

1 **Emission Characteristics of Particulate Matter and Particle-bound Metals**  
2 **from a Diesel Engine Generator Fueled with Waste Cooking Oil-based**  
3 **Biodiesel Blended with *n*-Butanol and Acetone**

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## Abstract

This study examines the emission properties of particulate matter and particle-bound metals from a diesel engine generator fueled with traditional fossil diesel (D100) with the addition of *n*-butanol (B), hydrous *n*-butanol (B'), acetone (A), hydrous acetone (A'), isopropyl alcohol (I) or waste cooking oil-based biodiesel (W). The fuel blends were B30W20D50 (abbrev. B30), B'30W20D50 (as B'30), A3I1W20D76 (as A3), A'3I1W20D76 (as A'3), B30A3I1W20D46 (as B30A3) and B'30A'3I1W20D46 (as B'30A'3) tested at 1.5 kW and 3.0 kW loads of the diesel engine generator. Experimental results indicate that adding B30, A3 or B30A3 reduces the PM mass concentration in the exhaust at both engine loads, in comparison with using only W20. Additionally, the PM emission concentrations are lower using B'30, A'3 and B'30A'3 than using B30, A3 and B30A3, respectively; in other words, replacing pure *n*-butanol/acetone with hydrated *n*-butanol/acetone in the blends further reduces the PM emission concentrations. However, B30 or B30A3 is more effective than A3 in reducing the PM emissions, irrespective of the water content in fuel blends. Conversely, using B30, B'30, A3, A'3, B30A3 or B'30A'3 reduces the metal contents in PM emissions at both engine loads compared with using W20. The major metal components in PM are Na, Mg, Al, K, Ca, Fe and Zn, accounting for about 97 wt.% of overall 21 metals. The remaining analyzed metals were dominated by Mn, Ni, Cu, Mo and Ba. Accordingly, adding biodiesel from waste cooking oil, and hydrous acetone/*n*-butanol, to diesel fuel for diesel engine generators reduces levels of PM and particle-bound metals. The waste hydrous acetone/*n*-butanol can be used for the recycling purpose in this process.

**Keywords:** Diesel engine generator; Waste cooking oil-based biodiesel; Acetone; *n*-Butanol; Particle-bound metal

## 39 1. Introduction

40 Diesel engine is a major power source of transportation and industrial activities, including  
41 on-/off-road vehicles, ships, construction and power utilities. However, researchers have reported  
42 significant negative effects of diesel engine emission on human health. Several studies in this  
43 decade have focused on the particulate matter (PM) emissions from diesel engines and  
44 alternative greener fuels, such as biodiesel. Moreover, the International Agency for Research on  
45 Cancer (IARC) classified diesel engine exhaust (DEE) as carcinogenic to humans (Group 1),  
46 based on sufficient evidence, in June 2012. Additionally, researchers consider diesel particulate  
47 matter (DPM) as a hazardous pollutant to human health and the environment. Conversely,  
48 biodiesel is reported as a diesel alternative to improve the combustion efficiency of diesel  
49 engines, and reduce emissions of carbon monoxide (CO), unburned hydrocarbon (HC),  
50 particulate matter (PM) and polycyclic aromatic hydrocarbons (PAHs) (Lin *et al.*, 2010; Tsai *et*  
51 *al.*, 2010; Xue *et al.*, 2011; Guido *et al.*, 2013; Wu *et al.*, 2016; Redfern *et al.*, 2017), as well as  
52 the reducing DPM biotoxicity (Tsai *et al.*, 2011; Tsai *et al.*, 2012).

53 Dwivedi *et al.* (2006) found that a mixture of 20% biodiesel with 80% diesel had higher  
54 emissions of Cr, Fe, Al, Zn and Mg, and lower Pb, Cd, Na and Ni, than 100% diesel in a diesel  
55 engine. Certain metals might be originally present in the food ingredients, such as Cu, Fe, Zn and  
56 Mn in vegetables (Kawashima and Valente Soares, 2003); Cu, Fe, Zn and Mn in meat

57 (Lombardi-Boccia *et al.*, 2005), and Cu, Zn and Cd in fish (Atta *et al.*, 1997). Additionally, some  
58 metals are released into cooking oil during high-temperature frying, or from cooking utensils  
59 (Kuligowski & Halperin, 1992). Metals can also be transported into waste cooking oil  
60 (WCO)-based biodiesel following transesterification. Consequently, combustion of WCO-based  
61 biodiesel potentially produces Fe, Zn, Cu and Mn emissions. Use of biodiesel in diesel engines  
62 has led to Na and K emissions, since these metals are components of major alkaline catalysts  
63 (KOH or NaOH) in transesterification of fatty acids. The metal content of PM in diesel engine  
64 exhaust may also be affected by factors including engine operation parameter, testing cycle, fuel  
65 property, fuel chemical composition and quality and engine wearing level (Wang *et al.*, 2016).

66 The fine particles emitted from diesel engines are easily inhaled through the respiratory  
67 system, and thus can penetrate deeply into the alveolus, be transported to epithelial mesenchyme  
68 and lymph nodes, and eventually accumulate in organs in the human body via the circulation  
69 system (Kreyling *et al.*, 2012). Some metal components, such as Cr, Cd, Pb, Ni, Mn, As, Zn and  
70 Cu, are poorly biodegradable, and are toxic to humans because they disrupt bio-metabolism and  
71 change blood composition, although they have fairly low contributions in PM<sub>2.5</sub> mass.  
72 Furthermore, these metals become increasingly concentrated in animal bodies up the food chain  
73 through bioaccumulation or biomagnification, and are thus especially toxic in the predators at the  
74 top of a food web (Adham *et al.*, 2011; Fang *et al.*, 2013; Abuduwaili *et al.*, 2015).

75 Adding 20% WCO-based biodiesel additive, a certain amount of acetone/butanol, or even a  
76 small amount of water, to diesel fuel reduces PM emission from diesel engine generators (DEG)  
77 (Tsai *et al.*, 2017). However, little information is available on whether using such  
78 multi-component fuels in DEG also reduces emissions of particle-bound metals. Hence, this  
79 work adopts traditional fossil diesel (D) as the base fuel, blended with *n*-butanol (B), 5 vol.%  
80 hydrous *n*-butanol (B'), acetone (A), 5 vol.% hydrous acetone (A'), isopropyl alcohol (I) or waste  
81 cooking oil-based biodiesel (W) to investigate the characteristics of PMs and their bound metals  
82 emitted from a DEG operated at 1.5 kW and 3.0 kW loads.

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## 84 **2. Materials and Methods**

### 85 **2.1 Engine System and Sampling**

86 Figure 1 shows the tested engine generator and stack sampling system. The diesel engine  
87 generator applied in this study was a four-stroke, single horizontal cylinder engine produced by  
88 Yanmar S. P. Co. The fuels were injected with 2845psi at BTDC 17.0° CA by a direct injection  
89 system, which was the same for each fuel. The generator loads were controlled with a variable  
90 resistance system comprising 100 parallel bulbs, which was equivalent to the maximum 5 kW  
91 load. The DEG was operated at two engine loads, 1.5 and 3.0 kW, both at 110 V with 60 Hz  
92 power output, to test its performances and exhaust during operation.

## 93 2.2 Testing Fuel Blends

94 The base fuel in this work was premium diesel fuel produced by Chinese Petroleum  
95 Corporation (CPC), Taiwan. Waste cooking oil-based biodiesel was provided by Chant Oil Co.,  
96 LTD. Analytical-grade acetone, *n*-butanol and isopropyl alcohol all had >99.5% purity, and were  
97 supplied by Sigma-Aldrich, Inc. The premium diesel fuel (D) was adopted as the base fuel and  
98 the control group in the current study. WCO-based biodiesel was tested as an additive (20 vol.%  
99 fraction) in diesel blend to form W20. *n*-Butanol was added to W20 to prepare B30W20D50  
100 (abbr. B30) (30 vol.%), while the 5 vol.% hydrous *n*-butanol (B') was used to prepare  
101 B'30W20D50 (abbr. B'30). Acetone was blended with W20 at a concentration of 3 vol.%, to  
102 which 1 vol.% isopropyl alcohol was added to stabilize the blend and form A3I1W20D76 (abbr.  
103 A3). Meanwhile, 5 vol.% hydrous acetone (A') was also tested as a stable diesel blend,  
104 A'3I1W20D76 (abbr. A'3). Finally, 30 vol.% of pure/hydrous *n*-butanol and 3 vol.% of pure or  
105 hydrous acetone were added to W20 with 1% isopropyl alcohol to form B30A3I1W20D46 (abbr.  
106 B30A3) or B'30A'3I1W20D46 (abbr. B'30A'3), respectively. The test data for W20 are in our  
107 previous study (Lin *et al.*, 2017).

