

Unignorable Emissions and Potential Health Effects of Unregulated Pollutants from Nonroad Engines Using Greener Fuels — A Review

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ABSTRACT

Nonroad mobile machinery such as construction equipment, agricultural machinery, ships, and generators emit significant pollutants. Countries worldwide have implemented stringent emission regulations targeting pollutants like CO, HC, NO_x, and PM. However, these machines also emit unregulated pollutants such as PAHs, VOCs, POPs, and metals, which severely impact human health and the atmosphere. A key strategy to reduce these emissions is using green fuels, including biodiesel, oxygenated fuels, metal additives, hydrogen, and ammonia. This paper analyzes the impact of these fuels on non-road machinery emissions of unregulated pollutants. Results indicate that green fuels can reduce some harmful emissions but may increase others. Biodiesel and oxygenated fuels significantly reduce PAHs and soot emissions, but their effect on VOCs varies with fuel type and engine load. Metal additive fuels effectively reduce EC/OC, smoke, and particle number emissions but may increase total PAHs and BaP_{eq} emissions. Currently, research on using metal additive fuels, hydrogen, and ammonia in generators is limited, focusing mainly on biodiesel and oxygenated fuels. Similarly, studies on unregulated emissions from ships using clean fuels are scarce. Most research concentrates on PAHs, PN, and EC/OC emissions, with less focus on POPs, N₂O, VOCs, and metal components.

Keywords: Nonroad engine, Oxygenated fuels, Metal additive fuels, Greener fuel, Unregulated pollutants

1 INTRODUCTION

General mobile machinery encompasses both road mobile machinery, such as cars, trucks, motorcycles, and buses, and Nonroad mobile machinery, including aircraft, ships, railway internal combustion engines, agricultural, and construction machinery. In recent years, with the growing concerns surrounding global air pollution and climate change (Karl and Trenberth, 2003), significant attention has been directed towards studying emissions from road vehicles (Hitchins *et al.*, 2000;

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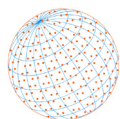
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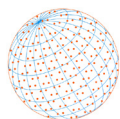


Wu *et al.*, 2017). However, research on Nonroad machinery remains relatively scarce. Studies have revealed that sulphur oxide (SO_x) emissions from shipping constitute approximately 60% of global transport SO_x emissions, while nitrogen oxide (NO_x) emissions contribute to about 40% of global freight NO_x emissions. Additionally, CO₂ emissions represent 2%–3% of global CO₂ emissions (Dalsøren *et al.*, 2009; Goldsworthy, 2010). Notably, in the Vancouver area, ship emissions have been associated with higher concentrations of SO₂, CO, PM_{2.5}, and VOCs (especially methyl compounds) in coastal areas compared to local inland concentrations of the same compounds (Lu *et al.*, 2006). Moreover, other Nonroad machinery, such as construction machinery, agricultural machinery, and generator sets, also produce significant pollutant emissions (Lindgren, 2004). For example, agricultural machinery in Switzerland emitted nearly 300 tonnes of particulate matter in 2010 (Hagan *et al.*, 2022), while construction machinery in London accounted for 15% of total PM_{2.5} emissions in 2016, with PM₁₀ accounting for 34% of emissions (Desouza *et al.*, 2020). Forestry/logging Nonroad mobile machinery, handheld generators and chainsaws (Lijewski *et al.*, 2017), trimmers, snow removal equipment (Millo *et al.*, 2002), etc., all highlight the significance of emissions from Nonroad mobile machinery.

As evident from the above, the use of Nonroad mobile machinery results in the production of a variety of pollutants. To mitigate further climate deterioration, various countries have implemented increasingly stringent emission standards to regulate harmful emissions from Nonroad machinery (MEE, 2007, 2008; European Commission, 2012). However, current emissions regulations primarily target particulate matter (PM), carbon monoxide (CO), nitrogen oxides (NO_x), and hydrocarbons (HC). Nonetheless, Nonroad machinery also emits numerous pollutants not subject to regulations, including soot (Ris, 2007), polycyclic aromatic hydrocarbons (PAHs) (Wang *et al.*, 2021), volatile organic compounds (VOCs) (Fang *et al.*, 2022), and persistent organic pollutants (POPs) (Tsai *et al.*, 2015), among others. These pollutants pose serious threats to human health and the atmospheric environment. The International Agency for Research on Cancer (IARC) classifies diesel engine exhaust as a human carcinogen (Group IIB) (IARC, 2012). Prolonged exposure to these unregulated pollutants can result in respiratory, nervous, and cardiovascular system disorders, leading to various diseases, including lung cancer, liver cancer, and stomach cancer (Rumchev *et al.*, 2007; IARC, 2019). Hence, it is imperative to control the emission of harmful pollutants from Nonroad machinery.

With the implementation of emission reduction policies in various countries, scholars have proposed various measures to mitigate emissions. One of the most significant measures involves the use of alternative fuels. Palm oil (Zhang and Balasubramanian, 2014b; Nguyen and Le, 2019), waste cooking oil (Tsai *et al.*, 2011), soybean oil (Tsai *et al.*, 2010; Jayaram *et al.*, 2011), microalgae biodiesel (Mwangi *et al.*, 2015), sugarcane biodiesel (Gysel *et al.*, 2014) and other green fuels serve as viable alternatives to fossil fuels for Nonroad machinery. Additionally, diesel/biodiesel fuel can be supplemented with oxygen additives or metal particle additives (Karthikeyan and Dharma Prabhakaran, 2018; Zhang *et al.*, 2022). Common oxygen-containing additives include methanol (Datta and Mandal, 2016) and ethanol (Merritt *et al.*, 2005; Lee *et al.*, 2011), butanol (Zhang and Balasubramanian, 2014a; Mwangi *et al.*, 2015), acetone (Lin *et al.*, 2010; Tsai *et al.*, 2014b), various types of ether fuels (Zhang and Balasubramanian, 2015; Agarwal *et al.*, 2022; Zacherl *et al.*, 2022) and emulsified fuels (Lin *et al.*, 2012; Tsai *et al.*, 2014a; Lin *et al.*, 2022). When used as alternative fuels, the options can effectively reduce the emission of PAHs, VOCs, POPs, and other unregulated pollutants in exhaust gases.

In recent years, numerous studies have focused on reducing unregulated pollutant emissions from non-road mobile machinery through alternative fuels. Due to the high costs and technical challenges of monitoring emissions from aircraft and railway engines, this paper only addresses other non-road mobile machinery. This paper systematically reviews the impact of green fuels—such as biodiesel, oxygenated additive fuels, metal additive fuels, and hydrogen/ammonia—on the emissions of unregulated pollutants (e.g., PAHs, VOCs, POPs) from generators, ships, and other non-road mobile machinery. By comparing different green fuels, the study reveals their advantages and disadvantages in reducing specific pollutant emissions, providing insights and recommendations for future emission reduction efforts.



2 EMISSIONS FROM THE TRADITIONAL FUELS IN NONROAD ENGINE

2.1 Emission Regulations for Nonroad Engines

Nonroad mobile machinery (NRMM) encompasses a wide array of equipment, including construction machinery (e.g., excavators, bulldozers, loaders, forklifts, road rollers, pavers, graders), agricultural machinery (e.g., tractors, combine harvesters, agricultural transporters, irrigation and drainage machinery), small general-purpose machinery (e.g., hand-held or non-hand-held lawnmowers, chain saws, hedge trimmers), diesel generator sets, ships (passenger and cargo ships), railway diesel locomotives, and civil aircraft (MEE, 2007; Hagan *et al.*, 2022). NO_x and PM emissions from Nonroad mobile machinery in China in 2022 are 82% and 339% of the corresponding pollutant emissions from road mobile machinery (motor vehicles) (MEE, 2022). This indicates the severity of the pollutant emission issue associated with Nonroad mobile machinery. To address this concern, countries worldwide have enacted increasingly stringent emission standards for Nonroad machinery. While EU emission regulations are generally more rigorous and standardized, China's restrictions on certain pollutants are stricter. Consequently, the following comparison focuses solely on EU and China's emission regulations for Nonroad mobile machinery.

In 2019, the European Union began implementing the fifth phase of Nonroad machinery emission standards (referred to as Euro V) (European Commission, 2012), while China commenced the implementation of the fourth phase of emissions standards (referred to as CH IV) in December 2022 (MEE, 2007). Additionally, countries have established emission regulations for marine engines. In 2009, the European Union initiated the implementation of the third phase of emission standards (referred to as Euro IIIA) (European Commission, 2012), whereas China began implementing the second phase of emission standards (below CH II) in July 2018 (MEE, 2008). These continuously updated and implemented emission standards progressively reduce emission limits and effectively mitigate pollutant emissions. Various regulations specify emission limits for carbon monoxide (CO), hydrocarbons (HC), NO_x, PM, etc., for different types of Nonroad mobile machinery. Fig. 1 and Fig. 2 provide comparisons of EU/China emission standards for Nonroad mobile machinery and ships, respectively. However, pollutants emitted by engine combustion not only include those regulated by these standards but also encompass various unregulated pollutants such as soot, sulfate, soluble organic matter (SOF), metal elements, volatile organic compounds (VOCs),

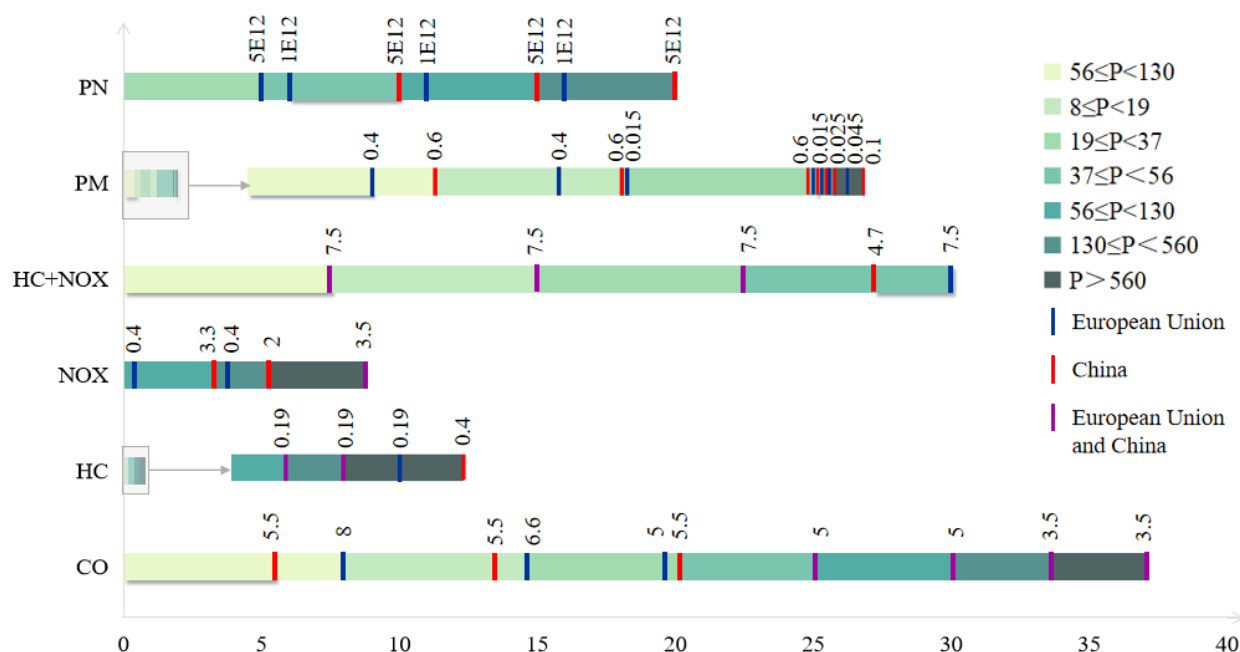


Fig. 1. China and the European Union for Nonroad mobile machinery emission standards.

