

# N<sub>2</sub>O Formation in Selective Non-Catalytic NO<sub>x</sub> Reduction Processes

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## ABSTRACT

*Nitrous oxide is not an environmentally regulated species in the U.S., but it does participate in the stratospheric ozone chemistry and contributes to the greenhouse effect. Nitrous oxide has been found to be a by-product of the selective non-catalytic reduction process. Chemical kinetic calculations demonstrated that the formation of nitrous oxide in the urea-based selective non-catalytic reduction process is linked to the conversion of NO by cyano species released from the process parent compounds. This conversion occurs within in temperature window between 850 and 1050°C. With urea injection, nitrous oxide emissions represent up to 20 percent conversion of the NO<sub>x</sub> reduced. The amount of nitrous oxide formed depends primarily on the process temperature, the amount of chemical injected, the initial NO<sub>x</sub> level, and the carbon monoxide level in the gas stream. These observations, which were based on the chemical kinetics of the process, should be considered in designing selective non-catalytic reduction systems to minimize nitrous oxide by-product formation.*

**Keywords:** Selective Non-Catalytic Reduction Process, Nitrous Oxide Emissions Chemical Kinetics

## 1. Introduction

While global climate changes have been associated with increased levels of carbon dioxide (CO<sub>2</sub>) in the atmosphere, there is growing concern about the role of other trace gas species such as methane (CH<sub>4</sub>), chlorofluorocarbons (CFC) and nitrous oxide (N<sub>2</sub>O). In fact, the trace gases are expected to equal the global warming effect of CO<sub>2</sub> within 50 years. Nitrous oxide affects the atmosphere at the troposphere level as well as at the stratosphere level. N<sub>2</sub>O is a relatively strong absorber of infrared radiation and is therefore implicated as an important contributor to the “Greenhouse Effect.” It is estimated that a mere doubling of its concentration will result in a 0.28°C contribution to global warming [1]. Since N<sub>2</sub>O is chemically stable in the troposphere, it can be transported to the stratosphere where it has also been responsible for ozone (O<sub>3</sub>) depletion (NO + NO<sub>2</sub> emitted into the troposphere are converted into nitrate and removed by rainfall). In the stratosphere, N<sub>2</sub>O is converted to NO, which is the primary species responsible for establishing the equilibrium stratospheric O<sub>3</sub> concentration. One estimate suggests that doubling atmospheric N<sub>2</sub>O concen-

tration would result in a 12 percent decrease in total column of ozone [2].

Mean global concentrations of N<sub>2</sub>O are approximately 300 ppb (about one-thousandth that of CO<sub>2</sub>) and have been increasing at a rate of 0.2 to 0.4 percent per year [1-3]. The latest Energy Information Administration – Emissions of Greenhouse Gases Report reports 2006 U.S. emissions of nitrous oxide of 375.7 million metric tons carbon dioxide (CO<sub>2</sub>) equivalent [4]. Although, the increase of N<sub>2</sub>O in the atmosphere has been attributed to anthropogenic sources, uncertainties in the source, sink, and accumulation terms have prevented a clear estimation of the importance of the various sources. References in the literature have suggested that combustion of fossil fuels results in N<sub>2</sub>O emissions which can account for the observed increase in N<sub>2</sub>O [1-5]. Measurements of nitrous emissions from electric utility power plants have indicated insignificant emissions from gas-fired flames (less than 10 ppm). On the other hand, field measurements of coal- and oil-fired power plants have shown a wide variation of N<sub>2</sub>O emissions, from 10 to above 200 ppm [6]. Some data have suggested that N<sub>2</sub>O molar emissions could be estimated as about 25 to 40 percent of the NO<sub>x</sub>

levels [7].

However, the characterization of N<sub>2</sub>O emissions from combustion sources is a complex issue due to reported findings of a major sampling artifact on measurements made on grab samples [8]. Reference [8] has demonstrated that processing combustion products containing SO<sub>2</sub>, NO<sub>x</sub> and water vapor can lead to N<sub>2</sub>O concentrations several hundred parts per million higher than those originally formed in the combustion process. The conversion of NO into N<sub>2</sub>O occurs by a reaction whose overall time constant is of the order of 2 hours. N<sub>2</sub>O emissions from combustion sources should be low given that gas-phase chemical reactions such as N<sub>2</sub>O + radicals = N<sub>2</sub> + radicals and stable species are extremely rapid at furnace temperatures (1500°C - 1700°C). Combined experimental results and kinetic modeling of sample combustion systems have shown that N<sub>2</sub>O formed early in the flame zone, is rapidly destroyed before it can reach the furnace exhaust [9].

