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Five-year Variations in Air Pollutant Emissions with Ultra-low Emission Retrofits in a 660 MW Coal-fired Power Unit

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ABSTRACT

China has enacted the ultra-low emission (ULE) transform in coal-fired power plants. Various studies have focused on the model simulation of pollutant emission variations on a national scale, while the specific data for a concrete generation unit was still lacking. We deployed a five-year online data collection campaign in a 660 MW unit to investigate the durative emission reductions of dust, NO_x, and SO_2, and increases in pollutant removal efficiencies. The result indicated that a time slag appeared between the ULE execution and meeting discharge standards. Since the ULE implementation in late 2014, the pollutant emissions exhibited a decreasing trend, while it was not until 2016 that the emission amounts reached the ULE standards with the actual emissions of 30.7 ± 4.43, 12.3 ± 3.49, and 1.80 ± 0.425 mg m^{-3} for NO_x, SO_2, and dust, respectively. It was particularly pointed out that emissions increased again in 2017 though they still met the ULE standards, indicating the comprehensive consideration should be taken between the emission reductions and high cost of the ULE policy. What's more, the ULE transformation weaken the correlation between pollutant emissions (in mg m^{-3}) and corresponding removal efficiencies of control devices, evidencing the pollutant emissions received complex impacts from coal components, emission control technologies and devices, and so on. More practical emission factors (EFs) expressed in kg h^{-1} and g MWh^{-1} relative to the ULE were updated and in this study. Unlike EFs expressed in mg m^{-3}, the most decreases of EFs in kg h^{-1} and g MWh^{-1} in 2015 by more than 97.0% compared to 2014 for three air pollutants arisen from reasonable air supply or enhancement in thoroughness of coal combustion.

Keywords: Coal-fired power plant, Ultra-low emission transformation, Emission factor, NO_x, SO_2

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1 INTRODUCTION

Over the last several decades, the developed countries have successfully enacted the regulation of air pollutants (Morino *et al.*, 2018). Consequently, emissions from the combustion sources, such as the coal-fired power plants (CFPPs), industrial factories, and motor vehicles, have decreased obviously over the last two decades (Granier *et al.*, 2011; Qi *et al.*, 2022). However, China have been experiencing frequent heavy air pollution episodes along with the rapid economic development and urbanization (Li *et al.*, 2022, 2023b). As a result, China has enacted the "Clean Air Action" in 2013–2017 to markedly reduce the $PM_{2.5}$ levels on a national scale through emission reductions from main anthropogenic sources (Cheng *et al.*, 2019; Liu *et al.*, 2022).

Chinese electric power industry has been developing rapidly along with the sustained economic growth within recent years. The thermal power plants owned the sharply increased total installed capacity from 3.98×10^7 kW in 1978 to 1.19×10^9 kW in 2019, and the CFPPs contributed more than 90% to the thermal power generation (National Bureau of Statistics, 2019). At the same time, coal-fired power plants consumed more than 50% of coal in 2020 (3.902 billion tons accounting for 54.3% of global total), which has an important impact on social economy, especially on air quality and human health (Liu *et al.*, 2019; Xie *et al.*, 2021). Stringent emission control measures had been enforced in the past decade (Li *et al.*, 2023a). To accommodate the national new emission standard, the ultra-low emission (ULE) retrofit was put to operation in Chinese thermal power plants (Tang *et al.*, 2019). In addition to the existing standard of GB13223-2011, China further issued even tougher emission standards for coal-fired power plants named as the ULE standards on September 12, 2014, covering all the existing and future coal-fired power generators, which are equivalent to those of natural gas-fired units (MEEPRC, 2015). This stricter policy required that 71% of the total installed capacity in 2014 must meet the ULE standards by 2020, and the new power generation units since 2015 must achieve the ULE goal (NDRC, 2014). Consequently, at least 80% of capacity for both preexisting and new power units can attain the ULE goal by 2030. It is undeniable that ULE has led to the additional costs used in monitoring power plants and supporting subsides by government, updating technologies, running pollution control devices, and closing inefficient units and building new units by power plants (Yang *et al.*, 2018b). At the same time, however, the ULE has been expected to bring about some significant co-benefits including air quality improvement, reduction of pollution related diseases, and progress in technologies of emission reduction due to the substantial decreases in pollutants (Wang *et al.*, 2018; Liu *et al.*, 2023; Tang *et al.*, 2023).

The CFPPs release air pollutants such as SO_2, NO_x, PM, and Hg, which has led to a series of atmospheric environmental problems (Shin *et al.*, 2022; Zhang *et al.*, 2023). As an important emission source of air pollutants, the CFPPs in China have been accordingly required to limit anthropogenic emissions of SO_2, NO_x, particulate matter (PM), and mercury (Hg) to mitigate environmental impacts (Wu *et al.*, 2023a). Furthermore, expunging the hazard of condensed particulate matter (CPM) was still a challenge for unconventional pollutants except for aforementioned conventional pollutants though the most CFPPs in China have come up to ultra-low emission standards (Morino *et al.*, 2018; Deng *et al.*, 2023; Sheng *et al.*, 2023). At present, various studies were conducted on changes of air pollutant chemistries along with the concrete emission control technologies based on a short-term observation (Han *et al.*, 2021; Tang *et al.*, 2022; Li *et al.*, 2023a; Liu *et al.*, 2023; Tong *et al.*, 2023; Zhang *et al.*, 2023). The ULE has implemented for a long time and obtained some environmental and health benefits, though Zhang *et al.* (2021) indicated that the ULE execution in coal-fired power plants (CFPPs) alone possessed minor benefit, with decreases of SO_2, NO_x, and dust by only 11%, 7%, and 2% of their total amounts in the Yantze River Delta Region. Therefore, a long-term observation campaign was urgently need to examine the ULE effectiveness for future policy optimization and improvement of emission control technologies and facilities (Zhang *et al.*, 2021; Li *et al.*, 2023a; Tong *et al.*, 2023).

