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Experimental approach concerning the selective catalytic reduction of NO_x for diesel engines of Romanian railway transport

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Transportation and its systems are one of the most used and important mobility systems worldwide, assuring the development of economy and society. For this reason it is important to have non polluting characteristics, especially in the case of being equipped with internal combustion engines (most diesel ones). In this case, it is extremely important to determine the chemical pollution produced by the large diesel engines and limit, by all means, the impact of the exhausted flue gas emissions on human health and environment. The Western part of Romania has the most developed rail network of the country concerning the diesel traction, and this article is focusing on *on line* NO_x concentration, generated by such diesel locomotives, used in the real regional railway transport. Representative measurements data set of NO_x from a diesel engine will be presented. Finally, the authors demonstrate that the SCR (selective catalytic reduction) after-treatment is a present necessity to reduce NO_x concentration and a best available technique to modernise the national railway fleet in Romania.

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1. Introduction

Modern civilization is characterized by two fundamental processes that generate pollution, namely: (i) industrialization and (ii) urbanization. Pollution is generated by continuous accumulation of dangerous pollutants from industrial activity & waste or household, by increasing noise pollution in industrial and urban areas, not at least by the traffic emissions, including engine equipped vehicles, and generating thus the degradation of natural landscape and human health, if legislative limits are not respected, in the exhaust. Among the many sources of pollution for air and water ecosystems, the road and railway transport, equipped with internal combustion engines are included. The transport sectors have registered an exponential development, first with the emergence of steam engine and, in following, the diesel engine were retrofitted. Historically, these have replaced, in turn, classic transport with animals. The vehicles powered by steam engines, and finally their replacement with diesel engine, were the result of the development of technique, generating not only technical performance, but also degradation of the environment.

It is known [1], that the air pollution is the main ecological impact environmental impact of transportation systems. Worldwide, means of transport, running with liquid fossil fuels, counts to contribute to air pollution and its quality degradation in large amounts, for example by 75 % with the dangerous carbon monoxide emissions, nitrogen oxides, and hydrocarbons species, not mentioning the particles [1]. The most important effects which transport has upon human health are related to the harmfulness exhaust gases that contain NO_x, CO, SO_2, CO_2, volatile organic compounds (VOC), and particle matter (PM), loaded with heavy metals (lead, cadmium, copper, chrome, nickel, selenium, zinc) [7]. The manner of coordinating and managing the transport system is also important for the environment. The biggest environmental effect produced by trains is the amount of carbon dioxide they emit. The carbon footprint left by a train compared to an automobile really depends on how many passengers are using it. A train full of passengers, for example, leaves a significantly smaller carbon footprint per capita than a car with just one person. A half-empty train, on the other hand, will leave a larger carbon footprint than a four-passenger carpool. Trains vary greatly in efficiency as well [23]. The comparison of a trip by train with one by car, for example, between Timisoara and Bucharest, applying the EcoPassaneger program [3] enables to calculate the energy consumption and exhaust atmospheric emissions; the results presented in Table 1 show that the train is less polluting then the car is, for similar conditions of freight.

Table 1. Energy consumption and exhaust atmospheric emissions.

Component	Unit	Train	Car
Carbon dioxide	kilograms	24.2	58.4
Energy resource consumption	Converted into liter petrol	13.7	27.3
Particulate matter	grams	9.3	18.8
Nitrogen oxides	grams	29.5	288.7
Nonmethane hydrocarbons	grams	2.6	28.2

Internal combustion engines emissions are harmful not only because their high noxious concentrations' amounts, but also because they are emitted at ground level, in close proximity to human habitat and activity zone, knowing that, in Romania, no special insulation zones, by vegetation, is limiting the dispersion of the gases from the mobile sources, neither on roads, nor on railway. Compounds formed in the exhaust gases contribute to air pollution both globally and locally, directly or indirectly causing chemical chain reactions in the atmosphere. Across the planet, increasing the concentration of the greenhouse gases (mainly CO_2, but not only) has led to climate changes, with unpredictable consequences on the environment, climate, and quality of life [1, 4, 5]. These are some resuming data arguments that emphasize the importance of studying the chemical pollution of air pollution produced by internal combustion engines. Although train travel generally is better for the planet than car travel, railroads do present a host of serious environmental problems. Most of these effects are considered localized issues, although their impact certainly is not limited to a particular region. Clean, efficient railway transportation may be crucial in the effort to reduce greenhouse gases and fuel consumption, but until this is achieved worldwide, trains will continue to pose a threat to the environment [2].

