

Impact of SCR Integration on N₂O Emissions in Diesel Application

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ABSTRACT

Significant reduction in Nitrogen Oxide (NO_x) emissions will be required to meet LEV III/Tier III Emissions Standards for Light Duty Diesel (LDD) passenger vehicles. As such, Original Equipment Manufacturers (OEMs) are exploring all possible aftertreatment options to find the best balance between performance, durability and cost. The primary technology adopted by OEMs in North America to achieve low NO_x levels is Selective Catalytic Reduction (SCR). The critical parameters needed for SCR to work properly are: an appropriate reductant such as ammonia (NH₃) provided as Diesel Exhaust Fluid (DEF), which is an aqueous urea solution 32.5% concentration in weight with water (CO(NH₂)₂ + H₂O), optimum operating temperatures, and optimum nitrogen dioxide (NO₂) to NO_x ratios (NO₂/NO_x). The NO₂/NO_x ratio is most influenced by Precious Group Metals (PGM) containing catalysts upstream of the SCR catalyst. Different versions of zeolite based SCR technologies are available on the market today and these vary in their active metal type (iron, copper, etc.), and/or zeolite type. To select an appropriate SCR type, the application's operating conditions as well as environmental factors must be considered. In order to fundamentally understand these differences, a study was conducted where various factors were investigated for the Tail Pipe (TP) N₂O emissions. They include 1) aftertreatment architectural differences, 2) individual component contribution to the overall N₂O make and 3) choice of specific component such as Ammonia Slip Catalyst (ASC) or its omission on the TP N₂O emissions. Results highlight the advantages and disadvantages of each factor to the overall TP N₂O emissions.

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INTRODUCTION

The introduction of Green House Gas (GHG) emissions mandates for methane (CH₄) in 2012 and nitrous oxide (N₂O) in 2015 for mid duty and heavy duty vehicles has prompted OEMs to revisit their TP GHG emissions plans. N₂O's radiative adsorption energy of 8000 to 20000 nm¹ allows the molecule to adsorb and emit radiation in thermal infra-red range of the electromagnetic spectrum. This, along with N₂O's long atmospheric life of 140-190 years², makes N₂O's global warming potential 298 times the GHG equivalency of CO₂. Due to the magnitude of this multiplier, even small amounts of N₂O emissions above the allowed emission levels become significant, which will move OEMs to significantly reduce the N₂O emissions from their vehicles.

The N₂O allowed levels are set at 50 mg/mile for Medium Duty Vehicles (MDV) based on a FTP-75/HFET combination cycle where 55% contribution is from FTP-75 emissions and 45% from HFET.³ For Passenger Cars (PC), Light Duty (LD) vehicles this allowed level is set to 10 mg/mile based on FTP-75 city cycle only⁴ and for Heavy Duty (HD) is set at 100 mg/hp-hr based on HD FTP-cycle.⁵

The high temperature and pressures prevailing in a diesel engine during the combustion cycle create conditions that are thermodynamically unfavorable to N₂O creation as shown in Figure 1. Therefore, N₂O formation is not as much an engine-out emissions issue as it is an aftertreatment one.

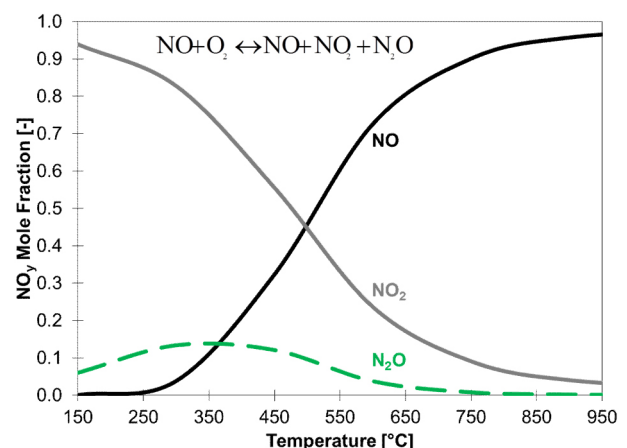


Figure 1. Thermodynamic equilibrium of nitrogen oxide species as a function of temperature. (Calculated using NASA CEA equilibrium code.6)

In general, three favorable modes can be identified for N_2O creation in a diesel aftertreatment system. The first referred to as “HC-DeNO_x” or “Lean Reduction” is a function of PGM containing oxidation catalyst such as a Diesel Oxidation Catalyst (DOC) where NO_x created through combustion process is reduced to N_2O through DOC by unburnt hydrocarbons used as reductant. Another known mode of formation is as the result of decomposition of surface nitrate species throughout the aftertreatment system. And thirdly, the formation of N_2O as the result of oxidation of ammonia, NH₃, through Ammonia Slip Catalyst (ASC) is fairly well known. Similarly, oxidation of NH₃ can take place within the SCR material regardless of the existence of an ASC. Each mode dominates based on the engine out NO₂ concentration, NH₃ concentration and excess and AT system temperatures.⁷⁻⁹

Krishna, et. al.⁷, identified the contribution of each aftertreatment component to the TP N_2O emissions based on conditions such as system temperature.⁴ They demonstrated that depending on the operating temperatures, one or another component dominates the formation of the N_2O and hence the actual mode under which the N_2O is formed.

