

http://pubs.acs.org/journal/acsodf

# Emission Characterization of Particulate Matters from the Combustion of Pulverized Coals in a Simulated Fluidized Bed Boiler

Jintuo Zhu, Haisong Sun, Wanxing Ren,\* Xinjian He,\* Liang Wang, Yongliang Yang, and Guoqing Shi





## ACCESS

III Metrics & More

**ABSTRACT:** Coal-fired power generation is one of the main causes of air pollution, and the fluidized bed technology is currently a commercially used coal-fired technology. Therefore, it is of great significance to investigate the characteristics of particulate matter released from the fluidized bed boiler. In this study, lignite, bituminous coal and anthracite with particle sizes of <75  $\mu$ m and 180–830  $\mu$ m were selected and burned completely at 700, 800, and 900 °C for the purpose of simulating the process of pulverized coal combustion in a small sized simulated fluidized bed boiler and exploring the effects of coal rank, particle size, and burning temperature on the characteristics of the released particulate matter. The results show that, under the same mass, bituminous coal combustion releases the most PM1, PM2.5, and PM10, followed by lignite and anthracite. For all combusted coals the released PM1 accounts for half and one-third of the PM2.5 and PM10, respectively, and the released PM2.5 accounts for half of the



Article Recommendations

PM10. A smaller particle size of pulverized coal and a higher burning temperature correspond to the release of more submicron to micron particulate matter. The mass concentration of released particulate matter for lignite and bituminous coal shows a bimodal distribution, with the two peak values in the ranges of 0.1-0.18 and  $3.2-10 \mu$ m, respectively. As the burning temperature increases and the particle size of pulverized coal decreases, the first peak value falls and the second peak shifts to a small particle size range. This study can serve as reference for diminishing the emission of submicron to micron particulate matter by coal-fired power plants and preventing air pollution.

### **1. INTRODUCTION**

According to the 2020 Global Air Quality Report, 82 of the 106 countries tested failed to meet the World Health Organization air quality standards, and air pollution remains one of the major threats to environmental health in the world.<sup>1-3</sup> Coal combustion is the main cause of air pollution in various countries. In China, coal-fired power generation is the main use of coal combustion, which accounts for about half of the total coal consumption.<sup>4-6</sup> In the process of coal combustion in power plants, massive micron particulate matter, known as PM10, is released, causing severe air pollution in most Chinese cities. Moreover, PM1 and PM2.5 with smaller particle sizes have larger specific surface areas, which can absorb heavy metal elements and are difficult to be captured by the dust removal equipment. Hence, they are more likely to cause greater harm to environment and human health.<sup>7-10</sup> China is a country that is rich in coal and lacks oil and gas. Such a basic national condition will guarantee the main status of coal in energy for a long time. Therefore, the control of submicron to micron particulate matter released from coal combustion is a major problem to be solved urgently.<sup>11–13</sup> According to different combustion methods, coal-fired power station boilers can be classified into pulverized coal furnaces, fluidized bed boilers, and chain furnaces. Among them, due to the advantages of low combustion

temperature, low investment, high thermal efficiency and wide fuel adaptability, fluidized bed combustion technology has been widely commercialized. At present, China has become the country with the largest number of fluidized bed boilers in the world.<sup>14,15</sup>

The emission characteristics of particulate matter from coalfired power plant boilers are affected by many factors, such as combustion temperature, combustion atmosphere, coal characteristics, and combustion time. Among them, combustion temperature and coal characteristics are the most important influencing factors. Therefore, it is very necessary to investigate the influence of combustion temperature and coal characteristics on the emission of particulate matter from different types of boilers. Through burning pulverized coal with different particle sizes and types in pulverized coal boilers at different temperatures, Lv and Li,<sup>16,17</sup> Wen et al.,<sup>18</sup> Seames et al.,<sup>19</sup> and

Received:August 8, 2022Accepted:October 4, 2022Published:October 14, 2022



Ninomiya et al.<sup>20</sup> explored the influence of coal type, coal particle size and combustion temperature on particulate emission characteristics of pulverized coal boiler. However, due to different combustion temperatures and combustion mechanisms, the experimental results of pulverized coal fired boilers (1100–1400 °C) may not be applicable to fluidized bed boilers (700–1000 °C). For fluidized bed boilers, there are few studies on the influence of combustion temperature and pulverized coal characteristics on particulate matter emission characteristics. Under laboratory fluidized bed conditions, Zhang et al.<sup>21</sup> burned the mixtures of peat and sludge at different combustion temperatures and investigated the influence of combustion temperature on particulate emission characteristics of peat and sludge co-combustion. Under the condition of pressurized fluidized bed (i.e., in a special pressurized environment), Wang et al.22 burned lignite, bituminous coal, and a mixture of lignite, bituminous coal, and biomass, and reported that coal samples containing more volatile components would produce more submicron particles. In addition, Wang et al.<sup>23</sup> selected three particle sizes of coal particles for combustion in a fluidized bed boiler and explored the effect of coal particle size on the particle size distribution of bottom slag. To date, the influence of combustion temperature and coal characteristics on the emission characteristics of particulate matter in fluidized bed boilers is still unclear.

