A statistical investigation of biodiesel effects on regulated exhaust emissions during transient cycles

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Evangelos G. Giakoumis[*]

Internal Combustion Engines Laboratory, Thermal Engineering Department,
School of Mechanical Engineering, National Technical University of Athens, Athens, Greece

ABSTRACT

In the present work, a statistical investigation is conducted in order to quantify the effects of biodiesel blending on the regulated exhaust emissions. To this aim, the available literature on biodiesel emissions during transient/driving cycles up to the end of 2011 was gathered and the reported measurements were statistically analyzed with respect to the biodiesel percentage in the fuel blend. From the analysis, collective results, statistical data and best-fit quadratic regression curves are derived based on the emission measurements from all driving cycles. Furthermore, the effects of engine type (heavy or light-duty), dynamometer schedule (chassis or engine), engine model year and biodiesel feedstock are deducted, with separate best-fit curves provided for each case and for each exhaust pollutant. The various trends observed are discussed and explained based on fundamental aspects of diesel engine combustion and emissions. It is believed that the results of this study can prove useful to administrations and international institutions by providing a good estimate of the vehicle fleet's expected emission changes when running on biodiesel blends over neat diesel, enabling long-term planning and accommodating decision making.

[*] Tel.: +30 210 772 1360; fax: +30 210 772 1343.
E-mail address: vgiakms@central.ntua.gr (E.G. Giakoumis)

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1. Introduction

Diminishing reserves and growing prices of crude oil are detrimentally affecting both the energy security of the non-oil producing countries and the world economy in general. These facts, coupled with the rising concern over global warming and environmental degradation have led to a considerable effort to develop alternative fuel sources, particularly for the transportation sector, with principal emphasis on biofuels that possess the pivotal advantage of being renewable [1]. To this aim, the European Parliament passed Directive 2009/28/EC [2] on the promotion of the use of energy from renewable sources that contains a specific mandate for Member States to include 10% (by energy content) of renewable fuel in the transport sector by 2020. In parallel in the US, the Energy Independence and Security Act of 2007 (EISA) increased the volume of renewable fuel required to be blended into transportation fuel from 34 billion liters in 2008 to 136 billion liters by 2022.

Biofuels made from agricultural products (oxygenated by nature) reduce the world's dependence on oil imports, support local agricultural industries and enhance farming incomes, while offering serious benefits in terms of sustainability, reduced emissions and increased energy and economic security. Among these, biodiesel (methyl or ethyl ester) is considered as a very promising fuel for automotive and truck engines, since it possesses similar properties with diesel fuel and can also be blended with diesel practically at any proportion and without modifications in the distribution infrastructure. Biodiesel is produced by transesterification of edible or non-edible vegetable oils, animal fats or recycled cooking oils, and consists of long-chain alkyl esters containing two oxygen atoms per molecule [3]. The more widely used biodiesels are rapeseed methyl ester (RME) in Europe and soybean methyl ester or methyl soyate (SME) in the US; other popular biodiesels are palm (mainly in Asia), sunflower, cottonseed, grease and tallow methyl esters, collectively known as fatty acid methyl esters (FAME).

The major biodiesel advantage relative to diesel fuel is its renewability. Life-cycle analyses have shown that the source-to-wheel CO_2 emissions from neat biodiesel combustion account for at least 60% savings with respect to petroleum diesel fuel, whereas for the most popular B20 blend it is of the order of 15−20% [2] This is an extremely hopeful fact in view of the increasing global warming contribution from the transportation sector, but other issues should be taken into account, such as food prices and biodiversity; not surprisingly, concerns over the latter have sparked the research on second-generation biodiesels [4]. It is well established today that biodiesel-blended fuels succeed to a large extent in reducing the amount of emitted PM or the smokiness from diesel engines. In general, similar positive effects have been noticed as regards hydrocarbon (HC) and carbon monoxide (CO) emissions (although contradicting results have been reported too), whereas a usually moderate negative impact is experienced with regard to nitrogen oxides (NO_x) [5,6].

In 2002, the US Environmental Protection Agency (EPA) published a comprehensive analysis of biodiesel impacts on exhaust emissions, where the available at the time amount of emissions data was collected and analyzed in order to quantify the effects of biodiesel blends on all regulated pollutants [7]. Although that work primarily concerned the US market, and at that time the vast majority of the investigations were from heavy-duty engines running on SME blends, the derived best-fit curves have since been used by researchers all over the world in order to demonstrate or even predict the expected emission benefit or penalty when biodiesel is added into the fuel blend. It is felt that it is time to update those emission predictions based on the following observations:

a) The EPA database included (with few notable exceptions) north-American and mostly heavy-duty engines, hence the effect of light-duty and European or Japanese vehicles was minimal;
b) Likewise, the majority of the data concerned the FTP cycle, from which a large percentage its ‘hot’ version, hence the effects of chassis-dynamometer and cold-started vehicular driving cycles were not adequately represented;
c) A significant percentage of the data concerned the, now almost obsolete, two-stroke truck engines;
d) 50% of the engines in the database were model year (MY) 1991−1993 (recall that in 1992 the EU launched the Euro 2 emission level), 29% of the engines were manufactured prior to 1990, whereas only 2% were post-1998 MY, hence the effect of modern antipollution systems such as EGR was practically absent; moreover, almost half of the data concerned one engine manufacturer;

e) More than 80% of these (north-American) investigations were conducted with soybean methyl ester, which did not allow the effects of other biodiesel feedstocks to be revealed; and

f) The reference diesel fuel during the 1980s and 1990s contained considerably higher amounts of sulfur, which is suspected to have increased the biodiesel PM benefit, particularly for high blends ratios, with respect to current regulations.

The target of the present work is to gather the regulated emission results from all studies conducted so far from engines running on various biodiesel blends during transient/driving cycles (i.e. truly transient conditions), to compare the available data and, where possible, identify trends that may exist in order to:

- quantify the biodiesel benefit or liability on all regulated exhaust emissions (primarily PM and NO_x but also CO, HC), and
- investigate the effect of engine type and characteristics (heavy or light duty, MY, EGR) or feedstock-related issues.

Moreover, and this is rather novel in such statistical analyses, an attempt will be made to discuss and explain the various trends observed based on fundamental aspects of diesel engine combustion and emissions mechanisms.

2. Methodology

Whereas for steady-state engine trials, each run is practically different from the other, the results during a transient/driving cycle (either engine or chassis dynamometer) are directly comparable owing to the common procedure and methodology applied. Moreover, transient cycles are inherently better suited to disclose the true (real-world) engine emission pattern of turbocharged diesel engines by incorporating some or all of the following driving conditions

- cold[1] and hot starting,
- frequent accelerations and decelerations,

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[1] It should be mentioned that, although the average temperatures in Middle Europe range between 7^o–11^oC, the NEDC cycle is normally performed at higher temperatures, of the order of 20^oC. Hence, the effect of realistically cold ambient temperatures is not fully incorporated in the transient test.

- changes of load,
- idling conditions typical of urban driving,
- sub-urban or rural driving schedule, and
- motorway driving.

By applying a transient cycle for the testing of new vehicles or engines, the complete engine operating range is tested and not just the maximum or some specific power or torque operating points. What is even more important is that the serious discrepancies that are experienced during abrupt transients (as a result of the turbocharger lag) are taken into account, a fact of key importance for diesel engines, which are nowadays almost exclusively turbocharged. The problems of turbocharged diesel engine operation are experienced during acceleration, typical in the every-day operation of automotive and truck engines, or after a load increase, more characteristic of heavy-duty, industrial, locomotive and marine engines; they originate in the inability of the turbocharger compressor to instantly supply the requested air-charge to match the increased fueling, and they manifest themselves as poor drive-ability and overshoot in particulate and gaseous emissions to levels way above their respective steady-state values [8,9]. Acknowledging these, nowadays well established, facts, various legislative directives in the European Union (EU), the US and Japan, have drawn the attention of manufacturers and researchers to the transient operation of diesel engines in the form of transient cycles certification for new vehicles [10,11].

In order to update the 2002 EPA formulas, a large amount of data was carefully collected from 67 papers, published in international Journals, well established Conferences and reports issued by renowned research centers, all dealing with transient/driving cycles experimentation [12−78]; fig. 1 illustrates the number of papers/reports used in chronological order. All these studies concern four-stroke, direct injection engines running on truly transient conditions, and have been conducted during the last 20 years. No steady-state cycles data have been incorporated (as was the usual case with earlier approaches), since these, without a doubt, fail to incorporate the serious emission discrepancies encountered during transients originating in the turbocharger lag [8,9,79]. Moreover, no two-stroke engine data were included, since these engines are becoming obsolete and are nowadays of little (if any) interest for long-term forecast. In general, it was our intention to include as much data as possible from newer production engines (generally speaking, from the last decade), in order for the obtained results to be more representative of the current state of things by incorporating the impacts of modern injection systems and EGR control. To this aim, all relevant works published up to the end of 2011, which provided clear and well founded results, have been, to the best of the author's knowledge, included. On the whole, the dataset contains a good mix of American, European and Japanese engines/vehicles, of real-world biodiesel feedstocks (see also Fig. 2 for a detailed list of all methyl esters investigated), and of engine (39%) and chassis-dynamometer (61%) cycles. Fig. 3 provides a list of the test cycles included in all the investigations (each biodiesel feedstock used in a paper or a report, or each transient cycle, corresponds to one investigation).

For the cases where two or more sets of measurements were available, average data were used, taking into account possible diesel fuel drift effects if these had been reported by the authors. For those cases where the vehicle was fitted with a diesel particulate filter (DPF), PM emission data were collected if they were available upstream of the DPF. The same was valid for the very few cases of engines equipped with SCR or NO_x adsorber; NO_x emission data upstream of these after-treatment devices were only taken into account. As regards the cases with a diesel oxidation catalyst (DOC), engine-out but also DOC-out data have been included in the database (sometimes both were available for the same investigation). On the other hand, some markedly 'extreme' data were only included in the database if the researchers had confirmed in their study the validity of these results with a second or third run. Since some of the authors preferred to present their results using bar charts (instead of providing the exact emission values), a careful and detailed digitization procedure was applied with the use of an appropriate software in order to acquire the emissions data. Data were not included from measurements where cetane improvers were applied, and from the few authors/studies that either decided to demonstrate their results in logarithmic-axis figures (hence a digitization procedure was highly questionable), or did not provide a comparison with the reference diesel fuel.

It should be pointed out that, in general, when diesel fuel data are compared with biodiesel data, the chi-square test should be carried out in order to verify if the observed differences are due to the fuel change or to other causes, such as sampling errors. This procedure is important especially when dealing with low blend ratios where the differences are usually small. However, it is not clear whether all the reported data in the literature have been treated in this manner. In view of this, the possibility that some of the data used were in fact not significant from a statistical point of view cannot be excluded.

Finally, for the cases where a Journal article or Conference presentation paper was based on a previous report, the original report's full set of results was used for a more complete and accurate update of the database, even-though the latter may not be referenced in the text.

3. Results and discussion

3.1. Overall results

Figures 4 to 7 summarize the effects of biodiesel blends on the regulated pollutants, during all transient cycles from the measurements reported in the surveyed studies. For estimation of the best-fit curves, a regression analysis was chosen in order to be able to also provide the coefficient of determination that indicates the degree of data variability. Specifically, a quadratic best-fit curve was chosen, which combines simplicity and relatively good regression capabilities. These quadratic approximations for all examined pollutants are plotted in each figure together with the whole set of original measurement points. As expected in Figs 4–7, the smaller the blend ratio, the narrower the range of reported values. However, since data from all kinds of transient and driving cycles, engines, model years and biodiesel feedstocks are included, unsurprisingly there exists a high degree of variation, particularly for the most popular biodiesel blending ratios B10, B20, B50 and B100. These figures are enhanced with the data provided in Table 1 that provides some basic statistical results derived from the collected measured data, i.e. average, minimum and maximum values, and standard deviation for the most investigated blend ratios. Despite the highly scattered data, a rather compelling decreasing trend for PM, CO and HC emissions with increasing biodiesel blend ratios can be established (Figs 4, 6 and 7), as well as a moderately increasing one as regards the emitted NO_x (Fig. 5).

