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Comparing diesel and GDiesel® exhaust exposures in an underground mining laboratory

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ABSTRACT: Diesel exhaust is a known carcinogen and has been linked to several negative health outcomes, yet it is commonly used in industrial settings such as underground mining. Alternative fuels such as GDiesel® (GD), a natural gas/diesel blend, have the potential to reduce health exposures and associated health effects. Mirroring a previous study in which use of GD in a 2005 Wagner load-haul-dump (LHD) vehicle with oxidation catalyst demonstrated significant reductions in diesel exhaust exposures, operator-location and area exposure samples were collected in an underground mining laboratory with diesel (D) and then GD fuel while operating a JCI LHD. Analytes of interest included total and respirable diesel particulate matter (tDPM and rDPM, respectively), total and respirable elemental and organic carbon (tEC, rEC, tOC, rOC, respectively), as well as formaldehyde (CH2O), nitric oxide (NO), and nitrogen dioxide (NO2). Use of GD resulted in non-significant reductions in median rOC, tDPM, tOC, and NO2 concentrations, non-significant increases in rDPM, rEC, tEC, and NO, and identical median CH2O concentrations. A significant decrease in NO2 exposure concentrations ($p=0.012$) and increase tEC and rEC exposure concentrations ($p=0.023$ and $p=0.024$, respectively) were observed. After controlling for environmental confounders there was no difference observed in analyte concentrations between the two fuels. Further research is needed to determine whether GD alone can significantly reduce human exposures across vehicles and pollution configuration types.

1 INTRODUCTION

1.1 *Background*

Diesel fuel (D) is used in a variety of applications for several industries, including transportation, agriculture, railroads, construction, and mining. In 2012 the International Agency for Research on Cancer designated D exhaust as a Group 1 carcinogen in humans (WHO 2012). Large longitudinal studies performed with underground miners have linked their exposure to D with increased risk of lung cancer (Attfield et al 2012, Silverman et al 2012). Exposure to D engine emissions is associated with adverse health effects including: chronic bronchitis, respiratory tract infections, asthma exacerbation, and increased cardiovascular morbidity and mortality (Bonauto et al 2007, Culp et al 2011, Kagawa 2002, Taylor 2006). Given the nature of D exposures in the mining industry and the many associated health effects (Attfield et al 2012, Silverman et al 2012, Sydbom et al 2001), reducing engine emissions has become an industrial hygiene priority. In the US, there are nearly 14,000 mine sites that employ over 210,000 individuals in the workforce (BLS 2017, MSHA 2014). Both underground and surface mines predominantly use diesel-powered vehicles at their operations to perform tasks such as drilling, mucking, and hauling. Miners are regularly exposed to D during their work, especially in areas where there is limited ventilation.

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1.2 *Regulatory compliance*

In 2008 the Mine Safety and Health Administration (MSHA) published updated standards for the allowable concentration of respirable (<1.0 μm with impactor) diesel particulate matter (rDPM) in an effort to control D exhaust exposures. The administration has also recently published a request for information (RFI) regarding diesel exhaust exposures and controls (MSHA 2016). The permissible exposure limit (PEL) of 160 $\mu g/m^3$ as total (combined inorganic and organic) carbon (CFR 2016) is monitored through air sampling and processing per the National Institute for Occupational Safety and Health (NIOSH) analytical method 5040. Despite the installation of controls such as mine ventilation, the underground rDPM exposure standard is frequently exceeded. Since 2000, MSHA has made detailed information regarding the occupational health exposure assessments performed during inspections available to the public. According to data since the new diesel exhaust standard, diesel particulate matter (DPM) had a higher median exposure concentration-to-PEL (ECP) ratio (0.47) than any other airborne contaminant (for which 100 or more samples were taken) except silver fumes (0.88). This ratio indicates that mine operators continue to struggle to control D exhaust exposures, and that governmental, national, and industrial organizations should focus on developing better control options.

