Synergistic Emission Reduction of Particulate Pollutants in Coal-fired Power Plants Using Ultra-low Emission Technology

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ABSTRACT

The total particulate matter (TPM) is a crucial indicator when evaluating flue gas emissions from coal-fired units. TPM contains solid and liquid contaminants and condensable particulate matter (CPM), which is mainly composed of sulfate and various anions, cations, and metal ions. This study selected three typical large-capacity coal-fired power plants in Shanghai retrofitted with ultra-low emission technology and monitored their emissions of PM$_{2.5}$, SO$_x$, and CPM during power generation. The results showed that the plants achieved comprehensive removal rates of 99.689–99.878% and 86.99–92.92% for PM$_{2.5}$ and SO$_x$, resulting in emitted concentrations of approximately 0.99–1.79 and 1.91–2.50 mg m$^{-3}$, respectively, which are considerably lower than those associated with the traditional flue gas process. Additionally, a significant decrease in the emitted filterable particulate matter (FPM) caused a simultaneous decrease in CPM. After being equipped with the ultra-low emission technology, the units displayed a 76% reduction in the emitted FPM concentration and a smaller FPM/CPM ratio (1:2 vs. 1:1). The emitted TPM (the sum of CPM and FPM) was reduced by 82%, with an average measured concentration of 7.36 ± 3.56 mg m$^{-3}$. The values we obtained are representative and provide basic data for environmental assessment and local environmental policy formulation.

Keywords: Ultra-low emission; Total particulate matter; Condensable particulate matter; PM$_{2.5}$; SO$_x$.

INTRODUCTION

China is a large energy-consuming country. In 2017, China’s total energy consumption was 4.49 billion tons of standard coal. Coal consumption accounted for 60.4% of the total primary energy (Han et al., 2018). Coal-fired power generation is crucial in China’s power industry and is expected to remain the primary source of power in the foreseeable future.

Coal combustion, being one of the main sources of air pollution in China, considerably affects the environment (Chen et al., 2019). According to the requirements of the standard GB13223-2011, “Air Pollutant Emission Standards for Thermal Power Plants,” the emission concentrations of particulate matter (PM), sulfur dioxide (SO$_2$), and NO$_x$ must be lower than the standard limits of 20, 50, and 100 mg m$^{-3}$, respectively. Therefore, all power plants in China had been equipped with flue gas denitrification (FGD), dust removal, and desulfurization systems by 2013. In the next 5–6 years, the development of ultra-low emission coal-fired units have gone through technical research, the demonstration of vital equipment (wet electrostatic precipitator [ESP], single-tower high-efficiency desulfurization and dust-removal integrated technology, among others), engineering demonstration, and large-scale promotion and application at the national level, especially in the central and eastern regions. Ultra-low emissions values of PM, SO$_x$, and NO$_x$ (5, 35, and 50 mg m$^{-3}$, respectively) have been realized from flue gas. Despite an increase in the unit capacity, a considerable reduction has been achieved in the pollutant emission levels in coal-fired power generation. The usage of ultra-low-pollutant-emission technology has been extended to steel, cement, and other industries. Chinese ultra-low-pollutant-emission technologies have become the leading environmental protection technology in the world.

The ultra-low emission retrofit for coal-fired units has reduced the effect of PM, SO$_x$, NO$_x$ on the environment (Zhao et al., 2015). The synergistic control effect of this technology promotes the considerable reduction of sulfur trioxide (SO$_3$), fine particulate matter (PM$_{2.5}$), condensable...
particulate matter (CPM), heavy metals, etc. This improves the ambient air quality, especially reducing the occurrence of smog, which is the focus of public scrutiny (Li et al., 2019a; Yang et al., 2019).

In this paper, the emission reduction effects of primary pollutants, which could form fine particles such as SO$_2$, PM$_{2.5}$, and CPM, were introduced. The effects and environmental benefits of the ultra-low emission units were discussed based on the total particulate matter (TPM) emission.

**TPM FROM FLUE GAS**

**TPM**

U.S. EPA Method 202 (U.S. EPA, 2010) proposed a combined test method for FPM and CPM. In this method, TPM is the sum of FPM and CPM, and the test data include all particulate pollutants from coal-fired flue gas (Fig. 2) (Pei, 2010). FPM consists of pollutants in particulate and liquid states, and CPM consists of gaseous and vapor pollutants that easily condense into nuclei in atmospheric environments.

