Similarities and Differences in $PM_{2.5}$, PM_{10} and TSP Chemical Profiles of Fugitive Dust Sources in a Coastal Oilfield City in China

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ABSTRACT

$PM_{2.5}$, PM_{10} and TSP source profiles for road dust, soil dust and re-suspended dust were established by adopting a re-suspension chamber in the coastal oilfield city of Dongying. Thirty-nine elements, nine ions, organic and elemental carbon were analyzed by multiple methods. The results indicate that Ca, Si, OC, Ca^{2+}, Al, Fe and SO_4^{2-} were the most abundant species for all the three types of dust within the three size fractions. OC/TC ratios were highest for dusts in Dongying when compared with those in the literature, which may be related to the large amounts of oil that is consumed in this area. Na, Mg and Na^+ in soil and road dust also exhibited higher mass percentages, indicating the influence of sea salt, as the city is close to the coast. Enrichment factors analysis showed that Cd, Ca, Cu, Zn, Ba, Ni, Pb, Cr, Mg and As were enriched, and these elements were more abundant in finer fractions of the fugitive dust. Similarities of the profiles for different size fractions and different types of dust were compared using the coefficient of divergence (CD). The profiles for different types of dust were different from each other, as the CD values were mostly higher than 0.30. In contrast, the profiles for different size fractions of each dust were similar to each other, with the CD values mostly lower or near to 0.30. From analyzing the relationship between crustal material's mass percentages and the total measured mass percentages of all chemical species, it can be concluded that the sources of road dust and resuspended dust are more complex, especially for fine particles.

Keywords: Source profile; Fugitive dust; $PM_{2.5}$; PM_{10}; TSP.

INTRODUCTION

Particulate matter emitted from simplex source types including both natural and anthropological sources, such as crustal material, volcanic ash, sea salt, coal combustion, vehicle emission (tire and brake wear, tail-gas exhaust, etc.), road surface wear, construction and demolition activities, other industrial activities and so on can enter into the air. Then they deposited on and relocated among various receiving surfaces such as road, topsoil, building surface to form road dust (RD), surface soil dust (SD) and re-suspended dust (RDB) by external forces, such as surface runoff, wind blowing, weathering, gravitation and human forces like traffic-generated turbulence. The chemical compositions in them may be influenced by each other. Fugitive dust from paved and unpaved roads, construction and demolition and bare ground sites are important contributors to PM_{10} (particles with aerodynamic diameters < 10 µm) and $PM_{2.5}$ (particles with aerodynamic diameters < 2.5 µm) in many urban areas (Ho *et al.*, 2003). To obtain reliable source apportionment results, detailed and local source profiles are needed.

Former studies have established various chemical source profiles for the three types of dust around the world (Watson *et al.*, 2001; Watson and Chow, 2001; Chow *et al.*, 2003; Ho *et al.*, 2003; Samara *et al.*, 2003; Chow *et al.*, 2004; Samara, 2005; Zhao *et al.*, 2006; Feng *et al.*, 2007; Gupta *et al.*, 2007; Cao *et al.*, 2008; Yatkin and Bayram, 2008). Chow *et al.* (2003) found that differences existed among six composited geological source profiles (including paved road dust, unpaved road dust, agricultural soil, dairy and feedlot soil, salt deposits and construction soil) and among subtypes within each source type. While no studies concerned SD, RD and RDB in $PM_{2.5}$, PM_{10} and TSP fractions synchronously. Therefore, whether the source profiles for the three types of dust were similar or dissimilar with each other is still not well addressed. Watson and Chow (2001) indicated that it

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was essential to characterize the particle sources locally for source apportionment studies with respect to the diversity in geographical locations, fuel type, combustion and emission control technologies and variation along with time.

Dongying is an oilfield city developed from 1960s. The dominated industries are oil and natural gas mining, machining and oil equipment manufacturing. In the year of 2008, the consumed raw coal, coke, oil and natural gas were 6.78 million tons, 16 thousands tons, 14.2 million tons and 473.6 million cubic meters, respectively which may release considerable amounts of dust to deposit on urban surfaces. In the year of 2005–2009, the primary air pollutant was PM_{10} in Dongying. The annual average mass concentrations of PM_{10} ranged from 132 μg/m^3 to 149 μg/m^3, about two times of the secondary standard values (70 μg/m^3). It should be also mentioned that the city locates at the estuary of Yellow River with large areas of bare land which may serve as a potential source of dust. However, there is no related information on source emissions of dust. Therefore, it is urgent to identify the sources of atmospheric particulate matter to make effective measures for controlling PM pollution by local government. A scientific research program was set to investigate the levels of TSP, PM_{10} and $PM_{2.5}$ and to identify their sources by chemical mass balance (CMB) model in different seasons of 2009–2010. This study is one part of the project. Other data is being analyzed by members in our group.

The main objectives were: (1) to develop chemical source profiles of three main types of fugitive dust that could be used for CMB model; (2) to analysis the similarities and differences among different types of dust and among different size fractions to give some implications for source apportionment studies.

METHODOLOGY

Study Area Description and Sampling

Dongying is located at the north of Shandong Province, China, with the latitude as N36°55′-38°10′ and longitude as E118°07′-119°10′. The climate of Dongying belongs to warm temperate continental monsoon system. In autumn (the sampling period), the average relative humidity and wind speed for Dongying were 56.0% and 2.25 m/s in 2006–2009. The average precipitation was 574.3 mm in 2000–2009. Its area and population are 8053 km^2 and 1.83 million with 0.58 million of people living at Dongying District (1156 km^2). The center of Dongying District can be further divided into western (old district, OD) and eastern parts (new district, ND) with an area of 66 and 44 km^2, respectively. The distance between them is 15 km. The OD was developed from 1960s along with the exploitation of oil while the ND was developed from 1980s owing to the city expanding with the population now less than 100,000. Most of commercial activities are concentrated in OD and the ND is defined as residential and government working area.

Total of fifty-nine dust samples were collected with the sampling locations shown in Fig. 1. RD and RDB samples were swept using a plastic brush and tray (Ho *et al.*, 2003). Fifteen RD samples (about 1 kg each) were collected from

the city′s main streets with seven samples in ND and eight in OD. Each sample was mixed of sub-samples, including the surface dust from vehicle and bycycle ways as well as footways. Fourteen SD samples were collected around the city including two corn soil samples, three wheat soil samples and nine cotton soil samples. Soils (0–20 cm depth) were collected using a pre-cleaned stainless steel scoop into pre-cleaned polyethylene bags. Before sampling, particles of small vegetation or/and litter were removed if necessary. Each sample was consisted of five sub-samples collected from the surrounding of each site (within 100 × 100 m^2, 1–2 kg each). Thirty RDB samples (about 1 kg each, ten for ND and twenty for OD) were collected from longtime un-cleaned windowsills or flat roofs which were 5–15 m high and were stored in polyethylene bags (Zhao *et al.*, 2006). All the samples were cooled in a self-designed and temperature-controlled box filled with ice bags during transport to the laboratory. Care was taken to reduce the disturbance of fine particles. Any obvious extraneous matters such as cigarette ends or other debris were not collected. Each brush was used once only before giving a thorough cleaning. The coordinates of the sample locations were recorded with a GPS.

Dust Pre-Treatment Method

Samples were pre-treated using re-suspension method (Chow *et al.*, 1994; Zhao *et al.*, 2006; Kong *et al.*, 2011a). Each sample was weighed after being dried in a vacuum freeze dryer. After drying, the sample was sieved through a 160-mesh Tyler screen to remove fibers and other large particles. About 0.5 g sieved material was placed in a 250 mL side-arm vacuum flask sealed with a rubber stopper. Air puffs into the flask introduced dust into a chamber (Ho *et al.*, 2003) where it was sampled through $PM_{2.5}$, PM_{10} and PM_{100} inlets with a flow rate of 20 L/min for about 1–2 minutes onto polypropylene and quartz-fiber filters. Prior to sampling, polypropylene and quartz fiber filters were calcined at 60°C for 0.5 h and 800°C for 2 h to remove organic compounds, respectively. After sampling, the filters were removed from the inlets and wrapped in a laminar-flow clean hood until use.

