

Diesel aftertreatment control technologies in underground mines: the NO₂ issue

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ABSTRACT: Extensive use of diesel-powered equipment makes reduction of underground miner exposure to diesel particulate matter and gaseous emissions from diesel-powered equipment a major challenge for the mining industry in the U.S. and worldwide. Improvements in mine ventilation are a commonly used remedy to meet more stringent emission limit for all pollutants. However, mine operators have other options to reduce diesel particulate matter emissions including the adoption of more advanced engine technologies, aftertreatment control strategies, and the use of biodiesel fuels. These strategies have been found to be effective in reducing the mass concentration of diesel particulate matter (DPM) but, as a side effect, they can modify the tailpipe emission of another pollutant, nitrogen dioxide (NO₂). If the concentration of NO₂ exceeds the regulatory enforced limit, an increase in ventilation rate becomes necessary with related increased power demand and/or the cost of supplemental air shafts. This paper provides an overview of what effects the different exhaust aftertreatment technologies and control strategies available to the mining industry have on the NO₂ emissions. Specific focus is given to their effect on the ventilation strategy in an underground mine environment.

1 Introduction

Diesel engines are the major source of exposure for underground miners to nitric oxide (NO) and nitrogen dioxide (NO₂). NO and NO₂ cause short-term (acute) and long-term (chronic) health problems, mostly pulmonary and cardiac related (Sunyer *et al.* 1997; Stieb *et al.* 2002). NO₂ is a potent oxidizer and can potentially interact with other pollutants and induce the formation of ozone (O₃) and secondary hazardous particles (Peters *et al.* 1999; WHO 2005).

The exposure of underground miners to NO and NO₂ is regulated by the Mine Safety and Health Administration (MSHA). The enforced TLV limits for NO and NO₂ are 25 ppm (TWA) and 5 ppm (ceiling) respectively for both metal/non metal mines (30CFR 57.5001) and coal underground mines (30CFR 75.322). The TLV-Ceiling limit is by definition the concentration which may not be exceeded for any length of time. MSHA used the ACGIH's 1972 (coal) and 1973 (metal and nonmetal) TLVs to set these standards (OSHA 1988). In US coal mines, the exposure limits of underground miners to pollutant concentrations determine the ventilation rate specific for certified diesel engines. The ventilation rates (VR) are based on the amount of fresh air needed to dilute CO, CO₂, NO, NO₂ in the undiluted exhaust gas to the TLVs when the engine is operated at the worst emission condition for that specific pollutant. The prescribed nameplate VR (ventilation rate listed on the nameplate for the engine) pertains to the worst engine operating conditions from an emissions toxicity standpoint and therefore represents a minimum ventilation value applicable to the area where the engine is operated (30CFR 7.88).

As engine exhaust emissions, NO and NO₂ are generally referred to as nitrogen oxides (NO_x) and together with the Diesel Particulate Matter (DPM), they represent the most difficult diesel engine emission products to control. NO is the main contributor to the NO_x concentration, and several factors influence the partitioning between NO and NO₂ in the diesel exhaust.

The NO_x and DPM emissions control technologies, in optional retrofit or in original equipment manufacturer (OEM) form, are gradually finding their way into underground mining. This manuscript will provide an overview of the effects of the various technologies used to control diesel emissions on NO, NO₂ and NO_x emissions. Emphasis will be placed on the technologies that are currently used by underground mining operators to control gaseous emissions and particulate matter emissions, such as diesel oxidation catalysts (DOCs) and diesel particulate filters (DPFs) systems. In addition, this manuscript will briefly examine the effects of other emission control technologies such as selective catalyst reduction (SCR), lean NO_x trap (LNT), and lean NO_x catalyst (LNC).

2 The Ventilation rate

Each specific engine model approved by MSHA and present in underground mines must have an approval plate that specifies the minimum volume of air required to ensure that the exhaust emission levels do not exceed the exposure limits for NO, NO₂, CO, and CO₂. The approval process involves emission testing on an engine dynamometer using the ISO 8178 C1 test cycle. Based on the test results, MSHA calculates the ventilation rates for

each approved engine model (30CFR 7.88). The ventilation rates are computed as the quantity of additional ventilation air that has to be provided to the mine in order to dilute raw exhaust emissions to the respective TLV levels. This procedure ensures that mine operators increase the amount of ventilation as more engines are introduced underground.

