

Diesel Oxidation Catalytic Converters for Underground Mining Applications

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The diesel oxidation catalytic converter (DOC) has been extensively used by the underground mining industry to reduce exposure of workers to carbon monoxide (CO) and hydrocarbons (HC) emitted by diesel engines. The effects of those devices on the gaseous and diesel particulate matter emissions strongly depend on catalyst formulation. Recently, certain formulations of catalytic coatings used in DOCs marketed to underground mining were scrutinized for their potential to adversely affect emissions of the highly toxic compound, nitrogen dioxide (NO₂). This study was conducted to get a better understanding of the effects that contemporary and emerging DOC technologies have on gaseous emissions. For test purposes, an EPA certified Tier 2 engine was retrofitted with two DOCs, one with a catalytic coating traditionally marketed to the underground mining industry (DOC 1), and the other with a novel coating designed to minimize undesirable NO₂ emissions (DOC 2). The evaluation was done for various steady-state engine operating conditions, generating exhaust with temperatures between 200 °C and 400 °C and one mining related transient cycle. Despite differences in catalyst formulations, both evaluated DOCs similarly reduced CO (63 to 98 %) and HC (16-32%) emissions. However, for the majority of test conditions, the NO₂ emissions were found to be adversely affected by DOC 1. Dramatic increases in NO₂ emissions were observed at conditions that produced exhaust temperatures above 300 °C. Conversely, DOC 2 was found to favorably affect NO₂ emissions for all test conditions. The findings of this study suggests that DOC catalyst formulations and systems can be successfully designed and optimized for underground mining applications to provide the desired reductions in CO and HC emissions without de novo generation of NO₂ and in certain circumstances, provide a reduction in NO₂ emissions..

Keywords: Diesel Oxidation Catalyst, Nitrogen Dioxide, Underground Mining.

1. Introduction

Diesel-powered vehicles and equipment are the backbone of production and transportation in the underground mining industry. However, they are also one of the primary sources of miners' exposure to fresh submicron aerosols and noxious gases such as carbon monoxide (CO), nitric oxide (NO), nitrogen dioxide (NO₂), and hundreds of various hydrocarbons (HC) including potentially carcinogenic and mutagenic polycyclic aromatic hydrocarbons (PAHs) and nitrated-PAHs (n-PAH).

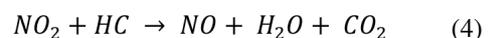
Over the past four decades DOCs have been extensively used in original equipment manufacturer (OEM) and retrofit applications to curtail some of the gaseous emissions from diesel powered equipment in underground mines [1-3]. Traditionally, the catalyst formulations and loadings used for underground mining DOCs have been optimized for effective curtailment of carbon monoxide (CO) and hydrocarbon (HC) emissions [4]. The major governing chemical reactions, oxidation of CO and HC are described by the equations 1 and 2.



Certain catalyst formulations, and primarily those containing Pt, promote the oxidation of nitric oxide (NO) to nitrogen dioxide (NO₂) while operated at temperatures typically observed for heavily loaded diesel engines [1,4-8]. That reaction is described by equation 3.



Under favorable conditions certain types of DOCs were shown to be effective in removing carcinogenic and mutagenic poly aromatic hydrocarbons (PAHs) [9,10], and reducing both particle and vapor-phase-associated mutagenic activity of exhaust emitted by older technology engines [10,11]. Certain types of DOCs were even found to reduce the organic fraction of diesel particulate matter (DPM) [12]. The effectiveness of a DOC as a DPM control is primarily dependent on the fraction of organic carbon (OC) present in the engine exhaust because the total DPM reduction efficiency increases with an increase in OC content of the exhaust [13,14]. The simplified reaction of NO₂ with HC and DPM [4,6,15] is described by equation 4.



It is important to note that the performance of a catalyst can degrade with age. Katare et al. [6] showed that in presence of reductants (HC, CO, DPM), certain

in-use DOCs with demonstrated low CO and HC conversion can be net consumers of NO₂.