Measurements at several full scale utility combustion sources, using continuous infrared measurement techniques, have associated utility combustion systems (firing natural gas, oil, and gas) with direct N<sub>2</sub>O emission levels below 10 ppm [10]. Furthermore, there was no correlation between the level of N<sub>2</sub>O emission and the NO<sub>x</sub> level in the flue gas.

While N<sub>2</sub>O emissions from conventional combustion equipment are nowadays recognized to be low, a number of other sources have been identified as responsible for significant N<sub>2</sub>O emission levels which could explain the increase on N<sub>2</sub>O in the atmosphere. Sources of N<sub>2</sub>O include denitrification of chemical fertilizers, biomass burning, and advanced combustion and emission control systems. The selective non-catalytic reduction (SNCR) process for NO<sub>x</sub> control has been associated with residual pollutant emissions. Possible by-products of the SNCR process included N<sub>2</sub>O, ammonia slip (NH<sub>3</sub>), ammonium sulfate salts, and increased CO emissions. The contribution of N<sub>2</sub>O from SNCR is expected to be small. Reference [11] has estimated that even if 50 percent of all fossil fuel burned by the U.S. utility industry used SNCR, the N<sub>2</sub>O produced would constitute only 0.5 to 2.0 percent of the total global production of N<sub>2</sub>O. However, it is prudent to be aware of the potential for N<sub>2</sub>O emissions from the SNCR process and to know the extent to which N<sub>2</sub>O is a process by-product, as well as, to know the process parameters and mechanisms leading to its emission. This information would be important for the development of control strategies for SNCR-produced N<sub>2</sub>O. This paper includes a discussion on N<sub>2</sub>O formation in SNCR processes from a chemical kinetics point of view, and it reports the results of parametric calculations of N<sub>2</sub>O emissions using a chemical kinetics model. Em-

phasis is placed on the urea-based SNCR process.

## 2. Selective Non-Catalytic NO<sub>x</sub> Reduction Processes

The SNCR process for NO<sub>x</sub> removal has been extensively applied in a range of industrial applications. SNCR is recognized as a NO<sub>x</sub> control method in the U.S. Environmental Protection Agency (EPA) NO<sub>x</sub> rules [12]. It involves the gas-phase reaction of a nitrogen-based reagent (typically ammonia (NH<sub>3</sub>) or urea) with NO<sub>x</sub> at elevated temperatures to form nitrogen and water. Several versions of this process have been developed. Anhydrous ammonia injection, the first generation SNCR process, works well in the temperature range of 930 to 1035°C (1700 to 1900°F). In chemistry terms, one mole of NH<sub>3</sub> is required to react with one mole of NO<sub>x</sub>. Aqueous ammonia SNCR is functionally similar to anhydrous NH<sub>3</sub>. The major difference is that the reagent is a water solution of NH<sub>3</sub> instead of a gas. In a urea-based SNCR system, an aqueous solution of urea compound (or amine salt) is injected into the oxygen-rich upper furnace and/or high temperature convective section of the boiler to promote the NO<sub>x</sub> reduction. The urea-based SNCR process takes place roughly at a temperature window between 870°C to 1150°C. Cyanuric acid and cyanic acid injection have also been used as reagents for SNCR processes. The temperature window for these acids is similar to the range of temperature in a urea injection system, 930°C to 1120°C.

## 3. Selective Non-Catalytic NO<sub>x</sub> Reduction Theoretical Model

### 3.1. Chemical Kinetics Model

A chemical kinetics model was developed to describe the time evolution of the urea-NO<sub>x</sub> process under homogeneous conditions. The numerical model describes an ideal, one-dimensional plug flow, with the temperature history approximated by a linear profile. The model assumes the reagent has already been atomized and the droplets are fully evaporated. Urea decomposition was modeled as a rapid and instantaneous one step breakdown to NH<sub>3</sub> and isocyanic acid (HNCO). The solution to this problem is simplified because the temperature history is known and it can be described by a set of ordinary differential equations of the following form:

$$\frac{dY_k}{dt} = \frac{w_k W_k}{\rho} \quad (1)$$

where  $Y_k$  is the mass fraction of the  $k$ th species,  $t$  is time,  $w_k$  is the molar production rate of the  $k$ th species by an elementary reaction,  $W_k$  is the molecular weight of the  $k$ th species, and  $\rho$  is the mixture density. The net chemical production rate  $w_k$  (the difference between the for-

ward and reverse reaction rates) of each species results from a competition among all chemical reactions involving that species. Each reaction proceeds according to the law of mass action and the rate coefficients ( $k$ ) are configured using a modified Arrhenius form:

$$k = AT^n \exp\left(\frac{-E}{RT}\right) \quad (2)$$

where the activation energy,  $E$ , the temperature exponent,  $n$ , and the pre-exponential constant,  $A$ , are parameters in the model formulation.

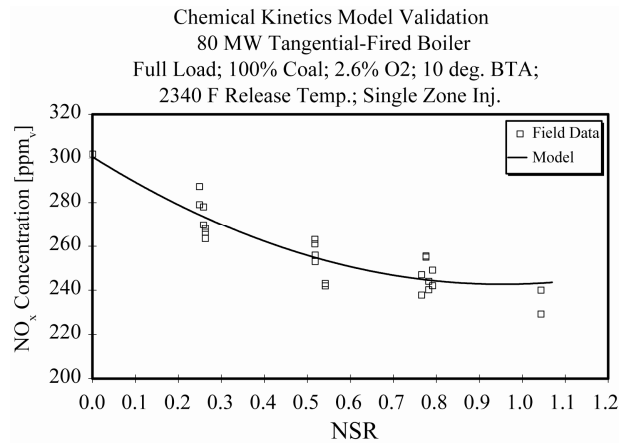
The model consists of a detailed chemical kinetic scheme involving NH<sub>3</sub> and HNCO oxidation reactions and the wet-CO oxidation reactions. The scheme is composed of 105 reversible reactions and 24 species. It comprises three complete major reaction subsets: 1) reactions of N/H/O species that are important in ammonia oxidation; 2) reactions important in the oxidation of CO; and 3) reactions involving HNCO and NCO.

Numerical solutions were calculated using Sandia's CHEMKIN Collection. The rate constants for reverse reactions were computed from the equilibrium constants, calculated using the CHEMKIN Thermodynamic database coefficients [13]. The method of solution used SENKIN [14] to predict the time-dependent chemical kinetics behavior of a homogeneous gas mixture. SENKIN uses the DASAC [15] software to solve the nonlinear ordinary differential equations which describe the species mass fractions for a system where the temperature is a specified function of time. The first-order sensitivity coefficients are computed with respect to the elementary reaction rate parameters.

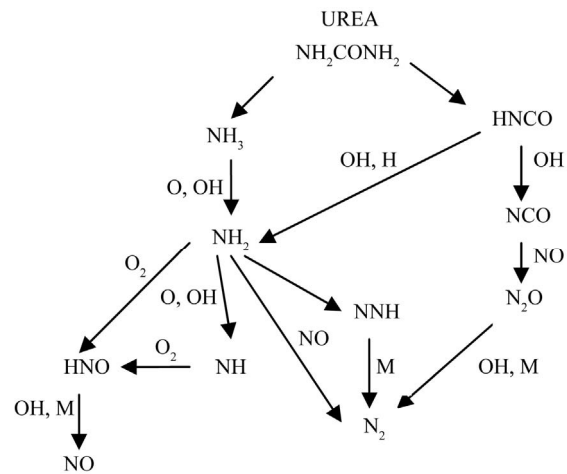
The kinetic model was validated with experimental data from a 80 MW coal-fired tangential boiler. **Figure 1** shows the comparison between measure and predicted NO<sub>x</sub> levels as a function of NSR (NSR is defined as the actual model ratio of urea to NO<sub>x</sub> divided by the theoretical stoichiometric ratio) for the unit operated at full load conditions. The data corresponds to single zone urea injection. Input to the theoretical model included CEM measurements of NO<sub>x</sub>, CO, CO<sub>2</sub> and O<sub>2</sub> in the economizer; urea injection flow; and temperature information obtained with an optical temperature probe. No measurable ammonia slip occurred at these temperatures and NSRs.

