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REVEALING HETEROGENEITY OF N_2O CONCENTRATION IN SELECTIVE CATALYTIC REDUCTION DENITRIFICATION: A MULTI-POINT SAMPLING STUDY IN COAL-FIRED POWER PLANT

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Abstract: Selective catalytic reduction (SCR) plays a critical role in achieving ultra-low emissions from coal-fired power plants by enabling highly efficient removal of nitrogen oxides (NO_x). However, the denitrification process may concurrently generate nitrous oxide (N_2O), a potent greenhouse gas often overlooked in emission control strategies. Crucially, conventional continuous emission monitoring systems (CEMS) focus solely on NO_x reduction efficiency without tracking byproduct formation, creating a potential blind spot for N_2O emissions when reduction reactions are incomplete. To address this gap, this study conducted multi-point on-site sampling monitoring within SCR system of ultra-low emission units at a 693 MW coal-fired power plant and a separate 2× 300 MW plant. A pivotal question was investigated: Are N_2O emissions inevitably elevated by SCR operation? Our findings reveal significant spatial heterogeneity in N_2O distribution: while concentrations increased at certain locations (e.g., upstream catalyst layers), they unexpectedly decreased at others (e.g., optimized mid-reactor zones), demonstrating position-dependent synergistic reduction of NO_x and N_2O under specific operational conditions. This work highlights the necessity of multi-dimensional emission assessment beyond standard CEMS protocols and provides actionable insights for co-controlling air pollutants and greenhouse gases in industrial air pollution control systems.

Keywords: Nitrous oxide; Nitrogen oxide; Selective catalytic reduction; Ultra-low emission; Coal-fired power plant

1 INTRODUCTION

Air pollution is one of the most important challenging problems in China and Egypt, which have a major impact on increasing threat of development in all fields [1-2]. The Chinese government prioritizes ecological and environmental quality improvement, having implemented comprehensive “ultra-low emission standards” in the power generation sector to mitigate air pollution’s adverse health impacts [3]. The main pollutants measured were sulfur Dioxide (SO_2), nitrogen oxides (NO_x), carbon monoxide (CO), ozone (O_3), and particulate matters with varied diameter (PM) [4]. It is known that various compounds exist in the nitrogen oxides depending on the bonding state of nitrogen and oxygen. Most of what is generated by combustion of fuels NO and NO_2, and is generally referred to as NO_x (Hereinafter referred to as NO_x) [5].

Figure 1 depicts a representative ultra-low emission control process for coal-fired power plants. Following emission from the boiler, flue gas undergoes sequential treatment: SCR for substantial NO_x abatement, followed by electrostatic precipitation (ESP) for particulate matter (PM) removal, and finally passes through a flue gas desulfurization (FGD) unit for sulfur dioxide (SO_2) elimination [6-7].

As the cornerstone NO_x control technology in ultra-low emission systems, SCR employs ammonia (NH_3) to catalytically reduce NO_x to nitrogen (N_2) in flue gas [8]. Nevertheless, SCR systems may simultaneously generate N_2O through competing reactions where NH_3 react with nitric oxide (NO) and oxygen (O_2) or with nitrogen dioxide (NO_2) [9-12]. This side-product formation warrants concern given N_2O’s dual role as both a potent greenhouse gas and ozone-depleting substance. In addition, to address the challenge of climate change, increasing renewable energy integration has necessitated deep load-cycling operations in coal-fired power plants, inducing frequent boiler load fluctuations. Consequently, flue gas temperature variability alters the intrinsic activity of the SCR catalyst, potentially compromising denitrification efficiency while affecting N_2O formation pathways [13-15].