Although many researchers experimented with higher than B50 blend ratios (mainly in an attempt to explore the 'extreme limits' of biodiesel combustion), it should be in any case emphasized that most of today's engines/vehicles, especially the light-duty ones, are not designed to burn fuels with biodiesel content higher than 20-30% (B20–B30). This is mainly due to problems associated with the injectors (e.g. faster coking) but also with the

ECU and EGR calibration. In fact in Europe, only a very small portion of the diesel-engined vehicle fleet is actually prepared for pure biodiesel combustion. Hence, since the results for higher than B50 blends could prove unpredictable and affect the obtained overall best-fit curves in a decisive manner, separate regressions were performed for the more ‘realistic’ blends up to B50. In general, biodiesel blend ratios higher then B50 have been investigated mainly for heavy-duty engines, and they comprise almost one fifth of the total observations (primarily B100 data). The best-fit curves for blends up to B50 are demonstrated in Figs 4–7 with the red-colored discontinuous lines, and they are very much close to the ones corresponding to the whole database (B0–B100) regressions. Interestingly, exclusion of the higher then 50% biodiesel blend results does not render the remaining data more cohesive. In fact, the coefficient of determination R^2 is now smaller (see Table 2 that summarizes the best-fit curves coefficients as well as the R^2 values for all investigated pollutants and for each transient cycle, engine type and biodiesel feedstock that will be discussed in the next sections), particularly so for PM and HC. On the other hand, the standard error is unsurprisingly smaller owing to the narrower range of the reported values for small blend ratios. As a result, the discussion that follows will concentrate on the whole available dataset, i.e. for all biodiesel blend ratios up to B100, since the suspicion that high blend ratios might have provided unpredictable results was not confirmed. In fact, from the comparison between the two derived best-fit curves in each of the Figs 4–7 it is established that the overall approximations are cohesive for the whole blending range, and are not dominated or even influenced by any particular blend ratio.

The obtained best-fit curves from Fig 4–7 are further demonstrated in Fig. 8 in comparison with the respective EPA trend-lines. Oddly enough, although a considerable amount of new data has been included in the current statistical analysis, most notably newer production engines with EGR control as well as many light-duty chassis-dynamometer results, the earlier EPA best-fit curve is only slightly altered for one of the most significant pollutants, PM, whereas for the other critical emission, NO_x, noteworthy coincidence is still observed for blend ratios up to 40–50%. One of the main reasons for the PM differences observed for high biodiesel blend ratios is believed to be located in the fuel sulfur content. For example, road commercial No 2 diesel in the US contained up to 500 ppm sulfur during the 90s in contrast to less than 50 ppm nowadays, or even less than 15 for ultra-low sulfur diesel (similar is the sulfur content of the diesel fuel in the EU), a fact that has accordingly altered (lowered) the respective diesel PM reference level.

As regards the NO_x emission, it should be pointed out that the behavior of older 2-stroke engines (in the EPA database) with biodiesel blends almost consistently led to increases compared with the neat diesel operation, whereas for their 4-stroke counterparts that are only included in this analysis, a variable trend is observed, with both increases and decreases reported. In order to support this argument it should be pinpointed that 'only' two thirds of all measured NO_x data concern increases over the reference diesel fuel with the remaining third regarding decreases, whereas for PM (but also for CO and HC) the trend is much clearer with almost 90% negative values (i.e. emission benefits with biodiesel combustion). As a result, R^2 for NO_x is very low, of the order of 0.13 (Table 2), whereas for PM it is 0.59.

For the CO and HC quadratic best-fit curves on the other hand in Fig. 8, although these remain comparable in their trend and development with the older EPA data, it is apparent that the biodiesel benefit has faded during the last years for all biodiesel blend ratios, particularly so for HC. As will be discussed later in the text this is primarily due to the fact that many cold-started measurements are included in the database, particularly from engines equipped with diesel oxidation catalysts.

B20 and B100 results extrapolated from Figs 4–7 are further provided in Table 3 in comparison with results from previous review studies [7,80–82] of both engines and vehicles test cycles relative to the neat diesel operation. Please note that whereas both the current study's and EPA's B20 and B100 values have been computed from the corresponding best-fit curves of Figs 4–8, the values from Refs [80–82] provided in Table 3 are the respective average ones for each blend studied, since those studies focused on B20 and/or B100 blend measurements only, and did not provide any regression curves.

3.2. Parametric study

3.2.1. Effect of engine and transient cycle type

The impact of the engine type (heavy-duty or light-duty) or the dynamometer schedule (chassis or engine) is analyzed in Figs 9–12, which illustrate the respective quadratic best-fit curves for all heavy-duty, heavy-duty engine-dynamometer, FTP-only, heavy-duty chassis-dynamometer and light-duty (mostly NEDC) measurements. There are four arguments that need to be made initially:

- The FTP results are by far the most cohesive as regards PM emissions, with the least variance and highest coefficient of determination R^2 of the order of 0.87; actually, these data deserve the label 'statistically significant'. This is probably due to the fact that these measurements have minimal disparity in engines and biodiesel feedstocks, since most of the FTP investigations have been conducted with a relatively small number of American manufactured engines running mostly on SME blends. Although these measurements span over two decades, most of them concern 1990s engines, with relatively few datasets added during the last years from more modern engines. This clear, from a statistical point of view, picture is not blemished at all if the few non-American, heavy-duty, engine-dynamometer cycles are included (ETC and WHTC), but is to a great extent altered if the chassis-dynamometer heavy-duty (mostly UDDS) results are taken into account (R^2 drops to 0.62 for all heavy-duty PM emissions irrespective of dynamometer schedule employed – Table 2). The latter effect is produced because many UDDS studies exhibited a 'reverse' emission pattern, with NO_x decreases and sometimes PM increases [e.g. 15,16,26]. It is known that light-loads (as are, for example, experienced during the UDDS cycle) result in bigger amount of fuel burned during the premixed phase of combustion, and this tends to produce NO_x benefits with rising biodiesel percentage in the fuel blend [45,83]. Likewise, the heavy-duty engine dynamometer data for NO_x (R^2=0.54), CO (R^2=0.83), and HC (R^2=0.78), are way more cohesive than their counterparts from the whole database (R^2=0.13, 0.43 and 0.60 respectively). As was the case with PM, the heavy-duty engine dynamometer data for CO and HC can be marked as statistically significant too.
- On the other hand, for the NEDC (which comprises the majority of the passenger car/light-duty chassis dynamometer data), results are only available during the last 3–5 years (Fig. 1), each investigation corresponds practically to a different engine/car, and there is a variety of tested methyl esters. As a result, the obtained data, many of which have been reported by the same research group, are noticeably scattered and often controversial, particularly so for CO and HC. It is also unclear whether many of the recently reported results (particularly those that

seem to differentiate by a lot from the ‘average’ trends) have been actually confirmed with a second or third run.

- A large percentage of the published data still corresponds to investigations carried out during the 1990s, where many FTP experimentations were limited to hot runs only. On the contrary, all the NEDC investigations have been carried out during the last years where a cold-started run is required by the EU legislation (Directive 98/69/EC). This means that the light-duty data fully incorporate the cold-starting effects, whereas the earlier FTP ones mask this effect to a large extent. As has been established [6,8], the higher initial boiling point of biodiesel with respect to conventional diesel fuel, which leads to more difficult fuel evaporation at low ambient temperatures, and the higher viscosity of biodiesel, which reduces the rate of spray atomization, lead to worse fuel–air mixing and, thus, more intense soot formation at low temperatures with increasing biodiesel percentage in the fuel blend. Similarly, for CO and HC (owing to the lower diesel oxidation catalyst’s efficiency, which in turn originates in the lower biodiesel exhaust gas temperature [84]), emission increases are often noticed when biodiesel is added into the fuel blend [57,58,67,70,76]
- The vast majority of the NEDC investigations have been carried out with engines equipped with EGR and with modern electronic injection systems. It has been well documented that the ECU strategy regarding the EGR control results in lower EGR valve positions and hence higher NO_x emissions, when a diesel-tuned engine runs on biodiesel blends, owing to biodiesel’s lower heating value, again elevating the biodiesel NO_x emission penalty [45,58].

In accordance with the arguments raised previously, the heavy-duty engine dynamometer and the FTP curves almost coincide (Figs 9 and 10) since very few data are available from non-FTP, heavy-duty engine-dynamometer cycles such as the WHTC [45] or the European ETC [74,75]. Not surprisingly, these data correlate very well with the earlier EPA trend-line which included mostly FTP results. On the other hand, inclusion of heavy-duty chassis-dynamometer data (mostly American and mostly UDDS, since there is no relevant European cycle and there is only one measurement available from the Japanese JE05 one [49]) shifts the PM benefit to much lower values. This occurs most probably owing to the fact that the chassis-dynamometer cycles are lighter loaded than the engine-dynamometer ones, hence with less aggressive transient schedules and milder turbocharger lag phases that are primarily responsible for turbocharged engines PM emissions, and where the beneficial effects of biodiesel (mainly its high oxygen content) prevail over conventional diesel combustion [8]. Likewise, the lightly-loaded passenger car chassis-dynamometer cycles exhibit lower PM benefit than the significantly higher-loaded heavy-duty engine dynamometer cycles.

Particularly as regards the heavy-duty chassis-dynamometer curve in Fig. 9 it should be pointed out that: a) owing to the quadratic best-fit approximation applied, b) the fact that data variance is not consistent for each biodiesel blend ratio, and c) because almost all data are available for B20, B35, B50 and B100 blends (in fact there are no measurements available for B60, B70, B80 and B90), the corresponding best-fit curve in Fig. 9 assumes a parabolic form, with the highest benefits suggested for B60 rather than B100. Similar arguments are valid for other ‘parabolic’ curves that will be presented and discussed later in the text.

Concerning the NO_x emissions, the light-duty NEDC differentiates from the rest of the curves again, this time considerably, exhibiting higher NO_x liability (and higher absolute NO_x emissions) for all the tested biodiesel blends. Compliant with the remarks made earlier, this behavior can be attributed to the following two reasons:

a) most of the NEDC experiments include modern passenger car engines fitted with EGR, which tends to increase NO_x emissions when an increasing biodiesel blend ratio is applied, and

b) this cycle has a higher portion of extra-urban segment, where the majority of NO_x is emitted, a fact that actually strengthens the previous point.

Unfortunately there are few non-European, passenger car or light-duty data available to support the previous arguments and confirm the trend observed in Fig. 10, since the majority of the investigations in the US have focused on heavy-duty engines and cycles.

An even more prominent differentiation from the general ‘average’ trend is exhibited by the heavy-duty chassis-dynamometer results/cycles, where a moderate negative trend for NO_x emissions with biodiesel blending is established (i.e. NO_x benefit over the reference diesel operation when biodiesel is added in the fuel blend). As mentioned earlier, the underlying reasons may be located in the light loading of these cycles (mostly the UDDS one), which tends to increase the premixed portion of combustion over the diffusion one, where the NO_x are primarily produced. In fact, the data in Fig. 13 confirm this trend by showing that only 40% of the total measured NO_x measurements from heavy-duty chassis-dynamometer cycles corresponded to emission increases when biodiesel is added in the fuel blend (i.e. an impressive 60% of the measurements showed NO_x benefits over the neat diesel operation), compared with 67% from all data and 78% from heavy-duty engine-dynamometer data.

Unlike the PM and NO_x results, for both CO and HC, the heavy-duty chassis-dynamometer and engine-dynamometer data are very cohesive, exhibiting an almost identical trend and only slight differentiation in the expected values from the best-fit curves (Figs 11 and 12 and Table 2).

