

# Evaluation of NH<sub>3</sub>-Slip from Diesel Vehicles Equipped with Selective Catalytic Reduction Systems by Neural Networks Approach

Mona Lisa M. Oliveira, Nara A. Policarpo, Ana Luiza B. P. Barros, Carla A. Silva

**Abstract**—Selective catalytic reduction systems for nitrogen oxides reduction by ammonia has been the chosen technology by most of diesel vehicle (i.e. bus and truck) manufacturers in Brazil, as also in Europe. Furthermore, at some conditions, over-stoichiometric ammonia availability is also needed that increases the NH<sub>3</sub> slips even more. Ammonia (NH<sub>3</sub>) by this vehicle exhaust aftertreatment system provides a maximum efficiency of NO<sub>x</sub> removal if a significant amount of NH<sub>3</sub> is stored on its catalyst surface. In the other words, the practice shows that slightly less than 100% of the NO<sub>x</sub> conversion is usually targeted, so that the aqueous urea solution hydrolyzes to NH<sub>3</sub> via other species formation, under relatively low temperatures. This paper presents a model based on neural networks integrated with a road vehicle simulator that allows to estimate NH<sub>3</sub>-slip emission factors for different driving conditions and patterns. The proposed model generates high NH<sub>3</sub>slips which are not also limited in Brazil, but more efforts needed to be made to elucidate the contribution of vehicle-emitted NH<sub>3</sub> to the urban atmosphere.

**Keywords**—Ammonia slip, neural-network, vehicles emissions, SCR-NO<sub>x</sub>.

## I. INTRODUCTION

ROAD transportation has contributed to increase emissions of conventional air pollutants (i.e., NO<sub>x</sub> – nitrogen oxides, HC - hydrocarbons, PM – particulate matter, NH<sub>3</sub>) and, consequently, to the increase of problems associated with the environment and human health [1], [2]. For example, the vehicle fleet in Ceará state, Brazil, has grown 180% over the last ten years. Fortaleza city is the capital of the state of Ceará, and has the seventh largest vehicle fleet in the country (387 vehicles per 1,000 inhabitants). At present, the vehicle fleet in Fortaleza Metropolitan Area (FMA) accounts more than 1 million vehicles. Totally, 8.4%of them are fueled with diesel (today with 8% v/v of biodiesel into mineral diesel) and 16% of these diesel vehicles have been fitted with NO<sub>x</sub>

aftertreatment systems since 2012 [3]-[5]. Although emission limits have become increasingly strict in the past years, road transport remains the most significant source of urban air pollution in Europe with respect to NO<sub>x</sub>, HC, and PM [22]. Furthermore, the presence of oxygen molecule in biodiesel might cause an increase in combustion gas temperature, which leads to an increase in NO<sub>x</sub> emissions, rising the flame temperature; this oxygen reacts with nitrogen and tends to form NO<sub>x</sub> [5], [20].

The Selective Catalytic Reduction (SCR) systems for NO<sub>x</sub> reduction by NH<sub>3</sub> has been the chosen technology by most of diesel-cycle bus and truck manufacturers in Brazil to meet the PROCONVE P7 [13] standards (EURO V equivalent). Significant NO<sub>x</sub> reduction is required, which demands higher amounts of NH<sub>3</sub>, increasing its probability to slip to the environment. Thus, more complex aftertreatment systems have been needed to prevent the NH<sub>3</sub>-slip. Two forms of NH<sub>3</sub> could be used in SCR systems: (i) pure anhydrous NH<sub>3</sub>, and (ii) aqueous NH<sub>3</sub>. Anhydrous NH<sub>3</sub> is toxic, hazardous, and requires thick-shell, pressurized storage tanks and piping due to its high vapor pressure. Aqueous NH<sub>3</sub> is less hazardous and easier to handle. Therefore, emission of NO<sub>x</sub> is reduced when introducing a reducing agent into the exhaust gases to convert NO<sub>x</sub> into harmless nitrogen and water, over an SCR catalyst. The commercialized SCR catalysts used in heavy-duty vehicles are monoliths catalyst based in V<sub>2</sub>O<sub>5</sub>-WO<sub>3</sub>-TiO<sub>2</sub> (vanadium pentoxide-tungsten trioxide-titanium dioxide) [5], [6]. The SCR catalytic formulation is coated on a ceramic or metallic catalyst support encased in a stainless steel cylinder. Both precious metal and base metals can be used for an exhaust temperature range of 230 °C to 430 °C. Due to its toxicity, a vanadium-based SCR catalyst is expected to be removed from the heavy-duty diesel market within few years. Almost all major heavy-duty vehicles manufacturers have decided to use this technology to meet the new emission legislation on NO<sub>x</sub> emissions. This reaction usually requires a urea solution as a reducing agent (~32.5 % wt. urea), with brand names such as AdBlue® (in Europe) or Arla® (in Brazil).