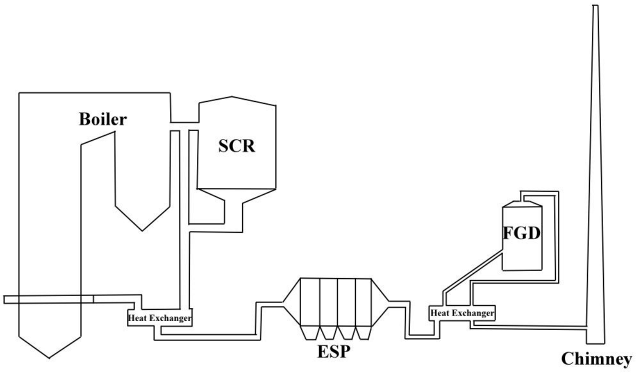

Figure 1 Flowchart of Ultra-Low Emission Technology in Coal-Fired Power Plants

Despite these operational complexities, systematic quantification of N_2O emissions from operational SCR systems remains scarce. Moreover, influential factors governing N_2O generation are inadequately characterized. This study bridges these knowledge gaps through field measurements quantifying N_2O concentrations at SCR inlets and outlets across multiple coal-fired plants. Furthermore, the correlative relationship between N_2O and NO_x concentrations in the flue gas were further examined to elucidate interactions mechanisms and evaluate co-control strategies for these interconnected pollutants.

2 EXPERIMENTAL METHODS

Figure 2 shows the field-deployed sampling setup used in the coal-fired power plant industrial environment. The vacuum pump operated at a flow rate of 10 L/min. The vacuum sampling box, with a total volume of 2 L, was connected to the vacuum inlet of the pump on its left side and to the sampler on its right side. A sampling bag was placed inside the box. The sampler is a steel tube approximately 2.5 meters in length, with a wall thickness of 2 mm and an outer diameter of 10 mm. One end of the steel tube was inserted into the sampling hole, while the other end was connected to the exterior of the vacuum sampling box.

Figure 2 In Situ Deployment of Plug-And Play Sampling Probes at Multi-Port SCR Monitoring Locations

The specific procedure for a single gas sample collection was as follows: First, with no sampling bag placed inside, the lid of the vacuum sampling box was securely closed, the pressure relief valve was tightened, and the vacuum pump was operated for approximately one minute to create a negative pressure environment. While keeping the vacuum pump running, the vacuum sampling box was then quickly opened, and the sampling bag was connected to the internal interface of the box. Subsequently, the valve of the sampling bag was opened, and the box was closed tightly again. Once the desired volume of gas had been collected, the vacuum pump was turned off, the pressure relief valve was loosened, and the vacuum sampling box was opened. Finally, the valve of the sampling bag was closed, and the bag was removed from the box.

3 RESULT AND DISCUSSION

Flue gas samples were collected from the SCR unit of a 693 MW coal-fired power plant located in China, featuring a supercritical once-through boiler with variable pressure operation and an integrated recirculating pump start-up system. Flue gas was collected into the gas bag from 10 sampling points located on both the A and B sides of the SCR reactor. N_2O concentrations were measured using gas chromatography. The collected flue gas samples were subjected to detailed analysis and characterization using gas chromatography (GC). The test results are shown in **Figure 3**. In **Figure 3a**, the green line represents the test results from the A side, the blue line represents the test results from the B side, and the black dashed horizontal line indicates the approximate background concentration of atmospheric N_2O (340 ppb). Correlation between N_2O and NO_x concentrations in flue gas samples were presented in **Figure 3b**. NO_x concentrations were measured using nitrogen oxide analyzers. The red line represents the linear fit between the measured N_2O and NO_x concentrations.

As shown in **Figure 3a**, significant spatial heterogeneity in N_2O concentrations was observed between across sampling point on both Sides A and B of the SCR unit, with approximately 50% of locations exceeding atmospheric background levels. **Figure 3b** further reveals a strong liner correlation between N_2O and NO_x concentrations.