As described in the following sections, several aftertreatment control technologies can have an effect on the NO₂ raw exhaust concentration. Periodic tailpipe NO₂ emission data can be collected while conducting the repeatable loaded engine test required for all heavy duty and permissible equipment under the weekly carbon monoxide (CO) check. This test measures the undiluted CO emissions of the engine operating under a loaded condition. This same test procedure can also be used to measure undiluted NO₂ emitted from the engine (MSHA 2002a).

This procedure allows the mine company to determine the variation of the NO₂ concentration caused by the aftertreatment control technology and to calculate a multiplier relative to this variation. In the case where NO₂ has increased, the mining company must obtain the MSHA ventilation rate for NO₂ (MSHA 2002a) and increase this rate by the same multiplier. If this calculated value exceeds the ventilation rate listed on the nameplate for the engine, the new value should be considered the air quantity necessary to dilute NO₂ emissions to safe concentrations.

It is important to underline that the NO₂ ventilation rate is different for each single approved engine, as is the prescribed nameplate ventilation rate, and that the same aftertreatment technology can have a different impact on the induced NO₂ tailpipe concentration for each engine. For this reason the NO₂ emission data collection needs to be performed for every single engine/control technology combination.

3 Diesel Oxidation Catalyst

Diesel Oxidation Catalysts (DOCs), are primarily designed to control carbon monoxide (CO), hydrocarbon (HC) and polycyclic aromatic hydrocarbons (PAH) emissions. Depending on catalyst formulation (described below), DOCs might have substantial effects on gas phase organics, but typically have limited effects on the organic fraction of particulate matter (Day *et al.* 2002). DOCs, initially introduced in underground mines to control the CO and HC emissions, became a widely used technology in mining applications. In recent years, DOCs have been often deployed to control emissions of hydrocarbons and the organic fraction of particulate matter emitted by mining vehicles fuelled with biodiesel.

A catalyst is a compound applied to the surface of the ceramic or metal DOC substrate, and it has the role of oxidizing CO and HC to CO₂ and H₂O. Several types of catalyst formulations are available and the most common formulations are based on oxides of cerium, vanadium,

platinum or palladium. The DOC activity is strictly dependent on exhaust temperature: a typical DOC shows low or no activity at low exhaust temperatures. The minimum temperature at which the DOC exhibits activity is called catalyst “light-off temperature”. To avoid the formation of sulphates and the consequent deactivation of the catalyst it is crucial to use fuel with low sulphur content.

DOCs typically have minor effects on NO_x emissions (Yamamoto *et al.* 2006). However, some of the catalysts used for DOC applications tend to promote the conversion of NO to NO₂ and they can substantially change the partitioning between NO and NO₂ emissions. The rate of oxidation of NO to NO₂ depends on several factors: catalyst formulation, monolith geometry, and exhaust temperature (Ambs *et al.* 1993; Farrauto 1997). The catalysts with platinum formulations are known to have high NO to NO₂ conversion potential. The catalysts with formulations based on vanadium and palladium were shown to suppress NO to NO₂ conversion (Farrauto 1997).

For some applications, such as the regeneration of a Diesel Particulate Filter (DPF), an elevated concentration of NO₂ in the exhaust is desired for the combustion of DPM. An 80% conversion of NO to NO₂ can be reached with an optimized DOC system operated at an average exhaust temperature of approximately 300 °C (Layrer 1994) A slight decrease of the NO₂ concentration has been observed at low load engine operating conditions for the catalyst formulations with low platinum loading (Ayala *et al.* 2001).

MSHA’s policy on this effect is that the increase in NO₂ emissions due to DOC use is tolerable as long as the engine’s approved ventilation rate is not violated. In other words, if the NO₂ ventilation rate is not higher than that listed on the engine approval, the aftertreatment technology should not degrade the mine environment significantly.

