smaller than those between cites of different regions. A value of 0.2 for COD was widely accepted as a cut-off point below which it is considered to be similar (Han et al., 2010; Wongphatarakul et al., 1998). According to the above-mentioned COD values, RD from cities within the same region could be aggregated together for analyzing similarity and conducting source identification (Table 3a). Large COD variations between regions suggested that urban RD could not be deemed as uniform or from a single source. For instance, COD between NWC and SWC was as high as 0.47, indicating that RD in these two regions were influenced by very different factors and had different chemical components. Treating RD samples as from a single source would lead to large errors in source apportionment analysis using receptor models (Reff et al., 2007; Watson, 1984). On average, lower COD values were found for CD than RD, indicating higher levels of similarity in CD, which may be due to the large usage of building materials.

Table 3a
Coefficient of divergence for road dust among the seven regions in China.

	NWC	NCP	NEC	CC	SC	SWC	EC
NWC	0						
NCP	0.32	0					
NEC	0.41	0.20	0				
CC	0.44	0.33	0.35	0			
SC	0.40	0.19	0.19	0.29	0		
SWC	0.47	0.33	0.32	0.34	0.26	0	
EC	0.42	0.27	0.25	0.30	0.22	0.29	0

Table 3b
Coefficient of divergence for construction dust among the seven regions in China.

	NWC	NCP	NEC	CC	SC	SWC	EC
NWC	0						
NCP	0.32	0					
NEC	0.37	0.16	0				
CC	0.43	0.34	0.31	0			
SC	0.39	0.23	0.18	0.23	0		
SWC	0.39	0.28	0.25	0.26	0.20	0	
EC	0.41	0.38	0.37	0.25	0.31	0.35	0

COD between RD and CD within and between northern and southern China were calculated (Table 3c). RD and CD similarity was prominent if considered Northern (COD value of 0.11) and Southern (0.09) China separately, but this is not the case if compared between norther and southern China (0.29–0.31).

3.5. Source identification of urban fugitive dust from other dust types

Ratios between the elements that comprise soil dust can be considered as markers to trace the origins of dust in various desert regions (Alfaro et al., 2003; Arimoto et al., 2004). In this study, the ratios of elements (Si, K, Ca, Ti, Mn, and Fe) to Al in fugitive dust samples were determined and are shown in Table 4. The diagnostic ratios in urban fugitive dust and desert soil dust reported in the literature are also listed in Table 4 for comparison.

The Si/Al ratios for RD (2.24 and 2.37 in samples from Northern and Southern China, respectively) were significantly higher than those for CD (1.68 and 1.66, respectively). These values were comparable to those reported for Hong Kong and Fushun fugitive dust (Ho et al., 2003; Kong et al., 2011a), but were much lower than those for UCC (3.83), Chinese loess soil (2.9) and cements (3.39–3.93). The Ca/Al ratios (considering RD and CD together) varied from 2.02 to 4.32 among the seven regions in China, and these values were much higher than that for UCC (0.35) but lower than those for cements (6.3–29.47). Higher Ca/Al ratios were found in CD than RD in all the regions, suggesting stronger influence of cements in CD than RD (Ho et al., 2006). As shown in Table 4, the Ca/Al ratios in Asian dust, which originated mainly from the natural aeolian process in desert regions, were much lower than those in urban fugitive dust samples (Arimoto et al., 2004; Kim et al., 2003; Kim et al., 2001; Xu et al., 2012). Our results highlighted that urban fugitive dust was influenced heavily by anthropogenic emissions, such as construction activities. Therefore, Ca/Al ratios should be taken as a tracer to distinguish the origins of urban dust from local sources or from long-distance transportation (such as Asian dust).

The ratios of K/Al and Fe/Al in dust samples both showed obviously higher values in Northern than Southern China. Soil dust from Northern China (Chinese loess) and desert dust (Asian dust) reported in literature had similar ratios to urban fugitive dust in Northern China reported in the present study, which implied that high Fe and K contents in urban fugitive dust in Northern China could be derived from soil and desert dust (Bi et al., 2007; Lee et al., 2008). The ratios of Ti/Al and Mn/Al in urban fugitive dusts were relatively consistent with those in UCC, Chinese loess, and Asian dust, which implied that these elements were

Table 3c
Coefficient of divergence for road and construction dust between Northern and Southern China.

	Northern China RD	Southern China RD	Northern China CD	Southern China CD
Northern China RD	0			
Southern China RD	0.30	0		
Northern China CD	0.11	0.29	0	
Southern China CD	0.31	0.09	0.29	0

Table 4
Comparison of ratios of selected elements to Al.