TPM is a crucial indicator of environmental quality. TPM contains previously unaccounted CPM in the current environmental monitoring system, and its pollutant component covers the majority of flue gas emissions. In addition to the investigation of the pollution reduction effects of CPM, SO$_2$, PM$_{2.5}$ in various facilities, boiler flue gas treatment facilities are crucial to the analysis of emission reduction and factors influencing TPM.

**CPM**

In China, flue gas emissions of PM, SO$_2$, NO$_x$, and Hg are monitored routinely. Some low-concentration pollutants, such as SO$_2$, PM$_{2.5}$, ammonia (NH$_3$), nitrates, volatile organic compounds, and heavy metals are generally neglected. However, these low-concentration pollutants also affect the environmental quality (Wang et al., 2016; Li et al., 2019b). Some pollutants are initially in the gaseous state in flue gas. Because of the decrease in the temperature and change in the pressure, the gaseous pollutants escaping from chimneys rapidly condense into small solid or liquid particles. These particles are known as condensable particulate matter (Li et al., 2019c). CPM contains sulfate ion (SO$_2^{2-}$), nitrate ion (NO$_3^-$), nitrate ion (NO$_2^-$), chloride ion (Cl$^-$), ammonia ion (NH$_4^+$) and Ca$^{2+}$, Mg$^{2+}$, Fe$^{3+}$, Na$^+$, K$^+$, and heavy metal ions such as Cu, Mn, Hg, As, Cd. Among them, SO$_2^{2-}$ proportion (Wang et al., 2011) is the highest (61.50–68.86%), and the metal element content is the lowest (1%). The analysis showed that SO$_2^{2-}$ is derived from SO$_2$/H$_2$SO$_4$ aerosol in flue gas. The current environmental monitoring system in China does not monitor these pollutants. According to the monitoring results of other countries, CPM emitted by fixed sources is equal to or higher than that of PM currently monitored in China. Therefore, CPM should be monitored.

U.S. EPA Method 202 defines CPM as “a vapor at stack conditions, that condenses and/or reacts upon cooling and dilution in the ambient air to form solid or liquid PM immediately after discharge from the stack, and is a component of primary PM.” That is, except traditional emission standards set for PM equalled to FPM defined in EPA Method 202, CPM is also present in flue gas existed usually in the condensed form. The aerodynamic diameter of CPM is less than 1 µm, and CPM is harmful both to the human body and environment. Therefore, PM emitted by coal-fired flue gas is higher than the PM concentration currently monitored.

The monitoring results showed that CPM contributes considerably to PM$_{10}$ in ambient air. Tests by Corio and Sherwell (2000) showed that the average concentration of CPM in flue gas from coal-fired boilers accounts for 76% of the total PM$_{10}$ (by mass). The inductive analysis of 18 coal-fired units in the United States showed that CPM accounted for an average of 49% (from 12% to 92%) of TPM, and FPM accounted for 51% (from 8% to 88%). It indicated that CPM emissions from coal-fired flue gas are equivalent to FPM emissions. Because of the generation mechanism, CPM consists of fine particles and has a larger share in PM$_{10}$ and PM$_{2.5}$. Pei (2000) showed that the average emission concentration of CPM from three coal-fired units in Shanghai (without the ultra-low emission retrofit) was approximately 41.8 ± 12.3 mg m$^{-3}$, and the average concentrations of FPM and CPM were 20.6 ± 10.0 and 21.2 ± 3.5 mg m$^{-3}$, respectively. CPM in TPM was approximately 50.7% (41.4–58.7%). These test results were similar to those reported in the United
CHARACTERISTICS OF PM REDUCTION

