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Effect of Fuel Properties on Particulate Matter and Polycyclic Aromatic Hydrocarbon Emission from Diesel Engine in Taiwan

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Abstract: In this study, a Cummins B5 diesel engine was set up to operate on a dynamometer under the US transient cycle. Twenty-four diesel fuels were tested. Regulated air pollutants (HC, CO, NOx, particulate matter) and polycyclic aromatic hydrocarbon (PAH) emissions were measured. PAH was analyzed by gas chromatography/mass spectrometer detector (GC/MS). The average emission factors were 0.32, 2.1, 4.9 and 0.09 g/BHP-hr for HC, CO, NOx and particulate matter, which were all lower than the emission standards in Taiwan. Total PAH (sum of 21 PAHs) emission factor was 3.6 mg/BHP-hr. The non-liner two-variable regression analysis was used to identify the fuel properties which may have influenced the diesel engine particulate and PAH emissions. The greatest influential factor on particulate emission was carbon content, followed by density and viscosity. A 1% increase in carbon content was associated with an increase of 63.1% in particulate emission. The first three important fuel properties affecting PAH emission were density, viscosity and carbon content. The influences of total aromatics and polyaromatic content on particulate and PAH emissions were insignificant.

Keywords: diesel engine; particulate matter; two-variable regression analysis; fuel property.

1. Introduction

It is well known that diesel engines have a superior fuel economy to spark ignition (SI) engines of the same power output, especially for a city-driving cycle. Diesel engines also have much lower carbon monoxide (CO) and unburned hydrocarbon (UHC) emissions than SI engines. However, particulate emissions from diesel engines are between one and two orders of magnitude higher than for a SI engine [1]. Diesel particulates are also considered a potential health hazard, mainly because of the polycyclic aromatic hydrocarbons (PAHs) present in the solvent [2, 3]. Some PAHs are potential mutagens and carcinogens and are probably the major culprits of high cancer rate [4]. It has been proposed that PAHs in diesel emissions arise primarily from diesel fuel PAH surviving the combustion

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process [1,5]. There is also evidence to suggest that PAH in diesel emissions may be synthesized during the combustion process [6]. Further emission control strategies will focus, in part, on improving engine design and on the continuing development of low emission diesel fuels.

The development of cleaner burning fuels is a particularly attractive method of reducing both particulate and PAHs while maintaining thermal efficiency and using current combustion technology. Some possible improvements in emissions by changing fuel properties are discussed by Miyamoto [7] and Bertoli [8]. Sulfur has been identified by several researchers, such as Lin [9], as an element which increases particulate emission. However, the effects of many other fuel components, such as aromatics, on emissions are still not clearly understood, making the development of new fuels a difficult task. The effect of aromatic content and composition is an area of continuing research, of which there remain many unsolved problems contributed by the difficulty of changing fuel properties independently. To change the aromatic content in the fuel, for example, will change the cetane number, density, viscosity and distillation characteristics.

Westerholm [10] suggested that by reducing fuel PAH contents in commercially available diesel fuel, the emissions of PAH to the environment would be reduced. For the reduction of PAH emission from diesel engine effectively, the practical work should be in regulating the aromatic content in the fuels rather than making modification on the treatment device of engine exhaust or the type of diesel engine [11]. Little published data are available regarding the homologue fraction on PAH in the diesel fuels to characterize the effect of PAH emission from the exhaust of diesel engine. In this study, the effect of fuel properties (aromatic structure especially) on particulate and PAH emission was investigated by statistical analysis on data collected in the burning of many diesel fuels.