1.3 *Biodiesel*

Given the health hazards associated with and MSHA's focus on D exposures, mine operators seek effective control methods for rDPM. Some have explored the use of biodiesel (B) fuel blends. Studies comparing D to B emissions and exposures report a decrease in total carbon output but an increase in organic carbon, aldehydes, and nitrogen dioxide (Turrio-Baldassarri et al 2004, Knothe, Sharp, & Ryan 2006, Li & McLaughlin 2005, Bugarski et al 2010, He et al 2009, Bugarski et al 2003). However, other research demonstrated an increase in aldehyde, nitrogen dioxide (NO_2), and organic carbon fraction concentrations with use of B mixtures (Bugarski et al 2010, Liu et al 2009). Despite the increasing usage of B, there is a lack of information on the human health effects of exposure to these emissions. Recent studies also suggest that exposure to B exhaust may be as or more toxic than D (Donoghue & Bates 2000, Bhavaraju et al 2013, Brak & Bates 2003, Bunger et al 2000, Armstrong et al 1998, Mehus et al 2015).

1.4 *GDiesel®*

GDiesel® (GD), an EPA-approved natural gas/diesel fuel mixture, became commercially available in 2011 and provides a second alternative to D fuel. GD is prepared by combining diesel with natural gas, using a proprietary charged-catalytic reaction. The end product is an American Society for Testing and Materials (ASTM)-designated D fuel with purportedly reduced tailpipe emissions of diesel particulate matter (DPM) and oxides of nitrogen (NO_x) compounds (NDEP 2010, ARC 2014). Our independent, simulated study using a 2005 Wagner load-haul dump (LHD) with a diesel oxidative catalyst (DOC) but no diesel particulate filter (DPF) at an underground mining laboratory showed a 66% reduction in rDPM and significant reductions for all other analytes except carbon monoxide (CO) associated with use of GD (Lutz et al 2015). LHDs are commonly used in underground mine settings and often associated with high exposures to D (Bugarski et al 2004, Bugarski et al 2003). While promising, this GD study represents a single vehicle, study design, and pollution configuration. Other studies have shown that D is influenced by a variety of factors including type of equipment, load, and pollution control equipment (Bugarski et al 2010, Liu et al 2009, Bugarski et al 2004, Durbin et al 2000, Tsai et al 2011).

1.5 *Objective*

The objective of this pilot study was to evaluate the exhaust exposures of GD, as compared to D, using a JCI LHD with Deutz engine and DOC but no DPF. We hypothesized that use of GD fuel in the JCI LHD would result in significant reductions for all analytes, comparable to those observed in our previous study.

2 METHODS

2.1 *Previous work*

Lutz et al (2015) conducted a pilot study at the University of Arizona San Xavier Underground Mining Laboratory (SX), a non-operational hard rock mine at which laboratory work is performed. Exposure to vehicle emissions was evaluated in the 'decline,' an unventilated portion of the SX with sloping underground opening for rubber-tired vehicle access. Study participants and research staff alternatively performed mucking activities with a University-owned 2005 Wagner B10-203 LHD vehicle with open cabin and DOC and observed from the decline rib. The D and GD fuels were loaded into separate fuel tanks, and the LHD was operated for approximately one hour after switching to the GD tank before beginning testing. Personal and area exposure samples to rDPM, rEC, rOC, tDPM, tEC, tOC, formaldehyde (CH_2O), acetaldehyde, naphthalene, NO, NO_2, and CO were collected. For a more detailed description, please reference the publication (Lutz et al 2015). The work by Lutz et al (2015) was limited to a single vehicle and pollution control configuration – factors that have been shown to influence both exhaust emissions and exposures (Bugarski et al 2003, Bugarski et al 2010, Durbin et al 2000, Tsai et al 2011).

