



Technical Note

Characterization of Inorganic Elements within PM_{2.5} and PM₁₀ Fractions of Fly Ashes from Coal-Fired Power Plants

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ABSTRACT

In this study 15 fly ash samples were collected from 15 large-scale coal-fired power plants (CFPPs) in China. The samples were then re-suspended through PM_{2.5} and PM₁₀ inlets and analyzed for the contents of 39 inorganic elements (IEs) using inductively coupled plasma-mass spectroscopy (ICP-MS) and inductively coupled plasma-optical emission spectrometry (ICP-OES). The results show that the particle size distributions for the 15 FAs samples exhibited bimodal patterns. The $\Sigma_{39}IEs$ (g g⁻¹) for the PM_{2.5} (0.292–0.564) in all the 15 CFPPs were higher than that of PM₁₀ (0.269–0.403). Except for Cu, all the other 38 IEs were more enriched in the PM_{2.5} with the PM_{2.5}/PM₁₀ ratios being 1.06–1.73. Considering 13 heavy metals, the same orders occurred between PM_{2.5} and PM₁₀ with Al >> Cr > Zn > Mn > Cu > V > Pb > Sn > Co > As > Sb > Tl > Cd. More attention should be paid to the high contents of Cr in both PM_{2.5} (1310 mg g⁻¹) and for PM₁₀ (1240 mg g⁻¹) from all 15 CFPPs. 23 IEs for PM₁₀ and 26 IEs for PM_{2.5} had the geo-accumulation index (I_{geo}) values higher than 0, indicating different pollution levels for them. On the other hand, there was moderate to extreme levels of pollution for Cr, Zn, Cu, Pb, Sn, Sb, Tl, Cd and Al based on I_{geo} values. The element profiles for PM_{2.5} or PM₁₀ from 15 CFPPs were similar based on low coefficients of divergence for PM_{2.5} (0.254 ± 0.038) and PM₁₀ (0.244 ± 0.054) according to the comparison between any two CFPPs. Most elements with low relative enrichment factors (REF) as less than 0.70 or 0.70–1.30 indicated no or weak condensation occurred for them during coal combustion, while Cr, Cu, Zn, Sn, W and Pb had REF values higher than 1.30 indicated that significant condensation occurred for these elements.

Keywords: Coal-fired power plant; Inorganic elements; Fly ash; PM_{2.5}; PM₁₀.

INTRODUCTION

Coal is still the most important energy source in the present China (Li *et al.*, 2014). More than half of coal consumed in China is used for power generation and this amount is projected to increase in the foreseeable future (You and Xu, 2010). Fly ashes (FAs) account for 80% of the total coal burning by-products in pulverized coal fired power plants

(CFPPs) (Janvijitsakul and Kuprianov, 2008). Large amounts of FAs originating from CFPPs are identified as problematic solid waste (Li *et al.*, 2016). China is the largest FAs producer and it is predicted that the production of 570–610 million tons will be reached by the year 2020 (Cao *et al.*, 2008; Li *et al.*, 2014). Clearly more and more FAs must be effectively disposed in the future in China (Li *et al.*, 2016). Although FAs have been widely used in many products and industries (Blissett and Rowson, 2012; Li *et al.*, 2014; Shaheen *et al.*, 2014; Li *et al.*, 2016), the utilization rate is still low in China. The rest 55% of FAs are stockpiled before landfill (Sahu *et al.*, 2009; Yao *et al.*, 2015; Li *et al.*, 2016). The reuse of FAs with different methods raises the concern about the characteristics of inorganic and organic pollutants in contained in the fly ashes (Baba *et al.*, 2010; Li *et al.*, 2014) including toxic compounds such as dioxins and PAHs (Cheruiyot *et al.*, 2015; Cheruiyot *et al.*, 2016).

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Coal contains a large number of IEs that are subsequently emitted in the FAs or react with surface chemicals of FAs during the combustion process (Choi *et al.*, 2002; Jegadeesan *et al.*, 2008). Higher levels of element contents in FAs are 4–10 times those of the feed coal, which should be an environmental concern (Akar *et al.*, 2012). The elements associated with stockpiled FAs may enter into soils and ground waters and result in adverse environment impacts (Li *et al.*, 2016). Padhy *et al.* (2016) found that the toxic elements such as Pb, Cr and Cd in plants that were grown in FAs amended soils tended to increase significantly under the influence of elements in FAs.

Atmospheric particulate matters (PM), especially particles with lower aerodynamic diameter, have received significant attention in the present China (Jiang *et al.*, 2015). The PM_{2.5} and PM₁₀ within FAs can be easily re-suspended by wind during their storage in the landfills and result in the atmospheric PM pollution. These PM_{2.5} and PM₁₀ can subsequently deposit on urban surfaces such as roads, soils, plants and building shells, then transported among environmental mediums by mixing force as wind, weathering, and gravitation and so on (Caravanos *et al.*, 2006; Zhao *et al.*, 2006; Shi *et al.*, 2008). IEs associated with these particles can accumulate in human body via inhalation, ingestion and dermal contact hence causing acute and chronic toxic effects such as neuropathies, rise in blood pressure, lung cancer and other diseases (Kong *et al.*, 2011; Li *et al.*, 2013; Dai *et al.*, 2015; Wei *et al.*, 2016; Zheng *et al.*, 2016). IEs associated with re-suspended PM_{2.5} and PM₁₀ from ash ponds by wind have been of major concern due to their toxicity and long residence time in atmosphere (Kong *et al.*, 2011; Li *et al.*, 2014). Laidlaw and Filippelli (2008) and Laidlaw *et al.* (2012) found the re-suspended particles from soils had become a persistent source of blood Pb for children in many USA cities.

The correlation between particle size of FAs and element contents had been studied widely due to most of elements were enriched in finer particles and hard to remove from flue gas (Sarkar *et al.*, 2006; Dai *et al.*, 2010). To our knowledge, no detailed data of elements within fractions of PM_{2.5} and PM₁₀ for FAs emitted from CFPPs was available (Xu *et al.*, 1997; Wang *et al.*, 1998; Li *et al.*, 2014; Liu *et al.*, 2015). So the study on inorganic elements abundant PM_{2.5} and PM₁₀ within FAs would be a novel study.

In this study, 15 FA samples were systematically collected from 15 CFPPs in China with mainstream individual block of power capacity of 600 MW. One paired of PM_{2.5} and PM₁₀ were sampled using re-suspension method for each FA for analysis of 39 IEs using ICP-MS and ICP-OES systems.

The main aims of this study were to investigate: 1) content distribution for sum of 39 IEs and individual element in both PM_{2.5} and PM₁₀; 2) geo-accumulation index (Igeo) assessment for analysis of pollution levels of individual element in PM_{2.5} and PM₁₀; 3) comparison of the similarity of the element composition profiles for both PM_{2.5} or PM₁₀ between any two CFPPs; 4) enrichment factors for each element were determined in order to analyze the behavior of particular elements during coal combustion.