In view of this, in this study, three typical ranks of coal most widely used in coal-fired power station boilers, i.e., lignite, bituminous coal, and anthracite, were selected as the experimental pulverized coal samples. Within the burning temperature range (700–900 °C) of fluidized bed boilers, 700, 800, and 900 °C were selected as the burning temperatures. According to the particle size range (0–10 mm) of pulverized-coal burning in the fluidized bed boilers, the experimental pulverized coal was treated to <75 and 180–830  $\mu$ m, so as to study the influence of coal rank, burning temperature, and particle size of pulverized coal on the submicron to micron particulate matter released from coal combustion. This study is expected to provide reference for the reduction of air pollution.

#### 2. EXPERIMENTAL STUDY

**2.1. Experimental Pulverized Coal Samples.** Lignite, bituminous coal, and anthracite, whose ignition points are 300–400, 400–500, and 550–600 °C, respectively, were selected as the experimental coal samples. Before the experiment, the coal samples were preprocessed. First, they were dried and crushed. Then, according to the particle size range of pulverized coal in fluidized bed boilers, a standard sieve was used to screen out those with particle sizes of  $<75 \,\mu$ m and  $180-830 \,\mu$ m. To ensure complete combustion of the pulverized coal, the mass of the coal sample for each experimental trial is 10 g. According to the Chinese national standard Proximate Analysis of Coal (GB/T212-2008),<sup>24</sup> proximate analysis results are given in Table 1.

**2.2. Experimental System.** As presented in Figure 1, the self-developed experimental system is mainly composed of a small-sized simulated fluidized bed boiler and a released particulate matter collection system. The simulated fluidized bed boiler was designed with reference to Zhang et al.<sup>21</sup> and Bhattacharya and Roy,<sup>25,26</sup> which consists of an air supply system (O<sub>2</sub> cylinder + N<sub>2</sub> cylinder + airflow meter + air

#### Table 1. Proximate Analysis Results of the Coal Samples

| coal sample     | moisture (%) | ash (%) | volatile (%) | fixed carbon (%) |
|-----------------|--------------|---------|--------------|------------------|
| lignite         | 18.00        | 10.20   | 41.54        | 30.29            |
| bituminous coal | 2.37         | 5.12    | 33.19        | 59.32            |
| anthracite      | 1.63         | 14.43   | 9.41         | 74.53            |

distributor), a coal feeding system (coal bunker + coal feeder), and ta emperature-controlled coal combustion system (combustion chamber + carbon silicon heater + PID temperature controller). The released particulate matter collection system is composed of a "cooling tube + multistage impact separation sampler + pressure monitor + vacuum pump". During the experimental process, mixed air consisting of 21%  $O_2$  + 79%  $N_2$ (volume ratio) was supplied at 10 L/min. The airflow was first preheated by the lower section of the carbon silicon heater, then evenly distributed by the air distributor, and finally released into the combustion chamber. The internal space of the combustion chamber was heated by the carbon silicon heater to the set temperature and controlled within  $\pm 5$  °C by the PID temperature controller. Through the coal feeder, the pulverized coal was fed at 10 g/min into the combustion chamber. Under the action of the pumped air flow, the smoke generated during the coal burning process was first cooled by the cooling tube and then the released particulate matter was separately collected by the multistage impact separation sampler.

2.3. Separation of Particulate Matter Released by Coal Combustion. The multistage impact separation sampler used in this study (model 110, TSI Inc., Shoreview, MN, USA) comprises a total of 11 stages, and the particle sizes after the classification include >18, 10-18, 5.6-10, 3.2-5.6, 1.8-3.2, 1.0-1.8, 0.56-1.0, 0.32-0.56, 0.18-0.32, 0.1-0.18, and 0.056–0.1  $\mu$ m in a descending order. It covers the particle size range of submicron to micron particulate matter that poses the greatest threat to human health and has the most significant effect on air pollution. The filter membrane used in this experiment is a smooth aluminum film without static electricity. Before the experiment, in order to prevent the rebound of collected particulate matter, a layer of aspirin was coated on the aluminum film first, and then the filter film was balanced in a constant-temperature-and-humidity chamber for 24 h at a balance temperature of 25 °C and relative humidity of 50%. Afterward, the aluminum film was weighed and recorded with a 0.1 mg analytical balance and then installed into the sampler for sampling. Subsequently, the aluminum film was placed in the constant-temperature-and-humidity chamber to reach equilibrium for 24 h under the same temperature and humidity conditions. Finally, the weight of the aluminum film after sampling was recorded with the same analytical balance, and then the mass of particulate matter collected by each stage of the aluminum film on the sampler was calculated.

**2.4. Data Process.** In order to reduce the experimental error, each experimental combination is repeated three times under the same conditions. In addition, this experiment compares the mass (mg) of particulate matter collected by all levels of filter membrane with the mass ( $g_{coal}$ ) of coal sample and obtains the emission factors (mg/ $g_{coal}$ ) of particulate matter in each particle size range. In order to analyze PM1, PM2.5, and PM10 released from coal burning, particulate matter in the particle size ranges of 0.056–1.0, 0.056–3.2, and 0.056–10  $\mu$ m collected by the impact separation sampler was approximately regarded as PM1, PM2.5, and PM10, respectively.