Filters were equilibrated in a relative humidity (35% ± 1%) and temperature (22.0°C ± 1°C) controlled environment for 48 h before gravimetric analysis. Filters were exposed to a low-level radioactive source prior to and during sample weighing to remove static charge. Then they were weighed both before and after sampling by a sensitive microbalance (Mettler M5). The balance sensitivity was ± 0.001 mg. Filter blanks were assessed in the same manner as the sampling procedure. After weighing, the filters were stored at –4°C until chemical analysis (Kong *et al.*, 2011b).

Sample Analysis Method

Inductively coupled plasma-mass spectroscopy (ICP-MS) (Agilent 7500a, Agilent Co. USA) and inductively coupled plasma-optical emission spectrometer (ICP-OES) were used for analysis of thirty-nine elements. For ions (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, F^-, Cl^-, NO_3^- and SO_4^{2-}) analysis, a Dionex DX-120 Ion Chromatograph (IC) was
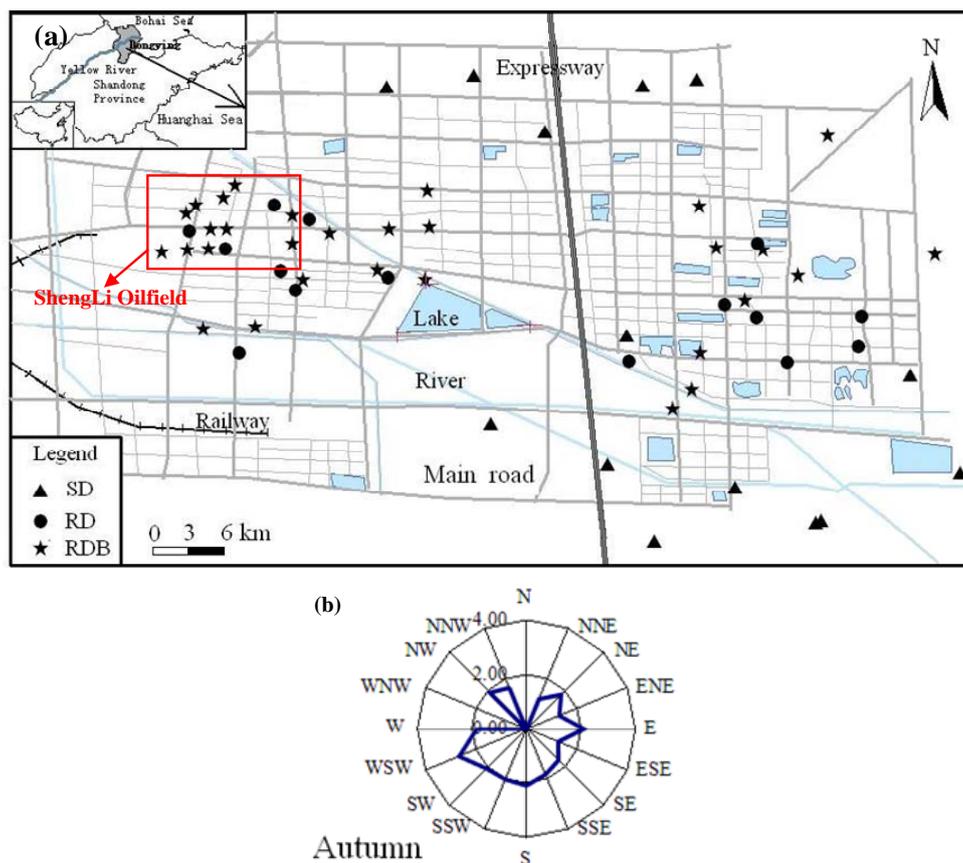
Fig. 1. (a) Sampling locations for road, soil and re-suspended dust in Dongying (SD: surface soil dust; RD: road dust; RDB: re-suspended dust deposited on building roofs or windowsills). The left of the expressway can be identified as old district and the right can be identified as new district. The red rectangle indicated sites located in the region of the ShengLi Oilfield. (b) Wind rose for autumn of Dongying in 2006–2009.

adopted. And for OC and EC analysis, Thermal/Optical Carbon Analyzer (DRI Model 2001) was used.

Thirty-four elements including Li, Be, Na, Mg, Al, P, K, Ca, Sb, Sc, Ti, V, Cr, Mn, Co, Ni, Cu, Zn, As, Rb, Y, Mo, Cd, Sn, Cs, La, Ce, Sm, W, Tl, Pb, Bi, Th and U were analyzed by ICP-MS. Before analysis, half of each polypropylene filter was placed at 100 mL polyfluortetraethylene beaker with 5 mL extractions (pH = 5.6) and a drop of HF (pH = 5.3) added. After covered, the solution was heated on a galvanothermy board at 220°C for 2.5 h. Then shut off the galvanothermy board, dipped the solution with 5 mL hydrochloric acid and put it into a 10 mL plastic bottle. Finally, the digested samples were diluted to 10 mL with deionized water. Two milliliter of solution was injected into the ICP-MS with flow rate as 2.0 mL/min.

Nine elements including Si, Al, Ca, Mg, Fe, Ti, Ba, Sr and Zr were analyzed by ICP-OES. Al, Ca, Mg and Ti were repeated with ICP-MS analysis results and we selected the data from ICP-OES analysis. Before analysis, half of each polypropylene filter was placed into nickel crucibles and then into a muffle with the temperature increasing from 0 to 300°C. Held on for 40 min and then increased the temperature to 530–550°C till the filters changed into ashes totally. Cooled the crucibles, added into some drops of ethanol and 0.2 g solid sodium hydroxide and then put the crucibles again into 500°C muffle for 10 min. Took the crucibles out, cooled them, added some hot distilled water and heated to boil. Then put the solution into a plastic tube with 2 mL hydrochloric acid (2:1 for acid and water). Washed the crucibles with 2% hydrochloric acid and put the washed solution into a plastic tube. At last, the solution was diluted with distilled water to 10 mL. One milliliter of solution was injected into the ICP-OES with flow rate as 1.0 mL/min.

The IC was consisted of a separation column (IonPac CS12A for cation and IonPacAS14 for anion) and a self-regenerating suppressed conductivity detector (IonPacCG12A for cations and Ionpac AG14 for anions) and a gradient pump (Dionex Ionpac GP50). The gradient weak acid eluent as 20 mmol/L MSA (1.0 mL/min) and weak base eluent as 3.5 mmol/L Na_2CO_3 +1.0 mmol/L $NaHCO_3$ (1.2 mL/min) were used for for cation and anion detection, respectively. Before analysis of ions, one-eighth of each quartz filter was cut into pieces into a vessel with scale, then was extracted with 5 mL distilled deionized water (18 MΩ) by ultrasonic and mechanical shaking in an ultrasonic cleaner (Type AS3120A, AutoScience Inc) at a frequency of 40 kHz for 15 min. One milliliter of solution was drawn into a syringe, filtrated by a 13 mm cellulose syringe filter (Xibosh) with pore size < 0.22 μm and injected into the IC with flow rate as 1.0 mL/min.

Quartz filter samples were also analyzed for OC and EC by using DRI Model 2001 (Thermal/Optical Carbon Analyzer). The IMPROVE A thermal/optical reflectance (TOR) protocol was used for the carbon analysis. Detailed descriptions could be found in our recent researches (Li *et al.*, 2012).

