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# A typical 300 MW ultralow emission coal-fired power plant: source, distribution, emission, and control of polycyclic aromatic hydrocarbons

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#### ABSTRACT

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Polycyclic aromatic hydrocarbons (PAHs) are one of the dominant contributors to toxic organic pollutants emitted from coal combustion. In this study, the PAHs and condensable particulate matter (CPM) were sampled from a 300 MW ultralow emission coal-fired power plant. The effect of air pollution control devices (APCDs) on PAHs migration regularity and the influence of coal species changes on the emission of PAHs were studied. Note that the APCDs have definite selectivity for the removal of PAHs with different phases and ring numbers. The low-low temperature electrostatic precipitator (LLT-ESP) presented the best removal effect on PAHs (PAHs in flue gas, 29.92%; PAHs in CPM, 94.76%). The concentrations of PAHs were reduced at the outlet of the furnace (PAHs in flue gas, decreased from 3.318 to 2.850  $\mu$ g/Nm<sup>3</sup>) and at the stack (PAHs in flue gas, decreased from 4.737 to 3.008  $\mu$ g/Nm<sup>3</sup>; PAHs in CPM, decreased from 0.554 to 0.429  $\mu$ g/Nm<sup>3</sup>) by replacing bituminous coal with blended coal (lower volatiles) as fuel. The adsorbent injection coupled with LLT-ESP had a preferable removal effect on the PAHs (decreased from 4.949 to 1.451  $\mu$ g/Nm<sup>3</sup>), which was nearly 40% higher than the efficiency without the spray adsorbent.

#### 1. Introduction

China's resource endowment is dominated by coal, and at present, China's proven reserves of coal exceed 1.6 trillion tons, accounting for more than 95% of fossil energy reserves. According to statistics [1], in 2020, coal accounted for 57% of China's primary energy consumption and provided 63% of the power generation, indicating that coal is still the basic energy in China and supports economic and social development. In a word, coal is the cornerstone of China's energy security. However, coal-fired power plants are considered the most important sources of air pollution, which emit large amounts of pollutants, such as particulate matter (PM), sulfur dioxide (SO<sub>2</sub>), and nitrogen oxides (NO<sub>X</sub>) [2-5]. In 2015, China officially launched a comprehensive ultralow emission work for coal-fired power plants and has now achieved the reduction of PM, SO<sub>2</sub>, and NO<sub>x</sub> emission concentrations to 10, 35, and  $50 \text{ mg/Nm}^3$  respectively [6,7]. Since the ultra-low emissions of conventional pollutants (PM, SO2, and NOX) in coal-fired power plants, the emission control of organic pollutants such as polycyclic aromatic hydrocarbons (PAHs), which are characterized by high toxicity and difficult to remove, have been valued. In fact, coal combustion has been identified as the primary origin of PAHs in the atmosphere [8-11].

PAHs are aromatic hydrocarbons containing two or more benzene rings, which are identified as major organic pollutants affecting human health due to their mutagenicity, carcinogenic, and teratogenic [12,13]. The U.S. Environmental Protection Agency (USEPA) specified 16 PAHs as priority test pollutants in 1976, which are shown in Table S1 [14,15]. In coal-fired power plants, in addition to the occurrence of PAHs in fuel coal, the incomplete combustion of volatiles in the feeding coal is the most significant source of PAHs [16-18]. In addition to the gas- and particulate-phase PAHs in flue gas, PAHs also occur in organic components of condensable particulate matter (CPM) from coal combustion [19-23]. CPM is material that is a vapor before discharge, but that condenses to form solid or liquid particulate immediately after it is emitted from the flue, including organic and inorganic substances [24]. Recent studies [25-27] have also shown the PAHs produced by coal combustion will be combined with solid wastes such as slag and fly ash, and also exist in liquid wastes produced by the desulfurization process. It is observed that the whereabouts of PAHs are very wide, so it is valuable guidance to clarify the source, distribution, and emission of PAHs in a typical coal-fired power plant for the formation of control technology.

At present, tremendous applied research has focused on the emission concentration of PAHs from coal combustion [28–30]. Lu et al. [31]

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found the emission concentrations of PAHs in flue gases from a coal-fired power plant and a coal-fired boiler were 28.4 and 43.4 µg/Nm<sup>3</sup>, respectively. The research of Li et al. [32] showed that the emission concentration of PAHs from a 1000 MW ultralow emission coal-fired power plant was only 0.87 µg/Nm<sup>3</sup>. Many previous reports proved that the concentration of PAHs emitted from coal-fired power plants reached microgram levels, and the pollution levels of sites around the coal-fired power plant were higher than those in other zones [33]. Nevertheless, the existing air pollution control devices (APCDs) in coalfired power plants have a limited removal effect on PAHs in the flue gas. The study of Chen et al. [34] showed that the removal efficiency of PAHs by APCDs adopted in a coal-fired power plant was only 18%. The effects of various APCDs on the migration and distribution of PAHs in the coal burning process are different; however, studies on the changes of gasand particulate-phase PAHs (with two to six ring numbers) throughout the flue gas purification process in coal-fired power plants are limited. Thus, research on the migration process of PAHs in flue gas during purification apparatus/systems installed in typical coal-fired power plants, and exploring the development of control technology bears practical significance.