The impact of biodiesel blends on the HC and CO emissions from passenger car engines running on light-duty chassis-dynamometer cycles is, on the other hand, another intriguing finding from the analysis, demonstrated in Figs 11 and 12. An extremely different pattern is established for both pollutants compared with the heavy-duty data. In accordance with the arguments mentioned earlier, it seems that data from

a) newer production, European engines with after-treatment control in the form of diesel oxidation catalysts, which seem to operate less efficiently with biodiesel,
b) many light-loaded passenger car cycles, and, predominantly,
c) cold-started runs

have gradually shifted the overall trends to lower CO and HC emission benefits. In fact, many of the researchers from light-duty engines/cycles have actually reported increases with the use of biodiesel. This can be further supported from the statistical data plotted in Fig. 13, where for CO and HC, whereas for all HD engines 92% of the measured data corresponded to decreases over the mineral diesel operation, for the LD engines/cycles this percentage drops considerably to 65% for CO and 78% for HC.

An interesting question has been raised by some researchers, whether the chassis-dynamometer (heavy-duty) studies are more representative than the engine-dynamometer ones in terms of more accurate real-world effects on emissions. McCormick et al. [41] based on the north-American studies reviewed up to that time and some new data reported, concluded that there does not appear to be a discrepancy between engine and chassis testing studies as regards the effect of B20 (which was the biodiesel blend under investigation) on NO_x emissions; they found that individual engines may show NO_x increasing or decreasing, but on average there appeared to be no net effect, or at most a very small effect of the order of ±0.5%. The current analysis, however, which includes a much larger dataset from all biodiesel blend ratios and engine origins does not support this theory, since as is obvious in Figs 9 and 10, chassis-dynamometer data from heavy-duty vehicles exhibit much different results and trends (and also much greater variance) than their engine-dynamometer counterparts.

3.2.2. Effect of engine MY

Another interesting comparison can be made on the basis of the investigated engine's model year (or emission level). Figs 14 to 17 demonstrate these effects by comparing, for each pollutant, the best-fit curves from all data, data for post 2000 MY (or Euro 3/III and later) and from post 2005 data (or Euro 4/IV and later). There are two main features that differentiate the engines with respect to the MY (since no two-stroke or IDI measurements are included in the dataset). The first is the antipollution system and the second is the fuel injection system.

It is most probably the EGR effects that differentiate the post 2000 MY results from the total regression data. Lower EGR valve positions dictated by the ECU during transients, result in lower amounts of recirculated gas, higher gas temperatures and higher NO_x but also lower PM [8]. However the data for engines/vehicles with post 2005 MY do not solidly confirm this trend at least not for all biodiesel percentages in the fuel blend. Since the minimal post-DPF or post-SCR data have been deliberately ignored from the database and the subsequent regressions, the only logical explanation as to the differentiation in the results must lie in the fuel injection system. One should not forget also that these newer data are much fewer (less than one quarter of the total, whereas the post-2000-MY data comprise 50% of the whole database), and primarily regard the highly-variable and scattered NEDC. Supporting the previous (EGR-related) argument, Fig. 18 shows that the percentage of negative NO_x values with biodiesel addition in the fuel blend over the reference petrodiesel operation seems to steadily increase over the years, with 87% negative values reported from post 2005 MY data relative to 77% from post 2000 ones and 66.5% from the whole dataset.

Particularly for the emitted CO, a previous investigation by Yanowitz and McCormick on north-American heavy-duty engines [81] showed that for B20 blends, the earlier pump-line-nozzle injection systems resulted in higher benefits over the reference diesel operation than their modern common rail counterparts, in a way confirming the results demonstrated in Fig. 16.

3.2.3. Effect of biodiesel feedstock

One of the most interesting points regarding the biodiesel impact on (NO_x) emissions is the effect of fuel properties and molecular structure. It is Peterson et al. [26] and McCormick et al. [30], studying the performance of heavy-duty diesel engines (1994 and 1991 MY respectively) during engine or chassis dynamometer cycles, who have conducted the most instructive research on the subject so far; this demonstrated that a highly statistical correlation between nitrogen oxides and degree of unsaturation exists (R^2=0.89 and 0.93 respectively). McCormick et al. [40], this time on a 2002-2003 common rail, heavy-duty diesel engine, showed that B100 blend tests, in general, confirmed the statistically significant trend of the earlier engines, although the R-squared value was found lower this time; it was argued that this might imply fuel injection system effects, primarily through the weaker influence of the biodiesel's higher bulk modulus of elasticity relative to neat diesel fuel, but EGR definitely had an important role too. On the contrary, the B20 test results of the newer engines were not found statistically significant (R^2 ranging from 0.32 to 0.49), possibly owing to the much lower absolute values of NO_x, which made feedstock effects more difficult to observe. In any case, a universal conclusion reached from all researchers that investigated feedstock effects on exhaust emissions was that no correlation between degree of unsaturation and PM, CO or HC could be reached.

Unfortunately no statistically significant direct comparison between the various biodiesel feedstocks is feasible from the available database in order to substantiate the previous findings, at least as regards the NO_x emissions. This is primarily due to the fact that although SME has been popular in all types of investigations (but mostly on American FTP), the rapeseed methyl ester has been primarily employed in European studies of the NEDC, all of which have been conducted during the last three years (see also Fig. 1), with a great variety of modern, EGR equipped engines/vehicles. Comparing the whole SME database with its RME counterpart would be, to a large extent, like comparing FTP with NEDC measurements, at least for the currently applied level of statistical analysis. To this aim, Figs 19 and 20 only provide, for the sake of completeness, a comparison between the vegetable, SME and non-vegetable (animal fat but also waste cooking derived) data from all available studies and engines.

The only cycle for which a healthy variety of methyl esters has been investigated is the NEDC, but one of the points raised earlier, i.e. the great disparity of the tested engines/vehicles, makes such a comparison dubious, that is why it has not been performed. Nonetheless, in Figs 21 and 22, a comparison is attempted as to the effect of biodiesel feedstock on PM and NO_x emissions from all heavy-duty data (engine and chassis-dynamometer ones), which, as was demonstrated in Table 2, have been found to exhibit the least data variance. It is the RME blends in heavy-duty experimentations that demonstrate the most ‘erratic’ behavior, with PM increases and NO_x emission decreases that differentiate considerably from the general tendency. For this trend, it is the results of Peterson et al. [15,16,26], who systematically used RME blends in their research, that are mostly responsible. Unfortunately, there are only a few RME FTP results available to confirm or refute this behavior. From the rest of the data, no clear conclusions can be reached as regards the effects of SME, animal fat (AFME) and waste cooking methyl esters (WCME) on the emitted PM and NO_x. In any case, it should be pointed that for both AFME and WCME only 16 different measurement datapoints were available (compared with 136 from SME); interestingly, all corresponded to PM decreases, whereas for NO_x a slightly mixed behavior was exhibited with both increases and decreases (Fig. 23).

Lastly, it was not considered feasible to investigate the effect of the injection system on emission effects from biodiesel use, although this actually constitutes a very intriguing subject, which might be able to disclose some masked at the moment emission mechanisms. Modern engines employ both modern injection systems and antipollution control such as EGR and diesel oxidation catalysts in contrast to older production motors (e.g. with mechanical-type injectors). Accordingly, it seems practically unachievable to isolate the effect of the injection system on the emission impacts, at least for the level of statistical analysis attempted in the current study.

Finally, Fig. 24 gathers all the measured PM and NO_x measurements, grouped with respect to the biodiesel percentage in the fuel blend, and documents the, well established during steady-state operation, contradicting behavior (trade-off) between PM and NO_x. The latter seems to apply reasonably well to transient cycles emissions too for the vast majority of biodiesel blend ratios, engines and transient cycles tested, although the data are not statistically significant (R^2=0.24).

4. Summary and conclusion

A large amount of data published in International Journals, well established Conferences and from renowned research centers up to end of 2011 was gathered and analyzed as regards the biodiesel blend impacts on regulated pollutants during transient cycles from passenger car/light-duty and heavy-duty engines/vehicles; only 4-stroke engine data were included from the last twenty years, and only transient measurements that are capable to disclose the true emission behavior of turbocharged diesel engines. The majority of the published work has focused on biodiesel effects during the American heavy-duty FTP and the European passenger car NEDC, and for B20 and B100 blends of soybean or rapeseed-derived methyl esters.

Correlations were reached for quantification of biodiesel benefits (or penalties) on regulated emissions relative to the neat diesel operation, updating an earlier EPA approach. The various trends observed were discussed and possible explanations were proposed based on fundamental aspects of diesel engine combustion and emissions, primarily for the two most critical pollutants PM and NO_x. Since data from all kinds of engines, driving cycles and biodiesel feedstocks and blends were used, the coefficients of determination, particularly for NO_x, were low, suggesting a high degree of variation of the available data. On the other hand, it is only the PM emissions that present the clearest picture as regards data cohesion for all types of engines, feedstocks and driving schedules. The concluding results for each pollutant are summarized in Fig. 25, where the overall trends are evident with a clear decreasing tendency for PM, CO and HC and a moderate increasing one for NO_x. For the more realistic scenario of biodiesel blends up to 50%, in general similar best-fit approximations were obtained, indicating that, although the majority of today’s cars is not prepared to burn pure biodiesel, the higher than B50 blends do not ultimately affect the general trend or the cohesion of the results.

However, apart from the general trends illustrated in Fig. 25, (sometimes completely) different individual trends were established depending on the specific engine or dynamometer schedule and the biodiesel feedstock. In particular, only the heavy-duty engine dynamometer (mostly FTP) data presented systematically high cohesion for all four regulated pollutants in the database. For most of the other measurements, however, the results were often scattered and sometimes controversial. This holds particularly true for passenger cars data, for PM data of heavy-duty chassis-dynamometer cycles, and also for

NO_x emission data in general. In view of this, it is postulated that a more sophisticated statistical analysis might prove better suited to deal with these data, possibly rejecting some measurements, and providing a more applicable best-fit approximation.

A vital question is whether the measured results and the computed in this study best-fit curves are actually representative of real-world driving conditions. It can be argued that dynamometer results, either chassis or engine ones, are representative to the same extent that the legislated transient/driving cycles are representative of real driving conditions. Particularly for the NEDC, from which a large number of data is available, a lot of criticism has been raised that this is actually a 'soft' cycle with linear and smooth accelerations and rather low loading for the majority of today's cars. It is thus anticipated that during real-world driving, the NO_x penalty from the use of biodiesel will be actually higher, since the more aggressive driving style results also in more abrupt accelerations, hence harsher turbocharger lag phases that in turn provoke lower EGR valve positions (and higher PM benefit with biodiesel fuel blends). Moreover, the effect of cold-starting on the real engine conditions is also expected to be more prominent that that incorporated in the current analysis, i.e. lower PM, HC and CO benefits are expected with biodiesel combustion over petrodiesel operation owing to the colder real-life temperatures compared with the ones under which the transient test cycles are usually performed.

In any case, it is believed that the results of the current study can prove valuable to administrations and international institutions by providing a good estimate of the expected emission impacts of biodiesel blended fuels on engine or vehicle fleets, enabling long-term planning and accommodating decision making on the delicate and timely matter of exhaust emissions from motor engines.

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Nomenclature

Abbreviations

AFME	animal fat methyl ester
DOC	diesel oxidation catalyst
DPF	diesel particulate filter
ECU	engine control unit
EGR	exhaust gas recirculation
EPA	Environmental Protection Agency
ETC	European transient cycle (for heavy-duty engines)
EU	European Union
FAME	fatty acid methyl ester
FTP	Federal Test Procedure (for heavy-duty engines)
FTP75	Federal Test Procedure (for light-duty vehicles)
HD	heavy duty
LD	light duty
MY	model year
NEDC	new European driving cycle (for light-duty vehicles)
PM	particulate matter
PME	palm methyl ester
RME	rapeseed methyl ester
SCR	selective catalytic reduction
SME	soybean methyl ester
UDDS	urban dynamometer driving schedule
US06	supplemental Federal Test Procedure (for light-duty vehicles)
VGME	vegetable methyl ester
WCME	waste cooking methyl ester
WHTC	world-wide harmonized transient cycle (for heavy-duty engines)

Table 1 – Basic statistical data for all regulated pollutants and for the most investigated biodiesel blends.

