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An Overview of the Influence of Biodiesel, Alcohols, and Various Oxygenated Additives on the Particulate Matter Emissions from Diesel Engines

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Abstract: Rising pollution levels resulting from vehicular emissions and the depletion of petroleum-based fuels have left mankind in pursuit of alternatives. There are stringent regulations around the world to control the particulate matter (PM) emissions from internal combustion engines. To this end, researchers have been exploring different measures to reduce PM emissions such as using modern combustion techniques, after-treatment systems such as diesel particulate filter (DPF) and gasoline particulate filter (GPF), and alternative fuels. Alternative fuels such as biodiesel (derived from edible, nonedible, and waste resources), alcohol fuels (ethanol, n-butanol, and n-pentanol), and fuel additives have been investigated over the last decade. PM characterization and toxicity analysis is still growing as researchers are developing methodologies to reduce particle emissions using various approaches such as fuel modification and after-treatment devices. To address these aspects, this review paper studies the PM characteristics, health issues, PM physical and chemical properties, and the effect of alternative fuels such as biodiesel, alcohol fuels, and oxygenated additives on PM emissions from diesel engines. In addition, the correlation between physical and chemical properties of alternate fuels and the characteristics of PM emissions is explored.

Keywords: biodiesel; particulate matter; PM; health effect; alternative fuel

1. Introduction

Recently, environmental issues have been exacerbated by harmful exhaust emissions and increasing levels of carbon dioxide (CO_2) from fossil fuels combustion [1]. Renewable fuels are a possible substitute for limited fossil fuel resources because the use of biobased fuels will not only help mitigate fossil fuel consumption, but also reduce greenhouse gas (GHG) emissions, and therefore help to address issues related to environmental protection and sustainable development [2]. The application of alternative fuels in internal combustion (IC) engines has gained interest over the years due to the potential for emissions reduction such as lowering particulate matter (PM) emissions [3].

In diesel engines the high temperature of compressed air allows the air/fuel mixture to reach the auto-ignition temperature and this combustion process causes the stored chemical energy to be released [4]. The products of an ideal combustion are H_2O and CO_2, while in reality a fraction of the fuel and lubricating oil remains unburnt. The incomplete combustion in diesel engines leads to emissions such as carbon monoxide (CO), hydrocarbons (HCs), nitrogen oxides (NOx), and particulate matter (PM), which are all regulated. There are also some unregulated emissions such as formaldehyde; acetaldehyde; 1,3-butadiene; ethene; ethyne; propylene; and benzene, toluene, and xylene (BTX) [5,6]. These byproducts of an incomplete combustion may contribute to many health issues such as asthma, chronic obstructive pulmonary disease (COPD), a decline in lung function, and pulmonary malignancy [7,8].

Thus, ongoing research activities are focussed on the abatement of hazardous emissions arising from diesel engines which include application of after-treatment devices such as diesel particulate filters (DPF), exhaust gas recirculation (EGR), lean NOx traps (LNT) and catalytic converters [9–12], and alternative fuels such as Fischer-Tropsch synthetic fuels [13], hydro-treated vegetable oils [14], and biodiesels [15,16]. In this paper, alternatives to diesel fuel and their impact on diesel engine PM emissions are reviewed.

The main focus of this review is PM, a byproduct of incomplete combustion, which is often composed of carbonaceous compounds along with adsorbed hydrocarbons. The chemical and physical characteristics of PM are significantly influenced by numerous parameters such as engine operating conditions (engine load and speed), fuel properties, injection parameters, and the lubricating oil used [17–20]. Thus, this paper has reviewed the health issues caused by the PM emissions and the influence of different fuels on the PM from diesel engines.

PM from diesel engines are primarily comprised of elemental carbon (EC), organic carbon (OC), soluble organic fractions (SOFs), as well as sulphates and ash. The metals in the exhaust gas when using heavy fuel oil used by marine engines are caused by metallic compounds (e.g., V, Ni, Fe) as well as from the lubricating oil (Ca, Zn), while for petroleum diesel most metals in the exhaust emissions are associated with the lubricants' composition [21]. In addition to the PM emissions, diesel engine exhaust also includes unregulated compounds such as benzene, alkanes, toluene, aldehydes, ketones, and xylene [22]. PM consists of very small sized particles (ranging from micrometre to nanometre) which enable easier penetration into the lungs and cause inflammation at the sites of deposition, and therefore make them hazardous [23–25]. Smaller sized particles such as nanoparticles (which have a size less than 50 nm) and ultra-fine particles (which have a size less than 100 nm) have a more hazardous impact than bigger particles which have a size less than 10 µm (PM_{10}) owing to the fact that diesel particulate filters are unable to filter the smaller sized particles.

Due to the negative effects of PM on the human respiratory system, the size of particles is a crucial parameter in engine emissions analysis [26]. PM characterization and toxicity analysis is still growing as researchers are developing methodologies to reduce particle emissions using various approaches such as fuel modification and after-treatment devices. To address these aspects, this paper presents a review of the characteristics and health effects of PM, as well as the effect of numerous fuels (biodiesel, alcohol fuels, and oxygenated additives) on PM emissions from diesel engines [27,28]. The main objective of this study is to review the impact of different oxygenated fuels (biodiesel from different feedstock, alcohol fuels, and oxygenated additives) to understand their impact on PM emissions. As the share of biofuels is increasing in the fuel market, it is necessary to understand the impact of biofuels on PM emissions from different perspectives (feedstock for biodiesel, chemical composition and engine type, and operation conditions). This manuscript is a systematic study to review the literature on the use of alternative fuels in diesel engines. The article covers the health effects of PM emissions, and their key chemical and physical properties. The following section, being core of this paper, deals with the influence of fuel type on PM emissions in relation to the role of fuel properties.

2. Health Issues Caused by Particulate Matter Emissions

Human health is vulnerable to air pollution from vehicles. PM emissions are considered to be hazardous due to their harmful environmental and health effects [29]. For this reason, PM has been categorized among regulated emissions and are consistently being given stricter regulatory guidelines by authorities such as the European Union and the United States Environmental Protection Agency (EPA) [6]. Numerous studies have demonstrated an association between PM emissions and both short- and long-term adverse health effects [30–33], including increased hospital admissions, morbidity, and mortality secondary to cardiorespiratory disease [34]. Wichmann [29] broadly classified the adverse health effects of PM into three types: acute (short-term exposure), chronic (long-term exposure) non-carcinogenic, and chronic carcinogenic effects.