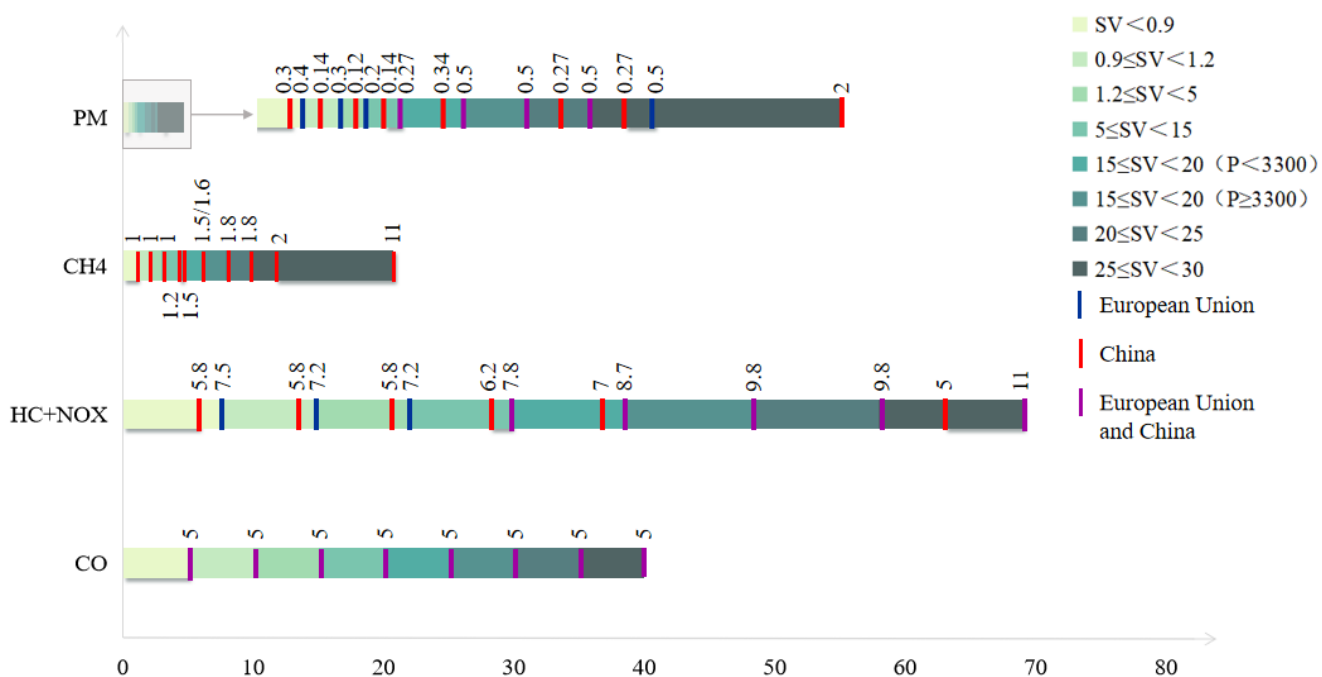
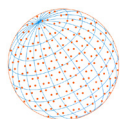


Fig. 2. China and the European Union for Inland Waterway Vessels emission standards.

and persistent organic pollutants (POPs) (Moldanová *et al.*, 2009), which can have potential impacts on the atmospheric environment and human health.

2.2 Effects of Regulated and Unregulated Pollutants on Human Health

Pollutants emitted by engine combustion are categorized into particulate matter and gaseous matter (Kittelson, 1998). Particulate matter includes soot (mainly composed of elemental carbon (EC) and organic carbon (OC)), sulfate, soluble organic matter (SOF), metal components, semi-volatile organic matter (heavy hydrocarbons, polycyclic aromatic hydrocarbons, etc.), polar organic compounds, etc. (Moldanová *et al.*, 2009). Gaseous substances encompass regulated gas pollutants, SO₂, volatile organic compounds (VOCs), persistent organic pollutants (POPs), among others. Different pollutants cause varying degrees of harm to human health (refer to Fig. 3). The National Cancer Research Center classifies the pollutants into four groups (WHO, 2020): I (Carcinogenic to humans); IIA (Probably carcinogenic to humans); IIB (Possibly carcinogenic to humans); III (Not classifiable as to its carcinogenicity to humans), as detailed in Table 1. The composition, formation process, and health effects of soot, PAHs, VOCs, POPs, and metal components will be discussed in detail in the following sections.

2.2.1 Soot and carbon

Soot, constituting nearly 50% of PM, is formed from unburned fuel. The remaining portion primarily comprises soluble organic parts (SOF), sulfates, metals, aldehydes, alkanes, olefins, aliphatic hydrocarbons, polycyclic aromatic hydrocarbons, and their derivatives. Generally, the transformation from fuel to soot involves six processes: pyrolysis, nucleation, surface growth, coalescence, agglomeration, and oxidation (Tree and Svensson, 2007). Soot primarily comprises carbonaceous components (EC and OC). EC originates from fuel droplet pyrolysis, while OC arises from unburned fuels, lubricants, and combustion byproducts (Shah *et al.*, 2004). SOF is a polycyclic aromatic compound containing oxygen, nitrogen, and sulfur. SOF in particles can transition from the gas phase to the particle phase through nucleation or adsorption (Zhang and Balasubramanian, 2014a). Most sulfur in fuels will oxidize to SO₂, while a small portion will oxidize to SO₃, resulting in exhaust particles containing sulfuric acid and sulfate. Prolonged exposure to soot particles can induce respiratory diseases such as bronchitis, pneumonia, and asthma (Lighty *et al.*, 2000); it can also impact the nervous system and cardiovascular function.

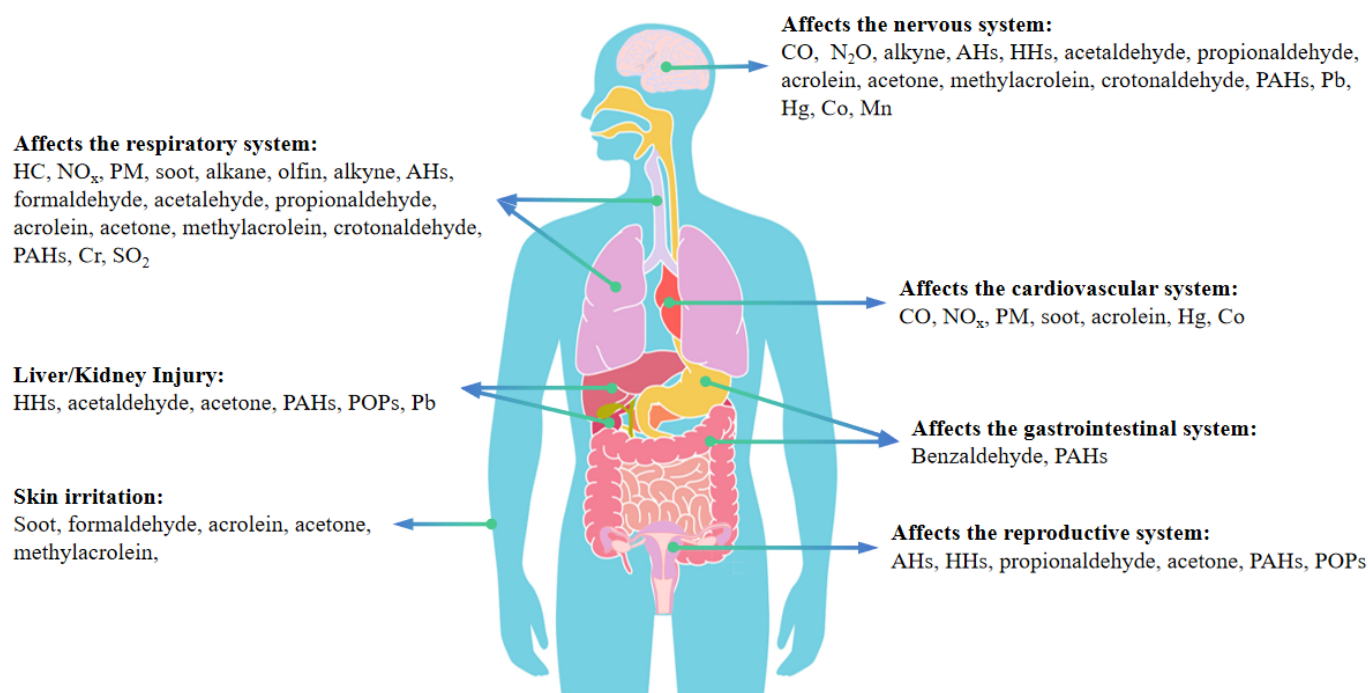
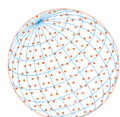


Fig. 3. Effects of different pollutants on human health.

Table 1. IARC classification of carcinogenicity of pollutants in engine exhaust.

Classification	Pollutants
I	1,3-Butadiene, Formaldehyde, benzo[a]pyrene; Polychlorinated biphenyls, Arsenic, Cadmium.
IIA	Styrene, Polybrominated biphenyls, Acrolein, Lead.
IIB	Ethylbenzene, Acetaldehyde, Crotonaldehyde, Naphthalene, Anthracene, Benzo[b]fluoranthene, Benzo[k]fluoranthene, Chrysene, Dibenzo[a,h]pyrene, Indeno[1,2,3-cd]pyrene, Nickel.
III	Sulfur dioxide, Propylene, Propionaldehyde, Acenaphthene, Coronene, Benzo[b]chrysene, Benzo[e]pyrene, Benzo[ghi]perylene, Pyrene, Fluoranthene, Fluorene, Phenanthrene, Polychlorinated dibenzofurans, Mercury, Chromium.

^I Carcinogenic to humans.

^{IIA} Probably carcinogenic to humans.

^{IIB} Possibly carcinogenic to humans.

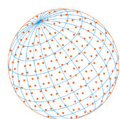
^{III} Not classifiable as to its carcinogenicity to humans.