The NO₂ ventilation rate data for an Isuzu 4JG1T engine have been recently released by MSHA along with the results of a fuel testing campaign (MSHA 2009). The approved ventilation rate for this engine is 2.83 m³/s (6000 cfm), based on the NO emissions. An increase in NO₂ emissions between the bare engine (Figure 1a) and the DOC equipped engine (Figure 1b) is clearly noted. This increase, while large, does not approach the ventilation requirement of the engine. With adequate ventilation in the mine area where the engine is operating (6000 cfm and above) it does not seem likely for NO₂ emissions to rise above a miner’s TLV levels for NO₂ exposure. In mine environments with inadequate ventilation where local “dead spots” exist in work areas, it would be possible for NO₂ levels to rise to problematic levels faster with use of a DOC than without. But if this event would occur, then other regulated gas levels would also be increasing and most likely exceeding their individual TLV limits as well.

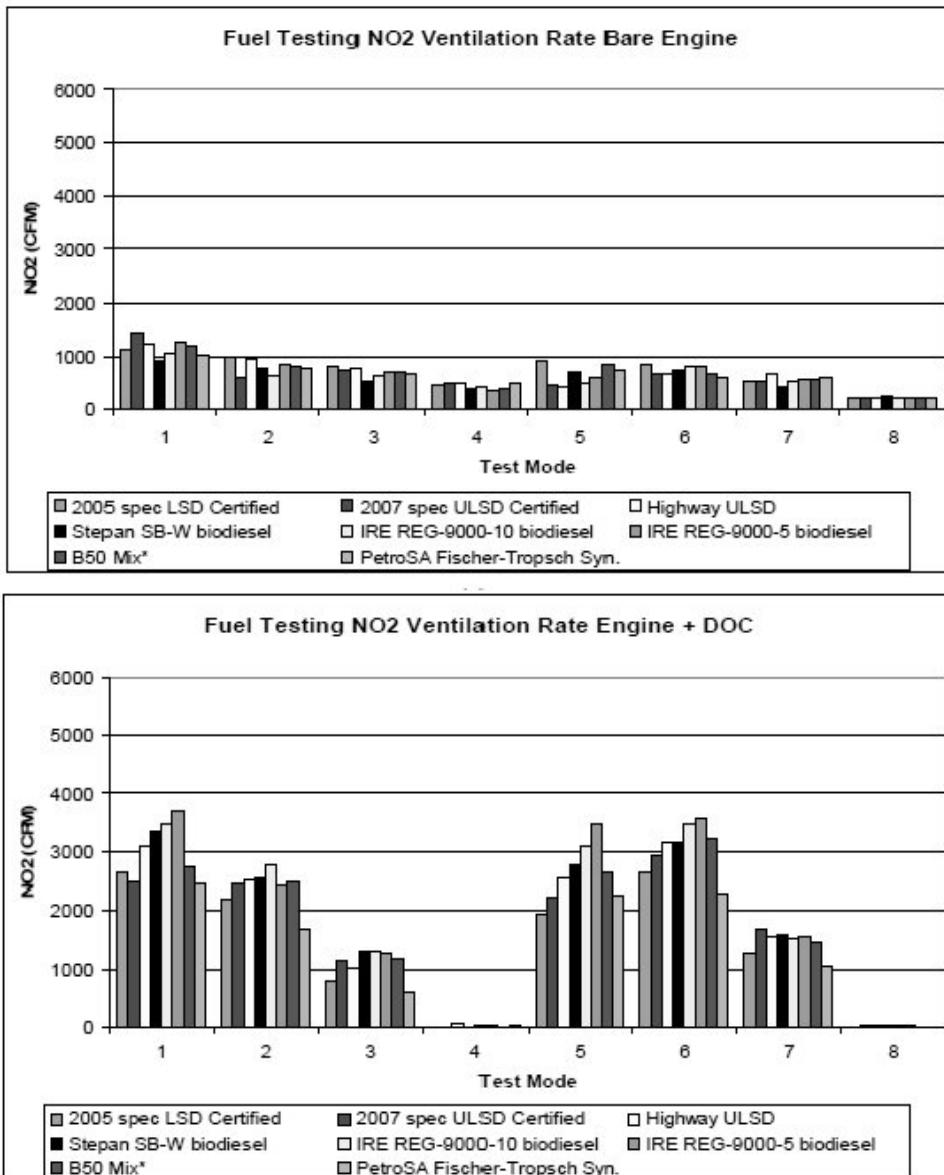


Figure 1 NO₂ Ventilation Rates for Bare engine (top) and Engine with DOC (bottom) during a Isuzu 4JG1T engine test campaign by MSHA (2009). Each bar is representative of a different fuel formulation used during the campaign. The results are subdivided in the eight engine modes used by MSHA to certify any approved engine.

It is important to underline that the DOC used for the MSHA testing has been found capable of converting at most 20% of NO to NO₂. The average reduction of CO emissions with the use of a DOC was in the range of 85%-100% for all the fuel tested. In this case, the reduction of the principal targeted pollutant, CO, by the DOC, does not induce an intolerable increase in NO₂ and it should not affect a well managed mine ventilation strategy.

4 Diesel Particulate Filter Systems

Diesel Particulate Filter (DPF) systems are designed to physically remove DPM from the exhaust of diesel engines. The majority of DPF systems currently present in the mining industry utilize, as a filtration strategy, the wall-flow system: the particles are trapped in the walls of the filter while the gases pass through them (Figure 2) (Corning 1999). The wall-flow system can provide up to