Acute irritation (e.g., eye, throat, and bronchial), respiratory symptoms (cough and phlegm), and neurophysiological symptoms (for example nausea) are common acute health effects of exposure to PM [29]. In addition, short term exposures to PM have also been proposed to increase cardiovascular risk, possibly by the activation of systemic inflammatory responses [35,36].

The chronic adverse health effects associated with long-term exposure to PM have been reported in numerous epidemiological studies. Specifically, long-term exposure to PM has been linked to the development and exacerbation of chronic respiratory diseases (such as asthma and COPD), lung carcinogenesis, and cardiovascular disease with a significant impact on morbidity and mortality [7,29,37,38].

Ristovski et al. [7] emphasized the importance of the chemical and physical properties of PM in the development of respiratory disease. Studies have reported that inflammatory injury, oxidative damage, and other biological adverse effects are heavily dependent on the size of emitted particles and their surface area [39–41]. The aforementioned effects are more extensive with exposure to smaller sized particles (ultrafine and nanoparticles) due to their ability to penetrate deep into lung tissue reaching the alveolar spaces, where 50% of particles can be retained [42].

Smaller sized particles also have increased specific surface area, which in addition to the adsorbed organic compounds, is a significant factor implicated in the mechanism by which they cause adverse health effects [7]. The chemical characteristics of PM such as organic and elemental carbon content, and the presence of trace metals and polycyclic aromatic hydrocarbons (PAHs), is strongly related to fuel quality and properties [17,43]. Thus, it is important to investigate the influence of fuels on PM emissions.

The adverse effects of PM can be attributed, at least in part, to the toxic substances absorbed on $PM_{2.5}$ (size less than 2.5 μm) such as PAHs, which mainly originate from incomplete combustion processes such as coal combustion, biomass burning, and vehicle exhaust, and are ubiquitous in the atmosphere. PAHs, which are rich in graphene, are formed mainly in regions of high local fuel/air equivalence ratio and high temperature due to enhanced residence time. PAHs adsorb various chemical components such as alcohol groups aliphatic C-H groups which affect soot reactivity [44].

The potential health risk posed by PAHs could be greater because they can undergo a series of chemical reactions in the atmosphere, thus producing more toxic derivatives. These include nitrated PAHs which have much higher mutagenicity, and hydroxylated PAHs which have higher cytotoxicity and oxidation potential than PAHs [45].

3. Properties of PM

3.1. Physical Properties of Diesel Particulate Matter (DPM)

DPM presents a complex mixture of various compounds and can be derived from different sources. According to the latest research, the composition of DPM is ultimately linked to the composition of their precursors. Size will govern their toxicity and lifetime in the atmosphere. Smaller particles will reside longer in the atmosphere, and thus will be more prone to atmospheric transformations that lead to the formation of secondary pollutants. Smaller particles will also be transported further and are generally considered to be more toxic than their bigger counterparts, due to their higher surface area and the fact that they can penetrate deeper in lungs. Larger particles are normally deposited close to their sources and are removed in the upper respiratory tract.

Coarse particles have diameters from 2.5 to 10 μm and originate mainly from mechanical processes, such as breaking, grinding, etc. Fine particles have diameters smaller than 2.5 μm, while particles smaller than 100 nm are called ultrafine particles. The main source of fine and ultrafine PM is combustion (biomass burning, traffic, waste burning, etc.). A typical size distribution of urban aerosols is characterized by three modes which are nucleation, accumulation, and coarse mode.

Another very important classification, commonly used by regulatory bodies and the general public, divides particles into the following four categories: $PM_{0.1}$, PM_1, $PM_{2.5}$ and PM_{10}, which are particles with diameters smaller than 0.1, 1, 2.5, and 10 μm, respectively. These size classifications are used in air quality standards and very often in public health-related studies. These size fractions are reported as mass concentration. As mentioned, the size of particles is an effective parameter when it comes to PM characterization.

The physical and chemical properties of PM generated through fuel combustion of diesel and alternative fuels will depend on different parameters such as fuel properties, engine type, operating conditions, and fuel injection characteristics. Primary pollution gets transformed quickly under the influence of atmospheric and meteorological factors. Oxidation in the air by atmospheric oxidants and the dilution will transform the primary pollution and lead to the formation of secondary pollution. Secondary pollutants, including PM and volatile organic compounds (VOCs), are a result of condensation of oxidized VOCs on particles, nucleated semi-volatiles or oxidized primary PM. Dilution is not chemically altering the composition of non-labile PM species, in other words, OC (organic carbon), BC (black carbon), heavy weight organic compounds, organic molecular traces, and metals. However labile species will be significantly affected by the first dilution stage, mainly through nucleation, coagulation, and condensation. Robinson et al. [46] argued that the organic component of PM depends on the dilution, whereas fuel-based emission factors of organics decrease with increasing dilution and related decreasing concentration. Semi-volatiles will experience nucleation and/or condensation increasing the chemical complexity of the primary PM. The EC/OC ratio varies for different fuels or combustion conditions, defined by different engine operating conditions. Moreover, VOCs either adsorb or absorb on the surface of EC or into the already existing organic layer coating the carbon, respectively. In addition, atmospheric aging by hydroxyl radical (OH·), ozone (O_3), and the nitrate radical ($NO_{3·}$) which are the most abundant oxidants in the atmosphere, will lead to a further transformation of primary emissions, which produces secondary emissions. Secondary inorganic emissions are associated with the formation of sulphates, nitrates, and ammonium which are fairly well understood, while the secondary organic aerosol formation is still not entirely understood.

3.2. Chemical Composition of DPM

Diesel exhaust emissions contain more than 20,000 different compounds [47]. Therefore, the chemical composition of DPM and biofuel PM is very complex and it is common in the literature to use the ratio of organic and elemental carbon (OC/EC) to indicate the bulk particulate composition. Organic fractions of DPM and biofuel PM originate from the incomplete combustion of fuel and lubricating oil and it is reported that the oxygenated fraction of PM, oxygenated organic aerosol (OOA), depends on the oxygenated content of the fuel [48]. Liang et al. reported that the main constituents of DPM are hydrocarbons, mainly cycloalkanes, alkanes, alkylbenzenes, and polycyclic aromatic hydrocarbons (PAHs) and their derivatives [49]. Engine operating conditions significantly influence the composition of DPM, primary PM, while meteorological and atmospheric factors, in general, further govern the composition of PM, secondary PM. For example, higher loads produce PM with higher EC content [50–52]. The incomplete combustion of a diesel engine results in smoke and PM emissions which mainly have three constituents: soot particles, organics condensed or adsorbed on soot particles, and sulphates [53].