## 108 2.3 PM Sample Analyses

### 109 *PM Mass Concentrations*

110 The PM and particle-bound metals were collected on a 47 mm-diameter quartz fiber filter

111 using a stack sampling system (Anderson Auto 5). The PM samples accumulated on the quartz  
112 fiber filters were stored and conditioned at  $23^{\circ}\text{C}\pm 1^{\circ}\text{C}$  at relative humidity  $40\pm 5\%$  in a clean  
113 room over 24h. Each filter was weighed twice with an electronic microbalance (METTLER  
114 TOLEDO Model XP2U) before and after sampling to minimize the inner-group error. Each PM  
115 mass concentration was calculated by subtracting the filter mass before sampling from the final  
116 filter mass (with PM sample), and further divided by the sampling volume (modified by the  
117 standard temperature and atmospheric pressure condition,  $\text{Nm}^3$ ).

#### 118 *Metal Elements Analysis*

119 The particle-bound metal contents were further measured by the analytical process described  
120 in standard method M105 from the Environmental Analysis Laboratory (EAL), Environmental  
121 Protection Administration (EPA), Taiwan. The weighed quartz fiber filters with PM sample were  
122 placed into 50mL graphite digestion tubes, soaked with 20mL nitric acid ( $\text{HNO}_3$ , 10% purity),  
123 and locked with cover. The graphite tubes were treated with 450W ultrasonic waves in a basin  
124 for 120 min, and further heated at  $80\text{--}85^{\circ}\text{C}$  for 30 min. After the heating process, the sample acid  
125 solutions were poured out and sieved using a  $0.45\mu\text{m}$  sieving plate. The metal contents in the  
126 pre-treated sample solution were quantified with an inductively coupled plasma mass  
127 spectrometer (ICP-MS, Jobin Yvon ULTIMA 2000). The calibration lines (with absolute error  
128  $<10\%$ ) were evaluated by using a standard solution. The analytical method had recovery levels in

129 the range 75–125%, which were checked every 10 samples by an extra standard sample solution.  
130 Additionally, the methodological blank sample was determined and found to be less than 2 times  
131 the method detection limit (MDL). The MDLs of the 21 target metals, namely Na, Mg, Al, K, Ca,  
132 Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Sr, Mo, Cd, Sn, Sb, Ba and Pb, were 6.12, 5.22, 3.16, 19.3,  
133 24.3, 0.36, 0.04, 0.14, 0.03, 2.22, 0.03, 0.31, 5.27, 0.31, 0.20, 0.99, 0.02, 3.15, 0.12, 0.51 and  
134 0.06  $\mu\text{g L}^{-1}$ , respectively.

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### 136 **3. Results and Discussion**

#### 137 **3.1 PM Mass Concentrations in the DEG Exhaust**

##### 138 *Effect of Engine Operation Load*

139 Table 1 shows the PM mass emissions from the tested DEG by using B30, B'30, A3, A'3,  
140 B30A3 and B'30A'3 at engine loads of 1.5 kW and 3.0 kW. The PM emission concentrations of  
141 using these blends at 1.5 kW were 33.9 $\pm$ 4.0, 28.8 $\pm$ 1.8, 40.6 $\pm$ 3.0, 35.2 $\pm$ 2.4, 30.9 $\pm$ 5.2 and  
142 30.2 $\pm$ 4.3  $\text{mg Nm}^{-3}$ , respectively, while those at 3.0 kW were 58.7 $\pm$ 3.3, 53.9 $\pm$ 1.7, 76.3 $\pm$ 7.0,  
143 70.2 $\pm$ 2.6, 50.8 $\pm$ 3.9 and 49.5 $\pm$ 2.8  $\text{mg Nm}^{-3}$ , respectively. For the diesel engine generator fueled  
144 with those blends, the PM emission factors based on fuel consumptions were in the range 11.1–  
145 15.9  $\text{mg L}^{-1}$  at 1.5 kW and 13.6–22.9  $\text{mg L}^{-1}$  at 3.0 kW. The above result indicates that the DEG  
146 fueled with these blends (B30, B'30, A3, A'3, B30A3, and B'30A'3) emitted higher PM mass



147 concentrations and emission factors at 3.0 kW than at 1.5 kW. This phenomenon likely resulted  
148 from the higher fuel consumption (FC) for the same fuel blend at 3.0 kW than at 1.5 kW, which  
149 increased the PM mass produced in the sample volume of exhaust gas (Tsai *et al.*, 2017). This  
150 speculation is observed by Lin *et al.* (2017), who studied the use of D100, W20 and W40 in a  
151 diesel engine.

### 152 ***Effect of Various Fuel Blends***

153 Figure 2 depicts the reduction rates (%) by using B30, B'30, A3, A'3, B30A3 and B'30A'3,  
154 in comparison with using W20. These diesel blends reduced the PM emission by  $35.1 \pm 7.7\%$ ,  
155  $44.8 \pm 3.5\%$ ,  $22.2 \pm 5.7\%$ ,  $32.7 \pm 4.5\%$ ,  $40.9 \pm 9.9\%$ , and  $42.1 \pm 8.3\%$ , respectively, compared with  
156 W20 at 1.5 kW, and  $38.0 \pm 3.5\%$ ,  $43.0 \pm 1.8\%$ ,  $19.4 \pm 7.3\%$ ,  $25.8 \pm 2.7\%$ ,  $46.4 \pm 4.1\%$  and  $47.7 \pm 2.9\%$ ,  
157 respectively, at 3.0 kW. Thus, B30, A3 and B30A3 had lower PM emissions than WCO-based  
158 biodiesel additives (*i.e.* W20). The reduction rate was even better with (5 vol.%) in *n*-butanol and  
159 acetone in fuel blends (*i.e.* B'30, A'3 and B'30A'3). Blending 30 vol.% anhydrous/hydrous  
160 *n*-butanol with diesel fuels led to greater reductions in PM mass (35.1–44.8%; average: 40.2%)  
161 emission than blending 3 vol.% anhydrous/hydrous acetone (19.4–32.7%; average: 25.0%). On  
162 average, the diesel blends comprising W20 and both *n*-butanol and acetone (*i.e.* B30A3 and  
163 B'30A'3) had the greatest PM reductions (40.9–47.7%; average: 44.3%) among all tested fuel  
164 blends.

165 The pure WCO-biodiesel had ~11 wt.% of oxygen, while acetone ( $C_6H_6O$ ) and *n*-butanol  
166 ( $C_4H_9OH$ ) had higher oxygen contents (27.5 wt.% and 21.6 wt.%, respectively). The oxygen  
167 contents of tested fuel blends in this work were listed in Table 2. The significant PM emission  
168 reductions at both DEG loads from adding anhydrous *n*-butanol/acetone into W20 (*i.e.* B30, A3  
169 and B30A3) could have resulted from the rise in fuel-oxygen content of fuel blend and the  
170 supply of additional oxygen radicals to initiate combustion, leading to more complete reaction  
171 and thus enhancing the combustion efficiency (Yang *et al.*, 2016; Saxena and Maurya, 2016).