**Figure 1.** Schematic diagram of experimental system. (1)  $O_2$  cylinder; (2)  $N_2$  cylinder; (3,4) Airflow meter; (5) Air distributor; (6) Coal bunker; (7) Coal feeder; (8) Combustion chamber ( $\Phi$ 50 \* 1000); (9) Carbon silicon heater; (10) Temperature controller; (11) Cooling tube; (12) Multistage impact separation sampler; (13) Pressure monitor; (14) Vacuum pump.



Figure 2. Emission factors of particulate matter with diverse particle size ranges for the combustion of different-rank pulverized coals.

#### 3. RESULTS AND DISCUSSION

**3.1. Influence of Coal Rank on the Submicron to Micron Particulate Matter Emission Factors.** Figure 2 shows the emission factors of particulate matter with diverse particle size ranges for the combustion of different-rank pulverized coals. As can be seen, the emission factors of bituminous coal in different particle size ranges are obviously higher than those of lignite and anthracite. As for the particle size distribution of emission factors, the emission factors of both lignite and bituminous coal show a bimodal distribution at three



Figure 3. PM1, PM2.5, and PM10 emission factors for the combustion of different-rank pulverized coals.

temperatures, with the first and second peak values appearing in the ranges of 0.1–0.18 and 3.2–10  $\mu$ m, respectively, and the second peak value is much higher than that of the first one, which is consistent with the classical theory:<sup>27</sup> the particulate matter released by pulverized coal combustion presents a bimodal distribution, with the first peak value appearing in the submicron area (near 0.1  $\mu$ m), and the submicron particulate matter is generated by the gasification and condensation of inorganics in the combusted coal particles. The second peak value occurs in the micron area (>1  $\mu$ m), and the micron particulate matter comes from the fragmentation and polymerization of coke particles during the combustion. However, in this experiment, the emission factors of anthracite at 700 and 800  $^\circ C$  showed a unimodal distribution with the peak occurring in the micron area, and at 900 °C the emission factors showed a bimodal distribution with the same peak ranges as lignite and bituminous coal. Wang et al.<sup>22</sup> and Huang et al.<sup>28</sup> found that the mineral composition of coal, especially the volatile content, has a significant effect on the generation of submicron particles,. Anthracite has a high degree of metamorphism and low volatile content (see Table 1). At lower combustion temperatures (700 and 800 °C), less submicron particulate matter is produced by gasification and condensation, resulting in failure to present the peak in the submicron region. With the increase of the combustion temperature, the gasification rate of the inorganic substances in the coal is accelerated, generating more submicron particulate matter, thus the emission factor peaks in the submicron region at a temperature of 900  $^{\circ}$ C.

Figure 3 presents PM1, PM2.5, and PM10 emission factors for the combustion of different-rank pulverized coals. It is found that bituminous coal corresponds to the highest emission factors, followed by lignite and anthracite. Specifically, the PM1, PM2.5, and PM10 emission factors of bituminous coal are 2.55-4.08, 2.68-3.99, and 2.74-4.23 times those of anthracite, and the emission factors of lignite are 1.27-2.60, 1.38-2.42, and 1.67-2.54 times those of anthracite, respectively. Naydenova et al.<sup>29</sup> and Wang et al.<sup>22</sup> explored the characteristics of generated particulate matter from the combustion of different fuels through a laboratory-scale fluidized bed and reported that the particulate matter emission was highly affected by the volatile content of coal. The higher the content of volatile matter, the higher the expansion rate of coke particles, and thus the more easily the coke particles break to generate more particulate matter. By comparing the proximate analysis results of the three ranks of coal (see Table 1), the volatile matter content of lignite is higher than that of bituminous coal, while the emission factors of lignite for PM1, PM2.5, and PM10 are lower than those of bituminous coal. This suggests that although the volatile matter



Figure 4. Emission factors of particulate matter with different particle size ranges for the pulverized coals burning at different temperatures.

promotes the generation of particulate matter to a certain extent, it is not the leading factor that determines the emission factors of submicron to micron particulate matter (PM1, PM2.5, and PM10) released during coal burning. Through the above analysis of the combustion of pulverized coal, it is concluded that the emission of particulate matter under the condition of fluidized bed is the result of the comprehensive action of pulverized coal characteristics and combustion conditions.