The urea consumption can vary from 1 – 5% v/v relative to diesel fuel consumption (average consumption of aqueous NH<sub>3</sub> is about 0.1 liters per 100 kilometers). If the NH<sub>3</sub> /urea introduced into the system does not match the NO<sub>x</sub> to be converted, then there will be some NH<sub>3</sub> slip. In practice, slightly less than 100% of the NO<sub>x</sub> conversion is usually targeted [7], [8], so that the aqueous urea solution hydrolyses

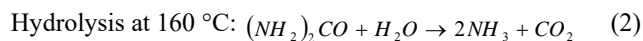
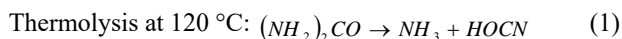
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to  $\text{NH}_3$ , under relatively low temperatures (via formation of cyanic acid –  $\text{HOCN}$ ), as shown in (1) and (2) [9]:



$\text{NH}_3$ -SCR provides a maximum efficiency of  $\text{NO}_x$  removal if a significant amount of  $\text{NH}_3$  is stored on its catalyst surface. This could produce  $\text{NH}_3$  desorption during high temperature lapses since it affects the catalyst storage capacity. Furthermore, in some conditions, over-stoichiometric  $\text{NH}_3$  availability is also needed, which increases the  $\text{NH}_3$ -slip even more. It should be noted that  $\text{NH}_3$  is an aggressive gas to the eyes, skin, and respiratory trajectory. It has a maximum allowable concentration in air of 20 ppm.  $\text{NH}_3$  can react with nitrate and sulphate in the exhaust gas and form respectively ammonium nitrate and sulphate that are emitted as secondary particles [10], [11].

The measurements also indicate that the  $\text{NH}_3$  is a precursor of fine particles, or  $\text{PM } 2.5 \mu\text{m}$ , which deteriorates urban air quality, affects human health and impacts the global radiation budget since vehicles have been important sources of  $\text{NH}_3$  in urban areas [2]. Experimental dynamometer  $\text{NH}_3$  slippage has presented average values of 10 ppm and maximum values of 100 ppm, as reported in literature. The adoption of SCR

system by the addition of urea or  $\text{NH}_3$  to diesel exhaust to meet  $\text{NO}_x$  emission standards could be also resulting in elevated  $\text{NH}_3$  emissions from traffic. From this time, Carslaw and Rhys-Tyler [11] reported that  $\text{NH}_3$  emissions in the United Kingdom are most important for older generation catalyst-equipped petrol vehicles and SCR-equipped buses [11]. More efforts are still needed to elucidate the contribution of vehicle-emitted  $\text{NH}_3$  to the urban atmosphere. Thus, in this work, emission estimates were evaluated for mobile emissions of  $\text{NH}_3$ -slip from diesel vehicles when these are equipped with SCR- $\text{NO}_x$ . Neural networks have been used to model the system. A model based on neural networks integrated with a road vehicle simulator that allows to estimate  $\text{NH}_3$ -slip emission factors for different powertrain configurations along different driving conditions was proposed.