City/area	Type	Si/Al	K/Al	Ca/Al	Ti/Al	Mn/Al	Fe/Al	Zn/Al	Pb/Al	Reference
NWC	RD	1.82	0.66	3.19	0.10	0.03	1.24	0.023	0.0082	This study
NCP	RD	2.36	0.41	2.50	0.09	0.02	1.02	0.021	0.0059	
NEC	RD	2.94	0.40	2.02	0.09	0.03	0.93	0.013	0.0045	
CC	RD	2.11	0.29	2.79	0.07	0.02	0.71	0.012	0.0083	
SC	RD	1.90	0.37	2.74	0.07	0.02	0.81	0.016	0.0077	
SWC	RD	2.10	0.32	2.81	0.12	0.02	0.89	0.017	0.0058	
EC	RD	2.24	0.30	2.38	0.08	0.02	0.84	0.017	0.0100	
Northern China	RD	2.37	0.47	2.55	0.10	0.03	1.05	0.020	0.0061	
Southern China	RD	2.09	0.32	2.70	0.09	0.02	0.82	0.016	0.0077	
NWC	CD	1.68	0.64	3.51	0.10	0.03	1.18	0.018	0.0076	
NCP	CD	1.66	0.35	2.98	0.08	0.02	0.83	0.010	0.0034	
NEC	CD	2.09	0.35	2.36	0.07	0.03	0.78	0.009	0.0023	
CC	CD	2.13	0.30	3.69	0.06	0.02	0.83	0.016	0.0107	
SC	CD	1.73	0.34	2.68	0.07	0.02	0.80	0.014	0.0075	
SWC	CD	2.03	0.34	4.32	0.12	0.02	0.83	0.009	0.0046	
EC	CD	2.19	0.27	3.77	0.07	0.02	0.68	0.008	0.0077	
Northern China	CD	1.68	0.64	3.51	0.10	0.03	1.18	0.018	0.0042	
Southern China	CD	1.66	0.35	2.98	0.08	0.02	0.83	0.010	0.0073	
Hong Kong	Cement	3.93	1.1	29.47	0.09	0.03	1.35	0.036	0.0115	Ho et al. (2003)
Hong Kong	Urban fugitive dust	2.56	0.4	1.93	0.05	0.03	1.54	0.148	0.0300	
Fushun	Cement	3.39	0.02	6.3	0.12	0.01	0.26	0.005	0.0002	Kong et al. (2011a)
Fushun	Road dust	1.79	0.09	2.89	0.06	0.01	0.14	0.009	0.0008	
Earth UCC	Bulk sample	3.83	0.35	0.37	0.04	0.01	0.44	0.001	0.0004	Taylor and Mclennan (1995)
Chinese Loess	Loess	2.9	0.47	2.5	0.08	0.03	1.05	0.003	0.0017	Cao et al. (2008)
Yulin, China	Asian dust	1.9	0.32	1	0.05	0.02	0.59	0.035	0.0250	Arimoto et al. (2004)
Yulin, China	Asian dust		0.46	0.81	0.07	0.02	0.51	0.005	0.0036	Xu et al. (2004)
Tongliao, China	Asian dust	4.06	0.44	0.76	0.07	0.02	0.77	0.09	0.0200	Shen et al. (2007)
Seoul, Korea	Asian dust		0.45	0.74	0.04	0.04	0.87	0.124	0.0704	Kim et al. (2003)
Gwangju, Korea	Asian dust		0.52	0.64	0.03	0.02	0.67	0.042	0.0230	Kim et al. (2001)

mainly from natural crustal matter. For heavy metals, the ratios of Zn/Al and Pb/Al were higher on average in RD than CD. The ratios observed in the present study were much higher than those in UCC and Chinese loess, but lower than those in Asian dust in Yulin, China observed by Xu et al. (2004). These results indicated that urban fugitive dust and Asian dust were influenced by anthropogenic emissions to some extent, and human activities influenced more on RD than CD. Previous studies have demonstrated that aerosol samples (even in desert regions) were influenced by non-crustal sources (Cao et al., 2005a; Zhang et al., 2003).

Ratios between water-soluble ions can also indicate the origins of soil dust in various desert regions (Arimoto et al., 2004; Shen et al., 2007). Relative ratios between selected water-soluble ions in this study and those determined in prior studies are listed in Table 5. The ratio of K^+/K ranged from 0.07 to 0.31 in RD and from 0.12 to 0.38 in CD. Water-soluble K was taken as a marker of biomass burning sources (Andreae and Merlet, 2001; Sun et al., 2017). Higher K^+/K ratios were previously observed during pollution episodes (0.55) as well as in straw burning-emitted particles (0.73) (Ni et al., 2017) than dust-dominated periods (Shen et al., 2007), Therefore, contributions from biomass burning sources to urban fugitive dust were very limited in the present study as indicated by the very low K^+/K ratios, and the majority of K in fugitive dust was insoluble.