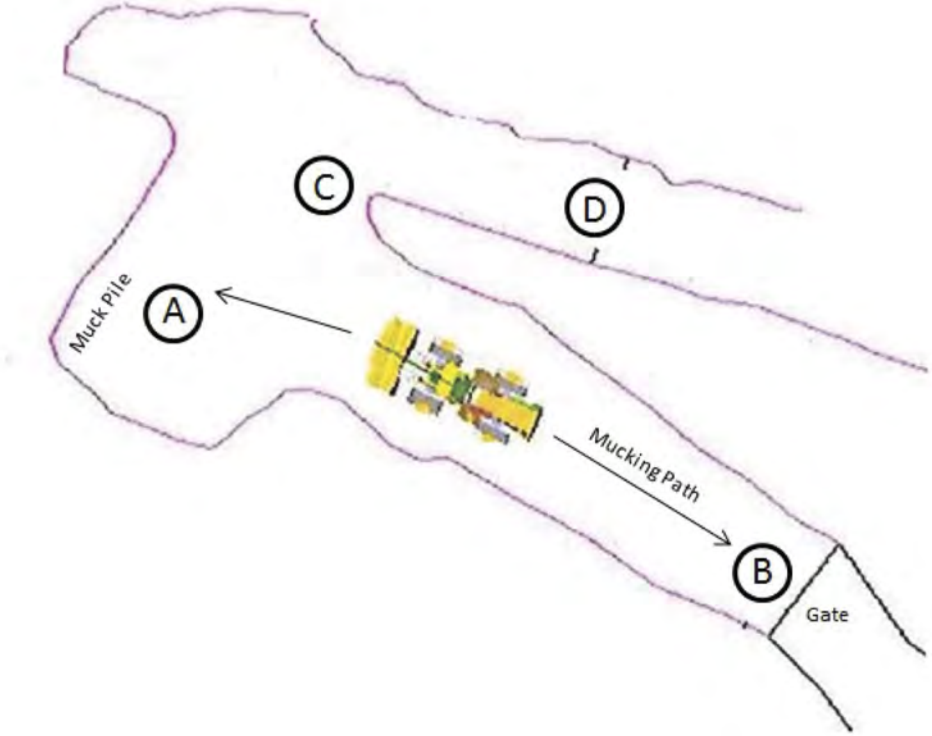

Figure 1. Depiction of the decline at the University of Arizona San Xavier Underground Mining Laboratory.

Note: Image not to scale.

2.2 *Fuels, equipment and location*

For the current study, ultra-low sulfur D #2 was obtained from a regional distributor (Arizona Petroleum, Tucson, AZ). The GD fuel #2 was purchased directly from the producer (Advance Refining Concepts, LLC., Reno, NV).

A JCI LHD with inline, 4-cylinder, 55 horsepower at 2300 RPM Deutz engine (motor number 7534-462) with an Engine Control Systems Purimuffler (part number A17-0313) DOC, but no DPF, was borrowed from an underground mine in the region. It also had an open cabin.

This study was conducted at the SX decline (see Figure 1 below). Similar to the previous study (Lutz et al 2015), the decline's gate (position B) was covered with a vinyl tarp and the metal door (position D) entering the 'adit' level was closed at the start of each exposure session to minimize airflow into and out of the decline. The mine fan was not operated during the study sessions.

2.3 *Procedure*

Research staff operated the vehicle for five consecutive days using D. Two 200-minute exposure sessions were completed each day, with one operator-location and one area sample taken during each session. In order to change from D to GD fuel, the LHD fuel tank was emptied using a fuel pump, filled with GD, and then driven for approximately one hour before beginning the first GD exposure session. The LHD was then operated for a total of four days with GD fuel. Due to water constraints in the mine and potential for significant dust dispersion, mucking was not performed. The LHD bucket was filled once at the beginning of the study and the equipment simply driven back and forth on the 30-meter decline mucking path by the same operator. The bucket was not emptied until the end of the study. Each day the vehicle was not allowed to idle before the first session and, before beginning the second, the tarp was removed for 30 minutes to allow natural ventilation to completely disperse carbon monoxide. CO dispersion was confirmed with an MSA 4X Altair gas monitor (Cranberry Township, PA).

2.4 *Exposure assessment*

Universal PCXR 8 (SKC West, Inc., Fullerton, CA) and Escort ELF (Zefon International, Inc., Ocala, FL) air sampling pumps, along with associated sampling tubing and holders or cassettes, were placed in pockets of or clipped to safety vests. Because the operator's cabin was small it was not possible for the operator to wear the vest. One was therefore placed in the cabin within approximately 36 inches of the operator's breathing zone. A second 'area' vest was placed on the rib wall at a height of approximately six feet (position C).