## II. METHODOLOGIES

The evaluation of  $\text{NH}_3$ -split from heavy-duty vehicles with SCR- $\text{NO}_x$  systems using neural-networks was done according to a rigorous procedure well documented in literature [6], [9]. Accordingly, the evaluation of the  $\text{NH}_3$  slip (in ppm or g/kWh) was carried out in a vehicle equipped with a SCR system under several traffic conditions, including one standard cycle as the Urban Dynamometer Driving Schedule - UDDS (Fig. 1) and real circuits measured both in Fortaleza-Brazil and Porto-Portugal cities (Fig. 2).

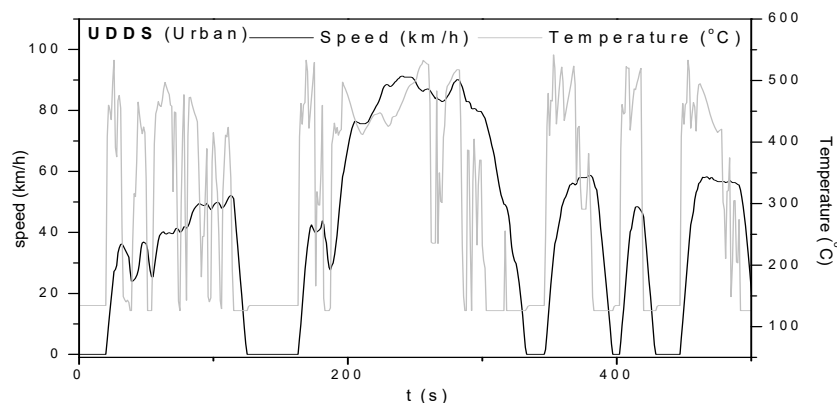


Fig. 1 Urban Dynamometer Driving Schedule (UDDS) driving cycle

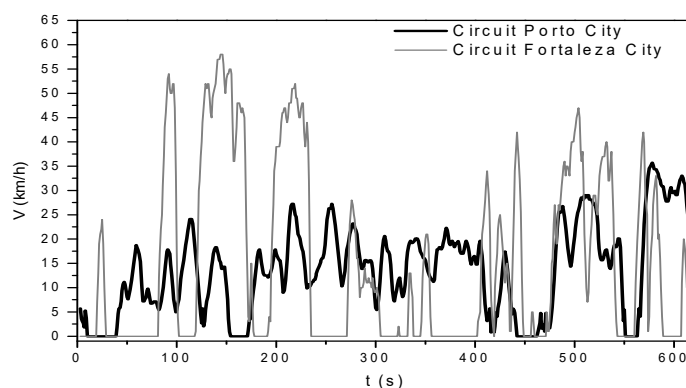


Fig. 2 Real driving cycle measured in Fortaleza and Porto cities

### A. Emissions Regulations for Diesel Vehicles

In Brazil, Euro V standard was fully applied in January 2012, setting new upper bounds for heavy duty vehicles. Therefore, the SCR system for NO<sub>x</sub> reduction by nitrogen compounds, such as NH<sub>3</sub> or urea, commonly referred to as simply “SCR”, has been developed for and well proven in industrial stationary applications. NO<sub>x</sub> maximum emissions are specially restricted, which leads to work under over-stoichiometric NH<sub>3</sub> conditions. It increases the chances for NH<sub>3</sub> to slip, requiring an assessment of NH<sub>3</sub>-slip when it does not have an exact catalyst to mitigate this exhaust gas. Euro V engines have had 60% lower NO<sub>x</sub> emissions. At present, the actual European standard is the Euro VI. This fact has driven to new emission standards, forcing a reduction of 2 g/kWh in

NO<sub>x</sub> emissions for heavy-Duty Diesel vehicles, as well as in Brazil [12]-[14].

