



A Review on the Effects of Biodiesel Blends on Compression Ignition Engine NO_x Emissions

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ABSTRACT

Biodiesels are produced by the transesterification of corresponding triglyceride feedstocks of vegetable (example: soybean, canola, palm, karanja) or animal fat sources. Currently, the leading feedstocks are soybean oil in the U.S., canola oil and rapeseed oil in Canada and Europe, and palm, karanja, jatropha and other oils in Asia. Due to the cost and production considerations of these biodiesels, blending biodiesels with the petroleum fuels appears to be a prudent option in the near-term. The use of biodiesels in compression ignition engines results generally in a reduction in the emissions of carbon monoxide (CO), hydrocarbons (HC), and particulate matter (PM), but a slight increase in the oxides of nitrogen (NO_x) emission. The reported NO_x emissions do not exhibit definitive trends and the results are significantly influenced by many factors, including engine type and design, test cycle, start of injection, ignition delay, fuel composition, adiabatic flame temperature, radiative heat transfer, fluid dynamics and combustion phasing. Due to appreciable variations in the physical properties and the highly nonlinear nature of the combustion process, the NO_x emission with biodiesel blends *does not vary monotonically with the percentage of biodiesel* in the blend. Hence, the intricate dependence of NO_x on biodiesel and its blending effect cannot be completely explained under all engine type and operating conditions. Although the literature contains several studies on the performance and emissions of compression ignition engines fueled with neat biodiesels, the information on the effects of blends is scattered and has not yet achieved a definitive status to explain the blending effect on NO_x. Hence, this work was motivated to review the available data with respect to the NO_x emission from engines fueled with the petroleum diesel/biodiesel blends.

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1. Introduction

Biodiesels, considered as alternative fuels to petroleum diesel, are defined as fatty acid methyl or ethyl esters derived from triglycerides of vegetable oils or animal fats. The utilization of biodiesels reduces greenhouse gas emissions, assists in sustainable energy development, and enhances energy independence due to the renewable and biodegradable nature of these fuels. Besides being close to environmentally carbon-neutral, biodiesels have properties similar to those of petroleum fuels and can be blended with petroleum fuels and used in existing engines without major modifications. Furthermore, they contain fuel-bound oxygen while being free of aromatic content; therefore, blends of biodiesels and petroleum fuels present the capability of reducing soot emissions from engines. Extensive studies have been conducted on the potential of biodiesels and the feasibility of commercializing them in the long run, as presented in comprehensive reviews [Van Gerpen, 2005; Meher et al., 2006; Agarwal, 2007]. With the current understanding of combustion characteristics of biodiesels, only blending of biodiesel (usually in volumetric percentage) with petroleum fuels is considered feasible in the near-term due to limited current availability of the commercial biodiesels and the lack of experience on the long-term effects of handling,

transportation, storage, and combustion of these biodiesels and blends on the engines and the environment. Currently, there are about 250 biodiesel fueling stations in the United States providing a range of biodiesel blends ranging from B20 to B100 (number indicating the volumetric percent of biodiesel) with B20 being the widely available blend [Alternative Fuels Data Center, 2015]. However, the commercial use of biodiesel has not been widely popular. One reason is a dearth of definitive understanding about the long-term impact of biodiesel and their blends with petroleum fuels on engines, especially on the engine power performance and regulated gaseous emissions. Previous studies have revealed that the use of biodiesels and their blends in a compression-ignition engine resulted in an appreciable reduction in the emissions of particulate matter, HC and CO, but an increase in NO_x emissions, compared to the use of diesel fuel. This increase in NO_x emission is termed the biodiesel NO_x effect, which has remained a prolonged issue over this decade. The US Environmental Protection Agency [2002] has categorized NO_x as one of the key pollutants in engine emissions that can affect the human respiratory system.

The biodiesel usage in the U.S alone is expected to reach 4000 million gallons (15000 million liters) in 2030. Hence, a comprehensive knowledge of the effects of biodiesels and their blends on NO_x is required for the use

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of biodiesels to become prevalent. Therefore, it is crucial to understand the effects of fuel properties and operating characteristics of the engines on biodiesel NO_x emission to develop enhanced mitigation and abatement techniques. The objective of this review article is to provide a comprehensive picture of the influence of fuel composition, engine calibration settings, fuel fluid dynamic parameters, and interactions of thermo-fluid-chemical parameters on the compression-ignition engine NO_x emission when fueled with biodiesel and their blends with petroleum fuels. Throughout this article, biodiesel blends are referred to as Bxx, where B stands for biodiesel irrespective of the feedstock and xx stands for the volumetric percent of biodiesel in the blend. For example, a B20 blend consists of 20% biodiesel and 80% petroleum diesel (by volume).

1.1. Statistics and significance of literature review

The United States Environmental Protection Agency, EPA [2002] has published a technical evaluation of the impact of biodiesel on exhaust emissions from compression ignition engines. EPA [2002] reported the averaged results of all these studies in which the use of biodiesel and its blends has a conspicuous and persistent favorable impact on the emissions of carbon monoxide (CO), hydrocarbons (HC), particulate matter (PM) and a relatively small negative influence on NO_x emission. The averaged results show a monotonic increase of up to 10% NO_x with neat biodiesel and a reduction of up to 50% and 68% of CO and HC respectively. Although the report provided an approximate trend of biodiesel effect on emissions of regulated pollutants, it does not contain sufficient experimental results for the blends having greater than 20% of biodiesel. The statistical

data employed for this study predominantly constitute emission results from B20 and B100. Moreover, these correlations have been developed based on a maximum likelihood curve-fitting approach instead of least-square regression. Hence, to provide a visual assessment of the goodness of fit, the report also compared the actual percent change in NO_x emissions of biodiesel and its blends from the corresponding emissions of the base fuel, which typically is the No. 2 diesel in most of the studies. The average effect of biodiesel on NO_x emission was seen to be small, but with a high variance, which resulted in difficulty in discerning a clear pattern. In the current review article, the engine NO_x literature has been updated with more recent works and findings along with the assessment compiled by Hoekman et al. [2011]; the total number of studies considered for this review with a breakdown of different volumetric proportion of blends and resultant change in NO_x emissions are presented in Table 1. This table contains the emission data from the engine tests reported and is grouped together as biodiesel (irrespective of the fuel feed stock), yet most of these studies employed biodiesel from soybean oil. Similar to the study by EPA, B20 constitutes the major data points with a scarce representation of higher biodiesel blend levels. We have plotted Figure 1 based on the data from Table 1 and Figure 2, an average value of the percentage variation of NO_x emission index (g/kW-hr) with biodiesel content in the blends. The wide scatter in the results from these studies reveals the indefinite trend of NO_x emission with the biodiesel content. This assessment again shows that the NO_x emission and its dependence on the fuel chemistry and content of biodiesel are difficult to ascertain since the effects are highly variable.

