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Crude Tall Oil based Renewable Diesel: Performance, Emission Characteristics and Storage Stability

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Crude Tall Oil based renewable diesel: performance, emission characteristics and storage stability

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Abstract

International policies aiming at replacing fossil fuels with biocomponents are getting stronger. Hydrotreating of bio-oils is a sustainable way to produce premium quality diesel fuels from completely renewable feedstocks. The Finnish forestry company UPM has developed an innovative production process based on hydrotreatment to convert crude tall oil (CTO) into a high-quality renewable diesel fuel that can be used as a blending component or as 100% fuel in all diesel engines without modifications. Paraffinic, high cetane CTO renewable diesel allows efficient and clean combustion, reducing harmful air emissions in addition to not releasing any new CO₂ into the atmosphere during their combustion. This study investigated the effect of CTO renewable diesel (BVO) on engine performance and exhaust emissions. Conventional market diesel served as a reference fuel. The research engine was a common-rail offroad diesel engine, operated by using the load points of the non-road steady state cycle C1 of the ISO8178 standard. The use of BVO reduced all regulated gaseous emissions (NOx -10%, CO -7%, HC -7%). A significant 26% reduction was obtained in cycle-weighted particulate number. In addition to low emissions, fuels are required to remain stable and of high quality even after long-term storage. Another target of the present study was to clarify the influence of long-term storage on CTO renewable diesel properties. In this context, the paper reports the comprehensive results of fuel analyses in a fresh state and after four years of storage, the focus being on parameters that may be affected by the formation of oxidation products. BVO did not show substantial storage stability problems.

1. Introduction

Today, most heavy-duty vehicles and non-road applications rely on diesel engine technology as their primary power source [1]. Liquid fossil fuels have become the predominant fuel for those segments due to their high energy density and ease of distribution and storage, and a vast global infrastructure has been built over the past century to support this system [2]. These segments will also be the most difficult to electrify due to the high energy demand [3]. The expected large battery capacity would burden vehicle weight and reduce the payload capacity. Moreover, the cost would be high, and the charging time would be long. For non-road machinery operating for extended periods far away from charging infrastructure, the issue of battery technology is even more complicated. Thereby, robust and efficient diesel engines are expected to continue to play a central role as a power source for heavy-duty and off-road applications [2,3,4,5].

At the same time, concerns on climate change and global warming are growing, and international policies aiming to reduce greenhouse gas (GHG) emissions are getting stronger. In September 2020, the European Commission decided on increasingly stringent GHG emission reduction targets. The new goal is to reduce EU GHG emissions by at least 55% by 2030, compared to 1990 levels [6]. This is a substantial increase from the earlier target of 40%. This level of ambition for the next ten years is expected to set Europe on a balanced pathway to becoming climate neutral by 2050 – an economy with net-zero GHG emissions.

There are still no real alternatives in non-road transportation that could compete with an internal combustion (IC) engine throughout the entire application range. Besides, IC engines are still undergoing continuous further improvements with, e.g., advances in combustion technologies [5]. However, the GHG reductions of the required magnitude cannot be achieved through energy efficiency measures alone, and large-scale deployment of advanced liquid biofuels is needed in parallel [3]. There is a huge potential to significantly and realistically improve the sustainability of IC engines in the short- and medium-term through the development and deployment of renewable liquid fuels. Extensive existing distribution infrastructure can support such initiatives without modification or investment [2].

In recent decades, various biofuels have been introduced to the existing fuel markets [7]. First-generation biofuels, such as biodiesels derived from corn, sugarcane or soybean [8], symbolize the step towards fossil-free energy production. However, from the sustainability perspective, first-generation biofuels face a number of sustainability challenges, the biggest drawback being the competition with food production through excessive land use [7, 8]. There are also concerns about potential negative impacts on biodiversity [9] and competition for water in certain areas.

