Fine particle and trace element emissions from an anthracite coal-fired power plant equipped with a bag-house in China

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Abstract

Fine particle and trace element emissions from energy production are associated with significant adverse human health effects. In this investigation, the fine particles and trace elements emitted from the combustion of pulverized anthracite coal at a 220 MW power plant were determined experimentally in the size range from 30 nm to 10 μm with 12 channels. The particulate size distributions and morphological characteristics before and after the bag-house were evaluated. The uncontrolled and controlled emission factors of particles are compared with the calculated values from the US Environment Protection Agency, AP-42. Size-classified relative enrichment factors of As, Hg, Se, Cd, Cr, Cu, Al, V, Zn, Mn, Fe were obtained. Relative distributions of trace elements between bottom ash, fly ash and flue gas are determined by mass balance method. The bag-house collection efficiencies of particles and trace elements in the particulate phase are obtained. Finally, the controlled and uncontrolled emission factors of elements of different particulate size fractions are obtained, which will provide useful information for $PM_{2.5}$ and PM_{10} emission inventory development, toxic and hazardous pollutant emission estimates and emission standards established for metal-based pollutants from a pulverized coal-fired boiler.
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1. Introduction

Coal is the major energy source in China and, even in 2020, it will account for 54% of total energy consumption [1]. In our country, about 45% of total coal consumed today is used in direct combustion by power plants; this is predicted to increase to 62% in 2010. The electric power industry accounts for most of the emissions of smoke and dust from among all industries [2]. At present in China, the emission standard of air pollutants for thermal power plants is based only on the concentration of total particulate matter (PM); however, the ambient standard is based on the concentration of particles with an aerodynamic diameter smaller than or equal to a nominal 100 μm (total suspended particulate, TSP) and 10 μm (PM_{10}). According to recent studies, PM_{10} from power plants is usually associated with significant toxic matter [3]. Fine and ultrafine particles are suspected to have a considerably stronger impact on human health than coarse particles. Therefore, fine and ultrafine particles emitted from coal combustion are the subject of significant investigation in China.

Although the concentration of trace metals in coal is typically below 100 ppm [4], the large quantities of coal consumed worldwide ensure that the aggregate, coal combustion contributes significantly to the total air burden of trace metals [5]. The fate of potentially toxic trace elements in combustion systems has received increasing interest from both regulatory authorities and scientists [6]. Based on 1983 emissions data from Western Europe, the United States, Canada, and the former Soviet Union, Nriagu and Pacyna estimated world-wide trace metals emissions from a variety of sources. They concluded that anthropogenic sources constituted 40–85% of the total annual

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atmospheric emissions for each of these elements [7], and electric utility coal combustion contributed 2–6% of the annual anthropogenic atmospheric arsenic emissions, 2–3% of the cadmium emissions, 14–17% of chromium emissions, 9–17% of the mercury emissions, 10–14% of the antimony emissions, and 6–13% of the selenium emissions [8]. In the United States, the Environmental Protection Agency (EPA) has stipulated that potentially toxic emissions of 11 metals from combustors and incinerators must be controlled, under Title III of the 1990 Clean Air Act Amendments [9]. In Europe, the EU has prescribed regulation limits for As, Cd, Co, Cr, Cu, Mn, Ni, Pb, Sb, Ti and V from waste incineration and co-incineration [10]. Emission standards are being developed for metal-based pollutants from pulverized coal fired boilers.

Presently, most coal-fired power plants in China have adopted electrostatic precipitators (ESP) to control the particulate emissions. By the end of 2000, about 1080 heat-engine units were equipped with ESP, representing more than 90% of total installed capacity. However, the collection efficiency of ESP sometimes is not sufficient to comply with the more strictly emission standards [11]. Bag-houses (BG) are used extensively in Australia, United States and South Africa et al. In Australia, all coal-fired boilers of power plants have gradually converted the ESP units into bag-house; in New South Wales, only four units are still equipped with ESP [12]. Some power plants in China began to adopt bag-houses instead of ESP, but that is still not widespread. Furthermore, particle and trace element emissions measurements from combustion sources in situ are difficult and complex, especially for fine particles. Therefore, it is infrequent to find field study reports for the size fractionating emission characteristics of fine particles and trace elements and the fractional collection efficiency of bag-houses in anthracite coal combustion in China.