2.2.2 Metal components

Inorganic elements in engine exhaust particles mainly comprise Zn, Fe, Cu, Si, S, Cl, Mo, Ni, Mn, and Ca. Enriched elements in PM include Zn, Fe, Mn, S, Ca, and Cr, with Zn and Fe exhibiting the highest concentrations. Zn in PM primarily originates from lubricant and bearing wear, while Fe results from the wear of cylinder liners, pistons, valves, gears, and other components. Mn derives from steel shafts and valves, S from fossil fuels and additives, Ca as a byproduct of lubricants and additives, and Cr from the wear of steel in diesel engines (Tsai *et al.*, 2018b). Additionally, the metal components of PM encompass Pb, As, Ni, Cd, Cr, and Hg, classified as human mutagens and carcinogens by the International Agency for Research on Cancer (IARC) (Table 1).

2.2.3 PAHs

Fossil diesel contains PAHs, and incomplete combustion of diesel, pyrolysis of aromatic compounds, and unburned fuels and lubricants may produce PAHs (Wang *et al.*, 2021). Initially in the gas phase, they adhere to the particle phase via diffusion as the exhaust gas cools, making the particle-phase polycyclic aromatic hydrocarbons primarily derived from unburned diesel fuel.



The U.S. Environmental Protection Agency (U.S. EPA) has prioritized 21 PAHs for testing based on their potential harm. PAHs can be divided into Low-molecular-mass PAHs (LVMW-PAHs) with 2–3 rings, Medium-molecular-mass PAHs (MMW-PAHs) with 4 rings, and High-molecular-mass PAHs (HMW-PAHs) with 5–7 rings. PAHs can significantly impact the human body and may lead to various cancers such as breast, lung, colorectal, stomach, and liver cancer. They can cause DNA mutations and accelerate cancer growth rates (Cogliano *et al.*, 2011).

2.2.4 VOCs

Volatile organic compounds are key precursors for ozone and PM formation. Produced by the incomplete combustion of fuel. Volatile organic compounds (VOCs) emitted by diesel engines can be divided into six categories: alkanes, olefins, alkynes, aromatics (AHs), halogenated hydrocarbons (HHs) and oxygenated volatile organic compounds (OVOCs). Alkanes, including n-hexane, n-heptane and n-undecane, are mainly derived from the pyrolysis oxidation of unburned alkanes and multi-carbon chain molecules in fuels (Zhang *et al.*, 2022). Alkenes, which mainly include propylene, pentene and hexene, are formed by the thermal cracking and partial oxidation of high-carbon chain molecules under high temperature and hypoxia conditions. Alkynes such as propyl are unsaturated aliphatic hydrocarbons. AHs include monocyclic aromatic hydrocarbons (such as benzene, toluene) and polycyclic aromatic hydrocarbons, which are precursors to soot formation. Most AHs comes from unburned AHs in fuel fractions, while the rest is generated by dehydrogenation, addition and cyclization reactions after pyrolysis at high temperature (Dandajeh *et al.*, 2017). HHs is formed when hydrogen atoms in hydrocarbon molecules are replaced by halogen atoms, which is mainly generated by the substitution reaction of hydrocarbons at high temperatures. In OVOCs, carbonyl compounds (CBCs) are more harmful. CBCs are a class of organic compounds with carbon - oxygen double bonds. There are many kinds of them, and the common ones are saturated aliphatic aldehydes (formaldehyde, acetaldehyde, propionaldehyde, butyraldehyde, etc.), saturated aliphatic ketones (acetone, butyl ketone, etc.), 2-pentanone, etc.), unsaturated aliphatic carbonyl compounds (acrolein, crotonaldehyde, etc.), aromatic carbonyl compounds (benzaldehyde, o-p-tolualdehyde, etc.), aromatic ketones (such as acetophenone), and dialdehyde (such as glyoxal) have serious effects on human health (Chai *et al.*, 2013) (see Fig. 3 and Table 1).

2.2.5 POPs

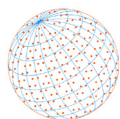
Persistent organic pollutants (POPs) formed by combustion include polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs), polybrominated dibenzo-*p*-dioxins and dibenzofurans (PBDD/Fs), polybrominated biphenyls (PBBs), and polybrominated diphenyl ethers (PBDEs). The formation of PCDD/Fs involves carbon formation and carbon oxidation stages (Huang and Buekens, 1995), with diesel engine exhaust containing PAHs and carbon particles potentially driving the formation of other toxic organic pollutants such as PBDD/Fs, PCBs, and PBDEs via precursors or de novo synthesis routes (Wang *et al.*, 2010).

2.3 Greener Fuels

It's evident from the preceding discussion that Nonroad machinery generates substantial emissions of unregulated pollutants, posing significant risks to both the atmospheric environment and human health. Therefore, it's imperative to control the emission of harmful pollutants, and one effective measure is the adoption of various green fuels such as biodiesel, oxygenated additive fuel, emulsified fuel, metal pellet additive fuel, hydrogen, and ammonia gas. The following section introduces common green fuels:

2.3.1 Biodiesel

Biodiesel can be produced through the transesterification of vegetable oils or animal fats with alcohol. Common biodiesel varieties include waste cooking oil (WCO), palm oil, soybean oil, rapeseed oil (Poulton, 1994), fish oil, and algae biodiesel, often used in marine engines, along with hydrogenated vegetable oil (HVO) frequently employed in Nonroad machinery. Compared to conventional fuels, biodiesel boasts a lower sulfur and aromatics content, a high cetane number, and a high oxygen content, making it burn more efficiently. Moreover, it is highly biodegradable



and renewable (Demirbas, 2009). Utilizing biodiesel as fuel can significantly reduce emissions of PM, HC, CO, etc. (Karavalakis *et al.*, 2009). However, its high viscosity can impair fuel atomization and, consequently, combustion efficiency. Additionally, biodiesel usage may elevate nitrogen oxide emissions (Karavalakis *et al.*, 2009). Typically, biodiesel is added as an additive to diesel fuel for combustion. It finds wide application in generators, ships, and Nonroad machinery, with its impact on unregulated pollutant emissions discussed in detail in subsequent sections.

2.3.2 Oxygenated fuel

Oxygen-containing additives, such as methanol, ethanol, butanol, acetone, acetone-butanol-ethanol (ABE), dimethyl ether (DME), diethyl ether (DEE), oxymethylene ether (OME), and various emulsified fuels, are commonly utilized as engine fuels. Notably, while these additives are seldom used as alternative fuels in marine engines due to technical feasibility and fuel economy considerations, they find wide application in generators and other Nonroad machinery. Compared to conventional fuels, alcohol fuels offer a higher octane-number and compression ratio, thus enabling greater thermal efficiency. Additionally, their lower carbon-hydrogen ratio compared to conventional fossil fuels facilitates reduced greenhouse gas emissions such as CO₂. However, their high boiling points may impact atomization during low-temperature combustion, potentially leading to increased PM and PN emissions. Consequently, alcohol fuel is typically employed as a fuel additive, mixed with conventional fuels. DME (Agarwal *et al.*, 2024), DEE (Anand and Mahalakshmi, 2007), OME (Zacherl *et al.*, 2022), ABE (Lin *et al.*, 2022) boast high cetane numbers, lower spontaneous combustion temperatures and boiling points, and high oxygen content, resulting in reduced engine ignition delay and enhanced combustion efficiency when used as fuel. Moreover, their lack of C-C bonds enables cleaner combustion and effectively reduces emissions of HC, CO, NO_x, and soot. The impact of these oxygen-containing additives on unregulated pollutant emissions in various Nonroad machinery is discussed in detail in subsequent sections.

2.3.3 Other green fuels

In addition, researchers have explored the addition of micrometer or nanoscale metal particles to conventional fossil fuels or biodiesel. Nano-fuel additives can enhance engine combustion performance while lowering pollutant emissions (Soudagar *et al.*, 2018). Both biodiesel and various additive fuels still contain carbon, leading to the production of greenhouse gases. In recent years, carbon-free fuels such as hydrogen and ammonia have garnered significant attention. Hydrogen boasts a substantially higher specific energy content (147 MJ kg⁻¹) compared to fossil fuels, more than three times that of many hydrocarbon fuels. As it lacks carbon atoms, burning hydrogen does not result in greenhouse gas emissions like CO and CO₂. However, its high volatility, low flash point, and susceptibility to explosions, coupled with stringent requirements for storage and transportation, present significant challenges (White *et al.*, 2006). Ammonia, containing 17.64 wt.% hydrogen, can serve as a carrier of hydrogen, offering more economical and safer storage and transportation options (Dimitriou and Javaid, 2020). In recent years, both hydrogen and ammonia have found wide applications in Nonroad machinery, particularly in ships. The effects of these fuels on unregulated pollutant emissions in various Nonroad machinery are discussed in detail in subsequent sections (Zincir, 2020; Mallouppas *et al.*, 2022).

3 UNREGULATED EMISSIONS FROM GENERATORS

3.1 Using Diesel and Biodiesel Blends

3.1.1 Soot, PAHs, and carbon

Incorporating biodiesel such as waste cooking oil (WCO) (Tsai *et al.*, 2011), waste edible oil (Tsai *et al.*, 2014a), soybean oil (Tsai *et al.*, 2010), palm oil methyl ester (PME) (Zhang and Balasubramanian, 2014b), rapeseed oil (Lu *et al.*, 2023) to diesel generators can effectively reduce emissions of soot, polycyclic aromatic hydrocarbons (PAHs), and total toxicity (see Table S4). However, the impact on PM-EC/OC varies, as detailed in Table S3 and Fig. 4. Specifically, PME, diethyl adipate (DEA), and dimethyl carbonate (DMC) reduce both EC and OC contents, which decrease with higher oxygenate fuel content (Zhang and Balasubramanian, 2015). In contrast, WCO and Soybean

