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17 pgs

IC 9197

Bureau of Mines Information Circular/1988

Effectiveness of Catalytic Converters on Diesel Engines Used in Underground Mining

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UNITED STATES DEPARTMENT OF THE INTERIOR

Library of Congress Cataloging in Publication Data:

McClure, B. T. (B. Thompson)

Effectiveness of catalytic converters on diesel engines useu in underground mining.

(Information circular; 9197)

Bibliography: p. 10-12.

Supt. of Docs. no.: I 28.27:9197.

1. Mining machinery--Catalytic converters. 2. Diesel motor exhaust gas--Purification. I. Baumgard, K. J. (Kirby, J.). II. Watts, W. F. (Winthrop F.). III. Title. IV. Series: Information circular (United States. Bureau of Mines); 9197.

TN295.U4 [TN345] 622 s [6221.8] 88-600120

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UNIT OF MEASURE ABBREVIATIONS USED IN THIS REPORT

°C	degree Celsius	m^3	cubic meter
ft	foot	mg/m^3	milligram per cubic meter
g	gram	min	minute
g/h	gram per hour	mm	millimeter
g/kW•h	gram per kilowatt hour	pct	percent
h	hour	ppm	part per million
L	liter	s	second

EFFECTIVENESS OF CATALYTIC CONVERTERS ON DIESEL ENGINES USED IN UNDERGROUND MINING

By B. T. McClure,[1] K. J. Baumgard,[2] and W. F. Watts, Jr.[3]

ABSTRACT

Oxidizing catalytic converters are sometimes used by underground mine operators as an emission control device to reduce odor, hydrocarbon (HC), and carbon monoxide (CO) emissions from diesel equipment. The objectives of this report are to quantitatively assess the effects of catalytic converters on diesel exhaust emissions and to make recommendations for their use. Information in this report is from a literature survey and contract research supported by the Bureau of Mines.

Catalytic converters are effective in reducing CO, HC, and odors when the exhaust temperature is high enough so that the converter remains above 250° C. Converter temperature is dependent upon engine duty cycle. Catalytic converters increse sulfate emissions and slightly increase oxides of nitrogen (NO_x) emissions. If low sulfur fuels are not used, the increase in sulfate and nitrogen dioxide (NO_2) emissions can offset any advantages of the catalytic converter as measured by the emissions quality index (EQI). In addition, in vitro bioassays have shown that catalytic converters produce soluble organic compounds, which have increased mutagenic activity with respect to untreated exhaust.

Based upon criteria recommended by a joint Canadian-United States research panel, catalytic converters should only be used in special circumstances in underground mines. Vehicles equipped with catalytic converters should operate under moderate to heavy load conditions, and use fuel with a sulfur content less than 0.1 pct.

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INTRODUCTION

This Bureau of Mines report quantitatively assesses the effects of catalytic converters on diesel exhaust emissions and makes recommendations for their use. Information in this report is from a literature survey and contract research supported by the Bureau.

Diesel-powered equipment is widely used in underground metal and nonmetal mines and its use in coal mines is increasing (1).[4] It is widely believed that diesels are more flexible, economical to operate, and are able to boost productivity over that of their electrically powered (battery, tethered, or trolly) counterparts (2).

However, diesel engines emit pollutants that reduce overall air quality. Pollutants emitted in diesel exhaust include CO, carbon dioxide (CO_2), HC, nitric oxide (NO), NO_2, sulfur dioxide (SO_2), and diesel particulate matter (DPM). Such pollution is particularly undesirable in a restricted environment as in an underground mine. Concentrations of CO, CO_2, NO, NO_2, and SO_2 are regulated by the Mine Safety and Health Administration. The standards are shown in table 1. DPM has no specific standard at this time, however, in coal mines DPM is indirectly regulated under the 2 mg/m^3 respirable dust standard.

TABLE 1. - Exposure limits for diesel exhaust pollutants

Pollutant	Noncoal		Coal	
	FSEL	STEL	FSEL	STEL
CO....ppm..	50	400	50	400
CO_2...ppm..	5,000	15,000	5,000	30,000
NO....ppm..	25	37.5	25	NAp
NO_2...ppm..	NAp	5	3	5
SO_2...ppm..	5	20	2	5

FSEL Full-shift exposure limit.
NAp Not applicable.
STEL Short-term exposure limit.

Because of the usefulness of diesels it is desirable to control their emissions, and catalysts are one method. Mine operators have been using catalytic converters in underground mines for more than a decade to reduce CO, HC (3), and odor, which is primarily associated with HC's (4). Converter use is prohibited in some areas of gassy noncoal mines, and coal mines, because of their high operating temperatures. Examples of these areas include the cutting face and return airways where gas and dust concentrations may be high and where permissible equipment is required.