Quality Assurance (QA) and Quality Control (QC)

Geochemistry reference matter (GSS-2) made by the Center of National Standard Matter was analyzed simultaneously to check the reliability of ICP-MS. GBW0741, GBW07404 and GBW07406 were adopted to check the reliability of ICP-OES analysis. The relative standard deviations between real values for standard materials and analyzing results were in the range of 1.5–14.7% and the detection limits were from 0.0001 to 1.39 μg for all the elements. Background contamination was routinely monitored by using operational blanks (unexposed filters, kept in clean hoods before and after sampling until analysis) which were processed simultaneously with field samples. One standard sample was analyzed per ten field samples to assure the recoveries were in the range of 80%–120%. One field sample was analyzed twice per ten samples to assure the relative errors (precisions) were less than 20% for both ICP-MS and ICP-OES methods. Detailed data for QA and QC were listed in supplementary file S1 and S2 for ICP-MS and ICP-OES, respectively.

For ions, standard solutions were prepared (Na^+: 1.64 mg/L, NH_4^+: 2.25 mg/L, K^+: 4.50 mg/L, Mg^{2+}: 1.85 mg/L, Ca^{2+}: 3.24 mg/L, F^-: 4.70 mg/L, Cl^-: 11.0 mg/L, NO_3^-: 32.1 mg/L, SO_4^{2-}: 43.8 mg/L) and were detected for three times. Results showed that the relative standard deviations for the nine ions were in 0.19% (for SO_4^{2-})–5.98% (for Ca^{2+}). The method detection limits (MDLs) for IC were 0.0258 mmol/L for Na^+, 0.0021 mmol/L for NH_4^+, 0.0066 mmol/L for K^+, 0.0053 mmol/L for Mg^{2+}, 0.0030 mmol/L for Ca^{2+}, 0.0051 mmol/L for F^-, 0.0144 mmol/L for Cl^-, 0.1215 mmol/L for NO_3^- and 0.0696 mmol/L for SO_4^{2-}, respectively. For TC and OC, every one sample in ten was detected for twice and the precision was less than 2%. Standard concentrations of CH_4/CO_2 mixed gases were used for calibration of the analyzer in each day before and after sample analysis.

Data Processing Methods

Enrichment factor (EF), calculated relative to the composition of the background values of Chinese soil, had been used in former studies (Cao *et al.*, 2008). It is a simple and semi-quantitative way of determining whether the elemental concentrations in the samples were enriched relative to what one would expect from the amount of crustal/mineral matter in the sample (Cao *et al.*, 2008). EF were calculated as:

$$EF_i = \frac{(X_i/Al)_{samples}}{(X_i/Al)_{crustal}} \tag{1}$$

where X_i indicated the concentration for elements of interest or reference elements referred to the concentration of crustal rock, most commonly Al or Si. In this study, Al was selected as the reference element (Cao *et al.*, 2008). The background geochemical compositions of the city soil

types can be adopted as the background values (Ji *et al.*, 2008). As background values were distinctly different among different soil types, values used in this study (for Na, Mg, Al, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ba, Pb) were referred to those of Jinan (Ji *et al.*, 2008) considering it was more adjacent to Dongying. For background values of Cd (9.7E-06%) and As (0.00111%), they were referred to Kong *et al.* (2011) for the background content of Chinese soil. If EF approached unity, crustal material was probably the predominant source for element X_i (Cao *et al.*, 2008).

Similarities between the profiles for the three types of dust were identified by coefficient of divergence (CD), a self-normalizing parameter used to measure the spread of the data points for two datasets. The parameter was determined as following:

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

where j and k were for the two profiles or sampling sites or size fractions; p was the number of investigated components; x_{ij} and x_{ik} represented the average mass concentrations of chemical component i for j and k (Wongphatarakul *et al.*, 1998; Feng *et al.*, 2007; Kong *et al.*, 2011a). If CD_{jk} approached zero, source j and k were similar, and if it approached one, they were significantly different. Wongphatarakul *et al.* (1998) pointed that the CD value 0.269 reflected the similarity between particles from two cities. Feng *et al.* (2007) concluded that for PM_{10} size fractions, no significant difference in chemical composition existed between topsoil and deep soil profiles of the same subtype with the CD values as 0.290, 0.110 and 0.174, respectively for bared hills, cropland of wheat, corn and vegetables and sandy soil of riverbeds, respectively. In Feng *et al.* (2007), for calculating CD values, Na, Mg, Al, Si, P, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Br, Ba, Pb, TC, OC, Cl^- and SO_4^{2-} were adopted except NH_4^+ and NO_3^- because some of their amounts were lower than the method detection limits. For calculating CD values in this study, the species with mean mass percentages less than 0.001% (including Be, Sc, Co, Mo, Cd, Sn, Sb, Cs, Sm, W, Tl, Bi, Th, U and NH_4^+ except for Co, Cd, Sn and Sb as they were important species for coal combustion) were excluded.

RESULTS AND DISCUSSION

PM$_{2.5}$, PM$_{10}$ and TSP Chemical Profiles of SD, RD and RDB in Dongying

Chemical abundances for each sample were blank subtracted and calculated by dividing each chemical concentration by the mass concentration (Chow *et al.*, 2004). The composite consisted of the average and standard deviation of species abundances for all individual profiles within a group (Watson *et al.*, 2002). Supplementary files S3, S4 and S5 listed the detailed chemical source profiles for the three types of dust within $PM_{2.5}$, PM_{10} and TSP fractions, respectively.

For SD, the mass unaccounted were in the range of 50.17% ± 4.87% (mean ± standard deviation (sd), the same for the following), 46.35% ± 4.13% and 37.82% ± 5.57% for TSP, PM_{10} and $PM_{2.5}$, respectively. For RD, the values were 45.54% ± 8.90%, 46.85% ± 3.06% and 41.21% ± 4.97% while for RDB, they were 47.01% ± 9.30%, 51.02% ± 3.67% and 49.35% ± 2.60%, respectively. Unaccounted mass were due to components not measured in the samples and oxygen (most major elemental components such as Si, Al, Fe, etc. existed in the form of oxides) (Samara *et al.*, 2003; Samara, 2005). For comparison, chemical species were further classified into crustal elements including Na, Mg, Al, Si, K, Ca, Ti, Fe and Mn, trace elements including elements except for crustal elements, nine water-soluble ions, OC and EC. Fig. 2 shows the chemical compositions of each group.

Crustal elements were the most abundant species, with mean mass percentages as 45.4%–48.5%, 38.4%–41.1% and 36.6%–46.1% for SD, RD and RDB, respectively. RDB held the highest mean mass percentages for trace elements as 1.21%, 1.18% and 1.10% for TSP, PM_{10} and $PM_{2.5}$, respectively which were 8.5%, 233.7% and 154.7% higher than those for RD and were 318.0%, 241.9% and 208.6% higher than those for SD, respectively. It may indicate that RDB was more obviously influenced by anthropogenic sources. Ions showed highest mean mass percentages for $PM_{2.5}$ as 9.74%, 9.48% and 7.41% for SD, RD and RDB which meant that they tended to concentrate in fine particles of dust. OC also exhibited the highest values for $PM_{2.5}$ as 3.54%, 7.41% and 4.38% for SD, RD and RDB, respectively. Samara *et al.* (2003) pointed out that OC and EC were major components of particles emitted particularly from combustion sources which may explain the higher mass percentages for OC in fine particles. For EC, its mean mass percentages ranged from 0.15% to 1.19% for RD and from 0.19% to 0.27% for RDB while for SD, the values were zero which may be associated with the measurable method from re-suspension procedure and the following TOR analysis. The carbonate and minerals in the resuspended filters may

bias the analysis of the trace level of EC which needed further investigations. Characteristic of source profiles were discussed below separately for each source type.