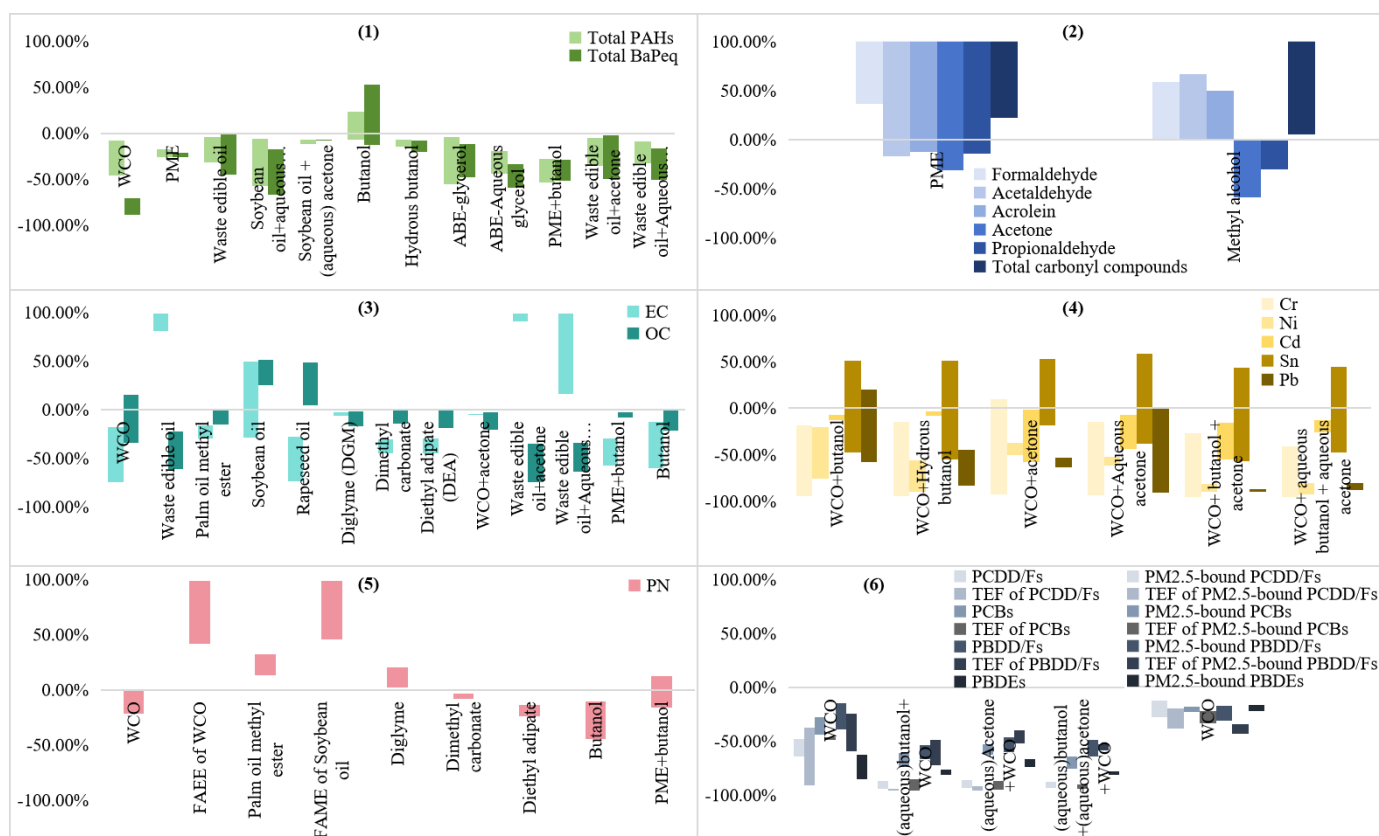
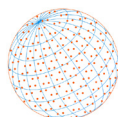


Fig. 4. Variation range of unregulated pollutant emissions when using various green fuels in generators compared to base fuels among (1) total PAHs and total BaP_{eq}; (2) main carbonyl compounds; (3) EC/OC; (4) toxic metal components; (5) PN; (6) POPs.

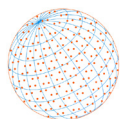
Oil significantly lower EC content, though OC is influenced by load and fuel type. Rapeseed Oil decreases EC while increasing OC, whereas Waste Edible Oil has an opposite effect on EC/OC. Thus, different biodiesel blends affect the carbon distribution in pollutants, but generally lower the total carbon content (Tsai *et al.*, 2014a).

Increasing the biodiesel content initially reduces pollutant emissions, peaking at B20 (Tsai *et al.*, 2011). However, excessive biodiesel (50%) raises pollutant concentrations (Tsai *et al.*, 2010). This is attributed to biodiesel's higher oxygen content, which, when added to petroleum diesel (e.g., B10 and B20), enhances autoignition and reduces ignition delay, improving combustion efficiency and suppressing PM formation. Moreover, biodiesel's low aromatic content dilutes aromatic compounds from fossil fuels, oxidizing PAH precursors (C₂H₂), leading to more complete combustion and degradation of H/MMW-PAHs into LMW-PAHs, thus lowering Total BaP_{eq} in diesel exhaust (Agarwal, 2007). However, excessive biodiesel addition can increase the fuel mixture's viscosity, affecting nozzle atomization and potentially leading to non-combustion or incomplete combustion of droplets in the combustion chamber, thereby increasing harmful substance formation (Tsai *et al.*, 2010).

3.1.2 Particle number concentration and size distribution

Biodiesel addition also affects particle number (PN) or particle size distribution (PSD), represented by the geometric mean diameter (GMD). With increased content of waste edible oil, diethyl adipate (DEA), and dimethyl carbonate (DMC), both PN and GMD decrease (Tsai *et al.*, 2014a; Zhang and Balasubramanian, 2015). Appropriate WCO levels reduce PN, with pure WCO showing the smallest peak diameter and total particle count (Betha and Balasubramanian, 2011). Different biodiesels have varying effects due to the types of alcohols used in production.

Adding methyl ester biodiesel like soybean oil fatty acid methyl ester (FAME) significantly reduces exhaust particles, with B20 showing the best reduction (56%), though further FAME



increases lessen the effect (Cosseron *et al.*, 2012). Methyl ester biodiesel lowers combustion temperature due to the oxygen in ester molecules, reducing non-volatile particles (soot emissions). Additionally, increasing biodiesel content reduces particles $< 0.1 \mu\text{m}$ while increasing those $0.1\text{--}1 \mu\text{m}$. Conversely, PME increases both PN and GMD (Zhang and Balasubramanian, 2014b, 2015). However, the use of ethyl biodiesel, such as fatty acid ethyl ester of waste cooking oil (FAEE), results in increased particulate matter emissions, which escalate with FAEE content (Cosseron *et al.*, 2012), as Table S3 and Fig. 4. Incomplete combustion of ethyl esters mainly produces carboxylic acids and alkenes in the initial stage. Subsequently, the combustion of ethyl oleate generates monounsaturated acids, which are further consumed by radicals ($\bullet\text{H}$, $\bullet\text{OH}$, $\text{HO}_2\bullet$, and $\bullet\text{CH}_3$ radicals) to produce intermediates. These intermediates form substances with fewer carbon atoms, such as olefins, CO_2 , unsaturated carboxylic acids, aldehydes, and ketones, promoting the formation of soot through subsequent reaction steps and leading to an increase in non-volatile particles. In summary, the use of methyl esters tends to promote the emission of smaller particles compared to ethyl ester biodiesel.

3.1.3 Metal component

Studies show that WCO-diesel blends dominate PM metal elements like Na, Mg, Al, K, Ca, Fe, and Zn ($> 90\%$ mass), with trace metals including Mn, Cu, Sr, and Pb (Lin *et al.*, 2017). WCO reduces metal emissions, but high biodiesel levels (B40) increase metal concentrations (see Fig. 4), possibly due to higher metal content in WCO than pure diesel. Biodiesel's high lubricity reduces friction in engine parts, lowering Fe emissions and improving lubrication reduces Cr emissions (Shah *et al.*, 2014).

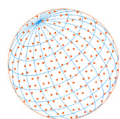
3.1.4 Carbonyl compound

Carbonyl compound emissions vary with biodiesel content. Lower biodiesel levels (B50 or $< \text{B50}$) reduce carbonyls (Bolin *et al.*, 2010), but pure biodiesel increases total emissions (Chai *et al.*, 2013). However, some studies report different findings, with PME (Liu *et al.*, 2009) and Soybean Oil FAME-diesel (Cosseron *et al.*, 2012) increasing carbonyls (Table S6 and Fig. 4). This phenomenon is attributed to the further reaction of monounsaturated acids generated from the combustion of ethyl oleate, leading to the formation of oxy-carbonyl compounds. Besides unsaturated carboxylic acids, various compounds including ethylene, propylene, 1-butene, formaldehyde, acetaldehyde, acetone, propionaldehyde, and unsaturated methyl esters are produced.

Biodiesel exhaust contains formaldehyde, acetaldehyde, acrolein, and acetone as major carbonyls, with others (e.g., propionaldehyde, crotonaldehyde, benzaldehyde) comprising 10–25% of total carbonyls (Chai *et al.*, 2013). As biodiesel content increases, the proportion of formaldehyde, acetaldehyde, and acrolein decreases, possibly due to increased production of carbonyl compounds during biodiesel combustion. Moreover, the emission rate of unsaturated aliphatic aldehydes like acrolein and crotonal in biodiesel blends significantly surpasses that of corresponding saturated aliphatic aldehydes such as propanal and butyraldehyde. This observation suggests that the oxidation of hydrocarbons to form carbonyl compounds may preferentially generate unsaturated carbonyl groups at the α -carbon bonds of carbon-carbon double bonds.

3.1.5 POPs

Using WCO-diesel reduces mass and toxicity concentrations of persistent organic pollutants (POPs) in exhaust (Tsai *et al.*, 2016) (see Table S7 and Fig. 4). This reduction is primarily due to the addition of biodiesel, which increases the oxygen content of the blended fuel and improves the combustion efficiency of the engine, thereby decreasing pollutant emissions. Moreover, biodiesel may contain fewer precursors of PCDD/Fs, such as aromatic compounds and C_2 radicals, which inhibit the de novo synthesis of persistent organic pollutants (Weber and Kuch, 2003). However, excessive amounts of biodiesel can lead to increased emissions of persistent organic pollutants due to its high viscosity, which affects fuel atomization and results in incomplete combustion. Additionally, blending biodiesel with diesel can significantly reduce emissions of POPs bound to $\text{PM}_{2.5}$. Research indicates that the addition of 20% WCO yields the most significant reduction in POPs emissions, with the reduction effect diminishing as the WCO content increases.



(Tsai *et al.*, 2018a). The mass emissions of these POPs were in the order PBDEs > PBDD/Fs > PCBs > PCDD/Fs, while the corresponding toxicity equivalent concentrations followed the order PCDD/Fs > PBDD/Fs > PCBs (Tsai *et al.*, 2018a).

In conclusion, biodiesel blends impact pollutant composition and emissions differently, with methyl esters generally promoting smaller particle emissions and reduced soot and PAHs, while excessive biodiesel increases viscosity, reducing combustion efficiency and raising certain pollutants. Critical evaluation and comprehensive comparison of various studies highlight biodiesel's potential and limitations, guiding its optimal use in diesel engines.

3.2 Utilizing Oxygenated Additives in Diesel/Biodiesel

3.2.1 Soot and PAHs

The addition of oxygen-containing additives, such as methanol (Datta and Mandal, 2016), ethanol (Lee *et al.*, 2011), butanol (Lin *et al.*, 2012; Zhang and Balasubramanian, 2014b; Tsai *et al.*, 2017), acetone (Lin *et al.*, 2010; Tsai *et al.*, 2014a, 2014b, 2017), ABE (Lin *et al.*, 2022) and ether fuels (Zhang and Balasubramanian, 2015), to diesel or biodiesel has been demonstrated to significantly reduce emissions of soot, total PAHs, and overall toxicity in diesel exhaust (Fig. 4, Table S4 and Table S5). This reduction generally increases with the higher content of biodiesel and oxygenated additives, due to the enhanced oxygen content in the fuel mixture. This enhancement improves combustion, lowering the concentration of PAH precursors (e.g., C₂H₂) and thus reducing PAH formation and emissions (Tsai *et al.*, 2014b). However, some studies have reported an increase in total PAHs and BaP_{eq} in particulate matter with higher butanol content (Zhang and Balasubramanian, 2014a), attributed to higher emissions of unburned hydrocarbons from butanol-diesel blends. Notably, when butanol content is below 10%, there is no significant change in total particulate PAH concentrations and corresponding BaP_{eq}. This indicates that the content of oxygenated additives critically influences pollutant emissions.