172 Moreover, water present in the hydrated *n*-butanol and acetone mixtures B'30, A'3 and  
173 B'30A'3 further lowered the PM emissions. This finding could be explained by both physical and  
174 chemical mechanisms. Physically, water has a much lower boiling point than either diesel or  
175 biodiesel. Therefore, the water tended to evaporate as steam at the very beginning of fuel droplet  
176 combustion. The sudden increase in volume of water (liquid to gas) destroyed the outer oil layer,  
177 and split each fuel droplet into much smaller ones, thus significantly increasing the specific  
178 intake air reaction area while initiating combustion. Therefore, this secondary atomization, called  
179 a micro-explosion (Ivanov and Nefedov, 1965), improved the combustion and decreased the PM  
180 emission (Tsai *et al.*, 2014a; Tsai *et al.*, 2014b). A fraction of water compound in steam are  
181 pyrolyzed to form  $OH\cdot$ ,  $O\cdot$ , and  $H\cdot$  radicals, which improve the oxidation of the fuel compound  
182 rapidly and completely, while the unburned hydrocarbon is also reduced, thus inhibiting PM

183 formation. Consequently, the anhydrous/hydrous oxygenated additives in this study lowered the  
184 PM emissions.

### 185 **3.3 Emissions of Particle-bound Metals**

186 The average emission concentrations of  $\Sigma$ Metal (sum of 21 particle-bound metals, namely  
187 Na, Mg, Al, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Sr, Mo, Cd, Sn, Sb, Ba and Pb) from using  
188 B30, B'30, A3, A'3, B30A3 and B'30A'3 were  $453\pm 264$ ,  $422\pm 75$ ,  $443\pm 108$ ,  $431\pm 68$ ,  $341\pm 103$   
189 and  $337\pm 41$   $\mu\text{g}/\text{Nm}^3$ , respectively, at 1.5 kW load (Table 3). Additionally, the high  $\Sigma$ Metal levels  
190 in exhaust were found to be  $1218\pm 116$ ,  $1047\pm 67$ ,  $1173\pm 139$ ,  $1004\pm 234$ ,  $869\pm 99$ , and  $853\pm 88$   
191  $\mu\text{g}/\text{Nm}^3$  by using B30, B'30, A3, A'3, B30A3 and B'30A'3, respectively, at 3.0 kW (Table 4).  
192 The above results show that B30, B'30, A3, A'3, B30A3, and B'30A'3 all resulted in lower  
193  $\Sigma$ Metal levels than W20.

194 The metal contents of the pure WCO-based biodiesel (W100), premium diesel fuel (D100),  
195 and lubricating oil (L100) were also determined (as presented in Fig. 3). To avoid the explosion  
196 caused by the high temperature condition in ICP/MS operation, W100, D100, and L100 were  
197 heated to  $150^\circ\text{C}$  to remove the volatile organic compounds, and were ultrasonically digested  
198 prior to the analysis of the 21 metals. The most abundant 7 metals were in the order  $\text{K} > \text{Na} > \text{Ca}$   
199  $> \text{Mg} > \text{Al} > \text{Fe} > \text{Zn}$  in W100, and  $\text{K} > \text{Na} > \text{Ca} > \text{Mg} > \text{Al} > \text{Sn} > \text{Fe}$  in D100. Obviously, the  
200 sums of the above 7-metal contents in fuels were similar (98.4% of W100 and 98.5% of D100).

201 However, the major metals in lubricating oil were Ca (69.3%) and Zn (30.2%).

202 The measurement results in Tables 3 and 4 indicate that the top 7 predominant  
203 particle-bound metals emitted when using W20 followed the order Fe > Ca > Na > Zn > K > Mg  
204 > Al, and Ca > Zn > Na > Fe > K > Mg > Cr (while Al is the 8<sup>th</sup> one) at 1.5 kW and 3.0 kW,  
205 respectively. The above dominant particle-bound metal compositions were very similar to those  
206 (K, Na, Ca, Mg, Fe, and Al) in the unburned W20 (comprising 20% of W100 and 80% of D100).  
207 Conversely, the top 7 dominant metals in the emitted PM when using B30, B'30, A3, A'3,  
208 B30A3, and B'30A'3 varied with the order Ca > Fe > Zn > Na > K > Mg > Al at 1.5 kW, and Ca  
209 > Zn > Fe > Na > K > Mg > Al at 3.0 kW loads. All emission had the metals Na, Mg, Al, K, Ca,  
210 Fe, and Zn.

211 The emissions of diesel engine were dominated by unburned black carbon (soot) and small  
212 amount of metals (ash) (Liati *et al.*, 2015). For diesel emission, metal might be harmful to human  
213 health, although they contribute relatively lower mass fractions. Those particle-bound metals  
214 could be linked with the lubricant, fuel, additive and detergent contents. The emission levels of  
215 metals generally rise with increasing content in fuels (Wang *et al.*, 2016). The particle-bound  
216 metallic pollutants in diesel engine emissions can be formed and released by several pathways:  
217 (1) the original metal contents are released by heating, and are adsorbed on the PM surface at a  
218 certain temperature and pressure; (2) the nuclei of metal components simultaneously move with

219 PM in the engine exhaust, and coagulate with each other to form metal-containing PM; (3)  
220 metals react with oxygen, halogens or organic components to form metallic compounds, which  
221 either become residue in the combustion ashes or evaporate as metallic fume, and (4) the metallic  
222 fume is further condensed on the colder surface of the PM or fly ashes during treatment of diesel  
223 engine emissions (Eddings *et al.*, 1994).

224 Shah *et al.* (2014) reported that Fe and Ca were the major particle-bound metallic  
225 components in fossil fuel combustion, while Na could be emitted from biodiesel combustion.  
226 Additionally, the blend of ultra-low-sulfur diesel and biodiesel caused the particle-bound metals  
227 to be dominated by As, Co, Al and Mn in diesel engine emissions, while they shifted to Cr, Cu,  
228 Fe, Ba, Zn, Mg, Ni and K when using pure biodiesel (B100). The increased Cu, Fe or Zn content  
229 in biodiesel fuel may be from the feedstock used in biodiesel preparation (Betha and  
230 Balasubramanian, 2011). The high lubricity of biodiesel decreases the friction of piston rings,  
231 cylinder liners, intake/exhaust valves and crank shafts during diesel engine operation, thus  
232 lowering Fe emissions. Additionally, the improvement lubricity also reduces Cr emission, and  
233 impedes the wear of gears, compression rings, and cam bearings (Agarwal, 2007). Nevertheless,  
234 the use of biodiesel was also reported to reduce Cr, Zn and Al emissions from fossil diesel fuel  
235 by improving the lubricity (Wang *et al.*, 2016).

236 The  $\Sigma$ metals content in PM by using B30, B'30, A3, A'3, B30A3 and B'30A'3 (1.11–1.48%)

237 were greater than by using W20 (1.02%) at 1.5 kW load (Table 3). However, the  $\Sigma$ metals content  
238 by using these blends (1.42–2.07%) were roughly lower than by using W20 (1.93%) at 3.0 kW  
239 load (Table 4). The above phenomenon may be related to the fact that the addition of  
240 pure/hydrous butanol and acetone will slightly restrain the lubrication of the engine oil. Shukla *et*  
241 *al.* (2017) indicated that biodiesel had worse nebulization of fuel spray than conventional diesel,  
242 due to its higher density, viscosity and surface tension. Bigger biodiesel droplets may even  
243 penetrate into the cylinder wall, becoming hydrocarbon residues. Moreover, the mechanical  
244 friction and wear between the piston ring and cylinder liner may lead to metallic emissions in  
245 diesel engine exhaust.

246 This investigation found that the emitted particle-bound metals by using B30, B'30, A3, A'3,  
247 B30A3, or B'30A'3 as the fuel of DEG were predominated (97% in mass) by Na, Mg, Al, K, Ca,  
248 Fe and Zn among the 21 analyzed metals, while Mn, Ni, Cu, Mo, and Ba were the main  
249 components among the remaining metals. The pure/hydrous *n*-butanol and acetone additives in  
250 W20 could further reduce the metal emissions during DEG operation.

251

#### 252 **4. Conclusions**

253 The main objective of this study is to investigate the effects of particle-bound metal  
254 emissions by using W20, pure/hydrous *n*-butanol or acetone additives as alternatives to

255 conventional diesel in a DEG. The findings of this investigation are summarized as follows.