Table 1: Statistics of the effect of biodiesel content in the blend on NO_x emission from studies considered in this review

Biodiesel blend level (vol. %)	^a Average reduction of thermal input (MJ/kg) from base (diesel) fuel with biodiesel blending (%)	^b Variation of NO _x with biodiesel content					
		No of studies that show an increase	No of studies that show decrease	No of studies that show no change	Total number of studies considered	Average percentage variation of NO _x from base (diesel) fuel	Standard deviation of percentage variation of NO _x from base (diesel) fuel
1	0.05	1	3	6	10	-1	2
2	0.10	0	2	0	2	-3	1
3	0.14	8	0	2	10	5	4
5	0.24	17	6	1	24	4	5
6	0.29	2	1	0	3	4	7
7	0.34	2	1	0	3	1	3
10	0.48	19	17	2	38	1	8
20	0.96	163	67	5	235	2	9
30	1.44	18	5	1	24	5	9
35	1.68	7	2	1	10	3	9
40	1.92	16	3	0	19	7	9
50	2.41	21	13	6	40	3	8
65	3.13	2	0	0	2	5	0
100	4.81	92	27	3	122	7	14
Total		368	147	27	542		

^a Considering heating value of diesel as 44MJ/kg and average value of biodiesel as 39.6 MJ/kg (from Table.3); ^b Includes data collected from [Hoekman et al., 2011]

In summary, a potential drawback in compiling these results from a wide range of experimental studies would be an inconsistent comparison of emission data in terms of engine type, load settings, engine speed settings, and fuel-air ratio during combustion which are not the characteristics of fuels alone. These presentations pose a risk of observing trends that may not correspond only to the parameters under study. Based on these developed correlations, it is difficult to confidently predict the amount of NO_x emitted from any biodiesel-fueled engine and to conclude the biodiesel blending effect on NO_x emissions.

2. NO_x formation mechanisms with biodiesels

Nitrogen oxides present in atmosphere induce photochemical smog formation. The nitrogen oxides of environmental interest are NO, N₂O and NO₂, among which the NO and NO₂ are collectively referred to as NO_x [Lissianski et al., 2000; Fattah et al., 2013]. The more stable NO always predominates over the other oxides of nitrogen in the combustion flue gas. The coupled concentrations of NO and NO₂ are decided by the fast radical reactions: NO₂ reacts with O, H and OH to form NO and NO

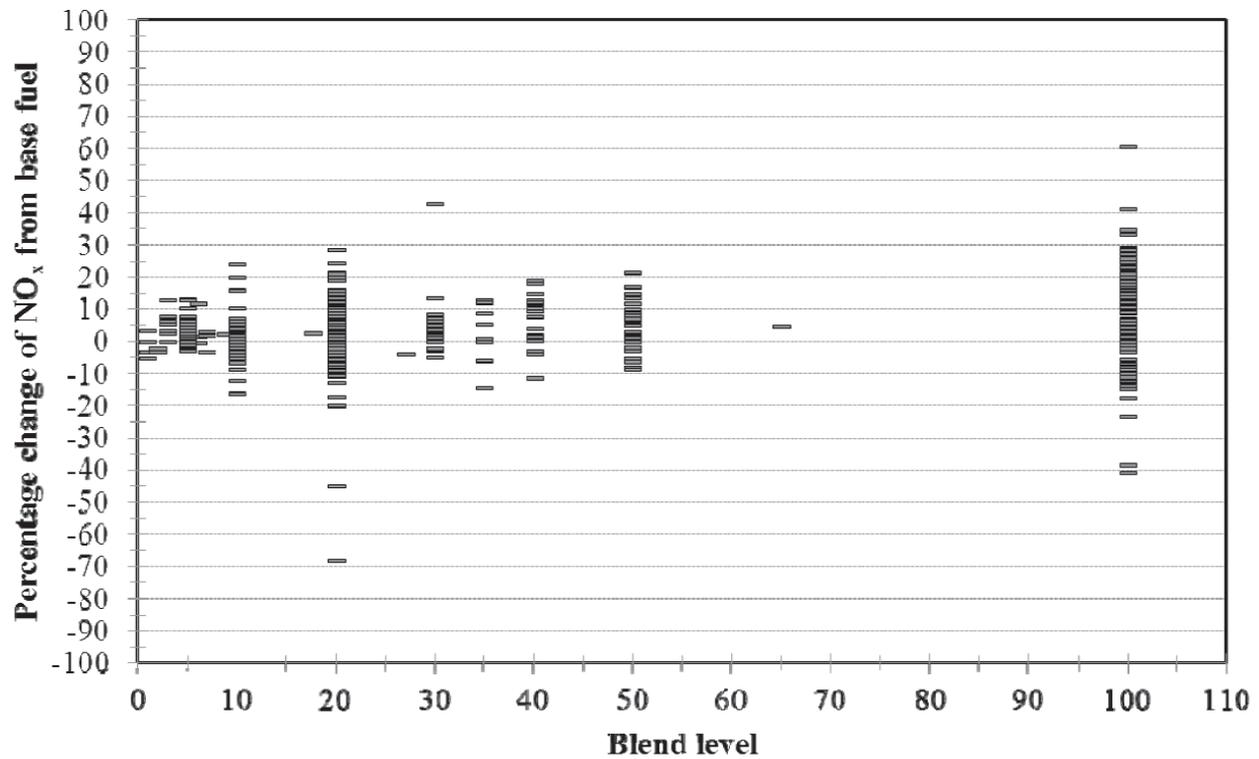


Figure 1 Percentage variations of NO_x emission from studies considered in this review