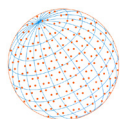
For acetone (containing water), the addition should not exceed 3 wt% (Lin *et al.*, 2010; Tsai *et al.*, 2014b) to avoid fuel instability and phase separation. Adding 1%–3% acetone can reduce generator exhaust emissions. Ethanol, due to its polar nature, causes phase separation in diesel when exceeding 5 vol% (Lee *et al.*, 2011). For butanol and water, the optimal additions are 0.5 wt.% water and 10% butanol to effectively reduce PM (Lin *et al.*, 2012; Zhang and Balasubramanian, 2014b).

Research indicates that emulsified fuels offer superior pollutant reduction. The emission reduction effect of each fuel on pollutants is ranked as follows: biodiesel - (water-containing) oxygen-containing additives > biodiesel - oxygen-containing additives > biodiesel (Lin *et al.*, 2012; Tsai *et al.*, 2014a; Lin *et al.*, 2022). The emulsification process enhances fuel combustion by increasing the contact surface area between fuel droplets and air, facilitated by micro-explosions and secondary atomization caused by water vaporization (Ithnin *et al.*, 2014). Additionally, water-containing fuels reduce PM formation through the pyrolysis of secondary atomized oil droplets during high-temperature combustion, leading to the formation of ionized carbon particles. These carbon particles may react with water vapor molecules, facilitating a water-gas transfer reaction as following Reactions (1) and (2), thereby improving combustion efficiency and reducing emissions of soot and other harmful pollutants (Tsai *et al.*, 2014a).



3.2.2 Carbon, PN, and PSD

The impact of different oxygenated additives on EC and OC varies. Butanol reduces EC content in PM while keeping OC content nearly unchanged (Zhang and Balasubramanian, 2014b, 2014a), though some studies report reductions in both EC and OC with butanol blends (Zhang and Balasubramanian, 2015). This reduction is attributed to improved combustion efficiency due to higher oxygen content and better fuel-air mixing (Zhang and Balasubramanian, 2014a). Adding



glycol dimethyl ether (DGM) decreases both EC and OC, while adding (aqueous) acetone to waste-edible-oil reduces OC but increases EC content, though overall total carbon (TC) is reduced (Tsai *et al.*, 2014a). Moreover, the emission concentrations of EC and OC in aqueous acetone-mixed fuel are lower compared to those in pure acetone-mixed fuel, and within a certain range (biodiesel content < 5 vol%), they decrease with an increase in aqueous acetone percentage. Nevertheless, when biodiesel percentage surpasses 5 vol%, the impact of water on EC and OC becomes insignificant. The introduction of butanol in the fuel mixture leads to particle size reduction, decreased PN, and predominantly lower emission of larger particles. Upon the addition of butanol to diesel fuel, the total number concentration of volatile and non-volatile particles notably decreases, consequently reducing particle condensation and agglomeration, resulting in a decrease in GMD (Zhang and Balasubramanian, 2014a; Saxena and Maurya, 2016). However, when ether fuel glycol dimethyl ether (DGM) is utilized, PN increases (Zhang and Balasubramanian, 2015). Specific emission reduction effects are presented in Fig. 4 and Table S3.

3.2.3 Metal components

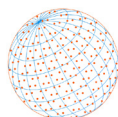
Adding butanol, acetone, and their aqueous forms to biodiesel effectively reduces metal emissions from generators, with exhaust metal compositions similar to those from WCO, including Na, Mg, Al, K, Ca, Fe, and Zn (Lin *et al.*, 2017). Compared to WCO-diesel blends, adding 30% aqueous butanol to WCO is more effective in reducing metal content (−35.44% to −16.57%) than adding butanol alone (−32.97% to −20.53%), with aqueous acetone also showing significant reductions. The most effective reduction is achieved with combined aqueous butanol and acetone (−53.05% to −36.53%) (Tsai *et al.*, 2018b). Notably, while WCO-butanol fuel increases metal content in PM, WCO-hydrated butanol and WCO-acetone/hydrated acetone blends lead to decreases (Fig. 4 and Table S6).

3.2.4 CBCs

Presently, limited research exists on the impact of oxygenated additive fuels on volatile organic compounds (VOCs) or carbonyl compound emissions. Blending methanol increases carbonyl compound (CBC) emissions in tail gas, particularly formaldehyde (Chao *et al.*, 2000). Fan *et al.* (2018) indicated that the methanol addition increases formaldehyde, acetaldehyde, acrolein, acetone, and crotonaldehyde emissions at low load (Table S7). This is attributed to incomplete combustion of carbonyl substances from hydrocarbon fuels (Agarwal *et al.*, 2016). The relatively low temperature not only promotes incomplete combustion of the fuel but also weakens further oxidation of the carbonyl species. Hence, under low-load conditions, methanol-diesel yields more formaldehyde and acetaldehyde compared to pure diesel. Conversely, under high load, the formaldehyde and acetaldehyde emissions from methanol-mixed fuel are similar to those of pure diesel, while propionaldehyde emission decreases. This is due to propionaldehyde production from straight-chain hydrocarbons such as hexane, 1-hexane, and octane during diesel combustion (Zervas *et al.*, 2006). The addition of methanol reduces the content of straight-chain hydrocarbons in the fuel, while the oxygen in methanol molecules likely promotes the oxidation of propionaldehyde precursors. Consequently, methanol-diesel exhibits lower propionaldehyde emissions than pure diesel.

3.2.5 POPs

Tsai *et al.* (2017) supplemented WCO fuel with (aqueous) butanol, (aqueous) acetone, and their mixtures based on prior research and observed significant reductions in PCDD/Fs, PCBs, PBDD/Fs, and PBDEs content in exhaust gas. Among these, the addition of aqueous butanol yielded the most favorable results, followed by (aqueous) acetone + (aqueous) butanol, and (aqueous) acetone had the least impact. However, both methods were superior to biodiesel alone (Fig. 4 and Table S8). Additionally, fuels containing water-based oxygenated additives demonstrated enhanced effectiveness, attributed to micro-explosions enhancing secondary atomization of fuel spray and combustion improvement (Chang *et al.*, 2013). In summary, the addition of butanol and acetone (both pure and aqueous) to W20 can further reduce POP emissions, demonstrating the effectiveness of oxygenated additives in improving fuel combustion and reducing pollutant emissions.



4 UNREGULATED EMISSIONS FROM SHIPS

4.1 Using Diesel/Biodiesel Blends

4.1.1 PAHs and carbon

Using WCO-marine gas oil blends in marine engines has been shown to reduce the concentrations of individual PAHs and total PAHs in exhaust emissions (see Fig. 5 and Table S9) (Su *et al.*, 2019; Yusuf *et al.*, 2022). This reduction diminishes with an increase in WCO content. Mixed fuels such as fish oil (Ushakov, 2012), jatropha oil methyl ester (JOME) and diethylene glycol dimethyl ether (DGM) (Nabi and Hustad, 2010) can lower soot emissions. However, the impact of soybean-based biodiesel varies with engine load, sometimes increasing total PAHs under specific conditions (Jayaram *et al.*, 2011).

Different biodiesel blends have varying effects on EC and OC in particulate matter (PM) (see Fig. 5 and Table S10). For instance, WCO usage slightly reduces OC below 50% load (Yang *et al.*, 2022) but increases EC. The sulfate and inorganic elements initially increase and then decrease. However, studies incorporating HEPT (ship exhaust after-treatment devices) have found that WCO use reduces both EC and OC emissions, which decrease further with higher WCO content (Yusuf *et al.*, 2022). Hydrotreated algae fuel-diesel blends significantly reduce PM_{2.5}, EC, and OC emissions. At low loads, EC decreases modestly while OC decreases significantly (25–28%). At medium to high loads, EC decreases substantially (30–34%), but OC changes are minimal or even increase (Khan *et al.*, 2012). Cane biodiesel notably reduces EC and OC without significantly affecting PM_{2.5} (Gysel *et al.*, 2014). Soybean biodiesel reduces PM_{2.5} and EC, with reductions correlating with biodiesel content. However, OC is influenced by both engine load and biodiesel content, increasing at 25% load and decreasing at the highest load (Jayaram *et al.*, 2011), thereby increasing the OC/EC ratio.

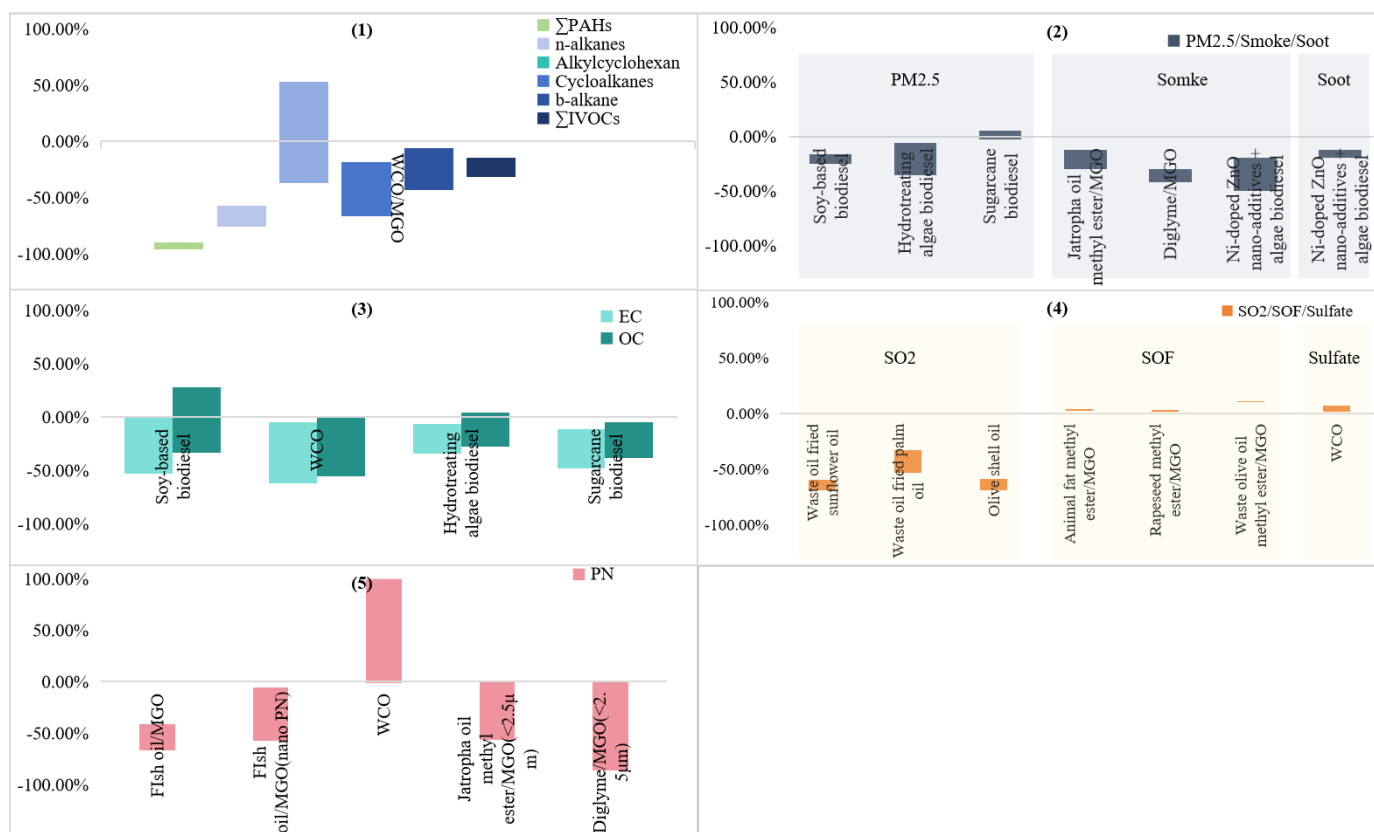
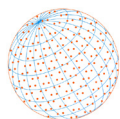


