




Supplementary Information

Biodiesel: An Overview II

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This paper is dedicated, *in memoriam*, to Professor Angelo da Cunha Pinto.

Table S1. Main studies recently published regarding pollutants exhausted through blended or unblended biodiesel fuel

Sampling year	Pollutant	Source	Observed result
2004/2005 ^{a 1,2}	polycyclic aromatic hydrocarbons	diesel gasoline biodiesel	high atmospheric concentrations of particulate PAHs from diesel-fueled road vehicles, exacerbated by accumulation in the daytime sea breeze circulation; burning diesel emits a higher concentration of PAH when compared to burning gasoline, which is the main PAH source; increasing use of biodiesel could reduce the emission of PAHs; benzo [b] fluoranthene (0.130-6.85 ng m ⁻³), the PAH with the highest concentration found in samples from diesel burning from ships, smaller boats and automobile traffic; chrysene (from 0.075 to 6.85 ng m ⁻³) was the one presenting higher concentrations from heavy duty diesel automobiles
2006/2007 ^{a 3}	polycyclic aromatic hydrocarbons	gasohol (gasoline with 24% of ethanol), neat ethanol, compressed natural gas (CNG), gasohol and ethanol, and diesel	Σ PAH accounted for 0.0018% of the TSP mass and 0.0012% of the PM ₁₀ mass; contributions of carcinogenic priority PAHs (B[a]An, B[b]F, B[k]F, B[a]Py, IPy) were 52% and 54%, for (DB[ah]A)
2008 ^{a 4}	PAH major ions	bus station fuel blend B3 (97% diesel and 3% biodiesel)	nitrate and sulfate were the highest, representing 21.2% of PM mass; nine PAHs were quantified; DBA concentrated in smaller sizes; all PAHs were found, particles with diameter lower than 0.25 μ m were the most abundant; biodiesel decreases the total PAHs emission however, it increased the fraction of fine and ultrafine particles when compared to studies with diesel fuel; evidences about particulate sulfate and mainly nitrate found in this study may also be attributed to the emission from diesel/biodiesel fuel blend (B3)

Table S1. Main studies recently published regarding pollutants exhausted through blended or unblended biodiesel fuel (cont.)

Sampling year	Pollutant	Source	Observed results
2008 ^{b 5,6}	carbonyl compounds	biodiesel-diesel (B2, B5, B10, B20, B50 and B75) and pure diesel, pure biodiesel as well; commercial pure diesel	formaldehyde and acetaldehyde presented the highest emission levels;
			acrolein increased for all blends while formaldehyde increased for all blends except B20 and B50; when considering total CC emissions, there is a consistent concentration decrease beginning at B20 up to B100 blends;
2009 ^{b 7,8}	CO CO ₂ NO _x	fuel blend B5 (commercial diesel oil), ethanol 99.5%, ethanol 95% with: methyl soybean ester (SB), methyl castor ester (AB) methyl residual oil ester (RB) B100 from transesterification of soybean oil and diesel	the formaldehyde, acetaldehyde, acrolein, acetone, propionaldehyde, butyraldehyde, and benzaldehyde emissions from the B2, B5, B10, and B20 mixtures were higher than those from neat diesel;
			the total carbonyl emissions from biodiesel blends were higher than those from diesel; the only exception was benzaldehyde, which a significative reduction was observed
	carbonyl compounds	fuel blend B5 (commercial diesel oil), ethanol 99.5%, ethanol 95% with: methyl soybean ester (SB), methyl castor ester (AB) methyl residual oil ester (RB) B100 from transesterification of soybean oil and diesel	formaldehyde, acetaldehyde, acetone, and propionaldehyde presented the highest emission concentrations;
			all fuel blends emitted more CC than pure diesel;
	CO CO ₂ NO _x	fuel blend B5 (commercial diesel oil), ethanol 99.5%, ethanol 95% with: methyl soybean ester (SB), methyl castor ester (AB) methyl residual oil ester (RB) B100 from transesterification of soybean oil and diesel	the fuel with castor biodiesel emitted the lowest CC concentration and when ternary blends contain vegetable oil, there is a strong tendency to increase the emissions of the high molecular weight CCs and decrease the emissions of the low molecular weight CCs;
			the highest acrolein concentration was observed when the fuel contains diesel, ethanol and biodiesel; with the exception of NO _x , the use of ternary blended fuels resulted on the increase of the studied compounds emission rates;
	CO CO ₂ NO _x	fuel blend B5 (commercial diesel oil), ethanol 99.5%, ethanol 95% with: methyl soybean ester (SB), methyl castor ester (AB) methyl residual oil ester (RB) B100 from transesterification of soybean oil and diesel	among fuel blends, the diesel/ethanol fuel showed higher reduction of the NO _x emission;
			the emission concentrations slightly decrease with decreasing engine loads
	CO CO ₂ NO _x	fuel blend B5 (commercial diesel oil), ethanol 99.5%, ethanol 95% with: methyl soybean ester (SB), methyl castor ester (AB) methyl residual oil ester (RB) B100 from transesterification of soybean oil and diesel	diesel has a higher relative contribution of formaldehyde than biodiesel, but biodiesel shows a comparatively high content of propionaldehyde and methacrolein;
			biodiesel, as an alternative fuel, has lower specific reactivity caused by carbonyls than diesel

Table S1. Main studies recently published regarding pollutants exhausted through blended or unblended biodiesel fuel (cont.)