2 METHODOLOGY

2.1 Description of the Investigated Power Plant

The investigated power plant is located in Zhejiang Province of China. A total of four supercritical once-through boilers (660 MW for each unit) are included in this plant. An electrostatic precipitator (ESP), a selective catalytic reduction system (SCR), and a wet flue gas desulfurization (WFGD) system were applied to remove dust, NO_x and SO_2, respectively. Before the ultra-low emission transforms in 2014, the designed pollutant concentrations at outlets of pollutant removal facilities were 30, 111, and 90 mg Nm^{-3} for dust, SO_2, and NO_x, respectively, and the actual emissions were 13, 120, and 85 mg Nm^{-3} under operation rates as 100%, 99.2%, and 99.2% of pollution control devices.

2.2 Main Renovation Measures

In 2014, the Zhejiang Provincial Government issued the “Zhejiang Air Pollution Prevention and Control Action Plan” (2013–2017). As a response, the designated emission standards in 2014 for

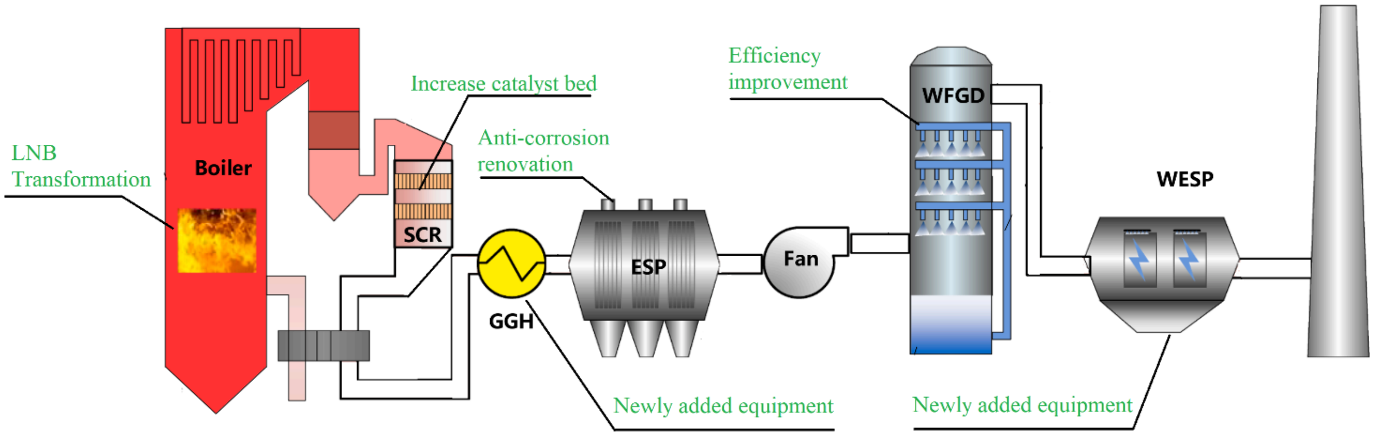

Fig. 1. Transformation measures for the ultra-low emissions (marked by green color).

the coal-fired power plants (CFPPs) were in agreement with those for gas-fired power plants, which were lower than 5, 35, and 50 mg Nm^{-3} for dust, SO_2, and NO_x, under the baseline oxygen content of 6% (Tang *et al.*, 2023). Fig. 1 showed the process flow diagram for air pollutant removal contained in the flue gas (FG), and the specific transformation measures marked by green color. A new catalyst bed on the basis of the original two layers was added in the selective catalytic reduction (SCR) reactor and the honeycomb catalyst layers comprising V_2O_5-TiO_2/WO_3 were equipped. The NO_x is converted to N_2 by the following equations when catalyst existed:

$$4NO + 4NH_3 + O_2 \longrightarrow 4N_2 + 6H_2O \tag{1}$$

$$6NO_2 + 8NH_3 \longrightarrow 7N_2 + 12H_2O \tag{2}$$

$$2NO_2 + 4NH_3 + O_2 \longrightarrow 3N_2 + 6H_2O \tag{3}$$

Furthermore, low nitrogen burner (LNB) reformation included laying additional NO_x nozzles or changing the nozzle height, and adsorption of new burner to ensure the NO_x concentration at the outlet of coal economizer lower than 280 mg Nm^{-3}.

Anti-corrosion treatment was carried out on the original ESP because of the decrease in inlet FG temperature with the newly added gas-gas heater (GGH). For WFGD, an additional slurry circulating pump was installed and three slurry spray layers were replaced by two interacting layers. Consequently, most of SO_2, HF, and HCl can be eliminated in WFGD. Wet WESP was a newly added system after WFGD for the ultra-low emissions (ULE). WESP was consist of two chambers with two electric fields to further eliminate gypsum droplets, soot particles, $PM_{2.5}$, SO_3 microdroplets, and mercury compounds, etc. Another added device was gas-gas heater (GGH), consisting of a FG heater, a FG cooler, and a steam heater. The GGH system enhanced the temperature of outlet FG of WESP by hot FG or steam after the air preheater to make the temperature of inlet FG of chimney higher than its dew point, avoiding the occurrence of white smoke and gypsum rain and improving dust removal efficiency of WESP. In addition, the temperature of inlet of FG of ESP decreased to slightly lower than the dew point to make SO_3 change into H_2SO_4 adsorbed in particle matter. Finally, SO_3 and particles were eliminated simultaneously in the ESP system.