The catalyst technologies gradually evolved to accommodate for advancements in diesel engine technologies, fuels, and exhaust aftertreatment technologies that resulted in generally lower CO, NO_x, HC, and DPM emissions. The DOCs found even new applications. Those with catalysts formulated to promote oxidation of NO to NO₂ have been extensively used in the diesel particulate filter (DPF) systems to increase the concentrations of NO₂ that is used in the DPF as a low-temperature DPM oxidizing agent [16,17] and in urea-based selective catalyst reduction (SCR) systems to increase the NO₂ concentration from engine levels to the levels needed to optimize the performance of the SCR catalyst [18]. Balancing the production and consumption of NO₂ in such systems over actual duty cycles, presents a major challenge. Without proper NO₂ slip control, these systems emit relatively high concentrations of NO₂ and are considered unsuitable for underground mining applications. The use of similarly catalyzed exhaust aftertreatment devices increased the ratio of NO₂ in NO_x in diesel emissions [3,8,19] and in the environment [2,7].

Nitrogen dioxide is a primary health concern because of its relatively high toxicity [20,21]. NO₂ is also of concern because of its potential role in the formation of nitro-PAHs [22] and atmospheric ozone-forming chemistry [23]. Due to technical and economic issues related to ventilation of confined spaces, the NO₂ emissions are of particular concern for the underground mining industry. The current regulations enforced by the Mine Safety and Health Administration (MSHA) in the U.S., limit NO₂ exposure of underground coal [24] and metal/nonmetal (M/NM) miners [25] to a ceiling value (CE) of 5 ppm. That level was established on the basis of the 1972 and 1973 American Conference of Governmental Industrial Hygienists (ACGIH®) threshold limit values (TLV®s), respectively. Certain states enforce the exposure limits for NO₂ more stringent than those enforced by MSHA. E.g., the ambient concentration for NO₂ in underground coal mines in West Virginia cannot exceed 3 ppm (CE) [26]. The ACGIH decision to reduce the TLV® for NO₂ from 5 to 0.2 ppm [21] reinvigorated the discussion on the cost of controlling DPM emissions using catalyzed systems that adversely affect NO₂ emissions.

The concern over de-novo formation of NO₂ emissions motivated development and evaluation of alternative catalytic formulations [4,27,28]. At relevant diesel exhaust temperatures, Pd-based and base metal-Pd based formulations were found to have a low tendency to catalyze the oxidation of NO to NO₂, while being quite effective, comparably to Pt-based formulations, in catalyzing the oxidation of CO and HC to CO₂ and CO₂ and H₂O, respectively [4,27]. However, some of the recent laboratory studies [3,29] showed that a surprisingly high number of DOCs currently used in underground mining use the catalyst formulations that promote undesirable secondary NO₂ emissions.

Therefore, this study was conducted to demonstrate the fundamental differences in the effects of two types of contemporary catalyst formulations on emissions and emphasize the need for careful selection and the potential optimization of DOCs for specific underground mining applications.

2. Methodology and Materials

Two DOCs supplied by AirFlow Catalyst Systems Inc. (Rochester, NY) were evaluated during this study, EZDOC (DOC 1) and MinNoDOC (DOC 2). Both DOCs are using identical metal substrates sized for the test engine. The major difference between units was in the oxidation catalyst formulations. The washcoat of DOC 1 was impregnated with a catalyst formulation representative of the coatings traditionally used for DOCs marketed to the underground mining industry, to curtail CO and hydrocarbon emissions from traditional light- and medium-duty diesel engines operated in very well ventilated areas of underground mines. The catalyst used for DOC 2 was specifically formulated to allow for effective control of CO and hydrocarbon emissions from contemporary diesel engines, and to suppress the formation of secondary NO₂ emissions.

Testing took place at the diesel laboratory of the National Institute for Occupational Safety and Health (NIOSH), Office of Mine Safety and Health Research (OMSHR). The DOCs were retrofitted to an electronically controlled, turbocharged, Mercedes Benz OM 904 LA rated at 174 hp. The emissions from this specific engine meet U.S. Environmental Protection Agency (EPA) Tier 2 standards. The engine was coupled to the SAJ AE400, 400 kW water-cooled eddy-current dynamometer. The testing was done using locally acquired ultralow sulfur diesel (ULSD). The test fuel was analyzed by Cashman Equipment Co. (Bentley Tribology Services, Sparks, NV). The results are summarized in Table 1.