In this work, we conducted a field sampling of PAHs in a typical 300 MW ultralow emission coal-fired power plant. The source, distribution, and emission of 16 kinds of PAHs from coal combustion were thoroughly investigated. And the control technology was explored emphatically. The research objectives of this paper are as follows: 1) study the migration and distribution of PAHs during the APCDs system; 2) study the effects of changes in feeding coal on the formation and emission of PAHs; 3) explore the control technology of PAHs in an actual coal-fired power plant.

#### 2. Experimental section

#### 2.1. Facility and sampling sites

A typical 300 MW ultralow emission coal-fired power plant was selected to make a comprehensive analysis of the source, distribution, emission, and control of PAHs from coal combustion. The overall flue gas purification process of the coal-fired unit and the information parameters of sampling sites are shown in Fig. 1 and Table S2. The coal-fired unit has been retrofitted with ultralow emissions and its APCDs consisted of a selective catalytic reduction (SCR) denitration device, a low-low temperature electrostatic precipitator (LLT-ESP), a wet flue gas

desulfurization (WFGD) system, and a wet electrostatic precipitator (WESP) for the removal of pollutants from coal combustion. During the sampling, the operating parameters of the coal-fired unit are shown in Table 1, the unit was maintained at full load (300 MW) and the APCDs were operated steadily. Noted, two types of coal (Table S3) were selected to explore the impact of fuel on the formation and emission of PAHs from coal combustion. PAHs in flue gas and CPM were collected at the other four sampling sites, except that only PAHs in the flue gas was sampled at the inlet of SCR. Moreover, solid (slag and gypsum) and liquid (desulfurization wastewater and slurry) samples were collected.

The field test for removing PAHs in coal-fired flue gas with adsorbent injection coupled with LLT-ESP was shown in Fig. S1 and Table S4. An adsorber injection device was installed before LLT-ESP and tested with two types of adsorbents (A<sub>1</sub> and A<sub>2</sub>). During the sampling, the coal-fired unit was kept operating at 60% load (180 MW) and the APCDs were operated steadily. The sampling sites were set at the inlet and outlet of the LLT-ESP. The adsorbent was sprayed under three conditions (A<sub>2</sub>, 60 kg/h; A<sub>2</sub>, 170 kg/h; A<sub>1</sub>, 170 kg/h), and the condition with the best removal effect on PAHs was selected. And then fly ash was collected from the three electric field ash hopper before spraying adsorbent, two hours after spraying, and four hours after spraying, respectively.

#### 2.2. Sampling equipment and methods

Fig. S2 shows the schematic of systems for PAHs and CPM sampling in the coal-fired flue gas. As shown in Fig. S2a, the XAD-2 resin in the adsorption chamber and membrane in the filter adsorb the PAHs in the gas phase and the particulate phase in the flue gas, simultaneously. Before the sampling, the surrogate standards (fluoranthene- $D_{10}$  and benzo[a]pyrene-D<sub>12</sub>) were added to the filter membrane surface drop by drop uniformly and quantitatively, then placed in the dark for 1 h, and started sampling pump. The sum of the concentration of PAHs in the two phases is the total concentration of PAHs in the flue gas. As shown in Fig. S2b, the flue gas is filtered by the Dekati PM<sub>10</sub> impactor so that CPM can be effectively captured by the condenser, short and long stem impactor, and CPM filter. Noted that the equipment before the condenser both in the two sampling systems should maintain the temperature at 120  $\sim$  130 °C. Besides, the mass concentration of PAHs in flue gas and CPM should be converted to the standard concentration under the conditions of 6% oxygen and drying standards. The specific function, parameter settings, and detailed operation process of sampling devices have been presented in previous studies [35,36].



Fig. 1. Schematic showing the APCDs and sampling sites in coal-fired power units. GGH(C), a cooling section of the gas-gas heat exchanger; GGH(H), a heating section of the gas-gas heat exchanger. Description: The unit is a circulating fluidized bed coal-fired boiler with an installed capacity of 1000 MW, a selective catalytic reduction (SCR) denitration device, a low-low temperature electrostatic precipitator (LLT-ESP), a wet flue gas desulfurization (WFGD) device, and a wet electrostatic precipitator (WESP) as APCDs. Ultra-low emission reformation: Yes. Purpose: Electricity generation. Location: South of China (Zhejiang Province).

#### Table 1

Operation parameters of the coal-fired unit.