Fig. 5. Variation range of unregulated pollutant emissions when using various green fuels in ships compared to base fuels among (1) PAHs and VOCs; (2) PM_{2.5}/Smoke/Soot; (3) EC/OC; (4) SO₂/SOF/Sulfate; (5) PN.



4.1.2 PN and GMD

The addition of JOME, DGM, fish oil (Ushakov, 2012), and soybean-based biodiesel (Jayaram *et al.*, 2011) has been shown to reduce both the volume mean diameter and total number concentration of particles (Fig. 5 and Table S10), with reductions correlating with increased biodiesel mixture ratios (Jayaram *et al.*, 2011). This phenomenon can be attributed to the high oxygen content of the blended fuel, which enhances the diffusion combustion phase of the combustion process. Additionally, oxygen-containing additives facilitate complete fuel combustion, leading to a more effective reduction in pollutant emissions, with DGM exhibiting superior efficacy compared to JOME (Nabi and Hustad, 2010). Conversely, the utilization of waste cooking oil (WCO) has been associated with an increase in particulate matter, which escalates with higher WCO content (Yusuf *et al.*, 2022). Initially, the use of WCO enlarges the particle size range before subsequently decreasing it. However, as WCO content increases, the number of particles smaller than 100 nm rises while the overall particle size diminishes (Yang *et al.*, 2022; Yusuf *et al.*, 2022).

4.1.3 VOCs

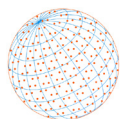
Carbonyl compounds are generated when palm oil-diesel blends are utilized and their concentration increases with the blend's content, except for B50 (see Fig. 5 and Table S9) (Nguyen and Le, 2019). This occurrence is attributed to the combustion of ester compounds present in biodiesel fuel. The combustion process of biodiesel involves breaking down the carbon bond of long carbon chain ester molecules into shorter carbon chains. In this process of ester molecular chain fragmentation ($R1-C=O-O-R2$), the C-O bond is particularly susceptible to breaking, facilitating the formation of carbonyl compounds ($R1-C=O-R2$). However, the emissions of carbonyl compounds from B50 declined. This is attributed to the high cetane number and elevated oxygen content of biodiesel fuel. A fuel boasting a high cetane number reduces the ignition delay of the fuel in the engine, intensifying the combustion process and resulting in fewer hydrocarbon emissions. Moreover, the high oxygen content facilitates more complete fuel combustion. Nonetheless, as the biodiesel content increases, carbonyl compound emissions continue to rise. This can be attributed to the higher viscosity of palm oil-diesel blends compared to diesel. Consequently, when the palm oil content is higher, the blend's viscosity increases, impacting the engine's fuel injection process and leading to incomplete fuel combustion, thereby elevating carbonyl compound emissions (Nguyen and Le, 2019). However, when soybean-based biodiesel was utilized, carbonyl compounds and their total emissions exhibited no significant change with increasing biodiesel blend ratios (Jayaram *et al.*, 2011).

4.1.4 POPs

There is limited research on emissions of POPs from ships. Swedish researchers conducted an investigation into the emissions of PCDD/Fs, hexachlorobenzene (HCB), and polychlorinated biphenyls (PCB) from three ships. They discovered that emissions of PCDD/F from ships were minimal and contributed relatively little, accounting for only 1–2% of the total emissions in Sweden in 2002 (Cooper, 2005).

4.1.5 SOF

As the addition of methyl ester increases, so does the emission of the SOF in PM. This uptick occurs because biodiesel's poor volatility, with its higher boiling point, leads to incomplete combustion of the fuel. Research by Karavalakis *et al.* (2008) examined three different types of methyl esters as fuels, revealing that SOF emissions followed this order: waste olive oil methyl ester (OME) > animal fat methyl ester (AFME) > rapeseed methyl ester (RME). On the flip side, using biodiesel tends to reduce SO₂ emissions in exhaust gas. This is because the emission of sulfur dioxide in exhaust gas closely correlates with the sulfur content of the fuel. Biodiesel, with its minimal sulfur content, effectively lowers the overall sulfur content when blended with other fuels, thereby diminishing SO₂ emissions. However, the extent of this reduction diminishes as the proportion of biodiesel in the blend increases. Nikolić *et al.* (2016) compared three types of methyl ester biodiesel and found that the SO₂ reduction effect ranked as follows: diesel oil and waste oil-fried sunflower oil > diesel oil and olive oil shell oil > diesel oil and waste oil-fried palm



oil. Nevertheless, it's important to note that sulfur in diesel contributes to lubricating the engine's moving parts, so reducing sulfur content may impact lubricity.

4.2 Using Other Green Fuels

When it comes to marine engines, besides incorporating biodiesel, various other green fuel alternatives exist, including metallic additive fuels, hydrogen, and ammonia, among others. Let's delve into the specific emission reduction effects of each green fuel on different pollutants:

4.2.1 Ni-doped ZnO nano additives

These additives exhibit effective reduction in PM, soot, and smoke emissions in algae biodiesel. The emission reduction tends to increase with higher additive content. Smoke typically arises in regions of the combustion chamber with insufficient air/oxygen. The mixed fuel, with its higher oxygen content, facilitates complete combustion (Fig. 5 and Table S10) (Karthikeyan and Dharma Prabhakaran, 2018).

4.2.2 Hydrogen and ammonia

These green fuels also demonstrate significant reductions in smoke and soot emissions in exhaust gas (Reiter and Kong, 2011; Dimitriou *et al.*, 2018; Jamrozik *et al.*, 2020). However, it's worth noting that the use of ammonia in internal combustion aboard ships may lead to increased emission of N₂O (Xu *et al.*, 2023). Nonetheless, research on the emission of other unregulated pollutants remains scarce.

5 UNREGULATED EMISSIONS FROM NONROAD ENGINES

5.1 Using Diesel/Biodiesel Blends

5.1.1 PAHs and carbon

When utilizing microalgae biodiesel (Mwangi *et al.*, 2015), palm oil (Phoungthong *et al.*, 2013), rapeseed oil (Vojtišek and Fenkl, 2010), HVO (McCaffery *et al.*, 2022) as fuel for Nonroad diesel engines, there is a notable reduction in total PAHs and Bapeq emissions from exhaust gas (Fig. 6, Table S12, and Table S13). However, when using palm oil-diesel blends, low molecular weight PAHs in the exhaust increase, while high molecular weight PAHs decrease. Benzene emissions also rise due to the high unsaturated compound content in RME. Aromatic compounds like benzene and alkylbenzenes depend on the unsaturation level of the fatty compounds, with highly unsaturated compounds forming more aromatics and thus increasing benzene emissions as RME content rises (Krahl *et al.*, 2002). Furthermore, HVO demonstrates a significant reduction in EC emissions, although its effect on OC reduction is less pronounced. It is noteworthy that excessive HVO content may diminish its effectiveness in reducing EC and OC emissions, as the reduction effect weakens with increased HVO content (Fontaras *et al.*, 2009).

5.1.2 PN and N₂O

Compared to diesel fuel oil, the utilization of rapeseed oil (Pietikäinen *et al.*, 2015) and RME (Krahl *et al.*, 2002) as fuel leads to an increase in the number of particles in exhaust gas (Fig. 6 and Table S11). When employing rapeseed oil, PN experiences a 10–100 fold increase, particularly in the ultra-fine particle size range (< 100 nm). Conversely, wood residue-based hydrotreated vegetable oil (Niemi *et al.*, 2016, 2019), crude tall oil-based renewable diesel fuel (Spoof-Tuomi *et al.*, 2021), and HVO (Pirjola *et al.*, 2017; McCaffery *et al.*, 2022) can reduce particle numbers. However, under certain conditions, pure HVO can increase total particle numbers. This may result from the high oxygen content in biodiesel converting carbon particles from ultrafine to nano-sized, and the high viscosity and low volatility of the fuel leading to poor atomization and uneven mixing with air, thereby increasing soluble organic fraction emissions and nucleation-mode particle formation (Sun *et al.*, 2019). Moreover, the adoption of wood residue-based hydrotreated vegetable oil and crude tall oil-based renewable diesel fuel can reduce the N₂O content in exhaust gas.