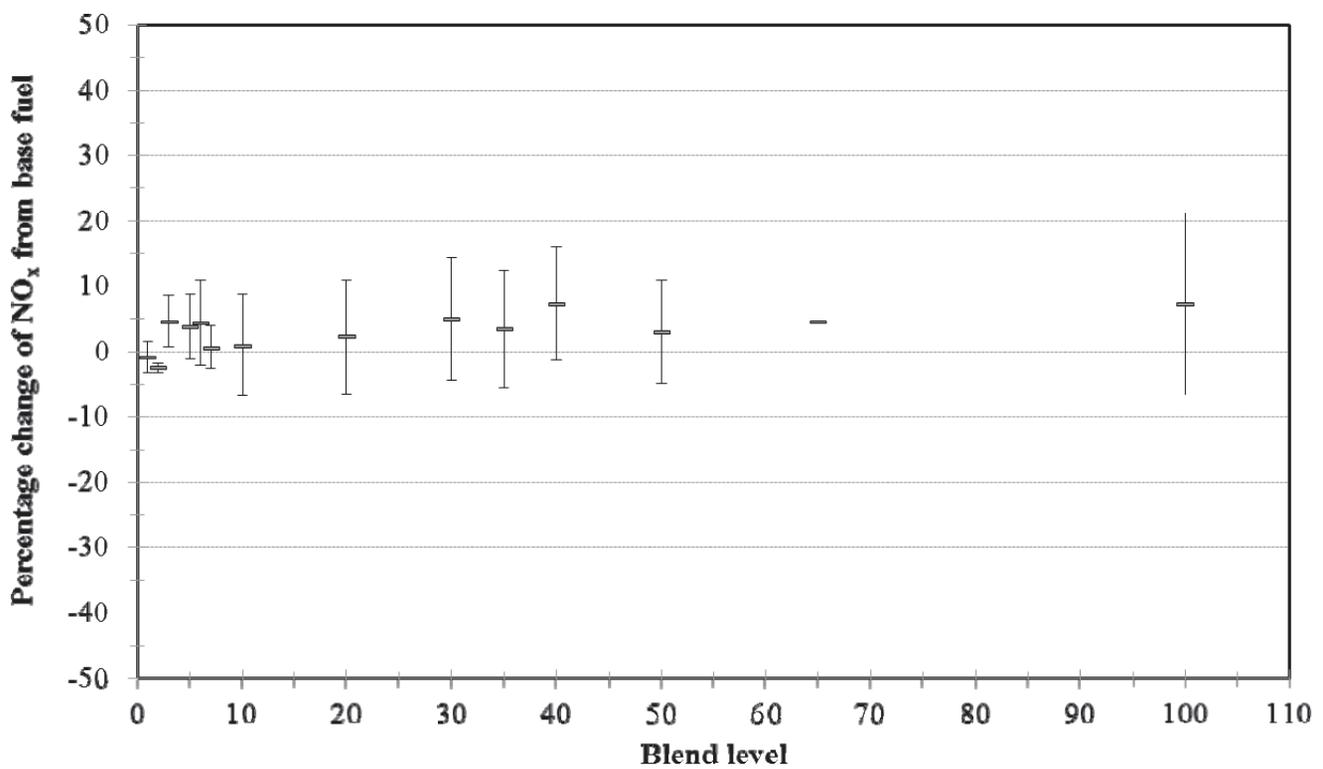


Figure 2 Average values of percentage variations of NO_x emission with standard deviation from tests considered in this review

reacts with HO₂ to form NO₂ [Lissianski et al., 2000]. In several high-temperature combustion processes, the predominant form of NO_x is produced as nitric oxide (NO), while the concentrations of NO₂ are typically less than 5% of total NO_x levels. NO_x abatement techniques require a basic understanding of the kinetics behind the NO_x-forming reactions. Although the NO_x reaction mechanisms have been detailed in the literature, the following section outlines some recent literature pertaining to the various mechanisms contributing to the formation of NO, and the general reactions involved in the formation of NO₂ and N₂O.

2.1 Nitric oxide (NO) emission

The atmospheric nitrogen is the prevalent source of nitrogen for NO production during the combustion of petroleum and FAME (Fatty Acid Methyl Ester)-derived fuels with air as the oxidizer. The thermal (Zeldovich), prompt (Fenimore), N₂O pathway, fuel-bound nitrogen and the NNH mechanism are the identified mechanisms for NO formation in diesel combustion which are summarized below.

2.1.1. Thermal mechanism

Thermal mechanism is the primary route by which NO formation occurs at temperatures typically above 1800K [Hoekman and Robbins,

2012]. At this high temperature, nitrogen (N₂) and oxygen (O₂) react through a set of chemical reactions in which the NO formation rate increases exponentially with temperature. The fundamental kinetic equations for thermal NO formation (R1-R3) along with their reaction kinetic parameters are presented in Table 2. The NO reaction rate is influenced by temperature, residence time and concentrations of nitrogen and oxygen in the combustion environment [Varatharajan and Cheralathan, 2012]. A correlation for the thermal NO emission index in automobile engines was developed by Saravanan et al. [2012]:

where NEI is the NO_x emission index (ppm), D is the density of fuel (kg/m³), C = 1 for loaded condition and C = 0 for unloaded condition, L is the load percentage (%), ID is the ignition delay (ms) and T_f is the flame temperature (K). It was observed that the NO_x emission index

$$NEI = 20956.69 * D * [1 + C[-9.91 * 10^{-3}L^2 + 1.608L - 18.431]] * \exp(2.9 * ID - 19.627) * \exp(4626.44/T_f) \quad (E.1)$$

predicted using this correlation agreed with experimental measurements within 20%.

Table 2 List of reactions with kinetic parameters*

	Reactions	A cm ³ /(gmol-s)	n	E _a / R _u K	Reference
R1	O + N ₂ ↔ NO + N	1.95E+14	0	38367	Loffler et al. [2006]
R2	N + O ₂ ↔ NO + O	6.40E+09	1	3160	Loffler et al. [2006]
R3	N + OH ↔ NO + H	3.80E+13	0	0	Loffler et al. [2006]
R4	CH + N ₂ ↔ HCN + N	4.40E+12	0	11060	Dean and Bozzelli [2000]
R5	CH ₂ + N ₂ ↔ HCN + NH	1.00E+13	0	37240	Dean and Bozzelli [2000]
R6	HCN + OH ↔ CN + H ₂ O	3.90E+06	1.83	5180	Dean and Bozzelli [2000]
R7	CN + O ₂ ↔ NCO + O	1.00E+13	0	0	Dean and Bozzelli [2000]
R8	O + N ₂ + M ↔ N ₂ O + M	4.13E+10	0	7890	Loffler et al. [2006]
R9	N ₂ O + O ↔ NO + NO	6.60E+13	0	13390	Loffler et al. [2006]
R10	O + NNH ↔ NO + NH	5.00E+13	0	0	Loffler et al. [2006]
R11	HO ₂ + NO ↔ NO ₂ + OH	2.20E+12	0	240	Dean and Bozzelli [2000]
R12	O + NO ₂ ↔ NO + O ₂	3.90E+12	0	120	Dean and Bozzelli [2000]
R13	H + NO ₂ ↔ NO + OH	1.30E+14	0	180	Dean and Bozzelli [2000]
R14	NH + NO ↔ N ₂ O + H	3.00E+18	-1.65	720	Dean and Bozzelli [2000]
R15	NCO + NO ↔ N ₂ O + CO	1.40E+18	-1.73	384	Dean and Bozzelli [2000]

*Rate constants are given by, $k = AT^n \exp(-E_a/R_u T)$

2.1.2. Prompt mechanism

Fenimore identified another important pathway resulting in NO formation which is termed as prompt NO mechanism. It is significant in some combustion environments where low-temperature fuel-rich conditions prevail, while the residence time is short [Fenimore, 1971]. Prompt NO is produced when hydrocarbon radicals react with nitrogen to form highly reactive cyano radicals such as HCN in the combustion chamber [Fernando et al., 2006]. Prompt NO is also more sensitive to the fuel chemistry than thermal NO because of the dependence on hydrocarbon radicals. Miller and Bowman [1989] studied the mechanism and modeling of nitrogen chemistry in combustion and concluded that the prompt NO mechanism was important in the NO formation and could not be neglected while estimating the total NO_x production, particularly in a fuel-rich combustion environment. The prompt NO mechanism involves a set of chemical reactions as shown by reactions R4 to R7 in Table 2. Prompt NO formation increases with equivalence ratio due to the availability of CH, CH₂ and HCN radicals that are prevalent in fuel-rich hydrocarbon flames, reach a peak, and decreases because of lack of oxygen. These hydrocarbon and cyano radicals are considered to be the significant contributors of prompt NO.