TYPICAL ULTRA-LOW EMISSION UNIT AND CHARACTERISTICS OF PM REDUCTION

Ultra-low Emission Process
The research of ultra-low emission technology for coal-fired units in Shanghai was carried out earlier in China. The performance of ultra-low emission technology in Shanghai was evaluated on the 1000 MW unit of Shanghai Shangdian Power Generation Co., Ltd., which was equipped with wet electrostatic precipitator first used in Chinese power stations. This precipitator was used for the study after approval from the National Development and Reform Commission. The resultant ultra-low emission process regional characteristics (shown in Fig. 3) and its technical features are as follows: 1) Cascade reduction design: PM is removed using the electrostatic precipitator and absorption tower, NOx is removed through low-nitrogen combustion in a furnace and high-efficiency selective catalytic reduction (SCR) flue gas denitrification, and SO2 is removed using a high-efficiency absorption tower. 2) Development and application of integrated absorption tower technology of high-efficiency desulfurization and dust removal: The absorption tower is equipped with a tray and wall ring, a three-stage high-efficiency defogging droplet reduction mechanism, and an optimized flow field. Because of the efficient PM-removal characteristics of the absorption towers, wet electrostatic precipitators are no longer used for end purification. 3) Full consideration of the synergistic reduction to multiple pollutants, mainly nonconventional pollutants such as SO3, PM2.5, CPM, and Hg: Higher emission reduction can be obtained using low-temperature electrostatic precipitators, high-efficiency integrated absorption towers, flue gas condensation, and low-temperature reheating, among other techniques. 4) Dry flue gas emissions do not harm the environment.

Test Objects and Test Methods
PM (including PM2.5, CPM, and SO3 in flue gas were monitored in three typical ultra-low emission units in Shanghai (Table 1) to calculate TPM.

The dust concentration in the electrostatic precipitator inlet was tested using Pitot parallel sampling gravimetric method (GB16157). The sample was measured using Malvern laser particle-size analyzer to calculate the PM2.5 concentration. PM from the outlets of electrostatic precipitator and absorption tower was tested using ISO 12141:2002 (determination of mass concentration of particulate matter [dust] at low concentrations—manual gravimetric method), PM2.5 by using EPA Method 201A (determination of PM10 and PM2.5 emissions from stationary sources). EPA Method 202 (dry impinger method) was used for determining condensable particulate emissions from stationary sources. The SO3 test uses EPA Method 8 (determination of sulfuric acid and sulfur dioxide emissions from stationary sources).

FPM
PM emitted by coal-fired flue gas contains dust and desulfurized gypsum, in which particle size is usually less than 10 µm. When the dust is carried with flue gas through a low-temperature electrostatic precipitator, approximately 99.9% of the dust accumulates. After passing flue gas through the high-efficiency desulfurization and dust-removal integrated absorption tower, a dust-removal efficiency of more than 75% can be achieved, and a PM emission concentration lower than 5 mg m⁻³, which contains desulfurized gypsum, is formed because of the escape of droplets.

Before passing through the electrostatic precipitator, the PM2.5 concentration in dust is relatively low. After cascade dust removal, the removal rate of the large particles is higher than that of the fine particles, and the final PM2.5 concentration in PM increases considerably (Jaworek et al., 2018). The test results of PM and PM2.5 for electrostatic precipitators and desulfurization absorption towers in the three ultra-low emission units are listed in Table 2. Fig. 4 illustrates the results of the PM particle-size distribution at the entrance of the electrostatic precipitator.

The test results showed that the PM2.5 concentration in PM at the inlet of the electrostatic precipitator varied between
The PM$_{2.5}$ content was affected by factors such as the coal quality, coal preparation, combustion condition, SCR catalyst and its flow field, SO$_3$ content, and escaped ammonia concentration. For fine pulverized coal, the SCR denitrification system was installed, and the concentrations of SO$_3$ and escaped ammonia were high, and the PM$_{2.5}$ content increased.

Using an electrostatic precipitator, dust and PM$_{2.5}$ removal efficiencies in the range of 99.77–99.83% and 99.00–99.53% were achieved for the three units. The removal rates are higher than that of the conventional electrostatic precipitator because the removal mechanism of the particles with different particle size is different in the electric field. Larger particles ($\geq 2.0$ $\mu$m) were removed because of the electric field charge, whereas fine particles ($\leq 0.1$ $\mu$m) were mainly removed because of the diffusion charge, and the particles between 0.1 and 2.0 $\mu$m were controlled using an electric field charge and diffusion charge mixing control zone. Therefore, the charging effect was inadequate, and the removal efficiency was relatively low in the conventional electrostatic precipitator.