Rated Capacities	Operating Loads	Total Fuel Consumption	Site Name	Coal Species	Concentration of NO <sub>X</sub> (mg/Nm <sup>3</sup> )	Concentration of SO <sub>2</sub> (mg/Nm <sup>3</sup> )	Concentration of PM (mg/Nm <sup>3</sup> )	Flue Gas Temperature (°C)
300 MW	292.25 ~ 314.88 MW	115.27 ~ 124.97 t/h	the Inlet of the SCR	Bituminous Coal	125.01 ~ 180.48			352.74 ~ 377.51
				Blended Coal	$114.04 \sim 157.13$			
			the Inlet of	Bituminous	29.21 ~ 30.56			$133.98 \sim 137.93$
			the LLT-ESP	Coal				
				Blended Coal	$27.64 \sim 31.14$			
			the Inlet of	Bituminous		848.37 ~ 940.12	$13.98 \sim 14.37$	$108.68 \sim 111.62$
			the WFGD	Coal				
				Blended Coal		891.42 ~ 992.52	$14.01 \sim 15.81$	
			the Inlet of	Bituminous				$48.40 \sim 49.55$
			the WESP	Coal				
				Blended Coal				
			the Inlet of	Bituminous	$24.71 \sim 25.45$	$12.71 \sim 18.43$	$1.03 \sim 1.11$	$76.01 \sim 77.21$
			the Stack	Coal				
				Blended Coal	$25.29 \sim 28.47$	$13.99 \sim 17.82$	$0.92 \sim 1.16$	

#### 2.3. Analytical procedure for samples

The categories of samples involved in this study include PAHs in flue gas (gas and particulate phase), PAHs in CPM, solid samples (coal, slag, gypsum, and fly ash), liquid samples (desulfurization wastewater and slurry), and adsorbent. Noted, surrogate standards (2-fluorobiphenyl and p-terphenyl- $d_{14}$ ) were added before the Soxhlet extraction. The detailed treatment and analysis of most of the samples have been reflected in previous studies [23,28,35,37], and this section is only briefly described.

#### 2.3.1. Analytical procedure for PAHs in flue gas

The gas-phase PAHs in the XAD-2 resins and the particulate-phase PAHs in the filter membranes were extracted with dichloromethane for  $18 \sim 24$  h by the Soxhlet extractor method. Then through the steps of rotary evaporation, solvent conversion, purification by silica gel column, and so on, the samples to be detected were finally concentrated to 1 mL.

#### 2.3.2. Analytical procedure for PAHs in CPM

CPM samples collected from the field require a series of pretreatment processes in the laboratory. As shown in Fig. S3, the inorganic and organic components in CPM were treated separately, and then through a series of steps such as ultrasonic extraction and rinsing, and finally concentrated the sample of the organic fraction to 1 mL. The PAHs in the organic components of the CPM are the PAHs in the CPM.

#### 2.3.3. Analytical procedure for PAHs in solid samples

The treatment of the four types of solid samples was similar to that of PAHs samples in flue gas, which were dried and ground to particles with sizes less than 0.15 mm. And 10 g of the sample was placed in a Soxhlet extractor by the method of coning and quartering, and then concentrated the sample to 1 mL through the steps of rotary evaporation, solvent conversion, and over silica gel column treatment.

#### 2.3.4. Analytical procedure for PAHs in liquid samples

The content of PAHs is extremely low in liquid samples and also requires enrichment treatment. At present, the most convenient and extensive pretreatment method is liquid-liquid extraction. Firstly, the particulate-phase insoluble substance was removed by filtration to reduce the interference of impurities and then collected filtrate. Then, 150 mL samples and 50 mL dichloromethane were added to the separatory funnel, and the separation was set aside after shaking extraction for 20 min. Finally, the operation was repeated twice. The samples that have completed the extraction were dehydrated by sodium sulphate and evaporated by rotation to concentrate the liquid sample to 1 mL.

#### 2.3.5. Quantitative analysis of PAHs

All samples with PAHs were eventually converted into liquid samples with n-hexane as a solvent for accurate quantitative analysis using a gas chromatograph/mass spectrometer (GC/MS) system (Agilent 7890B-5977A) equipped with an HP-5MS column (30 m  $\times$  0.25 mm  $\times$  0.25  $\mu$ m). The operating parameters of the GC/MS are shown in Table S5. The whole process blanks, laboratory blanks, and transport blanks were analyzed to ensure that the analytical method is accurate. The recovery rates of surrogate standards in the sampling process were 63 to 101% (fluoranthene- $D_{10}$ ) and 74 to 106% (benzo[a]pyrene- $D_{12}$ ), respectively. After extraction, purification, concentration, and analysis of the process, the recovery rates of 2-fluorobiphenyl and p-terphenyl-d<sub>14</sub> were 79 to 105% and 85 to 116%, respectively. PAHs standard solution was added to the blank samples, and the mean recoveries ranged from 83 to 114%. 15% of the total samples were analyzed for GC/MS replicates with an average relative deviation of 5.88%, indicating that the experiment had good reproducibility. The minimum detection limit of the GC/MS system used in this study reached 0.001  $\mu$ g/m<sup>3</sup> for 16 PAHs. Seven concentration levels (ranging from 0.05 mg/ml to 2.0 mg/ml) of standard solutions were prepared based on the emission concentrations of PAHs from coal-fired power plants. The correlation coefficient (R<sup>2</sup>) of the 16 PAHs standard curve was in a range of 0.990089 (IND)  $\sim$  0.999537 (NAP), and the R<sup>2</sup> of the PAHs standard curves were all greater than 0.99, which met the quantitative requirements of the experiment.

#### 2.3.6. Analytical procedure for adsorbent

The specific surface area, pore volume, and pore size distributions of adsorbent samples were analyzed by the N<sub>2</sub> adsorption-desorption test at 77 K (Micromeritics ASAP2020-C). The specific surface area of the samples was calculated by the BET-method measurement. The pore volume of the samples was calculated by the adsorption capacity of nitrogen at the relative pressure  $P/P_0 = 0.99$ . The micropore area and volume were calculated by the t-plot method. The micropore and mesoporous pore size distributions of the two adsorbents were calculated by HK and BJH methods, respectively.