		PM	NO_x	CO	HC
B5	Average	–5.62	0.62	–4.32	–5.23
	St.Dev	5.56	4.77	6.85	10.73
	Min–Max	–23.1÷9.1	–9.9÷9.5	–17.6÷7.1	–23÷24.5
	Count	20	24	19	20
B10	Average	–10.73	1.57	–6.54	–0.29
	St.Dev	10.19	7.80	9.79	16.65
	Min–Max	–32÷13.6	–16÷24	–32.3÷13.6	–25÷42
	Count	36	41	31	37
B20	Average	–14.94	1.07	–15.14	–18.25
	St.Dev	13.58	6.26	13.45	15.56
	Min–Max	–56.5÷28	–24÷28.5	–69÷25.1	–79.5÷35.2
	Count	146	150	134	134
B30	Average	–13.50	5.00	–9.22	–13.85
	St.Dev	11.09	7.54	18.71	14.37
	Min–Max	–52.7÷7.6	–12÷23.7	–58.8÷32	–56.5÷20
	Count	24	29	26	30
B50	Average	–23.72	3.30	–17.69	–22.20
	St.Dev	25.72	9.22	27.70	23.78
	Min–Max	–63.1÷35	–13.2÷22.4	–58÷42.7	–56÷55
	Count	32	35	34	32
B100	Average	–43.88	8.54	–33.01	–44.42
	St.Dev	31.88	17.17	30.09	24.87
	Min–Max	–80÷43.5	–23÷61	–72.4÷86	–96÷28.4
	Count	61	65	58	63

Table 2 – Summarization of best-fit quadratic curve coefficients A and B, coefficient of determination R^2 and standard error for all pollutants, transient cycles, engine types and methyl esters (quadratic best-fit curve: $y=Ax+Bx^2$).

	PM	NO_x	CO	HC
All data	A=–0.722 B=0.00323 R^2=0.59 Std. error=19.16	A=0.106 B=–0.000318 R^2=0.13 Std. error=10.24	A=–0.531 B=0.00236 R^2=0.43 Std. error=19.84	A=–0.613 B=0.00152 R^2=0.60 Std. error=20.03
All data for blends up to B50	A=–0.951 B=0.010 R^2=0.55 Std. error=14.56	A=0.1524 B=–0.00178 R^2=0.10 Std. error=7.45	A=–0.8566 B=0.01086 R^2=0.42 Std. error=15.46	A=–0.972 B=0.01075 R^2=0.48 Std. error=16.66
All data for engine MY>2000	A=–0.67 B=0.00191 R^2=0.64 Std. error=17.86	A=0.170 B=0.000384 R^2=0.48 Std. error=9.04	A=–0.302 B=0.0023 R^2=0.081 Std. error=21.56	A=–0.552 B=0.00191 R^2=0.43 Std. error=21.66
All data for engine MY>2005	A=–0.765 B=0.0055 R^2=0.48 Std. error=18.71	A=0.231 B=–0.000617 R^2=0.46 Std. error=8	A=–0.54 B=0.0054 R^2=0.14 Std. error=23.4	A=–0.550 B=0.00252 R^2=0.38 Std. error=21.36
All heavy-duty cycles	A=–0.734 B=0.00312 R^2=0.62 Std. error=20.04	A=0.007 B=0.000506 R^2=0.11 Std. error=8.15	A=–0.803 B=0.00453 R^2=0.68 Std. error=16	A=–0.779 B=0.00254 R^2=0.72 Std. error=18.99
All heavy-duty engine-dynamometer cycles	A=–0.840 B=0.00287 R^2=0.87 Std. error=12.6	A=0.068 B=0.000491 R^2=0.54 Std. error=5.75	A=–0.812 B=0.00416 R^2=0.83 Std. error=11.48	A=–0.811 B=0.00305 R^2=0.78 Std. error=16.46
All heavy-duty chassis-dynamometer cycles	A=–0.686 B=0.0049 R^2=0.31 Std. error=23.2	A=0.005 B=–0.000763 R^2=0.13 Std. error=7.87	A=–926 B=0.00573 R^2=0.64 Std. error=18.68	A=–0.829 B=0.0024 R^2=0.82 Std. error=14.49
All light-duty	A=–0.603 B=0.00235 R^2=0.61 Std. error=14.75	A=0.246 B=–0.00088 R^2=0.24 Std. error=13.3	A=–0.152 B=0.00186 R^2=0.01 Std. error=21.45	A=–0.529 B=0.0048 R^2=0.20 Std. error=19.34
All vegetable oils	A=–0.665 B=0.00334 R^2=0.50 Std. error=19.74	A=0.103 B=–0.00031 R^2=0.13 Std. error=9.71	–	–
All SME	A=–0.834 B=0.00353 R^2=0.73 Std. error=15.26	A=0.097 B=0.00014 R^2=0.3 Std. error=7.57	–	–

Table 3 – Comparative emission results from biodiesel combustion relative to neat diesel operation for European and American engine and vehicle test cycles from various review studies.

Engine Type / Model Year	Data up to	Biodiesel Blend	NO_x (%)	PM (%)	CO (%)	HC (%)
EPA [7] *	2000	B20	2.0	–10.1	–11.0	–21.1
Lindhjem and Pollack [80] **	2000		2.5	–9.0	–13.3	–18.2
Yanowitz and McCormick [81] ***	2006		1.0	–17.0	–16.0	–16.0
Anderson [82] ****	2009		4.7	–14.5	–5.4	–4.9
Current study *****	2011		2.0	–13.1	–9.7	–11.7
EPA [7] *	2000	B100	10.0	–48.0	–48.0	–67.0
Lindhjem and Pollack [80] **	2000		11.8	–51.0	–42.0	–69.7
Anderson [82] ****	2009		7.5	–	–	–13.4
Current study *****	2011		7.4	–39.9	–29.5	–46.1

* 86% of the EPA PM and 83% of the EPA NO_x database observations concern the FTP cycle; SME was the biodiesel used in almost 75% of the cases examined, which also included some ECE R49 stationary cycle results as well as many two-stroke engines data. Moreover, only 1.8% of the observations correspond to newer that 1998 engines, whereas 29% of the data are from engines manufactured during the 1980s

** Engine dynamometer tests only; two-stroke engines included

*** North-American, four-stroke, heavy-duty engines only

**** Average values from light-duty and heavy-duty dynamometer tests

***** Four-stroke engines only from all heavy-duty and light-duty engine and chassis dynamometer tests

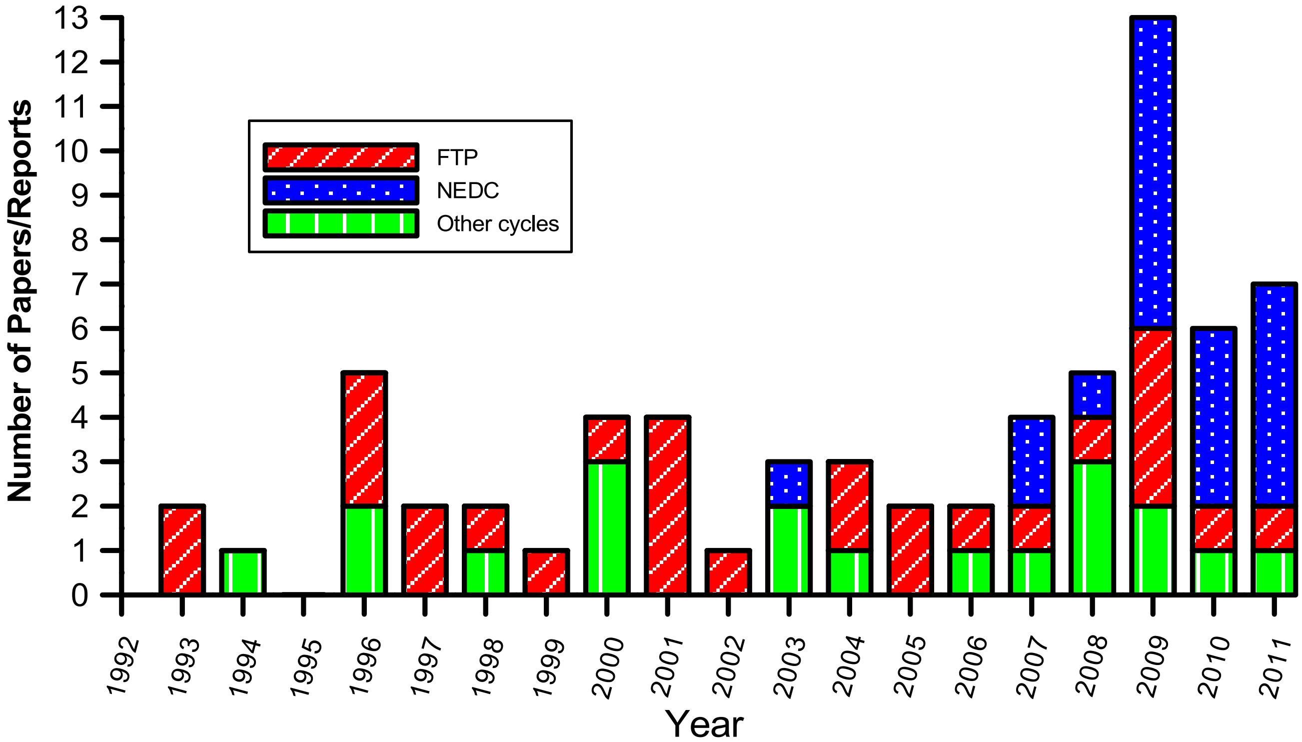

Fig. 1 – Number of published papers and reports on diesel engine emissions during transient cycles with biodiesel blends in a chronological order.

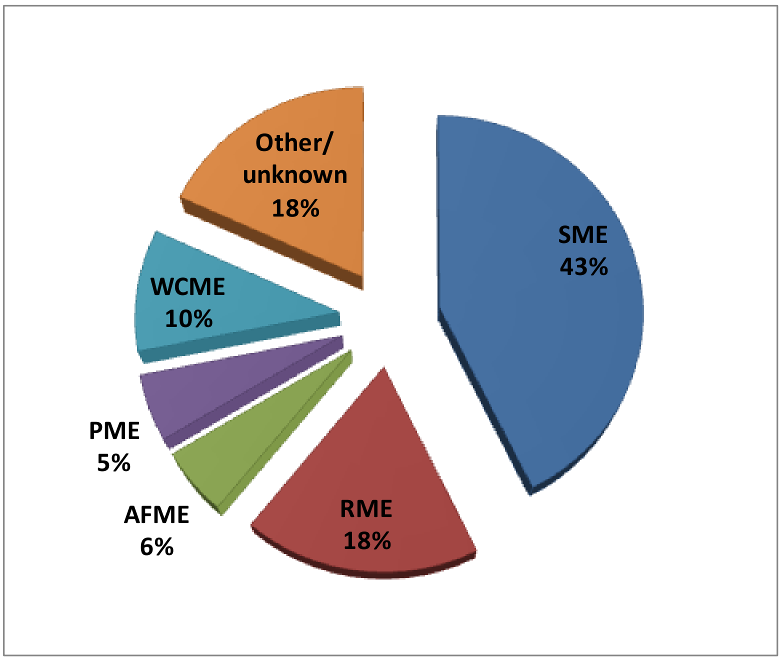

Fig. 2 – Investigations of diesel engine emissions during transient cycles with biodiesel blends based on the methyl ester used.