2.1.3. Fuel NO mechanism

Fuel NO is formed when the fuel-bound nitrogen reacts with excess oxygen during combustion and is negligible for both diesel and biodiesel combustion because of low nitrogen levels in the fuel; biodiesel has a

mean nitrogen concentration of only 0.02%. The presence of nitrogen-containing compounds such as pyridine, pyrrole etc. may also tend to form more fuel NO. This is due to the weaker C-N bond present in these chemical compounds compared to the N-N bond in molecular nitrogen. The fuel NO pathway involves the formation of nitro-radicals such as HCN, NH₂, NH, or CN, which then be oxidized to form NO [Fernando et al., 2006].

2.1.4. N₂O Intermediate mechanism

The intermediate mechanism (N₂O pathway) is another essential mechanism that becomes significant in high-pressure combustion processes [Dean and Bozzelli, 2000]. In this mechanism, the reaction occurs between N₂ and atomic oxygen to form intermediate N₂O by a three-body recombination reaction (R8) where the collision partner M collectively represents all the molecules present in the combustion medium and plays a key role in the execution of this reaction. The N₂O formed in reaction (R9) can then react to form NO.

2.1.5. The NNH mechanism

Under certain combustion regions like flame fronts where atomic concentrations are high, (R10) contributes to NO production. This mechanism requires interaction between hydrogen atoms and molecular nitrogen to form highly reactive NNH radicals which further react with atomic oxygen to form nitric oxide [Dean and Bozzelli, 2000].

2.2 Nitrogen dioxide (NO_2) emission

In the previous section, several reactions that lead to the formation of NO were discussed. Under certain conditions, a significant amount of NO can be converted to NO_2 . The fastest flame reaction forming NO_2 is R11 (Table.2), which is important whenever the concentration of HO_2 is significant, typically in the temperature range of 600 to 1000 K under fuel-lean conditions. However, at higher temperatures HO_2 dissociates quickly into H atoms and O_2 , and the higher prevailing concentrations of H, O and OH lead to more rapid NO_2 loss through the reactions (R12 and R13). This rapid loss of NO_2 is significant and is reflected as a lower NO_2 concentration in the combustion flue gas.

2.3 Nitrous oxide (N_2O) emission

At low combustion temperatures and high pressures, nitrous oxide formation by the ter-molecular reaction (R8) is significant. The N_2O formed in this reaction reacts with oxygen atoms exothermally to form NO (as discussed in section 2.1.4). Various nitrogen-containing radicals contribute to the formation of N_2O as in R14 and R15. However, the N_2O formed in these reactions undergoes further reactions forming NO which is more stable than N_2O .

In a compression ignition engine, the combustion reactions are characterized by high temperature, varying residence time depending on the injection timing, localized fuel rich conditions (though overall fuel lean) and high pressure. These reactions favor NO formation predominantly, in different pathways described above, out of which the thermal and prompt mechanisms are considered to be significant in the biodiesel engine combustion.

3. Experimental evidences of biodiesel impact on NO_x emission from compression-ignition engine exhaust

This section reviews the experimental studies on various aspects of engine operation, fuel properties and combustion processes and their influence on the emission of NO_x measured in the engine exhaust.

3.1. Effects of fuel composition

The fuel composition is a broad factor which comprises of influential parameters such as fuel feedstock, level of unsaturation, biodiesel blend level, oxygen content and cetane number. The following section discusses the effect of these parameters on the NO_x emission from the compression ignition engines.

3.1.1 Biodiesel feedstock / Level of unsaturation

Several studies reported differences in NO_x emission of biodiesels from different feedstocks [Graboski et al., 2003; Saravanan et al., 2009; Saravanan et al., 2010; Hoekman and Robbins, 2012; Varatharajan and Cheralathan, 2012]. Graboski et al. [2003] conducted experiments with neat methyl esters and ethyl esters in a 6- cylinder, 4- stroke, direct injection diesel engine to understand the effect of hydrocarbon chain length on the NO_x formation with saturated methyl esters based on their composition - lauric (C12), palmitic (C16) and stearic (C18) acids and found that esters with shorter carbon chain produced higher NO_x emissions. For example, the specific NO_x emission index of methyl stearate (C18) was about 8% lower than that of methyl laurate (C12). The authors also found a strong linear relationship between increasing NO_x with level of saturation (number of double bonds) in the fuel. For example, the specific NO_x emission index of C18 ester with three double bonds was 16% higher than that of C18 ester with one double bond. Possibly, the double bonds resulted in some pre-combustion chemistry that increased NO_x formation. Finally, the authors concluded that the NO_x emissions increased due to the decrease in average carbon chain length and increase in level of unsaturation; therefore, the most intrinsic way to revamp the emission performance was to alter the fuel chemistry, since molecular structure was the basis of fuel properties such as cetane number, density, boiling point and ignition delay. It is now widely acknowledged that increasing unsaturation and decreasing carbon chain length both lead to an increase in NO_x emission. This information has been developed from the experimental and numerical demonstration in several works [Graboski et al., 2003; Sun et al., 2010; Xue et al., 2011; Hoekman and Robbins, 2012; Varatharajan and Cheralathan, 2012; Palash et al., 2013] involving pure FAME components as well as practical biodiesel fuels employing a wide variety of engines and test cycles. Lin et al. [2009] found that palm oil methyl ester (POME) and palm kernel oil methyl ester (PKOME) resulted in a lower increase (15% and 5% respectively) in NO_x concentration (ppm) and a significant reduction (59% and 73% respectively) in smoke emissions than petroleum diesel and vegetable oil

methyl ester (VOME) fuels (soy methyl ester, peanut oil methyl ester, corn oil methyl ester, sunflower oil methyl ester, rapeseed methyl ester and waste fried oil methyl ester). The authors attributed this behavior to the more saturated carbon bonds of POME (about 50%) and PKOME (about 80%), when compared to other VOME fuels, and suggested that a fuel with more saturated carbon bonds would result in reduced NO_x emissions. The exact pathways by which the fuel composition effects influence NO_x are still ambiguous, but these parameters are recognized to critically influence the observed NO_x emissions.

3.1.2 Biodiesel content

Many publications in the literature suggest that NO_x emission increased with the biodiesel content in the blend. Lertsathapornasuk et al. [2008] noted that the NO_x emission index (g/kW-h) increased about 12% and 3% for neat palm biodiesel (B100) and B50 than diesel at 25% load condition; and an increase of about 26% and 9% in NO_x emission index for B100 and B50 while maintained the engine speed at 1500 rpm. A study by Luján et al. [2009] on high speed direct injection, 4-cylinder, diesel engine with high pressure common-rail fuel injection system during the standardized MVEG-A cycle (European Motor Vehicle Emission Group A) simulated the road load conditions. It was fueled by biodiesel (from vegetable oil) and its blends B30, B50 and B100. The authors observed that the increase in NO_x concentration (ppm) for B30, B50 and B100, compared to the base diesel fuel, was 21%, 26% and 45%, respectively. Similar trend was observed by Buyukkaya [2010] with rapeseed methyl ester/ diesel blends. Gumus and Kasifoglu [2010] tested three blends of apricot seed kernel oil methyl ester (B5, B20 and B50) with diesel fuel in a compression ignition engine and found a monotonic increase in NO_x concentration with the biodiesel content. They attributed this behavior to the fuel-bound oxygen of the biodiesel.