Personal integrated sample collection and analysis was performed in accordance with the NIOSH manual of analytical methods (NMAM). Specifically, GS-1 Respirable Cyclones with 37 mm jeweled impactor and flow rate of 1.7 L/min (NIOSH 2003a) were used for rDPM sampling. As rDPM is simply the sum of rEC and rOC, these were also obtained from the sample data. A 37 mm open face quartz fiber filter with flow rate of 2.0 L/min (NIOSH 2003a) was utilized to sample tDPM, tEC and tOC. A tandem triethanolamine/oxidizer with flow rate 0.025 L/min (NIOSH 1994) was used to collect NO and NO_2 samples. Sorbent tubes containing silica gel and a flow rate of 0.1 L/min (NIOSH 2003b) were used in formaldehyde sample collection. All sampling media were produced by the same manufacturer (SKC West, Inc., Fullerton, CA). Pre- and post-sampling calibration was performed using a Bios Drycal® Defender 520 calibrator (Mesa Labs, Inc., Butler, NJ) at the SX Training Center. An independent AIHA-accredited industrial hygiene laboratory performed laboratory analysis (Galson Laboratories, East Syracuse, NY).

2.5 *Data analysis*

Laboratory results were copied, pasted to, and then formatted in a database table. Reported concentrations were time-weighted over an 8-hour period (see Figure 1 below) to obtain the time-weighted average (TWA_8). Environmental data were averaged over the time period corresponding to each exposure session and paired using database relationships. All analysis was performed using STATA 12.0 (StataCorp, College Station, TX). Unadjusted differences in distribution were measured using the Wilcoxon rank-sum test. Multiple linear regression was used to identify which of the four environmental measures had the least potential to be a confounder. In every case except for formaldehyde, for which temperature had the least potential, the confidence interval for wind speed was most likely to contain zero. To adjust for environmental confounders, binary logistic regression was then performed using fuel type as the output, with the TWA_8 and three remaining environmental measures as inputs. An alpha error threshold of 0.05 was used throughout.

$$TWA_8 = \frac{\text{Concentration}\left(\frac{\text{mg}}{\text{m}^3}\text{ or ppm}\right) * \text{Sampling Time (minutes)}}{480\text{ (minutes)}} \tag{1}$$

Using fences at 150% of the interquartile range, seven outliers were identified, by fuel type, and removed from the D (rOC, NO_2) and GD (rDPM, rEC, rOC, NO_2) datasets. Statistical analysis was performed with and without outliers. In three of the four datasets there were no changes in conclusions observed when outliers were included versus not included. However, the rEC sample set changed when outliers were included. The rEC results with and without outliers are provided in the results section.

3 RESULTS

3.1 *Overview*

There were a total of 20 D and 16 GD samples collected. Variations in samples collected were due to LHD equipment malfunction during two of the GD sampling days. For the unadjusted rank-sum difference, median rOC, tDPM, tOC, and NO_2 TWA_8 exposure concentrations were reduced with use of GD (see Figure 2 below), but only NO_2 was significantly so (p=0.012). Use of GD resulted in increased median concentrations of rDPM, rEC, tEC, and NO, though only tEC and rEC (after removing outliers) were significantly greater (p=0.023 and p=0.024, respectively).