DPM, especially biofuel PM, usually exhibits bimodal size distribution. Nucleation mode mainly contains condensed VOCs and a small fraction of solid material [54]. Despite the large body of research, the composition of organic phase is not fully speciated, and the mechanisms of nucleation and the role of organics in the nucleation process are to be understood. On the other hand, accumulation mode particles are composed of higher hydrocarbons, metals, and compounds containing sulphur. The coarse mode particles come from the deposition and re-entertainment of materials from the engine cylinder and the exhaust manifold. Apart from the composition of DPM, diesel/biofuel exhaust contains a gaseous phase that contains sulphuric acid, SO_2, SO_3, H_2O, nitrates, and volatile organic compounds.

The chemical structure of the fuel influences the chemical composition of both the gas and particle phase of the exhaust. It has been established that the presence of oxygen in the fuel changes the composition and toxicity of the particle and gas phase [25]. Generally, this means that fuels with higher oxygenated content will produce PM with higher OC and gas phase with lower OC, as compared with diesel [55]. Carbon chain length and the degree of unsaturation are also very important fuel composition parameters for the composition of the resulting PM [55–57]. Linoleic acid, which is normally dominates biodiesel fatty acid composition, is known to readily oxidize and produce higher soot and NOx concentration.

4. Influence of Fuel Type

Alternative fuels such as straight vegetable oils, biodiesel, alcohol fuels (ethanol, n-butanol, n-pentanol), hydrogen, liquefied petroleum gas, methane, and compressed natural gas have gained the attention of researchers due to their similar performance and improved emissions characteristics as compared with diesel.

There have been numerous studies that focused on PM emissions from diesel engines fueled with alternative fuels [16,58–66]. This section discusses the effect of different types of fuels such as biodiesel, alcohol fuels, and oxygenated additives on diesel engine PM emissions. In general, the focus of this study has been on PM mass, particle number (PN) concentration, and particle size distribution for use of different fuels. The changes in PM emissions (particulate mass and size distribution) depend upon fuel properties, engine operating parameters such as speed, load, injection pressure, as well as engine ambient conditions and modern combustion techniques.

4.1. Biodiesel

Biodiesel is a key alternative fuel due to its renewable nature and its physico-chemical properties as compared with petroleum diesel [67,68]. Biodiesel is the alkyl ester from straight vegetable oils, nonedible oils, animal fats, microalgae or waste cooking oils [69–72]. The key characteristic of biodiesel is its renewable nature and lower GHG emissions.

There have been numerous studies that focused on the use of biodiesel from edible oils such as palm oil [73–75], soybean oil [74,76,77], and coconut oil [78–80]. The addition of soybean biodiesel to petroleum diesel resulted in a 2–3% decrease in PM emissions [9]. The higher oxygen content of biodiesel resulted in better combustion in localized fuel rich zones which reduced PM emissions [26]. However, the use of edible oils for biodiesel is potential competition for food crops, thus, it is contentious as a wholesale replacement for fossil fuels [81,82].

Recently, efforts have been made to produce biodiesel from nonedible oils and waste resources such as jatropha [17,83,84], karanja [22,85,86], waste cooking oil [65,87–89], cottonseed oil [90,91], and microalgae [56,87,92,93]. The higher oxygen content in biodiesel causes lower PM emissions [94]. With increasing biodiesel percentage in the fuel blend the oxygen content of the fuel increases, hence lower PM emissions occurs [11,95,96].

Zhu et al. [62] observed that the high-oxygen content in the fuel reduced PM emissions at high and medium engine loads. Satputaley et al. [97] reported oxygen content in micro-algae oil and biodiesel as the main reason for reduction in smoke emissions. The oxygen in the fuel blend improved the combustion process which resulted in lower PM emissions [98]. Similar observations were made by

Cheung et al. [99] for waste cooking biodiesel. Nabi et al. [100] used a wide range of oxygen content (from 0 to 14%) during European stationary cycle (ESC) and reported that PM emissions were decreased by the oxygen content in the fuel. Similar conclusions were made when a smaller range of fuel oxygen content was used [89]. Another study on the effect of oxygenated fuels used non-road transient cycle (NRTC) and a custom transient test (acceleration and load increase), and it was reported that the fuel oxygen content was the reason for the decrease of PM emissions [57].

The major factors that contribute to the lower formation of soot precursors and the reduction of PM emissions are the presence of strong double bonds in the ester groups in biodiesel (C and O), and the absence of aromatic compounds and sulphur [17,101,102]. Although some studies have concluded that there is a reduction in total PN emissions with the use of biodiesel blended fuels [55,62,100,103], an increase in nucleation mode particles has been reported for biodiesel in numerous studies [63,104,105]. A number of studies have recommended that in diesel engines that have a mechanical fuel pump, a higher injection pressure was required during combustion of biodiesel blended fuel due to the higher fuel density and viscosity. This led to higher nucleation mode particles [63,104,106,107]. Similar results were reported for the use of karanja biodiesel combustion [105]. It has been reported that lower availability of excess oxygen and higher in-cylinder temperature at higher loads promotes the soot nucleation and a higher number of nucleation mode particles [86].

In addition, some studies have studied the relationship of the variations in PN emissions with engine operating conditions. Man et al. [108] observed, for waste cooking oil biodiesel, that the number of particles having a smaller diameter was higher when the engine was operated at higher speeds or lower loads. For decreasing engine loads or increasing engine speed, the primary particle diameter decreased gradually. More unburnt biodiesel particles are accumulated at higher engine speeds due to the shorter residence time, thus, causing the formation of small diameter particles. Lower engine speeds result in more particle nucleation thus, allowing surface growth and resulting in the formation of larger sized particles [108]. Engines operated at lower loads have shorter combustion duration and lower in-cylinder temperature and pressure inhibiting the generation and growth of nucleation mode particles, which results in a higher formation of smaller particles.