Advanced (second and third-generation) biofuels overcome many of the sustainability issues associated with first-generation biofuels. Notably, they do not compete with food resources, as they are produced from non-food crops, such as agricultural and forest residues, and industrial wastes [10, 11]. Additional benefits of biofuels produced from residues and waste streams are improved process energy efficiency [11] and substantially reduced CO₂ production [9]. Major CO₂ savings are based on the fact that the production of biofuels from organic waste results in fuels that contain only biogenic carbon [12], i.e., atmospheric carbon that was captured via biomass growth. Combustion of these fuels is considered carbon neutral, as it does not release any new CO₂ into the atmosphere. A recent study by Soam & Börjesson [13] showed that substitution of fossil diesel by highergeneration biofuels produced from by- and waste products may result in GHG emission reductions of up to 90% over their life cycle.

Various types of fuels can be derived from renewable feedstock, such as fats and vegetable oils [14]. A common biofuel is fatty acid methyl ester (FAME), also referred to as biodiesel, produced via transesterification of vegetable oils [15]. Due to many disadvantages of FAME biodiesel, such as deposit formation and storage stability problems [16], alternative routes for producing a diesel substitute have been developed. One of the alternative processes to esterification is hydrotreating. Hydrotreating vegetable oils or animal fats leads to a deoxygenated and thus stable product that is fully compatible with petroleum diesel fuel [17]. To clarify the difference from ester-type biodiesels, the term 'renewable diesel' is used for hydrotreated vegetable oils (HVO) [16].

The development of hydrotreated vegetable oil type diesel fuels have been strong particularly in the Nordic countries. Several studies have proved the emission benefits of these types of fuels. In Niemi et al. [18], a non-road diesel engine was driven with different fuel blends of traditional fossil diesel fuel and CTO based renewable diesel. With neat renewable diesel, a reduction of 21% in HC, 15% in CO, and 27% in particle number (PN) compared with fossil diesel were reported. Dimitriadis et al. [19] and Murtonen & Aakko-Saksa [20] also reported remarkable reductions in engine-out HC, CO and soot emissions with HVO. In Aatola et al. [16], smoke emissions were reduced by 35% with HVO without any modification to the engine control. At the same time, a 6% decrease in NO_x emissions was detected. Sarjovaara [21] reported similar results. The opposite behavior compared to the traditional NO_x-PM trade-off outlined the benefits of this kind of fuel [16, 21].

In addition to emission efficiency, maintaining fuel quality during long-term storage is an important aspect [22]. Fuel storage stability is crucial especially for vehicles and stationary engines that are out of use for extended periods with fuel in their tanks, such as seasonal machinery, or military equipment and emergency generator sets. Thereby, the stable quality of diesel fuel even after years of storage is also relevant in terms of the security of supply. The term 'fuel storage stability' refers to fuel's resistance against degradation processes that can alter its physicochemical properties and form undesirable compounds upon prolonged storage [23]. For biodiesel, the degradative processes are largely related to the degree of unsaturation in the fatty acid alkyl ester chain, water content and environmental factors such as temperature and exposure to air during storage [24, 25]. Biodiesels are more susceptible to oxidative attack than fossil diesel fuel [25], and oxidative degradation, caused by exposure to oxygen in the air, is the primary concern for them. Oxidative degradation of biodiesel can lead to the formation of short-chain carboxylic acids, aldehydes, ketones, and polymeric sediments [22,25,26,27] that impair fuel quality. It may cause problems with, e.g., fuel filter clogging and reduce the lifespan of fuel delivery components due to increased metal corrosion and elastomer degradation [27]. In addition, biodiesel's storage stability may include issues of water contamination and microbial growth [26]. Currently, EN 590 diesel standard limits the maximum allowable FAME content at 7% [28].

Instead, the storage stability properties of hydrotreated oils are considered almost similar to those of fossil diesel. As those fuels contains little or no oxygen, the long-term oxidation stability remains fairly steady, resulting in good storage behavior [19]. Generally, hydrotreated fuels also has low tendency to incorporate dissolved water. As a result, the risk of microbial growth is similar to that of fossil diesel fuel, and no extra precautions regarding microbiological growth, water separation, or storage stability are required [29]. Hydrotreated renewable diesel fuels, with their chemical composition almost similar to fossil-based paraffinic fuels (e.g., n-, iso-, and cyclic paraffin), also have similar compatibility with fuel delivery components and materials as fossil diesel [30]. Extensive field trials, both at 100% hydrotreated fuels and various blending ratios, have not led to any operability issues or need for extra maintenance regarding fuel systems, engines, or exhaust after-treatment devices [29].