2. Materials and methods

2.1. Engine and dynamometer system

The diesel engine used in this study was a Cummins B5. with six-cylinders, four strokes, natural aspirated, directed injection, fuel injection sequence 1-5-3-6-2-4, bore and stroke 110 mm x 115 mm, total displacement 6557 mL, maximum horsepower 107 kW/@2800 rpm and maximum torque 570 N·m/@2000 rpm. This engine was mounted and operated on a Schenck GS-350 DC dynamometer with a DC-IV control system. The DC-current dynamometer with a fully automatic control system is capable of supplying maximum power and torque at 350 kW and 2000 N·m, respectively. It is also capable to switch from positive to negative torque values in a very short time. This enables the transient as well as the steady-state tests to be performed. In this study, a commercially available synthetic engine lubricating oil (API SG/CE^{+}; 15W/40) was used. The test was performed under the US transient cycle test. The US transient driving cycle is the legistive cycle used for heavy duty diesel engine certificates in Taiwan. The cycle sequence includes express way, congested urban and uncontested urban driving.

2.2. Fuels

Twenty-four diesel fuels were tested in this study. These fuels were bought from the gas stations selected randomly in Taiwan. The specifications of these diesel fuels are listed at Table 1. The average content of total- aromatics and poly-aromatic were 27.6 (21.5 - 33.3) wt% and 2.53 (1.10 - 4.70) wt%, respectively. The diesel fuel had a density of 0.84 g/mL. The cetane index was 52.9 and the viscosity at 40℃ was 3.09 CST. Among each experimental run for different fuels, the engine system was drained up to prevent fuel used in prior tests from influencing the results

of subsequent tests. Prior to a test on new fuel, the engine was operated at high idling condition for at least one hour to burn off the residual fuel remaining in the system from previous tests.

Table 1. The specification of the fuels used in this study

Fuel parameter	Mean	Range
Total aromatics content (wt%)	27.6	21.5 - 33.3
Poly-aromatic content (wt%)	2.53	1.10 - 4.70
Cetane index	52.9	49.7 - 60.3
Cetane number	52.2	48.3 - 59.8
Viscosity (CST@40℃)	3.09	2.64 - 3.57
90% distillation temperature	328	326 - 331
95% distillation temperature	346	341 - 352
Carbon residue (wt%)	0.06	0.04 - 0.07
Flash point (℃)	77.3	60.0 - 85.0
Nitrogen (wt%)	0.33	0.24 - 0.38
Carbon content (wt%)	86.4	84.9 - 87.0
Hydrogen content (wt%)	13.2	12.9 - 13.4
Lubricate (μm)	449	374 - 521
Sulfur content (wt%)	0.03	0.02 - 0.04
Density (g/ml @15.5℃)	0.84	0.83 - 0.85

2.3. Sampling system for engine exhaust

Particulate, particulate phase and gaseous phase PAH emissions were collected with a full flow CFV-type dilution tunnel. The tunnel is 350 mm in diameter. Dilution air was drawn into the dilution tunnel by a Spencer blower. The quantity of air in the dilution tunnel was controlled by critical flow venturi (CFV). Total engine exhaust gas was fed into the tunnel by a 10 cm diameter and 7.5 m long solid insulated pipe and mixed with the dilution air in a development section 10 diameters long to ensure good mixing and correct emissions species development. The tunnel air throughout was adjusted so that, at the rated power of the engine under test, the temperature of the fully diluted exhaust gas did not exceed 190℃ [12, 13].

Particulate and particulate phase PAHs were collected by glass fiber filters (pore size = 0.8 μm, diameter = 70 mm) at a temperature below 52℃. Two sets of filter holders were employed in this system. Back-up filters were used in each holder downstream the sampling filters to check the breakthrough effects at different engine conditions. A glass cartridge holder was installed after the back-up filter holder. A glass cartridge containing polyurethane foam (PUF) plug and XAD-2 resin was used to collect the gas phase PAHs. Before taking the samples, the glass fiber filters were placed in an oven at 450℃ for 8 hrs to burn off any organic compounds that might be present in the filters. Finally, the cleaned filters were stored in a desiccators for at least 8 hrs for the moisture equilibrium before weighing. After the experiments the filters were brought back to the laboratory and put in a desiccators for 8 hrs to remove moisture, they were weighed again to determine the net mass of particles collected.