COMMERCIAL CONVERTERS

Oxidizing catalytic converters are sold commercially for several applications, including wood-burning stoves, natural gas burners, and for heavy-duty diesel engines used in mining. The precious metals that are used as catalysts include platinum, palladium, and others, which are bonded to the surface of a substrate where the chemical reactions take place.

Converters used on underground diesels in the United States are typically one of two types, monolithic substrate or pellet-type. The monolithic converter uses a ceramic, monolithic honeycomb substrate to support the catalyst. The monolith has many small axial passages that extend the length of the honeycomb, and are separated from one another by thin porous walls approximately 0.15- to 0.3-mm thick. The monolith is usually cylindrical, sized to accommodate the exhaust flow of the engine, and enclosed in a stainless steel container adapted to fit the exhaust system.

The pellet-type converter uses spherical or cylindrical pellets, approximately 3 mm in diam which fit into a stainless steel container sized to fit the exhaust system. A typical pellet converter is larger and heavier than a monolithic converter.

OXIDIZING CATALYTIC CONVERTERS

An oxidizing catalytic converter is placed as near as possible to the exhaust manifold to ensure that hot exhaust

[4]Underlined numbers in parentheses refer to items in the list of references preceding the appendix.

gases pass over the catalyst at maximum temperature. The performance of converters is critically dependent on the exhaust temperature, which depends upon engine operation. If an engine is operated at low load, the low exhaust temperature allows soot to accumulate on the catalyst surface, thereby restricting access of the exhaust to the catalyst and inhibiting the desired oxidation. As the hot gases pass over the catalyst, oxidation of CO and HC to CO_2, water vapor takes place, thus reducing the amount of CO, HC, and offensive odors in the exhaust stream. At high exhaust temperatures the converter also oxidizes SO_2 and NO to more toxic sulfuric acid (H_2SO_4) and NO_2 (5-7).

Research conducted by the Engelhard Corp., Edison, NJ, under a Bureau contract (7) demonstrated that converter effectiveness is temperature dependent and that high-temperature operation increases NO_2 emissions. Figure 1 shows that greater than 50 pct reductions in CO and HC emssions can be achieved when catalyst temperatures exceed 250° C. This temperature is not achieved unless the engine is operated at moderate to high loads. Figure 2 shows that above 300° C this particular catalyst significantly increased the emission of NO_2. This occurs because at the higher temperatures the oxidation of NO to NO_2 is enhanced. Figure 3 shows that SO_2 is oxidized to SO_4 at catalyst temperatures above 300° C. SO_4 emissions are dependent upon fuel sulfur content and can be minimized by using fuel that has a sulfur content of less than 0.1 pct.

QUANTITATIVE EVALUATION OF CATALYTIC CONVERTERS

Laboratory tests have been performed by government agencies, private industry, and others to determine the effectiveness of using converters as an emission control device. These tests can be classified in categories; steady-state, transient, durability, and in-mine. Steady-state tests are performed by operating the engine at constant load and speed conditions and do not include periods of acceleration or deceleration. Transient engine tests, on the other hand, have periods of changing load and speed conditions, and more closely simulate equipment operation. Durability tests evaluate the long-term effectiveness of converters and in-mine tests evaluate the performance under actual working conditions.

ENGINE OPERATION TEST RESULTS

Eccleston (8) tested a monolithic converter on a Deutz F6M-212W engine using the Environmental Protection Agency 13-mode heavy-duty test cycle and No. 2 diesel fuel containing 0.20 pct sulfur. Emissions were measured before and after the converter. The results show that for the heavy-load conditions the converter decreased HC by 33 pct, CO by 78 pct, while NO_2 increased by 12 pct and SO_4 increased from 2.3 ppm to 25 ppm (983 pct). Under light-load conditions, HC decreased 28 pct, CO decreased 39 pct, SO_4 decreased 10 pct, and there was no significant change in NO_2. The average results over all 13 modes showed, that HC decreased 35 pct, CO 63 pct, NO_2 12 pct, and SO_4 increased 455 pct. These tests were repeated for a Deutz F6L-912W engine with similar results.

Acres (9) evaluated a ceramic substrate platinum catalyst at three different conditions; idle, medium, and heavy loads. At idle load and speed the catalyst had no effect on CO emissions, at medium loads the conversion efficiency was 80 pct, and at high loads the conversion efficiency was 84 pct. Acres also measured the NO_x concentration and found no significant effects.