Soil Dust

Si, Al, Ca, Fe and OC were most abundant constituents with mean ± sd as 20.05 ± 1.49%, 10.10 ± 2.46%, 9.82 ± 1.39%, 4.32 ± 0.90% and 3.54 ± 0.60% for $PM_{2.5}$, as 20.91 ± 1.67%, 7.92 ± 1.48%, 8.30 ± 0.63%, 4.12 ± 0.40% and 2.76 ± 0.47% for PM10 and as 25.40 ± 2.67%, 6.74 ± 1.21%, 6.13 ± 0.94%, 3.61 ± 0.76% and 1.86 ± 0.29% for TSP, respectively. Meanwhile for $PM_{2.5}$ and PM_{10}, Ca^{2+} was abundant as 6.77 ± 2.41% and 3.51 ± 1.55% which may be related to local intensive construction activities.

The OC/TC values observed in other SD profiles around the world were in the range of 0.64–0.99 (Watson *et al.*, 2001; Chow *et al.*, 2003; Ho *et al.*, 2003; Chow *et al.*, 2004; Gupta *et al.*, 2007) by the influence of motor vehicle emissions including brake, tire wear and oil drips (Ho *et al.*, 2003) and crop debris and agricultural chemicals (e.g., fertilizers, pesticides, herbicides) (Chow *et al.*, 1994; Cao *et al.*, 2008). While in this study, no EC was detected resulting in the OC/TC values high to 1.00 for all soil samples and the reason was explained before.

Samara (2005) studied that the Ca/Si ratio was less than 1 in the northwestern sites of Kozani Ptolemais-Florina basin, probably due to the presence of granites and gneisses; whereas it ranged between 1.5 and 2.0 in the southern sites where limestone was the prevailing formation. Chow *et al.* (2004) pointed that Al/Ca ratio > 3.8 were indicator of Sahara dust transport at eastern US IMPROVE monitors. Cao *et al.* (2008) calculated the elemental ratios of Si, K, Ca, Ti, Mn and Fe to Al in Asian dust source regions-Chinese Loess Plateau, compared the values to those found at downwind regions such as Korea, Japan and the North Pacific and concluded that these ratios can be used as fingerprints to trace the transport path of Asian dust. Dongying city was on the storm transport path and was thus an important exit point of the dust storms to the Pacific Ocean (Hsu *et al.*, 2005).
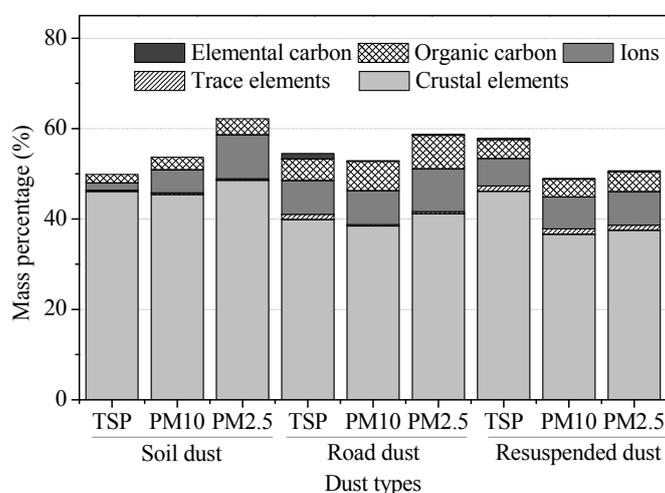

Fig. 2. Major chemical components for TSP, PM_{10} and $PM_{2.5}$ source profiles of soil dust, road dust and resuspended dust from Dongying.

Though soil profiles may change substantially with respect to geological origin, elemental ratios were calculated in Fig. 3.

The mean values for ratios of Si/Al, K/Al, Ca/Al, Ti/Al, Mn/Al, Fe/Al, Si/Ca, Si/Fe, Fe/Ca and K/Fe decreased mostly as the order: TSP > PM_{10} > $PM_{2.5}$, reflecting these elements tended to concentrate in coarse particles. The mean ± sd values for Si/Al, K/Al, Ca/Al, Ti/Al, Mn/Al, Fe/Al, Si/Ca, Si/Fe, Fe/Ca and K/Fe were 2.07 ± 0.41, 0.09 ± 0.04, 1.03 ± 0.29, 0.05 ± 0.01, 0.005 ± 0.002, 0.45 ± 0.12, 2.08 ± 0.34, 4.87 ± 1.39, 0.45 ± 0.11 and 0.23 ± 0.11 for $PM_{2.5}$, were 2.69 ± 0.31, 0.17 ± 0.08, 1.08 ± 0.21, 0.06 ± 0.01, 0.009 ± 0.004, 0.53 ± 0.07, 2.53 ± 0.62, 5.10 ± 0.39, 0.50 ± 0.06 and 0.33 ± 0.15 for PM_{10} and were 3.83 ± 0.51, 0.19 ± 0.07, 0.92 ± 0.16, 0.07 ± 0.02, 0.008 ± 0.003, 0.54 ± 0.12, 4.19 ± 0.44, 7.18 ± 0.91, 0.59 ± 0.05 and 0.36 ± 0.13 for TSP, respectively. The values were similar to those of dust from Chinese Loess Plateau, implying these crustal elemental ratios may be used to trace the aerosol transport during Asian Dust period.

Road Dust

Ca, Si, OC, Ca^{2+} and Al exhibited highest mean mass percentages with mean ± sd as 17.64 ± 2.59%, 11.57 ± 1.69%, 7.36 ± 1.75%, 5.45 ± 2.22% and 5.05 ± 1.21% for $PM_{2.5}$, 18.01 ± 2.27%, 10.20 ± 1.35%, 6.46 ± 1.04%, 3.17 ± 1.75% and 3.93 ± 0.54% for PM_{10} as well as 15.51 ± 2.13%, 13.37 ± 3.92%, 4.75 ± 1.07%, 2.32 ± 1.54% and 4.33 ± 1.47% for TSP, respectively. For TSP fraction, the contents of Na^+ (1.88 ± 0.88%) and Cl^- (2.41 ± 1.08%) were both highest than those for PM_{10} (1.49 ± 0.33% for Na^+ and 1.63 ± 0.39% for Cl^-) and $PM_{2.5}$ (1.22 ± 0.36% for Na^+ and 1.50 ± 0.28% for Cl^-) reflecting sea salt mainly influence the coarse particles. The percentages of Na^+ and Cl^- in TSP were 21.2% and 3.9% higher in ND than those in OD as the former was more close to sea (with the distances to sea as about 26 km and 40 km, respectively).

The OC/TC ratios for RD have always been calculated to identify the importance of vehicle emission in other studies (Watson *et al.*, 2001; Watson and Chow, 2001; Chow *et al.*, 2003; Ho *et al.*, 2003; Chow *et al.*, 2004). They were 0.97, 0.98 and 0.80 for $PM_{2.5}$, PM_{10} and TSP, respectively in this study which were higher than those as 0.92 in Hong Kong for $PM_{2.5}$ (Ho *et al.*, 2003), as 0.85 and 0.57 for residential

and industrial sites in Kolkata (Gupta *et al.*, 2007), as 0.90 in Colorado for $PM_{2.5}$ (Watson *et al.*, 2001), as 0.87 in San Joaquin Valley for PM_{10} (Chow *et al.*, 2003) and as 0.87 and 0.89 for $PM_{2.5}$ and PM_{10} in the Big Bend National Park (Chow *et al.*, 2004), respectively. The OC/TC ratios were all highest for the three fractions in Dongying than others which may be related to its energy structure in which lots of oil was consumed as discussed in the introduction. The OC/TC ratios of $PM_{2.5}$, PM_{10} and TSP in OD were 0.98, 0.98 and 0.85, all higher than those in ND (as 0.96, 0.97 and 0.75) which could be explained by the more heavy traffic in OD. It can also be found that the OC/TC ratios were highest for $PM_{2.5}$ implying the carbon components were mainly abundant in fine particles.