256 1. Using B30, A3 or B30A3 reduced PM emissions in comparison to using W20 at two DEG  
257 loads. Additionally, B'30, A'3, and B'30A'3 (*n*-butanol and/or acetone with 5 vol.% water)  
258 had higher PM emission reduction than pure B/A diesel blends. The blended fuels (B30/B'30  
259 and B30A3/B'30A'3) with higher B/A fractions exhibited lower PM emission levels than  
260 those (A3/A'3) with lower ones, regardless of the different water contents in the blended  
261 fuels.

262 2. The application of B30, B'30, A3, A'3, B30A3 and B'30A'3 decreased the particle-bound  
263 metal emissions when compared with adopting only W20 at both DEG loads.

264 3. The dominant metals in PM emissions among the 21 analyzed metals were Na, Mg, Al, K,  
265 Ca, Fe, and Zn . The sum of their mass contributed about 97 wt.% of total metal mass, while  
266 Mn, Ni, Cu, Mo and Ba were the main components of the remaining metals.

267 Consequently, the addition of waste cooking oil-based biodiesel and pure/hydrous  
268 acetone/*n*-butanol to traditional diesel is a potential green diesel alternative for DEGs to lower  
269 PM and particle-bound metal emissions. The waste hydrous acetone/*n*-butanol can be adopted  
270 for recycling in this operation process.

271

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276

## 277 REFERENCE

278 Abuduwaili, J., Zhang, Z.Y. and Jiang, F.Q. (2015). Assessment of the distribution, sources and  
279 potential ecological risk of heavy metals in the dry surface sediment of Aibi Lake in  
280 northwest China. *PLoS One* 10: e0120001.

281 Adham, K.G., Al-Eisa, N.A. and Farhood, M.H. (2011). Impact of heavy metal pollution on the  
282 hemogram and serum biochemistry of the Libyan jird, *Meriones libycus*. *Chemosphere* 84:  
283 1408-1415.

284 Agarwal, A.K. (2007). Biofuels (alcohols and biodiesel) applications as fuels for internal  
285 combustion engines. *Prog. Energy Combust. Sci.* 33: 233-271.

286 Atta, M.B., El-Sebaie, L.A., Noaman, M.A. and Kassab, H.E. (1997). The effect of cooking on  
287 the content of heavy metals in fish (*Tilapia nilotica*). *Food Chem.* 58: 1-4.

288 Betha, R. and Balasubramanian, R. (2011). Particulate emissions from a stationary engine fueled  
289 with ultra-low-sulfur diesel and waste-cooking-oil-derived biodiesel. *J. Air Waste Manage.*

290 *Assoc.* 61: 1063-1069.



- 291 Dwivedi, D., Agarwal, A.K. and Sharma, M. (2006). Particulate emission characterization of a  
292 biodiesel vs diesel-fuelled compression ignition transport engine: A comparative study.  
293 *Atmos. Environ.* 40: 5586-5595.
- 294 Eddings, E.G., Lighty, J.S. and Kozinski, J.A. (1994). Determination of metal behavior during  
295 the incineration of a contaminated montmorillonite clay. *Environ. Sci. Technol.* 28:  
296 1791-1800.
- 297 Fang, W., Yang, Y. and Xu, Z. (2013). PM<sub>10</sub> and PM<sub>2.5</sub> and health risk assessment for heavy  
298 metals in a typical factory for cathode ray tube television recycling. *Environ. Sci. Technol.*  
299 47: 12469-12476.
- 300 Guido, C., Beatrice, C. and Napolitano, P. (2013). Application of bioethanol/RME/diesel blend  
301 in a Euro5 automotive diesel engine: Potentiality of closed loop combustion control  
302 technology. *Appl. Energy* 102: 13-23.
- 303 Ivanov, V.M. and Nefedov, P.I. (1965). *Experimental investigation of the combustion process of*  
304 *natural and emulsified liquid fuels*, National Aeronautics and Space Administration,  
305 Washington, D.C., NASA technical translation F-258.
- 306 Kawashima, L.M. and Valente Soares, L.M. (2003). Mineral profile of raw and cooked leafy  
307 vegetables consumed in Southern Brazil. *J. Food Compos. Anal.* 16: 605-611.
- 308 Kreyling, W.G., Semmler-Behnke, M., Takenaka, S. and Möller, W. (2012). Differences in the

309 biokinetics of inhaled nano- versus micrometer-sized particles. *Accounts Chem. Res.* 46:  
310 714-722.

311 Kuligowski, J. and Halperin, K.M. (1992). Stainless steel cookware as a significant source of  
312 nickel, chromium, and iron. *Arch. Environ. Contam. Toxicol.* 23: 211-215.

313 Liati, A., Pandurangi, S.S., Boulouchos, K., Schreiber, D. and Dasilva, Y.A.R. (2015). Metal  
314 nanoparticles in diesel exhaust derived by in-cylinder melting of detached engine fragments.  
315 *Atmos. Environ.* 101: 34-40.

316 Lin, S.L., Lee, W.J., Lee, C.F. and Chen, S.J. (2010). Energy savings and emission reduction of  
317 nitrogen oxides, particulate matter, and polycyclic aromatic hydrocarbons by adding  
318 water-containing acetone and neat soybean oil to a diesel-fueled engine generator. *Energy*  
319 *Fuels* 24: 4522-4533.

320 Lin, S.L., Tsai, J.H., Chen, S.J., Huang, K.L., Lin, C.C., Huang, H.T., Hsieh, Y.C. and Chiu, C.H.  
321 (2017). Emissions of polycyclic aromatic hydrocarbons and particle-bound metals from a  
322 diesel engine generator fueled with waste cooking oil-based biodiesel blends. *Aerosol Air*  
323 *Qual. Res.* 17: 1579-1589.

324 Lombardi-Boccia, G., Lanzi, S. and Aguzzi, A. (2005). Aspects of meat quality: trace elements  
325 and B vitamins in raw and cooked meats. *J. Food Compos. Anal.* 18: 39-46.

326 Redfern, F.M., Lin, S.L., Wang, L.C. and Shih, S.I. (2017). Influences of waste cooking

327 oil-based biodiesel blends on PAH and PCDD/F emissions from diesel engines in durability  
328 testing cycle. *Aerosol Air Qual Res* 17: 1224-1233.

329 Saxena, M.R. and Maurya, R.K. (2016). Effect of butanol blends on nano particle emissions  
330 from a stationary conventional diesel engine. *Aerosol Air Qual Res* 16: 2255-2266.

331 Shah, A., Yun-shan, G., Shah, F., Mughal, H. and Naveed, A. (2014). Effect of biodiesel on  
332 particulate numbers and composition emitted from turbocharged diesel engine. *Int. J.*  
333 *Environ. Sci. Technol.* 11: 385-394.

334 Shukla, P.C., Gupta, T., Labhsetwar, N.K. and Agarwal, A.K. (2017). Trace metals and ions in  
335 particulates emitted by biodiesel fuelled engine. *Fuel* 188: 603-609.

336 Tsai, J.H., Chen, S.J., Huang, K.L., Lin, T.C., Chaung, H.C., Chiu, C.H., Chiu, J.Y., Lin, C.C.  
337 and Tsai, P.Y. (2012). PM, carbon, PAH, and particle-extract-induced cytotoxicity  
338 emissions from a diesel generator fueled with waste edible-oil-biodiesel. *Aerosol Air Qual.*  
339 *Res.* Vol. 12, pp. 843-855.

340 Tsai, J.H., Chen, S.J., Huang, K.L., Lin, W.Y., Lee, W.J., Chao, H.R., Lin, C.C. and Hsieh, L.T.  
341 (2014a). Emission reduction of NO<sub>x</sub>, PM, PM-carbon, and PAHs from a generator fuelled  
342 by biodiesels. *J. Hazard. Mater.* 274: 349-359.