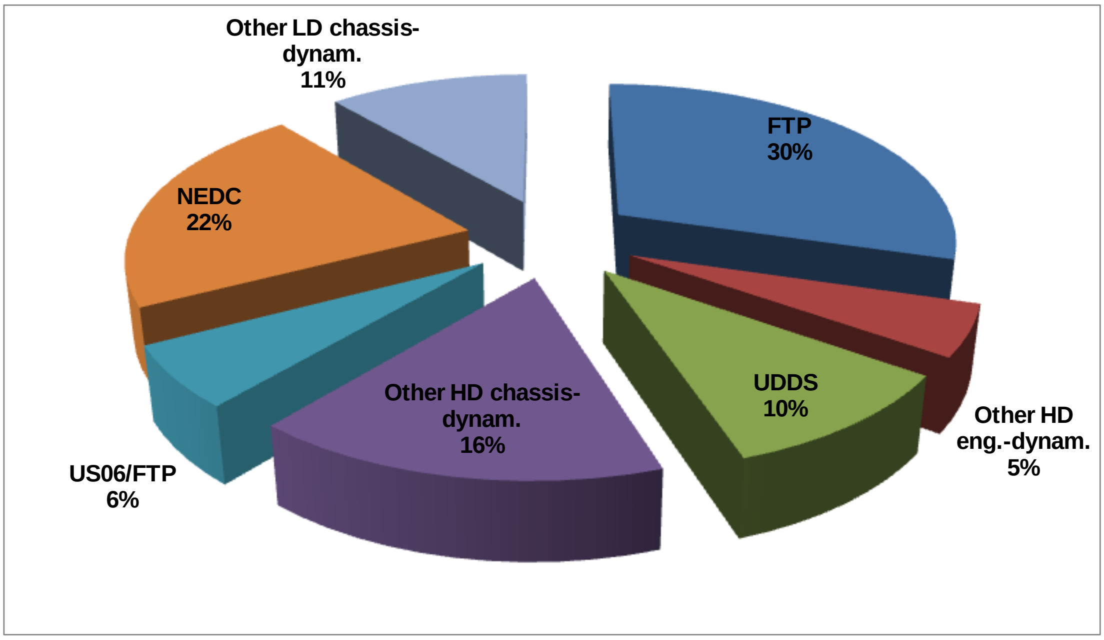

Fig. 3 – Investigations of diesel engine emissions during transient cycles with biodiesel blends based on the transient/driving cycle studied.

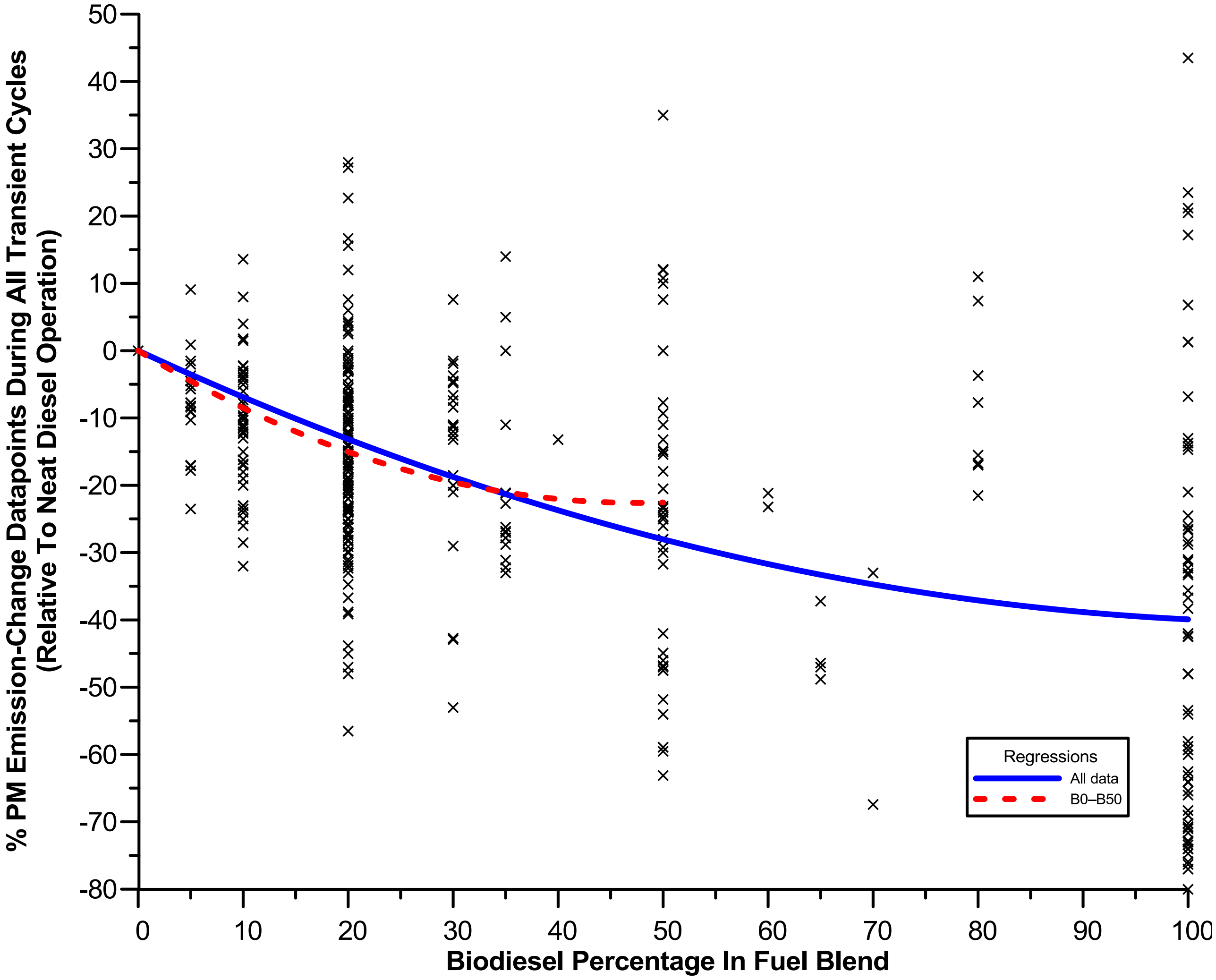

Fig. 4 – PM emission-change datapoints and best-fit curves when using various biodiesel-diesel blends during all transient cycles.

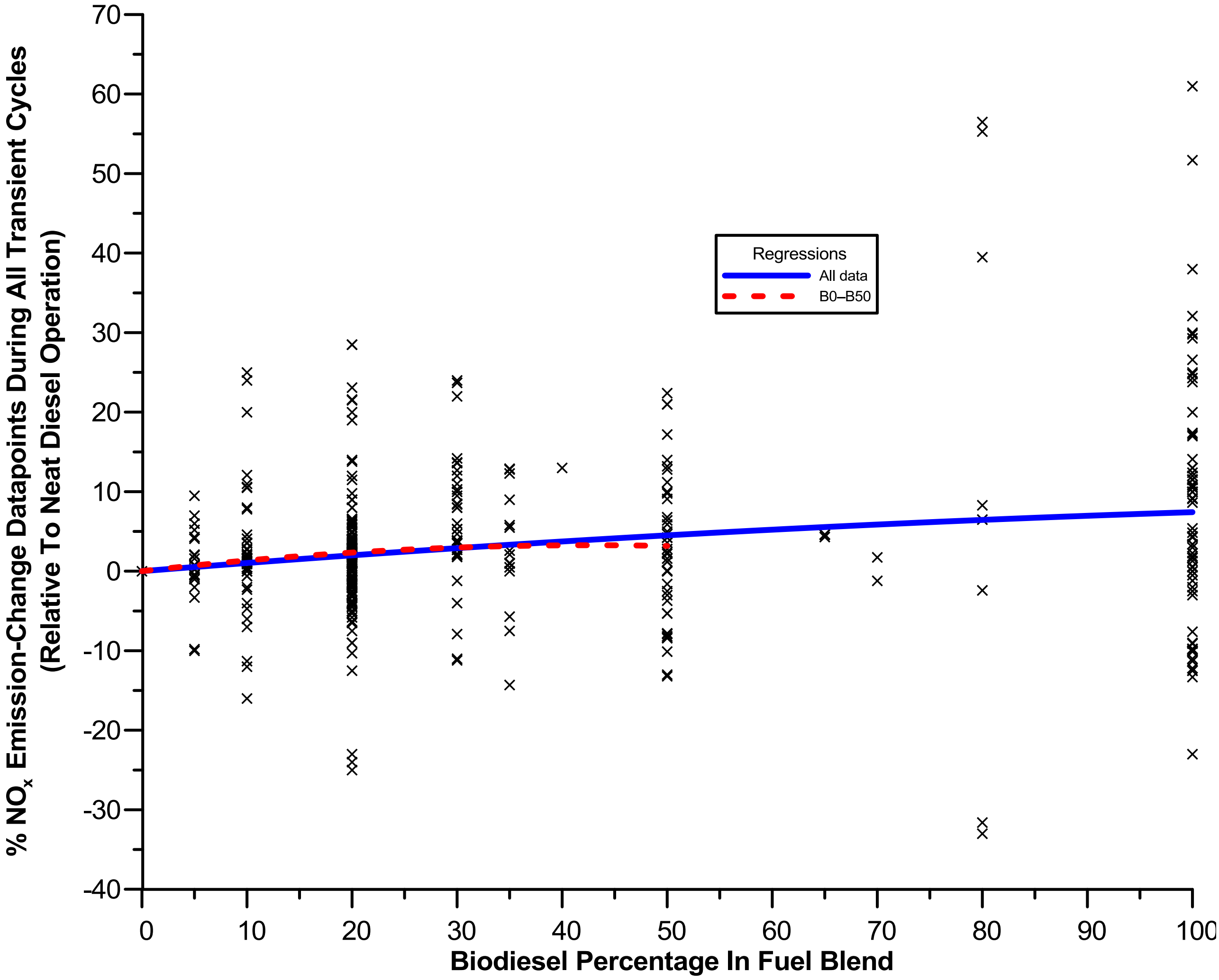

Fig. 5 – NO_x emission-change datapoints and best-fit curve when using various biodiesel-diesel blends during all transient cycles.

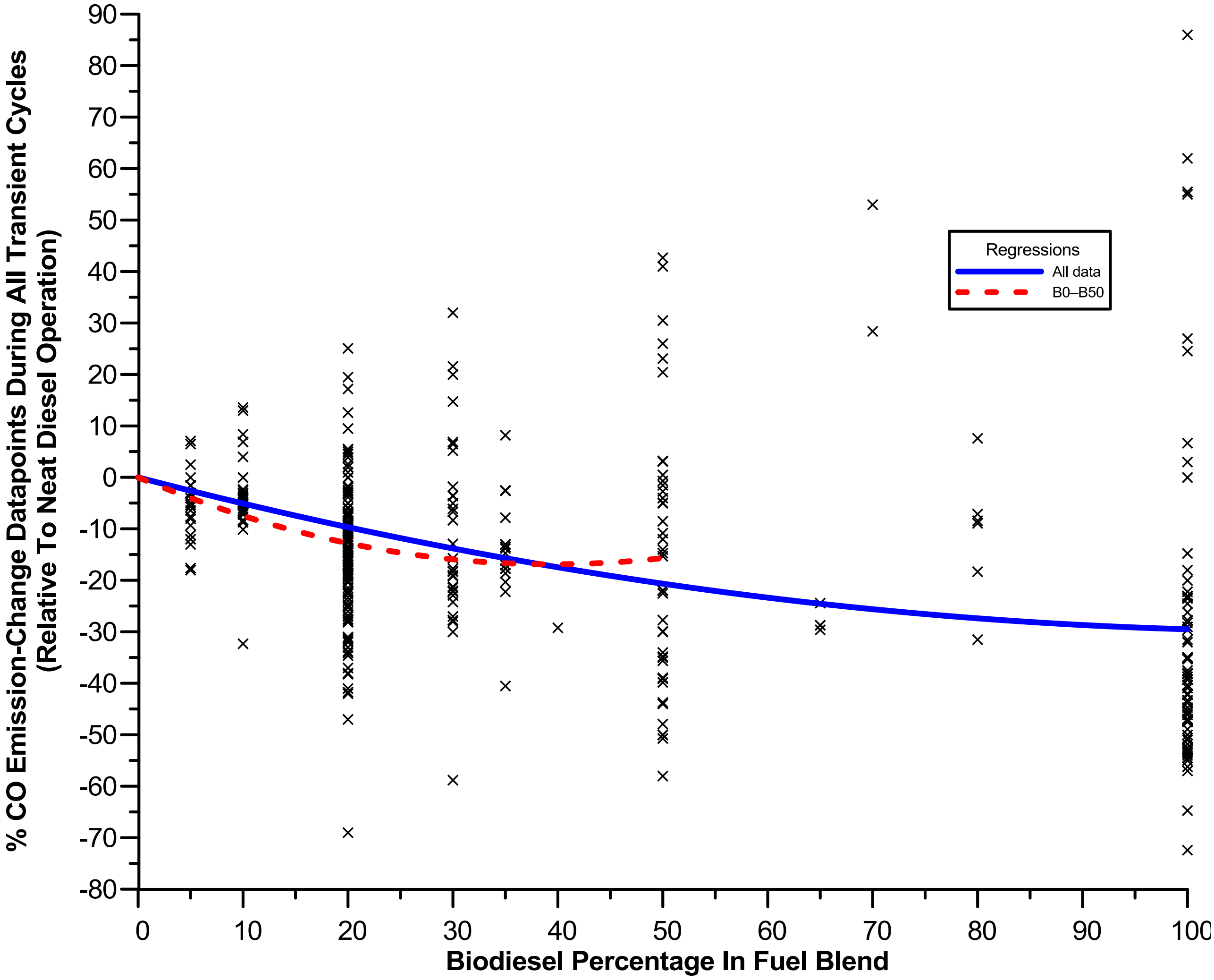

Fig. 6 – CO emission-change datapoints and best-fit curve when using various biodiesel-diesel blends during all transient cycles.