Sercombe (10) tested the effectiveness of two platinum catalysts using a direct-injection diesel engine following the California Air Resources Board 13-mode cycle. Emissions were observed in each mode, and cycle weighted averages were obtained. The results from the first catalyst tested indicate that the HC emissions decreased 68 pct and the CO emissions decreased 90 pct. The tests of a second catalyst showed a reduction of HC emissions of 64 pct and CO emissions of 87 pct. Neither catalyst showed a significant change in NO_2 emissions.

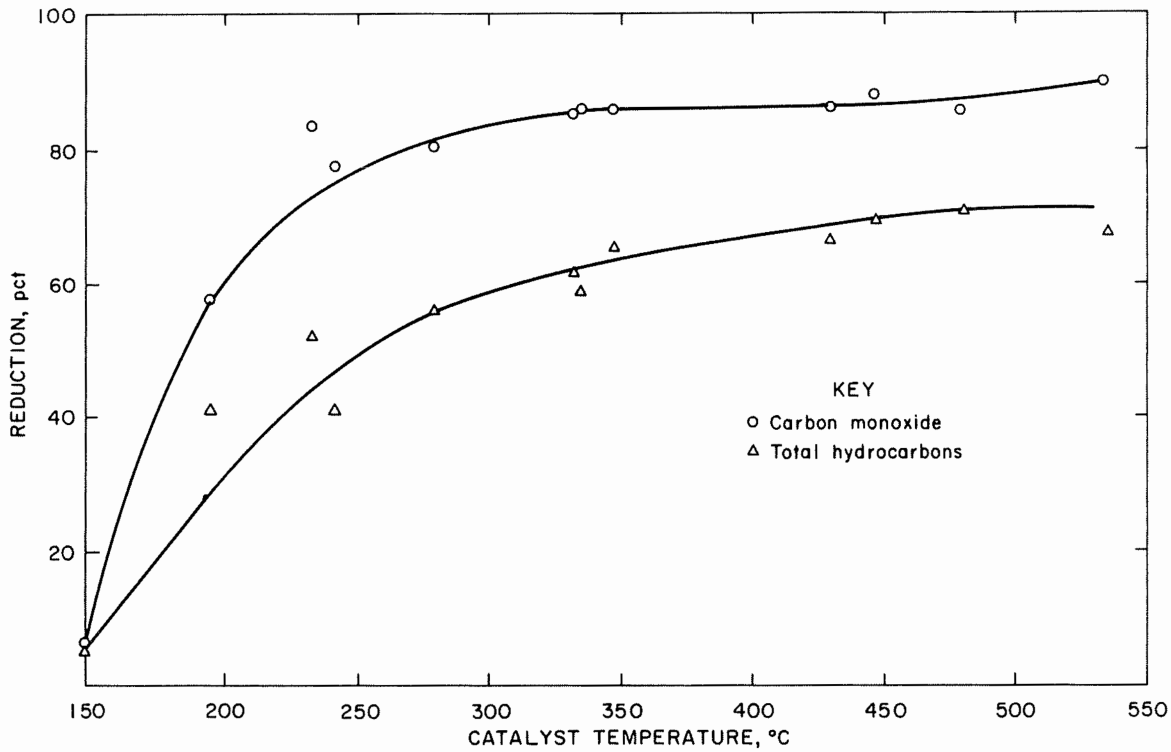

FIGURE 1.–Carbon monoxide and total hydrocarbons reduction as function of converter temperature.