343 Tsai, J.H., Chen, S.J., Huang, K.L., Lin, W.Y., Lee, W.J., Lin, C.C., Hsieh, L.T., Chiu, J.Y. and  
344 Kuo, W.C. (2014b). Emissions from a generator fueled by blends of diesel, biodiesel,

345 acetone, and isopropyl alcohol: Analyses of emitted PM, particulate carbon, and PAHs. *Sci.*  
346 *Total Environ.* 466: 195-202.

347 Tsai, J.H., Chen, S.J., Huang, K.L., Lin, Y.C., Lee, W.J., Lin, C.C. and Lin, W.Y. (2010). PM,  
348 carbon, and PAH emissions from a diesel generator fuelled with soy-biodiesel blends. *J.*  
349 *Hazard. Mater.* 179: 237-243.

350 Tsai, J.H., Huang, K.L., Chiu, C.H., Lin, C.C., Kuo, W.C., Lin, W.Y., Chaung, H.C., Yang, T.H.  
351 and Chen, S.J. (2011). Particle-bound PAHs and particle-extract-induced cytotoxicity of  
352 emission from a diesel-generator fuelled with soy-biodiesel. *Aerosol Air Qual. Res.* 11:  
353 822-836.

354 Tsai, J.H., Lin, S.L., Chen, S.J., Chang-Chien, G.P., Jheng, B.C., Huang, K.L., Lin, C.C. and  
355 Chiu, J.Y. (2017). Persistent organic pollutant reductions from a diesel engine generator  
356 fueled with waste cooking oil-based biodiesel blended with butanol and acetone. *Aerosol*  
357 *Air Qual. Res.* 17: 2041-2050.

358 Wang, Y., Liu, H. and Lee, C.F.F. (2016). Particulate matter emission characteristics of diesel  
359 engines with biodiesel or biodiesel blending: A review. *Renew. Sust. Energ. Rev.* 64:  
360 569-581.

361 Wu, G.Y.P., Lin, Y.F. and Chang, S.H. (2016). Emissions study and estimation of carbon  
362 dioxide production from *Jatropha curcas* oil biodiesel. *Aerosol Air Qual Res* 16: 1222-1233.

363 Xue, J., Grift, T.E. and Hansen, A.C. (2011). Effect of biodiesel on engine performances and  
364 emissions. *Renew. Sust. Energ. Rev.* 15: 1098-1116.

365 Yang, H., Li, X., Wang, Y., Mu, M., Li, X. and Kou, G. (2016). Pyrolysis characteristic analysis  
366 of particulate matter from diesel engine run on diesel/polyoxymethylene dimethyl ethers  
367 blends based on nanostructure and thermogravimetry. *Aerosol Air Qual Res* 16: 2560-2569.

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369 **Table Captions**

370 Table 1. PM mass concentrations and emission factors in DEG exhaust

371 Table 2. Oxygen contents of fuels.

372 Table 3. Particle-bound metal concentrations in DEG exhaust at 1.5 kW Load ( $\mu\text{g Nm}^{-3}$ ) (n = 3)

373 Table 4. Particle-bound metal concentrations in DEG exhaust at 3.0 kW Load ( $\mu\text{g Nm}^{-3}$ ) (n = 3)

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375 Table 1. PM mass concentrations and emission factors in DEG exhaust

Fuels	PM mass concentrations (mg Nm <sup>-3</sup> ) (n=3)		PM emission factors (mg L <sup>-1</sup> ) (n=3)	
	1.5 kW	3.0 kW	1.5 kW	3.0 kW
W20 <sup>a</sup>	<b>52.3</b> (± 2.7)	<b>94.7</b> (± 9.2)	<b>18.1</b> (± 1.9)	<b>28.5</b> (± 2.7)
B30	<b>33.9</b> (± 4.0)	<b>58.7</b> (± 3.3)	<b>13.8</b> (± 1.8)	<b>16.2</b> (± 2.0)
B'30	<b>28.8</b> (± 1.8)	<b>53.9</b> (± 1.7)	<b>11.1</b> (± 1.8)	<b>14.9</b> (± 0.9)
A3	<b>40.6</b> (± 3.0)	<b>76.3</b> (± 7.0)	<b>15.9</b> (± 1.2)	<b>22.9</b> (± 2.5)
A'3	<b>35.2</b> (± 2.4)	<b>70.2</b> (± 2.6)	<b>14.2</b> (± 1.3)	<b>20.6</b> (± 1.8)
B30A3	<b>30.9</b> (± 5.2)	<b>50.8</b> (± 3.9)	<b>12.7</b> (± 1.7)	<b>13.9</b> (± 3.1)
B'30A'3	<b>30.2</b> (± 4.3)	<b>49.5</b> (± 2.8)	<b>12.9</b> (± 2.1)	<b>13.6</b> (± 0.7)

<sup>a</sup> Cited from Lin et al. (2017)

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377

378 Table 2. Oxygen contents of fuels.

Oxygen contents (wt%)			
Anhydrous fuel blends		Hydrous fuel blends	
W20	2.31	–	–
B30	8.62	B'30	9.88
A3	3.34	A'3	3.46
B30A3	9.68	B'30A'3	11.1

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381 Table 3. Particle-bound metal concentrations in DEG exhaust at 1.5 kW Load ( $\mu\text{g Nm}^{-3}$ ) (n = 3)