It is clear that a fundamental understanding of the emission characteristics of the fine particles and trace element size distribution is an important step toward mitigating the environmental impact of PM emissions. The distribution of hazardous elements in different sized particles is needed to assess their potential mobility in the environment. Furthermore, knowledge of emission factors is a key element in the exposure assessment process. In this investigation, we sampled from anthracite coal-fired power plant at the inlet and outlet of the bag-house in situ, and describe size distributions, morphological characteristics, trace element emissions of the fine particles and the capabilities of bag-house. The experimental minimum particulate size reached was 30 nm.

2. Experimental methods

2.1. Sampling methods

Anthracite coal from Shanxi province was burned in a pulverized coal fired boiler equipped with a bag-house. The full electricity generating capacity is 220 MW and the testing load is 95%. Proximate analysis and ultimate analysis of coal are shown in Table 1.

Sampling positions are located at both the inlet and the outlet of bag-house, which respectively, represents the particle emissions from the boiler in non-controlled conditions and that emitted to the atmosphere after control. The flue gas temperature is 125–135 °C. During test run periods, the production equipments in the power plant were operating normally. The boiler testing load, and the fuel and the combustion operation mode did not vary; the emission concentrations of gaseous pollutants were also relatively stable.

The total PM is collected by a filter drum and measured by a TH-800 III microcomputer dust parallel sampling meter from Wuhan Tianhong Intelligence Meter Company. The size distribution of PM_{10} and size-classified PM_{10} samples were collected by Electrical Low Pressure Impactor (ELPI). The ELPI is designed for real-time monitoring of aerosol particle mass and number size distributions [13–15]. The ELPI measures aerodynamic size distribution in the size range of 30 nm to 10 μm using 12 channels. In order to decrease the particle concentrations and gas temperatures from the stack, a two-stage dilution system is applied to try to preserve the gas and particle conditions [16]. The sampling system for fine particles is shown in Fig. 1.

The sampling system consisted of the ELPI, an isokinetic sampler probe, pre-cut cyclone (cut-off diameter is 10 μm), two-stage dilution system and sample line to the instruments. All sampling lines were as short as possible to avoid losses of the large particles. Clean and dry pressurized air is used as dilution gas. The dilution ratio is about 1 to 90 in this work.

Different sampling substrates were selected for different research purposes. Cleaned and dried aluminum foils were used for sampling the PM_{10} size distribution. In order to minimize particle bounce, aluminum foils were coated with a thin layer of Apiezon-L grease [17]. The morphological and element samples were collected on a polycarbonate and twelve Teflon filters (Pall Corporation), respectively. Total PM and PM_{10} sampling follows the method prescribed in the regulation titled: "The determination of particulates and sampling methods of gaseous pollutants emitted from exhaust gas of stationary source" (GB/T16157-1996).

Pulverized coal, furnace bottom ash, and fly ash samples collected from the bag-house were gathered regularly during the testing period.

2.2. Analytical methods

After the morphological sample was collected, a small section of the polycarbonate filter was affixed to aluminum stubs with conductive adhesive. Then the sample was coated with 20 nm Au/Pd. Individual particle analyses was performed with a field emission source electronic microscope (FESEM) and energy dispersive X-ray microanalysis (EDX).