The first target of the present study was to examine engine performance and exhaust emissions with renewable diesel. The renewable diesel under investigation was CTO based diesel fuel (BVO) produced by the Finnish forestry company UPM. Conventional market diesel (DFO) served as a reference fuel. The experiments were carried out using a commercial diesel engine, AGCO 44 AWI, designed for off-road use. During a thorough test bench campaign, the test engine was run using the loads of the ISO8178 standard's non-road steady-state cycle C1. The engine had no exhaust after-treatment system. Both fuels were studied using similar engine settings, and no engine or parameter optimizations were applied with the studied fuels. Alongside the gaseous emissions, PN and particle size distributions (PSD) and the basic engine performance were determined. In addition, the study examined the influence of long-term storage on CTO renewable diesel properties. In this context, the paper reports the comprehensive results of fuel analyses in a fresh state and after four years of storage, the focus being on parameters that may be affected by the formation of oxidation products.

2. Experimental setup

The engine experiments were performed by the University of Vaasa at the IC engine laboratory of the Technobothnia laboratory unit in Vaasa, Finland.

2.1 Fuels

Hydrotreated fuel for this study was delivered by UPM, Finland. The studied CTO based renewable diesel fuel was produced at the end of 2016 in UPM's biorefinery in Lappeenranta. The production process of UPM's renewable diesel – known as UPM BioVerno – is based on hydrotreatment of crude tall oil, a wood-based residue of chemical pulping process. CTO is a mixture of fatty acids with a carbon chain of 16 to 20 carbons (36–58%), rosin acids (10–42%), and sterols and neutral substances (10–38%). The composition varies depending on many factors, such as tree species and the growing cycle and age of the tree, geographical location, time of the year, and the pulping conditions [7].

The CTO-based renewable fuel production process includes several phases, shown in Figure 1. In the process, crude tall oil – an organic, water-immiscible liquid from a pulp mill – is first pretreated to remove solid particles, elements and metals, and other contaminants before introducing it into the hydrogenation reactor. The amounts of these impurities have to be reduced to ppm level or less to ensure the hydrogenation catalysts' functionality. The actual hydrotreating is carried out in a plug flow reactor at the pressure range of 2–12 MPa and at the temperature range of 280–430 °C, using commercial catalysts capable of simultaneous hydrodeoxygenation, hydrodesulfurization, isomerizetion, hydrogenation and cracking, Next, the remaining hydrogen sulfide and incondensable gases are removed. Finally, the hydrotreated CTO "raw diesel" containing mid-distillate diesel components and lighter naphtha components is distilled, i.e., fractionated to renewable diesel and naphtha. [7].



Figure 1. CTO-based renewable fuel production process.

In the present study, BVO was studied as neat. Commercial Finnish low-sulfur diesel fuel oil served as a reference fuel. Table 1 shows the main specification of the studied fuels and also lists the limits of the European diesel specification EN 590. The fuel analyses came from the fuel supplier. For BVO, two different analytical results are presented. The newer fuel analysis was performed in 2020 in the context of this study, i.e., after four years of storage. The older fuel analysis was conducted in a fresh state in 2016. A key part of the study was to examine the impact of long-term storage on BVO quality.

BVO mainly met the requirements of the EN 590. The density was slightly below the minimum limit of 820 kg/m³, and 95% distillation slightly exceeded the maximum of 360°C defined in EN 590. BVO was treated with a lubricity additive.

- 1. Crude tall oil
- 2. Pretreatment
- 3. Hydrotreatment
- 4. Fractionation
- 5. Recycle gas removal
- 6. Distilling
- Renewable diesel and renewable naphtha

Oxidation stability, a key parameter of diesel fuel quality in terms of its storage stability, was determined following the DIN EN ISO 12205 [44]. ISO 12205 describes the procedure for measuring the inherent stability of middle-distillate petroleum fuels by bubbling oxygen through the heated sample (95°C) for 16 hours. The test simulates the storage of diesel fuel for one year. Following the exposure to accelerated oxidizing conditions, the fuel is filtered, and the level of insoluble material is measured. BVO resulted in an insoluble concentration well below the maximum limit of 25 g/m³ specified in the EN 590.