#### 3. Results and discussion

#### 3.1. Migration process of PAHs in flue gas and CPM during APCDs system

This section explored the migration process of PAHs in flue gas and CPM from bituminous coal during the air pollution purification system. In the combustion process of bituminite, PAHs in the flue gas and CPM were sampled at five sampling sites (the inlet of SCR – site A, the outlet of SCR – site B, the outlet of LLT-ESP – site C, the outlet of WFGD – site D, the outlet of WESP – site E). Note that the PAHs in the flue gas are almost all in gas-phase at the SCR inlet, and the concentration of 16 single

component particulate-phase samples is extremely low. Thus, samples with two phase states collected at site A were all processed together, while the PAHs at other sites were analyzed in the gas phase and the particulate phase. Furthermore, the removal efficiency of existing APCDs for PAHs and the distribution of PAHs in the waste produced by the devices were also explored, which would provide methods and a basis for the control and purification of PAHs from coal combustion.

#### 3.1.1. Migration process of PAHs in flue gas during APCDs system

Fig. 2 shows the change of concentration of PAHs in flue gas before and after APCDs. As shown in Fig. 2a and 2b, the concentration of PAHs at site A was 3.318 µg/Nm<sup>3</sup>, and the contribution of 2-ring and 3-ring PAHs reached 94.73%. The concentrations of PAHs at the following four sampling sites were 2.786, 1.952, 3.954, and 4.737  $\mu$ g/Nm<sup>3</sup>, respectively, and the total concentration of PAHs showed an obvious trend of decreasing first and then increasing. It is noteworthy that after site A, the concentration changes of monocomponent PAHs larger than the three rings were consistent with the tendency of total concentration. However, the dominant ring number of PAHs collected in coal-fired flue gas at each sampling site was different. The diagrams show that the dominant ring number of PAHs in flue gas increased with the purification process. It can be speculated that different purification equipment has considerable selectivity for the control of PAHs with different ring numbers in the flue gas. The removal of PAHs from APCDs and the regeneration of PAHs together affect the total concentrations.

As shown in Fig. 2c and 2d, from site B to site E, the concentration of the gas-phase PAHs was 1.29 to 2.18 times that of the particulate-phase

PAHs. Similar to studies [38–41], PAHs in flue gas from coal combustion were mainly in the gas phase, while the migration process of PAHs with two to six ring numbers in the two phases was different. Similarly, the variation of gas-phase PAHs with higher concentrations was consistent with that of total PAHs. However, compared with the change of gas-phase PAHs, the concentration of particulate-phase PAHs sampled at site E shows a downward trend. Results demonstrated that the existing apparatus also has definite selectivity for the removal of PAHs with different phases. The identification of the migration regularity of PAHs in the entire flue gas purification process provides a basis for the targeted removal of PAHs with different ring numbers and phase states at special sites.

#### 3.1.2. Migration process of PAHs in CPM during APCDs system

Fig. 3 shows the change of concentration of PAHs in CPM before and after APCDs. As shown in Fig. 3a and 3b, the concentration of PAHs in CPM at sites A and B were 7.185 and 9.070  $\mu$ g/Nm<sup>3</sup>, respectively, which is much higher than the concentration of PAHs in the flue gas. The results indicated that sufficient attention should be paid to the emission of PAHs in CPM from coal combustion, especially without the dedusting process, even if the concentration at site E was reduced to 0.554  $\mu$ g/Nm<sup>3</sup>. Combined with previous studies [22,23] on pollutant emissions from coal-fired power boilers and industrial boilers, the concentration level of PAHs in CPM emitted from coal combustion may cause considerable harm to human health and the environment due to the huge amount of coal burning. At five sampling sites, the 2-ring and 3-ring PAHs in CPM were all dominant components, which were



Fig. 2. Migration process of 16 kinds of PAHs (a), different ring numbers PAHs (b), gas-phase PAHs (c), and particulate-phase PAHs (c) in flue gas during APCDs system.



Fig. 3. Migration process of 16 kinds of PAHs (a), different ring numbers PAHs (b), and the percentage of PAHs with different ring numbers (c) in CPM during the APCDs system.

different from those in the flue gas. Fig. 3c shows the proportion of PAHs with different ring numbers in CPM during the process of coal-fired flue gas purification. At sites A to E, the proportion of 2-ring PAHs in the total concentration did not exceed 5%, while the proportion of 5-ring PAHs in the total concentration was the highest, reaching  $31.37 \sim 41.48\%$ . The proportion of 3-ring and 6-ring PAHs in the total concentration was similar, but there was no obvious variation rule. What draws special attention is that with the process of flue gas purification, the proportion of 4-ring PAHs gradually increased, and the proportion reached the highest at the stack. For the migration process of PAHs in CPM during the APCDs system, more targeted control of 4-ring and 5-ring PAHs is key to reducing the total concentration of PAHs in CPM.