A new study compares the "full life-cycle" emissions generated by 11 different modes of transportation in the US [6]. Unlike previous studies on transport emissions, it focuses beyond what is emitted by different types of car, train, bus or plane while their engines are running and includes emissions from building and maintaining the vehicles and their infrastructure, as well as generating the fuel to run them. The presented figures (Table 1 Analysis components [6]) indicate that Railway modes have the smallest fraction of operational to total energy due to their low electricity requirements per PKT (passenger-kilometer-traveled) relative to their large supporting infrastructures. The construction and operation of rail mode infrastructure results in total energy requirements about twice that of operational [8].

Life-cycle NO_x emissions are often dominated by tailpipe components, however, autos and electric rail modes show non-negligible contributions from other components. Non-operational NO_x emissions are due to several common components from the supply chains of all the modes: direct electricity use, indirect electricity use for material production and processes, and truck and rail transportation. With on-road modes, electricity requirements for vehicle manufacturing and maintenance as well as truck and rail material transport are large contributors. Through the use of life-cycle environmental assessments, energy and emission reduction decision-making can benefit from the identified interdependencies among processes, services, and products. The use of comprehensive strategies that acknowledge these connections are likely to have a greater impact than strategies that target individual components [8].

Romanian railway transport started as one of the first in Europe, based on the steam locomotives (by 1854) and was continuously developed, covering also the transition from steam engines to diesel engines. Romanian diesel engine locomotives production started by 1959 under Swiss license and a group of companies such as Sulzer Winterthur as diesel engine manufacturer, BBC Baden as manufacturer of electrical parts and SLM Winterthur as manufacturer of mechanical parts, etc. The most representative model was the 1544 kW diesel-electric locomotive, type 060-DA. Based and inspired on this type, the Romanian engineers manufactured a new locomotive, a diesel-hydraulic LDH-125 type 040-DHC, equipped with a Sulzer engine of 919 kW. This type of locomotive was successful used on freight and passenger hauling but also on handling activities. Current operating requirements imposed on the hydraulic diesel locomotives LDH 1250 HP (920 kW) modernization started by 1999, and consisted of equipping them with the latest mechanical and electrical technological aggregates.

From the modernising over these 40 years old locomotives were retrofitted and replaced by new diesel hydraulic locomotive LDH 1000 kW, equipped with Caterpillar 3508 B engines, characterised by a higher installed power, a modern design, both inside and outside, being more powerful, and used for hauling train on the principal lines for fast and IC (Inter City) trains.

Although trains are by and large more fuel efficient than automobiles, they still consume a tremendous amount of non-renewable fuel each year. Diesel trains consume fuel based on stopping patterns, the speed or throttle at which they are travelling and even weight. High-speed trains consume more fuel than freight trains. Trains consume fuel not just when they are running; fuel also is required to construct and maintain them.

The paper presents profiles of on board measured NO_x concentration from the exhaust gases of the diesel engines of the Diesel-hydraulic locomotive LDH 125 modernized and the environmental impact, in respect to this type of locomotive and its engine. Also it presents a possibility to reduce the amount of exhaust NO_x, using a technology that is already state of art for stationary combustion chambers (boilers) and is currently introduced in the engine clean technology.

Nitrogen oxides (NO_x) are binary compound of nitrogen and oxygen, or mixture between them like: nitric oxide (NO), nitrogen dioxide (NO_2), nitrous oxide (N_2O), nitrate radical (NO_3), dinitrogen trioxide (N_2O_3), dinitrogen tetroxide (N_2O_4) and dinitrogen pentoxide (N_2O_5) [12]. In atmospheric chemistry, air pollution and related fields, nitrogen oxides refers specifically to NO_x (NO and NO_2) [9]. All are harmful for the air and ecosystem and generate acid rains, global and irreversible temperature increase as result of the greenhouse effect, climate changes, ozone depletion, etc [10]. Nitric oxide from combustion originates from two sources: atmospheric nitrogen (N_2 – is the most important component in air used as combustion air) and nitrogen from the fuel (N_{fuel}), mainly of organic origin [9, 10]. The latest is generating the fuel based NOx and the prompt NOx, respectively. The first is known as thermal mechanism.