After passing through the desulfurization absorption tower, subsequent flue gas treatment facilities and flue gas reheating (Table 1), 79.12–84.50% and 68.82–74.02% removal rates were obtained for PM and PM$_{2.5}$. Tray synergism, high-efficiency defogging, and flow field optimization measures are implemented in the high-efficiency desulfurization and dust-removing integrated absorption tower. PM is subjected to high-efficiency washing of the liquid-holding layer of the tray in the tower and sprays with high liquid/gas ratios. The dust-removal efficiency of this method is considerably higher than that of the traditional absorption tower. The PM$_{2.5}$ removal effect was especially enhanced. In addition, under high-efficiency defogging and optimized flow fields,

**Fig. 3.** Schematic of typical process for ultra-low-emission of coal-fired units.

**Table 1.** Ultra-low-emission process of coal-fired units A, B, and C.

| Unit | Unit capacity | Ultra-low emission process |
|------|--------------|----------------------------|
| A    | 1000 MW      | low-nitrogen combustion → SCR denitrification → low-temperature electrostatic precipitator → desulfurization and dedust integrated tower → horizontal wet electric dust removal → flue gas reheater |
| B    | 1000 MW      | Low-nitrogen combustion → SCR denitrification → electric dust removal → high-efficiency desulfurization → flue gas condensation and water removal → flue gas reheater (low-temperature) |
| C    | 660 MW       | Low nitrogen combustion → SCR denitrification → low-temperature electrostatic precipitator → desulfurization and dedust integrated tower → horizontal mist remover → flue gas reheater |

**Table 2.** Removal rate of PM and PM$_{2.5}$ for ESP and FGD.

| Unit | Inlet PM$_{2.5}$ concentration (mg m$^{-3}$) | Outlet PM$_{2.5}$ concentration (mg m$^{-3}$) | PM$_{2.5}$ removal efficiency (%) | Inlet FPM concentration (mg m$^{-3}$) | Outlet FPM concentration (mg m$^{-3}$) | FPM removal efficiency (%) |
|------|---------------------------------------------|---------------------------------------------|----------------------------------|----------------------------------------|----------------------------------------|----------------------------|
| ESP  | A                                           | 811.90                                     | 3.81                             | 99.53                                  | 6526.90                                | 11.10                      | 99.83                      |
|      | B                                           | 1167.50                                    | 6.06                             | 99.48                                  | 6482.80                                | 14.90                      | 99.77                      |
|      | C                                           | 418.40                                     | 4.17                             | 99.00                                  | 7663.30                                | 12.26                      | 99.84                      |
| FGD  | A                                           | 3.81                                       | 0.99                             | 74.02                                  | 11.10                                  | 1.72                       | 84.50                      |
|      | B                                           | 6.06                                       | 1.79                             | 70.46                                  | 14.90                                  | 3.10                       | 79.19                      |
|      | C                                           | 4.17                                       | 1.30                             | 68.82                                  | 12.26                                  | 2.56                       | 79.12                      |
a. PM$_{2.5}$ accounted for 12.4% in unit A

b. PM$_{2.5}$ accounted for 18.0% in unit B

c. PM$_{2.5}$ accounted for 5.5% in unit C

Fig. 4. PM particle-size distribution at the inlet of ESP A, B, and C.

the concentration of the escaped droplet reduced from approximately 75 to $\leq$ 20 mg m$^{-3}$, which markedly reduced the escape rate of desulfurization gypsum, and an ultra-low PM emission value of 5 mg m$^{-3}$ was obtained.

From the experimental results, the removal rate of PM$_{2.5}$ using the traditional absorption tower is speculated to be higher than that obtained using the modified method. The removal rate of PM$_{2.5}$ is affected by the escape of the droplets, which reduces the PM$_{2.5}$ removal rate. Experimental results showed that PM$_{2.5}$ concentration exhibits a trend of increase. This is caused by the high escape rate of the fog droplets.

The analysis of the removal effect of PM with the ultra-low emission system revealed that the PM concentrations from the chimney in Units A, B, and C were 1.72, 3.10, and 2.56 mg m$^{-3}$ and total dust-removal rates were 99.97%, 99.95%, and 99.97%, respectively. The proportion of PM$_{2.5}$ in PM was 50.7–57.7%. The corresponding removal rates of PM$_{2.5}$ were 99.878%, 99.847%, and 99.689%, respectively. Thus, the removal rate of the ultra-low emission system was considerably higher than the conventional-unit PM$_{2.5}$ removal rate (95.68–98.47%) (Tao et al., 2018). The emission reduction effect of PM was obviously, and the higher emission-reduction efficiency was achieved.

