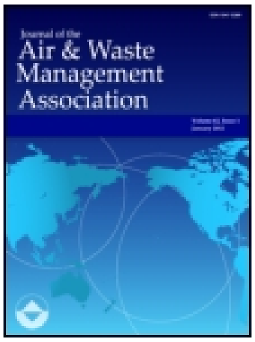

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The Effect of Diesel Fuel Sulfur Content on Particulate Matter Emissions for a Nonroad Diesel Generator

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ABSTRACT
The effect of sulfur content on diesel particulate matter (DPM) emissions was studied using a diesel generator (Generac Model SD080, rated at 80 kW) as the emission source to simulate nonroad diesel emissions. A load simulator was used to apply loads to the generator at 0, 25, 50, and 75 kW, respectively. Three diesel fuels containing 500, 2100, and 3700 ppm sulfur by weight were selected as generator fuels. The U.S. Environmental Protection Agency sampling Method 5 "Determination of Particulate Matter Emissions from Stationary Sources" together with Method 1A "Sample and Velocity Traverses for Stationary Sources with Small Stacks or Ducts" was adopted as a reference method for measurement of the exhaust gas flow rate and DPM mass concentration. The effects of various parameters on DPM concentration have been studied, such as fuel sulfur contents, engine loads, and fuel usage rates. The increase of average DPM concentrations from 3.9 mg/Nm^3 (*n* cubic meter) at 0 kW to 36.8 mg/Nm^3 at 75 kW is strongly correlated with the increase of applied loads and sulfur content in the diesel fuel, whereas the fuel consumption rates are only a function of applied loads. An empirical correlation for estimating DPM concentration is obtained when fuel sulfur content and engine loads are known for these types of generators: $Y = Z^m(\alpha X + \beta)$, where Y is the DPM concentration, mg/m^3, Z is the fuel sulfur content, ppm$_w$ (limited to 500-3700 ppm$_w$), X is the applied load, kW, m is the constant, 0.407, α and β are the numerical coefficients, 0.0118 $\pm$ 0.0028 (95% confidence interval) and 0.4535 $\pm$ 0.1288 (95% confidence interval), respectively.

IMPLICATIONS
This study indicated the proportional correlation of DPM emissions with fuel sulfur content and applied loads for nonroad diesel machines. The various operating modes should be considered for estimating DPM emissions and the implementation of control technologies. For this diesel generator, the fuel sulfur reduction alone from 3700 ppm to 500 ppm can reduce DPM emissions by >50%. The research method of correlating DPM emission with load and sulfur content can be applied to other nonroad diesel machines.

INTRODUCTION
Diesel particulate matter (DPM) has been regarded as carcinogenic by regulatory and research agencies, such as the U.S. Environmental Protection Agency (EPA) and National Institute for Occupational Safety and Health.[1] DPM has typical size ranges between 1 μm and 5 nm, with 90% of the particles being <1 μm in diameter, and has a mass median diameter of 0.2 μm.[2–3] In addition to elemental carbon, DPM also contains polycyclic aromatic hydrocarbons (PAHs), alkylated benzenes, nitro-polycyclic aromatic hydrocarbons, and a variety of polar, oxygenated, and nitrated derivatives.[3]

The use of diesel engines as power sources for nonroad applications such as construction, mining and agriculture, and so forth, is dramatically expanding every year. It is reported that in the United States, the 5-year sales volume of generator sets of size between 56 and 130 kW from the years 1996 to 2000 totaled 103,490 units, which represented 11% of the total sales volume of all nonroad diesel equipment sales of that size during that period.[4] This corresponds to ~1,000,000 various nonroad diesel equipment of this size range being sold during that time period.