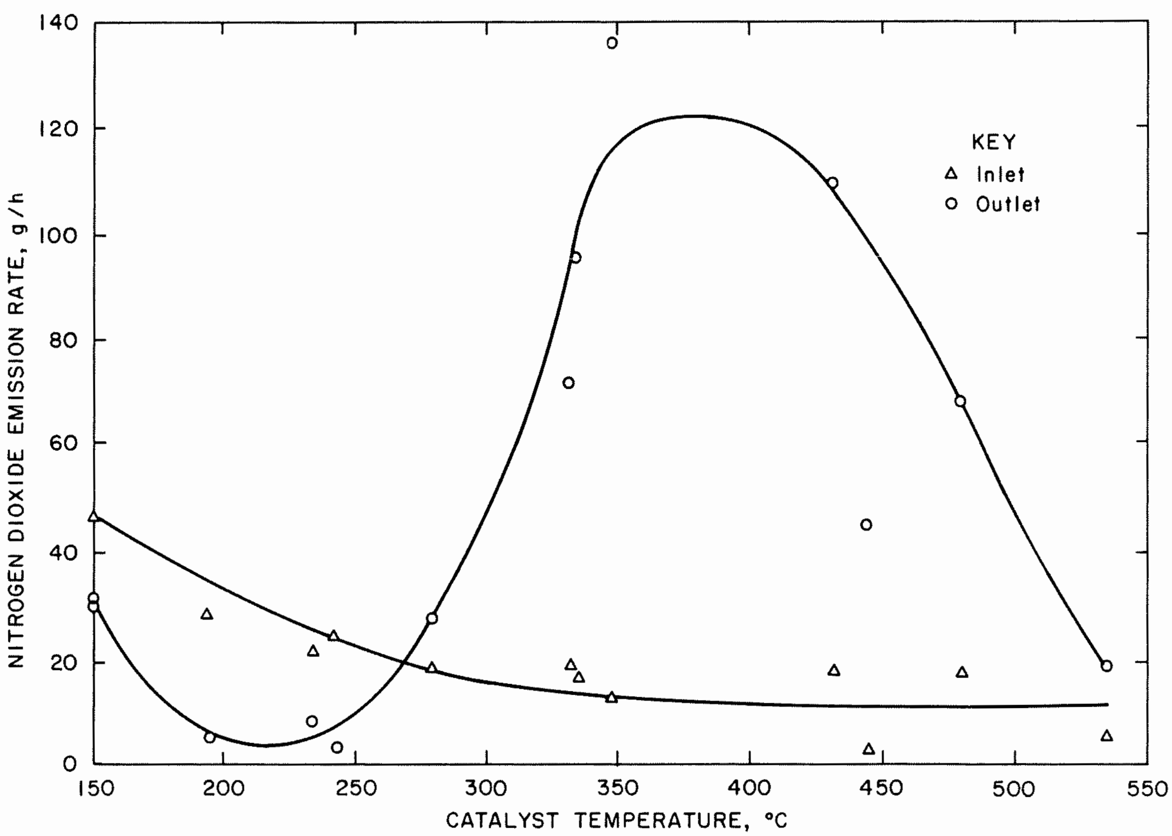

FIGURE 2.–Nitrogen dioxide emission as function of converter temperature.