#### 3.1.3. Removal effect of typical APCDs

Fig. 4 shows the removal efficiency of PAHs in flue gas and CPM from all APCDs (SCR system, LLT-ESP, WFGD system, and WESP) at the coalfired power plant. As shown in Fig. 4a, the removal efficiency of the PAHs with 2-ring and 3-ring in the flue gas exceeded 70% respectively, showing a good removal effect. However, the concentrations of three to six ring PAHs in the flue gas showed a significant negative increase after passing through the SCR system, even though the original concentration before the SCR system was very low. In addition, the concentrations of PAHs with different ring numbers in CPM all showed negative growth after passing through the SCR system. One possible explanation is that complex physical and catalytic surface chemical reactions in the SCR systems result in the conversion of 2-ring and 3-ring PAHs with lower melting and boiling points to PAHs with higher melting and boiling points, and the regeneration of PAHs in CPM. Combined with Fig. 4e, the removal efficiency of the SCR system to PAHs in flue gas and CPM was 16.04% and -26.23%, respectively. As shown in Fig. 4b, the dry-type precipitators, LLT-ESP had an excellent removal effect on PAHs from coal combustion, especially the removal efficiency of PAHs in CPM with different ring numbers was more than 90%. Besides, LLT-ESP had a considerable removal effect on PAHs in flue gas, except for the removal efficiency of 2-ring PAHs reaching 50%, the removal efficiency of the other components was about 30%. The removal efficiency of LLT-ESP to



Fig. 4. The removal efficiency of SCR (a), LLT-ESP (b), WFGD (c), WESP (d), four systems (e), and combined system (f) on PAHs in flue gas and CPM.

PAHs in flue gas and CPM was 29.92% and 94.76%, respectively. Previous studies [42,43] have shown that dry-type dust removal equipment, especially LLT-ESP, has the best removal effect on PAHs from coal combustion among existing purification devices. Fig. 4c and 4e show that the WFGD system presented the worst control effect on PAHs with different ring numbers in the coal-fired flue gas. Moreover, WFGD had a limited effect on the removal of the total concentration of PAHs in CPM. Fig. 4d and 4e show that WESP had a certain control effect on the 2-ring PAHs in flue gas and CPM from coal combustion. What is noteworthy is that WESP had a positive removal efficiency only for particulate-phase PAHs in coal-fired flue gas, while the removal efficiency for the remaining components was negative. In summary, as shown in Fig. 4f, the removal efficiency of PAHs in CPM by APCDs in the typical ultralow emission coal-fired power plant was 92.29%, while it had a negative effect on the removal of PAHs in flue gas with an efficiency of -42.77%.

Fig. S4 shows the mass fractions and mass concentrations of PAHs in some solid (slag and gypsum) and liquid wastes (desulfurization wastewater and slurry) produced by the coal-fired power plant. It can be found that the occurrence of single component PAHs in both solid and liquid wastes increased with the increase of ring numbers. It can be speculated that the 5-ring and 6-ring PAHs with higher melting and boiling points and more toxic are more likely to be enriched in the waste produced during coal combustion or flue gas purification in the process of generating electricity. As shown in Fig. S4, the mass fractions of PAHs in slag and gypsum were 0.878 and 0.883  $\mu$ g/g, and the mass concentrations of PAHs in desulfurization wastewater and slurry were 5.430 and 2.770 mg/L. With the deepening of the emphasis on the treatment of highly toxic organic pollutants in waste, it is of practical significance to study the whereabouts of PAHs in complex coal-fired power generation systems.

# 3.2. Effects of changes in feeding coal on the formation and emission of PAHs

This section explored the effect of different fuels on the generation and emission of PAHs employing coal blending. As shown in Table S3, in addition to the bituminous coal already used, blended coal was obtained through the blending of multiple coal species. In the combustion process of the two kinds of coal, PAHs in the flue gas were sampled at the outlet of the furnace, and PAHs in the flue gas and CPM were sampled at the chimney. Due to the limitations of the field conditions, the collection of CPM from blended coal combustion was not carried out at the furnace outlet. Based on this fact, this study focuses on the effect of volatiles on the formation and emission of PAHs from coal combustion, which provided basic data for source reduction technology of highly toxic PAHs.

#### 3.2.1. Effects of feeding coal species on the formation of PAHs in flue gas Fig. 5 shows the total mass concentrations of PAHs with gas and

particulate phase in coal-fired flue gas at the outlet of the boiler. Note that the total concentration of PAHs produced by combustion of blended coal decreased from 3.318 to 2.850 µg/Nm<sup>3</sup> compared to bituminite. The volatile content in raw coal should be an important influencing factor. Previous studies [44] have shown that the total PAHs emission of coal with volatile content of 7% ~30% during combustion increases with the increase of volatile content of the feeding coal. One possible reason is that the higher the volatile content in the coal, the more organic free radicals are produced by cracking under high-temperature combustion conditions, which greatly increases the number of PAHs synthesized by the reaction of free radicals in the furnace so that the total amount of PAHs generated increases. As shown in Fig. 5a, the four kinds of PAHs (Nap, Acp, Flu, and Phe) with concentrations above 0.2  $\mu$ g/Nm<sup>3</sup> were all lower during the combustion of blended coal compared with bituminous coal combustion, while the concentration changes of other PAHs were relatively limited. Fig. 5b shows the contribution of PAHs with different ring numbers to the reduction in total concentrations. Among them, the production of 3-ring PAHs decreased the most, accounting for 70% of the total reduced concentration, while the concentrations of 5-ring and 6-ring were virtually unchanged. In general, the PAHs released from the combustion of both fuels were mainly 2-ring and 3-ring.