2.3 Data Acquisition

The effectiveness and sustainability of the ultra-low emission system for a power generation unit (660 MW) in a 4 × 660 MW power plant was investigated in this study. China was tightening the emission standards, the emission limits for particle matter (PM), SO_2, and NO_x far decreased from GB13223 of 2003 (50, 400, and 450 mg Nm^{-3}) and GB13223 of 2011 (20, 50, and 100 mg Nm^{-3}) to "Ultra-low emission" of 2014 (5/10, 20, and 50 mg Nm^{-3}) (NDRC, 2014; MEEPRC, 2015) and "Near-zero emission" of 2019 (1, 10, and 20 mg Nm^{-3}). The GB13223 standards were still valid, which went into effect on July 1, 2014 and the emission limits of GB13223 were much lower than other jurisdictions such as the United States (20, 184, and 135 mg Nm^{-3}), and the European Union (30, 200, and 200 mg Nm^{-3}).

The extensive and detailed data about hourly emissions of dust, SO_2, and NO_x during August 1 to December 31, 2013, and January 1 to December 31, 2014, and corresponding data for daily emissions in 2015 and 2016, as well as monthly data in 2017, covering both the before and after the ULE implementation (BULE and AULE), were obtained from the supervisory information system (SIS) of this power plant.

3 RESULT AND DISCUSSION

3.1 Daily and Monthly Variations in Pollutant Emissions and Removal Efficiencies in 2014

The related data was available for totally 365 days in 2014. Fig. 2 showed the daily unit parameters, daily and monthly pollutant emission factors and removal efficiencies in 2014. No significant correlations were found between flue gas temperature/unit loads and NO_x, SO_2, and dust emission factors, which should be attributed to the fluctuations of coal components and unit operation conditions.

The monthly emission factors (in mg m^{-3}) for NO_x, SO_2, and PM were 80.1 ± 5.92, 103 ± 10.1, and 12.3 ± 2.73. The monthly dust and SO_2 removal efficiencies varied from 46.7% to 72.8%, and 91.7% to 92.6%, respectively, and they were averaged at 92.2% and 59.2%. The ULE standards