The glass cartridge was packed with 2.5 cm of XAD-16 resin sandwiched between a 5-cm upper PUF plug and a 2.5-cm bottom PUF plug. Silicone glue was used to seal and hold these two pieces of PUF to prevent resin from leaking out during sampling and extraction. After 8 hrs of drying, the PUF/resin cartridge was cleaned up by extracting for one day each with distilled water, methanol, dichloromethane and finally n-hexane for a total of 4 days. After clean-up, the glass cartridge was placed in a vacuum oven at 60℃ for 2 hrs to evaporate the residual solvent in the PUF/resin cartridge. After drying, each PUF/resin cartridge was individually wrapped in hexane-washed aluminum foil and was stored and transported in clean screw-capped jars with Teflon cap liners. Each glass fiber

filter was transported to and from the field in a pre-baked glass box which was also wrapped with aluminum foil.

2.4. PAH analysis

PAH samples were Soxhlet extracted with a mixed solvent (n-hexane and dichloromethane, 400 mL/L each) for 24 hours. The extract was then concentrated by purging with ultra-pure nitrogen to 2 ml for the cleanup procedure and then reconcentrated to 0.5 ml with ultrapure nitrogen.

The concentrations of the following PAHs were determined: naphthalene (Nap), acenaphthylene (AcPy), acenaphthene (Acp), fluorene (Flu), phenanthrene (PA), anthracene (Ant), fluoranthene (FL), pyrene (Pyr), cyclopenta[c,d]pyrene (CYC), benz[a]- anthracene (BaA), chrysene (CHR), benzo[b]-fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[e]pyrene (BeP), benzo[a]pyrene (BaP), perylene (PER), indeno[1,2,3,-cd]-pyrene (IND), dibenz[a,h]-anthracene (DBA), benzo[b]chrysene (BbC), benzo[ghi]perylene (BghiP) and coronene (COR).

A GC (Agilent 6890) with a MS detector (Agilent 5973N) and a computer workstation was used for the PAH analysis. This GC/MS was equipped with an Agilent capillary column (Agilent Ultra 2 - 50 m x 0.32 mm x 0.17 μm), an Agilent 7673A automatic sampler, injection volume 1 μL, split-less injection at 310℃, ion source temperature at 310℃, oven, from 50℃ to 100℃ at 20℃/min; 100℃ to 290℃ at 3℃/min; hold at 290℃ for 40 min. The masses of primary and secondary ions of PAHs were determined by using the scan mode for pure PAH standards. Qualification of PAHs was performed by using the selected ion monitoring (SIM) mode.

3. Results and discussions

3.1. Emission of air pollutants

Air pollutant emission from diesel engine, expressed in mass emitted per brake horse power–hour (BHP-hr), was listed in Table 2. The average emission factors were 0.32, 2.1, 4.9 and 0.09 g/BHP-hr for HC, CO, NOx and particulate matter, which were all lower than the emission standards in Taiwan. The results of VE-10 program, conduced by the Coordinating Research Council - Air Pollution Research Advisory Committee (CRC-APRAC) in 1994, reported that emission factors of HC, CO, NOx and particulate were 0.21, 0.51, 0.41 and 0.20 g/BHP-hr, respectively [14].

The emission factors of HC, CO and NOx in this study were 1.5, 4.1 and 12.3 times of than those obtained by CRC-APRAC. The emission standards of particulate will be 0.01 g/BHP-hr in the US in 2007. The emission of particulate must be reduced to fulfill the standard in the future. PAH emission factors were listed in Table 3. Nap was the dominating component in PAH emission. Average total PAHs (sum of 21 PAHs) emission factor was 3.6 mg/BHP-hr (Table 3), which was comparable to that (3.2 mg/BHP-hr) measured by Yang [13].