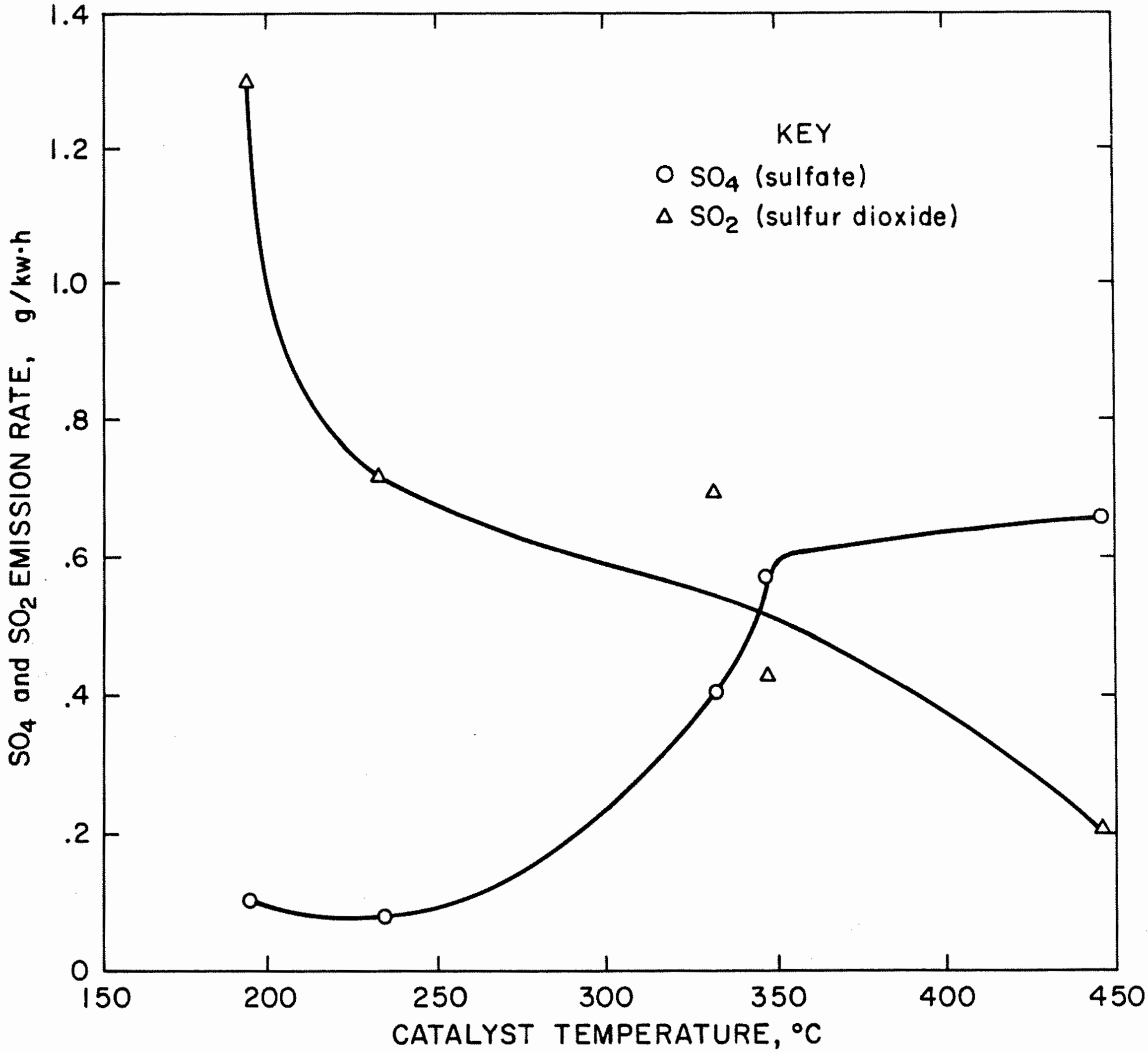

FIGURE 3.–Sulfate and sulfur dioxide emissions as function of converter temperature.

Evaluation of catalytic converters operated under steady-state engine conditions has shown that a high conversion efficiency of CO occurs when the engine is operated at high engine loads, but that the efficiency is significantly less for the lighter loads. Sulfate emissions are greatly increased by the converter for the heavy loads, but not for the light loads.

Bykowski (11) tested an eight-cylinder, indirect-injection, 5.7-L diesel engine equipped with a monolithic converter using the transient Federal test procedure and fuel with 0.29 pct sulfur. The catalyst decreased the emission of CO by approximately 90 pct and HC by approximately 60 pct. A slight increase in NO_x emissions and a 500-pct increase in sulfate emissions were observed. The same data were collected after the vehicle had been operated 190 miles. No change in converter operation was detected. During the tests, the maximum temperature at the converter inlet was 196° C and the minimum temperature was 76° C.

Steady-state data were used to estimate the cycle emissions for a mining load-haul-dump (LHD) vehicle equipped with a Deutz F6L 714 engine and a PTX 623 monolithic converter (5). The cycle was divided into six steady-state modes, each of different duration. The total time for one complete cycle was 210 s. The emissions, before and after the converter for each of the six modes, were averaged over time to yield integrated values for each pollutant for the entire cycle. The converter removed 85 pct of the CO and caused a small reduction in NO with a corresponding increase in NO_2. A small increase in DPM was accompanied by a decrease in SO_2 with a corresponding 44-pct increase in H_2SO_4.

The efficiency of a clean converter depends mainly upon the temperature of the catalyst, which in turn is dependent on the duty cycle. Figure 1 shows 50 pct conversion of CO and HC occurs at about 190° and 250° C, respectively. As higher temperatures are attained, the conversion efficiency increases. At low exhaust temperatures, soot and carbon accumulate on the converter substrate and performance degrades rapidly.

Tests of catalytic converters operated under transient test conditions resulted in increased NO_2 emissions. DPM emissions were increased by the formation of SO_x through oxidation of fuel sulfur. This can be mitigated if low-sulfur fuel is used.

CATALYTIC CONVERTER PERFORMANCE OVER TIME

The durability of a monolithic converter was investigated by Fleming (12). Tests were conducted on a naturally aspirated, direct-injection, 10.4-L displacement diesel engine using fuel with 0.14 pct sulfur. During the evaluation NO emissions were reduced by retarding the injection timing by three degrees and by employing exhaust gas recirculation. To reduce CO and HC emissions, a monolithic platinum converter was installed on each bank of cylinders. The injection nozzles were modified to aid in lowering HC emissions. At 125-h intervals, the emissions of CO, HC, and NO_2 were measured for thirteen 10-min periods and averaged. Samples of the exhaust were taken upstream of the converter during the first 5 min and downstream of the converter for the last 5 min during each period. The results indicated that the converter oxidized about 90 pct CO at the beginning of the test and about 88 pct after 1,060 h. Initially, 70 pct of the HC's were converted, but after 1,060 h only 60 pct were converted. The converters caused a small increase in fuel consumption, and smoke emissions increased slightly with operating time.