99% DPM mass filtration. The DPF must be periodically or continuously cleaned (regenerated) to avoid the occurrence of excessive engine back pressure. The regeneration is typically accomplished via oxidation of filtered DPM. Heat generated by exhaust temperatures in excess of the DPM ignition temperature is needed to initiate and support DPM oxidation. The DPM ignition temperature in a DPF is influenced by several parameters, but commonly it has to be higher than 250 °C (Cauda *et al.* 2007). Diesel engines operated over typical underground mining duty cycles rarely achieve exhaust temperatures sufficient to support regeneration of a DPF. However, the DPF regeneration can be promoted by the use of an active catalyst. The catalyst can be applied directly to the DPF wall using wash-coating technologies or it can be introduced in the system using fuel additives.

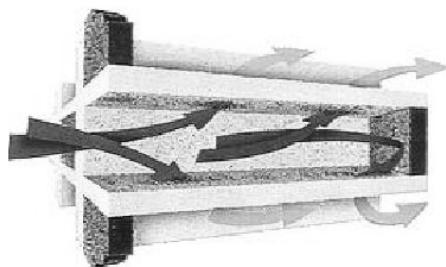


Figure 2 A ceramic wall-flow diesel particulate filter (DPF). The dark arrows represent the inlet flow, with high concentration of DPM, whereas the outlet flow, light arrows, presents a low DPM concentration. The DPM has been filtered by the walls of the DPF.

If an uncatalyzed DPF is utilized, the filtration process itself does not substantially affect the gaseous emissions. Under proper conditions the NO₂ was found to react with the filtered DPM in a process known as chemisorption and a net reduction of NO₂ is possible (Kalberer *et al.* 1999). On the other hand, several wash-coated catalytic DPFs demonstrated a high activity in converting NO to NO₂ (Chatterjee *et al.* 2004; Zhu *et al.* 2004; Czerwinski *et al.* 2007; Mogensen *et al.* 2009).

MSHA advises against using platinum catalyzed DPF systems in underground mines due to the potential for an increase in NO₂ emissions.(MSHA 2002b).

Other catalyst formulations, such as the metal based fuel borne catalysts, were found to have minor effects on NO₂ emissions (Richards *et al.* 2003). Nevertheless, a slight increase in NO₂ emissions is possible for the fuel additive formulations using a precious metal such as platinum (Czerwinski *et al.* 2007).

