

Occurrence and volatility of several trace elements in pulverized coal boiler

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Abstract: The contents of eight trace elements (Mn, Cr, Pb, As, Se, Zn, Cd, Hg) in raw coal, bottom ash and fly ash were measured in a 220 t/h pulverized coal boiler. Factors affecting distribution of trace elements were investigated, including fly ash diameter, furnace temperature, oxygen content and trace elements' characters. One coefficient of *Meij* was also improved to more directly show element enrichment in combustion products. These elements may be classified into three groups according to their distribution: Group 1: Hg, which is very volatile. Group 2: Pb, Zn, Cd, which are partially volatile. Group 3: Mn, which is hardly volatile. Se may be located between groups 1 and 2. Cr has properties of both group 1 and 3. In addition, the smaller diameter of fly ash, the more relative enrichment of trace elements (except Mn). The fly ash showed different adsorption mechanisms of trace elements and the volatilization of trace elements rises with furnace temperature. Relative enrichments of trace elements (except Mn and Cr) in fly ash are larger than that in bottom ash. Low oxygen content can not always improve the volatilization of trace elements. Pb is easier to form chloride than Cd during coal combustion. Trace elements should be classified in accordance with factors.

Keywords: occurrence; volatility; relative enrichment; trace element; group of elements

Introduction

Coal, composed of aroma and mineral, is a heterogeneous fuel containing varying amounts of inorganic substances and combustible metamorphosed prehistoric vegetation. Up to now, about 84 elements have been found in coal, which include many trace elements (TEs). Though the contents of trace elements in coal (currently less than 100 ppmw) are very low, environment pollution caused by trace elements has been inarguable fact. The toxicity of trace elements is due to their "d electrons" that cause strong chemical reactions in the human body. Catalysis and strong chemical reactions are the main causes of their serious toxicity. In fact, one previous research suggested that the contents of trace elements in soil around power plants exceeded the allowable data of US Environment Protection Agency (EPA) (Edward, 2000). Environmental influence of fluoride and selenium on the health of people is assessed with coal in south and southwest China.

Pollution control of trace elements during coal combustion has recently been a new field, and many scholars have studied on it. But detailed researches on occurrence and volatility of trace elements in a full-scale pulverized coal boiler from three aspects of bottom ash, fly ash and flue gas are rare. In the present study, contents of eight trace elements (Mn, Cr, Pb, As, Se, Zn, Cd, Hg) in raw coal, bottom ash and fly ash were measured in a 220 t/h pulverized coal boiler. The objectives of this study are to analyze the factors, which affect occurrence and volatility of trace

elements including fly ash diameter, furnace temperature, oxygen content and trace elements' characters. The objects also include an evaluation of distribution of trace elements in the combustion products of coal. In this study these elements can be divided into several groups according to their occurrence and volatility.

1 Experimental

1.1 Description of the boiler and base characters of coal

There are altogether 8 boilers, whose MCR are all 220 t/h, at Thermoelectric Power Plant of Yangzi Petrochemical Co. Each boiler is connected to a three-field electrostatic precipitator (ESP) whose removal efficiency is about 99%. The sixth boiler was chosen to test by reason of its representative. The boiler has 8 primary air nozzles and 12 second air nozzles. About 29.7 tones of crude coal per hour are consumed. The coal used in examination is Xuzhou bitumite. Average coal diameter is 35.1 μm (50% aerodynamic cutoff diameter) by using HYDAO2000 laser light particle. Proximate analysis and ultimate analysis of coal are shown in Table 1.

1.2 Experimental condition

Correlative apparatus were verified at the beginning of experiment. All of the parameters have been modified in the next part of this paper. Combustion parameters were varied during the experiment, including the excessive air coefficient (α) and furnace temperature. In order to get three different excessive air coefficients (α), air velocity and rotation speed

of coal feeder were adjusted and furnace temperature approximately kept constant (typically within 20℃). Next, air velocity and rotation speed of coal feeder were adjusted to gain three different furnace temperatures, while excessive air coefficient kept constant (typically within 0.01). During the experiments, boiler output was 200—220 t/h, and the exhausted flue gas temperature was 139—148℃. Results of this study were used to gain a better understanding of the distribution of trace elements in a pulverized coal boiler.