Table 2. Emission factors of criteria air pollutants (g/BHP-hr)

Air pollutants	Average	Range	Emission standard
HC	0.32	0.29 - 0.34	1.30
CO	2.10	1.90 - 2.40	10.0
NOx	4.90	4.80 - 5.00	5.00
Particulate	0.09	0.08 - 0.10	0.10

3.2. Correlation analysis

Table 4 lists the correlation coefficients between salient properties of the twenty-four fuels. Some important fuel properties are so closely intertwined that it is almost impossible substantively to vary one property independent of the others. For example, when the aromatic fraction of a diesel fuel is increased,

several other fuel properties change at the same time: the chemical structure changes, the cetane number tends to decrease and the density, viscosity and final boiling point all tend to increase. Many fuel properties selected in this study do not correlate highly each other, and it is possible to isolate some individual effects on exhaust emissions.

This study was to discover those fuel properties having significant influence on particulate and PAH emissions. The two-variable regression analysis was carried out in the next sections. Strictly speaking, each property pair should be made up of two properties that are not highly correlated, that is having a correlation coefficient of less than, say, 0.6. This would have ruled out the property pairs: viscosity with total aromatic content, viscosity with flash point, viscosity with density and density with flash point.

3.3. Fuel properties affecting particulate emission

The non-liner two-variable regression analysis was used to identify the fuel properties which may have influenced the diesel engine particulate and PAH emissions. Only a two-variable analysis was used, because of the limited number of test measurements available. The following non-liner regression equation was used [15]:

$$E = aP_1^b P_2^c \tag{1}$$

Where P_1 and P_2 are a selected pair of fuel properties, a, b and c are the regression coefficients to be estimated from the two-variable regression analysis and E is either the particulate or PAH emission.

Differentiating both sides with respect to P_1 in equation (1):

$$\frac{\partial E}{\partial P_1} = abP_1^{b-1}P_2^c \tag{2}$$

and dividing both sides by E/P_1 gives

$$\frac{\partial E/E}{\partial P_1/P_1} = \frac{abP_1^{b-1}P_2^c P_1}{aP_1^b P_2^c} = b \tag{3}$$

i.e.

$$\frac{\partial E}{E}\times 100\% = b\frac{\partial P_1}{P_1}\times 100\% \tag{4}$$

If $\left(\frac{\partial P_1}{P_1}\right)\times 100\% = 1\%$

(i.e. property P_1 changes by 1 percent of its current value),

then $\left(\frac{\partial E}{E}\right)\times 100\% = b\%$

(i.e. particulate or PAH emission changes by b per cent as a result)

Likewise, one can differentiate Equation (1) with respective to P_2 and obtain Equations (2), (3) and (4) for P_2. It is evident that the regression expression (1) has an advantage in that the parameters b and c are dimensionless and represent particulate and PAH sensitivities with respective to property 1 and property 2.

Table 5 shows the results obtained for the total aromatics pairs that were selected for regression analysis. The value of the particulate sensitivity to total aromatics (index b in Table 5) appears to be consistent. Its value is between -0.58 and 0.69 and the mean value is 0.04. In other words, a 1% increase in total aromatics produce only 0.04% increase in the particulate emission. The results suggest that the influence of total aromatics on particulate emission is insignificant.

Table 6 gives an overall summary of the particulate sensitivity obtained from the entire regression analysis of all the selected pairs. The greatest influence on particulate emission was carbon content (sensitivity 63.1), followed by density (sensitivity 17.5) and viscosity (sensitivity 7.16). The results of EPEFE (European Program on Emissions, Fuels and Engine Technologies) research program show that 4% particulate emission

was reduced while sulfur content was reduced from 450 ppm to 30 ppm for heavy duty diesel engine, a rather low insensitivity of about 0.043 [16]. Our study has also demonstrated the influence of sulfur content (0.02-0.04%) on particulate emission was not significant (sensitivity 0.003).