3.2. Influence of Burning Temperature on the Submicron to Micron Particulate Matter Emission Factors. Figure 4 displays the emission factors of particulate matter in different particle size ranges for the pulverized coal burning at different temperatures. With the rise of burning temperature, the emission factors of all three ranks of coal show an upward trend, except those of lignite and bituminous coal which fall in the range of 0.056–0.32  $\mu m.$  In addition, with the rise of burning temperature, the peak values of emission factors in the submicron area for lignite and bituminous coal first decrease then increase, and for all three coals, the particle sizes corresponding to the emission peak values in the micron area gradually shift to smaller particle size range. The two peak values change because lignite and bituminous coal have high volatile matter contents (see Table 1) which can produce a large amount of coal tar in the pyrolysis process.<sup>30</sup> At a low burning

temperature, coal tar may be gasified into flue gas without being completely burned. As the ambient temperature drops, the flue gas will condense into submicron aerosols. During this experiment at 700 °C, particulate matter released from lignite and bituminous coal combustion is collected on the filter membrane, and a certain amount of yellow viscous oil matter, namely, coal tar particles, are visible. These coal tar particles are mainly distributed on the 10th filter membrane, resulting in the large peak values of emission factors in the range of  $0.1-0.18 \,\mu m$ for lignite and bituminous coal. However, with the rise of burning temperature, the amount of incompletely combusted coal tar dwindles, and submicron coal tar particles are reduced (at 800 °C, almost no coal tar particles collected on the filter membrane), leading to a significant decrease in the peak values of emission factors in the submicron area for lignite and bituminous coal. As the combustion temperature increases from 800 to 900 °C, the inorganic substances in pulverized coal that vaporize and condense increases, and the released micron-sized particulate matter increases, leading to an increase in the peak emission factors in the submicron for lignite and bituminous coal. With the increase of combustion temperature, the thermal stress on coke particles increases and intensifies the collision between coke particles, thus the breakage of coke particles becomes more severe and the generated and released smaller



Figure 5. Emission factors of PM1, PM2.5, and PM10 released from coal combustion at different burning temperatures.

| Table 2. PM1/PM2.5, PM1/PM10, and PM2.5/PM10 of Particulate Matter Released from the Pulverized Coals Burnin | ig at |
|--|-------|
| Different Temperatures   | -     |

| coal sample                | temp   | (PM1/PM2.5)% | (PM1/PM10)% | (PM2.5/PM10)% |
|----------------------------|--------|--------------|-------------|---------------|
| $<75 \ \mu m$ lignite      | 700 °C | 60.8         | 38.4        | 64.7          |
|                            | 800 °C | 58.0         | 32.6        | 62.1          |
|                            | 900 °C | 54.9         | 34.3        | 63.3          |
| 180–830 $\mu$ m lignite    | 700 °C | 59.4         | 32.6        | 63.6          |
|                            | 800 °C | 52.5         | 33.1        | 59.4          |
|                            | 900 °C | 54.2         | 31.8        | 62.6          |
| <75 $\mu$ m bituminous     | 700 °C | 51.2         | 32.8        | 64.1          |
|                            | 800 °C | 46.1         | 33.3        | 72.2          |
|                            | 900 °C | 49.7         | 36.2        | 72.9          |
| 180–830 $\mu$ m bituminous | 700 °C | 58.8         | 30.9        | 52.5          |
|                            | 800 °C | 58.4         | 29.8        | 51.0          |
|                            | 900 °C | 46.2         | 30.0        | 56.2          |
| <75 $\mu$ m anthracite     | 700 °C | 60.0         | 38.4        | 63.8          |
|                            | 800 °C | 57.2         | 43.1        | 75.5          |
|                            | 900 °C | 52.4         | 38.8        | 74.2          |
| 180–830 $\mu$ m anthracite | 700 °C | 56.6         | 37.2        | 65.6          |
|                            | 800 °C | 50.8         | 31.9        | 62.7          |
|                            | 900 °C | 52.3         | 35.9        | 68.5          |



Figure 6. Emission factors of particulate matter in different particle size ranges released from the combustion of pulverized coals with different particle sizes.

size particles increases,<sup>31,32</sup> resulting in the particle size corresponding to the peak value of emission factors in the micron area shifts to a small particle size range.

Figure 5 presents the emission factors of PM1, PM2.5, and PM10 for the pulverized coal burning at different temperatures. As can be seen, under the fluidized bed condition, PM1, PM2.5 of lignite and PM1 of bituminous coal all decrease first and then increase with the increase of temperature. PM10 of lignite, PM2.5, PM10 of bituminous coal and PM1, PM2.5, PM10 of anthracite all increase with the increase of temperature. However, through the combustion of pulverized coal in pulverized coal boiler at temperature range of 900-1100 °C, Seames et al.<sup>19</sup> and Zhang et al.<sup>33</sup> found that, with the increase of combustion temperature, the evaporation rate of inorganic matters in pulverized coal increased, and more submicron particles PM1 were generated. By comparison, it can be seen that the influence of combustion temperature on particle emission of fluidized bed is partially different from that of pulverized coal boiler, and the difference is mainly caused by the influence of combustion temperature on the generation of submicron particles. The combustion temperature range of the fluidized bed is 700–1000 °C, and when pulverized coal was burnt at 700 °C, lignite and bituminous coal with high volatile matter contents (see Table 1) would produced submicron coal tar particles during the combustion process. By checking the filter membrane after sampling, it was found that the amount of submicron coal tar particles accounted for the vast majority of PM1. As the burning temperature increased, on the one hand, the evaporation rate of inorganics in coal accelerates, and the

generated PM1 increases, on the other hand, submicron coal tar particles decrease with the rise of temperature, and the reduction of PM1 is more than the increment, resulting in the decrease of the total production of PM1 with the increase of combustion temperature. However, the volatile matter content of anthracite is low, and coal tar particles are not generated in the combustion process. As the burning temperature rises, the evaporation rate of inorganics in anthracite accelerates, and more submicron particles are produced, which leads to an increase in the PM1 emission factors of anthracite.