MATERIALS AND METHODS

Sample Collection and Re-suspended for Obtaining of PM_{2.5} and PM₁₀

The FA samples were collected from 15 large-scale CFPPs during January to October, 2015. The same sampling method was adopted according to Li *et al.* (2016). The samples were collected from bottom ashes of electrostatic precipitators (ESP) using a stainless steel spade. The CFPP locations and corresponding administrative divisions where they are located are as shown in Fig. 1. Finally 1 kg of FA was collected from electrostatic precipitators of each CFPP and stored. All samples were dried using a vacuum freeze dryer, and then stored in brown glass bottles before being re-suspended for PM_{2.5} and PM₁₀ fractions.

The particles with aerodynamic diameters less than 2.5 μm (PM_{2.5}) and 10 μm (PM₁₀) were re-collected from fly ashes using re-suspension method (Chow *et al.*, 1994; Kong *et al.*, 2011). Fig. 2 showed the illustrative diagram of re-suspension chamber. For each paired of PM_{2.5} and PM₁₀, 0.5 g of FA was blow out from dust feeding bottle using a dust feeding pump and injected into the re-suspension chamber and then through particle size cutters (2.5 and 10 μm) and intercepted by Teflon filter (diameter 47 mm; Pall Co. USA). Suspension time was 5 min for each sample, and then 40 min was needed to eliminate the interference of last suspension sample. All the Teflon filters were equilibrated for 72 h in a room with constant temperature and humidity before and after sampling, and then weighed. The sampling amount was calculated based on the mass difference of the filters before and after sampling. The sampling amounts ranged from 1110 to 8030 μg for PM₁₀ and 1150 to 6040 μg for PM_{2.5}, respectively.

Particle Size Analysis for FAs and Element Analysis within PM_{2.5} and PM₁₀ Fractions

The particle size distribution of FA was characterized by laser-based analyzer (BT-9300H, Suzhou Qile Electronic Technology Co., China) and water was used as medium. The 30 IEs including Li, Be, Na, P, K, Sc, V, Cr, Mn, Co, Ni, Cu, Zn, As, Rb, Y, Mo, Cd, Sn, Sb, Cs, La, Ce, Sm, W, Tl, Pb, Bi and Th were analyzed by ICP-MS system (Agilent 7500a, Agilent Co. USA). The rest 9 IEs such as Si, Al, Ca, Mg, Fe, Ti, Ba, Sr and Zr were analyzed by ICP-OES system (Agilent 5100, Agilent Co. USA).

For ICP-MS analysis, the 1/2 of Teflon filter was cut into pieces by a ceramic scissors and stored in a Teflon crucible. The 5 mL of aqua regia (V:V = 1:5) and 1 drop of HF acid (V:V = 1:1) were added and heated at 120°C for 2 h and then raised to 130°C until it evaporated to dryness. Then 10 mL 2% of HCl acid was added and heated for 20 min and the extract was transferred into a plastic comparison tubes before analysis. For ICP-OES analysis, the remaining 1/2 filter was cut into pieces and stored in a Teflon crucible, then heated at 300°C for 40 min in a muffle furnace, then gradually heated up to 530–550°C for ashing completely. A few drops of absolute ethanol and 0.1–0.2 g NaOH were added and heated for 10 min at 500°C in a muffle furnace. Water was added and boiled on an electric heating plate and



Fig. 1. Location distributions of 15 CFPPs in Chinese administrative divisions.

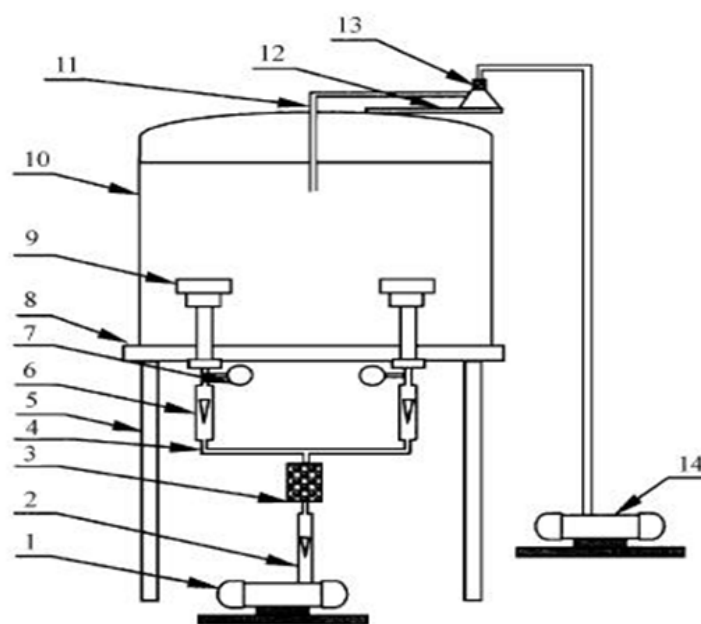


Fig. 2. Re-suspension sampling system of Nankai University. (1) Sampling pump, (2) Total flow meter, (3) Filter, (4) Sampling gas path, (5) Support shelf, (6) Branch flow meter, (7) Vacuum gauge, (8) Sampling platform, (9) Stainless steel particle size cutter, (10) Re-suspension chamber, (11) Gas path of dust injection, (12) Stainless steel bottle holder, (13) Dust feeding bottle, (14) Dust feeding pump.

then transferred into a polyvinyl chloride colorimetric tube with 2 mL HCl (V:V = 1:1) added. The extract was finally diluted to 10 mL with high pure water before analysis.

The method detection limits (MDLs) ranged from 0.100 ng for Tl, Bi and Th, Cs, Sm to 764 ng for Na with the mean value as 76.7 ± 234 ng for 30 IEs analyzed by ICP/MS. The MDLs for 9 IEs analyzed by ICP-OES were in the range of 4.00 ng for Sr to 1810 ng for Fe with the mean value as 459 ± 559 ng. The accuracy of ICP-MS and -OES was determined by detection of standard soil materials, GBW07446-GBW07457, purchased from Center for National Standard Matter. The relative standard deviation (RSD) values for 39 IEs such as Li, Be, Na, P, K, Sc, V, Cr, Mn, Co, Ni, Cu, Zn, As, Rb, Y, Mo, Cd, Sn, Sb, Cs, La, Ce, Sm, W, Tl, Pb, Bi, Th, Si, Al, Ca, Mg, Fe, Ti, Ba, Sr, Zr and U were 2.58%, 2.63%, 3.10%, 3.95%, 1.85%, 2.99%, 3.26%, 4.12%, 6.20%, 2.59%, 2.68%, 3.27%, 4.10%, 1.98%, 2.75%, 3.21%, 4.22%, 5.10%, 3.24%, 2.66%, 2.55%, 3.69%, 4.29%, 4.38%, 6.24%, 2.16%, 2.91%, 2.89%, 3.65%, 3.88%, 4.19%, 4.39%, 3.25%, 2.77%, 2.51%, 3.67%, 6.98%, 4.09% and 2.63%, respectively. The procedural and solvent blank experiments were routinely conducted to detect the background contamination. The instrument repeatability was assessed by detection of one standard material every 5 field samples. The recoveries for 39 elements by technique adopted in this study were in the range of 80–120%. In addition, one sample duplicate

experiment was conducted every 8 samples with the RSD less than 15%.