Table 1
Proximate analysis and ultimate analysis of coal

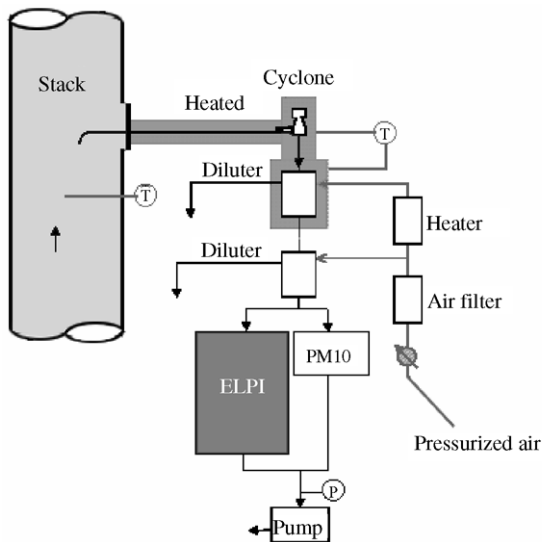| Carbon (%)	Nitrogen (%)	Oxygen (%)	Hydrogen (%)	Moisture (%)	Ash (%)	Volatile matter (%)	Sulfur (%)	LHV[a] (kJ/kg)
65 ± 5	0.79 ± 0.04	0.84 ± 0.08	2.10 ± 0.24	8.50 ± 0.85	31.2 ± 2.8	8.65 ± 0.67	0.54 ± 0.02	23600 ± 130

[a] Lower heating value.

Fig. 1. Sampling system for fine particles.

The element sample collected PM_{10} in twelve stages. The mass proportion of PM collected at each stage was determined through weighing Teflon filter papers by an analytical microbalance (0.001 mg precision). Afterward, the element samples were pretreated with the filters and analyzed separately. The amount of Cr, Cu, Al, V, Zn, Mn and Fe were measured using the inductively coupled plasma – atomic emission spectrometry (ICP-AES). As, Hg, Se and Cd were detected using atom fluorescent spectrometry (AFS).

3. Results and discussion

3.1. Concentration and size distribution of particulate matter

Fig. 2 shows the number and mass size distributions of PM_{10} both before and after the bag house.

The distribution of the number of particles from coal combustion display a bimodal distribution that contained the submicron mode and the coarse mode, which respectively represent two different particle formation mechanisms: 1. mineral transformation and char fragmentation and 2. vaporization, condensation and nucleation of inorganic constituents in the coal [18,19]. From Fig. 2, before the bag-house, the bimodal particle number distribution can be seen with a peak around 0.1 μm and 2 μm, respectively. In mass size distributions, the fine mode has a peak around 0.2 μm; however, the coarse mode with a peak larger than 10 μm is not evident because the size measurement

range of the instrument is exceeded. After the bag-house, the bimodal number size distribution is same as the incoming gas with the number concentration decreasing substantially. At the same time, in mass size distribution, the submicron mode shows low mass and the mean diameter of coarse mode is decreased to about 5 μm. Though PM_{10} emissions from coal-fired power plant have low mass concentration, the number of particles is high.

Because we are more interested in presenting emissions factors based on PM_{10} and $PM_{2.5}$ rather than on the entire particle emission range, the particulate mass size fractions of the total PM and emission factors with different particle sizes are calculated (Table 2).

From Table 2, although the fraction of PM_{10} before the bag-house contains less than 20% of the total mass, it represents approximately 70% of the total mass after it. Compared with the measured emission factors and the calculated values from US Environment Protection Agency (EPA) AP-42, the uncontrolled measured emission factors of PM and PM_{10} are higher than the calculated values, but the factors of $PM_{2.5}$ are lower. This indicates that the mass fraction of fine particles in this power plant is less than the mass fraction in the AP-42. However, the controlled measured emission factors are lower than the calculated values because the total collection efficiency of the bag-house in this work is higher than that of the AP-42 (99.8%).

3.2. Morphological characters

According to the morphological samples of the PM_{10} at inlet and outlet of the bag-house, there are many type particles at the inlet of bag-house, and most particles are coarse. Conversely, the particles are fine at the outlet. Most of the particles in both the submicron and super micron regions are spheres. Some spheres show signs of stretching, fracturing, or other shape-related irregularities. These particles have high surface-to-volume ratios compared to the typically smooth spheres found in the bulk fly ash. In addition, the vast majority of the particles are essentially nonporous in nature. The results are similar to those found by other authors [20,21].

Through image analysis system, the particle number size distributions based on equivalent spherical diameter of the particles were obtained, as shown in Fig. 3.