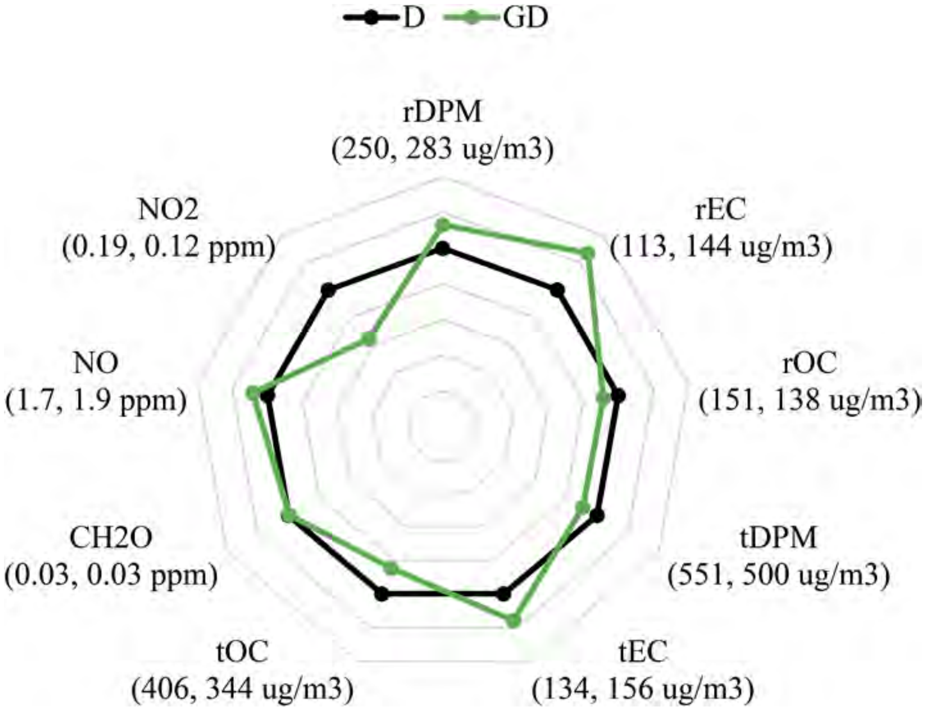

Figure 2. Median time-weighted average exposure concentrations by analyte. Note that Diesel (D) has been set as a reference and GDiesel (GD) marker placement represents a respective percentage compared to D. Abbreviations: respirable diesel particulate matter (rDPM), respirable elemental carbon (rEC), respirable organic carbon (rOC), total diesel particulate matter (tDPM), total elemental carbon (tEC), total organic carbon (tOC), formaldehyde (CH_2O), nitric oxide (NO), and nitrogen dioxide (NO_2).

Median formaldehyde concentrations were identical. For more detailed results, see Tables 1, 2 and 3 below. After controlling for confounders, the likelihood of a given TWA_8 being associated with either fuel was not statistically significant for any analytes.

Table 1. Descriptive statistics for gaseous analytes formaldehyde (CH_2O), nitric oxide (NO), and nitrogen dioxide (NO_2), by fuel. Abbreviations: diesel fuel (D), and GDiesel fuel (GD). All units in ppm.

Analyte	CH_2O		NO		NO_2	
Fuel	D	GD	D	GD	D	GD
N	19	16	20	16	20	16
Mean	0.035	0.033	1.946	2.976	0.345	0.159
SD	0.013	0.010	0.783	2.322	0.589	0.098
Median	0.030	0.030	1.725	1.868	0.186	0.120*
Q1	0.027	0.025	1.469	1.470	0.156	0.102
Q3	0.041	0.040	2.619	4.100	0.293	0.171
Min	0.020	0.020	0.413	1.000	0.088	0.076
Max	0.058	0.052	3.559	7.877	2.813	0.417

* Significantly different from D at $p<0.05$.

Table 2. Descriptive statistics for respirable particulates: respirable diesel particulate matter (rDPM), respirable elemental carbon (rEC), and respirable organic carbon (rOC), by fuel. Abbreviations: Diesel fuel (D), and GDiesel fuel (GD). All units in µg/m^3.

Analyte	rDPM		rEC			rOC	
Fuel	D	GD	D	GD	GD[a]	D	GD
N	20	15	20	15	14	20	15
Mean	274.9	305.2	115.6	137.6	144.4	159.3	167.8
SD	112.2	84.3	36.4	32.6	19.5	85.4	100.8
Median	250.4	283.4	113.4	140.9	143.8*	150.9	138.2
Q1	203.5	245.0	90.5	126.9	130.7	92.7	114.3
Q3	347.8	336.7	146.6	160.7	160.7	200.8	172.5
Min	111.9	217.8	50.8	41.4	115.3	53.9	93.5
Max	533.8	549.8	177.6	183.6	183.6	377.4	507.5

[a] With outliers removed,
* significantly different from D at $p<0.05$.

Table 3. Descriptive statistics for total particulates: total diesel particulate matter (tDPM), total elemental carbon (tEC), and total organic carbon (tOC), by fuel. Abbreviations: Diesel fuel (D), and GDiesel fuel (GD). All units in µg/m^3.