Table 1. Fuel properties

Fuel Properties	ASTM	Value
	Test Method	
Aromatics [vol %]	D1319	24.2
Olefins [vol %]	D1319	1.6
Saturates [vol %]	D1319	74.2
Flash Point, [°C /°F]	D93	62.5 /144.5
Sulfur, by UV [ppm]	D5453	7.4
Cetane Number []	D613	44.5
API Gravity @ 15.6 °C [°API]	D1298	36.9
Heat of Combustion [BTU/gal]	D240	139945

The DOCs were tested for several steady-state engine operating conditions and one transient cycle. The first set of steady-state test conditions was selected to generate exhaust gases with temperatures ranging between 200 to 400 °C. That was achieved by maintaining the engine speed at 1800 rpm while gradually increasing engine load in steps from 136 to 610 Nm (Table 2, Fig. 1). A subset of four modes of the

International Standards Organization for Standardization (ISO) 8-mode test cycle [30] was incorporated for the second set of steady state conditions (Table 2). The R100 (M1) and I100 (M5) modes are representative of heavy-duty engine operating conditions producing relatively high exhaust temperatures. The R50 (M3) and I50 (M7) modes are considered medium-duty engine operating conditions that produce low to medium exhaust temperatures (Fig. 1). The ISO 8178-C1 is used by MSHA for approval and certification of diesel engines for use in underground mines in the U.S. [31].

Table 2. Steady-state engine operating conditions

Engine Speed rpm	Engine Load		ISO 8178 C1
	lb-ft	Nm	
1800	100	136	N/A
800	200	271	N/A
1800	250	339	N/A
1800	300	407	N/A
1800	350	475	N/A
1800	400	542	N/A
1800	450	610	N/A
1400	235	319	M8 (I50)
1400	470	637	M6 (I100)
2200	190	258	M3 (R50)
2200	380	515	M1 (R100)

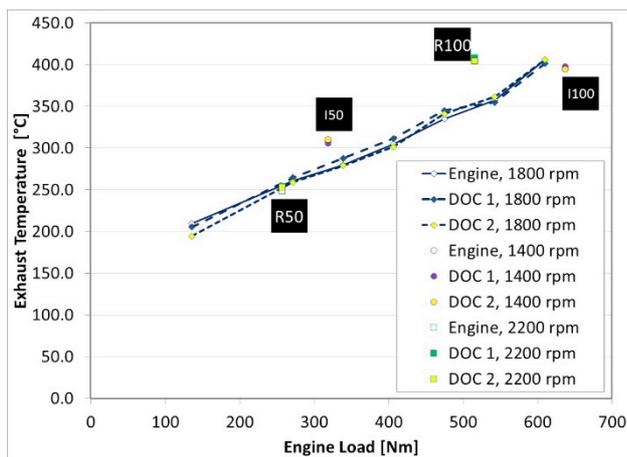


Fig. 1. Engine loads and corresponding exhaust temperatures at the inlet to DOC

In an attempt to quantify the effects of the DOCs for more production-representative conditions, the engine was operated over an 820-second transient mining cycle. This cycle has been recreated from field data to simulate operation of an engine in underground mining load-haul-dump vehicles.

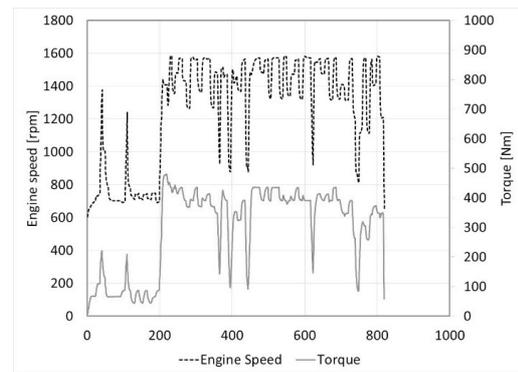


Fig. 2. Transient underground mining cycle

The concentrations of CO, CO₂, NO, NO₂, and hydrocarbons were measured upstream and downstream of the DOCs in undiluted exhaust using a Fourier transform infrared (FTIR) spectrometer (Gasetm, DX-4000). The concentrations of the following hydrocarbons were combined to obtain total hydrocarbon concentrations (THC): ethane, propane, butane, pentane, hexane, octane, ethylene, acetylene, propene, 1,3-butadiene, formaldehyde, acetaldehyde, benzene, and toluene.

3. Results

Generally, the contributions of DOCs to CO₂ concentrations are negligible, and the results of CO₂ concentration measurements may be used to examine the consistency of the test procedure (Fig. 3). As indicated by the graphs given below, the variability in measured CO₂ concentrations for all steady-state tests conducted at the corresponding conditions was within ± 3 percent (Fig. 3b, Fig. 3c).