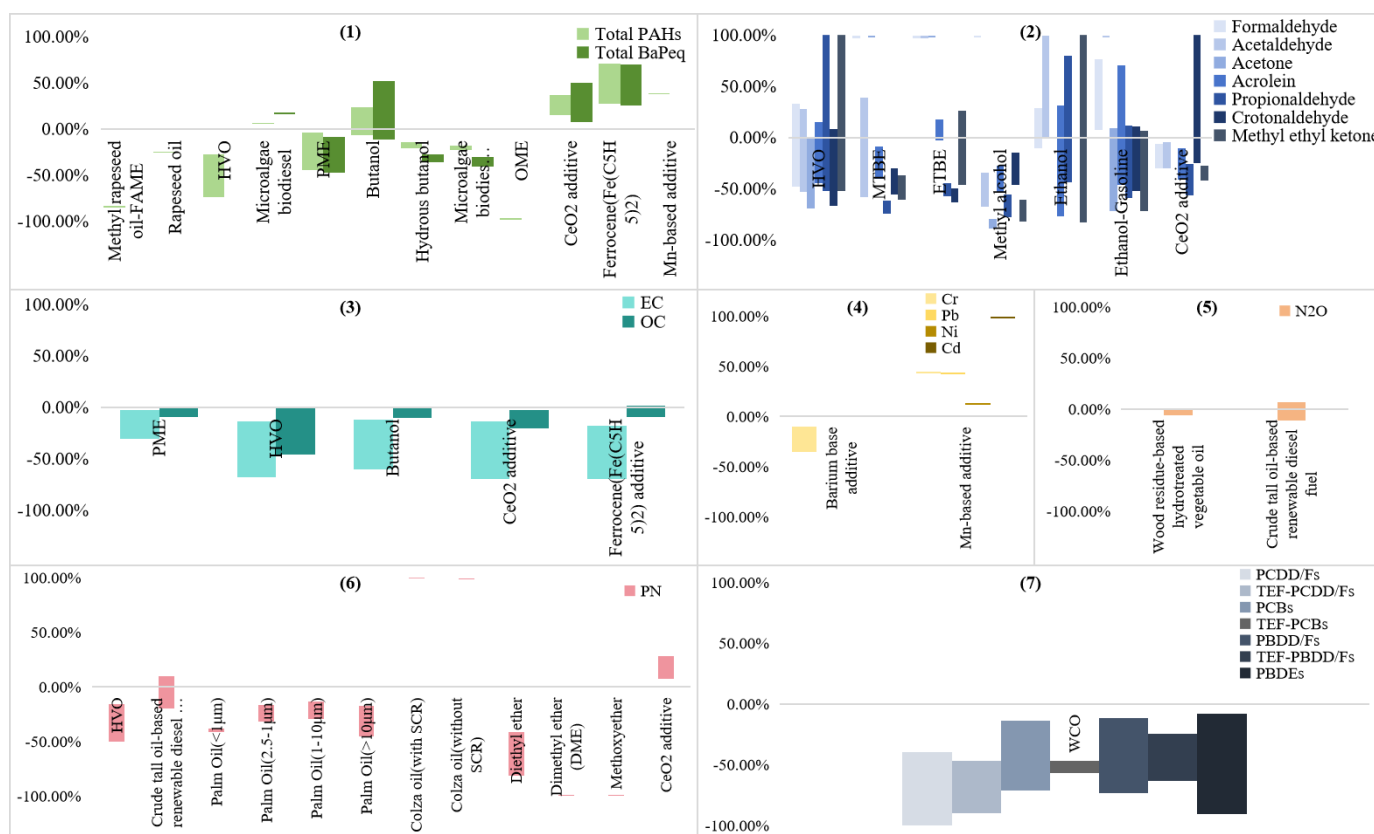
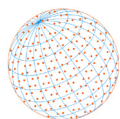


Fig. 6. Unregulated pollutant emissions when using various green fuels in remaining Nonroad engines compared to base fuels among (1) total PAHs and total BaP_{eq}; (2) main carbonyl compounds; (3) EC/OC; (4) toxic metal components; (5) N₂O; (6) PN; (7) POPs.

5.1.3 Metal components

HVO or HVO/biodiesel blends have varied effects on different metal emissions. Overall, compared to pure diesel, these blends generally reduce metal or element emissions associated with PM (McCaffery *et al.*, 2022).

5.1.4 CBCs

The addition of HVO to biodiesel significantly reduces the emissions of carbonyl compounds compared to pure diesel (Fig. 6 and Table S14) (McCaffery *et al.*, 2022). The higher the HVO content, the more pronounced the reduction in carbonyl compounds. However, using pure HVO as fuel can increase carbonyl emissions.

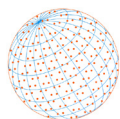
5.1.5 POPs

Incorporating an appropriate amount of waste cooking oil into diesel can effectively reduce the emission of POPs, with the reduction increasing as the amount of biodiesel added increases, with the optimal amount being 40% (Fig. 6 and Table S15) (Chen *et al.*, 2017). This reduction is attributed to two factors: the low content of POP precursors (e.g., aromatic compounds) in biodiesel, which inhibits POP formation (Chang *et al.*, 2014), and the high oxygen content (typically 10–12%) of biodiesel promoting more complete combustion. However, biodiesel usage in diesel engines is limited by its physical properties, such as viscosity, which affects fuel atomization quality and combustion efficiency. Therefore, excessive biodiesel content can negatively impact pollutant reduction, sometimes even increasing emissions.

5.2 Using Oxygenated Additives in Diesel/Biodiesel

5.2.1 PAHs, carbon, and PN

The addition of oxygen-containing additives such as ethanol (Merritt *et al.*, 2005), aqueous



butanol (Mwangi *et al.*, 2015), OME (Zacherl *et al.*, 2022) and DEE (Agarwal *et al.*, 2022) to diesel or biodiesel can effectively reduce the total PAHs and BaPeq emissions in exhaust gas (Fig. 6, Table S12, and S13), decreasing with the increase of oxygen-containing additive content. Ethanol addition increases the proportion of heavy PAHs in pollutant particles while decreasing the proportion of light PAHs (Merritt *et al.*, 2005). Moreover, emulsified fuels can further decrease pollutant emissions (Mwangi *et al.*, 2015), as discussed in detail in Section 3.2 regarding the reasons behind emission reduction with emulsified fuels.

Furthermore, certain amounts of butanol (Zhang and Balasubramanian, 2014a), DME (Agarwal *et al.*, 2024), DEE (Agarwal *et al.*, 2022) and OME (Zacherl *et al.*, 2022) can significantly reduce the total number concentration of volatile and non-volatile particles (Table S11), leading to decreased condensation and agglomeration of particles, resulting in a lower GMD (Zhang and Balasubramanian, 2014a). Among these, DME has been shown to reduce the total particle number (TPN), nanoparticle number (NP), nucleation mode particle (NMP), and accumulation mode particle (AMP) by over 99% (with slight variations under certain load conditions compared to diesel). However, the effect on count median diameter (CMD) is influenced by load and speed, with significant reductions at low speeds but varying effects at high speeds (Agarwal *et al.*, 2024). DEE reduces the total particle number, though its impact on particle number concentration (PN) is less significant (Agarwal *et al.*, 2022).

5.2.2 VOCs

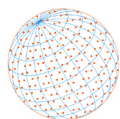
The impact of adding various oxygen-containing additives to diesel oil on VOCs emissions varies. The addition of DEE, methanol (Tian *et al.*, 2018) and ethanol (Merritt *et al.*, 2005) tends to increase VOCs emissions in exhaust gas (Fig. 6 and Table S14). However, when OME is added, the total VOC emission in the tail gas is significantly reduced (−69.89% to −52.78%).

DEE addition can lead to increased emissions of SO₂, formic acid (HCOOH), formaldehyde (HCHO), n-pentane, n-octane, and isobutene in the tail gas, with emissions increasing with the rise in DEE content (Agarwal *et al.*, 2022). DEE is typically produced by dehydrating ethanol in the presence of strong acids such as sulfuric acid, which may introduce sulfur contamination and increase SO₂ emissions. The high oxygen content of DEE promotes the formation of formaldehyde from the partial oxidation of fuel and lubricating oil. Increased alkane emissions result from the high oxygen content of DEE facilitating the breakdown of long-chain hydrocarbons into shorter ones (Agarwal *et al.*, 2022). Conversely, OME reduces VOCs emissions due to its absence of C-C bonds, forming oxygen-containing substances like formaldehyde and methyl ester during combustion, rather than soot precursors like acetylene (Zacherl *et al.*, 2022).

For non-road engines using gasoline as a base fuel, such as lawnmowers and chainsaws, the impact of adding oxygenated additives on VOC emissions has also been studied. Methanol (Ni *et al.*, 2014), ethanol (Magnusson *et al.*, 2002) and certain ether fuels like methyl tert-butyl ether (MTBE) and ethyl tert-butyl ether (ETBE) (Magnusson and Nilsson, 2011) added to gasoline significantly increase the emission of formaldehyde and acetaldehyde in the tail gas (Table S14). However, ethanol at concentrations below 15% has minimal impact on VOC emissions but significantly reduces emissions of compounds such as acetone, methyl ethyl ketone, crotonaldehyde, butyraldehyde, and propionaldehyde when its concentration increases (Magnusson *et al.*, 2002). The emission levels of formaldehyde for each oxygen-containing fuel are ranked as methanol > MTBE > ETBE > ethanol. As for acetaldehyde, the ranking is ethanol > ETBE >> MTBE ≈ methanol. And the total carbonyl emission levels are ethanol ≈ ETBE > Methanol > MTBE (Magnusson and Nilsson, 2011).

5.3 Using Other Green Fuels

Metal additive fuels are less commonly utilized in Nonroad machinery. However, given that most Nonroad machinery employs diesel engines, it's reasonable to assume that road diesel engines and Nonroad diesel engines experience similar emission reduction effects on pollutants when using metal additive fuels. Therefore, this study explores the effects of cerium (Ce) (Jung *et al.*, 2005), cerium oxide (CeO₂) (Zhang *et al.*, 2013; Zhang and Balasubramanian, 2017), ferrocene (Liu *et al.*, 2020), manganese (Mn)-based (Yang *et al.*, 1998) and barium (Ba)-based additive fuels (Zhang *et al.*, 2022) on unregulated pollutant emissions in road engines.



5.3.1 PAHs and carbon

The use of CeO₂, ferrocene, and Mn-based additive fuels tends to increase PAHs emissions from exhaust gas (Fig. 6, Tables S11, S12, and S13). However, CeO₂, ferrocene, and barium-based fuel additive fuels can enhance the complete combustion of fuel, reducing the mass concentration of OC and EC. Nevertheless, their impact on the percentage of EC and OC in total carbon species varies (Tables S10 and S11). Ba-based fuel additives heighten the oxygen concentration in the cylinder, facilitating the oxidation of dry soot and reducing the percentage of EC in particulate matter (PM). However, the Ba additive marginally reduces the exhaust temperature (Draper *et al.*, 1988). With soot reduction and decreased exhaust temperature, more unburned fuel condenses and binds to the resulting soot's surface, eventually leading to an increase in the mass fraction of OC in PM (Zhang *et al.*, 2022).

5.3.2 VOCs

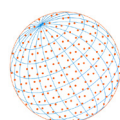
CeO₂ additive fuel tends to reduce CBCs emissions (Zhang *et al.*, 2013). However, Ba-additive fuels exhibit different effects on diesel engine emissions of alkanes, olefins, alkynes, aromatics (AHs), halogenated hydrocarbons (HHs), and oxygenated VOCs (Table S14). The use of Ba-AD reduces the concentrations of alkanes, olefins, and oxygenated VOCs while increasing the emitted alkynes concentration. This is attributed to the promotion by Ba ions in the additive of fuel decomposition into small molecular free radicals, intensifying the separation of hydrogen atoms in the reaction process. Consequently, a substantial number of hydrogen atoms inhibit the hydrogen-abstraction-C₂H₂-addition (HACA) reaction (Chu *et al.*, 2019), leading to increased acetylene pollutant emissions. Ba-AD also decreases AHs concentration due to the combustion promotion of the additives, reducing AHs emissions from unburned fuels. Moreover, the additives' inhibition of fuel molecule pyrolysis and small molecule dehydrogenation and cyclization contribute to the AHs concentration reduction. Additionally, barium-based fuel additives increase metal ion concentration in the cylinder, with halogens likely reacting with metals to form inorganic salts. Furthermore, HHs can react with metals to form organometallic compounds, thereby reducing HHs emission concentration.