The NO_x are produce during combustion, especially at high temperature, based on the air nitrogen was firstly described by Zeldovich, under the name of thermal generated NOx. The mechanism is described through the following elementary reactions [9]:

$$O_2 \rightarrow 2O \tag{1}$$

$$N_2 + O \rightarrow NO + N \tag{2}$$

$$O_2 + N \rightarrow NO + O \tag{3}$$

$$OH + N \rightarrow NO + H \tag{4}$$

The equations (2), (3) and (4) are known as the extended Zeldovich mechanism: N_2 in the presence of elementary oxygen (O) reacts and NO and atomic non stable N result. The next step reaction is between the molecular nitrogen (N) and O_2, resulting NO and O. Thus the chain reaction starts again.

The nitrogen atoms released during the reaction (2) are after that oxidized to nitric oxide, mainly by a hydroxyl radical (OH), through reaction (4). Reaction (2) needs very high activation energy and this is a factor limiting for the reaction rate, being also extremely sensitive to temperature. For this reason the nitric oxide formed according to the Zeldovich mechanism is commonly known as **thermal NO**, mostly activated over 1200-1300 degree C [13].

The thermal NO formation rate on Otto engines is not significant on temperature below 1700 K, but became strongly accelerated on temperature over 2000 K by λ=1,1 when the equilibrium concentration is reached. This rate can be reduce by lowering and controlling the temperature by direct steam injection into the cylinder dilution of the intake air with re-circulated exhaust gases, and humidification of the intake air with steam, late injection [9].

The proportion of NO_2 from total NO_x emissions to the gasoline engine is 1-10% and from diesel engines is 5-15%. Inside the engine, NO_2 and OH radicals are obtaining from NO by reaction with HO_2. The most likely equation is:

$$NO + HO_2 \rightarrow NO_2 + OH \tag{5}$$

At ambient temperature, the chemical equilibrium is almost completely to NO_2. The NO is reacting with ozone in the presence of light and therefore the equilibrium will be establish after a few hours or days – depending on the environmental conditions– established [9].

Prompt NO. Hydrocarbons radicals react with molecular nitrogen to form hydrogen cyanide and atomic nitrogen (6). This atomic nitrogen reacts with hydroxyl groups (7) or radicals in the flame to form NO and atomic hydrogen [11]:

$$N_2 + CH \rightarrow HCN + N \tag{6}$$

$$N + OH \rightarrow H + NO \tag{7}$$

If oxygen is present, the hydrogen cyanide (HCN) and the nitrogen atom (N) produced in the reaction react further to nitric oxide through several reaction phases. The main reaction sequence is:

$$HCN \xrightarrow{+O} NCO \xrightarrow{+H} NH \xrightarrow{+H} N \xrightarrow{+O_2,+OH} NO \tag{8}$$

The formation of nitric oxide is usually very fast, and the nitric oxide formed is therefore called prompt NO. In contrast to thermal NO, fast NO depends only slightly on temperature. In diesel engines, the contribution of prompt NO to the total NO emission is estimated to be minor, below 5% [10].

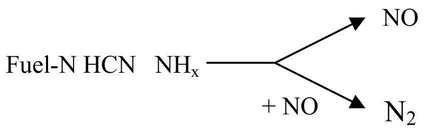
Nitrous oxidex (NO) are, according to the third mechanism for reaction of molecular nitrogen to nitric oxide between atomic oxygen (O) and N_2, forming also an unstable gas (N_2O, „laughing gas") according to the following reaction:

$$O + N_2 + M \rightarrow N_2O + M \tag{9}$$

where M represents any gas component. N_2O is a very dangerous (240 times more aggressive than CO_2) green gas. Further more, the „laughing gas" formed reacts again, either back to N_2 or to NO, depending on conditions. When the air ratio and temperature increase, the formation of nitric oxide also increases. The main reaction to nitric oxide is then [10]:

$$N_2O + O \rightarrow 2NO \tag{10}$$

Fuel NO. The nitrogen in the fuel is first converted to hydrogen cyanide, which then reacts to form NH species like ammonia and finally NO and N_2 [11].