It is important to note that NO₂ has a higher oxidative potential than oxygen at low exhaust temperatures (Cooper *et al.* 1989; Allansson *et al.* 2002) and it is useful in some applications to lower regeneration temperatures

and enhance the regeneration of the DPF. The typical low concentration of NO₂ in diesel exhaust is normally insufficient to support DPF regeneration. For this reason, a DOC is commonly employed ahead of the DPF to increase the concentration of NO₂ in the exhaust gas. A DOC and an uncatalyzed DPF are the main components of the Continuously Regenerating Trap (CRT), patented by Johnson Matthey (Cooper *et al.* 1989; Cooper *et al.* 1990).

Balancing NO₂ production with NO₂ consumption by the DPM plays a critical role in supporting regeneration while preventing a potential increase in NO₂ emissions from such a system, known as NO₂ slip (Hawker 1998; Ayala *et al.* 2001). A proper choice of catalyst and settings of a DOC+DPF system theoretically can minimize the NO₂ slip. NO₂ slip can also be reduced by deploying the NO₂ abatement strategies discussed in the next sections of this article.

DPF systems actively regenerated using electrical energy off board or on board of the vehicle are popular in the mining industry. Actively regenerated DPFs are typically uncatalyzed and, as described in the previous paragraphs, they do not tend to increase NO₂ emissions during their use. The regeneration of off board DPFs should be performed in well ventilated stations where emissions of NO₂ and other gases should not be an issue.

The challenges related to the regeneration of DPF systems led to the development of flow through filters (FTF). FTFs are devices that can capture and store carbonaceous PM material for a period of time sufficient for its catalytic oxidation, while having open flow-through passages that allow exhaust gases to flow, even if the DPM holding capacity is saturated. The characteristic of this type of DPF is lower engine back pressure but only up to 50% DPM mass filtration (Okawara *et al.* 2005; Choi *et al.* 2007). Most FTF are wash-coated with a catalyst to continuously regenerate the DPM trapped and at the same time acting as a DOC by converting CO to CO₂.

From a mine ventilation perspective, the increase of NO₂ emissions connected with the use of a DPF system is a serious threat: the use of DPFs has been indicated as one of the most effective strategies to reduce the emission of DPM and to meet the MSHA DPM requirements in both coal and metal/non metal mines. The introduction of DPFs represents a robust alternative strategy to the increase of the ventilation rate to meet the DPM requirements: a lower ventilation rate can translate to a lower energy consumption rate and relative economical positive effects for the mine company. Increased NO₂ emissions connected with the use of some DPFs can reverse these effects, if the ventilation rate has to be increased to meet the TLV levels for NO₂ exposure.

Recently, due to the NO₂ issue in the mine environment, and to a much greater extent interest in NO₂ from the Air Resource Board in California, manufacturers of DPFs have begun working to reformulate these devices to minimize NO₂ production. MSHA is constantly updating the list of DPM control technologies available

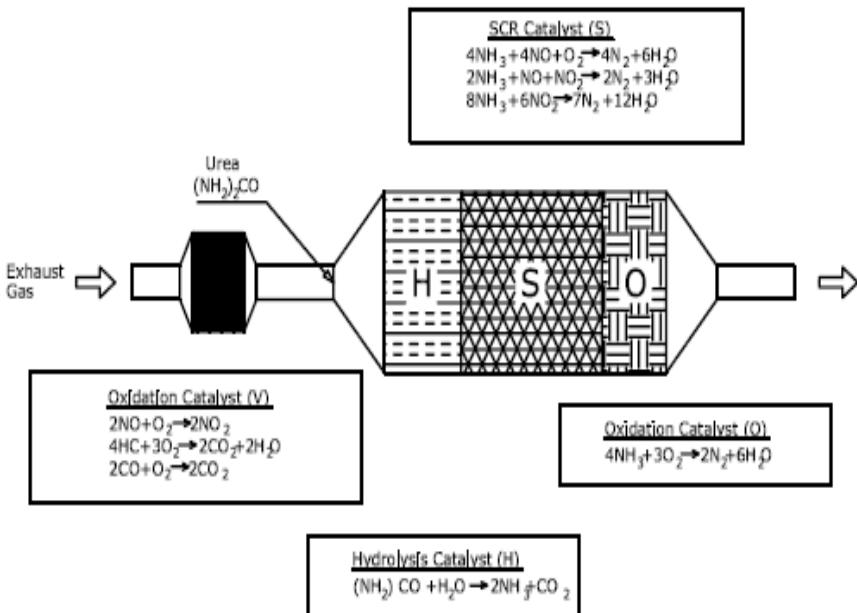


Figure 3 The scheme of a generic Selective Catalytic Reduction (SCR) system for mobile source