Marshall (13-14) tested several catalysts to determine their efficiency in oxidizing CO and HC, and also tested a platinum-based pellet-type converter for durability. For these tests, a duty cycle was chosen to simulate the operation of a utility vehicle with an overall load factor approximately 25 pct. The converter was most effective at reducing CO and HC concentrations when operated above 300° C and maintained a relatively constant conversion efficiency for the first 1,000 h. After that time, the CO and HC emissions increased both before and after the converter. The increase was attributed to fuel-injection nozzle malfunctions, such as a poor spray pattern or leakage of the injectors.

Inadequate engine maintenance can adversely affect emissions and degrade catalytic converter performance (15). All catalysts tested greatly increased the emissions of SO_3 and these increases were related to the fuel sulfur content. Because of these increased emissions, the investigators recognized that other control measures, such as ceramic particle filters or water scrubbers, might have to be used in conjunction with catalyst systems to ensure satisfactory performance.

These evaluations establish that catalytic converters maintain their performance up to 1,000 h under laboratory conditions, proper engine maintenance is required for best performance.

IN-MINE TESTS

A West German research group installed PTX-4D and 6D monolithic converters on mining vehicles (16), which were periodically taken out of service and checked for emissions after operation in two drifts. Drift A had eight PTX-4D catalysts and a total of 18,500 h of operation were accumulated on the eight converters for an average of 3,083 h each. Two converters failed during this test after 925 and 1,732 h, respectively. In drift B, one PTX-6D failed after 1,436 h and two PTX-4D failed after 1,156 and 1,432 h, respectively. The engines equipped with PTX-4D converters should have used the larger PTX-5D, but because of space constraints the 4D's were substituted. This may explain the high failure rate of the 4D's. The remaining eight PTX-6D's accumulated 17,500 h of operation for an average of 2,188 h each.

The converters were tested under two conditions, partial load and full load. For the PTX-4D, the initial conversion efficiency was 87 pct for CO, which decreased to 65 pct after 200 operating h. The efficiency dropped to 30 pct between 1,000 to 1,500 operating h at which point the converters were cleaned by burning off the collected soot at high temperatures, a process called regeneration. Regeneration resulted in improved efficiency, but the initial conversion efficiency was not restored. The PTX-6D converter tested at full load had an initial CO conversion efficiency of 90 pct, which gradually decreased to 50 pct after 2,000 operating h.

Table 2 summarizes the catalytic converter test results reported in the literature. These results were obtained on a variety of engines operated under different conditions, with different types of catalytic converters. Specific information can be obtained from the reference cited.

The average reduction for CO was 77 pct and 54 pct for HC for all tests summarized in table 2. The results for NO_2 and SO_4 indicate an increase in emissions.

TABLE 2. - Catalytic converter emission reductions, percent

	CO	HC	NO_2	SO_4		CO	HC	NO_2	SO_4
Reference 4.....	85	NG	INC	[1]+441	Reference 11....	90	60	NC	+500
Reference 8.....	63	35	12	+455	Reference 12....	90	70	NG	NG
	78	33	+12	+983		88	60	NG	NG
	39	28	NC	+10	Reference 14....	[2]752	[2]772	NG	NG
Reference 9.....	80	NG	NC	NG		[2]802	[2]452	NG	NG
	84	NG	NC	NG	Reference 16....	87	NG	NG	NG
Reference 10....	90	68	NC	NG		30	NG	NG	NG
	87	64	NC	NG	Reference 17....	90	NG	+25	+4,251

\+ Percent increase.
INC Increased.
NC No change.
NG Not given.
[1]Measured as H_2SO_4.
[2]Approximate values.

DISCUSSION

Diesel exhaust contains many substances, each of which may be affected by the use of a control device. To ensure that the use of a specific device to control one pollutant does not lead to an overall degradation of emissions, some means to judge the overall effectiveness of a treatment device is required. In 1981, three criteria were recommended by a joint Canadian-United States research panel (18). These were (1) an index for ventilation, (2) the Ames bioassay, which tests for mutagenicity, is one indicator of potential carcinogenity, and (3) measurement of the concentrations of six polycyclic aromatic HC, some of which are known carcinogens. These criteria are used to evaluate catalytic control systems.