Table 1 Proximate analysis and ultimate analysis of coal

	Proximate analysis			Ultimate analysis		
Total moisture	Mt	4.9%	C	Cad	51.82%	
Moisture	Marl	2.24%	H	Had	3.57%	
Volatile matter	Vad	25.74%	S	Sad	0.43%	
Fixed carbon	Cad	38.53%	N	Nad	0.9%	
Ash	Aad	33.49%	O	Oad	7.55%	
HHV	Q _h	20491 kJ/kg				
LHV	Q _{g ad}	19718 kJ/kg				

1.3 Sample collection

Due to practical reason, the samples in flue gas were not collected. A number of solid samples were collected from

various sites, which were pulverized coal classifier, bottom of boiler and each stage of electrostatic precipitators. Each test lasted 2—3 h. In order to keep samples unanimous, samples of raw coal, bottom ash and fly ash were collected at different time. Bottom ash was sampled after raw coal about one hour, and fly ash was sampled after bottom ash about one hour. After being sampled, the particle samples were decanted quantitatively into acid-washed, light high-density polyethylene(HDPE) containers.

1.4 Analysis

In order to determine the pathways of trace elements to the environment and their occurrence and volatility during each test, Mn, Cr, Pb, As, Se, Zn, Cd and Hg were analyzed. The graphite oven atomic absorption spectrometer (AAS) was selected for Se and Cd. Hg was analyzed using the cold vapor AAS-techniques and Mn, Cr, Pb, As, Zn were analyzed using X-ray fluorescence(XRF). In addition, the contents of ash in particle samples were analyzed. Results are shown in Table 2. Trace elements' average contents in the earth's crust are also listed.

Table 2 The contents of trace elements in raw coal, bottom ash and fly ash

No.	Furnace temperature	Excessive air coefficient	The contents of trace elements, μg/g or ppmw								Carbon content, %
			Mn	Cr	Pb	As	Se	Zn	Cd	Hg	
Coal	0—1		109	26	16	< 1	3.5	15	0.020	0.096	33.49
Bottom ash	1—1	1417	288	44	7	< 1	0.8	15	0.068	n. d.	13.60
	1—2	1437	274	36	15	< 1	1.0	21	0.039	n. d.	10.60
	1—3	1412	260	41	15	< 1	0.9	36	0.086	n. d.	9.50
	1—4	1363	253	41	21	< 1	1.8	22	0.052	n. d.	6.43
	1—5	1306	256	43	18	< 1	1.2	21	0.060	n. d.	4.10
Fly ash	2—1	1417	229	24	45	< 1	9.2	38	0.045	0.096	7.86
	2—2	1437	254	25	55	< 1	10	48	0.052	0.085	3.25
	2—3	1412	271	30	43	< 1	5.2	52	0.063	0.074	1.96
	2—4	1363	276	27	52	< 1	5.4	52	0.064	0.056	1.66
	2—5	1306	274	23	54	< 1	6.7	45	0.060	0.036	0.92
1st-field	3—1	1390	262	26	48	< 1	4.8	50	0.060	0.019	1.92
2nd-field	3—2		192	41	80	4.2	7.0	64	0.112	0.054	1.02
3rd-field	3—3		178	50	86	13	12	88	0.166	0.066	0.96
Contents in the earth's crust			950	100	12.5	1.8	0.05	70	0.2	0.043	

Notes: n. d. = Not determine

2 Results and discussion

2.1 Recovering trace elements from fly ash

Some trace elements' contents in fly ash are far greater than their average contents in the earth crust, especially Se (Table 2). The content of Se in crust is very little, which belongs to dispersive elements and scarcely individually presents in earth crust. So fly ash from electrostatic precipitator not only represents a significant potential source of toxic emissions to the environment, but also may be source, which can be abstracted some useful trace elements. A chemical extraction method in which soluble salts and trace elements are extracted in an acidic medium is an attractive medium. There have been progresses in this direction. It has attached higher importance to the progress in Japan. But most of the methods are still under development in laboratories (Tateda, 1997).