On the other hand, Kalligeros et al. [2003] found a monotonic decreasing trend of NO_x concentration in the biodiesel blends containing 10%, 20%, and 50% of two types of methyl esters from sunflower oil and olive oil, in a single-cylinder diesel engine. At 3.80 kW load, B10, B20 and B50 of sunflower oil methyl ester with marine diesel produced about 3%, 5% and 6% lower NO_x concentration, while B10, B20 and B50 of olive oil methyl ester produced about 8%, 14% and 14.5% lower NO_x concentration than marine diesel. The variation in the NO_x reduction potential between the different biodiesel feedstock was attributed to the difference in cetane number of biodiesels; it was hypothesized that the higher cetane number of olive oil methyl ester (CN: 61) than that of sunflower oil methyl ester (CN: 58) resulted in increased reduction of NO_x concentration; the cetane index of marine diesel was reported as 46.

Interestingly, some other publications in the literature showed that the NO_x emissions varied non-monotonically with the biodiesel content in the blend [Labeckas and Slavinskas, 2006; Lin et al., 2007; Pereira et al., 2007; Fontaras et al., 2009; Aydin and Bayindir, 2010]. The authors attributed these variations to the differences in the degree of evaporation and the combustion processes in the engine, since the calibration settings of the conventional diesel engine could have been not ideal for all proportion of biodiesel blends at all operating regimes. A non-monotonic NO_x variation was observed in a water-cooled direct injection diesel engine fueled with polanga methyl ester and its blends (20, 40, 60, and 80%) with diesel [Sahoo et al., 2007]. The NO_x concentration from B20 was found to be 2% higher than diesel while neat biodiesel produced 4% lower NO_x . The authors attributed the variations to the differences in compression ratio, residence time and temperature distribution among the tested fuels. Biodiesels derived from jatropha, karanja and polanga oil and their blends (B20, B50 and B100) were tested in a 3 cylinder compression ignition engine [Sahoo et al., 2009]. It was observed that the karanja and polanga biodiesels and their blends had a monotonically increasing NO_x with the biodiesel content (with peak NO_x occurred with neat biodiesels; KB100 and PB100 produced about 15% and 21% higher NO_x (g/kW-h) than that of diesel), while a non-monotonicity was observed in NO_x with jatropha oil biodiesel. JB20 showed a 20% increase in brake specific NO_x emission index (g/kW-h) compared to baseline diesel, whereas JB50 and JB100 showed an increase of 15% and 17% respectively when compared to diesel. Several of these studies suggested that it was not appropriate to linearize the biodiesel blending effect on NO_x emissions due to the varied operating conditions, engine calibration and combustion phases.

3.1.3 Oxygen content

The fuel-bound oxygen content of the blend is proportional to the volumetric concentration of biodiesel in the blend. Generally, the increased NO_x emissions in biodiesel and their blends are attributed to the higher

oxygen content in the biodiesel [Godiganur et al., 2010; Gumus and Kasifoglu, 2010] due to a linear increasing trend of NO_x with the increase in mass percentage of fuel oxygen. However, there are studies that do not agree with this linear increase; a linear decrease [Kalligeros et al., 2003] in NO_x emission or a non-monotonic increase or decrease with the oxygen content in the fuel [Sahoo et al., 2007; Fontaras et al., 2009; Aydin and Bayindir, 2010] have been documented.

Canakci [2005] studied No. 2 diesel fuel (no fuel bound oxygen), No. 1 diesel fuel (no fuel-bound oxygen), SME (11% oxygen by mass / 3.6% oxygen by volume) and B20 (20% SME and 80% No.2 diesel) (2.2 % oxygen by mass / 0.7 % by volume) in a turbocharged diesel engine and observed the brake specific NO_x index (g/kWh) of the SME and B20 blend were increased by 11% and 1%, respectively compared to the No. 2 diesel. Surprisingly, the NO_x emission index of No. 1 diesel fuel was 6% lower than that of No. 2 diesel fuel, while there was no appreciable difference of oxygen concentration in the exhaust between the fuels tested. Therefore, in addition to the fuel-bound oxygen effect of biodiesel, more research is required to identify the properties that impact the combustion reactions favoring NO_x emission. Puhan et al. [2005] questioned the availability of fuel-bound oxygen for NO_x formation and argued that the esters might decarboxylate and form CO_2 early during the combustion. It is evident that additional studies are essential to understand the significance of fuel-bound oxygen effect, especially their impact in the formation of NO_x during combustion in engines.

3.1.4 Effects of cetane and iodine numbers

Auto-ignition of the injected fuel is a critical factor in the performance and operation of compression ignition engines. Cetane number is a property of the fuel that quantifies the self-ignition characteristics and ignition delay time of a fuel in an engine cycle; the higher the cetane number, the shorter the ignition delay [Pulkrabek, 2004]. Cetane numbers for various esters of the saturated fatty acids ranging from C8 to C18 have been determined according to ASTM D-613. For methyl esters, the cetane numbers were found to increase nonlinearly with the fatty acid chain length [Klopfenstein, 1985]. The higher cetane number of biodiesel reduces the ignition delay and also leads to the advancement in combustion. Consequently, with the availability of oxygen, higher temperature together with longer residence time, may lead to the increase of NO_x emissions. Several researchers, e.g., Lim et al. [2014] used this reasoning to describe the increased NO_x emissions with biodiesel content. However, this reasoning is debatable. Higher cetane number results in not only an early onset of combustion, but also leads to lower amount of fuel burning in the premixed-combustion mode, which result in lower temperature and residence time in the combustion chamber, causing a lower NO_x formation. Wu et al. [2009] observed that the brake specific NO_x emission index (g/kWh) of palm methyl ester (PME) was 7% lower than that of waste oil methyl ester (WME), even though both had almost the same oxygen content of about 11.2% by mass. This was credited to the higher cetane number of PME (64 for PME compared to 56 for WME) which could reduce ignition delay and the amount of fuel consumed in the premixed phase, resulting in a reduction of in-cylinder temperature and subsequently reduced NO_x emission. Many authors [Graboski et al., 2003; Kalligeros et al., 2003; Puhan et al., 2005; Knothe et al., 2006; Karavalakis et al., 2009] concluded that NO_x emissions decreased with an increase in cetane number.

