

## Characterization and mutagenicities of emissions from industrial coal combustion based on particle size distribution

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**Abstract.** Total suspended particles (TSP), sulphur dioxide (SO<sub>2</sub>), polycyclic aromatic hydrocarbons (PAHs) and inorganic metal elements (IME) in the emission from coal boilers used in factory and power station, and the mutagenicities of extract materials of the stack samples were measured and discussed. When coal briquettes were burned instead of conventional raw coal for boilers, the TSP and SO<sub>2</sub> emission decreased approximately by 60–70% and 40–50% respectively, as well as the Ames mutagenicities of emissions from burning coal briquettes are lower than those from the latter.

**Keywords:** mutagenicity; coal briquettes; polycyclic aromatic hydrocarbons.

One important environmental problem of public concerns in China is the air pollution in urban area caused by coal combustion emission, which usually contains suspended particles-coal flyash, SO<sub>2</sub>, inorganic elements and some mutagenic organic compounds such as PAHs. The results introduced in this paper indicate that the coal briquettes (made of raw coal and 8% binding material, 3–5% SO<sub>2</sub> fixation agents under pressure and heating) as alternative of conventional raw coal for industrial boiler might be effective for controlling and abating air contamination.

### EXPERIMENTAL

#### *Sampling*

The amount of SO<sub>2</sub> in stack flue was measured with SO<sub>2</sub> chemiluminescence analyzer. For TSP, PAHs and inorganic metal elements, a special sampler was designed basing on the modified method 5 of USEPA (Cui, 1989) in order to collect not only pollutants on TSP surfaces, but also those in vapor phase smoke effectively (Fig. 1).

#### *Analysis*

The PAHs in the samples were collected by the above mentioned sampler and were analyzed by Shimadzu LC-3A high performance liquid chromatography (HPLC, Yao, 1986), while the metal elements were measured by PE 4000 USA atomic absorption spectrometry (AAS).

Protocol suggested by Ames with *Salmonella typhimurium* strains TA98 and TA100 was followed without changes (Ames, 1975). The couple of ultraviolet-fluorescence detector for HPLC determination has satisfied sensitivity for the majority of PAHs based on the facts that the PAHs which have less structural rings are stronger absorbers of UV while those with more rings, yield intense fluorescence (Fig. 2).