### B. Diesel Fleet Emissions: Fortaleza Metropolitan Area

In 2015, the total fleet of heavy-duty diesel trucks in FMA was more than 5,000 vehicles, 78% of which Fortaleza alone accounted. Only 16% of this fleet was fitted with SCR-NO<sub>x</sub> systems as of 2012 [4]. This fact might reflect directly on the reduction of NO<sub>x</sub> and PM emissions for this category, as shown in Fortaleza in Figs. 1 and 2 for NO<sub>x</sub> in 2010 and 2015, respectively. The emission decreases in recent years have been affected due to the introduction of aftertreatment systems into exhaust gases, as well as, the better quality of fuels [6], [15].

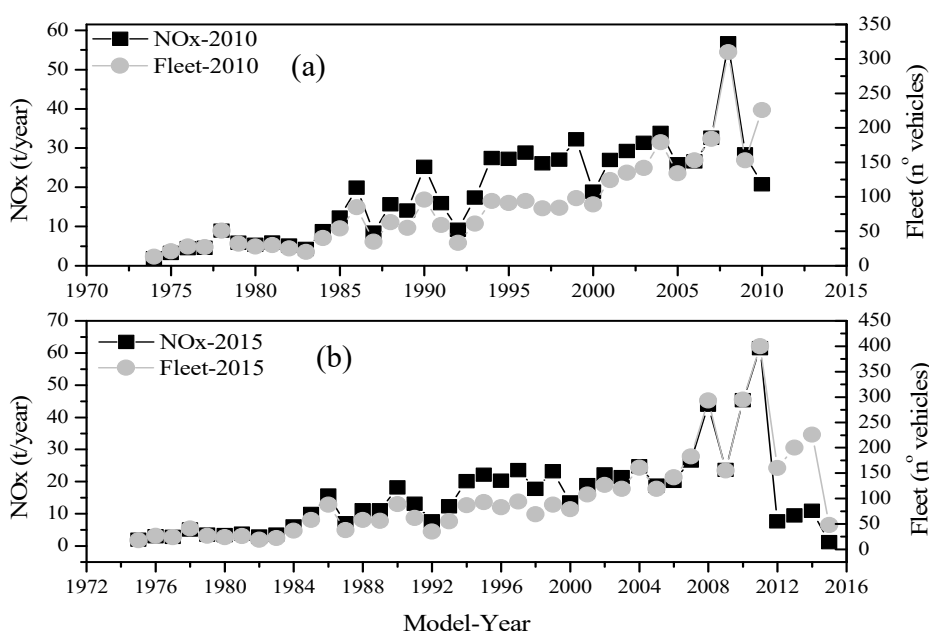


Fig. 3 NO<sub>x</sub> emissions and diesel vehicular fleet vs. model-year for the Fortaleza city/Brazil: (a) for year 2010 and (b) for year 2015

### C. NH<sub>3</sub>-slip by ANN Approach and ADVISOR Integration

The ANN was developed using the experimental dataset from typical exhaust gas characteristics of Diesel engines. The ANN was successfully trained accordingly to experimental data in SCR-NO<sub>x</sub> systems in order to predict transient changes in NO<sub>x</sub> emissions level in Diesel vehicle for operating conditions according to loads/engine speeds observed in driving cycles. NH<sub>3</sub> slippage heavy-duty vehicles along the standardized (UDDS) driving cycles and real measured driving cycle (in Fortaleza and Porto Cities), according to the model presented by Oliveira et al. [6]. Moreover, it was compared the performance of the other SCR catalysts investigated for UDDS and real driving cycle.

In this research, it was also used ADVISOR Version 2002, ran in the MATLAB® environment with Simulink, which is publicly available. ADVISOR does not cover any SCR-NO<sub>x</sub> model [6], [16], [17].