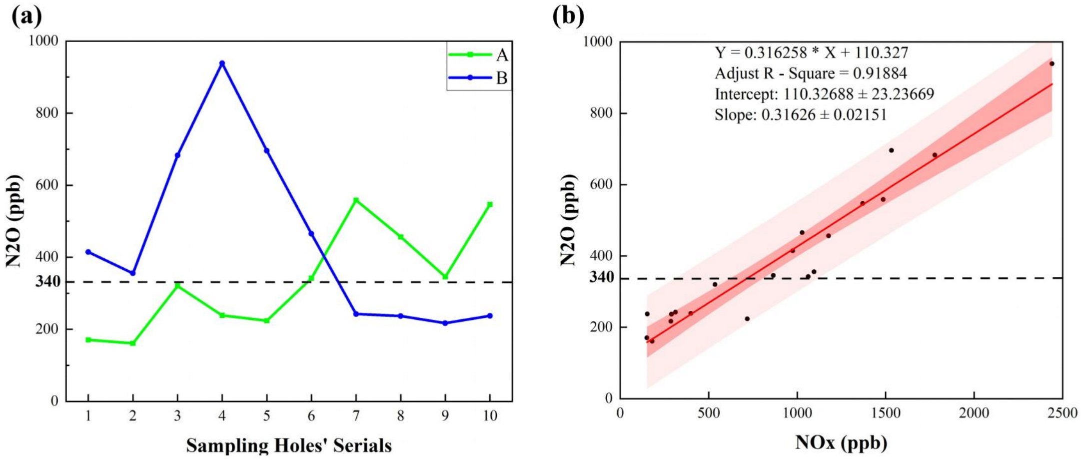

Figure 3 Flue Gas Sample Test Results. (a) N_2O Concentrations in Flue Gas From Different Sampling Holes

Complementing these findings, a parallel investigation was conducted at a coal-fired combined heat and power (CHP) facility (2 × 300 MW subcritical units). Flue gas samples were collected at the inlet and outlet of both Side A and Side B of the SCR unit associated with Unit #1 under two power generation loads: 210 MW and 105 MW. The results are summarized in Table 1. Key observations confirm consistent detection of both N_2O and NO_x at all sampling locations, alongside marked asymmetries in pollutant concentrations and removal efficiencies between SCR sides. Moreover, the concentrations of N_2O and NO_x emitted from the boiler were significantly higher under the higher power generation load compared to the lower load. Correspondingly, the removal efficiencies of these pollutants also varied with power load. Notably, the concentrations of N_2O and NO_x at the outlet were significantly lower than those at the inlet, unequivocally demonstrating SCR's capacity for simultaneous N_2O/NO_x abatement through synergistic removal mechanisms. This finding supports, to some extent, the results presented earlier.

Table 1 Flue Gas N_2O and NO_x Concentrations and Their Removal Efficiencies Under Different Power Generation Loads

Power Generation Load	Sides	Location	N_2O(ppb)	NO_x(ppb)	Removal Efficiency of N_2O (%)	Removal Efficiency of NO_x (%)
70%	A	Inlet	667	14745	87.56	88.75
70%	A	Outlet	83	1659		
70%	B	Inlet	598	13715	93.14	92.28
70%	B	Outlet	41	1059		
35%	A	Inlet	460	12710	84.78	77.07
35%	A	Outlet	70	2915		
35%	B	Inlet	288	11275	99.31	91.04
35%	B	Outlet	2	1010		

Note: "Power Generation Load" refers to the percentage of the actual load of the target unit during sampling relative to its designed maximum power generation capacity. Each installed SCR reactor has two sides, denoted in this study as "Side A" and "Side B". The terms "Inlet" and "Outlet" indicate the sampling locations: the former refers to the sampling port located upstream of the SCR reactor, while the latter refers to the port located downstream. "N_2O (ppb)" and "NO_x (ppb)" represent the relative concentrations of N_2O measured by gas chromatography and NO_x measured by a nitrogen oxide analyzer, respectively. The "Removal Efficiency" is calculated as: "Removal Efficiency" = ("Inlet Concentrations" – "Outlet Concentrations") / "Inlet Concentrations" × 100%, which applies to both N_2O and NO_x in this work.

4 CONCLUSION

In this work, a sampling-based measurement approach was employed to investigate the substantial spatial heterogeneity in N_2O concentration within ultra-low emission SCR systems while revealing their paradoxical capacity for simultaneous NO_x (88.75-91.04%) and N_2O (84.78-99.31%) abatement under optimized conditions, thereby establishing SCR's dual-function potential for carbon-pollutant synergy. These empirical insights advance co-governance strategies by demonstrating how existing pollution control infrastructure can mitigate greenhouse gases alongside conventional pollutants. Future research should integrate catalytic mechanism modeling with multi-plant field validation to develop sector-transferable frameworks applicable beyond power generation to energy-intensive industries like primary metal smelting and bulk chemical manufacturing.

COMPETING INTERESTS

The authors have no relevant financial or non-financial interests to disclose.

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