RESULTS AND DISCUSSION

Size Distribution of CFAs from 15 CFPPs

Particle size is one of the most critical features for fly ashes from CFPPs (Li *et al.*, 2014). Fine sized particles, especially $PM_{2.5}$ and PM_{10} can easily enter into the atmosphere by wind power during the storage of FAs and subsequently result in human respiratory system diseases (Harris and Davidson, 2009; Kong *et al.*, 2011; Zhang *et al.*, 2012; Kulshrestha *et al.*, 2014; Li *et al.*, 2014; Zhang *et al.*, 2015). Table 1S and Fig. 3 list and show the particle size distribution of FAs from 15 CFPPs. In ordinate of Fig. 3, the volume (%) was the volume percentage of particles with specific size. Results indicated that 50% particles for 15 CFPPs ranged from 12.2 μm for FA15 to 55.1 μm for FA6, 75% particles were of 31.5–95.2 μm and 90% particles were of 57.5–142 μm indicating a bimodal distribution. As shown in Fig. 3, there were two peaks in each particle size distribution curve. Generally these curves were firstly peaked at about 0.76–1.18 μm and secondly peaked at about 21.1–84.9 μm for 15 FA samples, the particle size span for second one was wider compared to the first one. The same bimodal distribution pattern was also reported elsewhere (Lind *et al.*, 2003; Arkar *et al.*, 2012; Li *et al.*, 2014; Verma

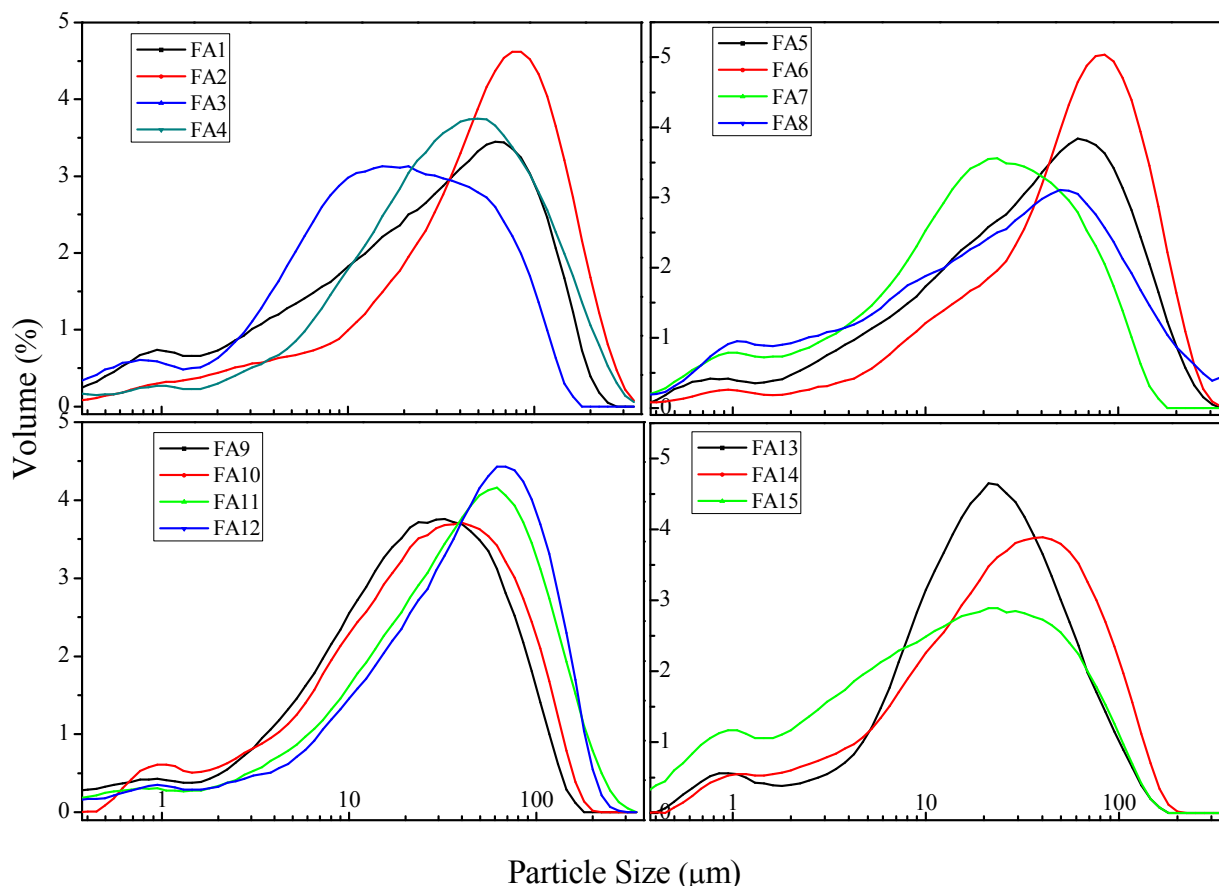


Fig. 3. Particle size distribution curves for fly ashes from 15 CFPPs.

et al., 2015). It is generally believed that evaporation, condensation and nucleation of inorganic components in feed coal is responsible for the first peak, while the second peak was results from fusion and coalescence of inorganic components (Linak and Wendt, 1994; Yu, 2004; Li *et al.*, 2014). Another hypothesis was proposed by Lin *et al.* (2002), whereby they asserted that the injection of limestone into furnace for desulphurization resulted in this bimodal particle size distribution. In addition, Li *et al.* (2015) reported there were four peaks in size distribution curve for FAs emitted from industrial solid waste incineration. The difference of combustion conditions and feed fuels could be the major reason for these observations.

Contents of 39 Elements and Individual Element for 15 CFPPs

As shown in Fig. 4, the highest $\Sigma_{39}\text{IEs}$ for $\text{PM}_{2.5}$ occurred for FA9 (0.56 g g^{-1}) and PM_{10} occurred for FA1 (0.40 g g^{-1}). The $\Sigma_{39}\text{IEs}$ varied from 0.27 to 0.40 g g^{-1} for PM_{10} and 0.29 to 0.56 g g^{-1} for $\text{PM}_{2.5}$, with the mean value being 0.29 ± 0.05 and $0.40 \pm 0.08 \text{ g g}^{-1}$, indicated the IEs were more enriched in lower sized particles. The same conclusions were drawn by other studies (Xu *et al.*, 1997; Wang *et al.*, 1998; Raclavska *et al.*, 2009; Kong *et al.*, 2011; Li *et al.*, 2014; Liu *et al.*, 2015). Xu *et al.* (1997) reported the contents for 22 IEs including Al, Fe, Ti, As, Co, Sc, Cu, Ga, Ge, Hf, Mo, Ni, Pb, Th, Sr, Li, U, V, Mo, Y, Zn, Be and Se increased with the decreasing of particle size of FAs from a Nanjing Power plant. Wang *et al.* (1998) studied the 16 IEs in coal fly ashes from different type of boilers and indicated the most elements increased with the decrease in particle size. Raclavska *et al.* (2009) indicated the contents for Ti, Fe, Na, Mn, Ca, Mg and P in FAs were in line with this trend. Kong *et al.* (2011) reported the sum of Zn, Cr, Cu, Pb, Ni, As and Cd in re-suspended dust from building surface followed the order as $\text{PM}_{2.5} > \text{PM}_{10} > \text{PM}_{100}$. Li *et*

al. (2014) reported Be, Cu, Ni, V, Se, Mo and Cd in FAs from a combined heating and power plant in Anhui also exhibited a similar trend. Liu *et al.* (2015) reported As, Cu, Pb, Zn and Cd were in accordance with this trend, while Hg did not adhere to this trend due to its high volatility.