ADVISOR integration was available to ANN as an important simulation tool to evaluate the performance of

vehicles equipped with SCR systems that use other catalysts, as well as possible to study the NH<sub>3</sub>-slip in driving cycles for Diesel vehicles. The relevant seven inputs were considered for the NH<sub>3</sub>-slip output: space velocity (SV, h<sup>-1</sup>), exhaust gas temperature (T, °C), oxygen concentration (O<sub>2</sub>, v/v%), water concentration (H<sub>2</sub>O, v/v%), sulphur dioxide concentration (SO<sub>2</sub>, ppm), NO<sub>x</sub> concentration (ppm), NH<sub>3</sub> concentration initial (NH<sub>3</sub>, ppm) (Fig. 4). The stochastic back propagation with the Levenberg-Marquardt algorithm (MATLAB®) was used to train the network [6], [9].

The SCR ANN was tested using 15% of data points measured by Oliveira et al. [6]. The other 85% of data (600 experimental points for each catalyst teste) were used to train and to validate several Network. The model is continuously updated with actual component test data from users and university validation efforts. It is flexible enough to model specific components and vehicle configurations for the needs of most users [21]. Taking all this into account, ADVISORs integration with the available ANN is an important simulation

tool to evaluate the performance of the vehicles equipped with  $\text{NH}_3$ -slip in driving cycles. Thus, it was possible to study the emissions

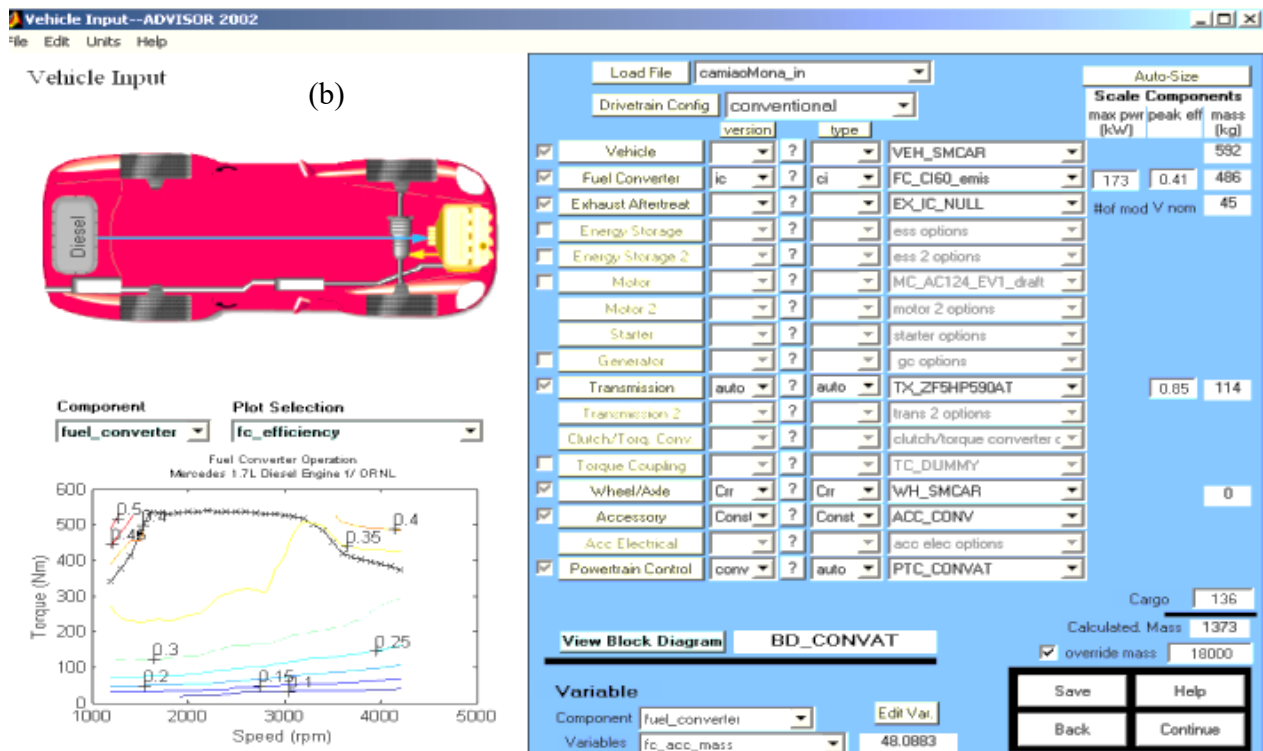
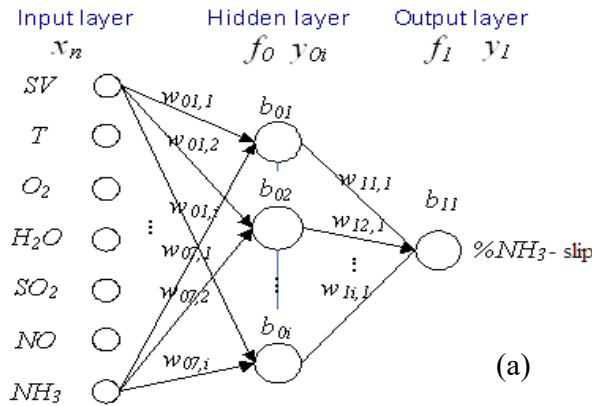