Sampling year	Pollutant	Source	Observed result
2010 ^{a 9-11}	PAH nitro-PAH quinones	bus station B4 mix as fuel (4% diesel and 96% biodiesel) for buses ethanol-to-gasoline (with any proportion) for light duty vehicles run	2-nitrobenzanthrone = 14.8 µg g ⁻¹ 3-nitrobenzanthrone = 4.39 µg g ⁻¹ PAH 0.06 to 15 ng m ⁻³ nitro-PAH < LOD to 69.4 ng m ⁻³ 0.32 to 3.38 ng m ⁻³ (nonderivatized form) 0.29 to 4.75 ng m ⁻³ (acetylated derivatized form) quinones 0.27 to 115 ng m ⁻³
2010 ^{c 12,13}	low-molecular weight carboxylate, water-soluble major ions and carbonyl compounds	fuel blend B5 (95% diesel and 5% biodiesel)	formate was the most abundant carboxylate species in both PM _{2.5} and PM ₁₀ followed by acetate and oxalate; formaldehyde, acetaldehyde and propanone were the most abundant; concentrations higher than 3% biodiesel-to-diesel demonstrated an improvement in the carbonyl concentration profile with high flow of heavy-duty vehicles
2011 ^{c 14}	carbonyls, polycyclic aromatic hydrocarbons, nitro-PAHs and oxy-PAHs	two fresh and two oxidized biodiesel fuels of different source materials were blended with an ultra low sulfur automotive diesel fuel at proportions of 10, 20, and 30% v v ⁻¹	CC emissions for biodiesel blends were significantly higher than those of diesel fuel; the use of the oxidized biodiesel increases in both light and heavy molecular weight carbonyls; CC emissions were higher over the NEDC and Artemis Urban as compared to road and motorway cycles; the addition of most biodiesel blends led to increases in low molecular-weight PAHs when compared to diesel fuel; PAHs emissions for the oxidized biodiesel blends were higher than those of diesel fuel and the other biodiesel blends; an adverse effect was observed for the nitro-PAH and oxy-PAH emissions with the use of oxidized blends; the higher exhaust temperature and thus the better performance of the oxidation catalyst led to lower PAH emissions. PAH emissions may increase during cold-start engine conditions

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Sampling year	Pollutant	Source	Observed result
2013 ^{a 15}	carbonyl compounds	fuel blend B5 (95% diesel and 5% biodiesel)	it was quantified two free carbonyl compounds and six bound carbonyl compounds
2013 ^{a 16}	water-soluble transition metals, PAHs, nitro-PAHs, quinones	fuel blend B7 (93% diesel S50 and 7% biodiesel)	biodiesel/diesel particles exhausted from heavy-duty vehicles showed oxidative potential levels similar to fossil diesel-emitted particles in other studies, with trace metals contribution at 89%
2014 ^{a 17}	PAHs	B4, B25, B50 (BX where X is % of biodiesel add to diesel) and B100	(+) biodiesel increase of PAH emission
2014 ^{c 18}	NO _x , CO, HCs, CO ₂ , PM	B100 (pure soybean oil Biodiesel), B5 and B5E6 (89% diesel, 5% biodiesel and 6% ethanol)	B5E6 increases HC emissions and the number of smaller particles; B100 reduces HC and CO emissions and increases NO _x emissions; B5E6 and B100 reduce power and increase fuel consumption, but energy efficiency could be similar to B5 fuel; CO ₂ emissions are statistically similar
2014 ^{b 19}	carbonyls	diesel, an animal-fat biodiesel (AF), 50 vol.% (AF50) and a blend of this one with tire pyrolysis liquid at 5 vol.% (5 TPL)	carbonyl emissions from biodiesel are higher than diesel; despite specific emissions were slightly higher for 5TPL than those for DC, their reactivity is lower; emissions of acrolein can easily reach the established limits of hazardous contaminant

Table S1. Main studies recently published regarding pollutants exhausted through blended or unblended biodiesel fuel (cont.)