Metals	W20 <sup>a</sup>	B30	B'30	A3	A'3	B30A3	B'30A'3
	Mean( $\pm$ SD)	Mean( $\pm$ SD)	Mean( $\pm$ SD)	Mean( $\pm$ SD)	Mean( $\pm$ SD)	Mean( $\pm$ SD)	Mean( $\pm$ SD)
Na	<b>81.4</b> ( $\pm$ 7.0)	<b>64.2</b> ( $\pm$ 16)	<b>74.8</b> ( $\pm$ 19)	<b>46.2</b> ( $\pm$ 15)	<b>69.3</b> ( $\pm$ 13)	<b>39.4</b> ( $\pm$ 4.7)	<b>35.5</b> ( $\pm$ 2.6)
Mg	<b>26.6</b> ( $\pm$ 4.1)	<b>20.3</b> ( $\pm$ 6.7)	<b>17.8</b> ( $\pm$ 14)	<b>17.5</b> ( $\pm$ 3.5)	<b>19.1</b> ( $\pm$ 18)	<b>16.2</b> ( $\pm$ 17)	<b>16.4</b> ( $\pm$ 3.8)
Al	<b>20.6</b> ( $\pm$ 2.3)	<b>19.1</b> ( $\pm$ 23)	<b>16.9</b> ( $\pm$ 1.7)	<b>18.0</b> ( $\pm$ 4.4)	<b>16.4</b> ( $\pm$ 4.7)	<b>14.4</b> ( $\pm$ 3.4)	<b>11.7</b> ( $\pm$ 2.8)
K	<b>56.1</b> ( $\pm$ 9.8)	<b>43.4</b> ( $\pm$ 13)	<b>40.2</b> ( $\pm$ 22)	<b>45.9</b> ( $\pm$ 8.3)	<b>45.3</b> ( $\pm$ 6.8)	<b>40.4</b> ( $\pm$ 22)	<b>36.1</b> ( $\pm$ 14)
Ca	<b>119</b> ( $\pm$ 12)	<b>148</b> ( $\pm$ 47)	<b>122</b> ( $\pm$ 41)	<b>135</b> ( $\pm$ 50)	<b>120</b> ( $\pm$ 44)	<b>113</b> ( $\pm$ 45)	<b>120</b> ( $\pm$ 27)
Ti	<b>N.D.</b>	<b>0.575</b> ( $\pm$ 0.31)	<b>0.575</b> ( $\pm$ 0.077)	<b>0.654</b> ( $\pm$ 0.099)	<b>0.646</b> ( $\pm$ 0.020)	<b>0.547</b> ( $\pm$ 0.078)	<b>0.563</b> ( $\pm$ 0.051)
V	<b>0.0719</b> ( $\pm$ 0.010)	<b>0.0923</b> ( $\pm$ 0.028)	<b>0.0844</b> ( $\pm$ 0.056)	<b>0.0801</b> ( $\pm$ 0.018)	<b>0.0834</b> ( $\pm$ 0.0098)	<b>0.0865</b> ( $\pm$ 0.014)	<b>0.0937</b> ( $\pm$ 0.015)
Cr	<b>2.03</b> ( $\pm$ 0.35)	<b>1.65</b> ( $\pm$ 0.84)	<b>1.73</b> ( $\pm$ 1.8)	<b>1.83</b> ( $\pm$ 2.4)	<b>1.74</b> ( $\pm$ 1.1)	<b>1.48</b> ( $\pm$ 0.54)	<b>1.20</b> ( $\pm$ 1.5)
Mn	<b>4.89</b> ( $\pm$ 1.3)	<b>1.13</b> ( $\pm$ 1.1)	<b>1.30</b> ( $\pm$ 0.55)	<b>1.66</b> ( $\pm$ 0.47)	<b>1.60</b> ( $\pm$ 0.18)	<b>1.12</b> ( $\pm$ 1.3)	<b>0.808</b> ( $\pm$ 0.10)
Fe	<b>152</b> ( $\pm$ 13)	<b>93.5</b> ( $\pm$ 141)	<b>84.2</b> ( $\pm$ 39)	<b>105</b> ( $\pm$ 53)	<b>89.5</b> ( $\pm$ 24)	<b>63.8</b> ( $\pm$ 27)	<b>62.7</b> ( $\pm$ 11)
Ni	<b>1.30</b> ( $\pm$ 0.20)	<b>1.03</b> ( $\pm$ 0.44)	<b>0.578</b> ( $\pm$ 0.46)	<b>0.81</b> ( $\pm$ 0.35)	<b>0.623</b> ( $\pm$ 0.65)	<b>0.136</b> ( $\pm$ 0.058)	<b>0.249</b> ( $\pm$ 0.24)
Cu	<b>1.00</b> ( $\pm$ 0.050)	<b>4.52</b> ( $\pm$ 4.4)	<b>6.27</b> ( $\pm$ 6.1)	<b>3.59</b> ( $\pm$ 0.95)	<b>5.84</b> ( $\pm$ 4.7)	<b>1.95</b> ( $\pm$ 0.065)	<b>1.87</b> ( $\pm$ 0.20)
Zn	<b>58.4</b> ( $\pm$ 3.7)	<b>50.2</b> ( $\pm$ 15)	<b>50.8</b> ( $\pm$ 20)	<b>59.7</b> ( $\pm$ 12)	<b>55.4</b> ( $\pm$ 3.4)	<b>44.3</b> ( $\pm$ 15)	<b>44.5</b> ( $\pm$ 1.6)
As	<b>N.D.</b>	<b>N.D.</b>	<b>N.D.</b>	<b>N.D.</b>	<b>N.D.</b>	<b>N.D.</b>	<b>N.D.</b>
Sr	<b>0.499</b> ( $\pm$ 0.11)	<b>0.455</b> ( $\pm$ 0.32)	<b>0.488</b> ( $\pm$ 0.12)	<b>0.612</b> ( $\pm$ 0.053)	<b>0.579</b> ( $\pm$ 0.056)	<b>0.597</b> ( $\pm$ 0.040)	<b>0.491</b> ( $\pm$ 0.23)
Mo	<b>0.350</b> ( $\pm$ 0.040)	<b>0.835</b> ( $\pm$ 0.38)	<b>0.694</b> ( $\pm$ 0.77)	<b>0.781</b> ( $\pm$ 0.11)	<b>0.738</b> ( $\pm$ 0.10)	<b>0.742</b> ( $\pm$ 0.16)	<b>0.765</b> ( $\pm$ 0.18)
Cd	<b>0.0658</b> ( $\pm$ 0.024)	<b>0.0608</b> ( $\pm$ 0.051)	<b>0.0604</b> ( $\pm$ 0.017)	<b>0.0280</b> ( $\pm$ 0.010)	<b>0.0368</b> ( $\pm$ 0.016)	<b>0.0299</b> ( $\pm$ 0.012)	<b>0.0571</b> ( $\pm$ 0.031)
Sn	<b>6.21</b> ( $\pm$ 3.4)	<b>3.28</b> ( $\pm$ 0.0027)	<b>2.81</b> ( $\pm$ 0.68)	<b>5.06</b> ( $\pm$ 0.12)	<b>3.82</b> ( $\pm$ 1.0)	<b>2.68</b> ( $\pm$ 0.18)	<b>3.24</b> ( $\pm$ 0.69)
Sb	<b>0.132</b> ( $\pm$ 0.049)	<b>N.D.</b>	<b>N.D.</b>	<b>N.D.</b>	<b>N.D.</b>	<b>N.D.</b>	<b>N.D.</b>
Ba	<b>0.411</b> ( $\pm$ 0.062)	<b>0.274</b> ( $\pm$ 0.21)	<b>0.393</b> ( $\pm$ 0.23)	<b>0.347</b> ( $\pm$ 0.206)	<b>0.303</b> ( $\pm$ 0.045)	<b>0.230</b> ( $\pm$ 0.016)	<b>0.301</b> ( $\pm$ 0.022)
Pb	<b>0.731</b> ( $\pm$ 0.10)	<b>0.878</b> ( $\pm$ 0.65)	<b>0.406</b> ( $\pm$ 0.18)	<b>0.343</b> ( $\pm$ 0.090)	<b>0.732</b> ( $\pm$ 0.081)	<b>0.0954</b> ( $\pm$ 0.038)	<b>0.145</b> ( $\pm$ 0.023)
$\Sigma$ metals	<b>531</b> ( $\pm$ 28)	<b>453</b> ( $\pm$ 264)	<b>422</b> ( $\pm$ 75)	<b>443</b> ( $\pm$ 108)	<b>431</b> ( $\pm$ 68)	<b>341</b> ( $\pm$ 103)	<b>337</b> ( $\pm$ 41)
$\Sigma$ metals content in PM (%)	<b>1.02</b> ( $\pm$ 0.10)	<b>1.35</b> ( $\pm$ 0.81)	<b>1.48</b> ( $\pm$ 0.35)	<b>1.11</b> ( $\pm$ 0.35)	<b>1.22</b> ( $\pm$ 0.14)	<b>1.14</b> ( $\pm$ 0.45)	<b>1.13</b> ( $\pm$ 0.23)

382 <sup>a</sup> Cited from Lin et al. (2017)