2.2 Relative enrichment factors(RE)

In order to investigate the change in the content of each trace element in the combustion products, relative enrichment facts(RE) are calculated. Scholars have put different REs. For example R. Cenni proposed a coefficient of Bottom Relative Enrichment (BRE) (Cenni, 1998): $BRE = \frac{C_{if} - C_{ib}}{C_{ib}}$. C_{if} represents the concentration of i trace element in fly ash and C_{ib} represents concentration of i trace element in bottom ash. This factor has advantage of putting relation only ash samples. Thus, it is free from the analytical uncertainties stemming from the interference of carbon in the sample matrix. The basic assumption in this factor is that the relative content of some elements(like Mn), which does not vaporize, must have the same concentration in fly ash as that in bottom ash.

One coefficient of RE defined by Meij is calculated as

the ratio between the concentration in fuel and combustion products(Meij, 1994). The mathematical expression of this factor is: $RE = \frac{C_{in}}{C_{ic}} \times \frac{1 - C_{adc}}{100}$. Here, C_{in} represents concentration of i trace element in different combustion products and C_{ic} represents concentration of i trace element in raw coal. C_{adc} represents content of carbon in raw coal. This factor only takes into account the influence of ash content in raw coal on RE , but does not include the influence of ash content in fly ash and bottom ash. Now, the $Meij$ factor is improved. That is $RE = \frac{C_{in}}{C_{ic}} \times \frac{1 - C_{adc}}{1 - C_{adn}}$. C_{adn}

represents content of carbon in different combustion products. RE describes the enrichment of trace metals in different products normalized to the ash content of the fuel. The improved coefficient of $Meij$ can more directly show element enrichment in combustion products. If RE equals 1, i trace element is not enriched nor depleted in the particular ash. If RE is larger or smaller than 1, i trace element is enriched or depleted respectively. RE s of trace elements are listed in Table 3.

Table 3 Relative enrichment facts of trace elements*

		Furnace temperature	Excessive air coefficient	Relative enrichment facts(RE)						
				Mn	Cr	Pb	Se	Zn	Cd	Hg
Bottom RE_b	1—1	1417	1.34	1.024	0.656	0.170	0.089	0.388	1.317	0
	1—2	1437	1.37	0.942	0.519	0.351	0.107	0.524	0.730	0
	1—3	1412	1.47	0.883	0.584	0.347	0.095	0.888	1.591	0
	1—4	1363	1.48	0.831	0.564	0.470	0.184	0.525	0.931	0
	1—5	1306	1.47	0.820	0.578	0.393	0.120	0.489	1.048	0
Fly ash RE_f	2—1	1417	1.34	0.764	0.336	1.022	0.955	0.921	0.818	0.363
	2—2	1437	1.37	0.807	0.333	1.190	0.989	1.108	0.900	0.306
	2—3	1412	1.47	0.849	0.394	0.918	0.508	1.184	1.076	0.263
	2—4	1363	1.48	0.862	0.354	1.107	0.525	1.181	1.090	0.199
	2—5	1306	1.47	0.850	0.299	1.141	0.647	1.014	1.014	0.127
1st-field RE_f	3—1	1390	1.49	0.821	0.341	1.024	0.468	1.138	1.024	0.068
2nd-field RE_f	3—2			0.596	0.534	1.692	0.677	1.444	1.895	0.190
3rd-field RE_f	3—3			0.552	0.650	1.818	1.159	1.984	2.807	0.232

Notes: Because the content of As in most of samples is less than the limit of X-ray fluorescence, discussion about As is leaved out in the next part of this paper

2.3 Influence of particle diameter on REs of trace elements

From Fig.1, it can be seen that most of trace elements have the trend to deposit on the smaller particles. The smaller diameter of fly ash, the more relative enrichment of trace elements(except Mn). That means there is a strong relationship between total particulate emissions from ESP and the emissions of specific trace elements. It can be concluded that Mn in raw coal in our experiment exists in some steady and high melting point coarse phase. Compared with other trace elements, Mn has adverse trend, i. e. the smaller diameter of fly ash, the less relative enrichment.