Sampling year	Pollutant	Source	Observed result
2014 ^{b 20}	NO _x , NH ₃ and N ₂ O	low-sulfur diesel (less than 50 ppm), ultra low sulfur diesel (less than 10 ppm) and a blend of 20% soybean biodiesel	the biodiesel blend presented lower concentrations in the exhaust fumes than using ultra-low sulfur diesel; The selective catalytic reduction system its reducing NO _x emissions; however, NH ₃ and N ₂ O emissions increased; N ₂ O increases exacerbate the greenhouse effect and the risk assessment indicated small values for non-cancer risks only for NH ₃
2015 ^{b 21}	sulfur	diesel (S10-A, S10-B S500-A and S500-B)	sulfur was quantified ranging from 161 to 5.6 mg kg ⁻¹ (mean values) for S500-A and S10-B, respectively
2015 ^{a 22}	polycyclic aromatic hydrocarbons, nitro-PAHs, and petroleum biomarkers	fuel (ultra-low sulfur diesel or ULSD, Swedish low aromatic diesel, and neat soybean biodiesel)	Swedish diesel, biodiesel and the DOC + DPF significantly reduced PM _{2.5} , PAHs, nitro-PAHs, hopanes and steranes emissions, although emissions of PM _{2.5} and several compounds (benzo[k]fluoranthene and 5-nitroacenaphthene) increased during idling with biodiesel; emission rates of PM _{2.5} and SVOCs increased with engine load, with the important exception that PM _{2.5} emissions increased during idling with B100; the toxicity of diesel exhaust was reduced using the alternative fuels and the DOC + DPF
2016 ^{b 23}	CO, NO _x , saturated hydrocarbon compounds, unsaturated hydrocarbons, aldehydes, alcohols, SO ₂ , formic acid and benzene	mineral diesel, Karanja biodiesel blends (KB5, KB20) and methanol blended (M5) with diesel	all test fuels lead to increase in CO emission at higher engine loads; HC emissions were observed to be higher from alternative fuels used in this study at lower engine loads; however, these alternative fuels resulted in reduction in HC emissions at higher engine loads; NO _x emissions were observed to be marginally lower from alternative test fuels; biodiesel blends emitted lower trace concentrations of methane while M5 emitted higher trace concentration of methane vis-a-vis baseline mineral diesel; aldehyde is also a major carcinogenic compound, and biodiesel blends emitted higher trace concentration; the fraction of unidentified hydrocarbons increased drastically at full load for all test fuels, which subsequently lead to increased HC emissions at higher engine loads

Table S1. Main studies recently published regarding pollutants exhausted through blended or unblended biodiesel fuel (cont.)

Sampling year	Pollutant	Source	Observed result
2017 ^{a 24}	organic carbonelemental carbon	B4 (soybean oil biodiesel), B50 (waste cooking oil biodiesel), B100 (waste cooking oil biodiesel).	(+) biodiesel > non-fractal particles with diffuse edges; morphological parameter showed agglomerates were self-arranged in fractal geometry, with similar fractal dimension values, regardless the fuel composition
2017 ^{a 25}	polycyclic aromatic hydrocarbon and inorganic ions	B20 fuel (20% waste cooking oil biodiesel)	the three aftertreatments were shown to reduce total PAH emissions; DOC + DPF (diesel particle filter), and DOC + CDPF (catalyzed diesel particle filter), can decrease total toxicity equivalent quantity, effectively with a sharp decrease in PAH mass; the DOC (diesel oxidation catalyst) increased the particles TEQ by 46.9%; the catalyst in the DOC increased some high molecular weight PAHs; catalysts in DOC and CDPF promotes the formation of SO ₄ ²⁻ and NO ₃ ⁻ , which leads to higher inorganic ion emissions with DOC than no aftertreatments and higher inorganic ion emissions with a DOC + CDPF than with a DOC + DPF
2017 ^{c 26}	carbonyl compounds, unsaturated hydrocarbons, aromatic compounds	B20, B50, B75, neat biodiesel from waste cooking oil (WCO) and pure diesel with 10 ppm by mass of sulfur	biodiesel increases the weighted brake specific emissions of formaldehyde, acetaldehyde, 1,3-butadiene, propene, ethene and benzene; the chemical composition and shorter combustion duration of biodiesel also contribute to the increased emissions of the aforementioned unregulated gases; but there are decreasing trends for the weighted toluene and xylene emissions when using biodiesel; very high correlation coefficients (higher than 0.9) are observed between weighted unregulated gaseous and particulate matter (PM) emissions and biodiesel content, which indicate that the weighted unregulated gaseous and PM emissions are proportional to the biodiesel content
2018 ^{a 27}	polycyclic aromatic hydrocarbon and nitro- PAHs	diesel (ULSD maximum of 10 ppm or mg kg ⁻¹ of sulfur) and B5 (ULSD) and B20 (ULSD)	the results indicated the use of selective catalytic reduction and the largest fraction of biodiesel studied may suppress the emission of total PAHs; the toxic equivalent was lower when using 20% biodiesel, in comparison with 5% biodiesel, reaffirming the low toxicity emission using higher percentage biodiesel; the use of SCR, suppress the nitro-PAHs compounds

Table S1. Main studies recently published regarding pollutants exhausted through blended or unblended biodiesel fuel (cont.)