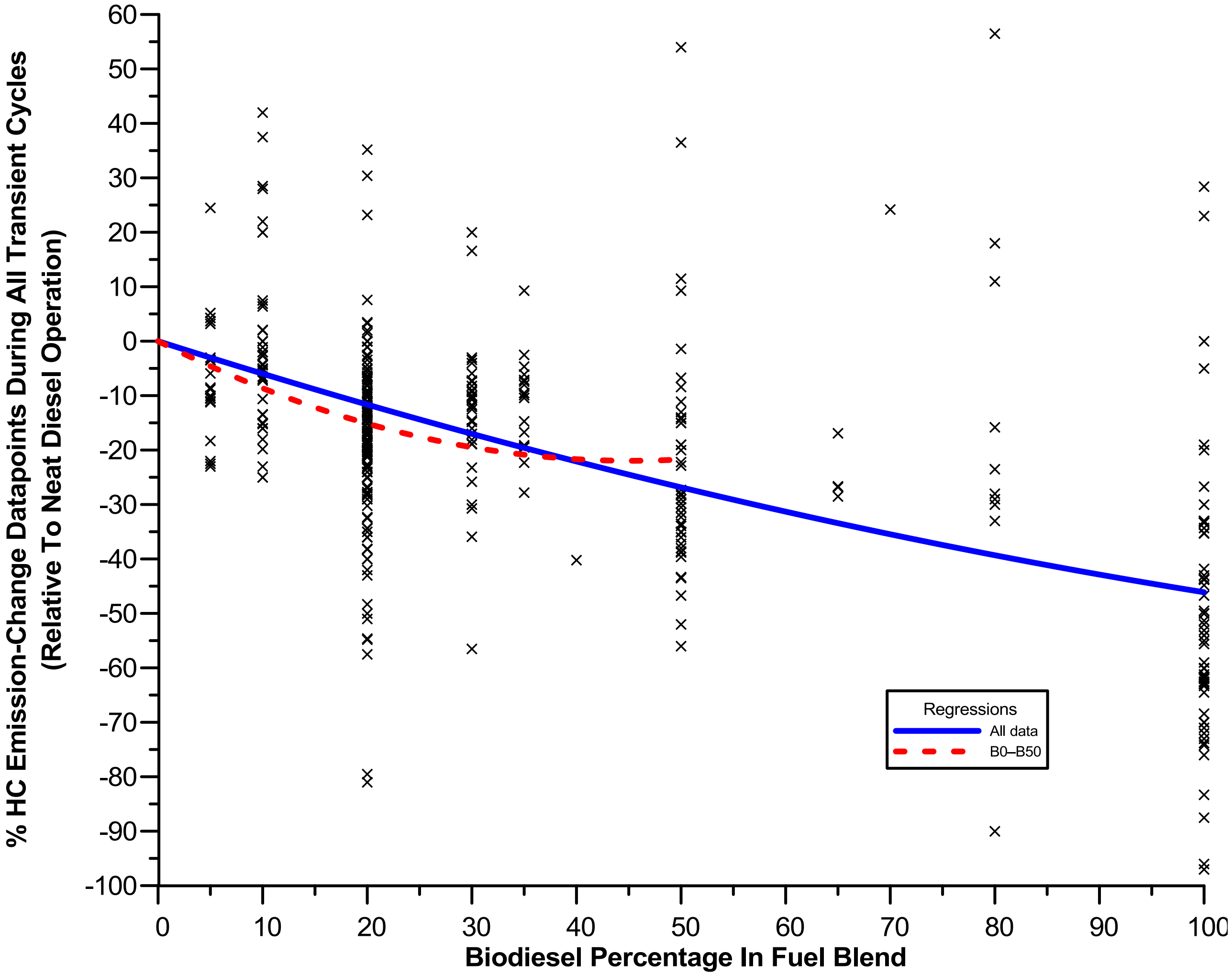

Fig. 7 – HC emission-change datapoints and best-fit curve when using various biodiesel-diesel blends during all transient cycles.

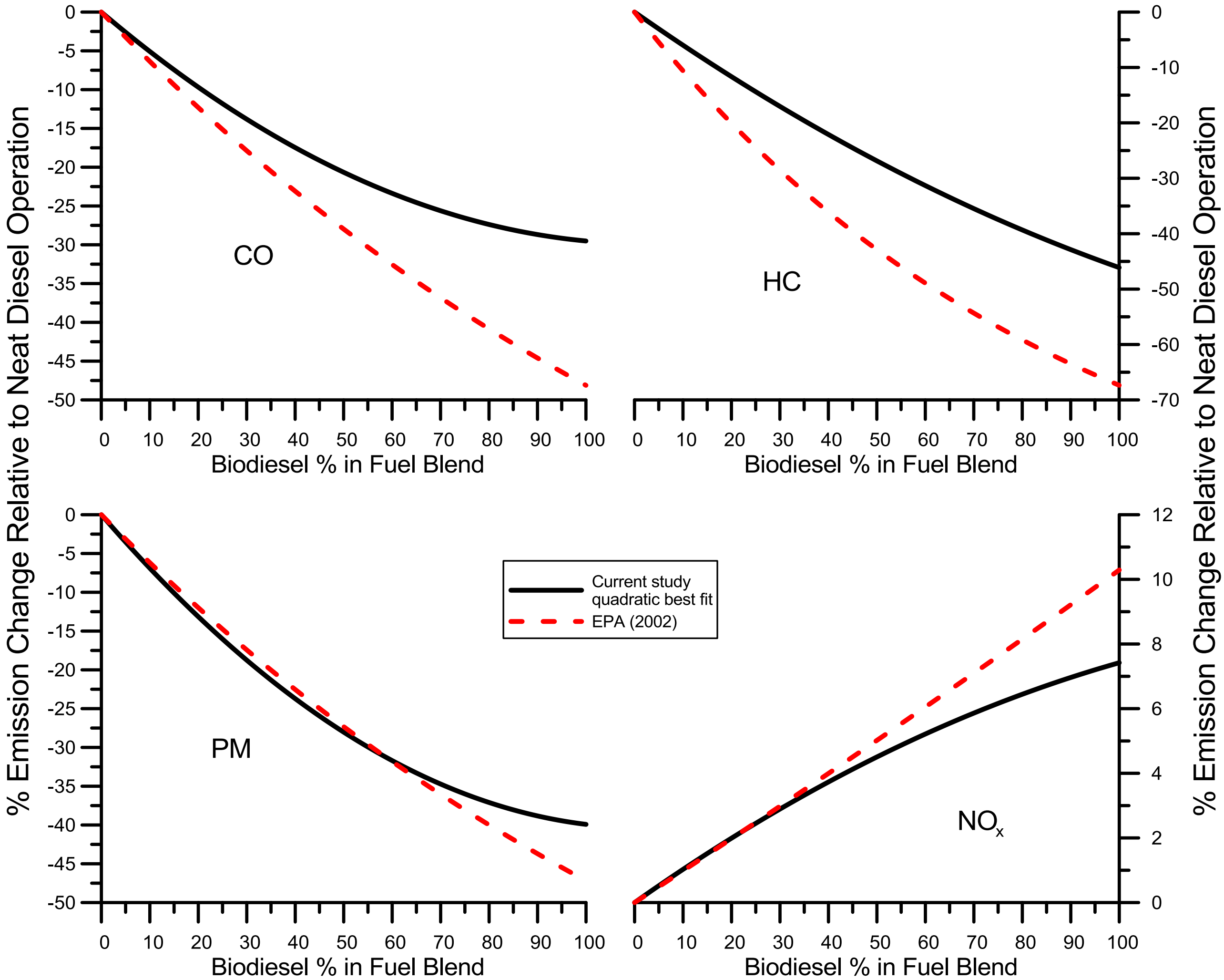

Fig. 8 – Comparison of collective best-fit curves for all regulated pollutants between the current investigation and the earlier (2002) EPA study.

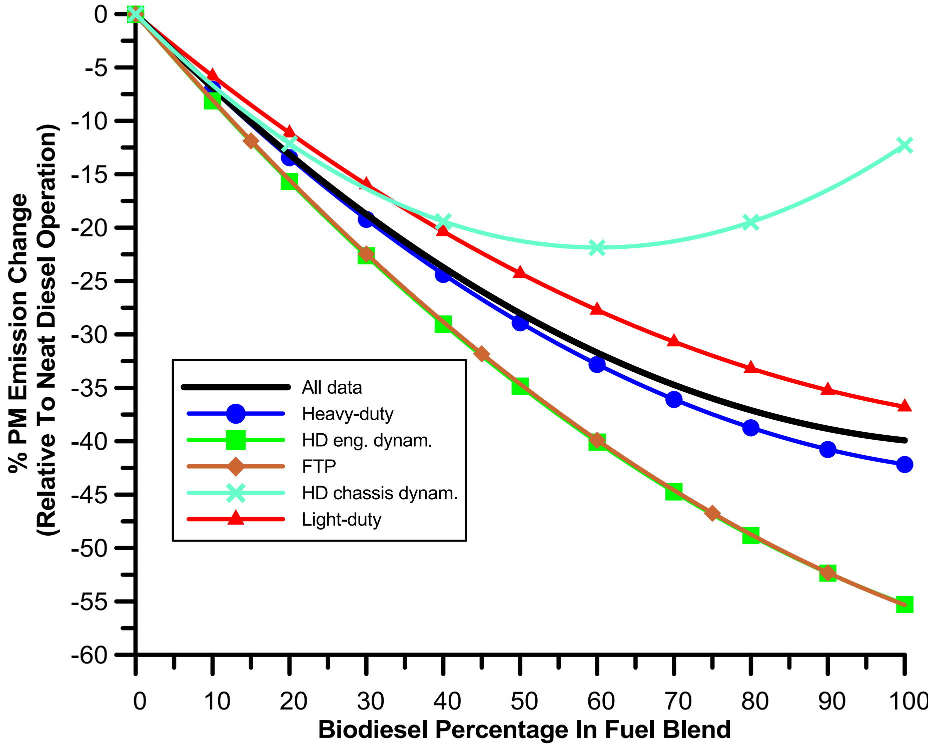

Fig. 9 – Comparative PM emission change best-fit curves for various transient cycles and engine types.