The figure shows that the spherical particles make up the main proportion, more than 80% of the particles. The peak value size of irregular particles is higher than spherical particles, which indicate that the irregular particles appeared in coarse particles more frequently than in fine particles.
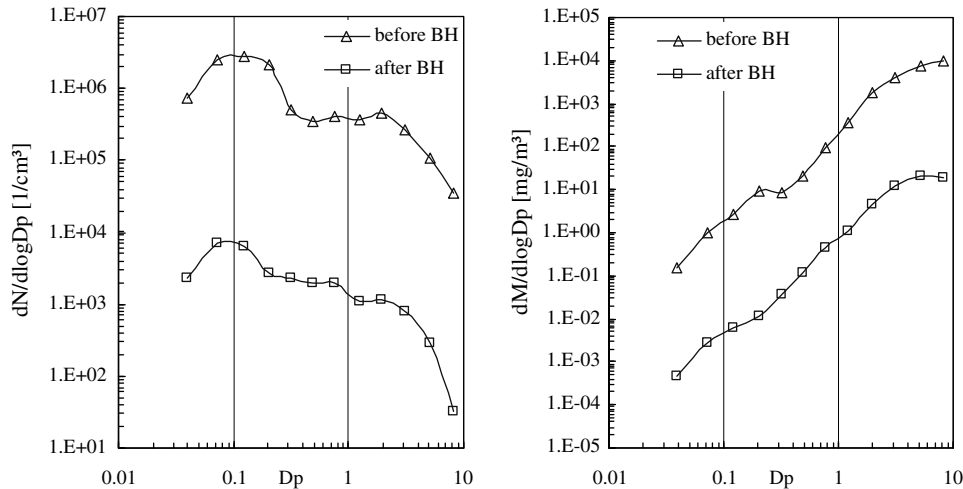
Fig. 2. Number and mass size distribution of PM$_{10}$ before and after the bag-house.

Table 2
Particulate mass size fractions of total PM and emission factors

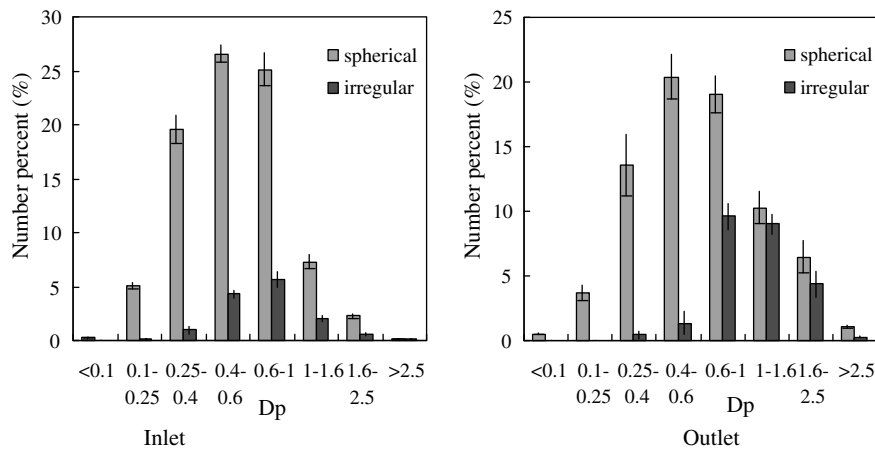| Dp	Before bag-house			After bag-house		
	PM$_{2.5}$	PM$_{10}$	PM	PM$_{2.5}$	PM$_{10}$	PM
Mass fractions (%)	1.7	16.7	100	8.6	71.3	100
Emission factors measured in this work (kg/t coal)	5	50	300	0.015	0.12	0.17
Values calculated by EPA AP-42 (kg/t coal)	8.5	32.7	142	0.09	0.19	0.28

Fig. 3. The particle number size distributions based on equivalent spherical diameter.