The results of steady-state and transient tests showed that both DOCs had favorable effects on CO emissions. For conditions that generated exhaust temperatures above 300 °C, the DOCs were found to be very efficient in oxidizing CO (Fig. 4a). At those conditions, CO reduction was several percentages better for DOC 2 than for DOC 1 (Fig. 4b). At exhaust temperatures below 300 °C, DOC 1 was somewhat more efficient than DOC 2. For both DOCs, the highest efficiencies were observed during the heavy duty portion of the duty cycle (Fig. 4c). The reductions in cumulative CO emitted during the cycle for DOC 1 and DOC 2 were estimated to be 87 and 91 percent, respectively (Fig. 4d).

Both DOCs had a relatively minor effect on NO_x (NO_x=NO+NO₂) emissions (Fig. 5a). The presence of catalysts in DOCs affects processes such as oxidation of NO to NO₂ and the reduction of NO₂ to NO. However, since those reactions are reversible, the total sum of those compounds (NO_x) typically remains in equilibrium: Increases in NO₂ concentrations are typically offset by concurrent decreases in NO concentrations. For steady-state tests (I50, I100, R50, and R100), the effects of tested DOCs on NO_x were found to be within ± 5 percent (Fig. 5b).

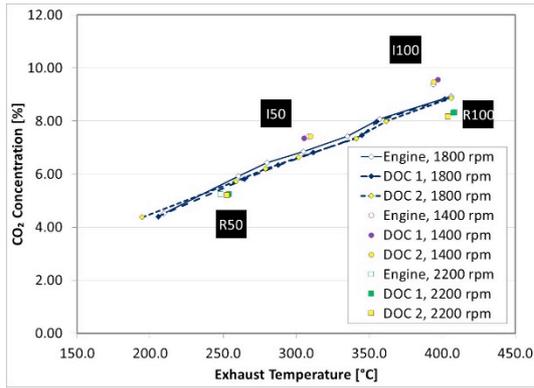


Figure 3A: CO₂ emissions for steady-state

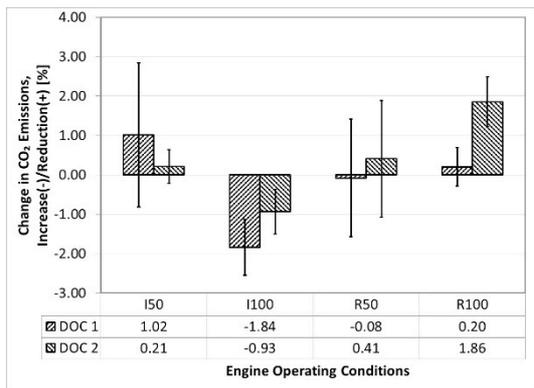


Figure 3B: changes in CO₂ emissions for I50, I100, R50 and R100 conditions

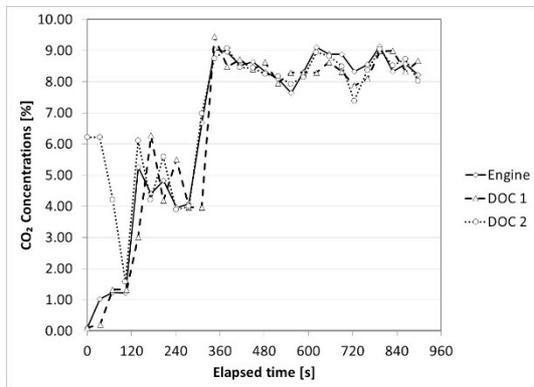


Figure 3C: CO₂ emissions for transient cycle

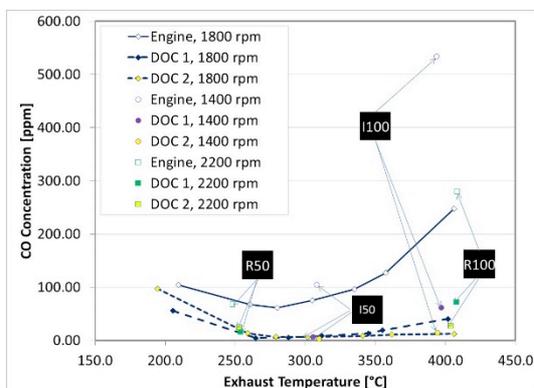


Figure 4A: CO emissions for steady-state conditions

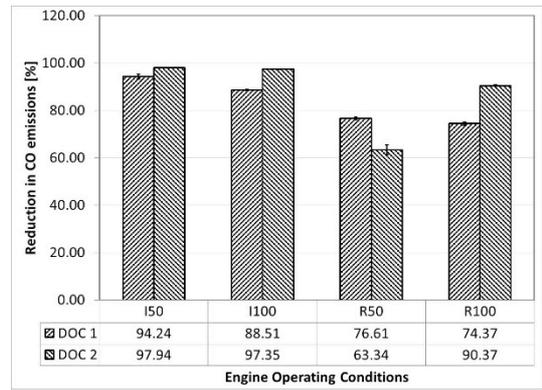