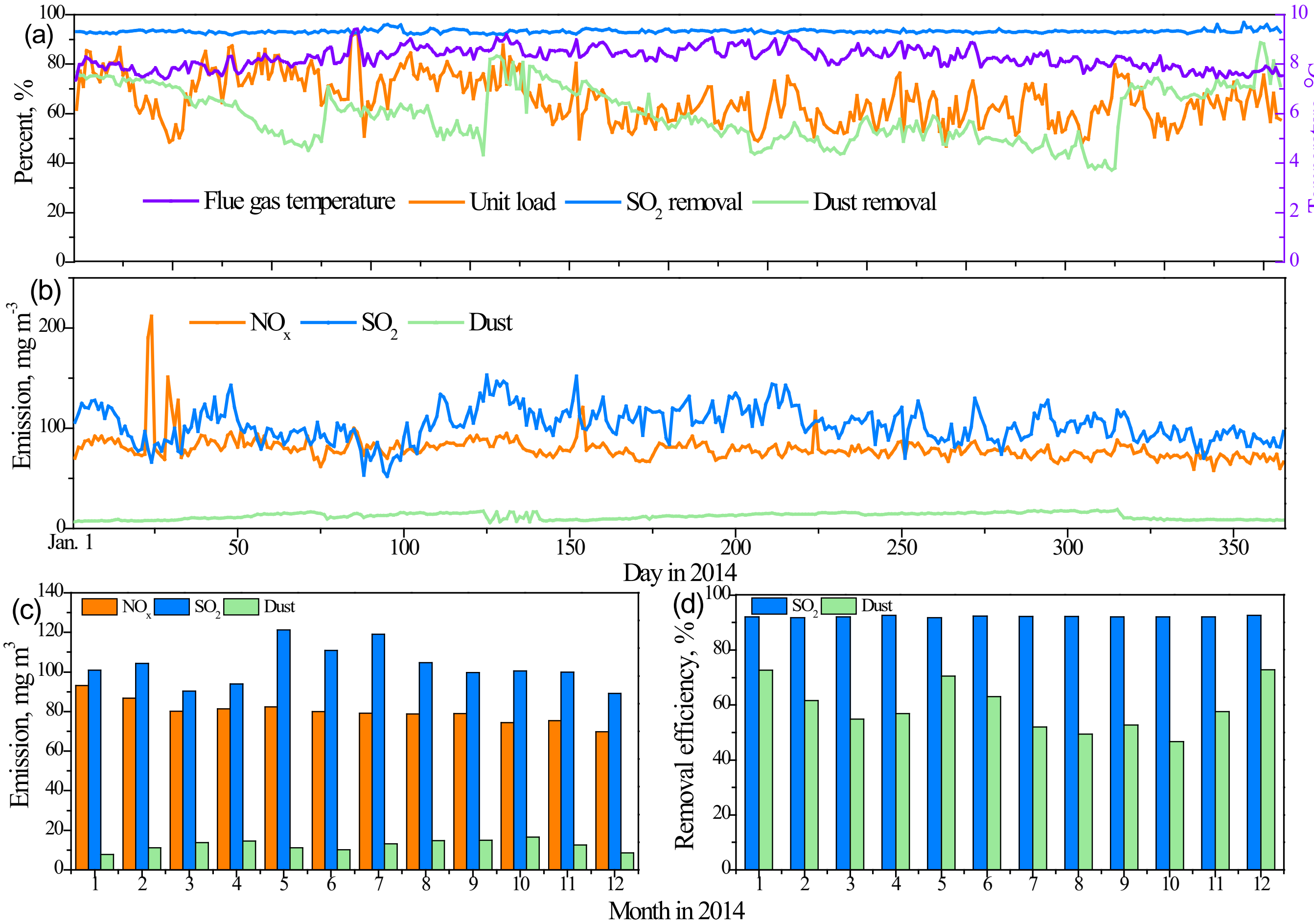

Fig. 2. (a) Daily unit parameters and pollutant removal efficiencies in 2014; (b) daily pollutant emissions in 2014; and monthly averages of (c) pollutant emission factors and (d) removal efficiencies.

were implemented since Dec 2014. In consequence, the lowest emission factors (69.9, 89.2, and 8.55 mg m^{-3}) of NO_x, SO_2, and dust, and the highest removal efficiencies (92.7% and 72.9%) for SO_2 and dust were found in December. At the same time, however, the emissions of NO_x and SO_2 in December were still far beyond the ULE standards (NO_x, 50 mg Nm^{-3}; SO_2, 20 mg Nm^{-3}). The dust emission basically met the ULE emission standard of 5/10 mg Nm^{-3}. What's more, the actual emissions were far away from the "Near-zero emission" (NZE) standards in 2019.

3.2 Daily and Monthly Variations in Pollutant Emissions and Removal Efficiencies in 2015

The online data for totally 173 days from Jan. to Aug. 2015 were available in this study. Daily and monthly emission factors and pollutant removal efficiencies in 2015 were shown in Fig. 3. Monthly data were averaged at 79.9, 91.8, and 10.2 mg m^{-3} for NO_x, SO_2, and dust emissions, which were lower than the corresponding 80.1, 103, and 12.3 mg m^{-3} in 2014. Furthermore, the lowest decrease of 0.249% was found for NO_x, which might be related to the difficulty in improvement of de-NOx efficiency. The newly added slurry circulating pump in WFGD, GGH, and WESP made significant increases in removal efficiencies in dust and SO_2. However, the dust and SO_2 removal rates were relative low in 2015 due to the hysteresis of operating condition adjustment comparing with the device update speed.

3.3 Daily and Monthly Variations in Pollutant Emissions and Removal Efficiencies in 2016

Daily and monthly emission factors and pollutant removal efficiencies in 2016 were shown in