The results of fuel analyzes are further discussed in Section 3.1

Table 1. Properties of the test fuels analyzed by ASG Analytik-Service GmbH. Abbreviations HFRR = High frequency reciprocating rig, CFPP = Cold filter plugging point.

	Test method	Unit	DFO	BVO in 2020	BVO in 2016	2016 Specification EN 590:2014	
						min	max
Cetane Number	DIN EN 17155 [31]*	-	57.5	61.6	60.8	51.0	-
Cetane Index	DIN EN ISO 4264 [33]	-	57.0	66.1	66.4	46.0	-
Density (15°C)	DIN EN ISO 12185 [34]	kg/m ³	836.4	812.7	812.6	820	845
PAH content	DIN EN 12916 [35]	% (m/m)	2.7	0.1	1.0	-	8.0
Total aromatics	DIN EN 12916	% (m/m)	23.8	4.6			
Sulphur content	DIN EN ISO 20884 [36]	mg/kg	7.9	<5	<5(<1)	-	10
Flash Point	DIN EN ISO 2719 [37]	°C	67.5	73.0	72.0	>55	-
Carbon residue (10% Dist.)	DIN EN ISO 10370 [38]	% (m/m)	< 0.10	< 0.10	0.02	-	0.30
Ash Content (775°C)	DIN EN ISO 6245 [39]	% (m/m)	< 0.001	< 0.001	< 0.005	-	0.1
Water content	DIN EN ISO 12937 [40]	mg/kg	32	22	<30	-	200
Total contamination	DIN EN 12662 [41]	mg/kg	<12	<12	6	-	24
Copper strip corrosion	DIN EN ISO 2160 [42]	Korr.Grad.	1	1	1	-	1
FAME content	DIN EN 14078 [43]	% (V/V)	< 0.1	< 0.01	< 0.1	-	7.0
Oxidation stability	DIN EN ISO 12205 [44]	g/m ³	<1	<1	7	-	25
filterable insolubles	DIN EN ISO 12205	g/m ³	<1		4	-	-
adherent insolubles	DIN EN ISO 12205	g/m ³	<1		3	-	-
HFRR (Lubricity at 60°C)	DIN EN ISO 12156-1 [45]	μm	380	380	361	-	460
Kin. Viscosity (40°C)	DIN EN ISO 3104 [46]	mm ² /s	3.3	2.8	2.8	2.0	4.5
% (V/V) recovery at 250°C	DIN EN ISO 3924 [47]	% (V/V)	22.7	33.2	33.1	-	<65
% (V/V) recovery at 350°C	DIN EN ISO 3924	% (V/V)	93.4	92.8	92.6	85	-
95% (V/V) recovery	DIN EN ISO 3924	°C	354	367	369	-	360
CFPP	DIN EN 116 [48]	°C	-12	-8	-9	-	**
Manganese (Mn)	DIN EN 16576 [49]	mg/l	< 0.5	<0.5	<0.5	-	2.0
Surface tension (20°C)	DIN EN 14370 [50]	mN/m	28.3	27.3			
Calorific value, lower	DIN 51900-2 [51]	MJ/kg	42.8	43.6	43.3	-	-

* In 2016: DIN EN 15195 [32]

** According national specifications

The test engine, an AGCO Power 44 AWI, was a high-speed fourcylinder diesel engine designed for off-road applications. It is turbocharged, intercooled and has Bosch common-rail fuel-injection system with a maximum pressure of 160 MPa.

Table 2 gives the main specification of the test engine. The engine was loaded with a Horiba eddy-current dynamometer WT 300. A Horiba SPARC controller platform was used to control engine speed, torque and throttle.

Table 2. Test engine specification.