According to the EDX analysis, typical particulate types from coal burning sources are aluminosilicate, siliceous, calcareous, ferruginous, and carbonaceous. Aluminosilicate particles existed in all samples, but there were some differences in the structures, such as solid smooth spheres, stretched spheres, cenospheres, and irregular dense and porous particles. The investigation indicated that solid smooth spheres and irregular dense particles are common not only in aluminosilicate particles with different sizes but also in siliceous, ferruginous and calcareous particles. The microstructures of these particles are similar. As for the carbonaceous particles, they can be divided into spherical char, cenospheres char, porous char, dense char and chain-like soot aggregates. In the fine size range (<2.5 μm), carbonaceous particles are mainly chain-like soot aggregates, and in the coarse size range (>2.5 μm) are mostly irregular porous and dense char.

3.3. Size-classified relative enrichment factor of trace elements

Trace elements are emitted from a coal-fired boiler as a consequence of vaporization during combustion. For many trace elements, condensation leads to concentration enrichment in the submicron particles because of the significant surface area-to-volume ratio of this particulate [22].

Surface reaction may lead to redistribution of trace elements away from the difficult-to-capture submicron particles [23,24]. In this research, the trends of the change of trace elements in the combustion particulate matter are described by relative enrichment factor (REF) [25], that is: $REF = \frac{C_{oj}}{C_{fj}} A_f$.

In above equation, C_{fj} and C_{oj} mean the mass fraction of trace element j in raw coal and ash sample, respectively, and A_f means ash content in raw coal.

The relative enrichment factors for trace elements of PM_{10} before and after the bag-house are respectively shown in Fig. 4.

From Fig. 4, elements may be classified into three groups according to the size-classified REF in PM_{10} before bag-house. Group 1 includes Fe, Al and Mn. The REFs of the three elements have a slight change in different size ranges, and approximate 1. It indicates that these three elements are approximately equally distributed and show no signifi-

cant enrichment or depletion in PM_{10}. Group 2 consists of Hg, Se and Cr. It is found that the REFs of these trace elements increase as particle size decreases. However, the REFs of these elements are all greater than 1 in the submicron particles and less than 1 in the coarse particles. It shows that these elements in the PM_{10} sampled from this power plant are enriched in the submicron range, but depleted among the coarse particle. Group 3 contains As, Cd, Cu, V, and Zn. The REFs of these elements all decrease with increasing particle size and are greater than 1 in the whole size range. However, the degree of enrichment of elements is enhancing with the increasing volatility of elements.