### 3.2. N<sub>2</sub>O By-Product Formation

Since the SNCR process involves chemical activity between NO<sub>x</sub> and nitrogen species in a temperature window between 870°C to 1150°C, it is possible for N<sub>2</sub>O to be present in the NO<sub>x</sub> chemical reduction process. **Figure 2** shows the overall chemical path leading to the reduction of NO and to the formation of N<sub>2</sub>O. This simpli-



**Figure 1.** Comparison between measured and predicted NO<sub>x</sub> levels.



**Figure 2.** SNCR chemical kinetics path.

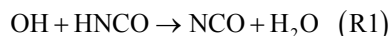
fication was based on sensitivity and rate-of-production analyses (evaluation of the order of magnitude of the reaction rates). The methods used in performing the sensitivity analysis have been described in detail in Reference [16]. Sensitivity coefficients are displayed as:

$$\beta_{ik}(x) = \frac{A_i}{X_k} \frac{\partial X_k}{\partial A_i}(x) \quad (3)$$

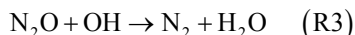
where  $\beta_{ik}(x)$  is the sensitivity coefficient for changes in the mole fraction of the  $k$ th species, the derivative of  $X_k$ , corresponds to changes in the temperature-independent factor of the  $i$ th reaction coefficient,  $A_i$ .

**Figure 2** shows that N<sub>2</sub>O is an intermediate step in the reduction of NO by the species formed from urea. There is a temperature window in the region from 870 to 1230°C, where the formation of N<sub>2</sub>O by the reaction of cyano species (HNCO, NCO) and NO occurs in high temperature combustion gases [17]. The N<sub>2</sub>O formation path (right-hand-path in **Figure 2**) involves the formation

of NCO which subsequently reacts with NO to form N<sub>2</sub>O as follows:



At relatively higher temperatures (greater than 1000°C), any N<sub>2</sub>O formed will tend to be rapidly removed by the reaction:

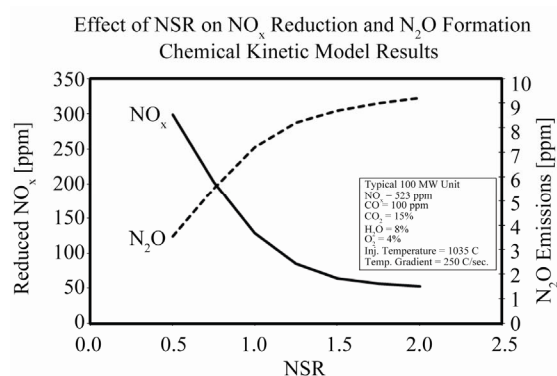


However, typical applications and SNCR target performance preclude the operation of the SNCR process at those higher temperatures. As a result, the forward reactions between NO and the cyano species evolving from reagent breakdown and species oxidation are not kinetically limited. Thus, any N<sub>2</sub>O formed in the chemical process can persist in the exhaust gases.