PM1/PM2.5, PM1/PM10, and PM2.5/PM10 in particulate matter released from coal burning at different temperatures are listed in Table 2. As can be seen, the PM1/PM2.5, PM1/PM10, and PM2.5/PM10 for all three coals lie in 46.1-60.8%, 29.8-43.1%, and 51.0–75.5%, respectively, suggesting that, during the combustion process of either lignite, bituminous coal, or anthracite, the released PM1 accounts for a half and one-third of the PM2.5 and PM10, respectively, and the released PM2.5 accounts for about half of the PM10. In addition, it is found that, with the increase of combustion temperature, released PM1/ PM2.5, PM1/PM10, and PM2.5/PM10 for the three coal samples decrease, which agrees with the finding of Lv and Li<sup>17</sup> on coal combustion in a pulverized coal boiler (900-1100 °C). However, the reasons for the decrease of PM1/PM2.5, PM1/ PM10, and PM2.5/PM10 are different. In the fluidized bed, with the increase of temperature, the incomplete combustion of coal tar is reduced, and the submicron coal tar particles produced are also reduced, while the amount of micron particles increases with the increase of temperature, resulting in the decrease of



Figure 7. Emission factors of PM1, PM2.5, and PM10 released from the combustion of pulverized coals with different particle sizes.

PM1/PM2.5, PM1/PM10, and PM 2.5/PM 10 with the increase of temperature. In a pulverized coal fired boiler, with the increase of temperature, submicron particles easily adhere to the surface of large particles and agglomerate, which reduces the amount of submicron particles and increases the amount of micron particles. Finally, PM1/PM2.5, PM1/PM10, and PM 2.5/PM 10 also decrease with the increase of temperature.

3.3. Influence of Burned Coal Particle Size on the Submicron To Micron Particulate Matter Emission Factors. Figure 6 presents the emission factors of particulate matter in different particle size ranges released from the combustion of pulverized coal with different particle sizes. The emission factors in all particle size ranges for the combustion of  $<75 \ \mu m$  pulverized coal are notably higher than those of 180-830  $\mu$ m. In addition, with the decrease of particle size of pulverized coal, the peak values of emission factors in the submicron area for lignite and bituminous coal decrease, and the peak values of emission factors in the micron area for lignite, bituminous coal and anthracite gradually occur in a smaller particle size range. The main reason may be that with the decrease of particle size, the pulverized coal combusts more completely, and fewer coal tar particles are released. As a result, the peak values of particulate matter in the submicron area released from the combustion of lignite and bituminous coal drop. Moreover, the smaller particle size of combusted pulverized coal causes a decline of average particle size of minerals in pulverized coal. Under the same combustion condition, the particle size of produced particulate matter dwindles accordingly, so that the peak value of emission factors in the micron area deviates to a small particle size range.

Figure 7 shows the emission factors of PM1, PM2.5, and PM10 released from the combustion of pulverized coal with different particle sizes. The three ranks of coal samples demonstrate the same rule at three different temperatures; that is, when the pulverized coal is finer, more PM1, PM2.5, and PM10 are released from the complete combustion. It can be seen that, under the condition of fluidized bed, the finer the pulverized coal is burned, the more submicron to micron particles will be released by complete coal combustion, and the greater the harm to the environment. This rule is the same as the influence of pulverized coal particle size on particle emission of pulverized coal boiler.<sup>20,34</sup> There are two main reasons for this: (1) The smaller the particle size of pulverized coal is, the larger the specific surface area is, and the lower the ignition temperature is, thus the more intense the combustion is. This increases the amount of gasification of inorganic elements and hence cause an increase in submicron particulate matter. (2)With the decrease of the particle size of pulverized coal, some of the internal minerals in the coal particles are converted into external minerals, and during the fluidized bed combustion process, the external minerals can be directly converted into micron sized particles, increasing the generation of micron sized particles.

Table 3 displays PM1/PM2.5, PM1/PM10 and PM2.5/ PM10 in particulate matter released from the combustion of pulverized coal with different particle sizes. As can be seen, with the decrease of particle size of pulverized coal, PM1/PM2.5, PM1/PM10, and PM2.5/PM10 for all three ranks of coal increase. The main reasons are as follows: on the one hand, the reduction of particle size of pulverized coal leads to more intense Table 3. PM1/PM2.5, PM1/PM10, and PM2.5/PM10 of Particulate Matter Released from the Combustion of Pulverized Coal with Different Particle Sizes