Fig. 4 (a) ANN architecture of the SCR model to  $\text{NH}_3$ -Slip and (b) ADVISOR integration tools

### III. RESULTS AND DISCUSSION

The correlation coefficients ( $R^2$ ) between measured data and test, training and validation data are always higher than 0.70. This indicates that the developed ANN-based model can make good predictions of  $\text{NH}_3$ -slip on SCR systems for a Diesel vehicle. These  $R^2$  values show a good adequacy of the developed mathematical correlations, for all studied catalysts (Table I), as reported by Oliveira et al. [6], [9] for % NOx conversion.

Simulated and measured data tests for  $\text{NH}_3$  slippage, for all studied catalysts, are presented in Fig. 5. Generalization performance is reasonable, indicating confidence in the training results and produced fairly accurate forecasts.

TABLE I  
 R2 VALUE FOR THE TEST, TRAINING AND VALIDATION DATA

Correlation coefficients ( $R^2$ )	Catalyst	$\text{NH}_3$ - Slip
Test	CATCO	0.80
	FeZSM5	0.72
	CuZSM5	0.80
Training	CATCO	0.82
	FeZSM5	0.76
	CuZSM5	0.90
Validation	CATCO	0.80
	FeZSM5	0.72
	CuZSM5	0.70

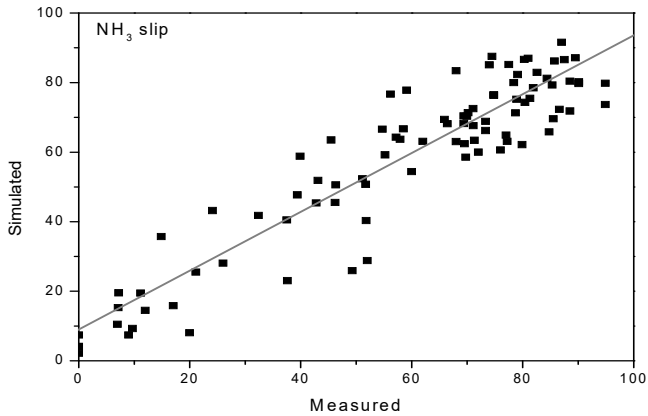


Fig. 5 Simulated and measured data for the test values of  $\text{NH}_3$ -slip for commercial catalyst (CATCO)

Emissions from diesel vehicles also can be expressed in grams of pollutant per unit of travelled distance (g/km) or

ppm. Thus,  $\text{NH}_3$ -slip (ppm) data after the SCR system and the exhaust gas temperature are obtained for all the studied catalysts for UDDS driving cycle for heavy-duty vehicle. For all catalysts used in this study, a significant concentration of  $\text{NH}_3$ -slip during a sample (500 s) for real circuit measured is shown in the Figures below. The maximum  $\text{NH}_3$ -slip concentration was estimated, approximately, 700 ppm for catalyst based in ZSM5 zeolite. Similar results were obtained for the remaining cycles and are summarized by Suarez-Bertoa et al. [2]. For all catalysts, a significant concentration of  $\text{NH}_3$ -slip during a sample of 500 s for real circuit is shown in Figs. 6 and 7.