Engine	AGCO POWER 44 AWI
Cylinder number	4
Bore (mm)	108
Stroke (mm)	120
Swept volume (dm ³)	4.4
Compression ratio	16.5:1
Rated speed (rpm)	2200
Rated power (kW)	103
Rated maximum torque at rated speed (Nm)	446
Maximum torque at 1500 rpm (Nm)	560

The engine was not equipped with devices for exhaust gas aftertreatment, meaning that raw engine-out emissions were recorded during the experiments.

2.3 Analytical procedures

The measurement setup for regulated gaseous emissions consisted of a chemiluminescence detector (CLD), used to measure the NO_x, a nondispersive infrared (NDIR) analyzer to measure CO and CO₂, and a heated flame ionization detector (HFID) for HC. In addition, several unregulated gaseous compounds were measured using Fourier transformation infrared (FTIR) equipment. PN and PSD were measured using an engine exhaust particle sizer (EEPS) spectrometer.

For the determination of the exhaust PN, the exhaust sample dilution before the EEPS was organized using a rotating disc diluter (model Matter Engineer MD19-3E). After the disk diluter using the constant dilution ratio of 60:1, the diluted sample was further diluted for the EEPS by purified air with a dilution ratio of 2:1. Thus, the overall dilution ratio was 120:1. Table 3 summarizes the methods and instruments adopted for the measurements. The schematic representation of the experimental setup is shown in Figure 2.

Table 3. Analytical instruments.

Parameter	Measuring device	Technology
NO _x	Eco Physics CLD 822 M hr	Chemiluminescence
CO, CO_2	Siemens Ultramat 6	NDIR
HC	J.U.M. VE7	HFID
O ₂	Siemens Oxymat 61	Paramagnetic
PN and PSD	TSI EEPS 3090	Spectrometer
Smoke	AVL 415 S	Optical filter
Unregulated gaseous	Gasmet DX4000	FTIR
emissions		
Air mass flow	ABB Sensyflow P	Hot-film anemometer
Cylinder pressure	Kistler KiBox	





The sensor data were collected using software made in the LabVIEW system-design platform, published by National Instruments, Austin, Texas. In addition to gaseous and particulate emissions, the recorded quantities included engine speed and torque and several fluid temperatures and fluid pressures, such as temperatures of cooling water, intake air and exhaust gas, and intake air and exhaust pressures. The engine control functions were monitored with AGCO SISU Power WinEEM4 software. No engine parameter optimizations were applied during the tests, and both fuels were studied using similar engine settings.

Engine warm-up and measurement procedures were identical for both fuels. Prior to measurements, the analyzers were manually calibrated once a day according to the instrument manufacturers' instructions. The brake-specific emissions results were calculated from the measured pollutant concentration data according to the ISO 8178 standard [52].

For combustion analysis, the heat release rate (HRR) and the mass fraction burned (MFB) were determined based on the in-cylinder pressure data, measured with a piezoelectric Kistler 6125C pressure sensor. The signal was filtered and amplified by a charge amplifier and transferred to a Kistler KIBOX combustion analyzer. A Kistler 2614B1 crank angle encoder recorded the crankshaft position. To smooth out irregular combustion, the cylinder pressure data were averaged over 100 consecutive cycles.

HRR and MFB were calculated using the AVL Concerto data postprocessing tool. The average values of in-cylinder pressures and the Thermodynamics2 macro with a calculation resolution of 0.2°CA were used to calculate HRR values. Thereafter, the HRR curves were filtered with a DigitalFilter macro and a frequency of 4,000 Hz. For MFB calculations, the pressure values were not averaged to determine the standard deviations for 100 consecutive cycles.

2.4 Experimental matrix

The performance and emission measurements were conducted at eight steady state load points. The loads were selected according to the ISO 8178-4 C1 test cycle, known as the non-road steady cycle (NRSC) [53]. The test engine's rated speed was 2200 rpm, and the intermediate

speed was 1500 rpm. At idle, the speed was 860 rpm. The loading points or modes and the corresponding engine speeds, loads and torques, as well as the NRSC weighting factors for the different modes, are listed in Table 4.