Bugarski et al. [109] observed up to a 7% reduction in count mean diameter (CMD) of particles and 13–24% lower PM emissions with the use of renewable diesel fuels as compared with ultra-low sulphur diesel in a turbocharged diesel engine. Behçet et al. [110] found smoke opacity for cooking oil and fish oil biodiesel reduced by 7.871% and 15.36% as compared with diesel fuel. Similarly, a reduction from 6.2–60.5% was observed in smoke by Buyukkaya [111] for a 10–50% blend of biodiesel in diesel. The major reason for reduction in smoke and PM emissions for biodiesel fuels has been mentioned as oxygen content in the fuel which enhances the soot oxidation process and PM formation is reduced during the combustion process [103].

A brief summary of observations made for typical biodiesel fuels is presented in Table 1.

Table 1. A summary of observations made for typical biodiesel fuels.

No.	Fuel Used	Engine Used	Operating Conditions	Findings	Reference
1.	Diesel and Jatropha biodiesel	Four-cylinder, 2520 cc direct injection (DI) engine with 40 hp rated power at 2300 rpm.	Engine was run at 1800 rpm for five different loads from 0 to 100%.	PM mass ranged from ~28–40 mg/m^3 for diesel and ~22.5–32 mg/m^3 for biodiesel blended fuel.	[17]
2.	Diesel and Karanja biodiesel	Four-cylinder common rail DI diesel engine with rated power of 84.5 kW at 3000 rpm.	Engine was run at different load conditions at 1800 rpm for diesel and 20% blend of biodiesel in diesel	Lower particulates were emitted with the use of biodiesel blended fuel. At higher engine loads, reduction in particulates was more significant.	[16]
3.	Diesel and rice bran biodiesel	Four-cylinder, 2520 cc DI engine with 40 hp rated power at 2300 rpm.	Engine was run for diesel and 20% blend of biodiesel in diesel at different load conditions from 0–100% load.	PM mass ranged 17–48 mg/m^3 and 22–59 mg/m^3 for biodiesel blended fuel and mineral diesel respectively.	[101]
4.	Diesel and rapeseed oil biodiesel	2720 cc, water-cooled, twin-turbocharged V6 diesel engine with rated power 152 kW at 4000 rpm.	16 different test conditions with torque varying from 4–41% and EGR (vol.%) varied from 7.4–61 as per New European Driving Cycle for neat diesel and 30% of biodiesel in diesel.	Average size of particles for biodiesel blended fuel reduced by 41% as compared with diesel.	[104]
5.	Diesel European Norm (EN) 90 and commercially available pure biodiesel	Six-cylinder DI engine with 160 kW rated power at 1900 rpm.	Three different conditions: 50% load at 1600 rpm, 25% load at 1900 rpm and 75% load at 1900 rpm.	For all operating conditions, particle number concentration reduced by 30.4–66.2 % for blends up to 75% blend of biodiesel in diesel as compared with diesel, and increased by 4.76–66.22% for higher blends.	[112]
6.	Diesel and Karanja biodiesel	3000 cc, four-cylinder, common rail DI diesel engine having maximum power 84.5 kW at 3000 rpm.	Engine was run at 1800 and 2400 rpm and different loads conditions from 0–100% for diesel and 20% blend of biodiesel in diesel.	PM mass for 7.9 and 17.2 mg for 20% blend of biodiesel in diesel as compared with 13.3 and 17.6 mg for neat diesel respectively at 1800 and 2400 rpm.	[102]
7.	Diesel and cottonseed oil biodiesel	Single cylinder diesel engine with rated power of 4.476 kW at 1800 rpm.	Engine was run with diesel and 20% biodiesel at different loads.	PM emissions reduced by 24% for biodiesel blended fuels.	[113]
8.	Diesel and Jatropha biodiesel	Four-cylinder common rail DI diesel engine with rated power of 79 kW at 3200 rpm.	Engine was run at three different speeds for varying load conditions.	Accumulation mode particle reduced, however, total particle number concentration increased with biodiesel blended fuels.	[114]

Table 1. *Cont.*

No.	Fuel Used	Engine Used	Operating Conditions	Findings	Reference
9.	Diesel and waste cooking biodiesel	Six-cylinder diesel DI engine.	Engine was run at different load conditions for 2–20% blend of biodiesel.	With increase in biodiesel percentage in fuel blend particle number concentration decreased for all loads.	[115]
10.	Diesel and soybean biodiesel	Four-cylinder turbocharged DI diesel engine with rated power of 103 kW at 4000 rpm.	Engine was run at five stationary operating conditions and a transient cycle for 10–50% biodiesel blends.	Geometric mean diameter reduced for all biodiesel blended fuels.	[116]
11.	Diesel and used cooking oil biodiesel	Four-cylinder diesel engine with rated power of 85.22 kW at 4000 rpm.	Engine was run at five different operation modes for 30 and 70% biodiesel blends.	Soot, PM emissions and particle number concentration reduced significantly with biodiesel.	[117]
12.	Diesel and Licella biofuel	Four-cylinder diesel engine with rated power of 100 kW at 4000 rpm.	Engine was run at four different loads for 5–20% of biofuel in diesel.	PM mass reduced for all biofuel blends. A maximum of 30% reduction was observed for 20% blend of biofuel.	[118]
13.	Diesel and waste cooking biodiesel	Four-cylinder DI diesel engine with rated power of 88 kW at 3200 rpm.	Engine was run according to Japanese 13-mode test cycle for different biodiesel blends.	A reduction of 18–48% in PM mass concentration for different biodiesel blends.	[119]
14.	Diesel and commercial fatty acid methyl ester	Six-cylinder diesel engine with 10,350 cc capacity and rated power of 160 kW at 1900 rpm.	Engine was run at different modes according to 13-mode steady state cycle.	Total number and volume of particle emissions were minimum for 75% of biodiesel blend in diesel.	[112]
15.	Diesel, waste cooking biodiesel and triacetin	Six-cylinder common rail turbocharged diesel engine with rated power of 162 kW at 2000 rpm.	Engine was run at a custom 13-mode steady state cycle.	With the increase in biodiesel in the fuel blend, PM, PN, and accumulation mode counter median diameter of the particles decreased gradually.	[103]

Although biodiesel is renewable in nature since it is derived from biobased resources and it offers a significant reduction in CO, HC, and CO_2 emissions, an increase in NOx emissions has been reported in the literature with the use of biodiesel [120–123]. In addition, low oxidation and storage stability of biodiesel are also problems [124,125].