Therefore, understanding of the actual mode of formation of N_2O is essential for the mitigation and reduction of its emissions. Decisions such as aftertreatment component type, PGM ratio and loading, architecture of the aftertreatment system and type of catalyst in addition to choices made in the operating strategies of the vehicle can change N_2O emissions significantly.⁸⁻⁹

In this study, attempts were made to look at the N_2O emissions from various products, architecture, catalyst type and operational strategy. Data have been collected and presented here in a normalized fashion in order to avoid disadvantaging or favoring a particular product based on their absolute emissions. Furthermore, the US06 cycle was selected as an example of a high load condition instead of a HFET as is the case for certification purposes, in order to create a comparable condition for all products without recreating the actual certification cycles.

EXPERIMENTAL

Details of Vehicles and System Configuration

Four diesel vehicles containing SCR aftertreatment components were purchased as they became commercially available. These vehicles represent various system architectures as intended for this study. As shown in [Figure 2](#), (Note: Schematics are generic and are NOT the true representative of the actual AT design of these vehicles) vehicle 1 and 2 have the following architectural configurations: Diesel Oxidation Catalyst (DOC)-Selective Catalytic Reduction (SCR)-Catalyzed Diesel Particulate Filter (CDPF), while vehicles 3 and 4 have the SCR downstream of the DOC and CDPF.

The SCR catalysts from vehicles 1 and 2 were analyzed and did not have any PGM containing zones, although the presence of a CDPF behind the SCR would be expected to play a role in the oxidation of NH₃ that might slip from the SCR under certain conditions. Vehicle 4's system components were analyzed and found to have a PGM containing zone on the rear of the SCR that would be expected to

serve as an Ammonia Slip Catalyst (ASC). Vehicle 3 also proved to have no slip zone on the rear of the SCR or any other provisions to mitigate NH₃ slip other than controls calibration.

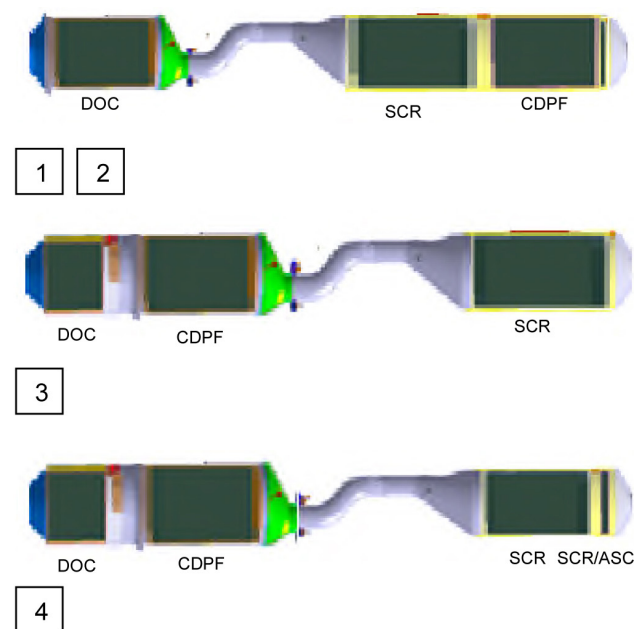


Figure 2. Generic schematic representation of the three different AT architecture from four different vehicles.

Effect of Aftertreatment's Architecture on TP N_2O Production

To obtain an understanding of the level of N_2O production from each vehicle, performance testing was conducted in a chassis cell with emission analyzers using identical procedure for all vehicles. The vehicles were prepared via a soot regeneration cycle, either stationary or running at a steady state speed depending on the options available on a scan tool, using a highway cycle. After cleaning the system, the SCR was saturated using the OEM's calibration through two UDDS cycles. This loading was followed by a minimum soaking of 12 hrs prior to emission testing using FTP-75 cycle followed immediately by a US06 cycle. In addition, in some cases 2 additional US06 cycles were run from a warm engine condition, where the final one was intended to represent ongoing high speed operation not influenced by NH₃ storage carried over from a colder cycle. The N_2O emissions of the FTP portion were weighted and the US06 portion reported directly.

The test cell utilized for this work employs a Horiba 150 HP single roll dynamometer and two Horiba 200 Series emissions benches calibrated for “modal” raw exhaust measurement. There is also a bag system that is used with another Horiba 200 Series emissions bench calibrated for the diluted samples. In this system, the exhaust flow calculation is based on a comparison of tailpipe “raw” CO₂ to tailpipe CO₂ “diluted” to generate a CO₂ dilution ratio to apply to the CVS flow value. At low flows and high air/fuel ratios such as idle on a diesel, this can lead to some exhaust flow measurement error, although effort was taken to minimize this effect. The two modal emissions benches were run in the engine out and dilution benches testing for this paper.