The burning up of feed coal and excessive oxygen were more important than the other factors such as coal type and plants design during the formation organic pollutants (Revuelta *et al.*, 1999; Cheruiyot *et al.*, 2015). For IEs, the incomplete combustion of coal may be a key factor. The TOC values were further detected for 15 FAs to assess the relationship between IEs and TOC. $\Sigma_{39}\text{IEs}$ for PM_{10} were well negative correlated with TOC of FAs ($R = -0.98$, $p < 0.005$), and were weak correlated with TOC for $\text{PM}_{2.5}$ ($R = -0.61$, $p < 0.01$), indicated the less evaporation amounts of IEs during incomplete combustion of coal.

Fig. 5 shows the average individual element content in PM_{10} and $\text{PM}_{2.5}$, generally the content distributions of 39 IEs were similar between PM_{10} and $\text{PM}_{2.5}$. For PM_{10} , the IEs contents decreased as shown in the following trend $\text{Al} > \text{Si} > \text{Ca} > \text{Fe} > \text{K} > \text{Mg} > \text{Na} > \text{Ti} > \text{Cr} > \text{P} > \text{Zn} > \text{Sr} > \text{Ba} > \text{Zr} > \text{Mn} > \text{Cu} > \text{V} > \text{Ce} > \text{Pb} > \text{Li} > \text{La} > \text{Y} > \text{Ni} > \text{Rb} > \text{Sn} > \text{Th} > \text{Co} > \text{Sc} > \text{As} > \text{U} > \text{Mo} > \text{Sm} > \text{W} > \text{Sb} > \text{Be} > \text{Cs} > \text{Bi} > \text{Tl} > \text{Cd}$, and it was $\text{Al} > \text{Si} > \text{Ca} > \text{Fe} > \text{K} > \text{Mg} > \text{Na} > \text{Ti} > \text{Cr} > \text{P} > \text{Sr} > \text{Ba} > \text{Zn} > \text{Zr} > \text{Mn} > \text{Cu} > \text{V} > \text{Ce} > \text{Li} > \text{Pb} > \text{La} > \text{Y} > \text{Ni} > \text{Rb} > \text{Sn} > \text{Th} > \text{Co} > \text{As} > \text{Sc} > \text{U} > \text{Mo} > \text{Sm} > \text{W} > \text{Be} > \text{Sb} > \text{Cs} > \text{Bi} > \text{Tl} > \text{Cd}$ for $\text{PM}_{2.5}$. The average contents for 39 IEs in $\text{PM}_{2.5}$ were correlated well with those in PM_{10} ($R^2 = 0.99$, $p < 0.005$). Higher Ca contents in $\text{PM}_{2.5}$ and PM_{10} in this study indicated the influence of desulphurization of coal by limestone injection into furnace in 15 CFPPs (Li *et al.*, 2014). Li *et al.* (2014) found CaO content accounts for as high as 6.97% of major oxides due to the addition of limestone, while it was only 1.23% for Chinese coal (Dai *et al.*, 2012).

Among 39 IEs, 13 heavy metals (HMs) such as Pb, Cr,

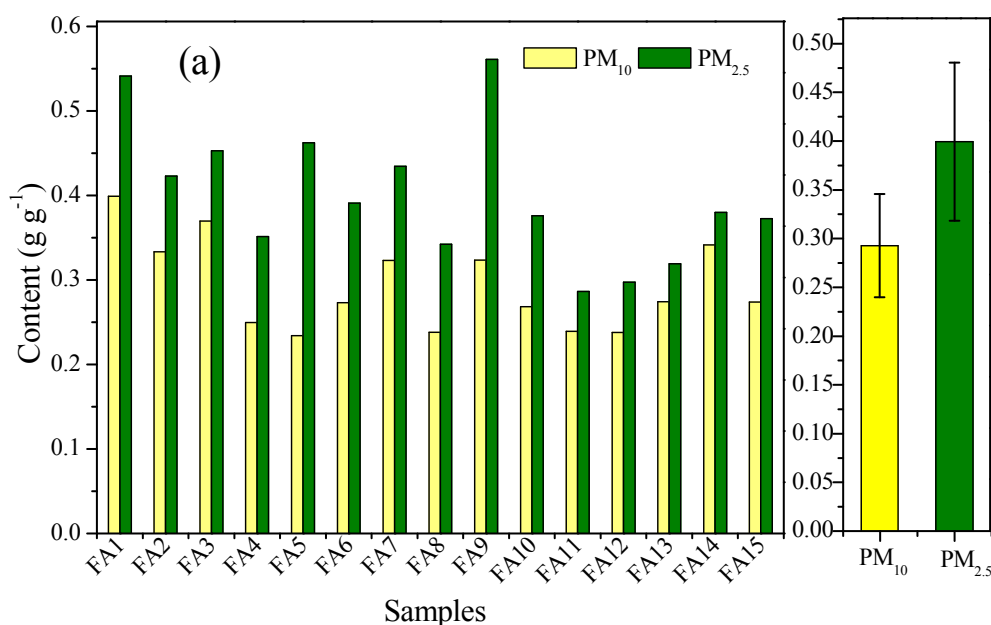


Fig. 4. (a) Contents of 39 IEs in $\text{PM}_{2.5}$ and PM_{10} from 15 CFPPs and (b) Mean value for $\text{PM}_{2.5}$ and PM_{10} .