The chemical composition of biodiesel plays a key role in determining its fuel properties. Biodiesel is derived from feedstock that is rich in unsaturated fatty acids and has relatively low oxidation stability [126]. Similarly, biodiesel produced from feedstock that is rich in saturated fatty acids has poor cold flow properties. Oxidation of biodiesel is accelerated as it comes in contact with oxygen in ambient air, metals (storage containers), and elevated temperature [127]. Although the use of additives like antioxidants or cold flow depressants may solve these problems, there are other fuel alternatives like alcohol fuels such as ethanol, n-butanol, and n-pentanol which are derived from biobased resources and have comparable oxidation stability to diesel fuel [128,129].

Later references reported that in addition to the operating conditions such as engine speed, load, fuel injection timing and pressure, the ambient conditions also played a key role in PM emissions of a diesel engine. It has been observed that engine operation at colder conditions suffered with more PM mass as compared with the normal conditions. Poor volatility characteristics of biodiesel fuels during cold-start of engine resulted in higher PM emissions. Figure 1 shows the comparisons among the different fuel blends on PM emissions of diesel engine in different ambient conditions.

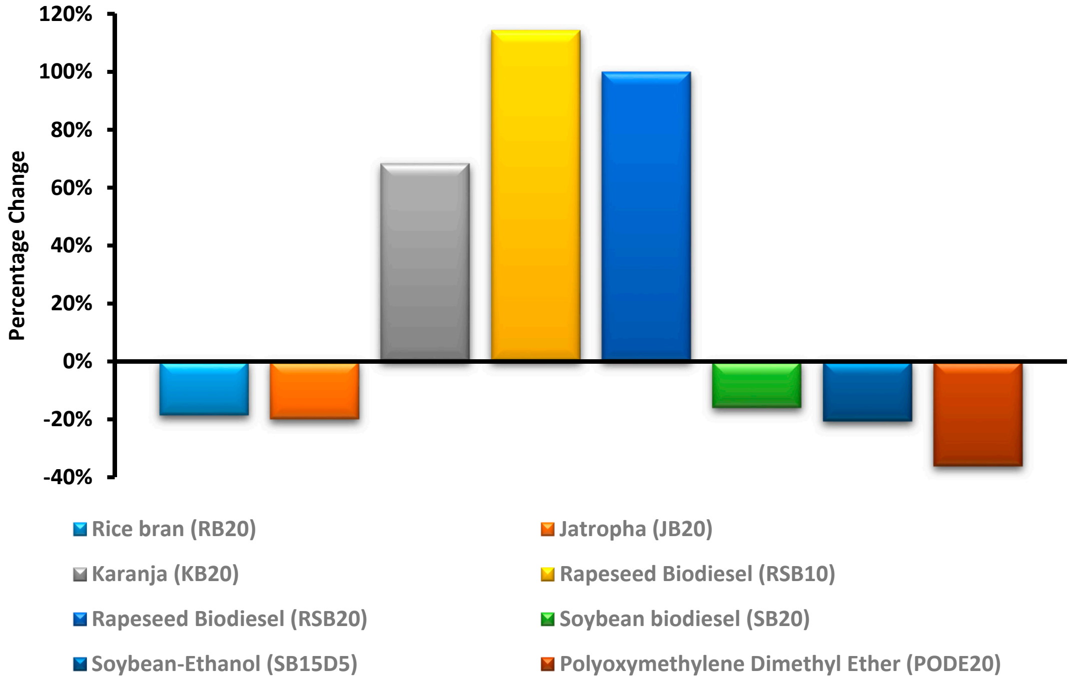

Figure 1. Percentage changes in particulate mass (PM) for different fuels for cold-start as compared with hot-start of engine [17,64,101,102,130,131].

As shown in Figure 1, blends of Karanja and Rapeseed biodiesel in diesel had higher PM emissions in cold-start conditions as compared with the use of other biodiesel fuels in hot-start or normal operating conditions. Engine operating conditions played a key role in this as it was run in cold ambient conditions (−7 °C). Inferior volatility characteristics of biodiesel as compared with petroleum diesel resulted in higher PM emissions during cold-start conditions. Longer cranking period, higher fuel injection quantity, poor lubrication, fuel evaporation, and combustion conditions are the crucial problems for the cold-start.

4.2. Alcohol Fuels

Alcohols such as ethanol can be derived from sugarcane wastes (e.g., molasses) and has already been adopted in a number of countries as an additive to diesel [70,132–137]. The use of longer-chain alcohols such as n-butanol and n-pentanol have also emerged as potential alternatives to diesel fuel [62,138–140]. Alcohols with longer carbon chain length have merit over shorter chain alcohols due to their higher calorific value, cetane number, improved miscibility with diesel, and superior cold flow properties [69,85]. The lower viscosity and density of alcohol blended fuels can improve the fuel atomization and fuel-air mixing consequently reducing the PM emissions [141–143]. A brief summary of observations made by different researchers for different alcohol fuels is presented in Table 2.

Table 2. Literature summary of observations made for different alcohol fuels.

No.	Fuel Used	Engine Configuration	Operating Conditions	Emission Findings	Reference
1.	Methanol, waste cooking biodiesel and diesel	4334 cc, four-cylinder diesel engine with rated power 88 kW at 3200 rpm.	Engine was run at steady speed of 1800 rpm for varying BMEP from 0.08 to 0.70 MPa for diesel, biodiesel, and 5–15% of methanol in pure biodiesel.	Geometeric mean diameter for methanol-biodiesel blended fuels reduced as compared with diesel.	[59]
2.	Ethanol, used cooking oil and diesel	425 cc single-cylinder diesel engine.	Engine was run at 1500 rpm and 600 kPa indicated mean effective pressure for conventional combustion and low temperature combustion for diesel, biodiesel, and 20% ethanol in biodiesel.	Particle number concentration in exhaust for different fuels can be ordered as diesel-biodiesel-diesel-ethanol blend.	[144]
3.	Ethanol, waste cooking and diesel	Four-cylinder naturally aspirated water cooled diesel engine.	Engine was run at steady speed of 1800 rpm for varying torque and BMEP from 28–240 Nm and 0.08–0.70 MPa for diesel, biodiesel, and 5–15% of ethanol in pure biodiesel).	Lowest PM emissions was found for ethanol and biodiesel blended fuels.	[65]
5.	Ethanol, biodiesel and diesel	Single-cylinder diesel engine with rated power 10 hp connected to generator at 1800 rpm.	No load and speed of 1800 rpm. Fuels: B5, B100, and biodiesel ethanol additive.	Lower PM emissions for ethanol additive fuel is due to its molecular structure better stability. Higher emission in biodiesel blended fuels as compared with ethanol blended which could be due to higher viscosity of biodiesel.	[145]
6.	Ethanol, waste cooking biodiesel and diesel	4334 cc, four-cylinder DI diesel engine with rated power of 88 kW at 3200 rpm.	Engine was run at steady speed of 1800 rpm for varying engine torque from 30 to 240 Nm and brake mean effective pressure (BMEP) from 0.09 to 0.70 MPa.	Ethanol and biodiesel blended in diesel had minimum particle mass and particle number concentration which could be attributed to lower aromatics and sulphur.	[88]
7.	Ethanol, soybean biodiesel and diesel	Four-stroke diesel, 1272 cc with speed rate of 1800 rpm.	Engine was run in stationary mode with 70% load for different fuel blends of ethanol, biodiesel, and diesel.	Significant reduction in particle number concentration and average size of particles with pure biodiesel and ethanol blended biodiesel.	[146]