| temp   | different-sized coal<br>samples | (PM1/<br>PM2.5)% | (PM1/<br>PM10)% | (PM2.5/<br>PM10)% |
|--------|---------------------------------|------------------|-----------------|-------------------|
| 700 °C | <75 $\mu$ m lignite             | 60.8             | 38.4            | 64.7              |
|        | 180–830 $\mu$ m lignite         | 59.4             | 32.6            | 63.6              |
| 800 °C | <75 $\mu$ m lignite             | 58.0             | 32.6            | 62.1              |
|        | 180–830 $\mu$ m lignite         | 52.5             | 33.1            | 59.4              |
| 900 °C | <75 $\mu$ m lignite             | 54.9             | 34.3            | 63.3              |
|        | 180–830 $\mu$ m lignite         | 54.2             | 31.8            | 62.6              |
| 700 °C | <75 $\mu$ m bituminous          | 51.2             | 32.8            | 64.1              |
|        | 180–830 μm<br>bituminous        | 58.8             | 30.9            | 52.5              |
| 800 °C | <75 $\mu$ m bituminous          | 46.1             | 33.3            | 72.2              |
|        | 180–830 μm<br>bituminous        | 58.4             | 29.8            | 51.0              |
| 900 °C | <75 $\mu$ m bituminous          | 49.7             | 36.2            | 72.9              |
|        | 180–830 μm<br>bituminous        | 46.2             | 30.0            | 56.2              |
| 700 °C | <75 $\mu$ m anthracite          | 60.2             | 38.4            | 63.8              |
|        | 180–830 $\mu$ m anthracite      | 56.6             | 37.2            | 65.6              |
| 800 °C | <75 $\mu$ m anthracite          | 57.2             | 43.1            | 75.5              |
|        | 180–830 $\mu$ m anthracite      | 50.8             | 31.9            | 62.7              |
| 900 °C | <75 $\mu$ m anthracite          | 52.4             | 38.8            | 74.2              |
|        | 180–830 $\mu$ m anthracite      | 52.3             | 35.9            | 68.5              |

combustion. As a result, the evaporation rate of inorganics in pulverized coal accelerates and more submicron particulate matter (PM1) is produced. On the other hand, under the same combustion conditions, the particle size range of particulate matter produced by small particle-sized pulverized coal correspondingly narrows. Under the combined action of these two conditions, the values of PM1/PM2.5, PM1/PM10, and PM2.5/PM10 of lignite, bituminous coal, and anthracite increase with the decrease of combusted coal size.

It is well-known that the smaller the particle size of pulverized coal is, the more intense the combustion becomes, the less the loss of incomplete combustion is, and the higher the combustion efficiency of the fluidized bed boiler is. However, this study reveals that the amounts of both submicron and micron particulate matter released from the combustion of finer pulverized coal grow significantly. Such particulate matter is often difficult to be captured and purified due to their small particle size. Moreover, their larger specific surface area can absorb more heavy metal elements, causing greater harm to the human body and the environment.<sup>35,36</sup> Therefore, when choosing the particle size of pulverized coal for the fluidized bed boilers, both economic benefits and environmental hazards should be considered, so as to seek the optimal pulverized-coal particle size.

#### 4. CONCLUSIONS

 Under the same mass, the combustion of bituminous coal releases the most submicron to micron particulate matter, followed by those of lignite and anthracite. The emission factors of PM1, PM2.5, and PM10 of bituminous coal are 2.55-4.08, 2.68-3.99, and 2.74-4.23 times those of anthracite, and the emission factors of lignite are 1.27-2.60, 1.38-2.42, and 1.67-2.54 times those of anthracite.

- (2) Particulate matter with different particle sizes released from the combustion of lignite and bituminous coal presents a bimodal distribution, with the two peak values occurring in the ranges of 0.1–0.18 and 3.2–10  $\mu$ m, respectively. In addition, with the rise of burning temperature and the decrease of particle size of pulverized coal, the first peak value goes down and the second peak value shifts to a small particle size range.
- (3) With the rise of burning temperature, the formation amounts of PM1, PM2.5 of lignite and PM1 of bituminous coal first decrease and then increase, while those of PM10 of lignite, PM2.5 and PM10 of bituminous coal, and PM1, PM2.5 and PM10 of anthracite increase. The values of PM1/PM2.5, PM1/PM10, and PM2.5/ PM10 in particulate matter released by the combustion of lignite, bituminous coal, and anthracite all drop with the rise of burning temperature.
- (4) A smaller particle size of pulverized coal corresponds to the generation of more PM1, PM2.5, and PM10. Additionally, PM1/PM2.5, PM1/PM10, and PM2.5/ PM10 for lignite, bituminous coal and anthracite all go up with the decrease of particle size of pulverized coal.
- (5) During the combustion process of either lignite, bituminous coal, or anthracite, the released PM1 accounts for half and one-third of the PM2.5 and PM10, respectively, and the released PM2.5 accounts for about half of the PM10.