Fig. 6 shows the mass fractions of PAHs in feeding coal. As shown in Fig. 6a, except for the almost non-existent Nap, the mass fraction of the other 15 kinds of PAHs decreased considerably in blended coal. It can be found in Fig. 6b that the mass fractions of PAHs in bituminous and blended coal were 9.133 and 5.016  $\mu$ g/g, respectively, and the content of PAHs deposited in blended coal with lower volatile was significantly reduced. Results demonstrated that 4-ring and 5-ring PAHs contributed the most to the PAHs that occurred in coal, which was different from the distribution of each ring number of PAHs in the coal-fired flue gas. Previous research found that the total PAHs concentration in bituminous coal was 9.305  $\mu$ g/g [45], and even the concentration was as high as 37.620  $\mu$ g/g in the study of Wang et al. [46]. So, in conclusion, the volatile content of blended coal can be reduced by blending coal species. Compared with bituminous coal with higher volatile content, the mass fraction of PAHs occurred in blended coal and the mass concentration of PAHs produced and emitted during combustion is lower. The field data obtained from the actual coal-fired power unit provides guidance for source reduction of PAHs in the coal-fired process. In addition to the blending of coal species, the influence of changes in parameters such as atmosphere and oxygen partial pressure in the furnace on the formation of PAHs needs further research.

3.2.2. Effects of feeding coal species on the emission of PAHs in flue gas

Fig. 7 shows the mass concentrations of gas-phase and particulatephase PAHs in flue gas emitted from the coal-fired unit. It is observed that the concentration of PAHs released by blended coal combustion



Fig. 5. Mass concentrations of 16 kinds of PAHs (a) and different ring numbers of PAHs (b) in flue gas at the outlet of the furnace.



Fig. 6. Mass fractions of 16 kinds of PAHs (a) and different ring numbers PAHs (b) in feeding coal.



Fig. 7. Mass concentrations of 16 kinds of gas-phase (a), particulate-phase PAHs (b), and different ring numbers PAHs (c), and TEQ of PAHs with different numbers of rings (d).

 $(3.008 \ \mu g/Nm^3)$  was significantly lower than that emitted by bituminous coal combustion  $(4.737 \ \mu g/Nm^3)$ . Fig. 7a and 7b show the emission concentration of 16 kinds of monocomponent PAHs with two phases from bituminous and blended coal combustion. It was found that the emission concentrations of monocomponent gas- and particulate-phase PAHs in flue gas were lower when blended coal was used as fuel. As shown in Fig. 7c and 7d, the distribution of PAHs with different ring numbers in the flue gas emitted by the combustion of the two coals was similar. For gas-phase PAHs, 3-ring PAHs were the dominant compounds; while for particulate-phase PAHs, the more toxic 5-ring PAHs

contributed more. Additionally, as in previous studies [22,28,47], 5-ring PAHs had the highest toxicity, and the TEQs of PAHs in flue gas emitted from bituminous and blended coal were 0.958 and 0.586  $\mu$ g/Nm<sup>3</sup>, respectively. It is worth noting that the combustion of blended coal with lower volatile content will also lead to the reduction of PAHs emission concentrations in the coal-fired flue gas.

#### 3.2.3. Effects of feeding coal species on the emission of PAHs in CPM

Fig. 8 shows the influence of two types of feeding coal on the emission concentrations of PAHs in CPM. It is observed that the total



Fig. 8. Mass concentrations of 16 kinds of PAHs in CPM (a), and the proportion of different ring numbers PAHs in concentration and TEQ (b).

emission concentration of PAHs in CPM decreased from 0.554 (fueled by bituminous coal) to 0.429 (fueled by blended coal)  $\mu$ g/Nm<sup>3</sup>. As shown in Fig. 8a, different from the effect on the emission concentration of single component PAHs in flue gas, the concentrations of some 4-ring and 5ring PAHs in CPM decreased significantly after switching to blended coal as fuel. Fig. 8b shows that the PAHs in the CPM emitted from two kinds of coal combustion were mainly 4-ring and 5-ring PAHs, and the 5ring PAHs contributed almost all the TEQs. The study [23] shows that the concentrations of PAHs in CPM emitted from two coal-fired power units were 1.43 and 2.08 mg/Nm<sup>3</sup>, which were higher than that of coalfired industrial units (0.17 and 1.31 mg/Nm<sup>3</sup>). This conclusion demonstrated that the concentration of pollutant emissions may be affected by the type of coal source. Although the emission concentration of PAHs in CPM is relatively lower than that in coal-fired flue gas, it is also of great practical significance to control the emission of highly toxic PAHs in CPM due to the increasing understanding of the properties and hazards of CPM.