Figure 4B: reductions in CO emissions for I50, I100, R50, and R100 conditions

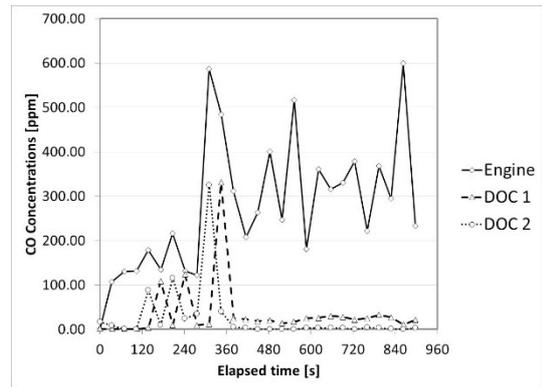


Figure 4C: CO emissions for transient duty cycle

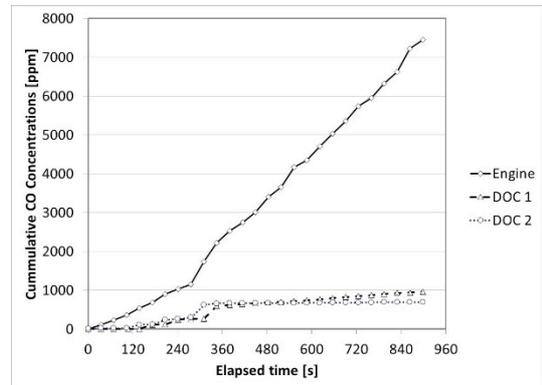


Figure 4D: cumulative CO emissions for transient duty cycle

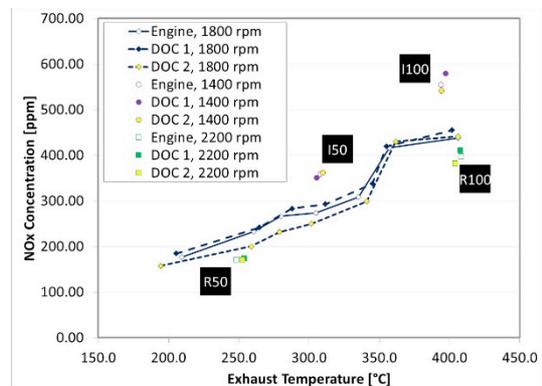


Figure 5A: NO_x emissions for steady-state conditions

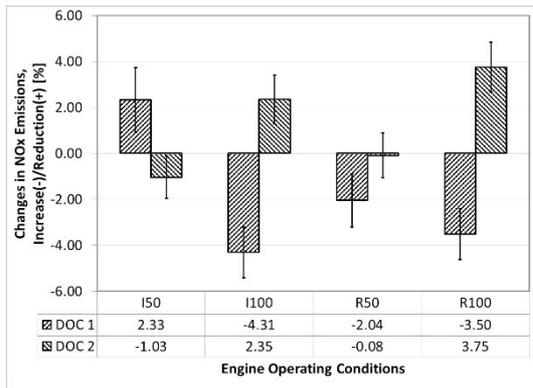


Figure 5B: Change in NO_x emissions for I50, I100, R50, and R100 conditions

In general, the NO₂ emissions were adversely affected by DOC 1, and favorably by DOC 2 (Fig. 6a, Fig. 6b). The effects of those DOCs on NO₂ emissions were found to be strongly dependent on exhaust temperature. In the case of DOC 1, the NO₂ emissions were above engine levels for exhaust temperatures exceeding 250 °C, exhibiting the highest values (up to 550 percent increase over engine emissions) at temperatures between 350 and 400 °C (Fig. 6a). The NO₂ emissions from DOC 2 were below engine levels for the entire examined range of exhaust temperatures (Fig. 6a). The cumulative emissions from the engine retrofitted with DOC 1 contained, approximately 129 percent more NO₂ than those from the engine alone, while the cumulative emissions from the engine equipped with DOC 2, contained approximately 77 percent less NO₂ than those from the engine alone (Fig. 6d). The NO₂ concentrations emitted from DOC 1 were higher throughout the cycle, but the bulk of NO₂ was emitted during the heavy-duty portion of the cycle (Fig. 6c). The NO₂ emissions from DOC 2 were relatively low and constant throughout the entire cycle (Fig. 6c).