Table 2. *Cont.*

No.	Fuel Used	Engine Configuration	Operating Conditions	Emission Findings	Reference
8.	Butanol, palm biodiesel and diesel	296 cc, single-cylinder four-stroke direct injection (DI) engine with 4.5 kW at 3000 rpm.	Engine was run at steady speed of 3000 rpm and load corresponding to 25, 50, and 75% of rated power for different load conditions for neat ultra-low sulphur diesel and different blends of diesel-biodiesel-butanol.	Reduction in elemental carbon for butanol and biodiesel blended fuels.	[3]
9.	Butanol, pentanol and diesel	296 cc, single-cylinder four-stroke DI engine with 4.5 kW at 3000 rpm.	Engine was run at steady speed of 3000 rpm and load corresponding to 25, 50, and 75% of rated power for different load conditions for diesel, butanol, and pentanol blended fuels.	Diesel-butanol blends resulted in more reduction in elemental carbon, solid and volatile particle number emissions as compared with pentanol-diesel blend.	[139]
10.	n-pentanol, waste cooking biodiesel and diesel	4334 cc, four-cylinder DI diesel engine with rated power of 88 kW at 3200 rpm.	Engine was run at steady speed of 1800 rpm for different torques from 18–224 Nm and BMEP 0.08–0.65 MPa for different fuels i.e., diesel, biodiesel (B100), and biodiesel-pentanol fuel (BP10, BP20, BP30).	Total particle number concentration decreased for biodiesel-pentanol blends as compared with biodiesel or diesel fuels.	[62]
11.	Butanol and diesel	Single-cylinder DI diesel engine with rated power of 3.5 kW at 1500 rpm.	Engine was run at different loads at 1500 rpm for 10–30% blends of butanol in diesel.	Smoke opacity, particle mass, and particle number concentration decreased significantly with butanol-blended fuels.	[147]
12.	Butanol and diesel	Six-cylinder diesel engine with maximum power of 162 kW at 2500 rpm.	ESC cycle (3 speeds under 4 different engine loads). Fuels were the blends of butanol and diesel (10, 20, and 30% butanol blended in diesel).	PM decreased by increasing the share of butanol in the blend.	[140]

There have been several studies which studied the effect of n-pentanol on PM emissions from diesel engines [62,138,139]. The total PM number concentration was found to be lower for n-pentanol blended fuels as compared with diesel and biodiesel [62]. The lower cetane number and viscosity of n-pentanol resulted in a longer ignition delay. The OH group and higher intensity of aliphatic groups assisted in inhibiting soot precursors. These factors all contributed to the reduction of PM emissions. The R-OH group present in n-pentanol was much more effective in inhibiting the formation of soot precursors as compared with the ester groups in biodiesel. Furthermore, with n-pentanol, a notable reduction in the number of nucleation mode particles as compared with biodiesel.

Soot oxidation is mostly caused by aliphatic C-H and oxygenated groups. The soot oxidation plays an important role in soot emissions from diesel engines. Internal burning and rapid oxidation reaction are key phases of soot oxidation in biodiesel combustion. Soot oxidation is sped up by the removal of the outer shell layer due to desorption of the surface oxygen group in the open edge sites. Once the core has internal burning, the oxidation process becomes even faster with a layered rearrangement [148]. Zhu et al. [61,65] stated that the higher heat release rate for ethanol combustion was the key reason for the lower PM formation as it increases the rate of soot oxidation. Higher concentrations of OH in methanol and ethanol can reduce soot precursor formation in the fuel rich zones, which promotes soot oxidation to CO and CO_2, and reduces the PM mass and PN concentration. The use of alcohol fuels has been proven to be a good alternative to petroleum diesel for reducing dependence on fossil fuel resources and in curtailing engine emissions [3,62,88]. However, the low energy density of alcohol fuels, especially ethanol, can affect the engine performance considerably [138].

4.3. Oxygenated Additives

The use of oxygenated additives as a blending agent not only reduces PM emissions, but may also change the physio-chemical properties of the particulates, hence, their toxicological characteristics [149,150]. To date, several oxygenated fuels such as dimethyl ether (DME) [151–155], 2,5-dimethylfuran (DMF) [156–165], dimethyl carbonate (DMC) [58,166–170], diethyl adipate (DEA) [5,58,170–172], triacetin [18,19,57,100,103,173–177], and diethylene glycol dimethyl ether (DGL) [178] have been studied for their application as additives to diesel or biodiesel fuels. Properties of different oxygenated additives are shown in Table 3 and compared with standard diesel. It is noted that all the additives have high oxygen content (by weight).

Table 3. Properties of different oxygenated additives [153,155,156,159–162].