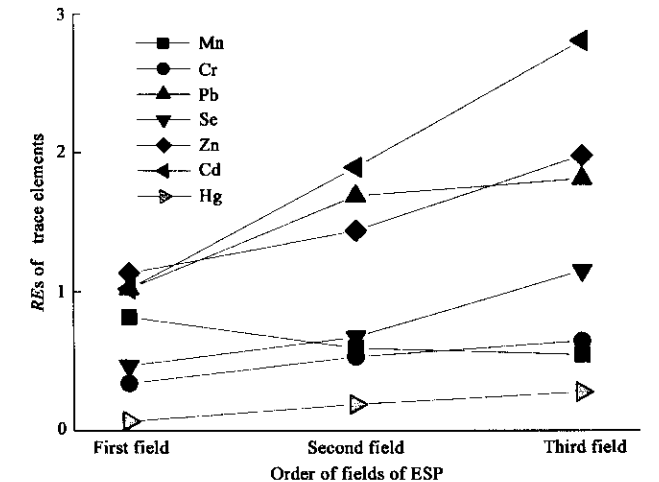


Fig.1 Relationship between REs of trace elements in fly ash and order of fields of ESP

From the first-field of electrostatic precipitator to the third-field, the particle diameter of ash becomes smaller and smaller and the surface area of ash becomes greater and greater. Metals vaporize from coal during the combustion process and re-condense in the post-combustion zone. Ash particles with diameters in the range of 0.01—0.1 μm are believed to be primarily formed via vaporization and condensation of major elements in coal such as silicon and iron(Flagan, 1978). The high surface area of sub-micron aerosol can cause a higher proportion of gaseous trace metal to deposit on the sub-micron ash as the flue gas cools, and results in an enrichment of certain element in the particles. Particles with diameters of approximately 0.1 μm are collected with the least efficiency in particulate collection devices such as electrostatic precipitators and bag-houses(Ylatalo,1998). Thus fly ash enriched in the fine particles represents a significant potential source of toxic emission to the air from coal-fired power plants.

Jyh-Cherng studied the adsorption mechanism of trace elements during incineration, and concluded that adsorption mechanisms of trace elements were related to the volatility of heavy metal species formed at high temperatures(Jyh-Cherng, 2001). For less volatile trace elements, chemical reaction is the major adsorption mechanism. But more volatile trace elements easily release at high temperatures and hardly react with ash to form thermostable compounds. Thus physical adsorption procession such as condensation, coagulation, and sedimentation are the important mechanisms for more volatile elements. So Cd, Zn, and Se adsorption on fly ash is

physical adsorption. Cr content in fly ash is less obvious trend along with particle diameter, which means Cr adsorption on fly ash is mainly chemical. Cr species is primary Cr_2O_3 at low temperature 500 K, which may react with CaO in fly ash: $\text{Cr}_2\text{O}_3 + \text{CaO} \rightarrow \text{CaO} \cdot \text{Cr}_2\text{O}_3$ (Leena, 1998). For Pb and As, chemical adsorption and physical adsorption are both important. When flue gas temperature is below 800 K, PbCl_2 is primary species of Pb (Flagan, 1978; Clemens, 1999), which may react with Al_2O_3 and SiO_2 in fly ash. Reaction is shown as follows: $\text{PbCl}_2 + \text{Al}_2\text{O}_3 + 2\text{SiO}_2 + \text{H}_2\text{O} \rightarrow \text{PbO} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 + 2\text{HCl}$ (Flagan, 1978). A. H. Clemens considered that As_2O_3 reacted with CaO under temperature 773 K (Clemens, 1999): $3\text{CaO} + \text{As}_2\text{O}_3 + \text{O}_2 \rightarrow \text{Ca}_3(\text{AsO}_4)_2$. Constance L and L. E. Bool also came to a conclusion that content of As in ash was coherent with that of Ca in fly ash (Constance, 2000; Bool, 1995). The correlation between As and Ca in ash suggests the formation of calcium arsenate.

At combustion temperature (about 1350°C), mercury completely releases from coal. This holds true also for most of the length of the cooling pips. Its concentration in the bottom ash is not detectable (Table 2). The concentration of Hg in fly ash contributes to the high surface area of fly ash and the unburned components (char) in ash (Cenni, 1998). The more carbon content in fly ash, the more REs of Hg. Since Hg species at outlet gas temperature (about 140°C) are mostly Hg, Hg_2Cl_2 and HgO , and these species' partial vapor pressures are below atmospheric pressures. Most of Hg presents vapor phase. Gas-solid adsorption has less distinctness than solid-solid adsorption, and the essential mechanism need to be farther researched.