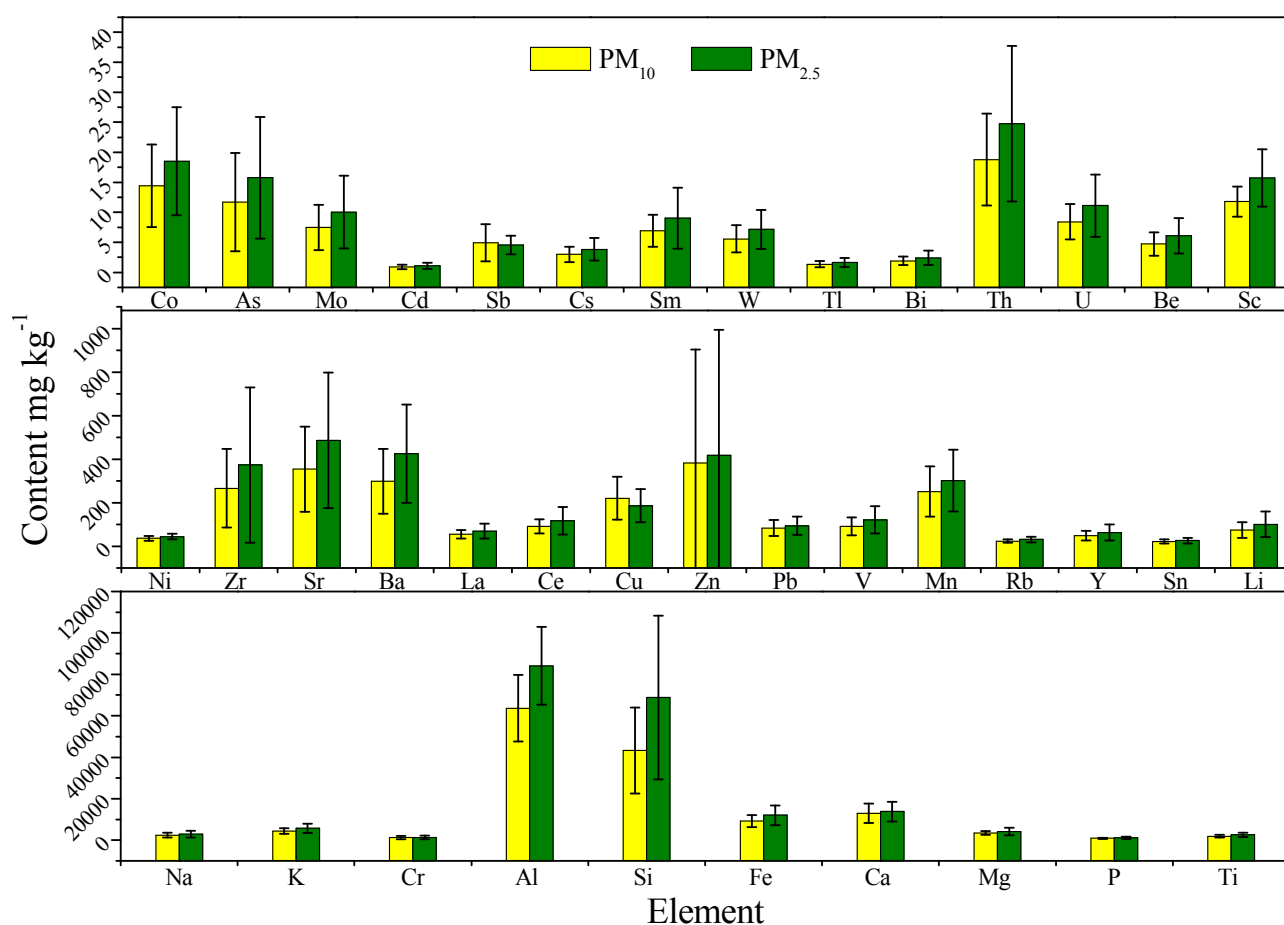


Fig. 5. Individual element content distribution for PM_{10} and $PM_{2.5}$ of 15 CFPPs.

Cd, As, Al, Co, V, Sb, Mn, Sn, Tl, Cu and Zn received more attention due to their adverse health effects to people. The contents ($mg\ kg^{-1}$) for these HMs were as follows Al (63700 ± 16100) > Cr (1240 ± 791) > Zn (383 ± 221) > Mn (251 ± 115) > Cu (220 ± 521) > V (92.1 ± 41.2) > Pb (83.6 ± 36.4) > Sn (21.9 ± 9.06) > Co (14.4 ± 6.86) > As (11.7 ± 8.19) > Sb (4.95 ± 3.11) > Tl (1.37 ± 0.55) > Cd (0.91 ± 0.38) for PM_{10} , and the same order were found for $PM_{2.5}$ as Al (84100 ± 18800) > Cr (1310 ± 888) > Zn (417 ± 576) > Mn (301 ± 142) > Cu (186 ± 74.5) > V (121 ± 62.4) > Pb (93.9 ± 42.5) > Sn (25.9 ± 12.6) > Co (18.5 ± 9.01) > As (15.8 ± 10.1) > Sb (4.56 ± 1.54) > Tl (1.63 ± 0.741) > Cd (1.08 ± 0.491). This order was significantly different from that as Cu > Zn > Pb > Cr > Ni > Sb > As for 13 bottom ashes during burning of various wastes, wooden and waste/wooden mixture, possibly explained by the difference in combustion conditions and feed fuels (Saqib and Bäckström, 2016). It should be mentioned that more attention should be paid to higher levels of Al, Cr, Mn and V in $PM_{2.5}$ and PM_{10} . Higher level of Cr was possibly attributed to the evaporation from the Cr containing stainless steel materials extensively used in boilers of CFPPs.

In order to investigate the particle size distribution of 39 IEs, the content ratio of $PM_{2.5}$ to PM_{10} for each element was calculated and showed in Fig. 6. The $PM_{2.5}/PM_{10}$ ranged from 0.89 for Cu to 1.72 for Si with the mean value as 1.29

± 0.13 . Except for Cu, $PM_{2.5}/PM_{10}$ ratios for the remaining 38 IEs were higher than unity, indicating that they were more inclined to be enriched in $PM_{2.5}$ compared to PM_{10} . The differences in volatilization and adsorption mechanisms among IEs could be the reasons for the observed variations. Seames (2003) reported that evaporated IEs were more easily condensed in particles with high specific surface area. The fine-sized particles had larger surface area, higher adsorption activity and ability compared to the coarse ones and resulted in higher contents of IEs in the fine particles (Xu *et al.*, 1997; Wang *et al.*, 1998; Liu *et al.*, 2015). IEs with relatively higher volatilization, evaporated from coal at high temperature and physically adsorbed by surface of FAs during cooling of flue gas, which was named as volatilization and condensation mechanism (Chen *et al.*, 2001). For elements with lower volatilization, chemical reaction adsorption between element and surface chemicals of FAs would be the main process (Li *et al.*, 2014).

Geo-accumulation Index Assessment for Each Element within PM_{10} and $PM_{2.5}$

Geo-accumulation index (Igeo) proposed by Muller has been widely used in element studies (Muller, 1969; Wei *et al.*, 2009; Kong *et al.*, 2011; Li *et al.*, 2013).