The minimum  $\text{NH}_3$  slip concentration was estimated, below 100 ppm for CATCO, while FeZSM5catalyst exhibited 250 ppm of  $\text{NH}_3$  slippage. CuZSM5 catalyst showed maximum value up to 550 ppm. Similar results were obtained by Suarez-Bertoa et al. [2].

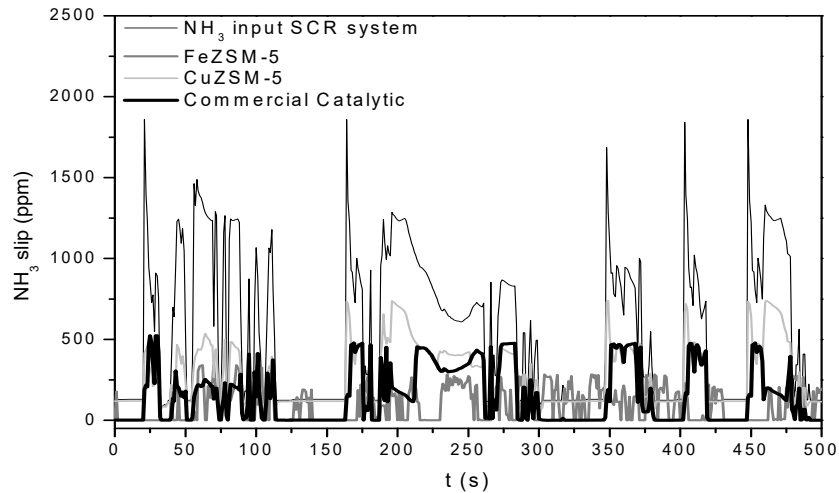


Fig. 6 Porto city real measured cycle when used in heavy-duty vehicle for  $\text{NH}_3$ -slip for metal-zeolite and commercial catalyst

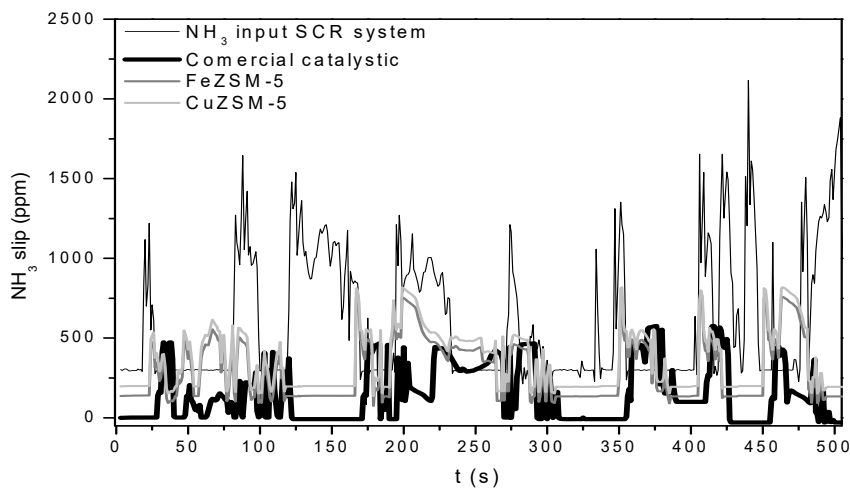


Fig. 7 Fortaleza city real measured cycle when used in heavy-duty vehicle for  $\text{NH}_3$ -slip for metal-zeolite and commercial catalyst

In the Porto cycle research, it was obtained a 100 ppm average and a maximum value of 500 ppm of  $\text{NH}_3$  that passed

through the SCR without reaction. It is noteworthy that the Fortaleza circuit shows slightly less than this value. At low exhaust temperatures, catalyst activity also falls sharply. This zeolite catalysts' behaviour could be explained not only by the support characteristics (i.e. acidity, framework structure, Si/Al ratio), but also by the metal ion-exchange. Consequently, this integration and simulation demonstrated to be a potential application tool in the development of emissions control strategies and emissions compliance or in the driving cycle test or real circuit validation of diesel vehicles. Fe-zeolites seem to be the best candidates for CATCO replacement. They reveal lower values of NH<sub>3</sub> slip tailpipe emissions.