Fuel NO is slightly dependent on temperature, and nitric oxide is easily formed from fuel-nitrogen at low temperatures too, below 1100 K [9].

From this NO forming methods, the most important is thermal; the others have relatively minor importance.

The preventive solutions to reduce thermal NO_x consist of influencing the temperature, residence time and oxygen content in the combustion area, or by secondary methods, referring to catalytic procedures. Avoiding the prompt NO_x is not very much to be influenced, literature indicates to use residual hydrocarbon radicals as reducers. Again a catalytic combustion represents a solution. For reducing the fuel originated NO_x hydro-denitrification [11] is recommended.

Environmental regulations and concern about clean air impact all of us. Protecting and preserving the environment is a core value by 2010 as the trend is putting into function only engines will achieve near-zero emissions of NOx, a greenhouse gas and smog causing compound. Two emissions technologies will be available, Selective Catalytic Reduction (SCR) and in-cylinder, Exhaust Gas Recirculation (EGR). Generally speaking, the SCR system operates downstream from the engine, removing much of the stress and heat rejection related to EGR-only engines to improve reliability and increase fuel economy. SCR is very simple. There are four major components to the SCR system: a Diesel Exhaust Fluid (DEF) tank, a DEF dozer, the SCR catalyst, and an ammonia (NH_3) system. There exist also other possibilities of DEF (basically with the compound NH_i). The components are all integrated into the exhaust system. Small amounts of DEF are injected into the catalyst where it is mixed and reacts with the NO_x found in the exhaust to produce nitrogen gas and water vapour, both of which are harmlessly released into the atmosphere through the vehicle's tailpipe. Compared to former technologies, for ex. the PACCAR technology applied in America mostly, according the emissions level, all engines using SCR after-treatment do not require significant increases in EGR flow rates to meet present NO_x limits. Because the pollutants are reduced to near-zero levels within the exhaust stream, an engine with SCR after-treatment will operate cooler, cleaner and more efficient than engines with higher EGR levels.

The catalyst is a material that increases the rate (molecules converted per unit time) of a chemical reaction while itself not undergoing any permanent change. For example, onto a specific sites on a platinum (Pt) catalyst and rapidly convert to carbon dioxide and water [8].

$$C_2H_4 + 3 \cdot O_2 \xrightarrow{catalyst(Pt)} 2CO_2 + 2 \cdot H_2O \qquad (11)$$

The SCR reactions takes place on temperature above a *limit* and in-between a window. For ex above 200 °C best results are obtained; and on temperature below 200 °C NH_3NO_3 can be explosively produced [8].

Another possibility for the additive is a combination, such as the known AdBlue, that is the registered trademark for AUS32 (Aqueous Urea Solution 32.5%) and is used in a process called Selective Catalytic Reduction (SCR). AdBlue consists of 32.5% of high purity urea and 67.5% of de-ionised water. The AdBlue trademark is currently held by the German Association of the Automobile Industry.

The SCR technology is still ongoing to be optimised for diesel engines, event the technology is studied from already 15 years in the automobile branch production. For ex. [14], some reasons for introducing the catalyst are given, according which temperature range, rapidity and simultaneous possibility to reduce other pollutants, as well, are the strengthen of the technology. Urea–selective catalytic reduction (SCR) has been a leading contender for removal of nitrogen oxides (deNO_x) from diesel engine emissions (according Urea thermolysis and NOx reduction with and without SCR catalysts [15]. Despite its advantages, the SCR technology faces some critical detriments to its catalytic performance such as catalyst surface passivation (caused by deposit formation) and consequent stoichiometric imbalance of the urea consumption. Deposit formation deactivates catalytic performance by not only consuming part of the ammonia produced during urea decomposition but also degrading the structural and thermal properties of the catalyst surface.

2. Experimental

The experiments were carried out on two levels:

1. On line on running engines in traffic (locomotives),
2. On line on a test rig, in laboratory conditions.

The experiments were run on an engine test bench - vehicle test rig (dynamometer) under real conditions. The on line measurements were performed on a 200 kW diesel engine with SCR system (Fig. 1).