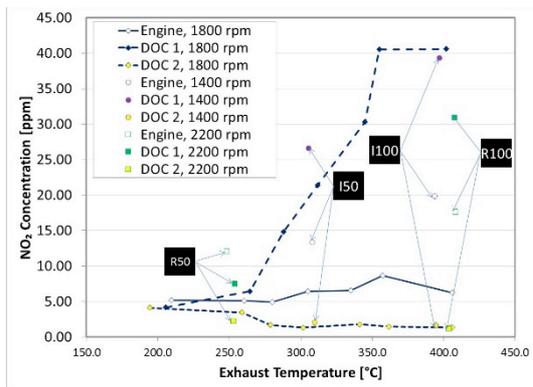


Figure 6A: NO₂ emissions for steady-state conditions

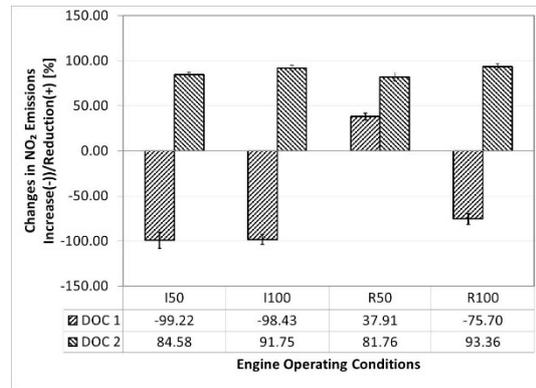


Figure 6B: changes in NO₂ emissions for I50, I100, R50, and R100 conditions

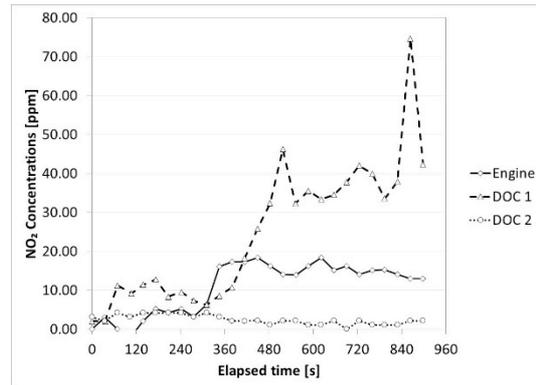


Figure 6C: NO₂ emissions for transient duty cycle

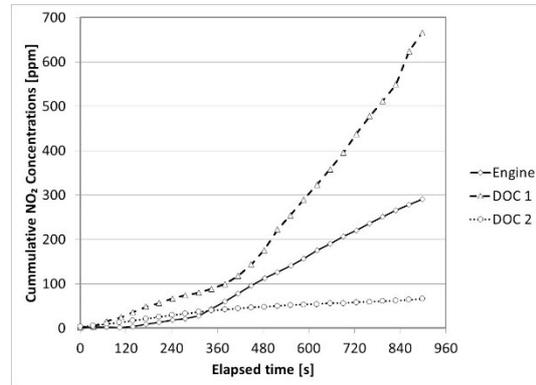


Figure 6D: cumulative NO₂ emissions for transient duty cycle

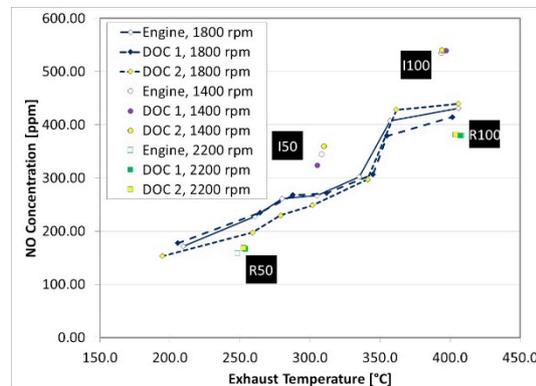


Figure 7A: NO emissions for steady-state conditions

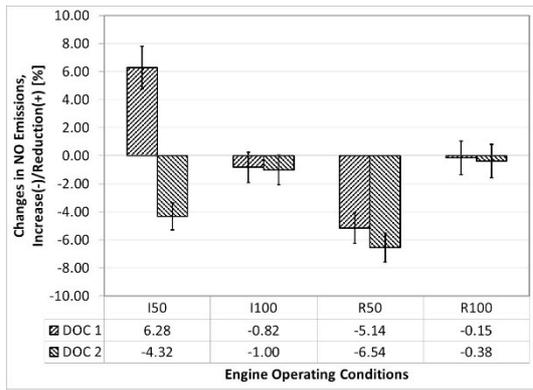


Figure 7B: changes in NO emissions for I50, I100, R50, and R100 conditions

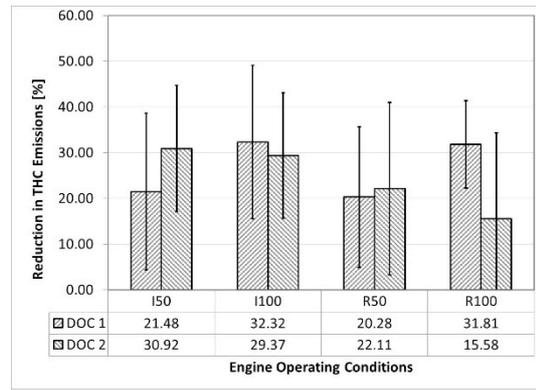


Figure 8B: changes in THC emissions for I50, I100, R50, and R100 conditions

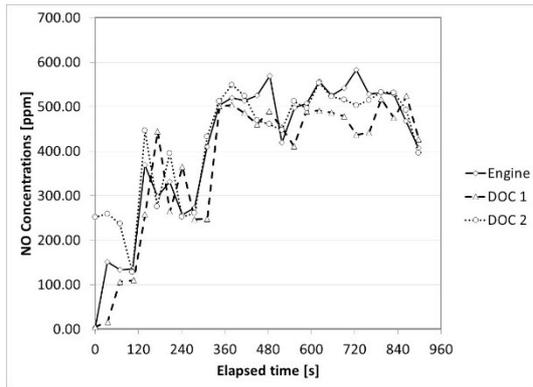


Figure 7C: NO emissions for transient duty cycle

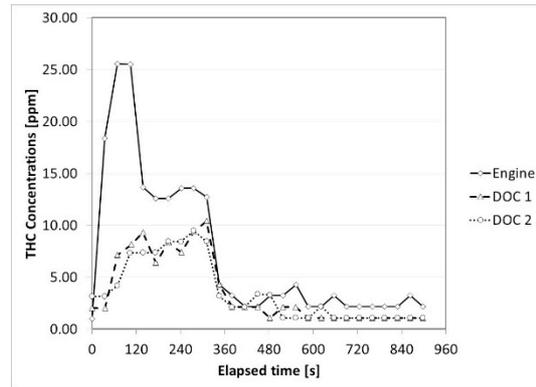


Figure 8C: THC emissions for transient duty cycle

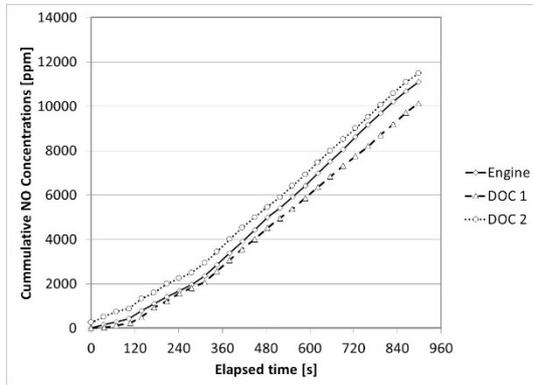


Figure 7D: cumulative NO emissions for transient duty cycle

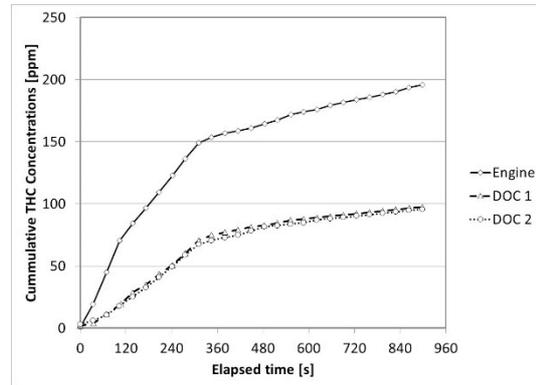


Figure 8D: cumulative THC emissions for transient duty cycle

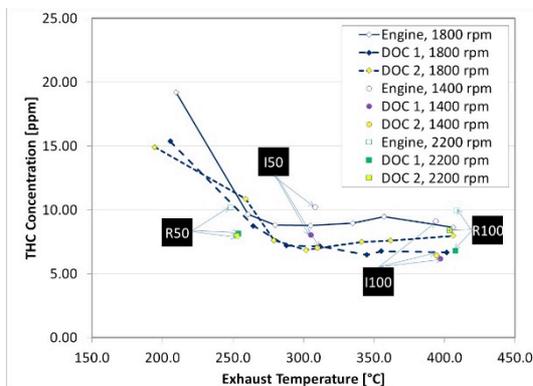


Figure 8A: THC emissions for steady-state conditions

NO comprised all but a few percent of the total NO_x emitted by the test engine and from DOC 2, and more than 90 percent of NO_x emitted from DOC 1. As a result, the major changes in NO_2 emissions resulted in relatively minor changes in NO emissions (Fig 7a, Fig 7b). The effects of DOCs on NO emissions were within ± 7 percent (Fig. 7b). In the case of transient duty cycle, DOC 1 reduced the cumulative NO emissions by 9 percent and DOC 2 increased the cumulative NO emissions by 4 percent over levels recorded for the untreated exhaust (Fig. 7c and Fig. 7d).