**Sulfur Trioxide**

Sulfur trioxide is the main component of CPM (approximately 60% or more). The efficient removal of SO$_3$ with ultra-low emission systems reduces both CPM and TPM. SO$_3$ in flue gas is affected by temperature, oxygen content, humidity, and catalytic substances (Zheng et al., 2019). After passing through the flue gas system, SO$_3$ concentration and morphology change. SO$_3$ test data for the process of the ultra-low emission system in Unit A showed that the comprehensive removal rate was as high as 96.0% because of process cascade reduction (Tao et al., 2018).

The sulfur contents of coal for Units A, B, and C were 0.21%, 0.36%, and 0.39%, respectively. The test results of SO$_3$ under full-load operation conditions in the process are depicted in Table 3. Experimental results showed that the concentration of SO$_3$ decreased gradually from the inlet of the electrostatic precipitator to the outlet of the chimney.
The SO\textsubscript{2} removal efficiencies of the electrostatic precipitators for A, B, and C were 54.08\%, 24.41\%, and 66.41\%, respectively. The test results were considerably affected by the operating temperature of the electrostatic precipitator (Table 1). This indicated a tendency of increase in the removal rate with the decrease in the smoke temperature. The measured data from FGD showed that the SO\textsubscript{2} removal rates of the three units were 65.94\%, 68.53\%, and 70.46\%, respectively. Among them, Unit C had one more secondary flue defogger. Thus, the ability of high-efficiency desulfurization absorption tower to reduce SO\textsubscript{2} synergistically is strengthened and the removal rate is relatively stable.

The unit emission data showed that the SO\textsubscript{2} concentration levels of the three units were similar, and the average emission concentration was 2.12 mg m\textsuperscript{-3}. The comprehensive removal rates were 92.92\%, 86.99\%, and 92.38\% respectively, with an average of 90.76\%. Similar to those of the ultra-low-reform unit shown by Tao et al. (2018), the emission concentration was 0.80–3.68 mg m\textsuperscript{-3}, and the average removal rate was 90.7\%.

**CPM**

CPM contains pollutants such as sulfur trioxide, ammonium salts, nitrates, volatile organic compounds, and heavy metals. Efficient step-by-step removal by the ultra-low emission systems considerably reduced most of these pollutants. The PM emissions of some units higher than 660 MW in Shanghai are listed in Table 4. The data in Table 4 shows that the emission FPM was approximately 1.17–3.93 mg m\textsuperscript{-3}, and the average concentration was 2.29 ± 1.20 mg m\textsuperscript{-3}. The CPM concentration was 1.65–8.56 mg m\textsuperscript{-3}, and the average concentration was 5.07 ± 2.62 mg m\textsuperscript{-3}. TPM is the sum of FPM and CPM, and its concentration was between 3.07 and 12.53 mg m\textsuperscript{-3}, and the average concentration was 7.36 ± 3.56 mg m\textsuperscript{-3}. The ratio of CPM to TPM was 53.7–83.3\%, with an average ratio of 67.6 ± 9.43\%.

The CPM emission concentration of ultra-low emission units was twice that of FPM. For the thermal coal quality at Shanghai, the TPM emission concentration of ultra-low emission units was approximately three times that of FPM. After ultra-low reform, the CPM emission concentration decreased by 76\%, and the ratio of FPM to CPM decreased from 1:1 to 1:2. But even so, because of the efficient removal of ultra-low emissions, the actual emission of TPM was lower than the traditional Shanghai power unit (Pei, 2010), and the emission reduction rate was approximately 82\%.