To sum up, the mass fraction of PAHs in blended coal decreased by 45.08% by blending different categories of coal. Compared with the use of bituminous coal as fuel, at the outlet of the furnace, the concentration of PAHs in flue gas from blended coal combustion decreased by 14.10%; at the chimney, the emission concentration of PAHs in flue gas from blended coal combustion decreased by 36.50%, and the concentration of PAHs in CPM decreased by 22.56%. Results demonstrated that the content of PAHs in blended coal can be effectively reduced by blending different coals, it has a relatively positive effect on the reduction of formation and emissions of PAHs. Noted, the development of source reduction technology for highly toxic pollutant PAHs can provide an important guarantee for the healthy development of China's electric power industry.

### 3.3. Control of PAHs from coal combustion by adsorbent injection coupled with electrostatic precipitator technology

This section explored the control method of PAHs emitted from large ultralow emission coal-fired power plants using carrying flow jet adsorption technology. The control technology was applied in the coal-fired power plant, and the adsorption material saturated in the flue was finally obtained, so it needs to be combined with the dust removal equipment. Therefore, the target flue of the spray gun was limited before the LLT-ESP, and the flue gas temperature was usually  $80 \sim 150$  °C. According to the application of this control technology in municipal solid waste incineration [48,49], the parameters such as the amount of injection and physical properties of adsorption material were selected. Table S4 shows the flue gas parameters and distribution of SM and SO<sub>2</sub> in the flue gas, the temperature was in the range of medium-high

temperature, so the adsorption environment had the characteristics of high sulfur, high dust, and medium-high temperature. Adsorbent injection technology coupled with electrostatic precipitators to remove highly toxic organic pollutants such as PAHs produced from coal combustion may become one of the research hotspots in the future, and has high guiding value for practical engineering applications, and will also be part of the precise control of future intelligent power plants.

#### 3.3.1. Parameter analysis of adsorbents

Two kinds of commercial activated carbon A1 and A2 were selected for engineering demonstration applications. Fig. S5 shows the surface morphological characteristics of the two adsorbents. Table S6 shows the properties of the two adsorbents. It can be found that the specific surface area of A<sub>1</sub> (1278.28  $m^2/g$ ) was slightly higher than that of A<sub>2</sub> (1013.12  $m^2/g$ ), but the total pore volume of A<sub>1</sub> (1.07 cm<sup>3</sup>/g) was much higher than that of  $A_2$  (0.60 cm<sup>3</sup>/g) while they have a quite similar micropore volume. Fig. S6 shows the N2 adsorption and desorption isotherms, micropore, and mesoporous size distributions of the two adsorbents. Obviously, as shown in Fig. S6a, the adsorption capacity of  $A_1\xspace$  was higher than that of A2, and the low temperature nitrogen adsorption isotherm of A1 conformed to the type IV isotherm defined by the International Union of Pure and Applied Chemistry (IUPAC), in which the type H4 adsorption hysteresis ring in the middle segment corresponded to the system with capillary condensation of porous adsorbent. However, A<sub>2</sub> conformed to the isotherm of type I, reflecting the micropore filling phenomenon on the micropore adsorbent. Fig. S6b and S6c show that the distribution of micropores smaller than 0.4 nm in the two adsorbents was similar, and the distribution of micropores larger than 0.4 nm in A1 was more abundant; besides, mesoporous pores larger than 2 nm and smaller than 30 nm were more widely distributed in A1 and that can be the reason why the pore volume of  $A_1$  was much higher than  $A_2$ , especially the external pore volume.

#### 3.3.2. The removal effect of the adsorbents on PAHs

Fig. 9 shows the removal efficiency of PAHs in coal-fired flue gas by adsorbent injection coupled with the LLT-ESP. In the absence of spray adsorbent conditions, the total concentration of PAHs in the flue gas was reduced from 4.949 to  $3.266 \ \mu g/Nm^3$  through LLT-ESP removal, and the removal efficiency was 34.01%, which was similar to the conclusion of the study on the migration process of PAHs in the previous chapter (29.92%). Under the condition that the injection volume of adsorbent A<sub>2</sub> was set at 60 and 170 kg/h, the removal efficiency of PAHs by the combined system increased to 46.61% and 57.33% respectively, indicating that increasing the spray of adsorbent before the LLT-ESP can effectively reduce the emission concentration of PAHs in flue gas, and appropriately increasing the injection volume of adsorbent was conducive to the increase of removal efficiency. It is worth noting that the



Fig. 9. The removal efficiency of PAHs from coal combustion by adsorbent injection coupled with an electrostatic precipitator.

removal efficiency of PAHs with LLT-ESP combined with  $A_1$  as an adsorbent was as high as 70.68%, nearly 40% higher than that without adsorbent injection, which is the best working condition for removal effect. The study [50] presents the mechanism of PAHs adsorption on activated carbon through fixed-bed adsorption tests and revealed the BET specific surface area had the greatest influence on PAHs adsorption. The adsorption of low molecular weight PAHs with 2-ring and 3-ring were dependent on the micropore volume, while middle molecular weight PAHs with 4-ring were more dependent on the mesopore volume. Thus, in contrast with  $A_2$ , the  $A_1$  has higher PAHs adsorption efficiency a higher BET surface area, and much higher pore volume, especially the external pore volume.