3.2. Influence of type of engine and test cycle

Engine type and its test cycles could strongly influence the NO_x emission of biodiesel. Tat [2003] demonstrated that the NO_x emission from compression ignition engines was significantly influenced by the variation in injection timing and advance due to the impact of fluid properties such as density, isentropic bulk modulus (compressibility) and viscosity. Karavalakis et al. [2009] studied the exhaust emission characteristics of ultra-low sulphur diesel and soy biodiesel blends at proportions of 10% and 30% by volume in a Euro 4 common rail injection diesel engine over various engine test cycles namely New European Driving Cycle (NEDC) and non-legislated Artemis driving cycles which simulate urban, rural and highway driving conditions in Europe. It is interesting to note that driving test cycles significantly influence the amount of NO_x emitted during the operation. Among the three Artemis driving cycle conditions, B10 and B30 produced higher NO_x (g/km) than the base line diesel. During rural driving condition, B10 and B30 produced the highest increase in NO_x as 14% and 18% respectively. However, in NEDC test cycle, NO_x emissions were slightly reduced for both B10 and B30 by 3% and 4% respectively. The authors attributed this observed reduction in NO_x to the smooth acceleration profile of NEDC, which dominated the NO_x emission

mechanism than the physicochemical characteristics of biodiesel. Moreover, the Artemis driving cycles were more aggressive and transient, which would inherently favor the increase of NO_x emissions. Hence, although the physicochemical properties and fuel chemistry properties such as saturation level and cetane number play certain roles in the NO_x emissions, their relative importance when compared to the engine parameters and the nature of test cycle in certain conditions are inconclusive. As a part of the program to quantify the effect of biodiesel fuels on engine NO_x emissions, EPA [2010] examined chassis dynamometer tests on heavy duty diesel engines to study the NO_x effects of soy-based biodiesel over light, medium and heavy duty cycles. The results indicated that brake specific NO_x index increased as a function of average cycle load, by about 5% for high loaded cycles; 2.5% increase in medium loaded cycles. However, in lightly loaded cycle, results showed inconclusive trends in NO_x emissions (where a decrease and increase in NO_x emissions was observed by 1% and 2% respectively in two different trials), and hence the load-dependent nature of test cycles and their influence on NO_x emissions could not be neglected. Osborne et al. [2011] tested soy biodiesel, diesel and their blends (B2, B10, B20) in a locomotive operated in two different cycles namely line haul and switch cycle. The changes in cycle weighted average of NO_x (g/kWh) for B2, B10 and B20 were comparable between two cycles, while B100 in switch cycle increased NO_x by about 15% over the line haul cycle. Fontaras et al. [2014] studied rapeseed methyl ester and its blends B10, B20 and B50 with diesel in three different vehicles (equipped with different exhaust after treatment technologies) over test cycles namely NEDC, which is a standard test cycle in Europe and real world testing cycles such as Artemis Urban and Artemis Road. In general, among all the tested conditions, NO_x (g/km) increased up to 20% with B50 in some cases while most showed an increase in the range of 1 to 10% depending on the blend and the vehicle. Serrano et al. [2015] tested diesel and 20% biodiesel blend (from soy and palm feedstock) in three different test cycles, namely NEDC, URBAN and EXTRA URBAN test cycle. In all three cycles, B20 had lower NO_x emission index (g/km) than diesel, but the reduction percentage varied with cycles as 10%, 20% and 5% respectively. In summary, all these studies reported different trends with different combination of engine test cycles, biodiesel feedstock and content. Hence the biodiesel and its blending effect on NO_x are inconclusive in terms of engine test cycle since several engine and combustion parameters influence the emission characteristics of a particular test cycle which can overshadow the actual emission potential of a particular fuel or fuel blend.

3.3. Effect of injection timing

The fuel injection is an important process in the engine operation and the timing of fuel injection into the combustion chamber is critical and affects performance and emissions to a large extent. The injection process is greatly influenced by the fluid dynamic properties of the fuel and NO_x formation appeared to be dependent on the start of injection timing in compression ignition engines. [Tat et al., 2000; Tat and Van Gerpen, 2003; Boehman et al., 2004; Agarwal et al., 2013]. Carraretto et al. [2004] studied the effect of advance in injection timing (at three crank angles namely, 21°, 24° and 27°) with neat biodiesel produced from vegetable oil in a 4-stroke diesel engine. They observed that NO_x concentration (ppm) increased with the injection advance by about 41% between 21° and 24° and by about 67% between 24° and 27° at both maximum torque and power speeds. Tsolakis et al. [2007] observed that the retarded injection timing by 3° reduced NO_x concentration (ppm) while testing with rapeseed methyl ester (by about 17%) and its 50% blend (by about 23%) with diesel in a diesel engine. Szybist et al. [2007] studied the influence of methyl oleate (mono-unsaturated fatty compound) present in soy biodiesel. The experiments were conducted at three different injection timings - early, mid and late. In early injection mode, where more premixed burn fractions occur, neat soy biodiesel and B20 produced about 11% and 3% more brake-specific NO_x (g/kWh) than ULSD. As the fuel injection timing was retarded, in mid and late injection modes, the differences in NO_x emissions between the fuels were reduced. In late injection mode, where more diffusion burn fractions occur, the NO_x emitted from all tested fuels were comparable. Hence, the authors suggested that retarding the injection timing was a potential way of reducing NO_x emissions. In agreement with this claim, a reduction in brake-specific NO_x emission index was observed with the retarded start of combustion (SOC) timing for SME/diesel blends [Moscherosch et al., 2010] and CME/diesel blends [Sequera et al., 2011].

3.4. Effect of engine speed

Engine speed also plays a critical role in the formation of NO_x in

compression ignition engines. Several studies proposed that NO_x emissions decreased with engine speed [Lin and Li, 2009; Imtenan et al., 2014]. The NO_x concentration (ppm) decreased by about 23% between engine speeds of 800 rpm and 2000 rpm for petroleum diesel and biodiesels from cooking oil and marine fish oil in a 4-stroke direct injection, naturally aspirated diesel engine [Lin and Li, 2009]. The authors concluded that, although the increased engine speed caused an increase in the temperature and pressure of burning gas, the reduction of ignition delay resulted in the reduction of residence time available for NO_x formation. Interestingly, different trends of NO_x emission was reported at two different engine speeds (2000 and 4000 rpm) at the full load condition when diesel, biodiesel from waste cooking oil and their blends of 10%, 20%, 40% and 60% (by volume) were studied in a common rail Euro 3 diesel engine [Zhang et al., 2008]. At 2000 rpm, a monotonic increase of NO_x concentration (ppm) was observed with the biodiesel content in the blend, especially with blends higher than 40% of biodiesel content. B100 had about 13% higher NO_x concentration than the diesel. However, at 4000 rpm, the NO_x concentration did not show any variation with biodiesel content. The authors attributed the observed trends to the common rail injection system where the impact of advanced injection of biodiesel due to higher bulk modulus, density and mechanical pump is no longer a significant factor.