Nonroad diesel vehicles contribute to approximately 44% of total DPM and 12% nitrogen oxide (NO_x) emissions from mobile sources nationwide.[5] DPM emissions from nonroad sources are significantly higher than on-road sources.[6] As a result, EPA is proposing stringent regulations to limit the DPM emission rates by means of fuel sulfur reduction. For highway diesel fuel, fuel sulfur content will be reduced from the conventional limit of 500 ppm$_w$ to 15 ppm$_w$ in June 1, 2006.[1] For the nonroad diesel fuel, it will be reduced from 3400 ppm$_w$ to 500 ppm$_w$ in

2007 and additionally to 15 ppm_w in 2010.[1] Diesel fuel with 500 ppm_w of sulfur for underground coalmine application has already been imposed since April 25, 1997.[1]

Although the use of stationary, nonroad diesel engines have increased rapidly, and the resultant DPM emissions are of significant quantities, there are a limited number of studies on DPM emission as a function of engine and fuel properties. Therefore, the objective of this article is to determine the correlation between DPM concentration and diesel fuel sulfur in association with engine load for a nonroad source. In this project, the stationary diesel generator was selected as the emission source because of the wide use of this type of equipment in construction and industry as a power back-up/supply.

Review of Previous Literature

A variety of sulfur compounds are present in diesel fuels, which include mercaptans, sulfides, disulfides, cyclic sulfides, alkyl sulfates, sulfonic acids, sulfoxides, sulfones, and thiophenes.[7–8] Nevertheless, benzothiophenes, dibenzothiophenes , and their alkylated homologues are the most abundant species in diesel fuel.[9] Diesel fuel sulfur is known to contribute to DPM emissions.[10] During diesel fuel combustion, fuel sulfur are oxidized to SO_x. Although the majority of sulphur oxides (SO_x) is SO_2, it has been reported that as much as 5% can be oxidized to SO_3, which then combines with water to form sulfuric acid aerosols.[11] Conversion rates of fuel sulfur to DPM sulfates on particulates are in the range of 1–3%.[12] In the exhaust tail pipe, sulfates, sulfuric acid, bound water, as well as hydrocarbons with relatively low vapor pressure will condense onto soot when exhaust gases are cooled, contributing to DPM growth. It is also believed that the amount of sulfates adsorbed is responsible for the mass of hydrocarbons, including PAHs, retained in the particles and emitted into the atmosphere.[13] A previous study reported that the adsorbed sulfuric acid on the surface of soot reacts with organic compound in the exhaust to form heavy hydrocarbons, which are likely to remain in the condensed phase.[14] This reaction is so-called the scrubbing effect.[13]

Regulatory agencies have been actively pursuing the reduction of sulfur in diesel fuel as a method of reducing both SO_x and DPM emissions. It is reported that a reduction of fuel sulfur content from 0.2 to 0.05 wt% resulted in a 7–12% particulate reduction of the Euro-I limit.[16] Baranescu conducted an experiment to evaluate the effect of fuel sulfur on the brake specific particulate emissions on medium and heavy-duty trucks. Three diesel fuels with 0.05, 0.19, and 0.29% sulfur were used. The results showed that for an increase of 0.1% in fuel sulfur, brake specific particulates increased by ~0.025 g/bhp-hr, because of the addition of soluble sulfates and bound water.[10] A previous study also reported that sulfates and bound water can make up as much as 20% of the total particulate matter (PM) produced.[16]

METHODOLOGY

Emission Source

A Generac diesel generator (Model SD080, direct-injection, turbocharging, and compression-ignition) rated at 80 kW, 60 Hz, 100 hp, and 1800 rpm was used as a stationary DPM emission source. The generator fuel injecting system was modified to allow for the measurement of fuel consumption rates. A load simulator (Merlin 100 manufactured by SIMPLX) was used to simulate loads by applying steady-state banks of heaters to the generator at 0, 25, 50 and 75 kW, respectively.

Diesel Fuel Selection

Three diesel fuels with different sulfur contents were selected to evaluate the effect of sulfur content on DPM concentration in the exhaust as shown in Table 1. The diesel fuels were supplied by Vulcan Oil Co. and Wardway Fuels Inc., the local companies in Cincinnati, OH. These diesel fuels contain 3700, 2100 and 500 ppm of sulfur by weight, respectively, as determined by ASTM method D4294, and are designated as fuel A, fuel B, and fuel C, respectively, in subsequent analysis. The purpose of opting for these ranges of sulfur is to have clear evidence of the sulfur influence on the DPM concentration. Moreover, fuel C was included in this test because EPA will soon be imposing its use on nonroad diesel vehicles.