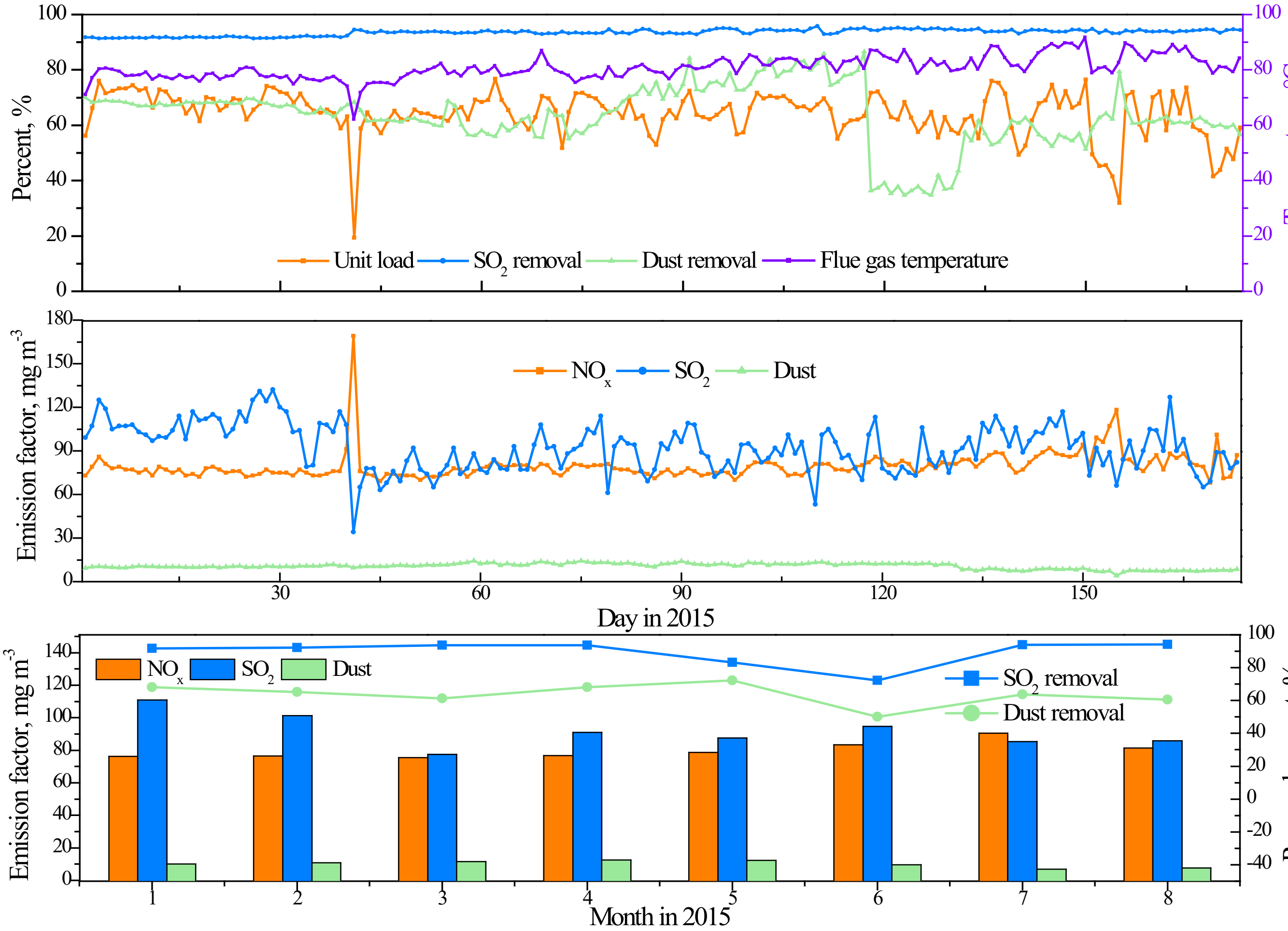

Fig. 3. Daily and monthly emission factors and pollutant removal efficiencies in 2015.

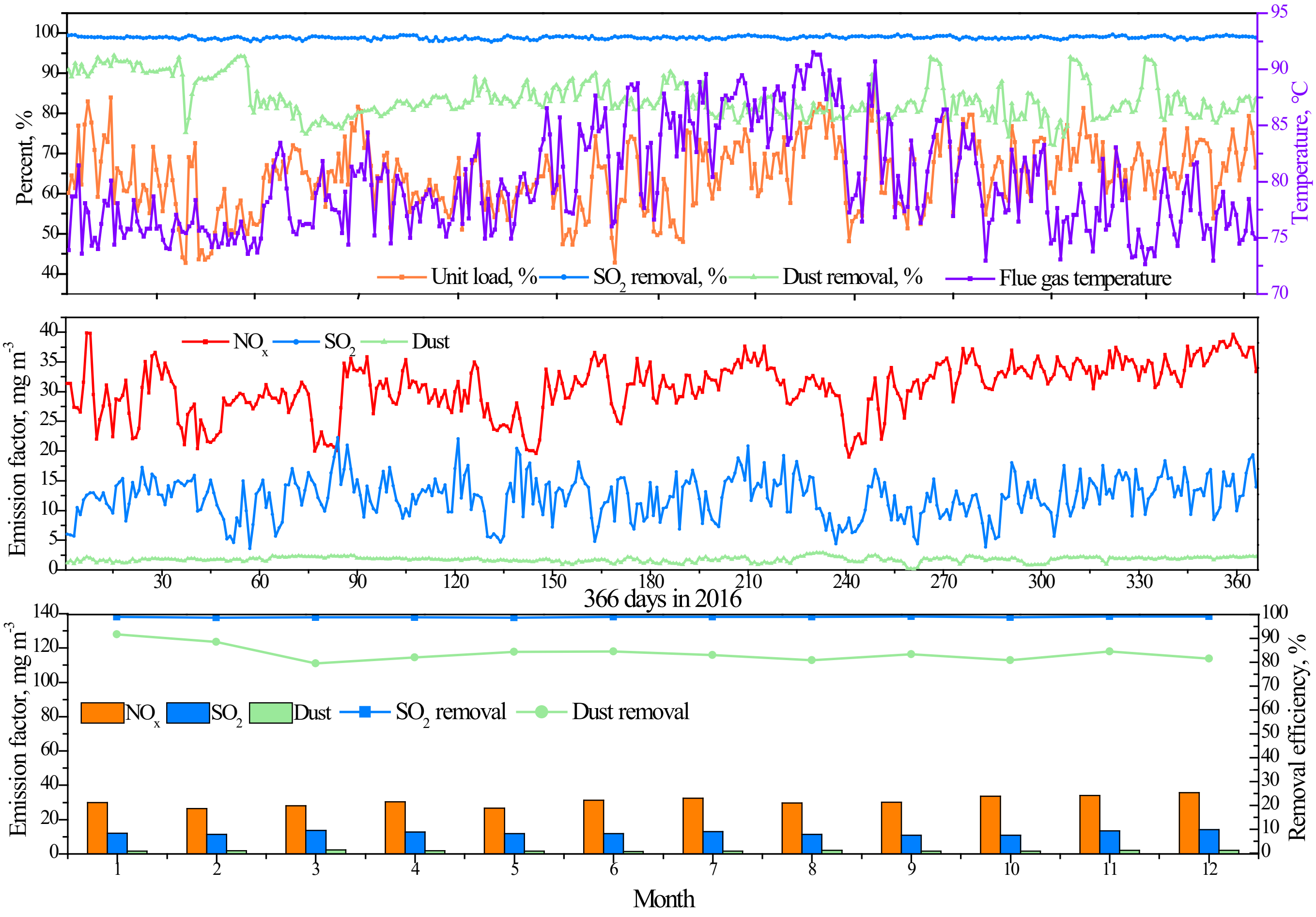

Fig. 4. Daily and monthly emission factors and pollutant removal efficiencies in 2016.