$$I_{geo} = \text{Log}_2(C_n/1.5B_n) \quad (1)$$

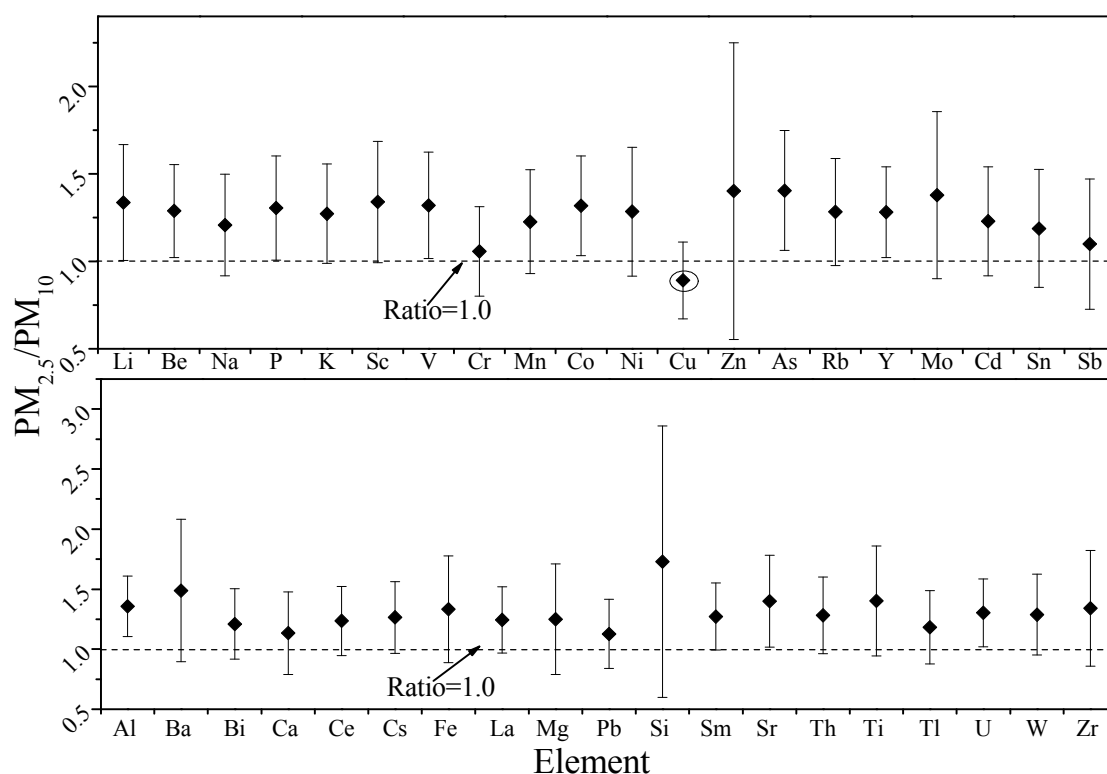


Fig. 6. Content ratio of each element between $PM_{2.5}$ and PM_{10} .

where, C_n is the measured content of element in FAs, B_n is the background value of elements in soil. The constant of 1.5 allowed evaluating natural fluctuation of a given element in environment and analyzing the slight anthropogenic effects (Wei *et al.*, 2009). In this study, the background values of IEs in soils for different Chinese administrative divisions where CFPPs located were presented in CNEMC. (1990) and listed in Table 2S. As shown in Table 1, the pollution levels of IEs were further identified as seven categories based on their Igeo values.

Among 39 IEs involved in this study, only 37 background values for corresponding IEs were found in CNEMC. (1990), so Igeo values were calculated for these 37 IEs. Generally Igeo values for 36 IEs except for Cu in $PM_{2.5}$ were higher than those in PM_{10} (Fig. 7). In case of PM_{10} , Class B including Tl, U, W, Y, Zn, Be, Li and Sr indicated uncontaminated to moderately contaminated, C including Sb, Pb, Bi and Mo indicated moderately contaminated, D including Cd, Sn and Cu indicated moderately to heavily contaminated state, E including Cr indicated heavily contaminated, G including K, Na, Ti, Fe, Mg, Ca and Al indicated extremely contaminated, and the rest 14 elements belong to A indicated uncontaminated state. For 37 elements in $PM_{2.5}$, 10 IEs as Th, Tl, W, Sm, Ce, La, Y, Ni, Be and Li were belong to B, 6 IEs as U, Zn, Sb, Pb, Sr and Mo were belong to C, 3 IEs as Cu, Sn and Cd were belong to D, Cr was belong to E, 7 IEs as K, Na, Ti, Fe, Mg, Ca and Al were belong to G, and rest 11 IEs were belong to A. For 13 HMs, 8 metals as Tl, Zn, Sb, Pb, Cu, Cd, Cr and Al in $PM_{2.5}$ and PM_{10} with Igeo > 0 indicated different pollution levels of them.

Similarity Comparison of Element Profiles for $PM_{2.5}$ and PM_{10} among 15 CFPPs

The fine-sized PM, especially for $PM_{2.5}$ and PM_{10} , would result in a series of adverse impacts on atmospheric quality and human health, such as visibility reduction, deterioration of ecosystem through deposit of toxic pollutants within them, change of earth radiation budget and global climate and so on (Kchih *et al.*, 2015). PM has become a severe environmental challenge to the present China. As a receptor model, chemical mass balance (CMB) model has been widely used for source apportionment of PM (Moreno *et al.*, 2009; Kong *et al.*, 2010). Accurate source profiles were the basis of the calculation result of the CMB (Kong *et al.*, 2010).

FAs emitted from CFPPs were an important PM source and establishment of element profiles for them from every CFPP was a heavy work. The similarity of element profiles among different CFPPs made they can replace each other, and then simplified this work. So the similarity of element profiles among different $PM_{2.5}$ or PM_{10} samples was identified by a parameter of coefficient of divergence (CD) in this study. CD was calculated as followed:

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

where j and k were the different sources (refer to different CFPPs in this study), p was the number of analyzed elements, and x_{ij} and x_{ik} were the mean masses of component i (refer to 39 elements in this study) for j and k (Wongphatarakul

Table 1. Seven categories of Igeo value for each trace element.

Class	Igeo index	Degree of contamination for element
A	$I_{geo} \leq 0$	uncontaminated
B	$0 < I_{geo} \leq 1$	uncontaminated to moderately contaminated
C	$1 < I_{geo} \leq 2$	moderately contaminated
D	$2 < I_{geo} \leq 3$	Moderately to heavily contaminated
E	$3 < I_{geo} \leq 4$	heavily contaminated
F	$4 < I_{geo} \leq 5$	heavily to extremely contaminated
G	$5 < I_{geo}$	extremely contaminated