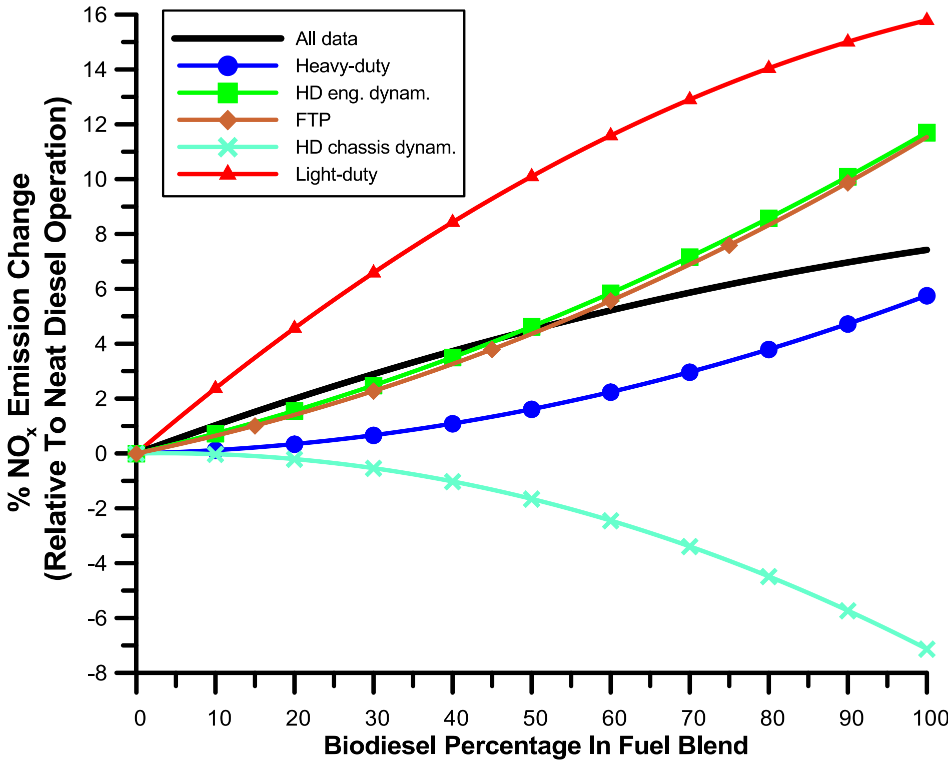

Fig. 10 – Comparative NO_x emission change best-fit curves for various transient cycles and engine types.

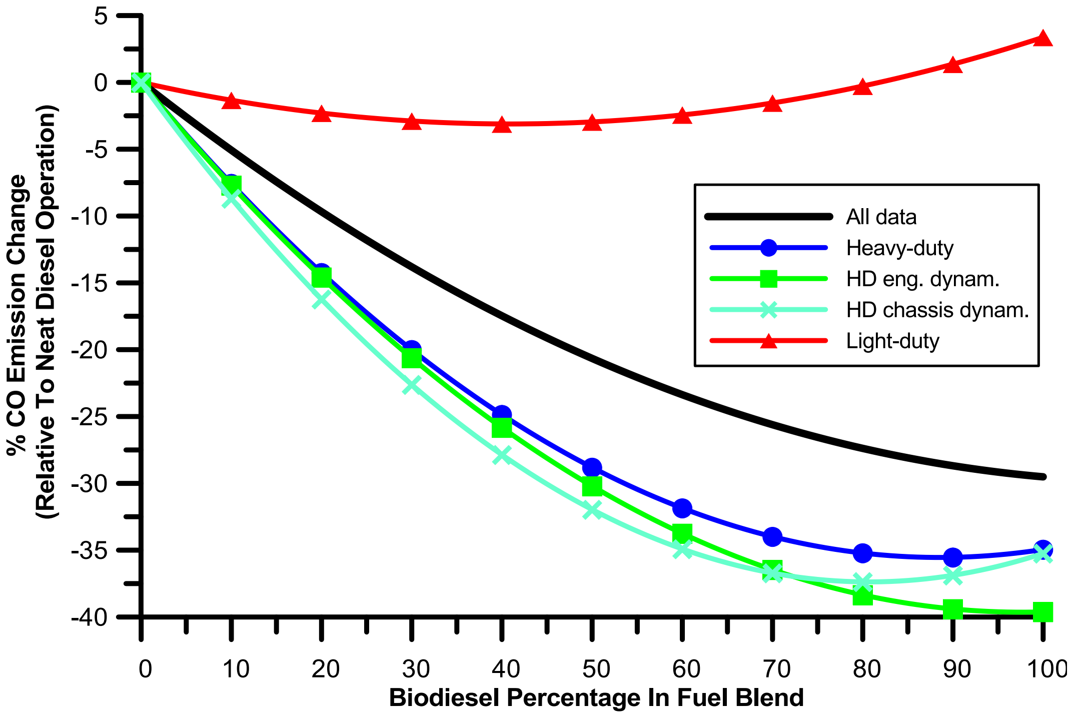

Fig. 11 – Comparative CO emission change best-fit curves for various transient cycles and engine types.

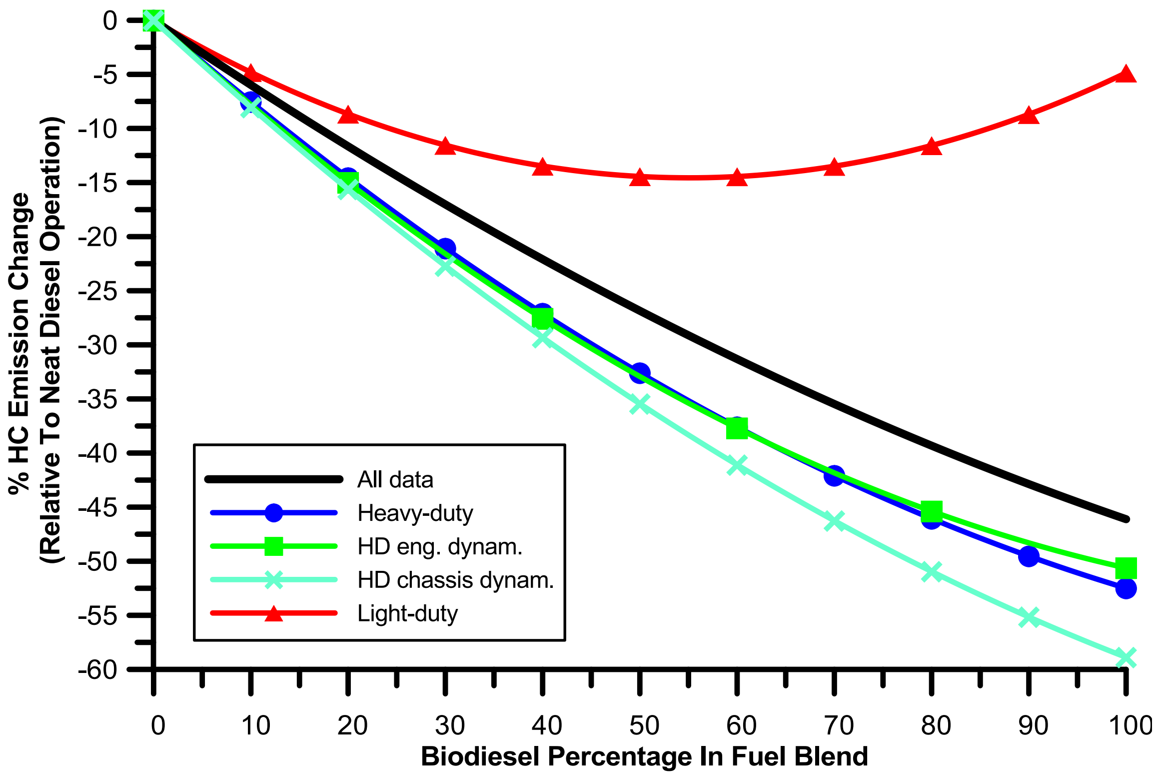

Fig. 12 – Comparative HC emission change best-fit curves for various transient cycles and engine types.

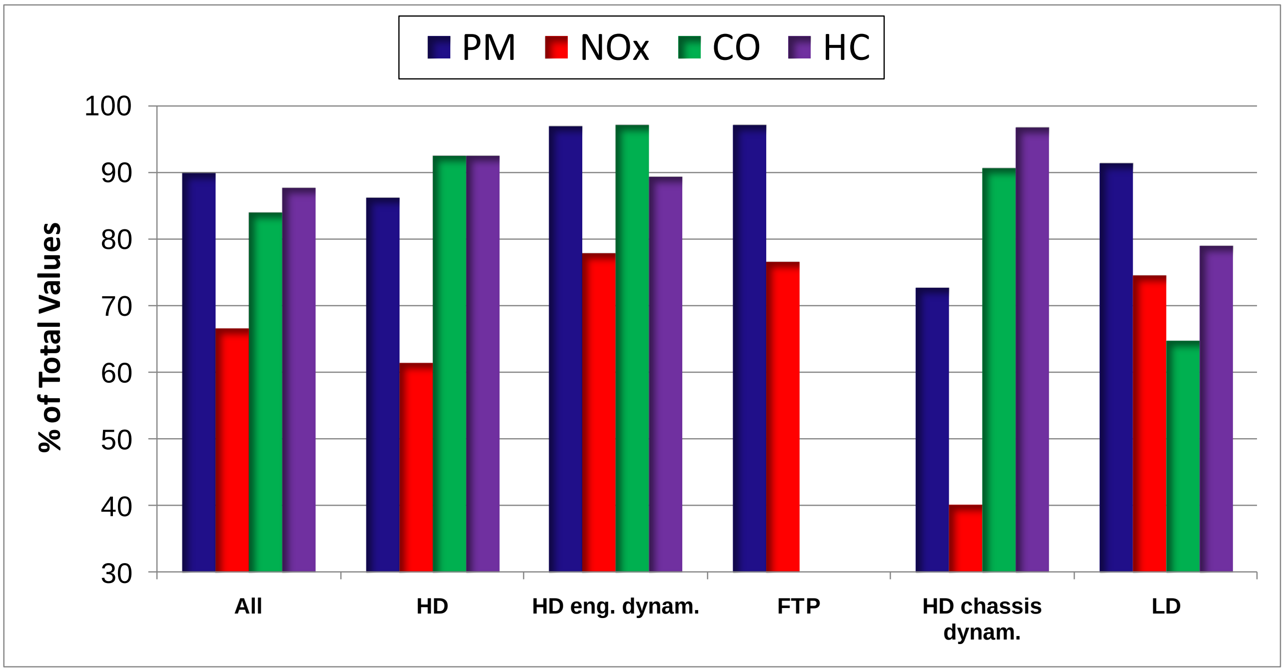

Fig. 13 – Percentage of total negative (PM, CO, HC) or positive (NO_x) measurement values for each engine cycle and engine type.

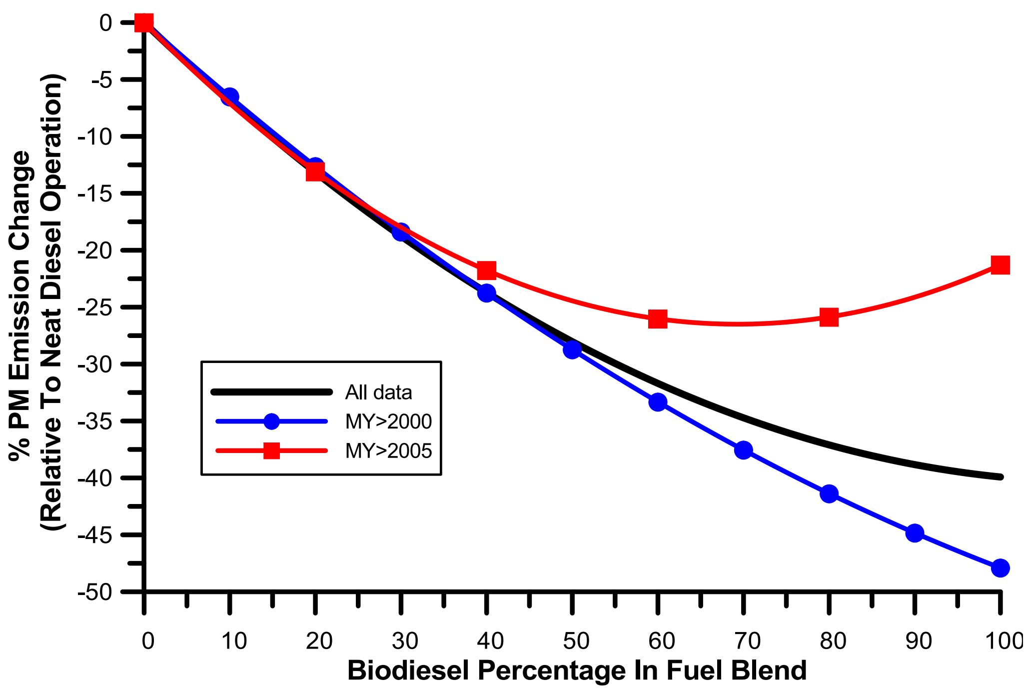

Fig. 14 – Comparative PM emission change best-fit curves for various engine model years.