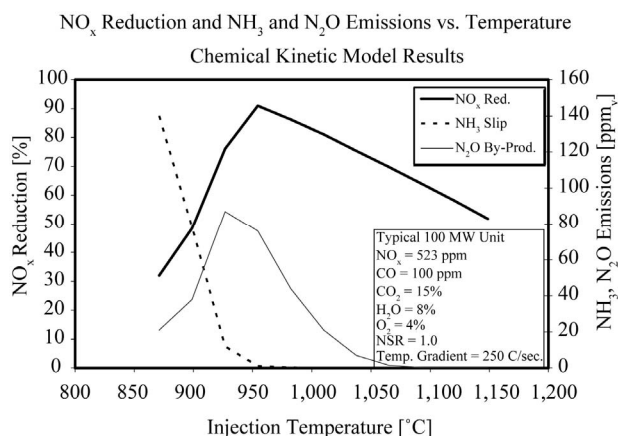
#### 4. Results and Discussion

A series of chemical kinetic calculations was performed to predict N<sub>2</sub>O formation from a utility boiler SNCR installation. The conditions used to generate the results included in **Figure 5** are typical 100 MW, NO<sub>x</sub> 523 ppm, CO 100ppm, CO<sub>2</sub> 15%, H<sub>2</sub>O 8%, O<sub>2</sub> 4%, NSR 1 and temperature gradient 250 C/sec. The results of these calculations are included in **Figure 3**. The NO<sub>x</sub> reduction vs. temperature curve of **Figure 3** is characterized by what is referred to as left- and right-side of curve. At the left-side of the curve, NO<sub>x</sub> decreases with increasing temperature because reaction rates and OH radical concentrations increase with temperature, which in turn, increases NO<sub>x</sub> reduction. At the right-side, the oxidation reactions of NH<sub>3</sub> and HNCO to NO<sub>x</sub> become a significant path for the urea-based reagent and they compete with the NO<sub>x</sub> reduction reactions. These oxidation reaction rates increase exponentially with temperature. For an NSR of one, the model indicates that N<sub>2</sub>O formation occurs in a temperature window (850°C to 1050°C) which is narrower than the temperature window needed for NO<sub>x</sub> reduction. This window coincides with the appearance of NCO, which confirms the formation of N<sub>2</sub>O in the urea-SNCR process through the right-hand-path in **Figure 2**. N<sub>2</sub>O peaks at about 930°C, which is consistent with the modeling and pilot scale results reported in Reference [11]. The model indicates that, although no N<sub>2</sub>O is present at temperatures above 1050°C, significant levels of N<sub>2</sub>O are present when the SNCR system is operated on the left-side of the NO<sub>x</sub> reduction vs. temperature curve.

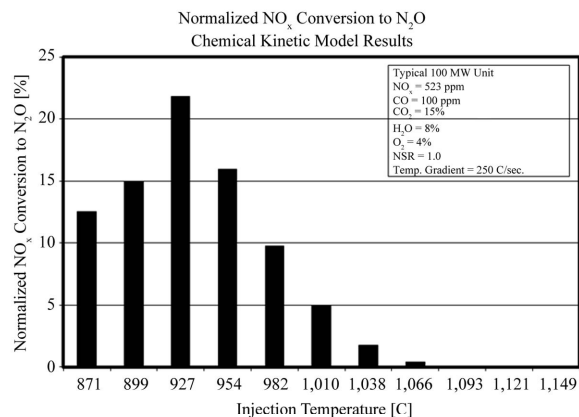
The model results were replotted as percent of N<sub>2</sub>O converted from NO<sub>x</sub> ( $\Delta\text{N}_2\text{O}/\Delta\text{NO}_x$ ). The calculations indicate a peak NO<sub>x</sub> to N<sub>2</sub>O conversion of approximately 20 percent (**Figure 4**). Again, these results coincide with typical results reported in the literature [18]. However, reductions of up to 40 percent have been reported in



**Figure 3.** Effect of NSR on NO<sub>x</sub> reduction and N<sub>2</sub>O formation.



**Figure 4.** NO<sub>x</sub> reduction and NH<sub>3</sub> and N<sub>2</sub>O emissions vs. temperature.



**Figure 5.** Normalized NO<sub>x</sub> conversion to N<sub>2</sub>O.

full-scale, utility boiler applications, which suggest that mixing has an effect on the N<sub>2</sub>O formation.

#### 4.1. Parametric Calculations

**Figure 6** shows the effect of baseline NO<sub>x</sub> on N<sub>2</sub>O formation. The urea flow required to achieve stoichiometric conditions (NSR = 1.0) is also included in the figure.

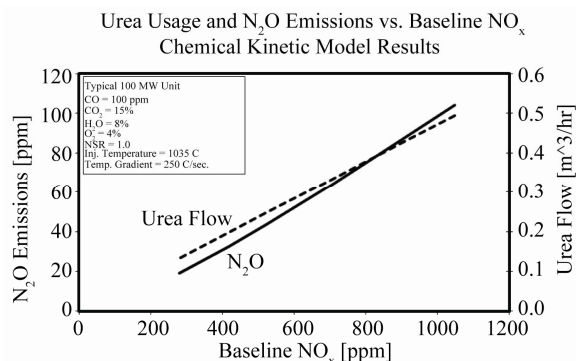


Figure 6. Urea usage and N<sub>2</sub>O emissions vs. baseline NO<sub>x</sub>.