The major sources for RD (paved or non-paved) were from tear of vehicle components (such as tires, brakes, clutches and exhaust system components), wear of pavement and unpaved areas (such as construction areas, unpaved roads, parking lots and shoulders), deposition of suspended particles from emission sources and water or wind erosion in adjacent areas (Ho *et al.*, 2003; Samara, 2005; Gupta *et al.*, 2007). The Pb abundance in RD of Dongying (mean ± sd as 0.006 ± 0.004% for $PM_{2.5}$, 0.005 ± 0.001% for PM_{10} and 0.005 ± 0.001% for TSP) was lower as only unleaded gasoline for motor vehicles were sold. Element Zn in RD may reflect contribution from tire wear and the use of zinc compounds in rubber production (Ho *et al.*, 2003; Samara, 2005). The mean ± sd values of Zn were 0.045 ± 0.022% for $PM_{2.5}$, 0.030 ± 0.019% for PM_{10} and 0.019 ± 0.007% for TSP, respectively which were about ten times lower than those in Hong Kong (Ho *et al.*, 2003). It may be related with the lower traffic density in Dongying than in Hong Kong.

Re-Suspended Dust

Till now, there was only one study concerning the source profiles of RDB by Zhao *et al.* (2006) in six northern Chinese cities and the major species were Si (12.1%–22.3%), Al (5.7%–8.7%), Ca (6.8%–15.9%), TC (6.7%–24.4%) and OC (6.9%–20.9%) (min-max values were listed). In this study, the dominated constituents were Si, Ca, Al, Fe, OC and SO_4^{2-} with the mean ± sd values as 13.08 ± 2.18%, 11.76 ± 2.15%, 5.53 ± 0.95%, 3.45 ± 1.14%, 4.38 ± 0.61% and 3.07 ± 1.83% for $PM_{2.5}$, as 13.08 ± 2.47%, 11.06 ±
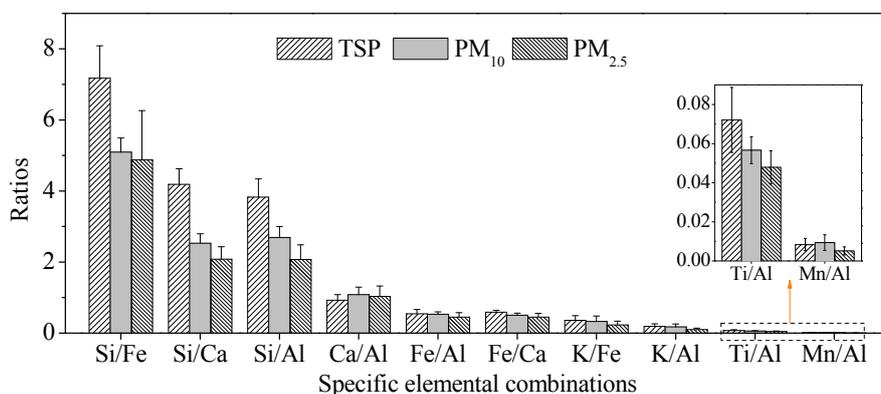

Fig. 3. Elemental ratios for soil dust from Dongying for TSP, PM_{10} and $PM_{2.5}$.

1.73%, 5.33 ± 0.10%, 3.49 ± 0.94%, 3.90 ± 0.68% and 2.71 ± 1.70% for PM_{10} and as 18.59 ± 7.76%, 11.51 ± 5.60%, 7.21 ± 4.00%, 4.61 ± 2.40%, 4.24 ± 1.79% and 2.57 ± 2.21% for TSP, respectively. The percentages of Na^+ in $PM_{2.5}$, PM_{10} and TSP were 31%, 31% and 13% higher in ND than those in OD; for Cl^-, they were 21%, 65% and 52% higher in ND. The content of SO_4^{2-} were in the range of 1.51–7.03% for PM_{10} profiles of RDB collected from six northern Chinese cities where coal combustion may contribute 14–66% to RDB in these cities (Zhao et al., 2006). This may explain the higher mass percentages of SO_4^{2-} in RDB from Dongying. The OC/TC ratios reported by Zhao et al. (2006) were 0.61–0.92 for RDB profiles, showing that TC was mainly in the form of OC and in this study, the ratios were 0.95, 0.95 and 0.94 for $PM_{2.5}$, PM_{10} and TSP, respectively. The reason may be that more oil were exploited and processed in Dongying than other cities.

Another interesting phenomenon should be emphasized was that though the OC/TC values for RDB within $PM_{2.5}$ for sites in or at the surroundings of the ShengLi Oilfield (one of the biggest oilfield in China) (OC/TC value was 0.92) were similar with those for other sites (OC/TC value was 0.97), the EC percentages for samples at the surroundings of ShengLi Oilfield were 2.6 times of those for other sites. The EC percentages at the surroundings of ShengLi Oilfield were 3.2 and 2.9 times of those for other sites for PM_{10} and TSP, respectively. It indicated that EC emitted from oilfield as production by the incomplete combustion of oil may influence its contents in surrounding dust.

Comparison with Others

To better understand the accuracy and representative of the source profiles established in this study, comparison were conducted with others as supplementary files S6, S7 and S8 shown for SD, RD and RDB, respectively.

For SD in Dongying, all the species were comparable with others except for Na, Mg and Na^+. Their mean mass percentages were 4.7, 10.2 and 5.8 times of those for TSP profiles, were 2.2, 2.4 and 4.0 times of those for PM_{10} profiles and were 3.2, 5.4 and 7.5 times of those for $PM_{2.5}$ profiles of soil from Chinese Loess Plateau, respectively. The reason may be related to its location as Dongying was more adjacent to the coastal line and dust here may be influenced more by sea salt. The extremely lower values for EC in SD in Dongying have been explained in section 3.1.

For RD, Na, Mg and Na^+ also exhibited higher mean mass percentages as 4.1–81.7, 1.3–4.9 and 2.1–18.8 times of others in literatures for PM_{10} fraction; while for $PM_{2.5}$, the values were 3.25–38.0, 2.5–4.6 and 1.0–8.66, respectively (min-max values were listed). Content of Na^+ in the dust from Hong Kong also exhibited higher values as 0.72% and 1.19% for PM_{10} and $PM_{2.5}$ (Ho et al., 2003) as it was also adjacent to sea. Meanwhile, the influence of sea salt was more obviously for coarse particles of RD in Dongying from the ratios listed above. Heavy metals like Cr, Mn, Co, Cu, Zn, Sn and Pb in RD from Hong Kong exhibited the highest mean mass percentages which were 3.7–6.1, 1.3–2.5, 4.8–11.9, 2.7–5.9, 3.6–17.0, 3.6–96.3 and 2.7–21.7 times of others for PM_{10} and were 2.1–2.7, 1.1–2.5, 23.8,

2.4–12.3, 3.5–14.2, 4.4–66.4 and 3.1–20.2 times of others for $PM_{2.5}$, respectively indicating vehicle emission was an more important source for RD in Hong Kong (Ho et al., 2003) (min-max values were listed). Higher OC and EC contents were also found for both $PM_{2.5}$ and PM_{10} profiles in Ho et al. (2003) and Chow et al. (2004) for paved RD. In Ho et al. (2003), RD samples were collected from highways, commercial/residential areas and tunnels; in Chow et al. (2004) samples were collected in Big Bend National Park in Texas where existed obvious human sources including coal-fired power plants and other urban/industrial sources. Therefore, the OC and EC levels in RD may be enhanced by vehicle emission, stationary sources and coal mining activities.

For RDB, all the species in Dongying were comparable with others in Bi et al. (2007) and Zhao et al. (2006) for PM_{10} profiles. Till now, there were eight northern Chinese cities established the profiles for RDB combined with this study. The major species were Si, Ca, OC, Al, Fe, SO_4^{2-}, Mg, K and Na with the mean ± sd values as 16.14% ± 5.15%, 8.72% ± 2.76%, 8.59% ± 3.78%, 7.16% ± 0.90%, 3.82% ± 1.24%, 3.68% ± 2.43%, 1.62% ± 0.64%, 1.36% ± 0.62% and 0.79% ± 0.32%, respectively. The higher SO_4^{2-} values indicated that in northern Chinese cities, coal combustion was still an important source for fugitive dust.