After the bag-house, the trends of the change of all the elements are basically the same as at the inlet of bag-house. The difference is that, to the elements that are enriched in submicron particles, their REFs are increase after the bag-house. This suggests that these elements will subsequently condense on the surfaces of fine particles during
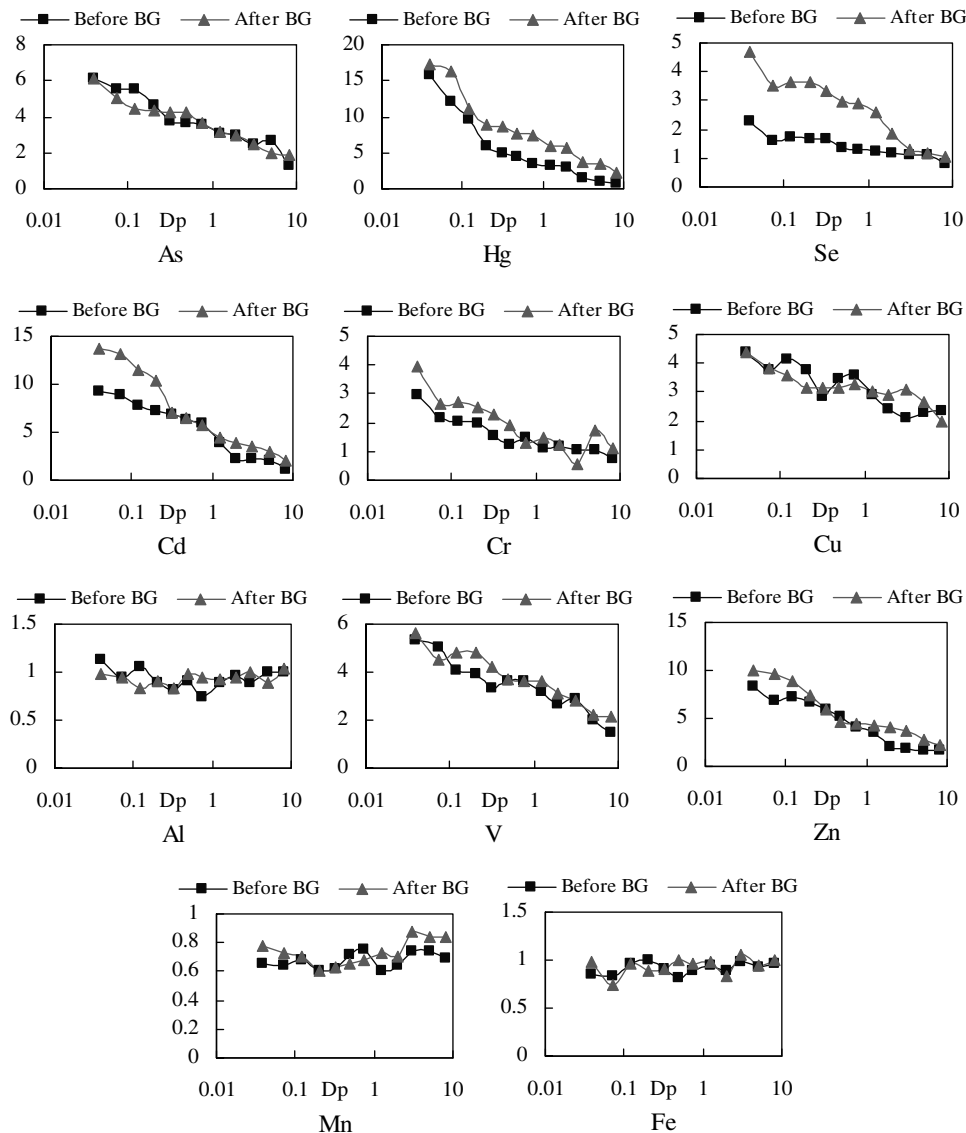

Fig. 4. Relative enrichment factors of trace elements of PM_{10} before and after the bag-house.

the flue gas travels through the bag-house. As a result, these submicron particles present more risks for environmental and health impacts.

3.4. Partition of trace metals in bottom ash, fly ash and flue gas

The mechanisms of trace element transformation during combustion are illustrated as following: the vaporized metals at high temperature near the combustion flame will subsequently nucleate or condense at a lower temperature downstream. The conversion of vaporized components into various solid and/or liquid forms is the key factor influencing the final trace elements' transformation/partitioning behavior. It is determined, basically, by three complex and interrelated processes: adsorption, condensation and chemical transformation. The trace element partitioning is often referred to the dispersion of elements among different emission streams: bottom ash, fly ash and flue gas [4].

Because the metal content of the flue gas from the power plant boiler was not measured in this study, we calculate it by a mass balance method from the following equation: $M_{ic} = M_{if} + M_{ib} + M_{ia}$, where M_{ic}, M_{if}, M_{ib}, M_{ia} represent the element i content in coal, fly ash, bottom ash and flue gas, respectively.

In order to reflect the quantitative trends of trace element partitioning in emission streams, data from other studies were adopted in this work. We assume that the element content of the combusted coal is the only input and the ash produced by the boilers is comprised of 20% bottom ash and 80% fly ash. In this work, the total mass efficiency of the bag-house is 99.94%, so the fine particle ratio in the flue gas is 80*(1–0.9994) of ash produced. The calculated results are showed in Fig. 5. The results of bottom ash and fly ash given for each element mass fraction are the mean value of the analysis of three samples.