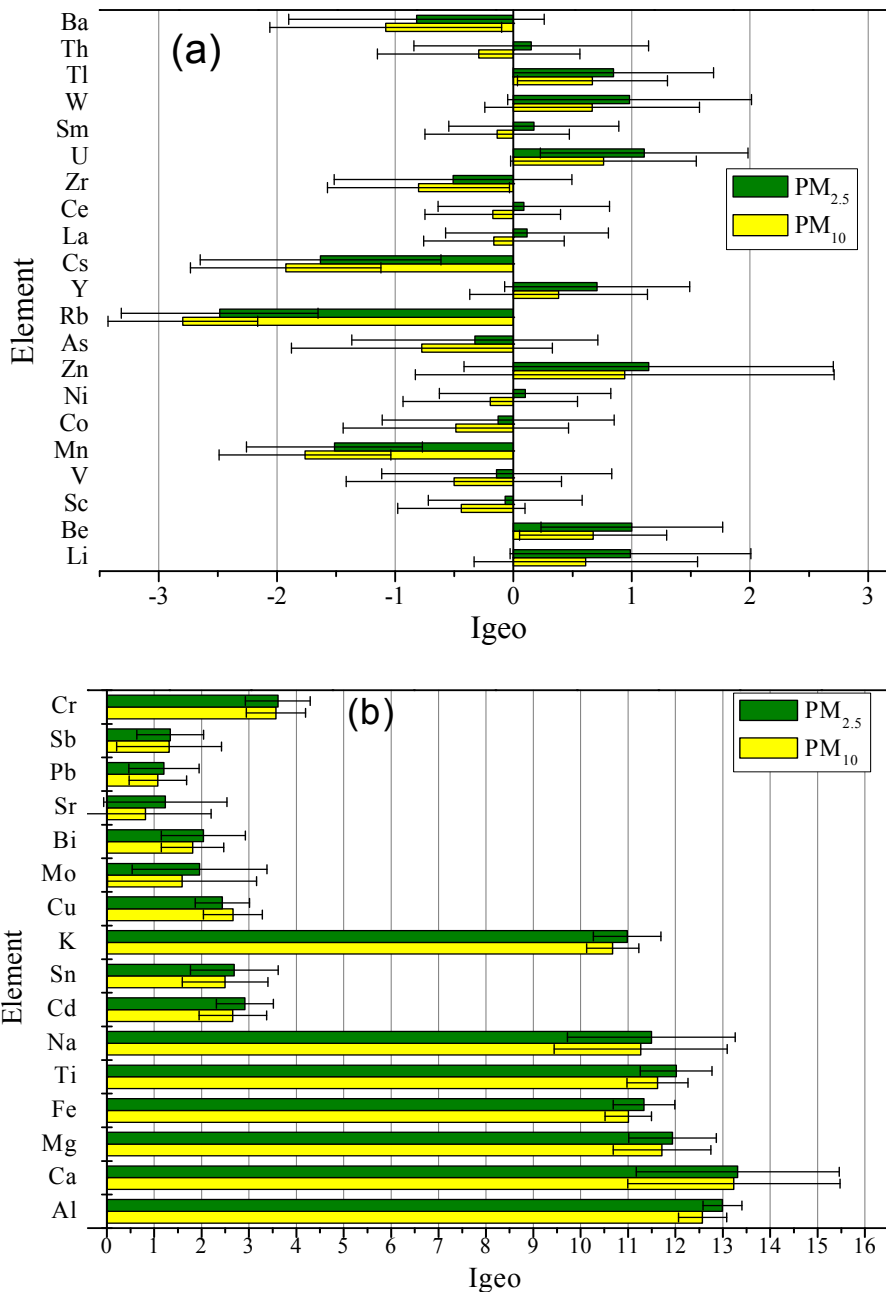


Fig. 7. Igeo values for each element in PM₁₀ and PM_{2.5}.

et al., 1998; Li et al., 2016). The CD value of 0 indicated source profiles of j and k were completely same, 1 indicated that they were completely different, and less than 0.3

suggested they were similar (Wongphatarakul et al., 1998).

In this study, CD values for 105 paired of element profiles for PM_{2.5} or 105 paired of those for PM₁₀ were calculated to

investigate the similarity of PM_{2.5} or PM₁₀ between any two CFPPs. Results showed that there were only 8 points higher than 0.30 among 105 points for PM_{2.5}, and were only 10 points higher than 0.30 among 105 points for PM₁₀ (Fig. 8). The CD values for 105 paired of data were 0.254 ± 0.038 for PM_{2.5}, and 0.244 ± 0.054 for PM₁₀, indicated the similarity of element profiles of PM_{2.5} or PM₁₀ from 15 CFPPs. In short, the element profiles for PM_{2.5} or PM₁₀ may be substituted for each other. This discovery would significantly simplify the work of element profiles of re-suspended particles from ash ponds.

Enrichment Factors Analysis

To evaluate the behavior of 39 IEs during coal burning process, a parameter named as relative enrichment factor (REF) was proposed by Meij (1994) and calculated for each element as follows:

$$REF = \frac{A}{B} \times C \quad (3)$$

where A refer to element content in FAs (mg kg^{-1}), B refer to that in coal (mg kg^{-1}), and C refer to ash content of coal (%). REF values were in the range of 0.70–1.30 indicated no significant enrichment and exhaustion occurred, higher than 1.30 indicated the significant enrichment occurred, and lower than 0.70 indicated the exhaustion occurred (Meij, 1994; Lu *et al.*, 2009). Li *et al.* (2014) reported there were three categories according to REF values: 1) elements with REF around 1 indicated they were not evaporated during burning process; 2) with REF around 0.7 indicated they were evaporated during burning process; 3) with a very low REF indicated they were completely evaporated during burning process.

Dai *et al.* (2012) reported the background contents of 51 elements and 10 element oxides for Chinese coal. The average ash content of 21.5% for Chinese coal was also reported.

Among these elements, 28 elements were involved in this study such as Li, Sc, V, Cr, Co, Ni, Cu, Zn, As, Rb, Sr, Y, Zr, Mo, Cd, Sn, Sb, Cs, Ba, La, Ce, Sm, Tl, Pb, Bi, Th, U and W with their contents (reported in mg kg^{-1}) as 31.8, 4.38, 35.1, 15.4, 7.08, 13.7, 17.5, 41.4, 3.79, 9.25, 140, 18.2, 89.5, 3.08, 0.25, 2.11, 0.84, 1.13, 159, 22.5, 46.7, 4.07, 0.47, 15.1, 0.79, 5.84, 2.43 and 1.08, respectively. The contents of Al, Si, P, Ti, K, Mg, Ca, Na, Mn and Fe were calculated by conversion of their oxides.

REF values for 39 elements were calculated by Eq. (3), and the results were shown in Table 2. According to the conclusion drawn by Meij in 1994 and for 39 IEs in PM_{2.5}: 17 IEs including Li, Be, Na, P, Mn, Co, La, Ce, Sm, Bi, Al, Mg, Ti, Ca, Fe, Ba and Si with REF < 0.70 indicated the evaporation, and no condensation or weak condensation occurred for them during coal combustion, 16 IEs including K, Sc, V, Ni, As, Rb, Y, Mo, Cd, Sb, Cs, Tl, Th, U, Zr and Sr with REF of 0.70–1.30 indicated no evaporation and condensation occurred for these elements during coal combustion, and 6 IEs including Cr, Cu, Zn, Sn, W and Pb with REF > 1.30 indicated significant condensation occurred for these elements. In case of 39 IEs elements in PM₁₀, 30 IEs were belonged to the first class with REF < 0.7, Cd, Sb, W, Pb and U were belonged to the second class with RE as 0.70–1.30, and Cr, Cu, Zn and Sn were identified as the third class with REF > 1.30. Generally the IEs in PM_{2.5} had higher REF values than corresponding elements in PM₁₀. It was should be mentioned, Cr in PM_{2.5} and PM₁₀ had REF as high as 18.3 and 17.4, may be explained significant condensation occurred during coal combustion or using of large amounts of Cr containing stainless steel materials in furnaces of 15 CFPPs.