Sampling and Analytical Methods

The generator was operated at steady-state conditions for four different test loads (0, 25, 50, and 75 kW). Exhaust particulate measurements were determined by isokinetic sampling[17] using EPA Method 5 “Sampling Method for Stationary Sources” along with Method 1A “Sample and Velocity Traverses for Stationary Sources with Small Stacks or Ducts.” Three tests were conducted at each test condition for better representation. In this method, the DPM was collected on an 82-mm glass micro fiber filter (Whatman 934AH by Fisher Scientific), operated at the

Table 1. Diesel fuel properties.

	Diesel Fuels		
Parameters	**A: 3700 ppm_w S**	**B: 2100 ppm_w S**	**C: 500 ppm_w S**
Carbon, wt %	85.5	83.5	77.1
Hydrogen, wt %	13.1	12.5	11.4
Nitrogen, wt %	0.01	0.01	0.02

controlled temperature of 120 °C (250 °F). The DPM laden filter and DPM mass collected from the probe washed by acetone were put into a desiccator for at least 24 hr to remove the moisture. The total DPM mass was determined by the gravimetric method. The DPM mass concentration was simply obtained by dividing the total mass with the total sample volume that was corrected to standard conditions (1 atm, 293 °K). The DPM from the 75-kW test sample collected from the three diesel fuels was also analyzed for their total sulfur content. The mass of total sulfur was quantified by using a LECO SC-132 Sulfur Determinator, Model 781–400.

RESULTS AND DISCUSSION

Percent Isokinetic, Diesel Fuel Usages and Overall DPM Concentrations

The summary of the isokinetic sampling efficiency, diesel fuel usage, and overall DPM concentrations is presented in Table 2. The isokinetic rates ranged between 90% and 110% for all of the tests. The fuel consumption rates to power the generator with fuels A, B, and C were measured to examine their correlation with DPM emissions. The results suggest that the fuel usage is slightly fluctuated at each operating mode regardless of diesel fuel sulfur contents. This proportional increase of fuel consumption with load is mainly related to the operation modes and engine characteristics and is also supported by the generation of similar amount of exhaust flow-rates when three diesel fuels were used. In contrast to the fuel usage rates, the overall DPM concentrations also increase as a function of sulfur contents in diesel fuels.

The overall DPM concentrations and applied load as a function of the different sulfur contents in the diesel fuels can be plotted as straight-lines as shown in Figure 1. The high R^2 values suggest that the correlation can be well explained by the use of a simple least-squared regression

Table 2. Summaries of experimental results.

	Parameters						
Test #	Avg. Fuel Consumption Rates (L/hr)	DPM Concentrations (Mg/Nm^3)	Stack Gas Flow Rates (Nm^3/hr)	DPM Emission Rates (g/hr)	Moisture Contents In Stack Gas (%)	Isokinetic (%)	Approx. Avg. Excess Air (%)
Diesel A: 3700 ppm Sulfur	—	—	—	—	—	—	—
A-0–1	4.57	15.40	277.58	4.28	1.20	99.98	~560
A-0–2	—	15.74	287.49	4.52	1.60	102.86	—
A-25–1	8.83	25.21	318.16	8.02	3.40	98.78	~270
A-25–2	—	25.00	326.61	8.17	4.20	98.10	—
A-25–3	—	25.79	337.13	8.69	3.60	102.92	—
A-50–1	12.45	31.88	349.78	11.15	5.40	100.84	~195
A-50–2	—	28.67	345.46	9.90	6.20	98.59	—
A-75–1	16.07	38.65	379.09	14.65	5.80	100.53	~145
A-75–2	—	37.23	378.44	14.09	6.80	98.57	—
A-75–3	—	34.46	365.61	12.60	7.10	99.58	—
Diesel B: 2100 ppm Sulfur	—	—	—	—	—	—	—
B-0–1	4.30	10.81	270.44	2.92	1.20	95.26	—
B-0–2	—	9.64	260.28	2.51	1.20	98.95	~560
B-25–1	8.12	13.00	314.65	4.09	3.60	108.90	—
B-25–2	—	14.34	317.67	4.56	4.00	98.36	~270
B-25–3	—	13.68	316.24	4.33	4.00	106.16	—
B-50–1	11.99	20.70	352.54	7.30	4.50	92.63	—
B-50–2	—	22.02	346.26	7.62	4.60	97.67	~195
B-75–1	15.76	31.96	397.63	12.71	4.40	98.63	—
B-75–2	—	30.58	392.79	12.01	4.10	100.55	~145
B-75–3	—	32.39	382.40	12.59	4.90	98.64	—
Diesel C: 500 ppm Sulfur	—	—	—	—	—	—	—
C-0–1	4.40	3.91	262.63	1.03	3.10	109.80	~560
C-25–1	8.20	9.63	286.05	2.76	3.50	107.32	~270
C-50–1	12.38	12.17	341.40	4.15	6.00	96.49	~195
C-75–1	16.12	17.97	381.99	6.86	7.20	91.48	~145