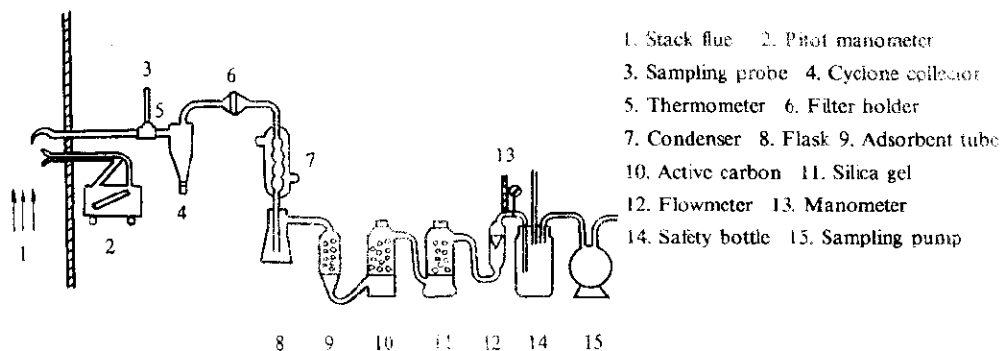


Fig. 1 Schematic diagram of the sampler

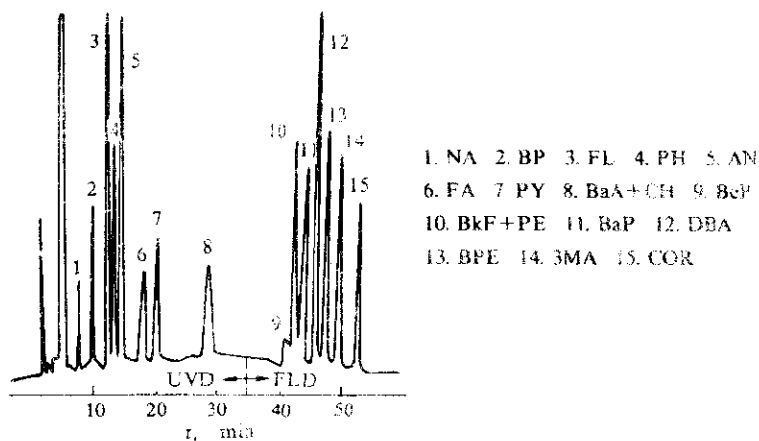


Fig. 2 HPLC chromatogram of 16 PAHs standards with dual-detectors (UV 254 and FLD 296/427nm) Zorbax-ODS  $C_{18}$  of  $250 \times 4.6$  mm I.D.  $CH_3OH/H_2O = 85/15$  for 35 min, then to 100%  $CH_3OH$  (2%/min)

## RESULTS AND DISCUSSION

*SO<sub>2</sub> and TSP*

Concentrations of SO<sub>2</sub> and TSP emissions from coal fired boilers are shown in Table 1. When coal briquettes are used to replace the raw coal, the apparent reductions of TSP and SO<sub>2</sub> (about 60–70% and 40–50%, respectively) have been observed (Table 1).

Table 1 SO<sub>2</sub> and TSP emissions from coal fired boilers

Boilers	KZL-2-8 (2T/h)	UG-35/39M (35T/h)
	for factory	for power station
Coal type	Beijing soft coal	Datong soft coal
Dust remover	Cyclone- remover	Water-remover
Binding materials	Asphaltic residue	Soda-lime
Coal type	Raw / briquettes	Raw/briquettes
SO <sub>2</sub> , g/Nm <sup>3</sup> *	0.994/0.500	0.686/0.315
TSP, g/Nm <sup>3</sup> *	1.404/0.584	1.081/0.301

\* g/Nm<sup>3</sup>: grams of pollutants per normal cubic meter stack gases

*PAHs and metal elements*Table 2 The concentrations of PAHs emitted from coal-fired boilers, µg/Nm<sup>3</sup>

Boilers Name of PAHs	KZL-2-8, 2T/h		UG-35/39M, 35T/h	
	Raw coal	Briquettes	Raw coal	Briquettes
Naphthalene	1.28	1.44	26.7	28.0
Biphenyl	2.05	2.23	27.6	15.1
Fluorene	0.04	ND	5.58	21.4
Phenanthrene	1.59	2.58	8.86	6.65
Anthracene	79.5	9.40	0.82	0.37
Fluoranthene	48.1	16.8	31.9	34.9
Pyrene	12.5	20.8	4.30	3.74
Benzo (a) anthracene*	3.10	1.90	2.25	3.12
Benzo (k) fluoranthene*	0.48	0.52	0.11	0.08
Benzo (a) pyrene	0.20	0.12	0.04	0.04
Dibenzo (ah) anthracene	0.52	0.28	0.18	0.38
Benzo (ghi) perylene	0.76	0.31	0.09	0.48
Coronene	0.61	0.06	0.06	0.06
Nitro-pyrene**	0.31	1.85	NA	NA

\* uneasy separated couple components of benzo (a) anthracene/chrysene and benzo (k) fluoranthene/ perylene are counted respectively as pure compounds

\*\* In micrograms of nitro-pyrene in each gram of TSP; NA means not analyzed.

Table 2 and Table 3 show that some PAHs and metal elements emitted from raw coal-fired boiler are higher in concentrations than those from using coal briquettes, while others are lower. The emissions of some metal elements such as aluminium, cadmium and iron and so on, are quite difference for different boilers. Whether it is related to the coal type needs to be further studied.

**Table 3** Contents of the metal elements measured in the emissions of coal-fired boilers,  $\mu\text{g}/\text{Nm}^3$

Metal elements	Boilers KZL-2-8, 2T/h		Boilers UG-35/39M, 35T/h	
	Raw coal	Briquettes	Raw coal	Briquettes
Aluminium	89.3	70.9	8140	6530
Arsenic	25.5	34.4	32	85
Barium	6.0	1.4	744	71
Cadmium	41.8	96.5	1080	1380
Calcium	4.2	7.8	5	5
Chromium	31.3	45.0	237	67
Copper	142	142	144	72
Iron	676	668	4680	4040
Manganese	8.9	6.1	178	31
Nickel	24.2	37.5	85	69
Lead	608	443	372	286
Selenium	2.4	5.3	100	74
Silver	2.1	1.4	ND	ND
Zinc	887	1109	678	1290

### Mutagenicities

The TSP collected from the sampler were extracted by ultrasonic wave with the solvent benzene and the mutagenicities of the extracts were measured by Ames test (Table 4).