Table 4. Experimental matrix.

Mode	1	2	3	4	5	6	7	8
Speed (rpm)	2200	2200	2200	2200	1500	1500	1500	860
Load (%)	100	75	50	10	100	75	50	0
Torque (Nm)	446	334	223	45	560	420	280	1
Weighting factor	0.15	0.15	0.15	0.1	0.1	0.1	0.1	0.15

Before initiating the measurements, it was always waited that the engine run had stabilized. The main criteria were that the intake air and exhaust gas temperatures were stable.

3. Results and discussion

3.1 CTO renewable fuel storage stability

The CTO based renewable diesel fuel was produced at the end of 2016, in Lappeenranta biorefinery. The fuel sample was then taken from a storage tank to be used in engine tests conducted by the VTT Technical Research Centre of Finland in the project that had a goal to optimize engine for paraffinic diesel fuels. The fuel batch used in these tests was stored in a chemical container that had a constant temperature of 20 °C. The fuel was kept in a 230-liter plastic barrel with an airtight lid through the four-years ageing period. Prior to storage, the fuel was treated with a lubricity additive

Renewable diesel used in these engine tests showed slight unstable behavior in the oxidation tests made in 2016, see Table 1. There was a small amount of deposit formation in the oxidation stability test of EN ISO 12205, where the sample is at an elevated temperature of 95°C and treated with oxygen. However, the result was well inside the limits set by the EN590 standard.

When the fuel was analyzed in autumn 2020, there was no deposit formation. The amount of measured polyaromatics in the sample had decreased from 1.0 m-% to 0.1 m-%.

Overall, the changes in the fuel properties were small, partly negligible, which shows that hydrotreated CTO-based renewable diesel can be stored under controlled conditions for extended periods without compromising the fuel quality. The next sections show, what kind of engine results this stored fuel generated.

3.2 Combustion analysis

In general, combustion progressed in a quite similar manner with both fuels, as shown in Figures 3 and 4. However, BVO consistently showed a slightly lower peak of heat release rate (HRR) in the premixed combustion phase. Presumably, the higher cetane number of BVO resulted in better fuel ignition in the cylinder and a shorter ignition delay, resulting in a lower premixed peak. At rated speed at 75% load, main and post-injections were in use, while at intermediate speed at 50% load, the pilot injection was also adopted.







Figure 4. HRR as a function of crank angle at intermediate speed at 50% load.

Table 5 shows 10%, 50%, and 90% mass fraction burned (MFB) points at rated speed at 75% load (Mode 2) and intermediate speed at 50% load (Mode 7). Standard deviations of crank angles at MFB 10%, MFB 50% and MFB 90%, calculated from 100 consecutive engine cycles, are also given. No significant differences in combustion durations (CD) were detected, defined herein as the crank angle duration from MBF 10% to MBF 90%. MFB 50% values were also very similar for both fuels.

Table 5. Mass fraction burned, standard deviations and combustion durations at rated speed at 75% load and intermediate speed at 50% load.

	MFB 10%	Stdev	MFB 50%	Stdev	MFB 90%	Stdev	CD MFB10- 90%
	°CA	°CA	°CA	°CA	°CA	°CA	°CA
Mode 2							
DFO	7.7	0.085	16	0.12	31	0.50	23
BVO	7.7	0.093	16	0.12	31	0.60	23
Mode 7							
DFO	8.7	0.063	14	0.078	27	0.27	18
BVO	8.8	0.065	14	0.084	27	0.33	18

The differences in maximum cylinder pressures between the studied fuels were minimal, as shown in Figure 5, also indicating that combustion propagated quite similar way with both fuels.



3.3 Efficiency

The engine brake thermal efficiency was very similar for both fuels at all loads (Fig. 6). DFO produced slightly higher values, but the difference was marginal by no more than 0.6 percentage points, and thus of the order of magnitude of the measurement accuracy.



speed. CTO renewable diesel after four-years ageing. Baseline fuel DFO.