Moreover, a non-monotonic variation of NO_x was observed with engine speeds in some studies. Usta [2005] observed different effects of engine speed on NO_x concentration (ppm) at different conditions, that is, as engine speed was increased (between 1500 rpm and 3000 rpm), the NO_x concentration increased by about 74% at full load, and increased by about 33% at three fourth of rated load, but gradually decreased by 28% at half load for both diesel and its blend (D82.5/TSOME17.5, by volume) with tobacco seed oil methyl ester. Non-monotonic variation of NO_x was also reported in several other studies which employed biodiesels different feedstocks ranging from waste frying oil [Utlu and Kocak, 2008], tall oil [Keskin et al., 2008], vegetable oil [Chokri et al., 2012], cooking oil [Arslan, 2011], calophyllum inophyllum linn oil which consists of mostly unsaturated fatty acids [Fattah et al., 2014]. A limited source of explanation has been reported in the literature on the engine speed effect on NO_x formation. Several studies attributed various parameters such as availability of oxygen, in-cylinder combustion temperature and residence time to the variation of NO_x between fuels; but no explanation was given to the non-monotonic effect of engine speed on NO_x emissions.

3.5. Effect of engine load

The biodiesel NO_x effect was significantly influenced by engine load when tested with biodiesel and its blends with diesel in diesel engines. Several studies reported a monotonic increase in NO_x with engine load due to higher temperature generated at higher engine load [Zhang and Boehman, 2007], while employing biodiesel blends from different feedstock as Neem oil [Sharma et al., 2009], Mahua and Fish oil [Godiganur et al., 2010], Jatropha oil [Tan et al., 2012; Padhee and Raheman, 2015] and Croton oil [Osawa et al., 2015]. The authors attributed this monotonic increase of NO_x emissions to the increased engine temperature and pressure, availability of oxygen and increased flow rate of the biodiesel blends.

However, Murillo et al. [2007] found a surprising decrease in NO_x emissions with load in a single-cylinder, naturally aspirated direct injection diesel engine. A substantial reduction of about 60% in the specific NO_x emission (g/kWh) between 25% load and full load when tested with B100 was observed. The authors attributed this trend to the increase in turbulence inside the cylinder, which contributed to a quicker combustion and resulted in lower residence time of the species in the high temperature zones. Agarwal and Rajamanoharan [2009] tested karanja biodiesel and blends (B10, B20, B50 and B75) with diesel in a single cylinder agricultural engine. The authors observed that the neat biodiesel and all the tested blends had comparable NO emission index (g/kWh), while they were about 45% lower than that of diesel at 20% load.

On the other hand, a non-monotonic response of NO_x for the use of biodiesels and their blends with diesel has been widely reported in the literature [Raheman and Phadatar, 2004; Sureshkumar et al., 2008; Dhar et al., 2012; Agarwal and Dhar, 2013; Chavan et al 2015]. A non-monotonic variation of NO_x with the biodiesel content over a wide range of loads was observed by Raheman and Phadatar [2004] while testing karanja methyl ester and its blends (B20, B40, B60, B80, by volume) with diesel in a single cylinder, 4-stroke, water-cooled direct injection diesel engine. At full load condition, B20 and B80 produced about 23% lower NO_x concentration (ppm) than diesel; while B60 and B100 produced about 38% lower NO_x than diesel and B40 recorded about 15

% lower NO_x than diesel. Hence, it is clear that NO_x is sensitive to load where changes in the loading conditions could possibly reverse the NO_x formation trends. Sureshkumar et al. [2008] also observed a non-monotonic variation in NO_x concentration with biodiesel content for a wide range of loading conditions while testing pongamia pinnata methyl ester and its blends (B20, B40, B60, B80, by volume) in a similar engine. At 75% load condition, B20 and B100 produced 8% and 25% lower NO_x concentration (ppm) than diesel; while B40, B60 and B80 produced about 38% lower NO_x than diesel. Though the NO_x concentration was decreased with the biodiesel content in the blend, the extent of reduction in NO_x was not proportional with the biodiesel content. Agarwal and Dhar [2013] tested karanja biodiesel and blends (B10, B50) with mineral diesel in a direct injection diesel engine. It was observed that B50 and B20 consistently produced higher NO (g/kWh) on an average of about three times than diesel, while B10 and B100 produced comparable or slightly higher NO_x than diesel. Chavan et al. [2015] studied the emission characteristics of jatropha biodiesel and its blends B10, B20 and B30 with petroleum diesel in a 4-stroke variable compression ratio engine at five different compression ratios (CRs) namely 14, 15, 16, 17 and 18 over a range of loads. It was observed that NO_x concentration (ppm) increased with increasing load and compression ratio. It may be due to the increase in temperature at high loads and lower ignition delay due to higher compression ratio that would result in increased pressure and temperature inside the cylinder. However the NO_x emission due to biodiesel and blending effect had neither a monotonic increase nor a monotonic decrease with load and CRs. For example at full load, diesel had lowest NO_x concentration (ppm) at CR 14 and CR 16; B100 had lowest NO_x concentration at CR 15; B30 had lowest NO_x concentration at CR 18 while diesel and B30 had lowest NO_x concentration at CR 17. Similarly, a non-monotonic trend was observed with different loads at a given compression ratio. Hence, the engine parameters and the associated phasing of combustion complicate the understanding of already entangled biodiesel and its blending effect on NO_x formation in compression ignition engines.

3.6. Fluid dynamics effects

Fluid dynamics of the fuel spray is an important and highly complex phenomenon that significantly influences the phasing of combustion [Sirignano, 1993]. The fuel spray characteristics such as injector penetration length, atomization and mean droplet size along with flow field and heat transfer interactions between droplets are critically influenced by various physical properties of the fuel. Especially, the differences in properties such as density, viscosity, surface tension, etc., between biodiesel and petroleum fuels influence the combustion process and can affect NO_x emission [Allen and Watts, 2000; Lee et al., 2005; Ejim et al., 2007; Suñ et al., 2007; Yuan et al., 2007]. The fuel viscosity and surface tension of fifteen neat biodiesels were reported in [Allen and Watts, 2000] using a regression model developed based on experimental results from five different biodiesels. The authors claimed that viscosity and surface tension, in turn the Sauter mean diameter (SMD) of the spray, could be predicted from the fatty acid composition of biodiesels. A maximum reduction of 50% in viscosity and 8% in surface tension between rapeseed methyl ester and coconut oil methyl ester was predicted and was attributed to the differences in carbon chain lengths; major fatty acid constituents in coconut oil methyl ester have shorter carbon chain length while rapeseed methyl ester has constituents with longer carbon chain length. The SMD of coconut oil methyl ester spray was comparable to that of petroleum diesel spray, also confirmed by Ejim et al. [2007], while rapeseed methyl ester spray had 40% higher SMD than that of diesel spray; other biodiesel sprays have about 25% - 29% higher SMDs than diesel fuel spray. It is also indicated that the discrepancies in the reported data among literature sources could not be verified since most of the studies did not report the extent of the reaction and the presence of triglycerides in the fuel after the transesterification process. The authors concluded that even a 6% by mass of triglyceride (canola oil) in the canola methyl ester could result in a 12% increase in viscosity of the biodiesel. The biodiesel blending effects on the viscosity and surface tension of the final blended fuel was studied by Lee et al. [2005] in a common-rail diesel engine with soy biodiesel and its 10%, 20% and 40% volumetric blends with petroleum diesel. It was observed that the kinematic viscosity and surface tension of the fuel linearly increased with the biodiesel content; however, the blending ratio had a minimal effect on the spray development. Lower injection velocity of biodiesel due to higher viscosity, and the associated increase in friction between biodiesel spray and nozzle surface causes shorter spray tip penetration; while higher SMD of the biodiesel spray causes a longer tip penetration. These two compensating effects resulted in a similar spray tip