**Table 4** Mutagenic activities of emission samples from UG-35/39 mol/L coal-fired boiler used in power station, Rev/ $\text{Nm}^3$

Coal Distribution	Raw coal				Coal briquettes			
	DF	FD	VS	Total	DF	FD	VS	Total
Strain TA98	2584	5015	4384	11983	783	4958	935	6676
Strain TA100	643	644	1746	3033	278	1044	884	2206

DF = dustfall

FD = floating dust

VS = vaporphase smoke

Table 4 indicates that the mutagenic activities of organic extracts emitted from coal

briquettes are 25–40% lower than those from raw coal. Furthermore, an noticeable tendency is that the mutagenic activities of floating dust and vaporphase smoke are high in comparison with the dustfall parts.

*The contribution of PAHs and inorganic elements emitted in each parts of the stack emission samples*

The stack emission were collected separately as mentioned above to give dustfall ( $5-20 \mu\text{m}$ ), floating dust ( $0.2-5 \mu\text{m}$ ) and the vaporphase smoke ( $<0.2 \mu\text{m}$ ), PAHs and metal elements in each part were also analyzed separately. The contribution of these pollutants is shown in Fig. 3. We can understand from these figures that the PAHs distributed in the vaporphase smoke are quite high in weight percentage, especially for those with less rings, but the majority of the PAHs with more structure rings, which generally are environmental toxic and detrimental to human health, more likely distribute in floating dust parts.

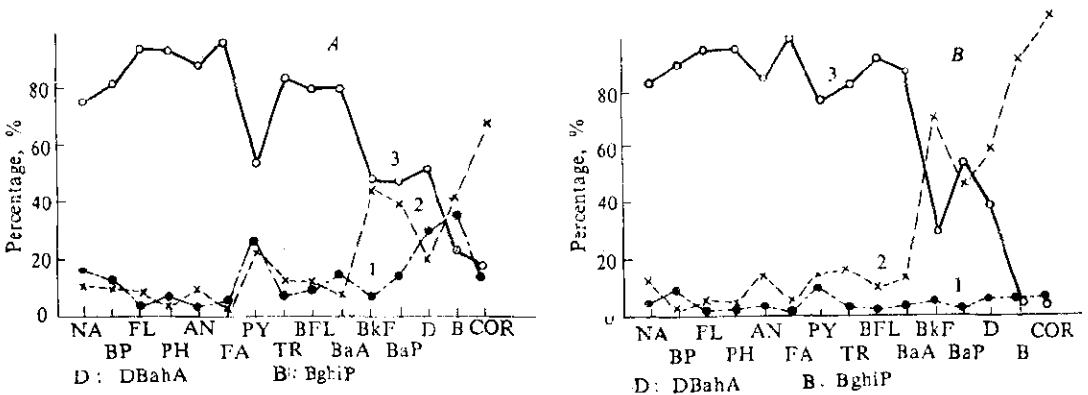


Fig. 3 Percentage distribution of PAHs on each part of the emission samples from UG-35M coal-fired boiler used in power station

A: raw coal B: briquettes

1. dustfall 2. floating dust 3. vaporphase smoke

## CONCLUSIONS

For investigating the distribution regulation of pollutants from the stacks of coal combustion, three parts (dustfall, floatingdust and vaporphase smoke) of the samples were collected separately. The results indicated that the majority of environmental significant PAHs of larger molecular structure distribute more likely on the floating dust, while very high percentages of PAHs with lower molecular weights remain in the vapor phases. When the coal briquettes are used for coal-fired boiler, the concentrations of TSP,  $\text{SO}_2$  and PAHs have remarkable reduction and the mutagenic activities of extracts of emission are also distinctly lower in comparison with those

from corresponding raw coal of same commercial sources. So the coal briquettes technology can be considered as one of the feasible interim measure for air pollution control and prevention.

**Acknowledgements** — The authors gratefully acknowledge the support of their research by Shan Xiaoquan, Shen Dixin, Zhu Naikai and Jin Zuliang of the Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences.

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(Received April 8, 1992)