3.4 Gaseous emissions

The use of BVO resulted in lower brake-specific NO_x emissions on all measured speed and load configurations compared to DFO, as seen in Figure 7. Over the measurement cycle, BVO reduced NO_x by 10% (Fig. 8). Throughout the cycle with weightings, the NO_x for DFO was

7.9 g/kWh and for BVO 7.1 g/kWh, also indicating that the engine was tuned for high NO_x – and high efficiency – and intended for the later use of various catalysts for thorough NO_x reduction.





Figure 7. NO_x emissions at different speeds and loads. CTO renewable diesel after four-years ageing. Baseline fuel DFO.



Figure 8. Cycle-weighed NO_x emissions for aged CTO renewable diesel. Baseline fuel DFO.

The main reason for improved NO_x outcome is that BVO is an exclusively paraffinic fuel with a high H/C ratio and low aromatic content. NO_x formation is reduced with fuels having lower aromatic content as aromatic compounds have higher adiabatic flame temperature and thereby produce higher local combustion temperatures [54, 55]. In addition, the higher cetane number of BVO most probably resulted in a shorter ignition delay, also concluded by [56]. A shorter ignition delay reduces the energy released during the premixed combustion phase resulting in lower maximum combustion pressure and temperature in the cylinder, which in turn leads to lower NO_x formation [55].

CTO renewable diesel also proved to be beneficial in terms of CO and HC emissions (Fig. 9). Cycle-averaged CO emissions decreased by 7% from 0.27 to 0.25 g/kWh. In absolute terms, the reduction was reasonably small because of the already low CO values. The percentage reduction of brake-specific HC emissions was also 7%, from 0.14 to 0.13 g/kWh. HC reductions were more pronounced at lower loads.

HC and CO emissions for both fuels were well below the EU Stage V and US Tier 4 emission limits for non-road engines (0.19 g/kWh for HC and 5.00 g/kWh for CO).





Figure 9. Cycle-weighted CO and HC emissions for aged CTO renewable diesel. Baseline fuel DFO.

The main reasons for lower CO and HC emissions were assumed to be the higher cetane number and the lower aromatic content of BVO, and thus better combustion characteristics. BVO's high cetane number reduces ignition delay, which favors the oxidation process of CO emissions and limits overmixing of fuel, and consequently, HC emissions [19].

From the recorded unregulated gaseous emissions, nitrous oxide (N₂O) concentrations were low at all loads and almost equal for both fuels. N₂O generally ranged from 0.4 to 0.8 ppm, but at full load at 1500 rpm, it was below 0.1 ppm.

The wet exhaust methane (CH₄) contents were also low at all loads. The highest concentrations were recorded at 10% load at 2200 rpm (0.9 ppm for DFO and 0.5 ppm for BVO) and at half load at 2200 rpm (0.4 ppm for DFO and 0.1 ppm for BVO). At higher loads, CH_4 content was always below 0.1 ppm for both fuels.

The wet exhaust formaldehyde concentrations peaked at 3.0 ppm (at half load at 2200 rpm). Otherwise, it varied between 0.6 and 2.1 ppm. No significant difference between fuels was detected.

3.5 PN emission and smoke

The total PN emissions within the particle size range of 5.6 to 560 nm for BVO and fossil EN590 diesel at different speed and load combinations are shown in Figure 10. Each bar represents the mean value of the total PN. Depending on the load, the total PN decreased by 1-74% with CTO renewable fuel. The largest reductions were detected at low loads.



Figure 10. Total PN emission within the particle size range of 5.6 to 560 nm at different speeds and loads. CTO renewable diesel after four-years ageing. Baseline fuel DFO.

Figure 11 depicts the weighted total PN over the ISO8178 C1 driving cycle. Compared with DFO, CTO renewable fuel produced a reduction of 26% in the total PN emission. The result comes in agreement with previous studies [18, 56].





PSD at high load and low load conditions are depicted in Figures 12 and 13. The averages of PN concentrations are presented as data points and connected by lines. The shaded area depicts the standard deviation. The first peak of the bimodal PSD was detected at a particle diameter of 10 nm, and the other at approx. 35 nm. BVO generally produced a lower PN average in the larger particle size range, but the average number of smaller particles (10 nm) was slightly higher than that of DFO at some loads. With BVO, the decrease in particle number at the size category of 10 nm was evident at half load at 1500 rpm and at low idle.



