Coordinated Control of Fine-Particle and Ozone Pollution by the Substantial Reduction of Nitrogen Oxides

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1. Introduction

In recent years, the air quality in China has improved significantly. In many cities, however, the concentration of fine particulate matter (PM2.5) remains higher than the secondary-level national ambient air quality standard (NAAQS level-2, 35 μg m⁻³, GB3095-2012 [1]) and much higher than the first-level NAAQS (15 μg m⁻³, GB3095-2012 [1]) and the World Health Organization (WHO) air quality guidelines (5 μg m⁻³). In addition, heavy haze pollution still occurs, especially in wintertime. The concentration of ozone (O₃) has increased in many regions of China. Indeed, O₃ has become the major air pollutant after PM2.5, especially in summertime. Thus, the coordinated control of PM2.5 and O₃, as part of the 14th Five-Year Plan for Economy and Social Development of the People’s Republic of China, remains a considerable challenge.

2. Substantial reduction of nitrogen oxides for the effective coordinated control of PM2.5 and O₃

2.1. Negative correlation between PM2.5 and O₃

Highly complex interactions occur under elevated concentrations of both PM2.5 and O₃, which affect radiation flux, quench free radicals, and drive oxidation, among other processes. In most regions of China with high PM2.5 levels, especially in northern China, the temporal variations of PM2.5 and O₃ (i.e., the hourly average concentrations) are negatively correlated in both wintertime and summertime. The same thing has occurred in the interannual variations of PM2.5 and O₃ in recent years [2]. This relationship has only disappeared in a few regions of southern China, such as the Pearl River Delta, where the concentration of PM2.5 is close to NAAQS level-2, which is also the WHO first-stage interim target. In every city of China, PM2.5 and O₃ vary in opposite directions, unless the concentration of PM2.5 falls below a certain threshold (e.g., 50 μg m⁻³) [2], as indicated by the correlation coefficients of PM2.5 and O₃ in Fig. 1. Therefore, reducing the concentration of PM2.5 to the threshold value in order to disrupt the negative correlation is an essential precondition for the coordinated control of PM2.5 and O₃.

2.2. Reduction of nitrogen oxides for the effective control of PM2.5

From 2013 to 2019, the concentrations of PM2.5 and sulfur dioxide (SO₂) in 74 key Chinese cities decreased by 47% and 75%, respectively, while the concentration of nitrogen dioxide (NO₂) only decreased by 23% [3]. Thus, nitrogen oxides (NOₓ) emissions remain poorly controlled, which limits further decreases in PM2.5. NOₓ convert to nitrate, which is the most abundant chemical component of PM2.5 in many cities [4]. Furthermore, high concentrations of NOₓ result in a high atmospheric oxidation capacity for heterogeneous and aqueous phase reactions [5–7], which is an important driving force for the growth of PM2.5. According to the analysis of the observational data from the China National Environmental Monitoring Center (CNEMC), the sensitivity of PM2.5 to NO₂ is much higher than its sensitivity to SO₂ (Fig. 2). For example, in China, research has indicated that a reduction of 1 μg m⁻³ NO₂ will reduce PM2.5 much more than a reduction of 1 μg m⁻³ SO₂ [2]. In 2020, the concentration of NO₂ decreased significantly during the coronavirus disease 2019 (COVID-19) lockdown [8]. Although the concentrations of SO₂ and carbon monoxide (CO) did not markedly decrease, the concentration of PM2.5 decreased significantly and showed highly similar spatial and temporal distribution characteristics with the decrease in NO₂ [8]. This finding indicates the high effectiveness of NOₓ reduction in PM2.5 control.