The air quality index (AQI) was formulated (19) to provide a single qualitative indicator of the risk associated with exposure to diesel emissions. AQI was defined as,

$$\begin{aligned} AQI = {} & (CO)/TLV \text{ for } CO \\ & + (NO)/TLV \text{ for } NO \\ & + (RCD)/TLV \text{ for } RCD \\ & + 1.5\ [(SO_2)/TLV \text{ for } SO_2 \\ & + (RCD)/TLV \text{ for } RCD] \\ & + 1.2\ [(NO_2)/TLV \text{ for } NO_2 \\ & + (RCD)/TLV \text{ for } RCD], \end{aligned}$$

where the gaseous components are expressed in parts per million and respirable combustible dust (RCD) is expressed in milligrams per cubic meter. In practice AQI is determined at different locations in the mine, and AQI decreases as the exhaust is diluted.

The denominator values for CO, NO, NO_2, and SO_2 were originally the 1978 recommended American Conference of Governmental Industrial Hygienists (ACGIH) threshold limit values (TLV). The current TLV's should be used; 50 ppm for CO, 25 ppm for NO, 3 ppm for NO_2, and 2 ppm for SO_2 (20). There is no TLV for RCD so the respirable coal mine dust standard of 2.0 mg/m^3 was proposed for RCD. The index applies the ACGIH additive principle for the presence of multiple pollutants (20) and incorporates two factors, 1.5 and 1.2, to account for possible synergistic effects.

It was suggested that (19) an AQI value between 3 and 4 be interpreted as posing a moderate health risk, and a value greater than 4 poses a serious health threat, which required either reductions in pollutant concentrations or increased ventilation. In any case no individual pollutant should exceed its TLV. For more detailed information on the AQI refer to references 19 and 21 through 23.

Mogan (24) extended the use of the AQI by applying the definition to raw and treated diesel exhaust and refers to this measure as the EQI. Since the goal for the AQI value in underground mines was 3, measuring the concentrations of the contaminants at the exhaust pipe with and without emission controls, calculating the EQI, and dividing by 3 provides an estimate of the number of equivalent volumes of fresh air needed to dilute the exhaust to achieve an acceptable AQI. This application of the EQI assumes that the chemical composition and quantity of exhaust products does not change with dilution.

Mogan (17) reported that the EQI for a bare engine was 215; for a converter equipped engine it was 397. The higher EQI was mainly due to the increase in RCD caused by the increase in sulfate. Sulfate emissions are directly related to the fuel sulfur content, and the increase in RCD owing to an increase in sulfates can be minimized by using a low-sulfur fuel. The fuel used in these tests contained 0.25 pct sulfur. Diesel fuel with less than 0.1 pct sulfur content should be used to minimize sulfate emissions (6, 25).

Similar tests were performed (5) on a Deutz F6L 714 and a Detroit 8V71N diesel engines equipped with monolithic converters. The emissions were measured for each mode and averaged to yield integrated values that were substituted into the EQI expression. For the Deutz engine, the EQI increased from 140 to 417 when the converter was used, and it increased by a factor of 2.5 for the Detroit engine. The increase in the AQI was due to increases in the emissions of NO_2 and sulfates.

Mogan (26) summarized estimates derived from data from a number of investigations by using the EQI to compare catalytic converter performance on engines tested with No. 2 diesel fuel. The results are shown in table 3. The percent of baseline columns in the table are determined by dividing the EQI for the exhaust control by the EQI for the bare engine and multiplying by the percent of baseline for the bare engine. This amounts to defining the performance of the untreated indirect-injection engine as a unit of EQI. Catalytic converters increased the EQI in some cases by more than a factor of 2. The table shows the benefits are derived when converters are used with low-sulfur fuel and water scrubbers.

Hunter (27) conducted tests to determine the effect that a monolithic oxidative catalytic converter had on emissions from a Caterpillar 3208 diesel engine using No. 2 fuel. Test results from four steady-state engine modes, showed that large increases in the amounts of DPM and sulfates and reduction of the soluble organic fraction (SOF) occurs when converters are used. In the mode exhibiting the most extreme case, SO_4 emission increased from a fraction of gram per kilowatt hour to more than 2.5 g/kW·h as the exhaust passed through the catalyst. Similar results were obtained for the other three modes.

TABLE 3. - Effect of catalytic converters on EQI

Exhaust treatment	DI engine		IDI engine	
	EQI	Pct of baseline	EQI	Pct of baseline
Bare	224	113	199	100
Monolithic converter	314	158	484	243
Pellet converter	314	158	292	147
Monolithic converter plus low-sulfur fuel	102	51	197	99
Pellet converter plus low-sulfur fuel	102	51	84	42
Monolithic converter plus water scrubber[1]	103	52	197	99
Pellet converter plus water scrubber	103	52	84	42
Monolithic converter plus water scrubber and low-sulfur fuel	74	37	133	67
Pellet converter plus water scrubber and low-sulfur fuel	74	37	55	28

DI Direct-injection. IDI Indirect-injection.
[1]For coal operations, a water scrubber is required to cool the exhaust.