Fig. 4. The daily unit loads and flue gas temperature in 2016 were averaged at 64.4% ± 8.67% and 80.3 ± 4.50°C, respectively. Furthermore, the daily emission factors of NO_x, SO_2, and dust were averaged at 30.7 ± 4.43, 12.3 ± 3.49, and 1.80 ± 0.425 mg m^{-3}, which had achieved the emission goal defined in the ULE standards (NO_x, 50 mg m^{-3}; SO_2, 20 mg m^{-3}; Dust, 5/10 mg m^{-3}), implying that the ULE retrofit played a positive role. At the same time, the monthly average removal efficiencies of SO_2 and dust far increased compared to those in 2014 and 2015, which were as high as 98.9% ± 0.346% and 83.7% ± 4.82%, respectively. The newly added slurry circulating pump was installed and replacement of three slurry spray layers by two interacting layers largely enhanced the de-SO_2 efficiencies and the maximum de-SO_2 rate reached 99.5%. Yang *et al.* (2021) indicated that the highest de-SO_2 efficiency was up to 99.7% for the two-absorber layer WFGD devices from 95 coal-fired power plants that have completed the ULE renovations.

3.4 Inter-annual Variations in Pollutant Emission Factors and Removal Efficiencies

We compared the data in same month among different year in Fig. 5. Also, the fluctuations of annual averages were shown in Fig. 5. The ULE planned to complete the transformation of coal-fired power plants producing 580 million kilowatts of energy by 2020, therefore, the pollutant emission reductions and enhancements of corresponding removal efficiencies of control devices and technology exhibited a lagging trend. NO_x and SO_2 decreased by 44.0% and 16.4% in 2014 compared to those in 2013, indicating that pollutant control in 2014 was mainly focused on NO_x and SO_2. At the same time, however, dust increased from 6.26 mg m^{-3} of 2013 to 12.4 mg m^{-3} of 2014, which might be ascribed to the increased de-SO_2 slurry enhanced Ca containing dust in flue gas inlet the chimney and the decreased removal efficiency of dust. Furthermore, SO_2 and NO_x

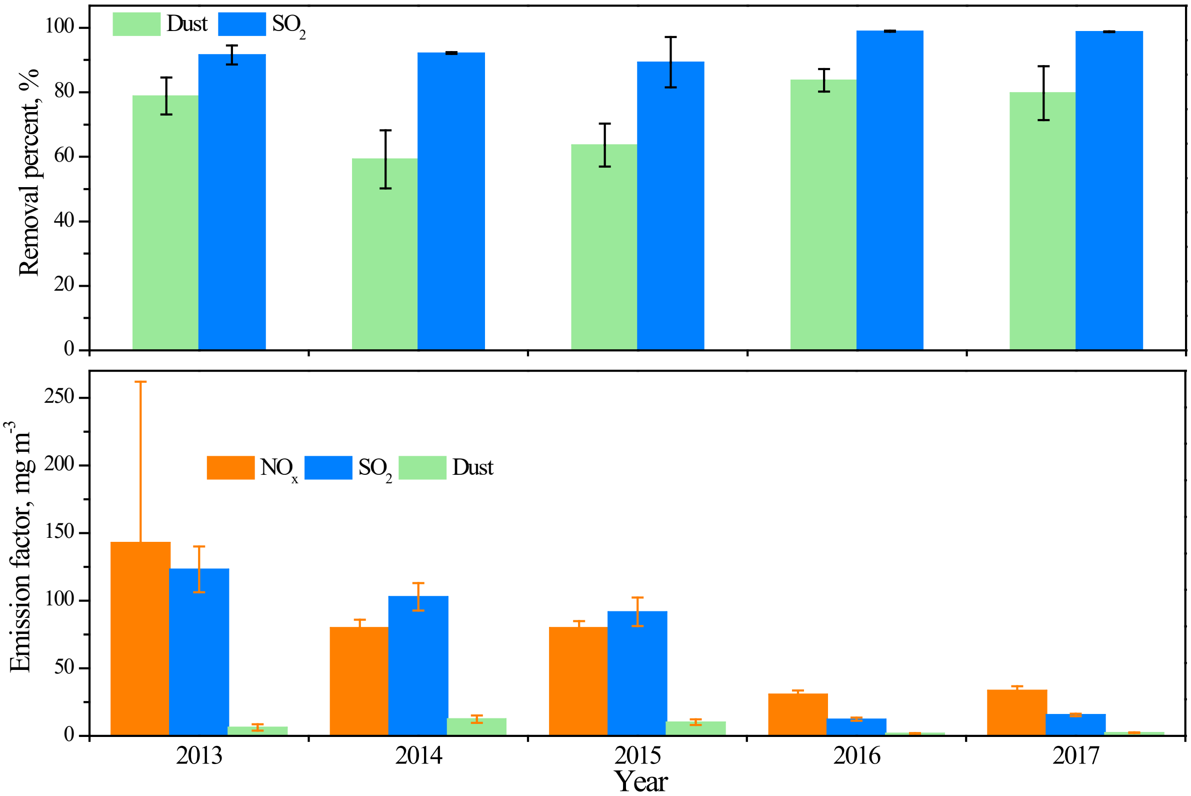

Fig. 5. Variations in pollutant emissions and removal rates from 2013 to 2017.