#### AUTHOR INFORMATION

#### **Corresponding Authors**

- Wanxing Ren Key Laboratory of Coal Methane and Fire Control, Ministry of Education, National Professional Laboratory for Fundamental Research of Mine Gas and Dust Control Technology, School of Safety Engineering, and School of Safety Engineering, China University of Mining and Technology, Xuzhou, Jiangsu 221116, China; ⊙ orcid.org/ 0000-0001-8231-2822; Phone: +86-13775883564; Email: rwxcumt@cumt.edu.cn
- Xinjian He Key Laboratory of Coal Methane and Fire Control, Ministry of Education, National Professional Laboratory for Fundamental Research of Mine Gas and Dust Control Technology, School of Safety Engineering, and School of Safety Engineering, China University of Mining and Technology, Xuzhou, Jiangsu 221116, China; Phone: +86-15206264911; Email: xinjian.he@cumt.edu.cn

#### Authors

- Jintuo Zhu Key Laboratory of Coal Methane and Fire Control, Ministry of Education, National Professional Laboratory for Fundamental Research of Mine Gas and Dust Control Technology, School of Safety Engineering, and School of Safety Engineering, China University of Mining and Technology, Xuzhou, Jiangsu 221116, China
- Haisong Sun Key Laboratory of Coal Methane and Fire Control, Ministry of Education, National Professional Laboratory for Fundamental Research of Mine Gas and Dust Control Technology, School of Safety Engineering, and School of Safety Engineering, China University of Mining and Technology, Xuzhou, Jiangsu 221116, China
- Liang Wang Key Laboratory of Coal Methane and Fire Control, Ministry of Education, National Professional Laboratory for Fundamental Research of Mine Gas and Dust Control Technology, School of Safety Engineering, and School

of Safety Engineering, China University of Mining and Technology, Xuzhou, Jiangsu 221116, China; © orcid.org/ 0000-0001-6815-9737

- Yongliang Yang Key Laboratory of Coal Methane and Fire Control, Ministry of Education, National Professional Laboratory for Fundamental Research of Mine Gas and Dust Control Technology, School of Safety Engineering, and School of Safety Engineering, China University of Mining and Technology, Xuzhou, Jiangsu 221116, China; © orcid.org/ 0000-0003-0293-1194
- Guoqing Shi Key Laboratory of Coal Methane and Fire Control, Ministry of Education, National Professional Laboratory for Fundamental Research of Mine Gas and Dust Control Technology, School of Safety Engineering, and School of Safety Engineering, China University of Mining and Technology, Xuzhou, Jiangsu 221116, China

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.2c05080

#### Notes

The authors declare no competing financial interest.

#### ACKNOWLEDGMENTS

The authors gratefully acknowledge financial support from the National Natural Science Foundation of China (Nos. 51904291, 52174222, 51674252 and 51974300), the Basic Research Program of Jiangsu Province (No. BK20190638), the Project funded by China Postdoctoral Science Foundation (No. 2020M681781), and the Jiangsu Planned Projects for Postdoctoral Research Funds (No. 2020Z076).

#### REFERENCES

(1) IQAir. 2020 World air quality report, https://www.iqair.cn/cn/ world-air-quality-report; 2020 [accessed 21 March 2021].

(2) McNutt, M.; Dzau, V. Academies' call to action: Air pollution threatens global health. *Ann. Glob. Health.* **2019**, *85* (1), 145–145.

(3) O'Connor, A. The invisible killer: The rising global threat of air pollution - and how we can fight back. *Lancet Respir. Med.* **2019**, *7*, 17–18.

(4) Huang, R.; et al. High secondary aerosol contribution to particulate pollution during haze events in China. *Nature* **2014**, *514* (7521), 218–222.

(5) Zhou, R.; Yan, C.; Cui, M. Research status and prospects on source apportionment of atmospheric fine particulate matter in Shandong Province. *China Environ. Sci.* **2021**, *41*, 3029–42.

(6) National Bureau of Statistics of China. *Statistical Yearbook*; China Statistics Press, 2018; Vol. 2018, pp 5–7.

(7) Hu, X.; Zhang, Y.; Ding, Z.; et al. Bioaccessibility and health risk of arsenic and heavy metals (Cd, Co, Cr, Cu, Ni, Pb, Zn and Mn) in TSP and PM2.5 in Nanjing. *Atmos. Environ.* **2012**, *57*, 146–152.

(8) Wu, H.; Pan, D.; Jiang, Y.; et al. Improving the removal of fine particles from desulfurized flue gas by adding humid air. *Fuel.* **2016**, *184*, 153–161.

(9) Wang, C.; Liu, X.; Li, D.; et al. Measurement of particulate matter and trace elements from a coal-fired power plant with electrostatic precipitators equipped the low temperature economizer. *Proc. Combust. Inst.* **2015**, 35 (3), 2793–2800.

(10) Geng, G.; Zheng, Y.; Zhang, Q.; et al. Drivers of PM2.5 air pollution deaths in China 2002–2017. *Nat. Geosci.* **2021**, *14* (9), 645–650.

(11) Wu, X.; Braun, D.; Schwartz, J.; et al. Evaluating the impact of long-term exposure to fine particulate matter on mortality among the elderly. *Sci. Adv.* **2020**, *6* (29), 5692–5701.

(12) Shostya, A. Ambient air pollution in China: Predicting a turning point. *Int. Adv. Econ. Res.* **2016**, *22* (3), 295–307.

(13) Dong, G. Ambient air pollution in China. *Respirology.* **2019**, *24*, 626–627.