Analyte	tDPM		tEC		tOC	
Fuel	D	GD	D	GD	D	GD
N	20	16	20	16	20	16
Mean	551.7	526.4	136.2	161.6	413.3	361.4
SD	141.8	75.4	34.0	20.0	120.6	52.8
Median	551.1	500.0	134.3	155.8*	405.9	343.6
Q1	464.1	466.4	118.4	149.1	330.4	321.4
Q3	620.5	566.7	159.4	176.3	484.1	396.7
Min	283.3	460.6	66.7	123.3	212.5	298.8
Max	843.3	683.3	195.5	195.5	690.0	469.8

* Significantly different from D at $p<0.05$.

4 DISCUSSION

4.1 *Findings*

This study demonstrated little statistical difference between D and GD exhaust exposures in a JCI LHD with DOC at the SX mine. The results of this study are inconsistent with results observed by Lutz et. al. (2015), while using the Wagner LHD with DOC. Previously rDPM, rEC, rOC, tDPM, tEC, tOC, CH_2O, NO, and NO_2 were significantly reduced, whereas this study observed a significant decrease in NO_2 alone, with significant increases in rEC and tEC. These findings are also inconsistent with those described by the manufacturer of GD (NDEP 2010, ARC 2014). However, the Nevada Division of Environmental Protection (2010) reported that use of GD may cause small increases in total hydrocarbon emissions. This research is consistent with previous work that has demonstrated variability in exposure reductions based on the use of different equipment and pollution control configurations (Bugarski et al 2004, Bugarski et al 2003, Bugarski et al 2010, Durbin et al 2000, Tsai et al 2011).

Compared to regular D fuel, this pilot study demonstrated a reduction in NO_2 and increase in tEC and rEC with use of GD, with no change in rDPM or other analytes. Given that the MSHA standard is based on rDPM exposures, this work makes it unclear whether use of GD would likely increase compliance with the federal regulation. Despite MSHA's focus on rDPM, other components of D contribute to adverse health effects. Insufficient data are available to indicate whether reducing one or some of the contaminant exposures from D influences observed health effects.

Use of compressed natural gas in D engines has been shown to reduce particulate emissions, polycyclic aromatic hydrocarbons, formaldehyde, and genotoxic activity, compared to D fuel Fritz & Egbuonu 1992, Greenwood et al 1996, Ayala et al 2002, Turrio-Baldassarri et al 2006). Despite this study's findings, a reduction in exposures with GD fuel would seem reasonable given the apparently cleaner burning qualities of natural gas.

Differences observed between fuel types are likely a function of the fuels' chemical composition, vehicle engine conditions, such as temperature of combustion and concentration of oxygen present, and emission source controls, such as DOCs and DPFs. For example, differences in DPM could be due to more complete combustion from higher engine temperatures, or impurities and hydrocarbon chain lengths found in each fuel. In addition, EC is released directly from the incomplete combustion of fossil fuels while OC, an aggregate of hundreds of individual compounds spanning a wide range of chemical and thermodynamic properties, is formed by a variety of processes, suggesting perhaps more complete combustion with D in the JCI LHD. Variation in NO_x concentrations could be due to higher combustion temperatures, as well as interaction with a DOC. Higher engine temperatures and increased oxygen content may lead to conditions that increase the formation of short hydrocarbons, such as formaldehyde and acetaldehyde. Increased combustion temperature may therefore explain the decrease in DPM but increase in aldehydes associated with B in some studies.

4.2 *Limitations*

The limitations of our pilot study include use of a single vehicle and single pollution control configuration (DOC, no DPF). In addition, this study occurred at a naturally ventilated underground laboratory, rather than an operational mine where other pieces of equipment and active, forced ventilation are typically present. During two of the scheduled GD exposure sessions the LHD would not start due to heating of the starter solenoid. Heating of the engine over time, as well as the chosen order of fuel use, may have biased these results. While efforts were taken to minimize airflow into and out of the mine, outdoor environmental conditions such as wind speed and barometric pressure may have influenced our observations.

4.3 *Conclusions*

The current lack of information regarding the alternative fuel's emissions suggests that further research is needed to determine whether GD can significantly reduce occupational and environmental exposures across vehicle and pollution configuration types. We recommend research that utilizes a crossover design, targets several of the most commonly used diesel-fueled vehicles in the mining industry and monitors exposures and health effects over an extended period. The current study, coupled with previous research [28], demonstrates how changes to the types of equipment used, driving conditions and environmental conditions make predicting exposure risks from diesel exhaust extremely difficult.

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