This finding may be caused by the small variability in sulfur content of the 24 test fuels ranges 0.02-0.04 wt%, which compromised the ability of regression to discern the actual influence. Though total aromatics will be regulated (36 vol%, max) starting in the year of 2007 in Taiwan. Because the influences on particulate emission for total aromatics (sensitivity 0.04) and polyaromatic content (sensitivity 0.06) were insignificant, new regulation will contribute little to the improvement in particulate emission abatement.

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Table 3. Emission factors of PAHs (mg/BHP-hr)

PAHs	Average	Range
Nap	2.40	1.73 - 3.06
AcPy	0.165	0.127 - 0.203
Acp	0.087	0.070 - 0.103
Flu	0.202	0.162 - 0.241
PA	0.200	0.167 - 0.232
Ant	0.034	0.028 - 0.039
FL	0.021	0.018 - 0.023
Pyr	0.023	0.022 - 0.023
CYC	0.017	0.007 - 0.027
BaA	0.004	ND - 0.007
CHR	0.002	0.002 - 0.002
BbF	0.130	0.078 - 0.181
BkF	0.024	ND - 0.038
BeP	0.082	0.035 - 0.129
BaP	0.012	0.007 - 0.017
PER	0.083	0.048 - 0.117
IND	0.015	0.010 - 0.020
DBA	0.009	0.002 - 0.015
BbC	0.121	0.014 - 0.228
BghiP	0.011	0.002 - 0.020
COR	0.003	0.001 - 0.004
Total	3.64	2.72 - 4.56

ND: Not detectable

3.4. Fuel properties affecting PAH emission

There have been many studies on the effects of aromatic compounds on PAH emission from diesel engines [17,18,19]. Yet, the overall picture is still in a state of flux. According to Table 7, the results suggest the following: First, density was the greatest influential factor on PAH emission (sensitivity 28.3), followed by viscosity (sensitivity 11.5) and carbon content (sensitivity 2.88).

Table 4. Matrix of correlation coefficients between fuel properties

	Total aromatics content	Polyarmatic content	Cetane index	Viscosity	90% distillation temperature	Carbon residue	Flash point	Carbon content	Sulfur content	Density
Total aromatics content	1.00									
Polyaromatic content	0.37	1.00								
Cetane index	-0.01	-0.17	1.00							
Viscosity	-0.61	0.25	-0.35	1.00						
90% distillation temperature	-0.28	-0.44	0.10	0.06	1.00					
Carbon residue	-0.33	-0.13	-0.09	0.20	0.08	1.00				
Flash point	-0.05	0.32	-0.22	0.65	-0.32	-0.13	1.00			
Carbon content	0.15	0.11	0.004	0.01	0.26	0.23	0.01	1.00		
Sulfur content	-0.53	-0.20	-0.52	0.54	0.08	0.33	0.07	-0.23	1.00	
Density	-0.23	0.46	-0.45	0.85	-0.21	0.12	0.75	0.06	0.42	1.00

Table 5. Results of regression analysis for the influence of total aromatics on particulate emission