(14) Yue, G.; Lv, J.; Xu, P.; Li, J. The Up-To-Date Development and Future of Circulating Fluidized Bed Combustion Technology. *Electr. Power.* **2016**, *01*, 1–13.

(15) Wu, C.; Zhan, B.; Hong, Z.; et al. Hydration behavior of circulating fluidized bed fly ash (CFBFA) as a cementitious binder. *Constr. Build. Mater.* **2022**, 314, 125625.

(16) Lv, J.; Li, D. Emission features of primary particulate matters after different pulverized coals combustion. *J. Combust. Sci. Technol.* **2006**, *12* (06), 514–518.

(17) Lv, J.; Li, D. Study on primary pm features influenced by pulverized coal combustion at different burning temperature. *Proc. Cess.* **2007**, 27 (20), 24–29.

(18) Wen, C.; Gao, X.; Yu, Y.; et al. Emission of inorganic PM10 from included mineral matter during the combustion of pulverized coals of various ranks. *Fuel* **2015**, *140*, 526–530.

(19) Fix, G.; Seames, W.; Mann, M.; et al. The effect of combustion temperature on coal ash fine-fragmentation mode formation mechanisms. *Fuel* **2013**, *113* (6), 140–147.

(20) Ninomiya, Y.; Zhang, L.; Sato, A.; Dong, Z. Influence of coal particle size on particulate matter emission and its chemical species produced during coal combustion. *Fuel Process. Technol.* **2004**, *85* (8–10), 1065–1088.

(21) Zhang, L.; Jia, L.; Wang, Y. Effects of co-firing of municipal sludge and bituminous coal washery tailing on particulate matter emission characteristics under fluidized bed conditions. *J. China Coal Soc.* **2021**, 1–10.

(22) Wang, Y.; Qiu, X.; Niu, X.; et al. Particulate matter formation mechanism in pressurized fluidized bed combustion of various solid fuels. *Journal of the Energy Institute J. Energy Inst.* **2022**, *105*, 167–175.

(23) Wang, Q.; Xu, Z.; Liu, Y. Test study on the influence of coal particle size upon ash-slag formation in cfb boilers. *Therm. Power Gener.* **2011**, *10*, 17–20.

(24) China Coal Industry Association. *GB/T212-2008 Industrial* analysis methods for coal; China Standard Press, 2008; pp 1–15.

(25) Roy, B.; Bhattacharya, S. Oxy-fuel fluidized bed combustion using Victorian brown coal: An experimental investigation. *Fuel Process. Technol.* **2014**, *117*, 23–29.

(26) Roy, B.; Bhattacharya, S. Release behavior of Hg, Se, Cr and as during oxy-fuel combustion using Loy Yang brown coal in a bench-scale fluidized bed unit. *Powder Technol.* **2016**, *302*, 328–332.

(27) Sarofim, A. F.; Howard, J. B.; Padia, A. S. The physical transformation of the mineral matter in pulverized coal under simulated combustion conditions. *Combust. Sci. Technol.* **1977**, *16* (3–6), 187–204.

(28) Huang, Q.; Li, S.; Li, G.; et al. Mechanisms on the size partitioning of sodium in particulate matter from pulverized coal combustion. *Combust. Flame* **2017**, *182*, 313–323.

(29) Naydenova, I.; Sandov, O.; Wesenauer, F.; et al. Pollutants formation during single particle combustion of biomass under fluidized bed conditions: An experimental study. *Fuel.* **2020**, *278*, 117958.

(30) Hayashi, J. I.; Takahashi, H.; Doi, S. Reactions in Brown Coal Pyrolysis Responsible for Heating Rate Effect on Tar Yield. *Energy Fuels* **2000**, *14* (2), 400–408.

(31) Huang, J.; Xu, M.; Yu, D. Research on thermal stress breaking during coal combustion. *J. Huazhong Univ. Sci. Technol.* **2004**, 32 (05), 78–80.

(32) Wen, C.; Xu, M.; Wang, J. Formation characteristic of particulate matters during coal and char combustion. *J. Eng. Thermophys.* **2011**, 32 (05), 879–882.

(33) Zhang, L.; Ninomiya, Y.; Yamashita, T. Formation of submicron particulate matter (PMI) during coal combustion and influence of reaction temperature. *Fuel.* **2006**, 85 (10/11), 1446–1457.

(34) Liu, X.; Xu, M.; Yu, D. Study on the influence of coal granule size on emission of inhalable particles during combustion. *Power Eng.* **2005**, 25 (04), 593–598.

(35) Yao, S.; Cheng, S.; et al. Effect of wet flue gas desulfurization (WFGD) on fine particle (PM2.5) emission from coal-fired boilers. *J. Environ. Sci. (China)* **2019**, *77*, 32–42.

(36) Liu, X.; Xu, Y.; Zeng, X.; et al. Field Measurements on the Emission and Removal of PM2.5 from Coal-Fired Power Stations: 1. Case Study for a 1000 MW Ultrasupercritical Utility Boiler. *Energy Fuels* **2016**, 30 (8), 6547–6554.