2.3. The feasibility of substantial NOₓ reduction for the coordinated control of O₃

From 2013 to 2019, the concentration of O₃ in 74 key Chinese cities increased by 29% [3]. Atmospheric O₃ is mainly generated via photochemical reactions between NOₓ and volatile organic compounds (VOCs) 

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compounds (VOCs) and is mainly a problem during summer conditions. As a result, controlling O3 requires the control of these two main precursors. There is a significant nonlinear relationship between O3 and the emissions of NOx and VOCs [3,9]. Although O3 formation is usually controlled by NOx concentration on a regional scale, it is sensitive to VOCs in most urban areas of China. However, it is usually very difficult to achieve a substantial emission reduction of VOCs in a short time. VOCs emitted by biogenic sources are natural processes and are difficult to regulate. Moreover, anthropogenic emissions of VOCs are highly complex and dispersed, so their effective control remains a great challenge. In a VOC-controlled O3 formation regime, a slight reduction in NOx emissions may cause an increase in O3, whereas a substantial reduction in NOx will change the VOC-controlled regime to a NOx-controlled regime and thus effectively reduce O3. In the United States, the historical control of O3 supports the reduction of NOx emissions: In the early stages, the California Government initially attempted to control VOCs, but the effect on O3 was limited; after increasing NOx emission control efforts, however, O3 pollution gradually decreased [10], indicating the key role of reducing NOx in O3 pollution control [9]. The change in air quality during the COVID-19 lockdown in early 2020 in China also provides solid evidence. During the initial lockdown, the slight decrease in NO2 and PM2.5 concentrations resulted in an obvious increase in O3 concentrations. However, under the strictest lockdown conditions, the NO2 concentration decreased by about 70% in some regions of China, which resulted in a significant reduction in PM2.5 and a cessation of the increase in O3 [8]. These results indicate that strengthening the reduction of NOx emissions is an effective and feasible measure for the coordinated control of PM2.5 and O3 in China. This conclusion is also supported by modeling studies. For example, in a case study in the Beijing–Tianjin–Hebei (BTH) region, a reduction in NOx emissions of greater than 60% was proposed in order to ensure the effectiveness of PM2.5 and O3 control [11]. Recently, WHO updated the global air quality guidelines. The new guidelines recommend a much lower level (10 μg/m^3) for NO2 concentration than 2005 guidelines (40 μg/m^3). The updates indicate an international consensus on substantial NOx reduction for further improvement of air quality and protection of human health.

3. Nitrogen oxides: Key pollution precursors with high reduction potential

NOx are the key precursors of PM2.5 and O3 pollution and demonstrate high reduction potential in China. They are mainly emitted from stationary combustion plants and internal combustion engines of transportation vehicles. Therefore, reducing NOx emissions is also highly consistent with the current carbon peaking and carbon neutralization policies. At present, the selective catalytic reduction (SCR) of NOx by NH3 (NH3-SCR) technique is...
widespread in coal-fired power plants, satisfying the ultra-low NO\textsubscript{x} emission limit [12]. However, for NO\textsubscript{x} reduction in non-electric industries, such as the steel, nonferrous metal, cement, glass, and ceramic industries, the NH\textsubscript{3}-SCR technique requires improvement, such as low-temperature activity and a tolerance to sulfur. Thus, different denitrification technologies need to be developed.

Meanwhile, remarkable progress has been made in NO\textsubscript{x} reduction from automotive exhausts [13]. For example, using a three-way catalytic technique, NO\textsubscript{x}, hydrocarbons, and CO can be efficiently removed from gasoline vehicle exhausts. The SCR of NO\textsubscript{x} by urea (urea-SCR) has also been successfully developed for diesel vehicle NO\textsubscript{x} emission control [14]. Under real driving conditions, however, NO\textsubscript{x} emissions from diesel vehicles equipped with the urea-SCR system often exceed the standard limits. Firstly, the messages of on-board diagnostic (OBD) systems regarding NO\textsubscript{x} emissions could be artificially shielded. Of course, this illegal situation is being corrected through the technical means of online monitoring. Secondly, certain diesel vehicles (e.g., buses and garbage trucks) often run at low speed. In this case, the urea-SCR system cannot operate efficiently. At present, urea-SCR catalyst with wide temperature window is being developed to solve this technical problem. Finally, if an unqualified urea solution (commonly known as AdBlue) is used in the SCR system to reduce cost—for example, urea solution with a low concentration of urea and/or containing impurities—it results in NO\textsubscript{x} emissions that exceed the standard limits. In addition, the domestic technologies for engine control systems and high-pressure common rail injection systems are still relatively undeveloped, which further impedes the implementation of China VI emission standards for diesel heavy-duty vehicles and the formulation of the next stage of emission standards. Thus, the actual effectiveness of NO\textsubscript{x} reduction technology is still far from perfect, leaving considerable room for improvement. Furthermore, China’s targets are to achieve peak carbon dioxide emissions before 2030 and carbon neutrality by 2060. These targets require a great change in the energy structure, which will fundamentally solve the NO\textsubscript{x} emission problem in the future. For example, the number of electric vehicles is increasing rapidly in China. With electricity being generated from sustainable energy, a significant reduction in both NO\textsubscript{x} and VOC emissions from vehicles can be expected [9].