on the market for underground mine applications (MSHA 2007): the list is organized in order to provide valuable information about the risk of higher concentrations of NO_2 connected with the use of some of these technologies.

5 NO_x control strategies.

Different NO_x emissions control technologies are gradually finding their way into underground mines, mostly in original equipment manufacturer (OEM) form. This introduction is strictly connected with the development of integrated OEM engine systems produced by the engine manufacturers to meet the upcoming heavy-duty off-road Tier 4 standard.

The integrated OEM engine systems will assure lower NO_x emissions for the equipment present in underground mines and for this reason the mining ventilation strategy should receive overall benefit by the introduction of these systems. The complexity of these integrated engine systems and the necessity of more fine tuning for the engines are the potential threats connected with their use underground.

One of the most promising NO_x control strategies is the employment of a Selective Catalytic Reduction (SCR) technology. The conversion of both nitrogen oxides, NO and NO_2 , in the SCR system follows a complex path of chemical reactions (Cho 1994).

The combustion in a diesel engine is considered lean combustion, with a large excess of oxygen: for this reason, the reduction of NO and NO_2 is only possible via

the use of a specific reductant compound, such as ammonia or urea, injected in the exhaust ahead of the SCR system. A complete SCR system (Figure 3) includes an auxiliary oxidation catalyst (V), employed to modify the NO_x ratio, a main SCR catalyst (S) where the NO_x are converted, and a second oxidation catalyst (O) to prevent the leak of ammonia at the exit of the tailpipe, known as ammonia slip. Due to toxicity and the difficulties in handling and storage, ammonia is generally found to be impractical for mobile applications. The aqueous solution of urea is currently the reductant of choice for mobile SCR systems: the urea is then converted to ammonia in the first part of the SCR system, the hydrolysis catalyst (H). The nitrogen oxides react with ammonia over the catalyst in the main monolith to form elemental nitrogen. Reduction of NO_x on the SCR monolith surface is dependent on catalyst choice, temperature, and the NO_2/NO_x ratio. NO_2 plays an important role in the SCR system. The optimum NO_2/NO_x ratio was reported to be about 50% (Gieshoff 2000; Koebel *et al.* 2002). The concentration of NO_2 in diesel exhaust can be increased by using a proper auxiliary DOC, usually based on precious metals (e.g. platinum).

The reported NO_x conversion efficiencies in heavy-duty engines range from 55% to 90%, depending on the application and test method (Havenith *et al.* 1997; Fritz 1999; Miller *et al.* 2000). The SCR monolith activity is not selective towards NO and NO_2 , and the same reduction is attainable for both compounds.

Overall the adoption of an SCR as an OEM integrating engine system will reduce the required

ventilation rate for both NO and NO₂. However, considering the increased NO₂ concentration upstream of the main catalyst by the auxiliary DOC, the resulting reduction in required NO ventilation rate will be greater than the one for NO₂.

Two non-OEM aftertreatment technologies, alternatives to SCR, are under development and may become available for underground equipments in the near future: NO_x adsorber and LNC.

The strategy of any NO_x adsorber technology involves two distinct phases: the storage of NO_x on a catalytic surface during an engine lean condition, with high oxygen content, and the desorption/reduction of NO_x at a rich condition, with high content of hydrocarbons and low concentration of oxygen in the exhaust. The engine operating mode plays an important role in the shift from the adsorption to desorption/reduction phase (Epling *et al.* 2004) and there is an extensive use of catalysts for both phases. It is still unclear whether the trapping materials adsorb NO₂ more effectively than NO, or whether NO₂ may be the required intermediate compound for NO adsorption. In any case, the system promotes the oxidation of NO to NO₂ on active oxidation catalyst sites in the first step of the adsorbing phase. In general, NO_x adsorber catalysts can exhibit high NO_x conversion efficiencies in excess of 80-90% (Brogan *et al.* 1998).