383 Table 4. Particle-bound metal concentrations in DEG exhaust at 3.0 kW Load ( $\mu\text{g Nm}^{-3}$ ) (n = 3)

Metals	W20 <sup>a</sup>	B30	B'30	A3	A'3	B30A3	B'30A'3
	Mean( $\pm$ SD)	Mean( $\pm$ SD)	Mean( $\pm$ SD)	Mean( $\pm$ SD)	Mean( $\pm$ SD)	Mean( $\pm$ SD)	Mean( $\pm$ SD)
Na	<b>225</b> ( $\pm$ 16)	<b>146</b> ( $\pm$ 12.3)	<b>96.7</b> ( $\pm$ 13)	<b>132</b> ( $\pm$ 6.1)	<b>106</b> ( $\pm$ 24)	<b>73.4</b> ( $\pm$ 5.6)	<b>79.0</b> ( $\pm$ 11)
Mg	<b>85.5</b> ( $\pm$ 4.7)	<b>64.7</b> ( $\pm$ 6.1)	<b>51.7</b> ( $\pm$ 3.8)	<b>58.1</b> ( $\pm$ 8.0)	<b>49.3</b> ( $\pm$ 11)	<b>49.6</b> ( $\pm$ 8.3)	<b>50.7</b> ( $\pm$ 23)
Al	<b>22.2</b> ( $\pm$ 0.40)	<b>28.7</b> ( $\pm$ 35)	<b>24.1</b> ( $\pm$ 3.6)	<b>27.5</b> ( $\pm$ 20)	<b>27.2</b> ( $\pm$ 1.2)	<b>23.0</b> ( $\pm$ 0.59)	<b>18.5</b> ( $\pm$ 3.5)
K	<b>101</b> ( $\pm$ 7.6)	<b>72.4</b> ( $\pm$ 9.1)	<b>56.7</b> ( $\pm$ 8.8)	<b>83.7</b> ( $\pm$ 21)	<b>65.9</b> ( $\pm$ 9.0)	<b>50.0</b> ( $\pm$ 7.6)	<b>44.4</b> ( $\pm$ 17)
Ca	<b>596</b> ( $\pm$ 83)	<b>398</b> ( $\pm$ 27)	<b>365</b> ( $\pm$ 11)	<b>378</b> ( $\pm$ 44)	<b>337</b> ( $\pm$ 71)	<b>302</b> ( $\pm$ 58)	<b>320</b> ( $\pm$ 55)
Ti	<b>N.D.</b>	<b>0.899</b> ( $\pm$ 0.19)	<b>0.844</b> ( $\pm$ 0.14)	<b>1.02</b> ( $\pm$ 0.33)	<b>0.978</b> ( $\pm$ 0.22)	<b>0.808</b> ( $\pm$ 0.084)	<b>0.791</b> ( $\pm$ 0.12)
V	<b>0.184</b> ( $\pm$ 0.021)	<b>0.173</b> ( $\pm$ 0.045)	<b>0.165</b> ( $\pm$ 0.033)	<b>0.208</b> ( $\pm$ 0.17)	<b>0.201</b> ( $\pm$ 0.014)	<b>0.154</b> ( $\pm$ 0.044)	<b>0.148</b> ( $\pm$ 0.022)
Cr	<b>68.9</b> ( $\pm$ 4.6)	<b>4.07</b> ( $\pm$ 4.6)	<b>3.93</b> ( $\pm$ 2.8)	<b>4.87</b> ( $\pm$ 0.84)	<b>4.25</b> ( $\pm$ 0.19)	<b>3.45</b> ( $\pm$ 2.1)	<b>2.96</b> ( $\pm$ 0.30)
Mn	<b>7.90</b> ( $\pm$ 0.093)	<b>2.82</b> ( $\pm$ 0.84)	<b>2.65</b> ( $\pm$ 1.6)	<b>3.96</b> ( $\pm$ 3.8)	<b>3.70</b> ( $\pm$ 1.1)	<b>2.50</b> ( $\pm$ 2.7)	<b>2.25</b> ( $\pm$ 0.53)
Fe	<b>204</b> ( $\pm$ 27)	<b>159</b> ( $\pm$ 44)	<b>152</b> ( $\pm$ 29)	<b>187</b> ( $\pm$ 49)	<b>147</b> ( $\pm$ 25)	<b>123</b> ( $\pm$ 2.0)	<b>106</b> ( $\pm$ 2.7)
Ni	<b>5.83</b> ( $\pm$ 0.48)	<b>1.39</b> ( $\pm$ 1.0)	<b>0.597</b> ( $\pm$ 0.042)	<b>2.88</b> ( $\pm$ 0.24)	<b>2.26</b> ( $\pm$ 0.45)	<b>1.08</b> ( $\pm$ 0.372)	<b>0.426</b> ( $\pm$ 0.12)
Cu	<b>2.75</b> ( $\pm$ 0.97)	<b>8.13</b> ( $\pm$ 2.1)	<b>7.55</b> ( $\pm$ 2.6)	<b>13.0</b> ( $\pm$ 6.0)	<b>9.61</b> ( $\pm$ 6.6)	<b>8.02</b> ( $\pm$ 1.7)	<b>5.38</b> ( $\pm$ 0.54)
Zn	<b>482</b> ( $\pm$ 20)	<b>316</b> ( $\pm$ 5.1)	<b>272</b> ( $\pm$ 30)	<b>266</b> ( $\pm$ 37)	<b>237</b> ( $\pm$ 111)	<b>221</b> ( $\pm$ 57)	<b>211</b> ( $\pm$ 17)
As	<b>N.D.</b>	<b>N.D.</b>	<b>N.D.</b>	<b>N.D.</b>	<b>N.D.</b>	<b>N.D.</b>	<b>N.D.</b>
Sr	<b>1.15</b> ( $\pm$ 0.36)	<b>1.05</b> ( $\pm$ 0.051)	<b>0.870</b> ( $\pm$ 0.090)	<b>1.27</b> ( $\pm$ 0.78)	<b>1.04</b> ( $\pm$ 0.39)	<b>0.887</b> ( $\pm$ 0.26)	<b>0.810</b> ( $\pm$ 0.25)
Mo	<b>2.25</b> ( $\pm$ 0.850)	<b>1.29</b> ( $\pm$ 0.24)	<b>1.27</b> ( $\pm$ 0.27)	<b>1.30</b> ( $\pm$ 0.60)	<b>1.29</b> ( $\pm$ 0.092)	<b>1.17</b> ( $\pm$ 0.24)	<b>1.18</b> ( $\pm$ 0.058)
Cd	<b>0.125</b> ( $\pm$ 0.0057)	<b>0.109</b> ( $\pm$ 0.33)	<b>0.121</b> ( $\pm$ 0.060)	<b>0.123</b> ( $\pm$ 0.031)	<b>0.116</b> ( $\pm$ 0.0082)	<b>0.105</b> ( $\pm$ 0.041)	<b>0.093</b> ( $\pm$ 0.027)
Sn	<b>5.46</b> ( $\pm$ 0.71)	<b>8.24</b> ( $\pm$ 0.40)	<b>8.25</b> ( $\pm$ 0.85)	<b>8.37</b> ( $\pm$ 0.25)	<b>8.68</b> ( $\pm$ 0.62)	<b>7.82</b> ( $\pm$ 1.6)	<b>7.90</b> ( $\pm$ 0.39)
Sb	<b>0.170</b> ( $\pm$ 0.019)	<b>N.D.</b>	<b>N.D.</b>	<b>N.D.</b>	<b>N.D.</b>	<b>N.D.</b>	<b>N.D.</b>
Ba	<b>0.365</b> ( $\pm$ 0.15)	<b>1.48</b> ( $\pm$ 0.80)	<b>1.33</b> ( $\pm$ 0.29)	<b>1.59</b> ( $\pm$ 0.55)	<b>1.44</b> ( $\pm$ 0.93)	<b>0.775</b> ( $\pm$ 0.20)	<b>0.542</b> ( $\pm$ 0.053)
Pb	<b>5.89</b> ( $\pm$ 2.8)	<b>2.50</b> ( $\pm$ 0.39)	<b>1.00</b> ( $\pm$ 0.15)	<b>2.16</b> ( $\pm$ 0.32)	<b>0.563</b> ( $\pm$ 0.23)	<b>0.609</b> ( $\pm$ 0.037)	<b>0.731</b> ( $\pm$ 0.17)
$\Sigma$ metals	<b>1817</b> ( $\pm$ 45)	<b>1218</b> ( $\pm$ 116)	<b>1047</b> ( $\pm$ 67)	<b>1173</b> ( $\pm$ 139)	<b>1004</b> ( $\pm$ 234)	<b>869</b> ( $\pm$ 99)	<b>853</b> ( $\pm$ 88)
$\Sigma$ metals content in PM (%)	<b>1.93</b> ( $\pm$ 0.17)	<b>2.07</b> ( $\pm$ 0.14)	<b>1.94</b> ( $\pm$ 0.07)	<b>1.53</b> ( $\pm$ 0.05)	<b>1.42</b> ( $\pm$ 0.28)	<b>1.72</b> ( $\pm$ 0.26)	<b>1.73</b> ( $\pm$ 0.27)

384 <sup>a</sup> Cited from Lin et al. (2017)