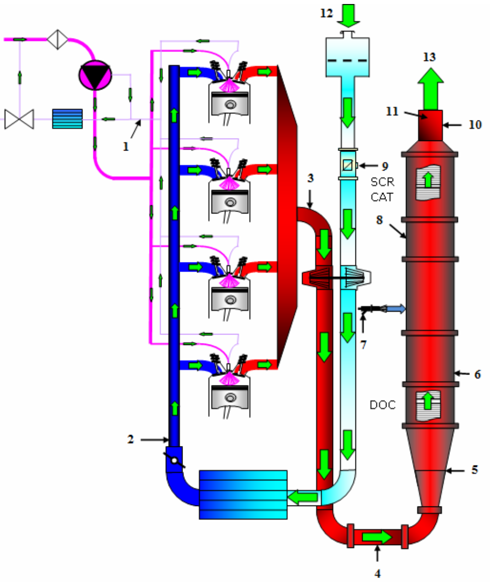


Fig. 1. Engine test bench configuration: 1-fuel circuit; 2-intake; 3-exhaust; 4-NOx raw; 5-temperature DOC up; 6-temperature DOC down; 7-AdBlue injection system; 8-pressure, temperature, and NO_x SCR up; 9-mass air flow;10- pressure, temperature, and NO_x SCR down; 11-presure SCR down; 12-air filter; 13-exhaust evacuation.

3. Results and discussion

The tests that have been made were NRTC (Non Road Transient Cycle) cycle according to the European STAGE norms [16]. Also test have been made on diesel hydraulic locomotive LDH 1000 kW on two different routes.

NRTC cycles were run twice, once with the cold engine and one with warm engine. AdBlue dosing started at temperature above 210 °C and α=1 (α is the injected ratio NH_3/NO_x).

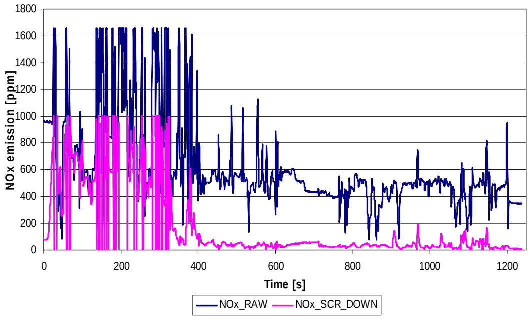


Fig. 2. NRTC cold run.

From the cold run presented in Fig. 2 ca be observed that in the first part of the graph, values of NOx concentration are similar in both points of measurement followed by an instability zone caused by fluctuations of NO_x concentration and the amount of AdBlue injected. After about 400 seconds NO_x emission values become more uniform due balance between NO_x emission and AdBlue injected, but also due to the presence of this substance in the SCR CAT. During this run the average NO_x reduction value resulted is 77.25 %.

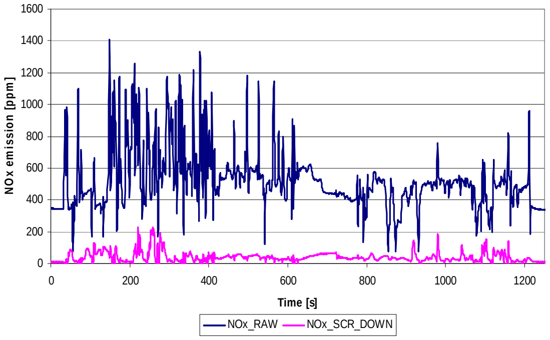


Fig. 3. NRTC warm run.

The hot engine functioning has a major influence on NO_x emissions, fact that can be seen in Fig. 3. This phenomenon is due to lower absorption of heat by the engine block (cylinder walls and cylinder head), the heat produced by compressing fuel is used in the combustion process, comparing with the first run where a significant part of this heat was transferred to the engine block causing an incomplete burn and as result higher NO_x emission are formed. Emission values of NO_x from warm run were reduced up to 96 % by mass.

The efficiency of SCR system applied on diesel engines was proved also in his researches by Dr. Eberhard Jacob [17], under similar conditions.

In order to conclude about the necessities for the Romanian diesel locomotive fleet, one run also several experiments for NOx measurement under real functioning. The measurements from the locomotive are divided in two operating modes: static and dynamic. First one was made in locomotives depot, andfive loading was applied for three times. The results are presented in Fig. 4.