An explanation offered for the increase in DPM is that it is caused by the dehydrogenation of the organic compounds present in the exhaust (27). This reaction results in the formation of solid carbon and low hydrogen-to-carbon ratio hydrocarbons. The fact that the SOF was decreased in all test modes is consistent with this explanation.

A similar catalyst was tested (28) on a single-cylinder diesel engine operated under different conditions and a 70-pct reduction of both DPM and SOF was obtained when catalytic temperatures exceeded 250° C. For polyaromatic HC and other high-molecular-weight compounds in the SOF, a 70-pct reduction was achieved at temperatures above 170° C. This result was attributed to the high-molecular-weight compounds adsorbed onto the carbon particulate and thereby being brought into direct contact with the catalyst surface.

The effect of monolithic and pelletized catalytic converters on mutagen levels in a salt mine was assessed (29), and a fivefold increase in mutagen level was observed when the monolithic converter was used. This finding is similar to laboratory data reported by Hunter (27). In Hunter's study the Ames bioassay showed that mutagenicity per gram of soluble organic material increased when the catalyst was used, but was partially offset by a reduction in the mass of SOF emitted.

The pelletized catalytic converter resulted in a thirtyfold increase in mutagen levels over the untreated engine despite a substantial decrease in polynuclear aromatic HC. This decrease in polynuclear aromatic HC accompanied by an increase in mutagenic activity results from nitration of the polynuclear aromatic HC, which are trapped and later released by the pelletized converter. Evidence suggests that very strong mutagens such as the dinitropyrenes are formed (30-31). Even though there is no direct relationship between Ames activity and adverse health effects it has been recommended (32) that the Ames test and other in vitro tests be used to screen engineering alternatives and that the information obtained be combined with data from animal and epidemiological studies to aid in the decision making process.

CONCLUSIONS AND RECOMMENDATIONS

1. Catalytic converters are effective in reducing CO, HC, and odors when the exhaust temperature remains above 250° C. Converter temperature is dependent upon engine duty cycle, and engines operating under moderate to heavy loads generally have high enough temperatures to allow efficient converter operation.

2. Catalytic converters slightly increase NO_x emissions.

3. Sulfate emissions are increased by the use of catalytic converters, but this increase can be offset by using fuel containing 0.1 pct sulfur or less. If low-sulfur fuels are not used, the increase in sulfate and NO_2 emissions can offset any advantages of the catalytic converter as measured by the EQI.

4. Mutagenic activity of the SOF is increased when catalytic converters are used.

Based upon criteria recommended by a joint Canadian-United States research panel (18), catalytic converters should have very limited use in underground mines. Converters can reduce CO, HC, and odor emissions, but these reductions are frequently offset by increased levels of the more toxic pollutants, NO_2 and sulfates, thus causing an increase in the EQI. In addition, studies have shown that catalytic converters produce soluble organic compounds with high mutagenic activity. Although mutagenic activity is not directly linked to adverse health effects, these assays have been recommended for screening engineering alternatives and mine operators should be aware of the results. Vehicles equipped with converters should operate under moderate- to heavy-load conditions and use fuel with a sulfur content less than 0.1 pct.

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APPENDIX.--ABBREVIATIONS AND SYMBOLS USED IN THIS REPORT

This listing does not include unit of measure abbreviations, which are listed after the table of contents, or abbreviations that are used and identified in the tables.

ACGIH - American Conference of Governmental Industrial Hygienists.

AQI - Air quality index.

DPM - Diesel particulate matter.

EQI - Emissions quality index.

LHD - Load-haul-dump.

RCD - Respirable combustible dust.

SOF - Soluble organic fraction.

TLV - Threshold limit value.

CO - Carbon monoxide.

CO_2 - Carbon dioxide.

HC - Hydrocarbon.

H_2SO_4 - Sulfuric acid.

NO - Nitric oxide.

NO_x - Oxides of nitrogen.

NO_2 - Nitrogen dioxide.

SO_x - Oxides of sulfur.

SO_2 - Sulfur dioxide.

SO_3 - Sulfur trioxide.

SO_4 - Sulfate.

U.S. GOVERNMENT PRINTING OFFICE: 1988 — 547-000/80,068

INT.-BU.OF MINES,PGH.,PA. 28765