emissions in both 2013 and 2014 exceeded their limits of 50 and 100 mg m^{-3} in GB13223 in 2011, while dust emission was much lower than 20 mg m^{-3} in GB13223. Variations in pollutant emissions indicated that the considerations of equilibrium in control processes of three kinds of target pollutants (Tang *et al.*, 2023). Compared with the emissions in 2014, NO_x in 2015 almost unchanged, SO_2 and dust decreased slightly by 10.8% and 18.0%, indicating that the control measures in 2015 were mainly focused on SO_2. In addition, the removal efficiencies of dust and SO_2 exhibited the slight increase and decrease in 2015. The most increases of dust and SO_2 removal rates by 31.5% and 10.7% were found in 2016 compared with those in 2015. Accordingly, the dust and SO_2 removal rates in 2016 were up to 83.8% and 98.9%, respectively. The NO_x, SO_2, and dust emissions far decreased by 61.6%, 86.6%, and 82.3%, which were as low as 30.7, 12.3, and 1.80 mg m^{-3}. The highest pollutant removal efficiencies and lowest emissions in 2016 evidenced that the ULE retrofit had played the most positive role in 2016. The related pollutant emissions exhibited a significant enhancement for three pollutants in 2017 compared to those in 2016, with the increases of 9.84%, 26.3%, and 30.3% for NO_x, SO_2, and dust, respectively. However, these emissions were lower than the limits in ULE standards. Overall, there was a lag in the reductions of pollutant emissions and improvement of pollutant removal rates compared to the ULE retrofit. ULE transformation was conducted since December in 2014, while all three kinds of pollutants met the emission standards of ULE until 2016, which was mainly attributed to the debugging and optimizing operating parameters, and organizational coordination of existing and newly added pollutant control devices (Li *et al.*, 2023a; Liu *et al.*, 2023). Finding ways to use coal efficiently and reduce related pollutant emissions was a long-term task that would take sustained work (Tang *et al.*, 2023).

It should be noted that there was a weak correlation between the pollutant emissions and corresponding removal efficiencies based on four-year data. The comprehensive impacts from the unit loads, flue gas temperature, coal components, pollutant control technology and devices, and etc. (Wang *et al.*, 2020). Wu *et al.* (2021) indicated that the filterable particle (FP) emission held weak correlation with gas temperature, while condensable particles (CP) correlated well with the temperature. Dios *et al.* (2013) and Feng *et al.* (2021) reported that the coal components possessed a significant impact on dust emissions including both CP and FP. Liu *et al.* (2023) indicated that pollutant control devices clearly affected the dust emission and associated water-soluble ions. Chen *et al.* (2022) illustrated that the ULE dust removal devices exhibited varied removal efficiencies for particles of different sizes. Yang *et al.* (2021) described that SO_x removal was largely affected by the number of tray absorber. Xu *et al.* (2022) reported that the NH_3-SCR device held the negative effects on dust emission, especially on condensable particle emission. Yang *et al.* (2018a) indicated that fuel types largely affected the dust emission and its components.

3.5 Emission Variations in Expressions of kg h^{-1} and g MWh^{-1} from 2013 to 2017

At present, there were large discrepancies in the existed emission factors for three pollutants (Ma *et al.*, 2016; Chen *et al.*, 2019; Wu *et al.*, 2023b). The current emission factors for coal-fired power plants were obtained in different expressions including emission mass per coal mass (kg t^{-1}) (Chen *et al.*, 2019), product of coefficients and contents of sulfur and ash in coal expressed as Skg t^{-1} and Akg t^{-1} (Zhao *et al.*, 2010), emissions per unit of power generation (g MWh^{-1}) (Li *et al.*, 2018), and emission concentrations in flue gas (mg m^{-3}) (Tang *et al.*, 2023). To improve practicality and renewal of emission factors, we further calculated the pollution emission factors expressed in kg h^{-1} and g MWh^{-1} in this study. Fig. 6 showed the inter-annual changes (2013–2017) in pollutant emission factors for three kinds of air pollutants. NO_x and SO_2 emissions exhibited the similar variation trends and they far decreased in 2015 due to the execution of the ULE transformation in December, 2014. The emissions of NO_x, SO_2, and dust in kg h^{-1} and g MWh^{-1} far decreased by 98.1%, 98.3%, and 97.2%, and 98.0%, 98.1%, and 97.2%, respectively, in 2015 compared with those in 2014. For instance, the emissions of NO_x, SO_2, and dust in kg h^{-1} significantly reduced from 134, 165, and 11.4 in 2014 to 2.50, 2.84, and 0.324 in 2015. It should be