**TPM and Emission Concentration**

Table 4 provides the TPM emissions data of some coal-fired units in Shanghai. The following aspects are crucial: 1) The data provided are based on the emission monitoring data of 660 MW and 1000 MW coal-fired units for Shanghai’s thermal coal quality. The concentration range and average value of these data are representative to some extent. The average value of 7.36 mg m\textsuperscript{-3} can provide basic data for environmental assessment. 2) The test results showed that the TPM concentration of the ultra-low emission unit was approximately three times that of the FPM concentration. The emissions data appeared to be underestimated. Compared with the traditional flue gas treatment process that does not have the ultra-low emission retrofit, TPM was considerably reduced. Compared with the FPM emission concentration before the reform, the emission reduction rate was higher than 60\%. Substantial emission reduction was achieved from coal-fired units. 3) The key to TPM reduction in the ultra-low-scale flue gas process is efficient synergistic removal of PM and unconventional contaminants. Optimizing the ultra-low flue gas process is a crucial task in the post-ultra-low era.

### Table 3. Change of flue gas SO\textsubscript{2} concentration with unit process (mg m\textsuperscript{-3}).

| Unit | ESP inlet | ESP outlet | FGD outlet | WESP outlet | Chimney outlet |
|------|-----------|------------|------------|-------------|----------------|
| A    | 26.98     | 12.39      | 4.22       | 1.99        | 1.91           |
| B    | 19.21     | 14.52      | 4.57       | 2.83\*      | 2.50           |
| C    | 25.60     | 8.60       |            | 2.54**      | 1.95           |

\*refers to the outlet of the condensing dehydrator; **refers to the outlet of the flue demister.

| Unit | FPM concentration (mg m\textsuperscript{-3}) | CPM concentration (mg m\textsuperscript{-3}) | TPM concentration (mg m\textsuperscript{-3}) | CPM/TPM (%) |
|------|---------------------------------------------|---------------------------------------------|---------------------------------------------|--------------|
| A    | 1.38                                        | 6.87                                        | 8.25                                       | 83.3         |
| B    | 1.63                                        | 2.30                                        | 3.93                                       | 58.5         |
| C    | 1.42                                        | 1.65                                        | 3.07                                       | 53.7         |
| D    | 1.17                                        | 3.93                                        | 5.10                                       | 77.1         |
| E    | 1.61                                        | 3.41                                        | 5.02                                       | 67.9         |
| F    | 3.95                                        | 7.18                                        | 11.11                                      | 64.6         |
| G    | 3.21                                        | 6.86                                        | 9.89                                       | 67.5         |
| H    | 3.97                                        | 8.56                                        | 12.53                                      | 68.3         |
| average | 2.29 ± 1.20                              | 5.07 ± 2.56                                | 7.36 ± 3.56                               | 67.6 ± 9.43  |
| Average value of three conventional units (Pei., 2010) | 20.6 ± 10.0 | 21.2 ± 3.5 | 41.8 | 49 |
CONCLUSION

1) PM in coal-fired flue gas contains FPM and CPM, the sum of which equals TPM. Whereas FPM comprises filterable solid and liquid contaminants, CPM consists mainly of SO$_2$ and plus various anions, cations, and metallic ions, and precipitates in the form of condensed nuclei after the discharge of the flue gas. TPM is a vital indicator of flue gas emissions from coal-fired units, which must be considered when evaluating the air quality of the atmospheric environment, including the severity of haze.

2) The PM$_{2.5}$ in FPM strongly affects the air quality. The PM$_{2.5}$ removal rates of the tested power plants considerably improved after they were equipped with ultra-low emission technology, resulting in emitted concentrations of 0.99–1.79 mg m$^{-3}$, or comprehensive removal rates of 99.689–99.878%. The SO$_2$ in the flue gas, which is the primary source of CPM, decreased to 1.91–2.50 mg m$^{-3}$ in concentration, exhibiting comprehensive removal rates of 86.99–92.92% (averaging 90.76% for the three tested units). Thus, both the PM$_{2.5}$ and the SO$_2$ were significantly reduced.

3) The ultra-low emission retrofit of the tested units noticeably reduced the generated FPM and thus CPM, producing a smaller FPM/CPM ratio (1:2 vs. 1:1). The emitted concentrations of CPM and TPM dropped by 76% and 82%, respectively. The decrease in PM$_{2.5}$ and SO$_2$ also caused a decrease in CPM.

4) The TPM emission values obtained in this study, which investigated typical ultra-low emission units in Shanghai, are representative. Therefore, they provide basic data for environmental assessment and local environmental policy formulation.

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