Fig. 5 illustrates that the mass percent of volatile trace metals such as Hg and Se in flue gas are greater than other elements. These trace elements are completely vaporized at combustion temperature, and are sufficiently volatile to remain partly in the vapor phase as a very high vapor pressure at typical stack outlet temperature. Therefore, the air
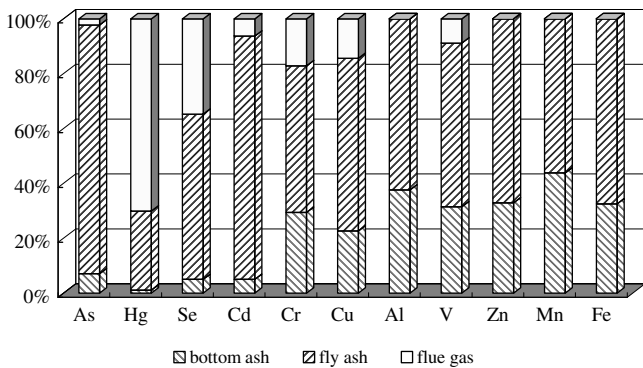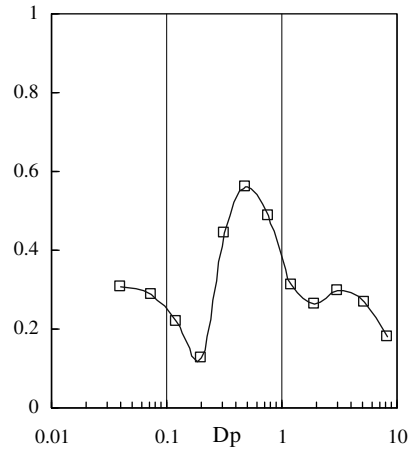

Fig. 6. Penetration of PM_{10} through the bag-house.

pollution control devices have limited capturing efficiency for these elements because most of them were discharged as gaseous phase with the flue gas. As, Cr, and Cd are partially vaporized at flame temperatures and subsequently condense on the surfaces of fly ash particles generated during combustion. Other elements basically condensed on the fly ash and bottom ash. Hence, a major proportion of these elements could be captured effectively by air pollution control devices.

3.5. Collection efficiency of bag-house

Bag-house collection of PM is highly efficient, reaching 99.94%. However, the collection efficiency of PM_{10} and $PM_{2.5}$ is lower, 99.76% and 99.72%, respectively. The measurements of the penetration of particles through bag-house are provided in Fig. 6. Although bag-house efficiency is higher than that of the ESP, the removal efficiency of particles is also least efficient in an intermediate size range from 0.1 μm to 1 μm. In this size range, bag-house penetration of PM_{10} is 0.46%.

The collection efficiencies of trace elements in different particulate size fractions are shown in Fig. 7.

Fig. 5. Relative distribution of trace elements between bottom ash, fly ash and flue gas.
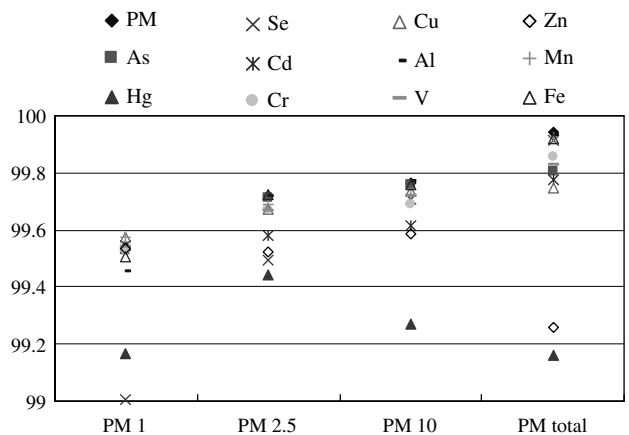

Fig. 7. The collection efficiencies of trace elements in different particulate size fractions.