The NO_3^-/SO_4^{2-} ratio has been used as an indicator of the relative importance of stationary versus mobile sources to aerosol particles

Table 5
Comparison of diagnostic ratios of selected water-soluble ions.

Location	Sample type	K^+/K	SO_4^{2-}/K^+	NO_3^-/SO_4^{2-}	Cl^-/K^+	SO_4^{2-}/Ca^{2+}	Reference
NWC	RD	0.13	8.03	0.20	4.24	0.43	This
NCP	RD	0.24	5.02	0.23	1.41	0.29	study
NEC	RD	0.13	7.64	0.10	1.13	0.23	
CC	RD	0.29	4.08	0.09	0.75	0.10	
SC	RD	0.31	2.65	0.09	0.65	0.15	
SWC	RD	0.27	2.43	0.00	0.39	0.07	
EC	RD	0.07	12.58	0.10	7.25	0.13	
Northern China	RD	0.16	6.92	0.19	2.62	0.33	
Southern China	RD	0.24	3.75	0.08	1.11	0.11	
NWC	CD	0.12	6.44	0.20	3.17	0.28	
NCP	CD	0.13	9.18	0.16	1.76	0.24	
NEC	CD	0.19	3.72	0.11	0.54	0.19	
CC	CD	0.30	3.46	0.07	0.67	0.08	
SC	CD	0.34	2.22	0.10	0.51	0.12	
SWC	CD	0.23	3.24	0.03	0.27	0.10	
EC	CD	0.38	4.85	0.02	2.36	0.10	
Northern China	CD	0.14	6.06	0.16	1.92	0.24	
Southern China	CD	0.30	3.23	0.05	0.82	0.10	
Tongliao	Dust storm	0.15		0.31		0.15	Shen et al. (2007)
	Normal days	0.34		0.27		0.34	
	Pollution episode	0.55		0.52		0.55	
Beijing	Aerosol		11.01	0.67	1.98	10.47	Wang et al. (2005)
Source	Straw burning	0.73	3.31	2.00	0.62	3.11	Ni et al. (2017)
	Fly ash of coal combustion	0.05	172.6	0.10			Chow et al. (2004)

(Arimoto et al., 1996; Shen et al., 2008). In the present study, the NO_3^-/SO_4^{2-} ratio in dust samples was 0.19 in RD and 0.16 in CD in Northern China and was 0.08 and 0.05, respectively, in Southern China. These ratios were relatively low when compared to aerosol samples in Beijing (0.52) and Tongliao (0.27–0.52) (Wang et al., 2005; Shen et al., 2007). It can thus be concluded that urban fugitive dust was mainly from stationary emission sources.

The ratios of SO_4^{2-}/K^+ and SO_4^{2-}/Ca^{2+} in dust samples were higher in Northern China than in Southern China, suggesting more contributions from coal combustion to fugitive dust in Northern than Southern cities in China. It is noted that the NO_3^-/SO_4^{2-} ratios in dust samples from Southern China were even lower than that from coal combustion source (0.10 in fly ash from coal combustion) reported by Chow et al. (2004), suggesting the possibility of additional sulfate-rich sources contributing to fugitive dust in Southern China (i.e. industry and power plant emission).

4. Conclusions

The chemical profiles of urban fugitive dust from 21 selected cities distributed in seven regions in China were determined and compared with those reported in relevant literature. Soil dust comprised as much as 50% of urban fugitive dust from Southern cities and as much as 70% from Northern cities, indicating more influence from anthropogenic emissions in Southern than Northern cities. The WSI proportions in RD and CD were slightly higher in Southern China (10.3 ± 2.7% and 12.2 ± 2.4%, respectively) than in Northern China (9.0 ± 4.0% and 10.2 ± 3.3%, respectively), although the differences in RD were not significant. The total carbonaceous fractions in urban fugitive dust samples were dominated by OC (>80%), with higher percentages in Northern than Southern China. In contrast, EC showed a reverse spatial gradient than OC, indicating that OC and EC were partly produced from different sources. Among the elements, Ca, Si, Al and Fe were the most abundant in urban fugitive dust samples. High ratios of SO_4^{2-}/K^+ and SO_4^{2-}/Ca^{2+} and low ratios of NO_3^-/SO_4^{2-} revealed that urban fugitive dust was heavily influenced by stationary sources. COD results showed that regional difference for both RD and CD were significant. Further studies should focus on measuring organic matter (e.g., Polycyclic Aromatic Hydrocarbons) in dust samples to generate more comprehensive chemical profiles of urban fugitive dust.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.08.374.

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