Both DOCs were found to be fairly effective in controlling THC emissions (Fig. 8). The reductions in THC emissions were only slightly affected by exhaust temperatures (Fig. 8a). The average reductions observed during steady-state tests did not exceed 32 percent (Fig.

8b). Over the transient cycle, DOC 1 and DOC 2 reduced the cumulative THC concentrations by 50 and 51 percent, respectively (Fig. 8c, Fig. 8d).

4. Summary

The DOCs were found to be very effective in controlling CO emissions, and fairly effective in controlling THC emissions. The major difference was in the effects of those DOCs on NO₂ emissions. Due to the adverse effects on NO₂ emissions, DOC 1 appears not suitable for use on underground mining vehicles when operated over duty cycles where exhaust temperatures exceed 250 °C. DOC 2 appears to offer reasonable reductions in CO and THC emissions with a decrease in NO₂ emissions for applications where exhaust temperatures are between 200 and 400 °C.

Due to the wide range of operating conditions, DOC catalyst formulations need to be optimized for specific applications by taking in account all targeted control parameters as well as exhaust physical properties and chemical composition. Properly optimized DOCs with catalyst formulations specifically designed for underground mining applications should provide the desired reductions in CO and HC emissions, while also reducing engine out NO₂ emissions.

Disclaimer