Enrichment Factors Analysis

Fig. 4 showed that Cd and Ca were significantly enriched with EFs values higher than 5 for all the three types of dust except for PM_{10} and $PM_{2.5}$ fractions. While for RD, EFs values were higher than 2 for Cu, Zn, Ba, Ni, Pb, As, Cr and Mg and for RDB, Zn, Pb, Cu, Ba, As, Ni, Mg and Cr exhibited EFs values higher than 2 indicating moderate enrichment (Han et al., 2006). The higher EFs values for Cd, Ca, Cu, Zn, Ba, Ni, Pb, Cr, Mg and As suggested they were mainly influenced by anthropogenic sources including fossil fuel combustion, vehicle emission, construction and manufacturing activities as discussed before. Meanwhile, the Cd percentages for sites at the surroundings of ShengLi Oilfield were 1.6, 1.1 and 1.2 times of those for other sites for $PM_{2.5}$, PM_{10} and TSP, respectively. While, the enrichment factor of Cd for $PM_{2.5}$, PM_{10} and TSP were 1.2, 0.85 and 0.96 times of those for other sites. Therefore, coal combustion or incineration may influence the Cd content more obviously in fine dust particles. The EFs for Fe, Ti, Mn, Co, K, Na and V were mostly lower than 2 indicating deficiency to minimal enrichment and they tended to concentrate in coarse fraction with EFs values decreased as TSP > PM_{10} > $PM_{2.5}$ (Fe, Ti, K, Na and V for SD; Mg, Na, Fe, Mn, K and V for RD; Ba, Co, Ti, V and K for RDB). While for anthropogenic-related elements, they tended to concentrate in finer fraction with EFs values exhibiting decreasing trends as $PM_{2.5}$ > PM_{10} > TSP (Cd, Zn, Cu, Pb, Cr, As and Ni for SD; Cd, Cu, Zn, Ni, Pb, As and Cr for RD; Cd, Zn, Ca, Pb, Cu, As, Ni and Cr for RDB).

Similarities of Fugitive Dust Profiles with Different Types and Different Fractions

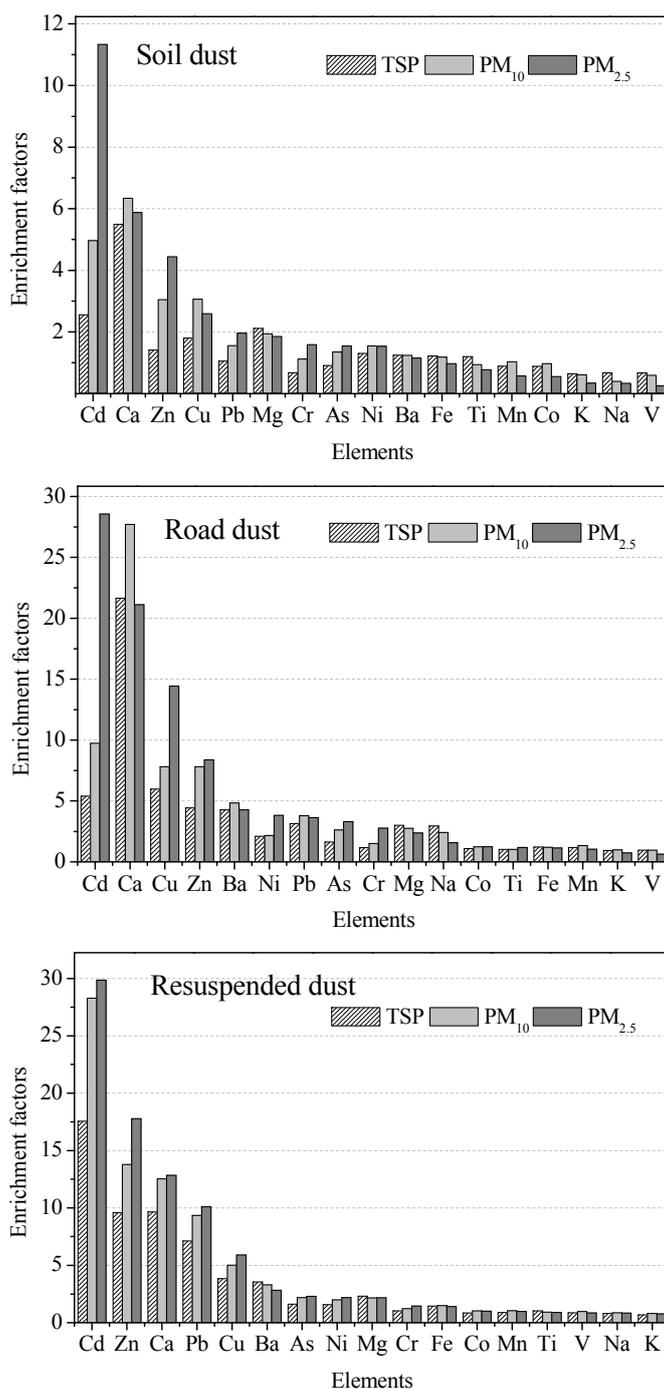The mass percentages scatter diagrams for different size

Fig. 4. Enrichment factors for soil dust, road dust and resuspended dust. Al was used as the reference element.

fractions and different types of dust were shown in Fig. 5 and Fig. 6 with CD_{jk} values listed.

For the comparison among different types of dust, the CD values were 0.39, 0.40 and 0.47 for RD-SD, RD-RDB and SD-RDB for TSP, were 0.33, 0.33 and 0.38 for RD-SD, RD-RDB and SD-RDB for PM_{10} and were 0.27, 0.32 and 0.35 for RD-SD, RD-RDB and SD-RDB for $PM_{2.5}$, respectively indicating that the profiles were different from each other for the three types of dust with an example as RD-SD for $PM_{2.5}$ excluded. In all the three fractions, the CD values were lowest for RD-SD and were highest for SD-

RDB implying the soil dust and road dust may influenced by some similar sources as the surface soil dust were mainly collected close to major roads in this study.