The LNC system can be a valid alternative to the SCR system when the employment of urea and ammonia can be problematic. These systems were first commercialized for light-duty diesel engines but ongoing development efforts target the more challenging heavy-duty diesel engine applications.

In the LNC system, the reduction in NO_x is achieved by reacting NO_x with hydrocarbons in the presence of a catalyst. In a few applications, unburned hydrocarbons from the engine chamber are sufficient to obtain the desired NO_x conversion, otherwise a post-injection of hydrocarbon in the exhaust is necessary (Dorriah 1999). The conversion of both NO and NO₂ depends mostly on the catalyst chemical formulation and configuration (Truex 1994; Kharas *et al.* 1998) and on the temperature window at which the catalyst has the highest activity. A maximum achievable NO_x conversion by LNC systems for diesel engines operated over real duty cycles is 60% (Kharas *et al.* 1998). NO₂ does not have a crucial role in the process and its conversion is proportional to the conversion of overall NO_x.

Both NO_x adsorbers and LNC could be useful tools to reduce the NO and NO₂ emissions from modern diesel engines in the mine environment. Unfortunately the high complexity of the chemical reactions employed by these systems and the requirement of a constant emissions monitoring and tuning, can delay the introduction of these technologies in mining equipment. While the reduction of required ventilation rate for both NO and NO₂, a result of the reduced tailpipe emission, can be an advocate for their introduction, the dangerous misuse or malfunctioning of these technologies could potentially lead to an increase in

NO₂ emissions which could translate to a NO₂ exposure level well above the miner's TLV value.

In general it is important to underline that SCR systems have been developed in particular for heavy duty applications, considering the temperature required and the space necessary for the system. On the other hand, LNC and NO_x adsorbers, due to performance and durability requirements have been developed for passenger cars and light duty applications.

6 Conclusions

This paper provides an overview of the effects which different aftertreatment technologies have on NO₂ emissions and the relative effect on the mine ventilation strategy. Emphasis was given to the technologies viable for controlling the exposure of underground miners to diesel particulate matter and gases. Although a wealth of information is available in the scientific literature on the effects of control technologies on NO_x emissions, limited information is available on the effect of those technologies on NO₂ emissions.

According to the literature, technologies such as uncatalyzed or base metal catalyzed filters have minor effects on NO₂ emissions. On the contrary, DOC and noble metal catalyzed filters were shown to have the potential to substantially increase NO₂ emissions. NO₂ slip was reported to be a particular problem for the system that depends on NO₂ for DPF regeneration. The NO₂ slip can be controlled with NO_x abatement systems. The NO_x abatement systems such as SCR and NO_x adsorbers were reported to be very effective for controlling total NO_x emissions. Those systems were shown to preferentially remove NO and therefore be less efficient in controlling NO₂ emissions.

The majority of the technologies described in this paper have been developed for on-highway and off-highway surface applications using design parameters different from those for underground mining applications. Due to the confined rugged environment and occupational rather than environmental regulations, the implementation of emission control technologies in an underground mine presents a unique challenge.

The MSHA prescribed ventilation rate for NO₂ can be influenced by the introduction of any of the aftertreatment control technologies described. If the modified NO₂ ventilation rate exceeds the MSHA nameplate ventilation rate, the adoption of the aftertreatment control technology can induce an increase in the air quantity necessary to dilute NO₂ concentrations below the TLV level. This effect has been noticed especially with the use of highly catalyzed DOCs and catalyzed DPFs. A misuse of more complex NO_x control strategies can have similar disadvantages.

Substantial research is currently being conducted on new technologies designed to reduce diesel emissions including NO₂ and on their effect on the ventilation requirements in underground mines.

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