In general, the results of the research show that the correlations obtained with ANN integrated with a road vehicle simulator, produce average NH<sub>3</sub> slippage results for commercial vanadium based catalyst consistent with experimental data and can be one more powerful tool for SCR simulation of diesel vehicles equipped with advanced after treatment systems, also studied by Faghihi and Shamekhi [18].

Emissions for diesel vehicles from NH<sub>3</sub>-slip are shown in Table II by grams of pollutant per unit of travelled distance. It is also assumed the average of % NO conversion for all catalysts studied to present UDDS standard and real circuit cycles.

TABLE II

PERFORMANCE OF THE SCR SYSTEM AND NH<sub>3</sub>-SLIP FOR HEAVY-DUTY DIESEL VEHICLE

Catalyst	Cycles Driving	% NO Conversion Average	NH <sub>3</sub> -slip (g/km) Average
	UDDS	89.5	2.29
CATCO <sup>(1)</sup>	Real Urban Circuit of Fortaleza city	87.3	2.62
	Real Urban Circuit of Porto city	83.7	2.97
	UDDS	77.7	2.30
FeZSM5 <sup>(2)</sup>	Real Urban Circuit of Fortaleza city	72.5	3.03
	Real Urban Circuit of Porto city	70.1	4.12
	UDDS	27.0	4.10
CuZSM5 <sup>(3)</sup>	Real Urban Circuit of Fortaleza city	25.6	5.07
	Real Urban Circuit of Porto city	23.0	6.40

<sup>(1)</sup> Commercial catalyst, <sup>(2)</sup> iron zeolite catalyst and <sup>(3)</sup> copper zeolite catalyst

These driving cycles were used in the present work because contrasting with the remaining cycles, they are very demanding in terms of accelerations rates, low average speed, aggressive topography and ambient temperature [19]. Complementing, these results show the performance of the SCR systems concerning the NH<sub>3</sub>-slip (g/km) for the remaining (non-homologation) driving cycle simulated. Moreover, the remaining catalysts exhibited high NH<sub>3</sub>-slip. Some authors have attributed this difference of behavior to characteristics of catalysts in function of temperature or probably to the reach of the maximum absorption limit of NH<sub>3</sub> and characteristic of the real circuit presented, as shown in both Fortaleza circuit (2.62-5.07 g/km) and Porto circuit (2.97-6.40 g/km) tested. The performance of SCR systems can

be improved with thermal management to increase exhaust temperatures [20]. Start-stop systems are also effective at keeping the SCR system warm by avoiding the cooler exhaust temperature of idling conditions [7], [9], [16].

#### IV. CONCLUSION

The Euro V standard fixes the maximum levels of NO<sub>x</sub> released by heavy duty vehicles and it was being fully applied in Europe since January 2012, while the Brazilian equivalent regulation was established only in January 2014. NO<sub>x</sub> emission is especially further restricted, which needs more severe urea injection in order to let the NH<sub>3</sub>-SCR storing significant amount of NH<sub>3</sub> for NO<sub>x</sub> reduction. In general, the results of the research show that the correlations obtained with ANN integrated with a road vehicle simulator produce average NH<sub>3</sub> slippage results for commercial vanadium based catalyst consistent with experimental data and can be one more powerful tool for SCR simulation of diesel vehicles equipped with advanced after treatment systems. The minimum NH<sub>3</sub> slip concentration was estimated; below 100 ppm for commercial catalytic and FeZSM5 zeolite catalytic exhibited 250 ppm NH<sub>3</sub>-slip, while CuZSM5 showed maximum value up to 550 ppm. Similar results were obtained for the remaining tests.

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