For comparison among different size fractions of each dust, the CD values were 0.27, 0.43 and 0.26 for TSP-PM_{10}, TSP-$PM_{2.5}$ and PM_{10}-$PM_{2.5}$ for SD, were 0.19, 0.33 and 0.21 for TSP-PM_{10}, TSP-$PM_{2.5}$ and PM_{10}-$PM_{2.5}$ for RD and were 0.11, 0.18 and 0.10 for TSP-PM_{10}, TSP-$PM_{2.5}$ and PM_{10}-$PM_{2.5}$ for RDB, respectively indicating that the profiles were similar to each other for the three size fractions with TSP-$PM_{2.5}$ for SD and RD excluded. It indicated that the
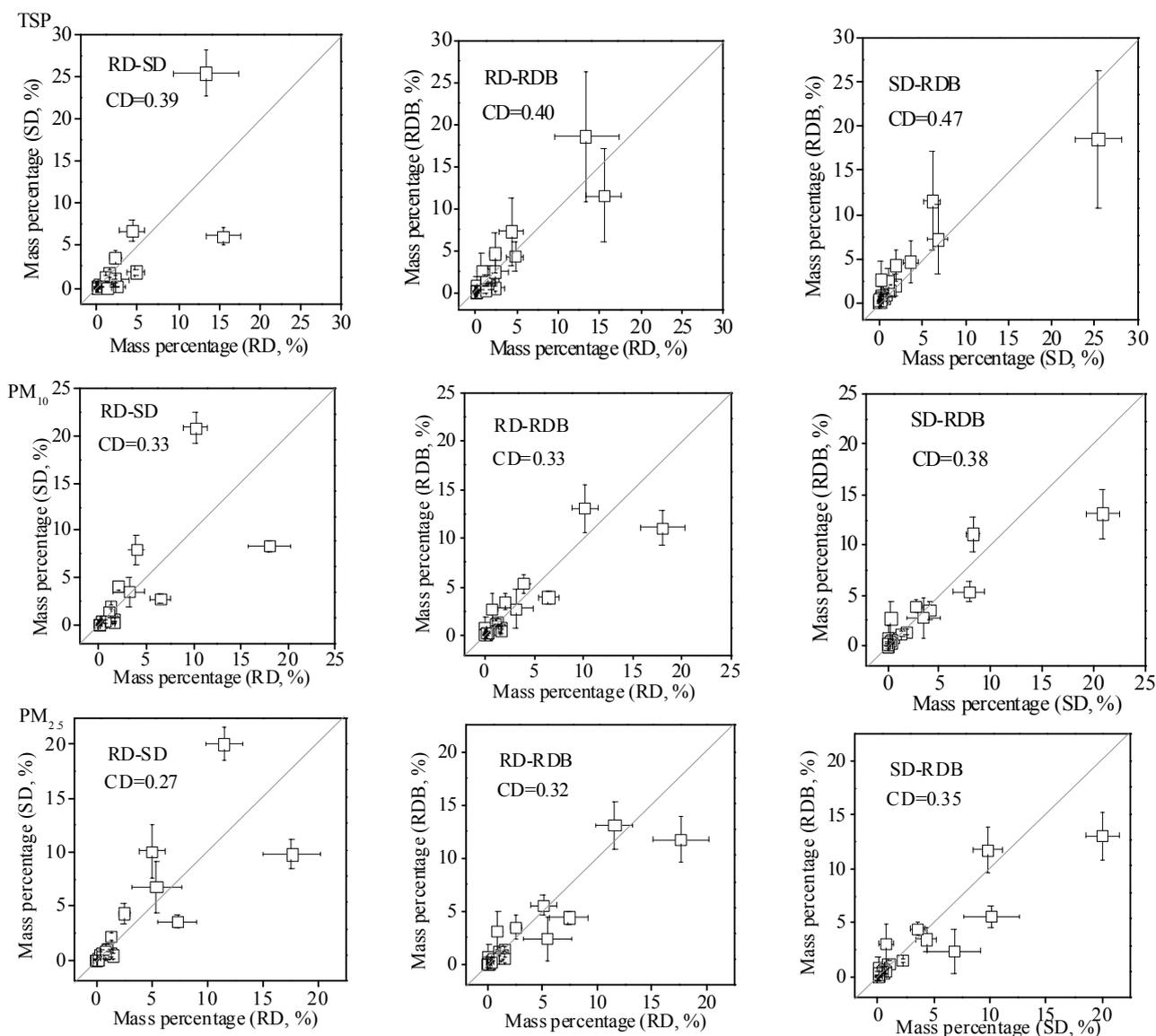
Fig. 5. Scatter diagrams for $PM_{2.5}$, PM_{10} and TSP profiles of different types of dust. SD: surface soil dust; RD: road dust; RDB: re-suspended dust.

source profiles for RDB within the three fractions could be substitute for each other while for RD and SD, the TSP-PM_{10} and PM_{10}-$PM_{2.5}$ profiles could substitute for each other, respectively. The differences in $PM_{2.5}$-TSP were more obvious than those in $PM_{2.5}$-PM_{10} and PM_{10}-TSP. For RDB, the CD values were all lower for PM_{10}-TSP, $PM_{2.5}$-TSP and $PM_{2.5}$-PM_{10} than those corresponding values of RD and SD indicating the diversities were low for this type of dust within different size fractions. Ho *et al.* (2003) also concluded that $PM_{2.5}$ and PM_{10} chemical abundances for a given source type were similar for most species.

Reconstructed versus Measured Mass

In Ho *et al.* (2003), crustal material was reconstructed by using the concentrations of elements including Al, Si, Ca, Ti and Fe to estimate the unmeasured oxides and was compared with the measured mass concentrations (by using

gravimetric measurement) with following equation used:

$$\text{Crustal material} = 1.16\,(1.90\text{Al} + 2.15\text{Si} + 1.41\text{Ca} + 1.67\text{Ti} + 2.09\text{Fe}) \qquad (3)$$

where the factor 1.16 was to compensate for the exclusion of MgO, Na_2O, K_2O and H_2O from crustal mass calculation (Ho *et al.*, 2003). Ho *et al.* (2003) concluded that only soil samples (PM_{10}) have good agreement with the calculated values and large variations of vehicular pollutants (OC, EC, Zn, and Pb) in paved road dust made the calculated values not match with the measured mass concentrations. Fig. 7 showed the scatter plots of calculated mass percentages versus measured mass percentages for SD, RD, and RDB in Dongying within $PM_{2.5}$, PM_{10} and TSP fraction, respectively.

Crustal material's mass percentages for SD of TSP, PM_{10} and $PM_{2.5}$ fractions exhibited good agreement with total
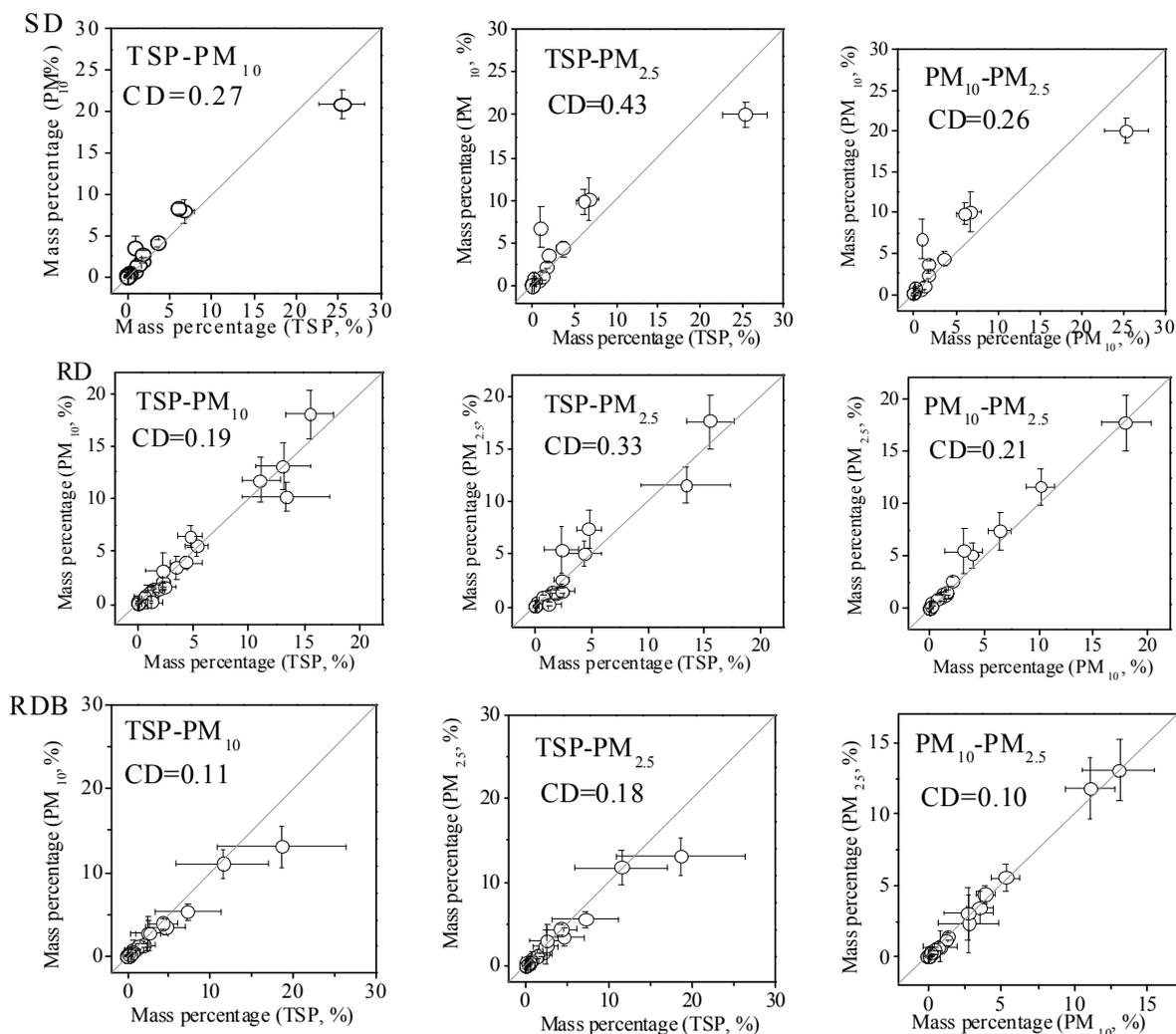
Fig. 6. Scatter diagrams for profiles of road, soil and re-suspended dust within different size fractions. SD: surface soil dust; RD: road dust; RDB: re-suspended dust.