2.4 Influence of furnace temperature on REs of trace elements

The volatilization of Cr, Zn, Hg rises with furnace temperature when excessive air coefficient between super-heater and economizer keeps approximately constant (comparing 2—3, 2—4, 2—5 sample). In the examination, the influences of temperature upon trace elements cannot be found.

2.5 Influence of excessive air coefficient on REs of trace elements

Comparing 1—1, 1—2 sample with 1—3 sample and comparing 2—1, 2—2 sample with 2—3 sample, we can see that the reducing oxygen increases the volatilization of some trace elements like Se, Hg under the condition of approximately invariable furnace temperature. But other elements do not abide the rule. Essential mechanism and more complex analysis need to be further researched. Rong Yan also considered that reducing conditions did not always enhance the trace elements volatilization, which depended on characters of both trace elements and coal (Rong, 2001).

2.6 Influence of characters of trace elements on REs

From Table 3, it is concluded that the REs of most of trace elements in fly ash are larger than those in bottom ash (except Mn, Cr). Because some mineral like K, Na compound which volatiles from raw coal at high temperature recoagulates on fly ash during flue gas cooling. So Mn and Cr have slight enrichment in bottom compared with fly ash. But

the RE of Mn is different from that of Cr, i.e. RE of Mn approaches 1, while RE of Cr is far less than 1. It is to interpret that in this experiment some Cr is associated with the organic part of the coal. This part of Cr volatiles at the beginning of coal burning, which causes concentration of Cr in bottom ash to decrease. In the post-combustion zone, organic Cr may homogeneously nucleate and forms sub-micron particle that directly emit from chimney. Constance L and Stanislav V also considered that in their investigations coal, part of Cr was associated with the organic (Constance, 2000; Stanislav, 2001).

Table 3 also shows that though boiling point of Pb (2032 K) is less than that of Cd (1042 K), REs of Pb in fly ash is higher than that of Cd. It is to explain that Pb has a stronger affinity to Cl in flue gas than Cd, and boiling point of PbCl_2 (1223°C) is far less than that of CdO (1773°C). It means Pb is easier to form chloride than Cd during coal combustion. These results are in agreement with previously published results (Rong, 2001; Stanislav, 2001).

2.7 Sort of trace elements

Combustion products are divided into three parts: bottom ash, fly ash and flue gas. Each element abides mass conservation: $M_{ic} = M_{if} + M_{ib} + M_{ia}$. M_{ic} represents total mass of i element in coal. M_{if} represents total mass of i element in fly ash. M_{ib} represents total mass of i element in bottom ash. M_{ia} represents total mass of i element in flue gas. Because it is difficult to obtain the distribution of ash in big power plants, we assume that ash distribution among bottom ash, fly ash and flue gas is 10:89:1 according to empirical data. The percent of trace elements in gas have been estimated mathematically using the following assumption. For a give trace element: $R_{ia} = 1 - R_{if} - R_{ib}$, $R_{ia} (M_{ia}/M_{ic})$ represents the percent of i element in gas. $R_{if} (M_{if}/M_{ic})$ represents the percent of i element in fly ash and $R_{ib} (M_{ib}/M_{ic})$ represents the percent of i element in bottom ash.

Now, representative samples 0—1, 1—4, 2—4 were chosen to calculate the distributions of trace elements in combustion products. Results are shown in Fig. 2.

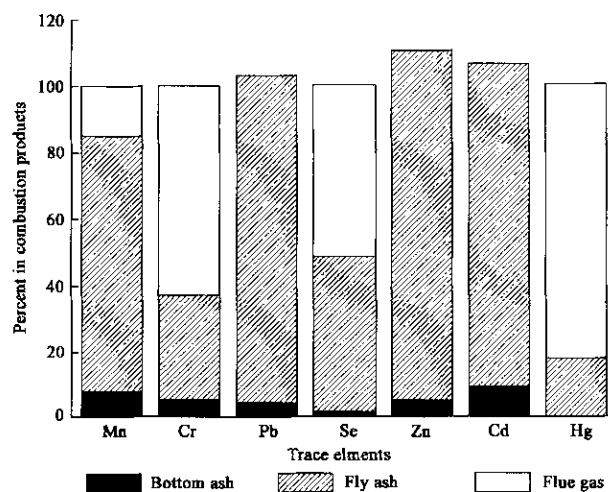


Fig.2 Trace elements percent in combustion products

Total percent of some elements excess 100. This phenomenon can be explained by two reasons. Firstly, there