Property	Diesel	DMF	DME	DGL	DEA	Triacetin	DMC
Oxygen content (wt%)	0	16.7	34.8	35.8	36.7	44	53.30
Chemical formula	C_9 to C_{25}	C_6H_8O	C_2H_6O	$C_6H_{14}O_6$	$C_{10}H_{18}O_4$	$C_9H_{14}O_6$	$C_3H_6O_3$
Molecular weight (g/mol)	~96	96.12	46.07	134.17	-	218.20	90.08
Density (kg/m^3)	800–840	890	660–668 (at 20 °C)	-	1005 (at 20 °C)	1159 (at 15 °C)	1069–1073 (at 15 °C)
Cetane number (-)	40–55	9	55–60	126	15	15	35
Lower heating value (MJ/kg)	40–45	33.7	28.43	24.5	25.5	16.78	15.7
Flash point (°C)	≥55	−1	−41	67	-	-	18
Stoichiometric air fuel ratio	14.5	10.79	9.0	11.4	-	-	4.59
Auto ignition temperature (°C)	-	285.85	-	187.78	-	-	195

4.3.1. Dimethyl Ether

DME is quite volatile and is a liquid at pressure higher than 0.5 MPa. Its liquid nature makes handling and storage of DME straight forward. There are a number of factors which makes DME useful as a fuel additive:

1. It is safe to store and transport because it does not form peroxides which are explosive in nature.

2. It is highly oxygenated (35% oxygen by molecular weight) which can result into lower PM mass emissions.
3. DME's higher cetane number leads to improved combustion resulting in lower emission of PM, toxic gases, CO, and hydrocarbons [154].

DME can be produced using indirect or direct synthesis methods. DME can be generated indirectly through a dehydration reaction involving methanol, while direct synthetic methods produce DME directly from natural gas [153]. The chemical structure of DME is as shown in Figure 2.

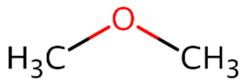

Figure 2. Structural formula of DME.

Nabi and Hustad [179] studied the performance and emission characteristics of a diesel engine with different fuels such as marine gas oil (MGO) as baseline fuel, 10% blend of jatropha biodiesel in diesel (JB10), and 10% blend of diethylene glycol dimethyl ether in diesel (DME10). It was concluded that PM emissions for different fuels were in the order of MGO > JB10 > DME10. The reduction in PM mass emissions was attributed to a lack of aromatics, lower sulphur content, and higher oxygen content in JB10 and DME10. Similar results were obtained in another study where PM mass and PN emissions were lower for oxygenated fuels as compared with diesel fuel [144]. Sirignano et al. [180] studied the impact of DME as an oxygenated additive and found a reduction in particulate matter with a 10–30% addition of DME.

4.3.2. 2.5-Dimethylfuran

Recently, it has been discovered that DMF (structure shown in Figure 3) could be an alternative fuel for diesel engines due to its potential for mass production. In contrast to ethanol and n-butanol, the higher calorific value of DMF results in higher engine power. In addition, DMF does not readily absorb moisture from the atmosphere which is beneficial for storage [163].

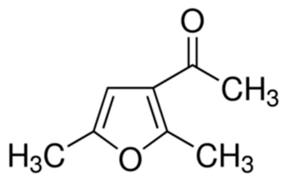

Figure 3. Structural formula of DMF.

Chen et al. [160] identified that due to the combined effect of higher oxygen content and longer ignition delay, a 30% blend of DMF was more effective in reducing PM emissions as compared with 30% butanol. The longer ignition delay with DMF enhanced the time to premix fuel and air resulting in a reduction of the local equivalence ratio which resulted in soot reduction. Zhang et al. [156] compared the effect of different blends of DMF in diesel on PM emissions and observed that a 40% blend of DMF in diesel was more effective for PM reduction as compared with using 20% DMF in the blend and also as compared with the neat diesel. This was attributed to its higher oxygen content, longer ignition delay, and enhanced diffusive combustion.

4.3.3. Dimethyl Carbonate

DMC (structure shown in Figure 4) is an ester that can be derived from reacting waste CO_2 and methanol from power stations in the presence of a catalyst such as potassium chloride.

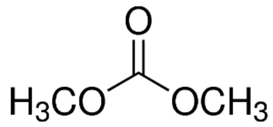

Figure 4. Structural formula of DMC.

DMC is biodegradable, nontoxic, and has good miscibility with diesel fuel [168], as well it has high oxygen content (53% by weight) which is useful for the reduction of PM emissions. The oxygen content of fuel impacts its behaviour during combustion by increasing the local air/fuel ratio in fuel rich zones of the combustion chamber thus affecting in-cylinder temperature and emission products. In the structure of DMC, each O atom is paired with a C atom which results in the formation of CO avoiding the C-C bonding which is a key source of smoke formation. In addition, the higher oxygen content makes up for the low local air/fuel ratio with OH radicals, and therefore oxidizes the unsaturated hydrocarbon compounds inhibiting the growth of soot particles. The molecular structure of DMC includes oxygen atoms bonded to carbon atoms forming CO. Hence, the absence of carbon-carbon bonds in the fuel moiety contributes to hydrocarbon oxidation rather than participation in soot growth reactions [167,168,181].

4.3.4. Diethyl Adipate

DEA (structure shown in Figure 5) can be derived from the esterification of adipic acid and ethanol in the presence of concentrated sulphuric acid [175].

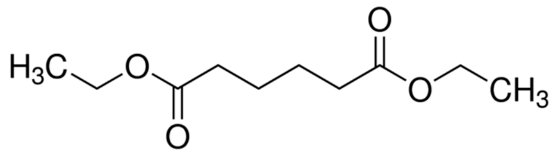

Figure 5. Structural formula of DEA.

DEA emerged as a potential additive due to its high oxygen content and superior miscibility in diesel, low sooting tendency, and fuel properties comparable to diesel fuel [171]. Moreover, DEA is a colourless liquid that has relatively low toxicity, corrosivity, reactivity, and less sooting tendency as compared with petroleum diesel. Zhang et al. [58] reported that a longer ignition delay with fuels having DEA as an additive improved the combustion in premix mode and improved the engine performance. The addition of 2% and 4% DEA in palm oil biodiesel resulted in 9.4% and 18.2% reduction in PM emissions. In addition, 29.5% and 44.9% reductions were observed for EC (soot) emissions. It was suggested that pyrolysis, decomposition of oxygenates, and the rise in free radical concentration in premix flames inhibit the growth of soot precursors. A surprising observation was made in the PN concentration of nanoparticles. The addition of DEA to the fuel resulted in an increased number of nanoparticle emissions at a low engine load, whereas, at high and medium engine load, there was not a substantial difference.