CONCLUSTIONS

Thirty nine IEs such as Li, Be, Na, P, K, Sc, V, Cr, Mn, Co, Ni, Cu, Zn, As, Rb, Y, Mo, Cd, Sn, Sb, Cs, La, Ce, Sm,

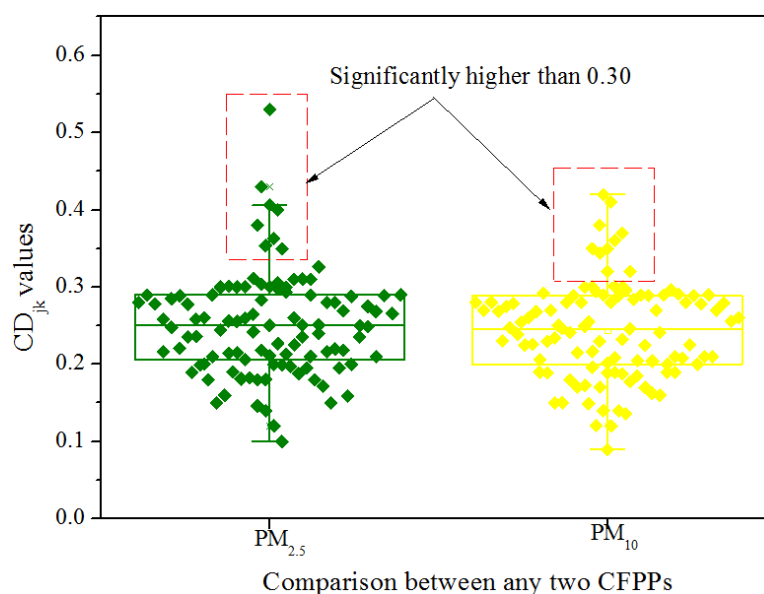


Fig. 8. Box chart for coefficient of divergence values of PM_{2.5} and PM₁₀.

Table 2. Relative enrichment factors of 39 elements in PM₁₀ and PM_{2.5}.

Element	REF (mean ± SD)		Element	REF (mean ± SD)	
	PM ₁₀	PM _{2.5}		PM ₁₀	PM _{2.5}
Li	0.506 ± 0.237	0.682 ± 0.344	Cs	0.569 ± 0.234	0.734 ± 0.361
Be	0.483 ± 0.192	0.624 ± 0.246	La	0.525 ± 0.187	0.661 ± 0.322
Na	0.450 ± 0.195	0.546 ± 0.292	Ce	0.423 ± 0.145	0.539 ± 0.289
P	0.470 ± 0.155	0.621 ± 0.276	Sm	0.367 ± 0.137	0.479 ± 0.269
K	0.608 ± 0.177	0.781 ± 0.316	W	1.11 ± 0.436	1.43 ± 0.541
Sc	0.581 ± 0.119	0.773 ± 0.234	Tl	0.629 ± 0.242	0.745 ± 0.339
V	0.565 ± 0.244	0.743 ± 0.383	Pb	1.19 ± 0.502	1.34 ± 0.606
Cr	17.4 ± 10.7	18.3 ± 12.4	Bi	0.526 ± 0.194	0.660 ± 0.328
Mn	0.465 ± 0.206	0.559 ± 0.264	Th	0.693 ± 0.272	0.913 ± 0.478
Co	0.439 ± 0.202	0.563 ± 0.274	U	0.746 ± 0.252	0.985 ± 0.457
Ni	0.571 ± 0.172	0.706 ± 0.191	Zr	0.641 ± 0.419	0.898 ± 0.856
Cu	2.71 ± 1.16	2.29 ± 0.941	Al	0.433 ± 0.106	0.572 ± 0.128
Zn	1.99 ± 0.424	2.17 ± 0.584	Sr	0.545 ± 0.290	0.754 ± 0.478
As	0.664 ± 0.449	0.896 ± 0.576	Mg	0.567 ± 0.133	0.694 ± 0.290
Rb	0.554 ± 0.179	0.718 ± 0.302	Ti	0.204 ± 0.067	0.276 ± 0.116
Y	0.576 ± 0.261	0.745 ± 0.441	Ca	0.392 ± 0.134	0.417 ± 0.145
Mo	0.524 ± 0.255	0.702 ± 0.422	Fe	0.059 ± 0.017	0.076 ± 0.031
Cd	0.781 ± 0.320	0.933 ± 0.423	Ba	0.404 ± 0.195	0.576 ± 0.305
Sn	2.24 ± 0.694	2.65 ± 1.08	Si	0.229 ± 0.106	0.365 ± 0.210
Sb	1.27 ± 0.568	1.17 ± 0.194			

W, Tl, Pb, Bi, Th, Si, Al, Ca, Mg, Fe, Ti, Ba, Sr, Zr and U within PM_{2.5} and PM₁₀ fractions of FAs collected from 15 CFPPs were analyzed using ICP-MS and ICP-OES. The sum of 39 IEs within PM_{2.5} (0.292–0.564 g g⁻¹) were higher than those of PM₁₀ (0.269–0.403 g g⁻¹), indicated the IEs were more inclined to be enriched in fine sized particles. For individual element content, all the 39 IEs except for Cu and Sb, were higher in PM_{2.5}. The content ratios in PM_{2.5} to PM₁₀ for 39 IEs except for Cu were higher than 1.0, and ranged from 1.10 for Sb to 1.72 for Si. Higher levels of Cr for all the PM_{2.5} and PM₁₀ should be paid more attentions.

The analysis result for particle size distribution of FAs from 15 CFPPs indicated 50% particles for 15 CFPPs ranged from 12.2 μm for FA15 to 55.1 μm for FA6, and 90% particles were of 57.5–142 μm. All the 15 size distribution curves had two peaks with the first one occurred at 0.762–1.18 μm and second one occurred at 21.1–84.9 μm.

Geo-accumulation index values for 23 IEs in PM₁₀ and 26 IEs in PM_{2.5} were higher than threshold value of 0, indicated their pollution levels ranged from moderately to extremely contamination for them. Igeo values for 8 heavy metals including Tl, Zn, Sb, Pb, Cu, Cd, Cr and Al in PM_{2.5} and PM₁₀ were higher than 0, especially for Cr, it was 3.57 and 3.61 for PM₁₀ and PM_{2.5}.

The element profiles of PM_{2.5} between any two CFPPs may be substituted for each other based on lower coefficient of divergence as 0.254 ± 0.038. The same situation was occurred for PM₁₀ (0.244 ± 0.056). The relative enrichment factors (REF) for 35 and 33 elements in PM₁₀ and PM_{2.5} were less than 0.70 or in the range of 0.70–1.30, indicated evaporation, no condensation or weak condensation processes occurred for them during the coal combustion. 4 IEs including Cr, Cu, Zn and Sn in PM₁₀, and 6 IEs including Cr, Cu, Zn, Sn, W and Pb in PM_{2.5} had the REF values higher than 1.30

indicated significant condensation occurred for them. Cr had the highest REF as 17.4 and 18.3 for PM₁₀ and PM_{2.5}, respectively.

ACKNOWLEDGEMENTS

This study was supported by National Natural Science Foundation of China (21407048), the Science and Technology Plan Project of Hebei Province of China (15273706D), the Fundamental Research Funds for the Central Universities (2014MS155), the Fundamental Research Funds for the Central Universities (2015ZD24) and Tianjin Research Program of Application Foundation and Advanced Technology (14JCYBJC23000).

SUPPLEMENTARY MATERIAL

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

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Received for review, February 27, 2017

Revised, March 13, 2017

Accepted, March 13, 2017