measured mass percentages. For RD, crustal material of TSP and $PM_{2.5}$ fractions showed significant relationship with total measured mass percentages. While for RDB, it was only for TSP fraction. The correlation coefficient also exhibited decreasing trends with particles size increased for the three types of dust except for the values of PM_{10} and $PM_{2.5}$ for RD. It was the same to Ho *et al.* (2003) who pointed out that fine fraction of soil samples in Hong Kong made the calculated values not matched with the measured mass concentrations for all samples. Conclusion could be drawn that, for RD and RDB, their sources were more complex especially for finer particles in Dongying.

CONCLUSION

Chemical components including 39 elements, 9 ions, organic and elemental carbon of soil dust, road dust and re-suspended dust were determined in Dongying to obtain $PM_{2.5}$, PM_{10} and TSP source profiles. Ca, Si, OC, Ca^{2+}, Al, Fe and SO_4^{2-} were the most abundant components for all the three types of dust within the three size fractions. The

OC/TC ratios were highest for the dusts in Dongying which was related to its energy structure with lots of oil exploited and consumed. The mean mass percentage of Na^+ in TSP fraction of road dust was higher in new district than that in old district as the former was more close to sea. Meanwhile, the influence of sea salt was more obviously for coarse particles of road dust in Dongying. It was the same to Na^+ in re-suspended dust with the mean values as 31%, 31% and 13% higher in new district for $PM_{2.5}$, PM_{10} and TSP, respectively.

The higher EFs values for Cd, Ca, Cu, Zn, Ba, Ni, Pb, Cr, Mg and As suggested they were mainly influenced by anthropogenic sources including fossil fuel combustion, vehicle emission, construction and manufacturing activities. They tended to concentrate in finer fraction. The EFs for, Fe, Ti, Mn, Co, K, Na and V were mostly lower than 2 indicating deficiency to minimal enrichment and they tended to concentrate in coarser fraction.

Similarities of the profiles for different types of dust and different size fractions were compared by coefficient of divergence. Results showed that the CD values were mostly
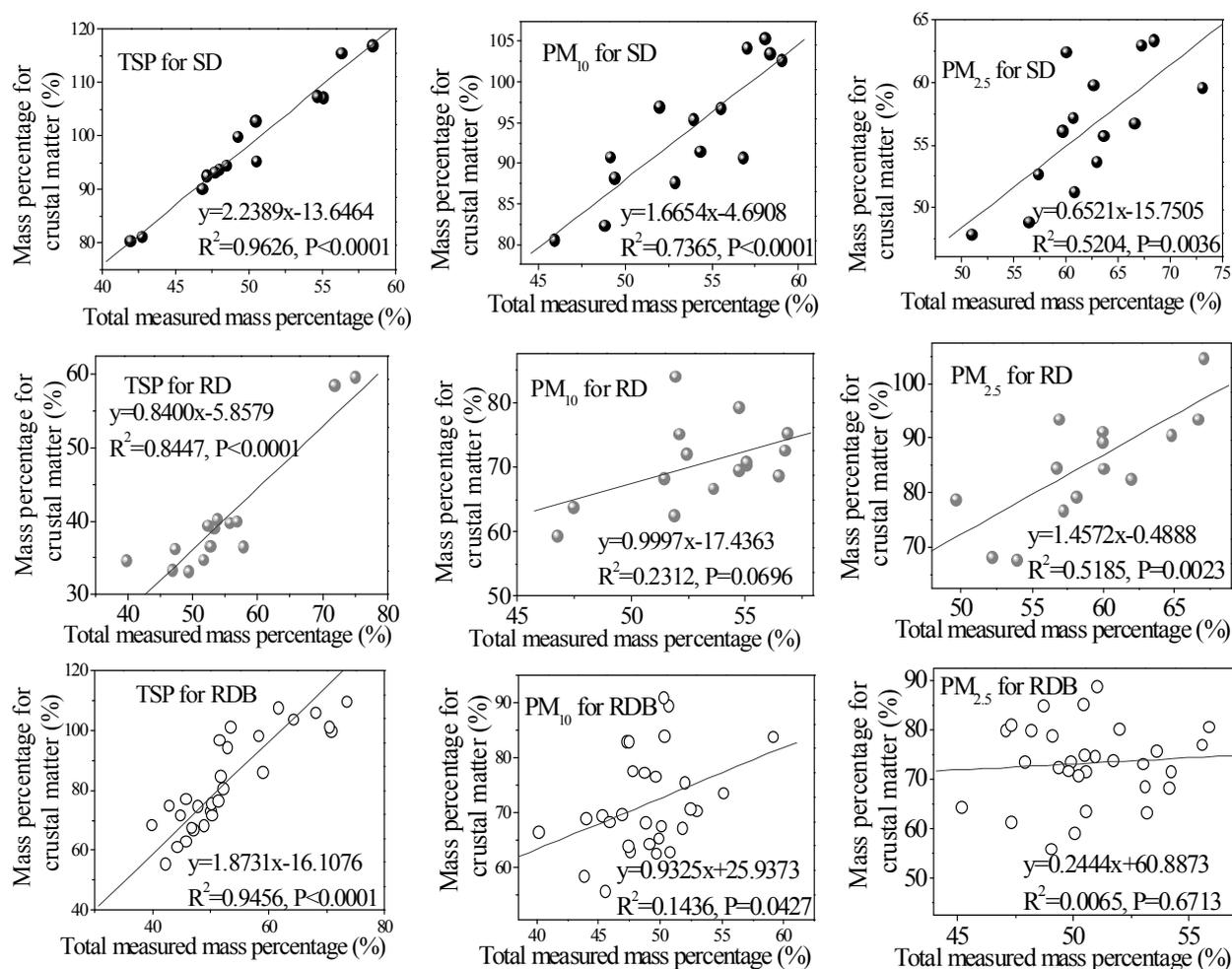
Fig. 7. Calculated crustal material mass percentages versus total measured mass percentages for soil dust, road dust, and resuspended dust with TSP, PM_{10} and $PM_{2.5}$ fractions. SD: surface soil dust; RD: road dust; RDB: re-suspended dust.

higher than 0.30 for comparison among different types of dust indicating that the profiles were different from each other. The CD values were lowest for SD-RD implying the soil dust and road dust may be influenced by some similar sources. For the comparison among different size fractions of each dust, the CD values were mostly lower or near to 0.30 indicating that the profiles were mostly similar to each other for the three size fractions. The source profiles for RDB within three fractions could be substitute for each other while for RD and SD, the TSP-PM_{10} and PM_{10}-$PM_{2.5}$ profiles could substitute for each other, respectively.

For road dust and resuspended dust, their sources were more complex especially for finer particles in Dongying according to the relationship analysis between crustal material mass percentages and the total mass percentages for all measured species. This study would be useful for understanding the similarities and differences of fugitive dust profiles and will benefit the source apportionment studies on fine particles.

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SUPPLEMENTARY MATERIALS

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

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