385 **Figure Captions**

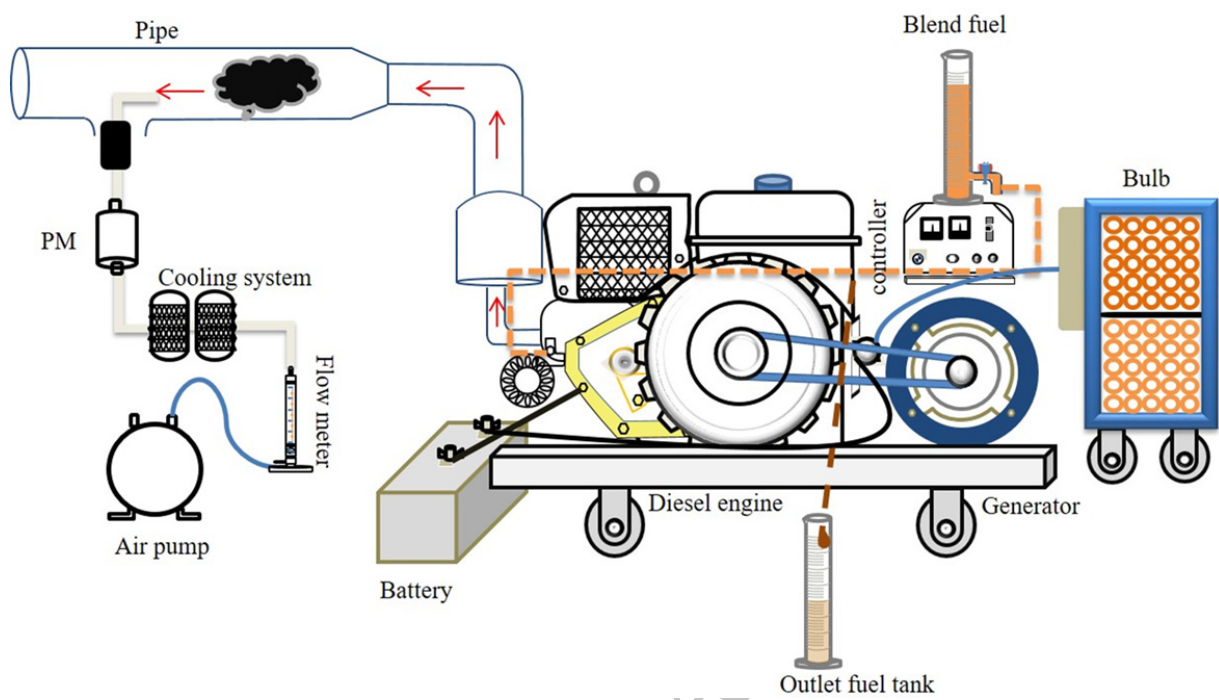
386 Fig. 1. The testing Diesel Engine Generator (DEG) system in this study.

387 Fig. 2. Reduction rates of PM emissions using various fuels in comparison with W20.

388 Fig. 3. Metal compositions of used fuels and lubricating oil.

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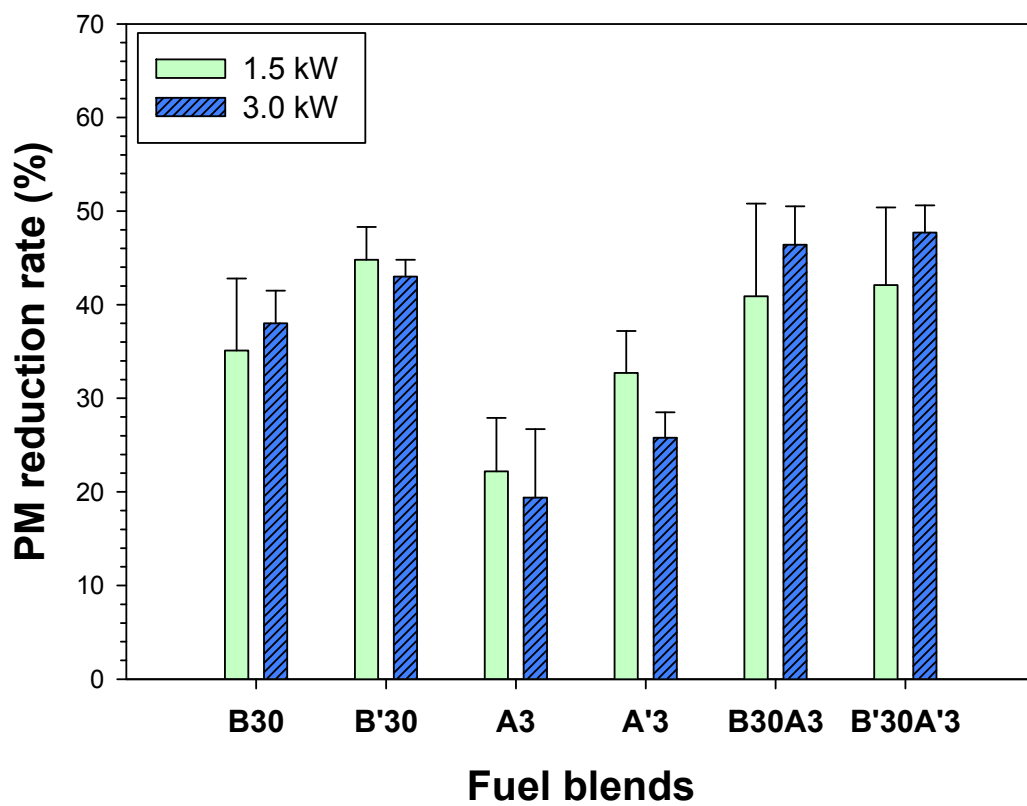
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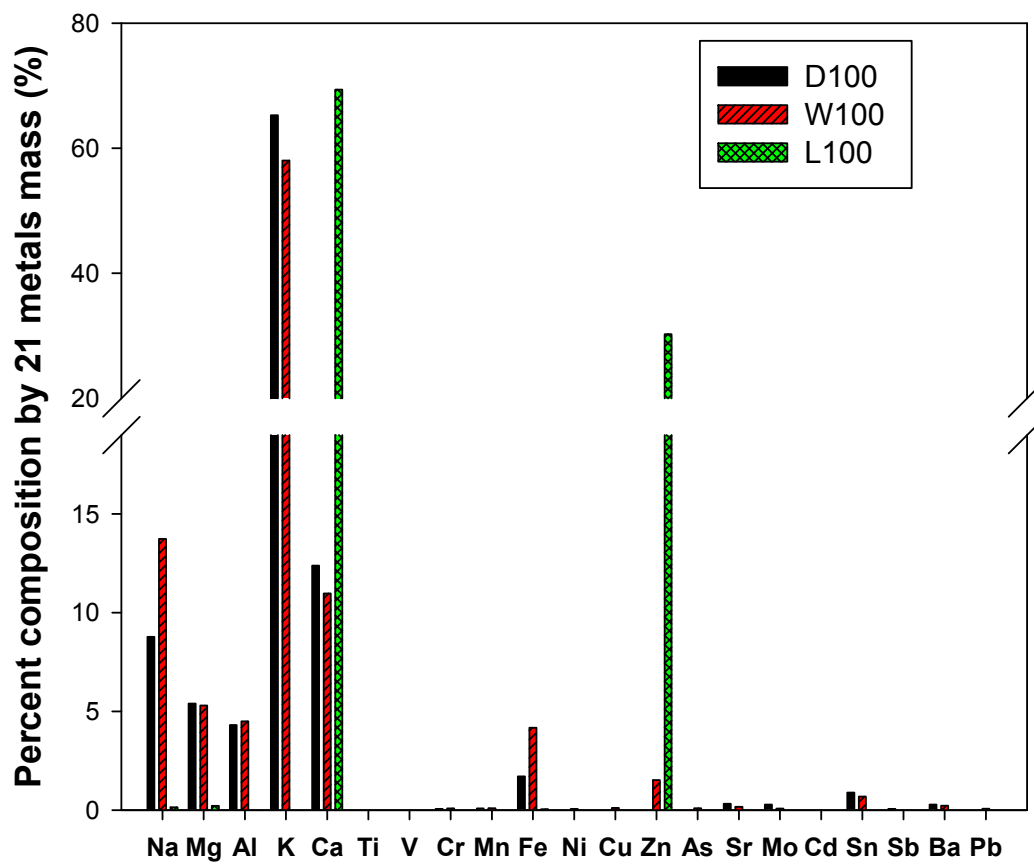
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Fig. 1.



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Fig. 2.



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Fig. 3.