Properties	Fuel properties	Coefficients	Estimate	Standard error	t-ratio	R^2
		lna	-0.996	0.90	-1.11	
P_1	Total aromatics	b	-0.46	0.28	-1.63	0.77
P_2	Polyaromatic content	c	0.11	0.07	1.63	
		lna	10.4	1.55	6.69	
P_1	Total aromatics	b	-0.28	0.06	-4.92	0.99
P_2	Cetane index	c	-3.00	0.38	-7.87	
		lna	-5.11	1.56	-3.28	
P_1	Total aromatics	b	0.37	0.30	1.22	0.87
P_2	Viscosity	c	1.26	0.54	2.33	
		lna	82.5	34.0	2.43	
P_1	Total aromatics	b	-0.58	0.23	-2.57	0.88
P_2	90% distillation temperature	c	-14.3	5.78	-2.48	
		lna	-2.48	0.68	-3.66	
P_1	Total aromatics	b	0.38	0.31	1.21	0.86
P_2	Carbon residue	c	0.40	0.18	2.26	
		lna	-14.9	8.81	-1.69	
P_1	Total aromatics	b	0.24	0.38	0.63	0.74
P_2	Flash point	c	2.67	1.78	1.50	
		lna	-493.5	469.8	-1.05	
P_1	Total aromatics	b	-0.02	0.35	-0.06	0.60
P_2	Carbon content	c	110.1	105.2	1.05	
		lna	0.13	2.32	0.06	
P_1	Total aromatics	b	0.69	1.03	0.67	0.55
P_2	Sulfur content	c	1.34	1.46	0.91	
		lna	-0.12	0.06	-1.89	
P_1	Total aromatics	b	0.01	0.01	0.74	0.99
P_2	Density	c	13.5	0.38	35.5	

Second, the sensitivities of PAH emission to total aromatics and polyaromatic content were 0.19% (-1.09 - 2.16) and 0.09 (0.08 - 0.11), respectively. In other word, a 1% increase in total aromatics and polycyclic content were associated with a 0.19% and a 0.09% increase in PAH emission, which were insignificant.

3.5. Fuel properties affecting PAH emission

There have been many studies on the effects of aromatic compounds on PAH emission from diesel engines [17-19]. Yet, the overall picture is still in a state of flux. According to Table 7, the results suggest the following: First, density was the greatest influential factor on PAH emission (sensitivity 28.3), followed by viscosity (sensitivity 11.5) and carbon content (sensitivity 2.88). Second, the sensitivities of PAH emission to total aromatics and polyaromatic content were 0.19% (-1.09 - 2.16) and 0.09 (0.08 - 0.11), respectively. In other word, a 1% increase in total aromatics and polycyclic content were associated with a 0.19% and a 0.09% increase in PAH emission, which were insignificant.

4. Conclusions

Particulate and PAH emissions from a diesel engine using twenty-four fuels were measured. Besides, the influences of fuel properties on particulate and PAH emission were investigated by non-liner regression model. The results showed that the emission factors of HC, CO, NOx and particulate were 0.32, 2.1, 4.9 and 0.09 g/BHP-hr, respectively. The important fuel properties affecting particulate emission include carbon content (sensitivity 63.1), density (sensitivity 17.5) and viscosity (sensitivity 7.16). Density (sensitivity 28.3), viscosity (sensitivity 11.5) and carbon content (sensitivity 11.5) were the first three important properties affecting PAH emission. The influences of total aromatics and polyaromatic content on particulate and PAH emissions were insignificant.

Table 6. Sensitivity of particulate emission to fuel-properties

Fuel properties	Average	Range
Total aromatics content	0.04	-0.58 - 0.69
Polyaromatic content	0.06	-0.11 - 0.24
Cetane index	-2.16	-5.33 - 4.62
Viscosity	7.16	5.54 - 24.6
90% distillation temperature	-1.37	-14.3 - 19.9
Carbon residue	-1.77	-13.8 - 0.39
Flash point	1.60	-0.93 - 2.67
Carbon content	63.1	7.17 - 110
Sulfur content	0.33	-0.22 - 1.34
Density	17.5	13.4 - 41.1

Table 7. Sensitivity of PAH emission to fuel properties

Fuel properties	Average	Range
Total aromatics content	0.19	-1.09 - 2.16
Polyaromatic content	0.09	0.08 - 0.11
Cetane index	-2.93	-14.5 - 4.62
Viscosity	11.5	0.08 - 71.6
90% distillation temperature	-0.82	-15.5 - 12.57
Carbon residue	-2.02	-22.1 - 3.60
Flash point	-0.80	-26.0 - 9.80
Carbon content	2.88	-62.0 - 173
Sulfur content	1.96	1.29 - 24.7
Density	28.3	15.9 - 41.1

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