In a similar study [170], it was found that the total PN concentration for DEA additive fuels (2% and 4%) was reduced by 21.3% and 36.7% as compared with diesel. In addition, there was a significant reduction in the number of accumulation mode particles, whereas, a marginal increase in the number of nucleation mode particles was observed. Consequently, the gradual mean diameter of the particles reduced by 7.45% and 13.92% with the addition of 2% and 4% DEA, respectively. Zhu et al. [171] blended 8.1–33.8% DEA in diesel to vary the oxygen content in fuel blends by 3–12%. It was observed that PM and PN emissions reduced significantly for all fuels, whereas, SOFs increased with the increase in DEA. There were diverse results observed in particle size distribution for different blends. For medium and high loads, more particles of size within a 60–100 nm range were observed.

4.3.5. Triacetin

Triacetin is a triglyceride (1, 2, 3-triacetoxypropane) and also is often known as glycerol triacetate. It is the tri-ester of acetic acid and glycerol. Figure 6 shows the structural formula of Triacetin.

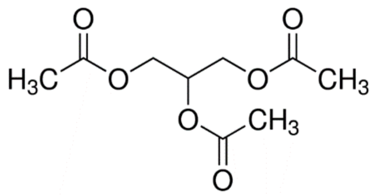

Figure 6. Structural formula of Triacetin.

The high oxygen content and antiknocking characteristics of triacetin makes it a potential fuel additive [182]. Furthermore, it has the ability to enhance cold flow properties and oxidation stability of biodiesel [183]. High selectivity and high conversion rate for producing triacetylglycerol from the byproduct glycerol can also be obtained using a two-step method. Firstly, during biodiesel production in trans-esterification reaction, glycerol is produced. Secondly, glycerol is esterified with acetic acid over resin and zeolites, and Amberlyst-35 has been found to be an excellent catalyst [183]. The addition of 4, 8, and 10% of triacetin to biodiesel increased the oxygen content of B100 (10.93%) to 12.25, 13.57 and 14.23%, respectively [103]. A gradual reduction in PM emissions was observed for engine combustion with oxygenated fuels. Figure 7 shows that the use of waste cooking biodiesel instead of diesel decreases the PM emissions significantly. Furthermore, it is shown in the figure that the addition of triacetin to WCO biodiesel decreases the PM emissions. Fuel oxygen content was reported as the main reason for this reduction. It was also reported that using triacetin as an oxygenated additive decreases PN emissions significantly and the emitted particles were smaller than for diesel.

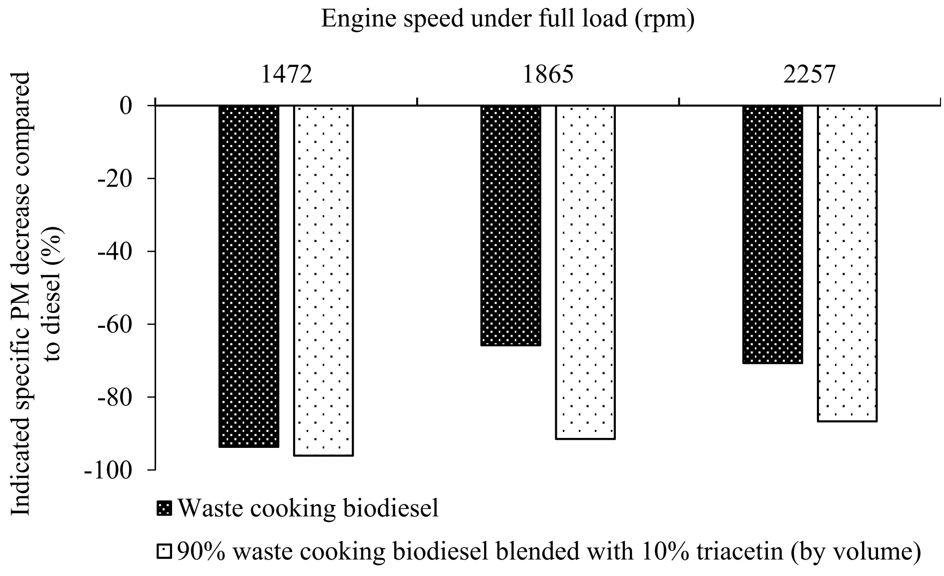

Figure 7. Indicated specific PM variation as compared with diesel [103].

5. Summary

The rising population, rapid industrialization, and stringent emission standards have caused researchers to investigate alternative fuels to fossil diesel. The increased use of biodiesel and alcohol fuels has had promising results for vehicle industry in recent times. In contrast to petroleum diesel, low sulphur levels, higher oxygen content, and the lack of aromatics make these fuels suitable as an alternative to diesel fuel. In addition, numerous oxygenated additives such as DME, DMC, DMF, DEA, DGL, and triacetin have emerged as additives to biodiesel and diesel fuels as they have high levels of oxygen and an ability to reduce PM mass emissions significantly without affecting physiochemical properties of the base fuel. An attempt has been made to review the influence of different fuels on PM and PN emissions from diesel engines. Henceforth, the following general conclusions can be drawn from the literature review:

1. The feedstock from which biodiesel is produced plays a key role in PM emissions. The high viscosity of biodiesel fuel leads to poor atomization of biodiesel fuel resulting in high soot formation in the middle of the combustion process. The presence of strong double bonds (C and O) in the ester structure of biodiesel, results in lesser availability of carbon, causing lesser formation of soot precursors, thus reflecting in lower PM emissions for the biodiesel combustion. Increasing the fatty acid chain length results in an increase in nucleation mode particles.
2. Although some researchers have reported a reduction in overall PN concentration for engine combustion with biodiesel and alcohol fuels, there is an increase in the number of nucleation mode particles. This is a serious concern for operating engines with biofuels because smaller sized particles are more hazardous to human health because they penetrate deeper into the respiratory system and blood cells.
3. Among different alcohols, n-butanol has the best ability to reduce PM emissions due to its highest oxygen content.
4. Oxygenated additives such as triacetin are more effective in reducing the PM mass as compared with pure biodiesel.

6. Future Prospects

Considering the rising share of biofuels in the market and the development of after-treatment devices, it is necessary to understand the formation of soot particles and their reactivity. The PM from combustion of fuel is trapped in the diesel particulate filter. The trapped PM has to be removed for proper function of the filter and this process involves the oxidation of soot particles (also called the regeneration of filter). The regeneration of filter relies upon the reactivity of soot particles. As future legislations are making it mandatory to use biofuels, it is essential to investigate the morphology, nanostructure, and oxidation reactivity of soot particles emitted from the combustion of biofuels. This will serve as a platform to better understand filtration efficiency, regeneration, and design of diesel particulate filters.

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