4. Suggestions for substantial reductions in NO\textsubscript{x}

4.1. Accelerating research and development of high-efficiency NO\textsubscript{x} control technologies for various industrial furnaces

To achieve ultra-low NO\textsubscript{x} emissions in industries other than thermal power plants, great effort should be devoted to the development of high-efficiency denitrification technologies with low cost and the promotion of large-scale application. These technologies include low-intermediate-temperature SCR technologies with high efficiency and stability [15] and selective non-catalytic reduction (SNCR) combined with SCR and oxidative denitrification [16]. Moreover, to achieve impactful abatement of NO\textsubscript{x} and the simultaneous reduction of carbon dioxide (CO\textsubscript{2}) emissions, embedded denitrification technologies that suit the temperature distribution inside furnaces should be developed, which would help to reduce energy input (e.g., coal, gas, and electricity). In addition, for the simultaneous reduction of NO\textsubscript{x} and CO\textsubscript{2} emissions in industries with long and multiple manufacturing processes, such as the steel and nonferrous metal industries, the industrial structure should be upgraded by developing clean and short-process smelting, as well as by conducting comprehensive process optimization.

4.2. Formulating ultra-low emission limits and strengthening assessment and supervision for key industries

The formulation of ultra-low emission limits and the development of technological guidelines for NO\textsubscript{x} control are urgently needed for non-electric industries such as building materials and nonferrous metals, according to their flue gas characteristics. Comprehensive evaluation after the implementation of ultra-low emission technologies in the iron and steel industry also needs to be strengthened, and techno-economic analysis of different ultra-low NO\textsubscript{x} emission technologies in such industries should be emphasized [17,18]. In addition, clarifying the benefits and shortcomings of such technologies should provide an important reference for proposing and improving ultra-low NO\textsubscript{x} emission technologies and schemes suitable for different industries. Finally, in combination with China’s economic development and energy consumption trends, it is necessary to predict total NO\textsubscript{x} emissions and their distribution by industry in the future, and to evaluate the reduction potential of NO\textsubscript{x} emissions in various industries and the environmental impacts induced by NO\textsubscript{x} reduction.

4.3. Promoting the research and development of key technologies for diesel engine emission control, and enhancing joint management and control of vehicle fuel

Firstly, efforts must be made to advance engine control systems, high-pressure common rail injection systems, turbocharger systems, and after-treatment catalysts and to establish independent and controllable diesel engine emission control technology chains [19]. Secondly, the next stage of national emission standards for diesel heavy-duty vehicles, off-road diesel engines, and vessels should be formulated as soon as possible. The implementation of these standards should encourage the adoption of new technologies to further reduce diesel engine exhaust emissions. Furthermore, more stringent standards for diesel, additives, AdBlue, and lubricating oil should be formulated and implemented to ensure the efficient and stable operation of diesel engines. Thirdly, the research and development of remote online diagnostic systems, remote sensing, and portable emission measurement technologies should be promoted. By integrating these advanced technologies, an intelligent digitized system can be developed for the supervision and management of in-use vehicle emission control. With this complete system, the closed-loop control of remote real-time vehicle emission monitoring, inspection, and maintenance can be achieved.

Acknowledgments

This work was financially supported by the National Key Research and Development (R&D) Program of China (2017YFC0211100), the consulting research project of the Chinese Academy of Engineering (2020-XY-22), and the Cultivating Project of Strategic Priority Research Program of Chinese Academy of Sciences (XDPB1901 and RCEES-CYZX-2020).

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