Larger urea flows are required to maintain an NSR of 1.0 for increased baseline NO<sub>x</sub> levels. For the range of baseline NO<sub>x</sub> calculated, reduced NO<sub>x</sub> levels were in the range of 73 ppm ± 3ppm. N<sub>2</sub>O emissions from the urea-based SNCR process increases almost linearly with baseline NO<sub>x</sub>. At a release temperature of 980°C, reducing baseline NO<sub>x</sub> by half (from 1000 to 500 ppm) can reduce N<sub>2</sub>O emissions by up to 50 percent. In this case, this effect is kinetically related to the NO<sub>x</sub> concentration available for reaction R2.

The effect of carbon monoxide is shown in **Figure 7**. The concentration of CO in the flue gas inhibits the NO<sub>x</sub> reduction at relatively higher temperatures [19] (greater than 900°C). Increased CO levels also have the same effect on N<sub>2</sub>O formation. The critical step in CO oxidation is CO + OH → CO<sub>2</sub> + H. Because the urea-based compounds compete for the OH radical, larger concentrations of CO slow down the right-hand reaction branch in **Figure 2**.

## 5. Conclusions

Nitrous oxide is not an environmentally regulated species in the U.S., but it does participate in the stratospheric ozone chemistry and contributes to the greenhouse effect. N<sub>2</sub>O has been found to be a by-product of the selective non-catalytic process for NO<sub>x</sub> control for combustion processes. The SNCR process contribution of N<sub>2</sub>O to the atmosphere is expected to be small - less than 2 percent of the total global production. However, for particular operating conditions discussed in this work, the process can increase N<sub>2</sub>O emissions by almost tenfold, as compared to that produced by the combustion source. Full-scale applications of the SNCR [20-23] have verified the formation of significant levels of N<sub>2</sub>O, and indicated that N<sub>2</sub>O by-product emission is very closely associated with NO<sub>x</sub> reduction. The literature reports that for utility size applications provided with urea injection, N<sub>2</sub>O emissions are typically 10 to 20 percent of the NO<sub>x</sub> reduced, although they can be as high as 30 to 40 percent.

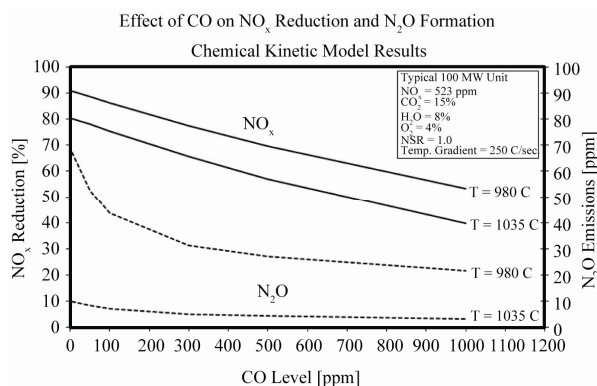


Figure 7. Effect of CO on NO<sub>x</sub> reduction and N<sub>2</sub>O formation.

Chemical kinetic calculations demonstrated that the formation of N<sub>2</sub>O in the urea-based SNCR process is linked to the conversion of NO by cyano species released from the SNCR parent compounds. This conversion occurs within a temperature window of 850 and 1050°C, which coincides with the temperature range where NCO (an intermediate species in the decomposition of urea) is produced. Maximum N<sub>2</sub>O production was found to take place at a temperature of 930°C. This reiterates that the N<sub>2</sub>O production is a problem at the low-temperature side of the SNCR NO<sub>x</sub> reduction vs. temperature curve. The amount of N<sub>2</sub>O formed depends primarily on the process temperature, the amount of chemical injected (*i.e.*, NSR), the initial NO<sub>x</sub> level, and the CO level in the gas stream. An explanation for these observations was given based on the chemical kinetics of the process. With urea injection, N<sub>2</sub>O emissions represent up to 20 percent conversion of the NO<sub>x</sub> reduced.

To date, no techniques have been offered commercially to control N<sub>2</sub>O emissions from the chemical processes used in the SNCR technology. The results of this work indicate that controlling SNCR operation could help to minimize SNCR-N<sub>2</sub>O formation. Operation at the right-hand side of the NO<sub>x</sub> reduction vs. temperature curve will translate in lower N<sub>2</sub>O and NH<sub>3</sub> emissions. Reduction of baseline NO<sub>x</sub> level will contribute to reductions in N<sub>2</sub>O formation and reagent usage.

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