6 OTHER METHODS OF PREVENTION AND CONTROL OF POLLUTANTS

In addition to cleaner fuels, various control methods are currently employed to reduce pollutant emissions from Nonroad mobile machinery. These methods encompass both in-machine purification technologies and external purification methods.

In-machine purification technologies involve optimizing the structural parameters of the combustion chamber, utilizing high-pressure injection, exhaust gas recirculation, delaying fuel injection, or improving fuel injection regularity (Feng *et al.*, 2022). These approaches aim to enhance combustion efficiency and reduce engine pollutant emissions. External purification methods primarily involve the use of aftertreatment devices. Common aftertreatment devices for marine engines include the HePT device, electrostatic precipitator (ESP), fabric filter (FF), wet electrostatic washing (WES), and others. The HePT device typically comprises a diesel exhaust fluid (DEF), diesel particulate filter (DPF), and selective catalytic reduction (SCR) equipped with a rapid exhaust pre-heater. External purification methods primarily involve the use of aftertreatment devices. Common aftertreatment devices for marine engines include the HePT device, ESP, FF, WES, and others. The HePT device typically comprises a DEF, DPF, and SCR equipped with a rapid exhaust pre-heater (Feng *et al.*, 2022).

In generators, four-unit converters such as the NO_x Storage Reduction catalyst (NSR) and Diesel Particulate NO_x Reduction catalyst (DPNR) are commonly employed as post-processing units. Research indicates that installing a four-element catalyst behind the generator can achieve a PN reduction rate exceeding 95% (Cosseron *et al.*, 2012). For Nonroad diesel engines, exhaust after-treatment technologies like the diesel oxidation catalyst (DOC), DPF, and SCR are widely utilized. These technologies significantly reduce emissions of CO, HC, NO_x, PM, PN, and N₂O (Pietikäinen *et al.*, 2015; Jung *et al.*, 2019; Hu *et al.*, 2021).



7 CONCLUSION

Non-road engines produce unregulated emissions that pose significant risks to human health and the atmospheric environment. Consequently, numerous scholars have conducted research in this domain. This study primarily analyzes the impact of different green fuels on various unregulated pollutants emitted by generators, marine engines, and other non-road engines. The results are summarized in Table 2, leading to the following conclusions:

- (1) Biodiesel: The use of biodiesel in non-road engines reduces the emissions of total PAHs/BaP_{eq}, EC/OC, and POPs, with the optimal blend ratio being B20. However, it also increases particle number (PN) emissions. The effect on carbonyl compounds (CBCs) is influenced by both the fuel type and engine load, potentially causing a significant rise in certain pollutants such as acetaldehyde and crotonaldehyde. Additionally, the use of biodiesel in marine engines increases SOF/SO₂/sulfate emissions. Biodiesel's high oxygen content promotes more complete combustion, thereby contributing to lower POP emissions.
- (2) Oxygenated Fuel Additives: The overall use of oxygenated fuel additives generally reduces the

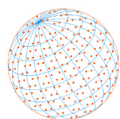
Table 2. Influences of different fuels on different pollutants in Nonroad machinery.

Fuels	EC	OC	N ₂ O	Soot	PN/PSD	Metals	VOCs	PAHs	BaP _{eq}	POPs
LNG	○	○	○	▲	▲	○	○	▲	▲	○
GTL	○	○	○	○	○	○	○	▲	▲	○
WCO	○	○	○	○	○	▲	○	▲	▲	▲
Waste edible oil	▼	▲	○	▲	▲	○	○	▲	▲	○
Soybean oil	▲	○	○	▲	▲	○	○	▲	▲	○
Palm oil	▲	▲	○	▲	▼	○	▼	▲	▲	○
Colza oil	▲	▼	○	▲	▼	○	○	▲	▲	○
Sugarcane biodiesel	▲	▲	○	○	○	○	○	○	○	○
DEA	▲	▲	○	○	▲	○	○	○	○	○
DMC	▲	▲	○	○	▲	○	○	○	○	○
Fish oil	○	○	○	▲	▲	○	○	▲	▲	○
JOME	○	○	○	▲	▲	○	○	▲	▲	○
Hydrotreating algae fuel	▲	▲	▼	○	○	○	○	○	○	○
Microalgae biodiesel	○	○	○	○	○	○	○	▲	▲	○
HVO	▲	▲	▼		○	▲	▲	▲	▲	○
Methanol	○	○	○	▲	○	○	▼	▲	▲	○
(Aqueous) ethanol	○	○	○	▲	○	○	▼	▲	▲	○
(Aqueous) butanol	▲	▲	○	▲	▲	○	○	▲	▲	▲
(Aqueous) acetone	○	▲	○	▲	○	▲	○	▲	▲	▲
ABE	○	○	○	▲	○	○	○	▲	▲	○
OME	○	○	○	▲	▲	○	○	▲	▲	○
DEE	○	○	○	▲	▲	○	▼	▲	▲	○
DME	○	○	○	▲	▲	○	○	▲	▲	○
DGM	▲	▲	○	▲	○	○	○	▲	▲	○
MTBE	○	○	○	○	○	○	▼	○	○	○
ETBE	○	○	○	○	○	○	▼	○	○	○
Ni-doped ZnO	○	○	○	▲	○	○	○	○	○	○
Ce	○	○	○	○	▲	○	○	○	○	○
CeO ₂	▲	▲	○	○	○	○	▲	▼	▼	○
Ferrocene	▲	▲	○	○	○	○	○	▼	▼	○
Mn	▲	▲	○	○	○	○	○	○	○	○
Ba	▲	▲	○	○	○	○	▲	○	○	○

▲ Positive effects.

▼ Negative effects.

○ Uncertain effects, influenced by operating conditions, or indicates different results in different literatures.



- emissions of total PAHs/BaPeq, EC/OC, PN, and POPs, with some exceptions such as butanol. In generators and other non-road engines, these additives tend to increase CBC emissions. The impact on heavy metal components varies, but there is an overall trend towards reduction.
- (3) **Metal-Based Additives:** Although metal-based additives are uncommon in non-road machinery, studies indicate that additives like cerium oxide (CeO₂), ferrocene, and barium-based compounds can enhance combustion efficiency and reduce the emissions of OC, EC, soot, and PN. However, these additives also increase PAH emissions, necessitating a balance between improved combustion efficiency and pollutant formation.
 - (4) **Carbon-Free Fuels:** The use of carbon-free fuels such as ammonia and hydrogen in non-road internal combustion engines is very limited. These fuels can significantly reduce soot emissions but may increase N₂O emissions.
 - (5) **Overall Impacts:** The effects of different green fuels on unregulated pollutants vary considerably. While some pollutants may decrease, others may increase, depending on the specific circumstances. Therefore, the selection of appropriate fuels and additives should be based on detailed consideration of the particular application and its emission characteristics.

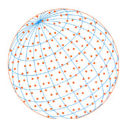
8 FUTURE PROSPECT

This study emphasizes the potential and actual effects of green fuels in non-road machinery. It systematically evaluates the advantages and disadvantages of different green fuels, focusing on unregulated pollutants and the development of new green fuels, providing insights for industry development and policy formulation. However, there are still significant deficiencies in current research. Future studies should consider the following directions:

1. In conclusion, the existing research on the effects of green fuels on emissions from Nonroad machinery remains limited. While studies primarily focus on generators and similar equipment, maritime vessel emissions receive less attention due to regulatory complexities. Common pollutants studied include PAHs, PN, and EC/OC, with fewer investigations into POPs, N₂O, VOCs, and metal components. Non-carbon-based fuels like hydrogen and ammonia, although increasingly used, lack comprehensive research, especially in Nonroad machinery applications. Hydrogen/ammonia combustion engines, while emitting harmless H₂O, produce NO_x, PM, and other harmful substances, highlighting the need for further exploration in this area.
2. In addition to the aforementioned green fuels, nano particle additives and oxygenated additives are potential options for road machinery to mitigate harmful emissions. However, research in this area is scarce for Nonroad machinery. Furthermore, unregulated pollutants like organic acids, hydrogen cyanide, and NH₃ are generated by road machinery, yet little research exists on their impact in Nonroad applications.
3. Other combustion sources such as coal, biomass, and waste also produce various pollutants like tar, N, S, K-based compounds, asphaltic compounds, and PBDEs, which can pose significant health risks. Therefore, future research should focus on understanding and mitigating these pollutants in the context of Nonroad machinery.
4. Conduct more field measurements under real operational conditions, as most current research is limited to laboratory settings, thereby restricting the practical applicability of findings.
5. Systematically assess the long-term health impacts of highly toxic pollutants under actual usage conditions, providing scientific bases for public health policies.

NOMENCLATURE

Abbreviations	Definition
CBCs	carbonyl compounds
CO	carbon monoxide
CO ₂	carbon dioxide
DEE	diethyl ether
DME	dimethyl ether
EC	elemental carbon



GMD	geometric mean diameter
HC	hydrocarbons
HCB	hexachlorobenzene
HMW-PAHs	high-molecular-mass PAHs
HVO	hydrogenated vegetable oil
IARC	International Agency for Research on Cancer
LMW-PAHs	low-molecular-mass PAHs
MMW-PAHs	medium-molecular-mass PAHs
N ₂ O	nitrous oxide
NO _x	nitrogen oxides
NRMM	nonroad mobile machinery
OC	organic carbon
PAHs	polycyclic aromatic hydrocarbons
PBBs	polybrominated biphenyls
PBDD/Fs	polybrominated dibenzo- <i>p</i> -dioxins and dibenzofurans
PBDEs	polybrominated diphenyl ethers
PCBs	polychlorinated biphenyls
PCDD/Fs	polychlorinated dibenzo- <i>p</i> -dioxins and dibenzofurans
PM	particulate matter
PN	particulate number
POPs	persistent organic pollutants
SO ₂	sulfur dioxide
SOF	soluble organic matter
VOCs	volatile organic compounds
WCO	waste cooking oil

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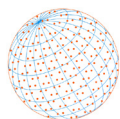
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SUPPLEMENTARY MATERIAL

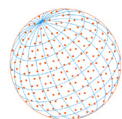
Supplementary material for this article can be found in the online version at <https://doi.org/10.4209/aaqr.240074>

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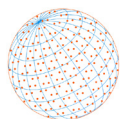
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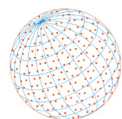
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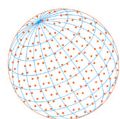
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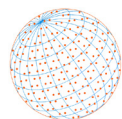
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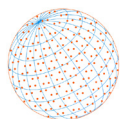
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