Note: Test # represents the diesel fuel used, the engine load (e.g., 25–1 denotes the load of 25 kW), and the sample number, respectively.

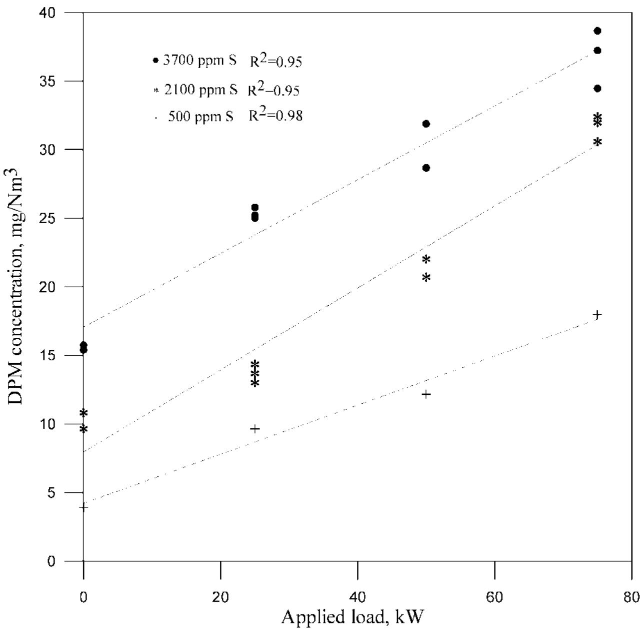

Figure 1. Overall DPM concentration with different diesel fuels.

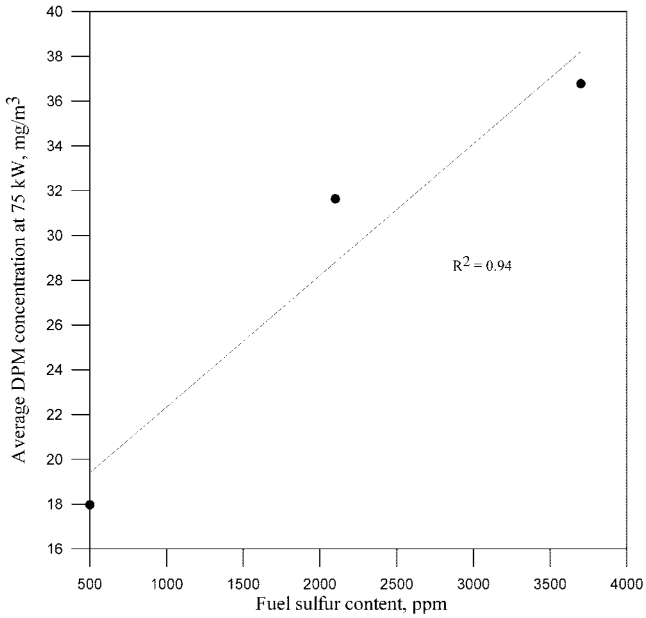

Figure 2. Average DPM concentration at the 75-kW load with different sulfur contents.

model. The average DPM concentrations increase from 15.6 to 36.8 mg/m^3, 10.2 to 31.6 mg/m^3, and 3.9 to 18 mg/m^3 corresponding with the sulfur content in diesel fuel of 3,700, 2,100, and 500 ppm, respectively, when the operating mode changes from 0 to 75 kW. When comparing the DPM emissions from the three diesel fuels at the same load, it can be seen that the increase in DPM concentrations is related to higher sulfur content in the diesel fuels, which will be discussed subsequently. The increase in the engine-operating load, which in turn causes the increase in the generator fuel usage, is also found to have a strong effect on the increase of the DPM concentration. The effects of moisture in DPM emissions can be relatively insignificant in this application, because the filter used for DPM collection was heated to 120 °C under Method 5 requirements, which will be unlikely to result in water condensation.