are inevitable scatter errors in measure background. Secondly, fly ash from electrostatic precipitator is magnified so other ash (e.g. economizer bottom ash, sediment ash in equipment wall, sedimentation ash in chimney, and so on) in calculation is ignored. Most of trace elements appear to enter fly ash from electrostatic precipitator and flue gas. Because mass of fly ash is far more than that of bottom ash, trace elements percent in bottom ash is very little compared with that in fly ash.

The elements can be divided into three groups according to their occurrence and volatility: Group 1: Hg, which is very volatile and emit almost totally in the vapor phase and whose characters are $RE_f \gg RE_b$ and $R_a > 80\%$. Group 2: Pb, Zn, Cd, which are vaporized at intermediate temperature and emit mostly in fly ash and whose characters are $RE_f \gg RE_b$ and $R_f > 80\%$. Group 3: Mn, which is hardly vaporized and so are equally distributed between bottom ash and fly ash and whose character is $RE_f < RE_b$. In addition, Se is located between Group 1 and 2. Because some of Cr is associated with organic in coal, Cr puts up peculiar characters and simultaneity possesses characters of Group 1 and 3 during combustion. Both R. Cenni and Rong Yan classified Cr to Group 3 (Cenni, 1998; Rong, 2001). So it is concluded that group of Cr should depend on its form in coal. That is to say if most of Cr in coal is associate with mineral, Cr belongs to Group 3, while if most of Cr in coal is associate with organic, Cr should belong to special group.

Content of As in coal and particle ash is too low to classify it. A. H. Clements considered As was located between Group 2 and 3 (Clemens, 1999), while Rong Yan classified Cr to Group 2 (Rong, 2001). The behavior of As during combustion will be determined by whether As is present as a substitution element for sulfur in pyrite or is present in oxidized form as As minerals. Thereby trace elements should be classified in accordance with many factors such as characters of trace elements, combustion conditions, type of coal, and type of furnace.

3 Conclusions

The improved coefficient of *Meij* can more directly show element enrichment in combustion products. Most of trace elements have the trend to deposit on the smaller particles. The smaller diameter of fly ash, the more relative enrichment of trace elements (except Mn). Some trace elements' contents (especially Pb, Se) in fly ash are far greater than their average contents in the earth crust, which has double meanings: a significant potential source of toxic emissions to the environment and a source abstracted some useful metals.

Cd, Zn and Se adsorption on fly ash is physical and Cr adsorption on fly ash is mainly chemical, while chemical adsorption and physical adsorption are both important for Pb and As.

The volatilization of Cr, Zn, Hg rises with furnace temperature when excessive air index (α) between superheater and economizer keeps approximately constant. The reducing oxygen increases the volatilization of some trace elements like Se, Hg under the condition of approximately invariable furnace temperature. But low oxygen content can not always improve the volatilization of trace elements.

Because some mineral like K, Na compound which volatiles from raw coal at high temperature recondenses on fly ash during flue gas cooling, Mn and Cr have slight enrichment in bottom compared with fly ash. But the *RE* of Mn is different from that of Cr. Other trace elements have different characters.

Though boiling point of Pb is less than that Cd, *RE* of Pb in fly ash is higher than that of Cd and *RE* of Pb in bottom ash is less than that of Cd. Pb is easier to form chloride than Cd during coal combustion.

As content in coal and particle ash is too low to classify it. Other elements (except As) may be classified into three groups according to their occurrence and volatility: Group 1: Hg. Group 2: Pb, Zn, Cd. Group 3: Mn. Se may be located between Groups 1 and 2. Cr has properties of both Group 1 and Group 3. Trace elements should be classified in accordance with many factors such as characters of trace elements, combustion conditions, type of coal, and type of furnace.

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