Table 3
The emission factors of elements of different particulate size fractions (g/t coal)

	Uncontrolled emission factors				Controlled emission factors			
	PM_{total}	PM_1	$PM_{2.5}$	PM_{10}	PM_{total}	PM_1	$PM_{2.5}$	PM_{10}
PM	3,00,300	300	5005	50045	173	1.4	14.8	124
Al	8040	8.3	164	1690	5.7	0.05	0.5	4.1
Fe	3060	2.9	54.6	600	2.5	0.02	0.2	1.8
Zn	172	0.7	6.2	44.3	0.6	0.012	0.09	0.4
As	5.5	0.02	0.24	1.7	0.011	0.0002	0.0016	0.009
Hg	0.1	0.0013	0.015	0.06	0.0009	0.0000	0.0003	0.0009
Se	2.9	0.005	0.08	0.65	0.0025	0.0000	0.0004	0.002
Cd	0.2	0.0012	0.009	0.06	0.0005	0.0000	0.0001	0.0004
Cr	11.6	0.03	0.34	2.7	0.017	0.0004	0.004	0.013
Cu	20.4	0.08	0.95	8.5	0.044	0.0007	0.006	0.036
V	12.9	0.06	0.8	6	0.023	0.0005	0.0034	0.018
Mn	65	0.1	1.4	11.7	0.05	0.0005	0.005	0.034

The collection efficiencies trend of trace elements in different particulate size fractions is consistent with that of PM, that is, the collection efficiencies of PM_{total} and PM_1 are maximum and minimum, respectively. However, in same particulate size fractions, it is different from the overall PM collection efficiency. As mentioned before, the element is equally distributed, enriched or depleted in different size range according to its transformation during combustion and emission. As a result, the capture efficiencies of relatively non-volatile elements such as Al and Fe approach the overall particulate collection efficiency, whereas more volatile elements such as Hg, Zn and Se are captured less efficiently than the total PM. Note that the collection efficiencies mentioned above aimed at partitioning the elements in the particulate phase. The elements in gaseous phase are not involved in this investigation. Therefore, a majority of elements in particulate phase could be captured effectively by the bag-house, but elements in gaseous phase will escape.

3.6. Emission factors of trace elements in particles

It is important to quantify trace elements emissions from combustion sources for regulatory and control purposes in relation to the air quality, especially for elements in fine particles. Measurement is the preferred means to obtain data. The controlled and uncontrolled emission factors of elements of different particulate size fractions are obtained in this study (Table 3). The results will provide useful information for emission inventory development, toxic and hazardous pollutants emission estimation and emission standards establishment for metal-based pollutants from a pulverized coal fired boiler.

4. Conclusions

The bimodal number size distribution peaks between 0.1 μm and 2 μm before and after the bag-house. The uncontrolled measured emission factors of total PM and PM_{10} are higher than the calculated values of the EPA AP-42, but the emission factor of $PM_{2.5}$ is lower. However, the controlled emission factors are lower than the calculated values. Most of particles in both the submicron and supermicron regions are spheres and essentially non-porous in nature. The irregular particles appeared in coarse PM more frequently than in the fine fraction. Typical types of particulates from coal burning sources are aluminosilicate, siliceous, calcareous, ferruginous, and carbonaceous. The microstructures of these particles are similar. Carbonaceous particles are mainly chain-like soot aggregates in the fine size range and mostly irregular porous and dense char in the coarse size range. According to the size-classed relative enrichment factors of As, Hg, Se, Cd, Cr, Cu, Al, V, Zn, Mn and Fe in PM_{10} before the bag-house, these elements may be classified into three groups. After the bag-house, the trends of change of all elements are basically the same as the inlet of the bag-house. However, the elements that are enriched in submicron particles will subsequently condense on the fine particles during the flue gas travel through the bag-house. The bag house collection efficiency of PM is 99.94% and 99.57% to PM_{10}. The minimum collection efficiency of bag house (99.4%) is appeared in the particle size range of 0.1–1 μm. The capture efficiencies of relatively non-volatile elements approach the overall particulate collection efficiency, whereas more volatile elements are captured less efficiently than the total PM.

In coal-fired power plants of China, about 90% of air pollution control devices are ESP, and fewer plants are equipped with low efficiency particulate controls such as cyclones and wet scrubbers. In order to control fine particles and trace elements emissions and develop the establishment of the metal-based pollutant emission standard, research on characteristics of formation and emission for fine particles and trace elements from coal combustion should be pay more attention in China.

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