Eventually, under the condition that the spray volume of sorbent A<sub>1</sub> was set at 170 kg/h, the total emission concentration of PAHs in the flue gas was reduced to 1.451  $\mu$ g/Nm<sup>3</sup> after passing through the LLT-ESP. The results of the field test and the characterization analysis of the two adsorbents are mutually verified, which guides the control methods of PAHs emitted from coal-fired power plants in the future.

## 3.3.3. The characteristics of fly ash in three electric fields under the LLT-ESP

According to the conclusion in the above section, adsorbent injection combined with LLT-ESP had the highest removal efficiency for PAHs from coal combustion when the spray volume of adsorbent  $A_1$  reached 170 kg/h. And, in consequence, fly ash was collected from the ash hopper under a three-level electric field of the LLT-ESP before and 2 h, and 4 h after the adsorbent  $A_1$  injection (170 kg/h), and all samples were characterized (SEM and EDS). Noted, in Table S7, the mass fraction of PAHs in fly ash collected four hours after the adsorbent injection was also measured.

As shown in Fig. S7, the fly ash was mainly composed of regular spherical particles, and there was basically no obvious pore structure on the surface of the fly ash, but there were many irregular flocs which are beneficial to the mutual adhesion between the fly ash. Under the condition of no adsorbent injection, there was more fly ash with a diameter of fewer than 2  $\mu$ m in the third electric field hopper, while there was more fly ash with a large size (about 10  $\mu$ m) in the first and the second electric field hoppers, which may be due to the collision between adsorbent A<sub>1</sub> and fly ash in the flue gas and increase the turbulent agglomeration effect of the coal-fired flue gas. When adsorbent A<sub>1</sub> was sprayed continuously, the size of fly ash in the first and second electric field ash hoppers decreased, but the size of fly ash in the third electric field ash hopper increased. The massive structure, which was obviously

different from the shape of fly ash, was found with sharp edges and corners around, which may be the adsorbent  $A_1$ . Table S8 shows that after 2 h of continuous injection of adsorbent  $A_1$ , the carbon content of fly ash in the primary and secondary electric fields increased significantly; after 4 h, the carbon content of fly ash in the primary, secondary, and tertiary electric fields increased. In addition, the oxygen content of fly ash decreased, but the changes in nitrogen content and sulfur content were not obvious. Combined with Fig. S8, it can be speculated that the increase of carbon content in fly ash is mainly caused by the injection of adsorbent  $A_1$ , which means that the adsorbent would be mainly captured by the primary and secondary electric fields of LLT-ESP.

The mass fractions of PAHs in fly ash from the three electric fields are shown in Fig. S8. The total mass fractions of PAHs in fly ash collected in the three electric fields of the LLT-ESP were 0.916, 0.486, and 0.446  $\mu g/g$ . The total PAH concentration decreased from the first to the third electric field. Similar to Li et al. [43] 's study, the first electric field plays a crucial role in the removal of PAHs from the coal-fired flue gas. Due to the decrease of flue gas temperature, when flue gas passed through the first electric field, part of PAHs was adsorbed by adsorbent, and part of PAHs was adhered to fly ash. Finally, PAHs were removed together with adsorbent and fly ash by LLT-ESP. The occurrence of PAHs in fly ash also proved that the adsorbents were mainly collected by the first and second electric fields of LLT-ESP.

#### 4. Conclusions

PAHs in flue gas and CPM from a 300 MW ultralow emission coalfired power plant equipped with multiple types of APCDs (SCR, LLT-ESP, WFGD, and WESP) were sampled. The effects of coal species on PAHs generation and emissions, the migration of PAHs during the flue gas purification process, and the removal effect of APCDs on it were investigated. Furthermore, this study explored the control method of PAHs from coal combustion by adsorbent injection coupled with electrostatic precipitator technology. The analysis results are as follows: (1) PAHs in flue gas from coal combustion were mainly in the gas phase, and the existing APCDs have definite selectivity for the removal of PAHs with different phases and rings numbers. The proportion of 4-ring PAHs increased in CPM with the process of flue gas purification, and control of 4-ring and 5-ring PAHs is key to reducing the total concentration of PAHs in CPM. (2) The LLT-ESP has the best removal effect on PAHs, the removal efficiency of the apparatus to PAHs in flue gas and CPM was 29.92% and 94.76%, respectively. However, the WFGD system presented the worst control effect on PAHs from coal combustion. (3) The occurrence of PAHs can be effectively reduced by blending different coals. Replacing bituminous coal with blended coal as fuel can reduce the concentration of PAHs emitted at the outlet of the furnace (PAHs in flue gas, decreased by 14.10%) and at the chimney (PAHs in flue gas, decreased by 36.50%; PAHs in CPM, decreased by 22.56%). (4) The adsorbent injection coupled with LLT-ESP had a favorable removal effect on the PAHs in the coal-fired flue gas, and the removal efficiency reached up to 70.68%, which was nearly 40% higher than the efficiency without the spray adsorbent.

#### CRediT authorship contribution statement

Yujia Wu: Conceptualization, Formal analysis, Data curation, Writing – original draft. Zhenyao Xu: Software. Xinlei Huang: Writing – original draft. Siqi Liu: Investigation. Minghui Tang: Methodology. Shengyong Lu: Supervision.

#### **Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.fuel.2022.125052.

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