penetration between diesel, biodiesel and their blends. Ejim et al. [2007] reassessed the findings by Allen and Watts [2000] and reported comparable SMDs among neat palm, soybean, cotton seed, peanut and canola biodiesel and their corresponding B5 and B20 blends with No.2 diesel. In a computational study by Yuan and group [2007], for a given engine speed and load, the maximum spray cone angle of diesel and soy biodiesel was found to be 47.5° and 30° and soy biodiesel produced about 8% higher brake-specific NO_x than diesel. When the spray cone angle of soy biodiesel was matched with diesel (47.5°), the brake-specific NO_x was reduced by 15% between cone angles of 30° and 47.5° of soy biodiesel spray. The authors concluded that a narrow spray angle could significantly emit higher NO_x since the narrow sprays induce strong stratification of fuel vapor which brings about local rich or stoichiometric regions that contribute to higher NO_x. Yuan and Hansen [2009] predicted that NO_x (g/kWh) decreased up to 3.5% when the viscosity of SME was decreased to match with petroleum diesel fuel. They also suggested that smaller spray cone angles and advanced start of injection were the main reasons for increased NO_x emission of biodiesel. They concluded that the decreased spray cone angle and increased spray penetration might increase NO_x emission. Agarwal and Chaudhury [2012] investigated the spray characteristics in a constant volume spray chamber with diesel, karanja biodiesel and their blends, B5 and B20, and concluded that B100 had highest spray tip penetration, cone angle and spray area followed by B20, B5 and diesel.

In addition to density, surface tension and viscosity, the boiling point of biodiesel could also significantly alter the spray characteristics; higher boiling point of biodiesel increases the combustion duration and cylinder gas temperature both of which could favor NO formation [Oszezen et al., 2008]. In a recent study, the spray, combustion and exhaust emission characteristics of soy biodiesel in a direct injection common-rail diesel engine were investigated by Yoon et al. [2009] who observed that biodiesel produced larger droplet size (about 12%), similar spray structure, and longer spray tip penetration (about 8% higher) into the cylinder than conventional diesel, which resulted in an increased indicated specific NO_x emissions of soy biodiesel by about 19% higher than that of diesel. Ye and Boehman [2010] studied the effect of engine injection strategies on the biodiesel NO_x effect with a direct injection diesel engine fueled with ultra-low sulphur diesel and its blend with soy biodiesel (B40). For a given speed and load, a higher volume of biodiesel has to be supplied because of its lower heating value compared to petroleum diesel. The increase of fuel consumption could be accomplished by either higher injection pressure or injection duration, which would result in increased NO_x emission. Injection characteristics such as mean injection rate, mean injection pressure, injection delay and injection duration increase with the biodiesel content, which could favor NO_x formation at certain conditions. Hence, the above discussed physical properties and their effects on the fluid dynamics of the fuel spray and atomization have critical impact on the combustion and emission characteristics of a fuel at any particular operating condition.

4. Conclusions

In this article, the key parameters that influence NO_x emissions in diesel engines fuelled with biodiesel blends were identified and broadly classified as fuel chemistry effects, engine control effects and fluid dynamics effects. In most engine studies, with a few exceptions, biodiesel or biodiesel blends resulted in increased NO_x emission compared to baseline diesel; however, *the change in NO_x emission for blends varied non-monotonically* with the biodiesel content due to a complex dependence of NO_x formation on various factors at various conditions. The relative significance and extent of influence of various physico-chemical factors reviewed in this article differ with engine type, cycle and operating conditions. Hence, a holistic approach is needed to investigate the biodiesel blending effect on NO_x emission by discerning the influential factors and then integrating the appropriate individual findings along with the corresponding coupled effects of thermo-fluid-chemical interactions for the particular combustion environment.

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Table 3 Physical and Chemical properties of various methyl esters of fatty acids*

Biodiesel Type	Density kg/m ³	Kine-matic Viscosity at 40°C mm ² /s	Surface Tension mN/m	Iodine value	Cetane number	Boiling point K	Flash point K	Pour point K	Heating value MJ/kg	Saturated fatty acids %w	C %m	H %m	O %m	S %m
Canola	912	4.02	27.21	110-120	52	607-623	519		39.7	7.0	77.00	12.2	10.8	
Coconut	918	2.73	24.01	8-10	68		410	269	36.9	81.5				
Corn	910	4.36	28.86	103-128	55	620-649	550		39.5	12.7	76.31	11.14	10.9	0.0003
Cottonseed	915	4.00	26.81	90-119	51	617-652	483	258-279	39.4	23.8			10.6	0.000005
Fish oil	860	7.20			51				41.4	37.3	80.01	12.75	7.2	
Jatropha	940	4.80	30.10	95-105	52	> 633	421	277	38.7	26.2	76.22	12.49	11.29	0.000011
Linseed	924	3.75		168-204	55		514		39.3	7.9				
Mahua	882	3.98		88	51	563-583	471	275	39.4	46.2				
Neem	919	5.21		65-80	51				39.6	39.6				
Olive	880	4.50	28.55	75-94	57		383	270	32.8	20.2				0.001
Palm	879	4.95	26.16	44-58	56	575-622	408	287-289	39.9	45.6	75.66	12.17	11.3	0.00005
Peanut	903	5.25	26.93	84-100	54		466		39.7	20.8	75.39	11.98	11.0	0.0011
Polanga	869	3.99			54		413	277	41.4					
Karanja	936	4.37		117	51		436	278	42.1	29.2	76.83	11.83	11.34	0.000009
Rapeseed	885	4.59	27.82	94-120	54	638-647	420-443	261	39.9	4.34	76.05	12.14	10.8	0.000006
Rice-bran	919	4.96		99-108	51					14.2-21.1				
Soybean	885	4.08	27.15	117-143	46	616-625	414-440	266-272	39.7	15.22	77.00	12.18	10.8	<0.005
Sunflower	885	4.22	28.96	110-143	47	673-657	383	270	39.9	9.34	76.38	11.3	10.9	0.0004
Tallow		4.82	28.39	54	59	610-613				47-63				
Yellow grease	872	5.92		80-100	63				40.1	36.51	76.66	12.33	11.0	<0.005

*Kalligeros et al., 2003; Yuan et al., 2005; Ejim et al., 2007; Sahoo et al., 2007; Benjumea et al., 2008; Sureshkumar et al., 2008; Lin and Li, 2009; Lin et al., 2009; Oszezen et al., 2009; Sahoo et al., 2009; Sanin et al., 2009; Wu et al., 2009; Yuan and Hansen, 2009; Karmakar et al., 2010; Pugazhavadu and Sankarnarayanan, 2010; Varatharajan and Cheralathan, 2012; Chhetri and Watts, 2013; Fattah et al., 2013; Palash et al., 2013; Sonar et al., 2015; Arbab Iqbal et al., 2015; Mistri et al., 2016.

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