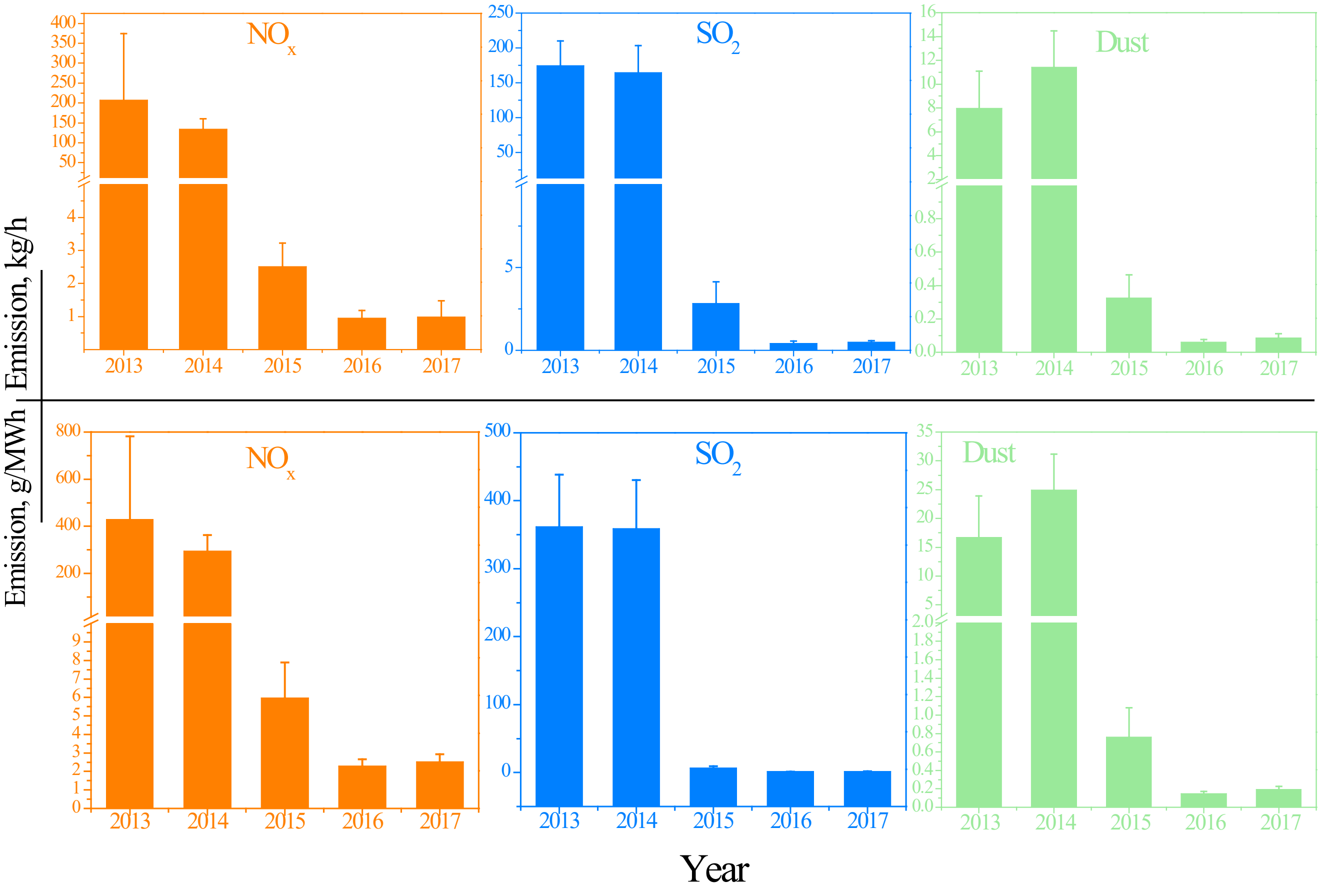

Fig. 6. Variations in emissions presented by kg h^{-1} and g MWh^{-1} of three pollutants from 2013 to 2017.

noted that the emissions in kg h^{-1} or g MWh^{-1} in 2015 compared with those in 2014 had more decreases than emissions in mg m^{-3}, which should be ascribed to the more reasonable air supply or enhancement in thoroughness of coal combustion due to the ULE or LNB transformation. Furthermore, the lowest emissions of pollutants occurred in 2016 and they were as low as 0.948, 0.422, and 0.061 kg h^{-1}, and 2.28, 1.02, and 0.148 g MWh^{-1}, respectively. In addition, Attributing to the inertness and high cost in the ULE implementation, the emissions in 2017 exhibited slightly increasing trends for all the air pollutants with the increases of 3.41%, 18.5%, and 40.4%, and 11.1%, 27.2%, and 30.1% for emissions in kg h^{-1} and g MWh^{-1}. However, the emissions of all three pollutants in 2017 still met the emission limits defined in the ULE standards. In a word, the ULE transformation played a positive role in emission reductions.

4 CONCLUSIONS

To evaluate the impacts of ultra-low emission (ULE) transformation on emission reductions of air pollutants, a systematic field observation was performed in a 660 MW power generation unit of a coal-fired power plant. We obtained the online monitoring data for five years from 2013 to 2017, which included the unit loads, flue gas temperature, hourly flue gas generation, and emission amounts in mg m^{-3}. Together, we calculated the emission factors in kg h^{-1} and g MWh^{-1} for NO_x, SO_2, and dust based on the aforementioned data. Some interest conclusions were deduced and shown as follows:

Before the ULE execution, the emissions of NO_x and SO_2 far exceeded their limits as 20 and 50 mg Nm^{-3} in the ULE standards. For example, their actual emissions were 143 and 123 mg m^{-3} for NO_x and SO_2, respectively. Focusing on the emission reductions in NO_x in 2014, NO_x decreased most by 44.0% compared with that in 2013 and followed by 16.4% of SO_2. However, dust emission increased by 98.1% and they were 6.26 and 12.4 mg m^{-3} in 2013 and 2014, respectively. Generally, removal of SO_2 and NO_x was more difficult than that of dust.

The actual emission reductions exhibited a lag relative to the ULE reclamation measures. The ULE transformation was put into effect in December of 2014. The emissions of NO_x, SO_2, and dust in December still failed to meet the ULE standards though they were the minimum values among twelve months. It was not until 2016 that the actual emissions of all three kinds of pollutants met the ULE standards. At the same time, however, all pollutant emissions exhibited an increasing trend in 2017 compared to 2016 due to the execution inertia and high cost of the ULE innovation.

No significant correlation appeared between air pollutant emissions and their removal efficiencies of control facilities. Attributing to the ULE transformation, the comprehensive impacts on air pollutant emissions from unit loads, flue gas temperature, coal components, pollutant control technology and devices, and unit running parameters had led to above weak correlation. Therefore, the simulation of complex multi-linear relationships among aforementioned factors was necessary for further reduce the pollutant emissions and improve the air quality.

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