Influence of the Fuel Sulfur Content on the Average DPM Concentrations at the 75-kW Applied Load

To better understand the effects of the fuel sulfur contents on DPM emissions, the average DPM concentrations taken from the 75-kW load were plotted against the sulfur contents in diesel fuels as shown in Figure 2. It indicates that the DPM concentration increases linearly as a function of the fuel sulfur content with a high correlation coefficient (R^2 = 0.94). Together with Table 1, it can be estimated that the DPM emission rate increases by ~0.022 g/hr-hp for the increase of 0.1% in fuel sulfur. These results agree quite well with the previous study by Baranescu[10] on diesel trucks.

Comparison of the Sulfur Content in DPM Sample with the Fuel Sulfur

The straight-line plot in Figure 3 indicates an acceptable correlation between the sulfur content in the 75-kW DPM samples with the sulfur content in diesel fuels. The sulfur content in DPM sample was analyzed by using the sulfur determinator (expressed in % S by weight). The results show that the ratio of DPM sulfur is somewhat proportional to that of the fuel sulfur, and this may indicate that the increase in DPM concentration at the same load is possibly because of the increase of the fuel sulfur content. If assuming the total sulfur in the 75-kW DPM samples is

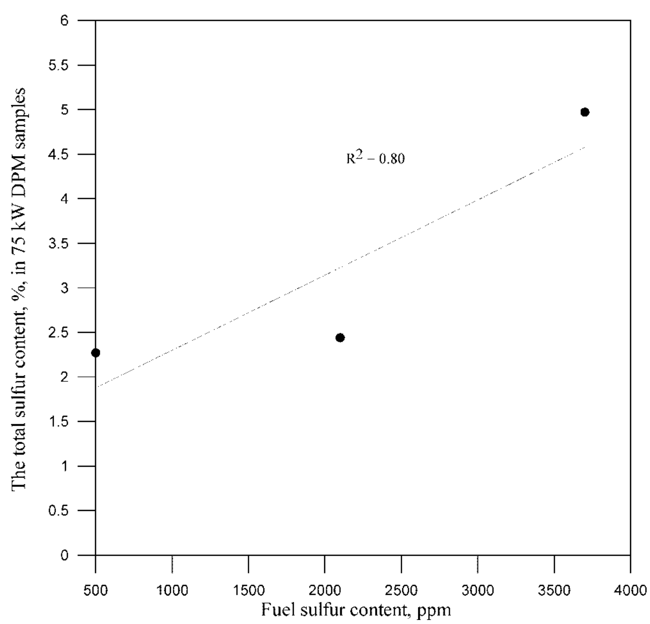

Figure 3. Comparison of DPM sulfur and fuel sulfur.

primarily sulfate, the sulfate constitutes 7–15% of the DPM mass, which is consistent with other published results.[15]

Correlations between Average Normalized DPM Concentrations for Fuel Sulfur and the Applied Loads

Through the use of the least-squared regression method, the empirical correlation of DPM emission to the operating mode, and fuel sulfur content is obtained.

$Y = Z^m(\alpha X + \beta)$, where Y is the DPM concentration, mg/m^3, Z is the fuel sulfur content, ppmw (limited to 500-3700 ppm$_w$), and X is the applied loads, kW, m is the constant, 0.407, α and β are the numerical coefficients, 0.0118 ± 0.0028 (95% confidence interval) and 0.4535 ± 0.1288 (95% confidence interval), respectively. The solid line in Figure 4 represents the normalized DPM concentrations with the above equation. It agrees reasonably well with experimental measurements under various loads, and the high R^2 value of 0.994 indicates a strong correlation. The 95% confidence interval on the average normalized DPM concentrations is also constructed. As a result, it can be 95% confident that the average of the measured DPM concentrations normalized for diesel sulfur lie within this interval. The parity plot of the average DPM concentrations obtained from measurement at each load and the DPM concentration obtained from the empirical equation are also shown in Figure 5.