Figure 12. Particle size distributions at high load conditions for CTO renewable diesel after four-years ageing. Baseline fuel DFO.

The improvement in the total PN emission can be explained by examining the fuel properties. The higher H/C ratio of BVO most likely led to a more complete combustion, also concluded by [57, 58]. Furthermore, due to the BVO's almost aromatic free composition, less PAH, i.e., precursors of soot, was formed [59, 60]. The significant PN reduction at the particle size category of 10 nm at low idle can be explained by BVO's near-zero sulfur and low aromatics content [61] and the improved ignition quality [62].

With both fuels, smoke was very low at all loads. For DFO, filter smoke number (FSN) values varied from 0.018 to 0.070 and for BVO from 0.013 to 0.037, respectively. The reason for lower engine smoke

with CTO renewable diesel was assumed to be the lower aromatic content of the fuel. According to Aatola et al. [16], a high cetane number can also have a beneficial effect on engine smoke.



Figure 13. Particle size distributions at low load conditions for aged CTO renewable diesel. Baseline fuel DFO.

4. Conclusions

This paper investigated CTO-based renewable diesel. The main target was to examine the effect of renewable diesel on the performance and exhaust emissions of an off-road diesel engine. In this context, raw engine-out emissions were recorded. CTO-based renewable fuel was studied in neat form, and conventional fossil diesel was used as the baseline fuel. An additional goal was to examine the effect of longterm storage on the properties of this renewable diesel.

From the fuel analyzes, the following conclusion could be drawn:

• After four years of storing, the changes in the fuel properties were negligible and showed that hydrotreated CTO-based renewable diesel fuel can be stored under controlled conditions for extended periods of time without compromising the fuel quality.

Engine performance and exhaust emissions were characterized over the eight-mode steady state cycle that followed a standardized NRSC driving cycle C1. Based on the measurements, the main findings were:

- The engine's brake thermal efficiency was very similar for both fuels.
- The use of BVO resulted in lower brake-specific NO_x emissions on all measured speed and load configurations compared to DFO. Over the NRSC cycle, BVO reduced NO_x by 10%.
- The main reason for lower NO_x emissions was assumed to be the high H/C ratio and low aromatics content of BVO. In addition, the higher cetane number of BVO most probably resulted in a shorter ignition delay, leading to lower NO_x formation.
- Over the eight-mode cycle, CO emissions decreased by 7% with BVO. HC emissions also decreased by 7%. HC reductions were more pronounced at lower loads.
- The main reasons for lower CO and HC emissions were assumed to be the higher cetane number and the lower aromatic content of BVO, and thus better combustion characteristics.
- The wet exhaust gas concentrations of nitrous oxide, methane, and formaldehyde were negligible with both fuels.
- Fueling with CTO renewable fuel produced a reduction of 26% in the total PN emission compared with DFO. The main reasons for decreased total PN emission were assumed to be the improved ignition quality, the beneficial hydrocarbon structure and near-zero sulfur and low aromatics content of BVO.

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Abbreviations

CA	crank angle
CD	combustion duration
CFPP	cold filter plugging point
CH ₄	methane
CLD	chemiluminescence detector
со	carbon monoxide
СТО	crude tall oil
DFO	diesel fuel oil
EEPS	engine exhaust particle sizer
EU	European Union
FAME	fatty acid methyl ester
FSN	filter smoke number
FTIR	Fourier-transform infra-red
GHG	greenhouse gas
НС	total hydrocarbons
HFID	heated flame ionization detector
HFRR	high frequency reciprocating rig
HRR	heat release rate
HVO	hydrotreated vegetable oil
IC	internal combustion
ISO	International Standard Organization
MFB	mass fraction burned

N ₂ O	nitrous oxide
NDIR	non-dispersive infra-red
NO _x	oxides of nitrogen
NRSC	non-road steady cycle
РАН	polyaromatic hydrocarbons
PN	particle number
PSD	particle size distributions