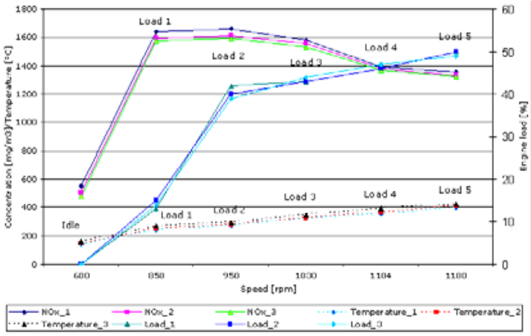


Fig. 4. NO_x concentration, temperature, and engine load variation.

The NO_x concentration is higher on the start point after that it is going down, because of the turbo system that raise the intake air pressure and the combustion is more complete resulting less NO_x.

The second operating mode, meaning dynamic conditions, was accomplished between to railway station from the Western part of Romania, on five different days. The measurements resulted are presented in Fig. 5.

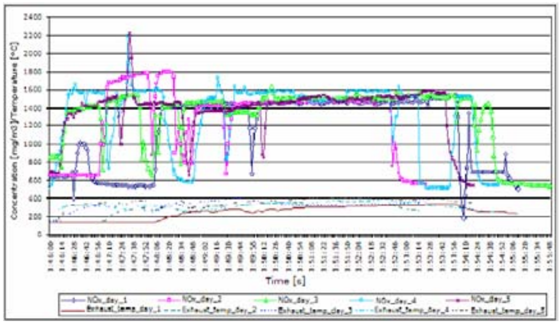


Fig. 5. NO_x concentration and temperature variation.

The train was a two passenger cars with a total tonnage of 96 tones, the maximum velocity was 50 km/h. Analyzing the data resulted that more than 85 % of the traveling time, the NO_x concentration were above 1,400 [mg/m^3] and the temperature average was over 325 °C. It is tremendous necessary to reduce these large NO_x values and thus the reduction technology is proposed.

SCR system efficiency was studied also by Wilfried Müller [18] and Christopher S. Weaver [19]. They proved that an SCR installed on a locomotive reduced the NO_x level by 80 %. Considering in comparison the level of exhaust concentrations obtained from the tested diesel engine having a SCR system and the data obtained in real conditions from diesel-hydraulic locomotive NO_x (concentration and exhaust temperature) one can conclude that a SCR system is the appropriate solution to reduce NO_x concentration for the retrofitting of the Romanian locomotives running diesel engines. It is absolutely necessary to implement such technologies, that must reduce the NOx concentrations more than by 90 -95 % by mass, in order to meet the environmental limits for function.

4. Conclusions

SCR is a method for converting nitrogen oxides, NO_x, through a catalyst in nitrogen, N_2 and water, H_2O. The conversion reduction takes place in the presence of a reducing gas, which is based on NH_i. Usually urea solution (32.5%) and high purity water (distilled water) known as AdBlue is industrially used, but other solutions such as anhydrous ammonia, aqueous ammonia which are sprayed into the exhaust gas flow and are absorbed into the catalyst, are also possible.

From the beginning, this method had a relatively wide range of application such as industrial boilers, solid waste incinerators, combustion plants, industrial gas turbines and engines which has proved effective, with a percentage reduction of emissions NO_x between 70-95 % by mass. In this context, the application of the technology for engines occur problems with the necessity of space, as all parts of the SCR system, from reducing agents that has to be stored in separate tank and injected with a performant injection system into exhaust flow, must find the proper space and shape.

As result of implementing the SCR methods for the denox of the flue gases from the diesel engines of locomotives in Romania, the air quality in the vicinity of the vehicle daily run is enhanced, and the maximum values for the national and international standards (emission and imission) are more probable respected. As result also air quality standards in the vicinity of railway cross roads is of better quality.

European legislation requires vehicle manufacturers to use this technology to reach the reduced levels of pollutants. The use of AdBlue helps not only the environment by reducing the emissions created by diesel engines, but it also helps transport companies and drivers reduce fuel costs, by reducing fuel consumption [20, 21, 22].

As main disadvantage of the technology, the toxicity of the reducing agent is noted. AdBlue is very susceptible to contamination from foreign matter as well as incorrect material selection. Therefore appropriate equipment should be used to handle AdBlue correctly and increase the lifespan of your SCR system.

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