CONCLUSIONS

This study shows that the DPM emission from the stationary, nonroad diesel generator is a strong function of the operating mode and the fuel sulfur content. The fuel usage rate is found to be independent of the fuel sulfur content. The correlation of the DPM concentration, fuel sulfur (with a power constant 0.407), and applied loads can be well represented by the least-squared regression method. The DPM concentration increases significantly when the applied loads and the sulfur in diesel fuel increase. On the other hand, the sulfur reduction from 3,700 ppm to 500 ppm can result in >50% of DPM reduction. The percentage of sulfur content in the DPM is proportional to that in the fuel sulfur to some extent. The empirical equation obtained could be used for estimating of DPM concentration when the operating modes and fuel sulfur contents are known. However, if the characteristics of the diesel generator and the fuel properties are substantially different from what were used in this work, variations of the predicted DPM concentrations from the measured values may be expected.

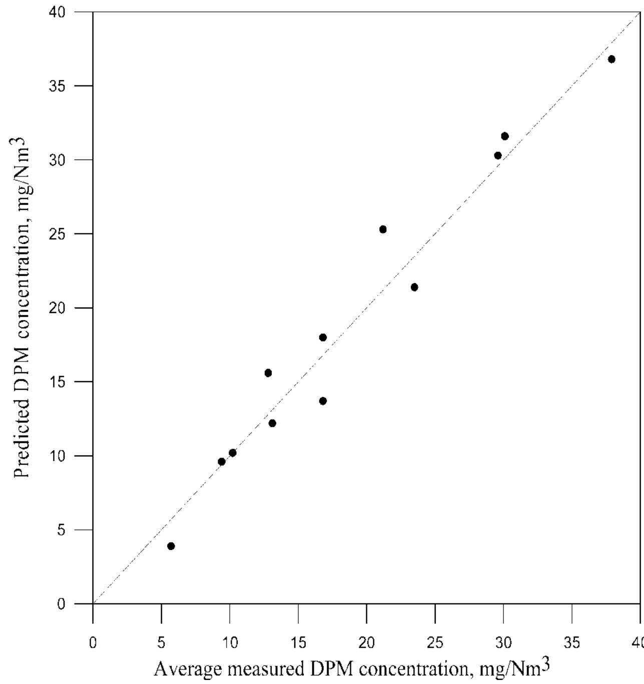

Figure 5. Comparison of measured and predicted DPM concentrations.

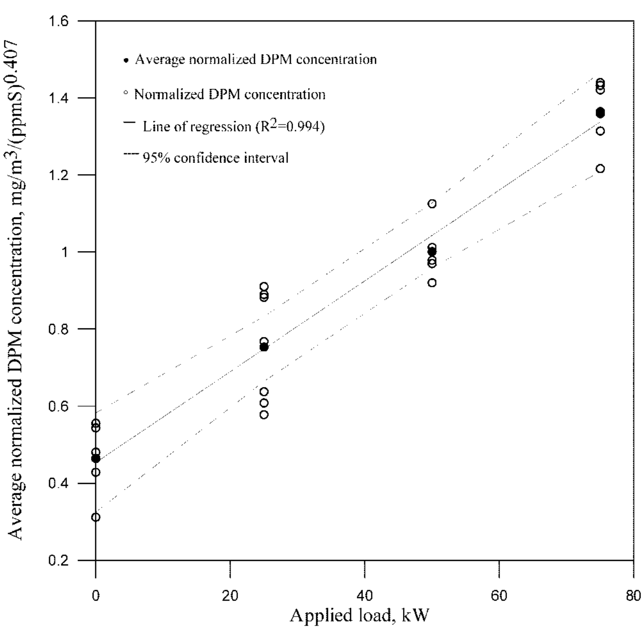

Figure 4. Normalized DPM concentration for fuel sulfur with different loads.

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