The findings and conclusions in this manuscript are those of the authors and do not necessarily represent the views of NIOSH. Mention of company names or products does not constitute endorsement by the Centers for Disease Control and Prevention.

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Acronyms

ACGIH	American Conference of Governmental Industrial Hygienists
API	American Petroleum Institute
ASTM	ASTM International, an international standards organization that develops and publishes voluntary consensus technical standards
CE	celling value
CO	carbon monoxide
CO ₂	carbon dioxide
DOC	diesel oxidation catalytic converter
DPF	diesel particulate filter
DPM	diesel particulate matter
EZDOC	diesel oxidation catalytic converter supplied by AirFlow Catalyst Systems Inc.
FTIR	Fourier transform infra-red
HC	hydrocarbons
I50 (ISO M8)	intermediate speed 50 percent load
I100 (ISO M6)	intermediate speed 100 percent load
ISO	International Organization for Standardization
MinNoDOC	diesel oxidation catalytic converter supplied by AirFlow Catalyst Systems Inc.
M/NM	metal/nonmetal
MSHA	Mine Safety and Health Administration
NIOSH	National Institute for Occupational Safety and Health
nitro-PAHs	nitrated polycyclic aromatic hydrocarbons
NO	nitric oxide
NO ₂	nitrogen dioxides
NO _x	nitric oxides (NO _x =NO+NO ₂)
OEM	original equipment manufacturer
OMSHR	Office of Mine Safety and Health Research
PAH	polycyclic aromatic hydrocarbons
R50 (ISO M3)	rated speed 50 percent load
R100 (ISO M1)	rated speed 100 percent load
SAE	Society of Automotive Engineers
SCR	selective catalyst reduction
THC	total hydrocarbons
TLV	threshold limit values (ACGIH)
TR	transient
ULSD	ultralow sulfur diesel
U.S. EPA	U.S. Environmental Protection Agency