Sampling year	Pollutant	Source	Observed result
2019 ^{a 28}	organic carbon, elemental carbon, polycyclic aromatic hydrocarbons, <i>n</i> -alkanes, fatty acids and inorganic ions	B5, B10 and B20 (biodiesel from waste cooking oil) and the petroleum diesel	biodiesel resulted in lower particle number emission and the reduction increased linearly with the biodiesel ratio, but the proportions of nucleation mode particles were enhanced; CO emissions decreased with the biodiesel index, while the EC increased; biodiesel reduced PAHs emissions and the toxic equivalent; lower particle number emission, low emissions of <i>n</i> -alkanes and fatty acids and higher major ion emissions were observed with the use of biodiesel; the use of biodiesel caused higher major ions emissions including Cl ⁻ , NO ₂ ⁻ , NO ₃ ⁻ , SO ₄ ²⁻ , Na ⁺ , NH ₄ ⁺ , K ⁺ and Ca ²⁺ in the exhaust particles
2019 ^{c 29}	polycyclic aromatic hydrocarbons and carbonyls	palm oil biodiesel, animal fat biodiesel and soybean oil biodiesel; a blend of 80% biodiesel; Chilean diesel (A1 grade)	palm oil biodiesel emissions being the least harmful, the animal fat biodiesel and soybean emissions were as toxic as the diesel emissions; carbonyl emissions were higher in the two biodiesels than in diesel because of the increase in oxygen content in the fuel mixture; although PAHs were reduced up to 66%, they were still present; high PAH emissions in soybean oil biodiesel were related to the higher content of unsaturated methyl esters (double bonds) in its composition
2019 ^{b 30}	CO, CO ₂ , O ₂ , total hydrocarbons, NO _x , aldehyde and alkenes	ULSD (ultra-low sulfur diesel), B20, B50 B75 and B100 (waste cooking oil biodiesel)	the pure biodiesel led to increase in brake thermal efficiency and decreases in THC, CO and PM emissions; however, increased the brake specific fuel consumption and unregulated emissions (except toluene and xylene) at low engine loads, and increases in CO ₂ , NO _x and 1,3-butadiene at high engine loads
2019 ^{a 31}	organic pollutants and polycyclic aromatic hydrocarbons	D100 (pure diesel), B20, B40, B60, B80 and B100 (WCO-based biodiesel)	the EURO IV diesel engine showed PM and toxic organic pollutant emissions were reduced with the increase in the blending ratio up to B60 scenario when compared to the D100 scenario; EURO III engine had improvement in combustion but using biodiesel resulted in greater polychlorinated dibenzo- <i>p</i> -dioxins and dibenzofurans reductions

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Sampling year	Pollutant	Source	Observed result
2020 ^{a 32}	polycyclic aromatic hydrocarbons	diesel fuel and glycerine fuel; a blend composed of the diesel (80% v/v) and a biofuel called o.bio [®] (20% v/v); o.bio [®] is a blend with fatty acid methyl ester derived from waste cooking oil (69.6% v/v), a fatty acid glycerol formal ester (FAGE, 27.4% v/v) and acetals (3% v/v)	the particle-bound PAH emissions from a residual glycerine-derived fuel blend are lower than those measured using a conventional diesel fuel; for both fuels tested, the emissions of these particle-bound PAH increase with the engine speed and with the EGR rate in the Mo.bio tests due to the reduction in the local temperature (as well as in the oxygen concentration in the diesel combustion chamber); the degree of carcinogenic potential of these emissions is higher for the mineral diesel fuel than for the biofuel mixed with FAGE; this result is attributed to the greater emission of compounds such as benzo[a] pyrene (BaP) and dibenz[a,h]anthracene (DahA) (with a TEF = 1) in the conventional diesel fuel

^aParticulate phase; ^bgaseous phase; ^cparticulate and gaseous phase. PAH: polycyclic aromatic hydrocarbon; HC: hydrocarbon; PM: particle material; TSP: total suspended particulate; DBA: dibenzo(a,h)anthracene; CC: carbonyl compounds; NEDC: New European Driving Cycle; LOD: limit of detection; SVOCS: semi-volatile organic compounds; DOC: diesel oxidation catalyst; SCR: selective catalytic reduction; EC: elemental carbon.

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