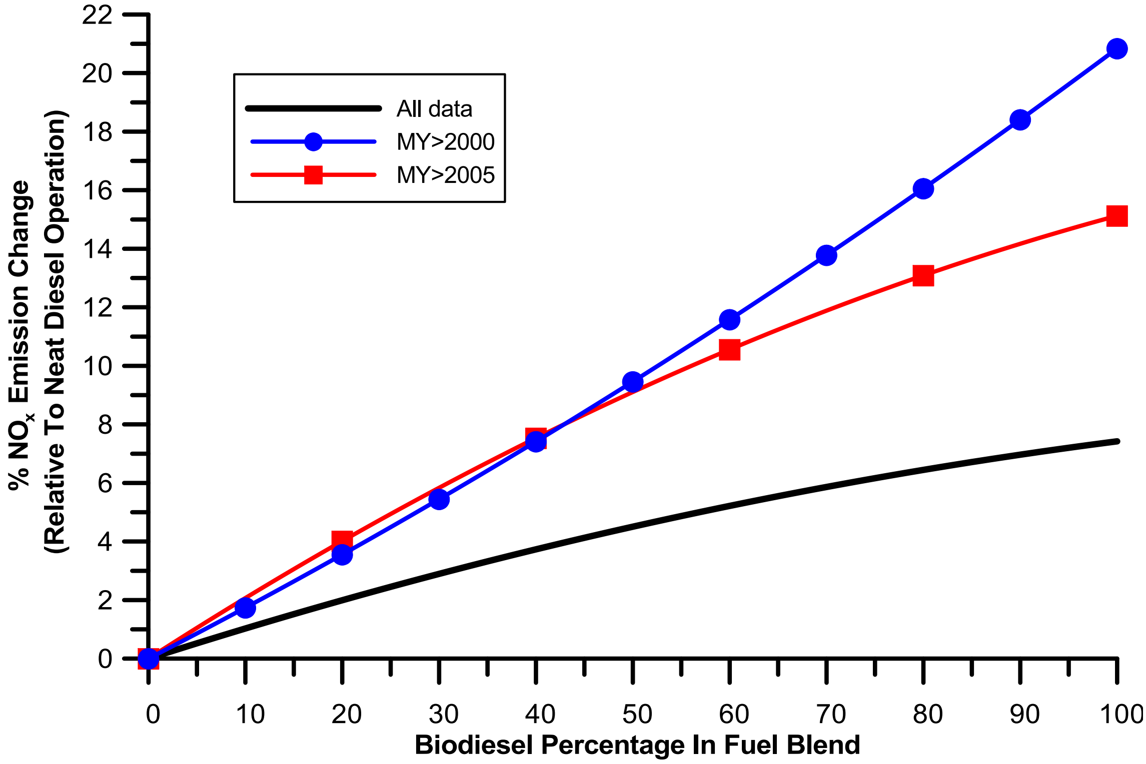

Fig. 15 – Comparative NO_x emission change best-fit curves for various engine model years.

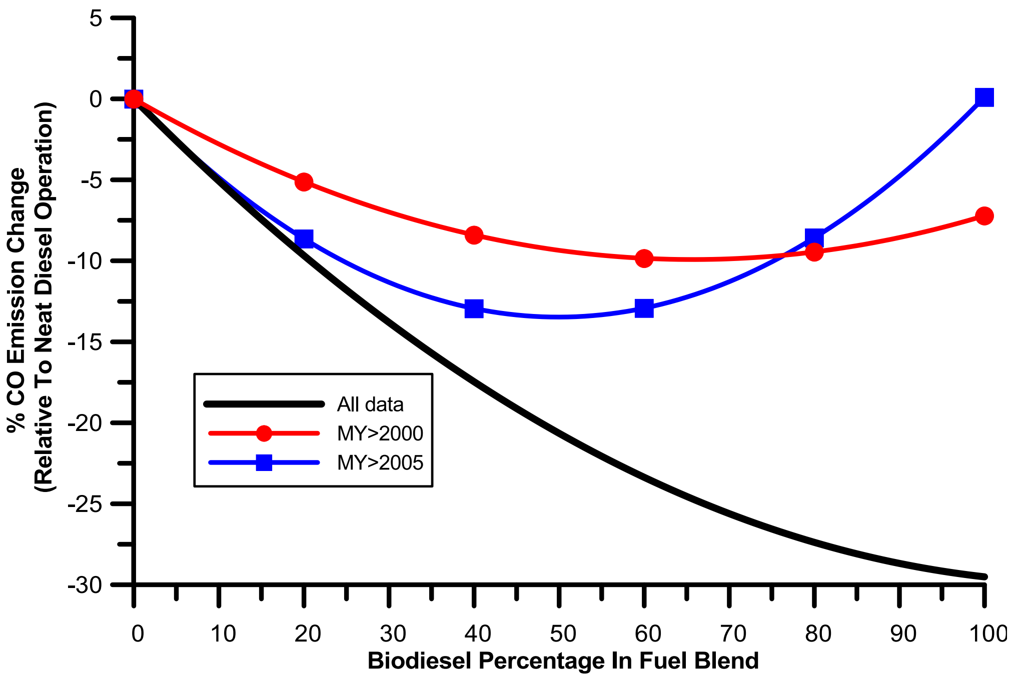

Fig. 16 – Comparative CO emission change best-fit curves for various engine model years.

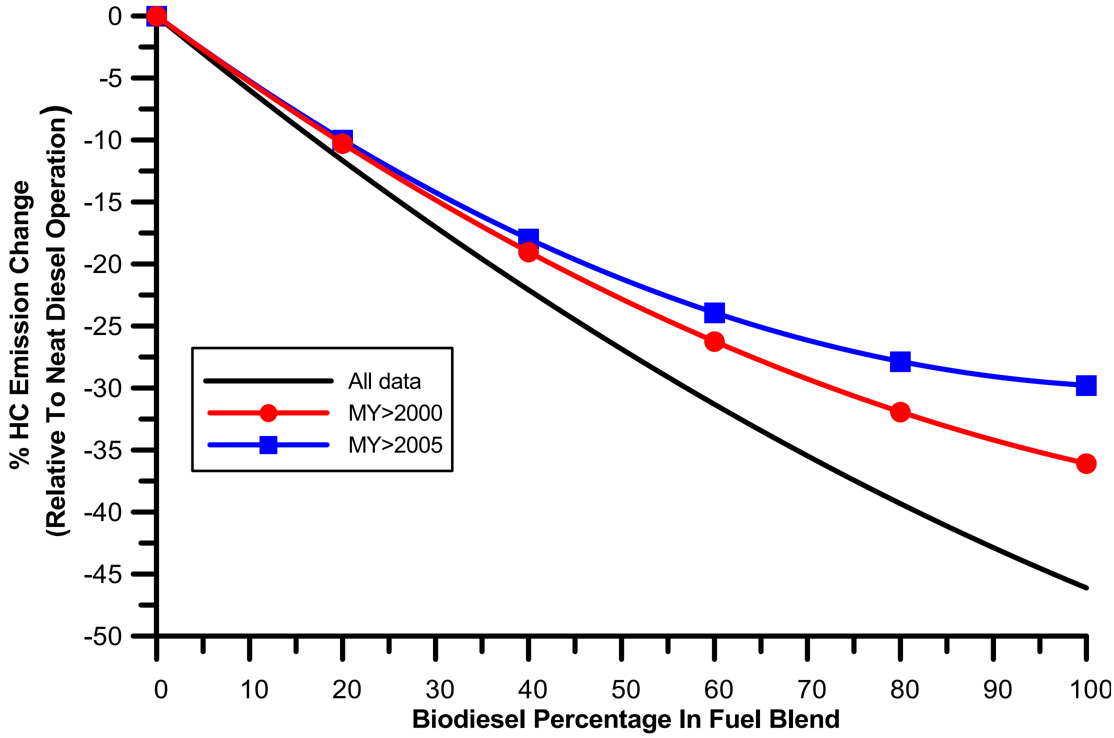

Fig. 17 – Comparative HC emission change best-fit curves for various engine model years.

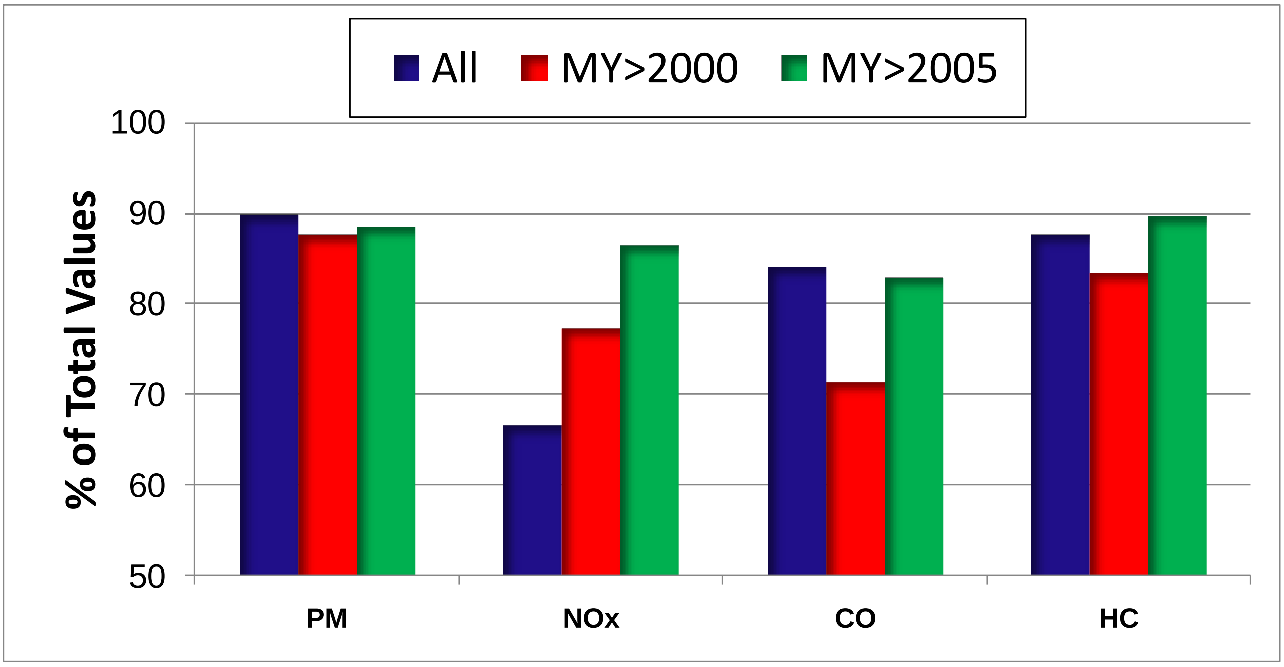

Fig. 18 – Percentage of total negative (PM, CO, HC) or positive (NO_x) measurement values for engine model year.

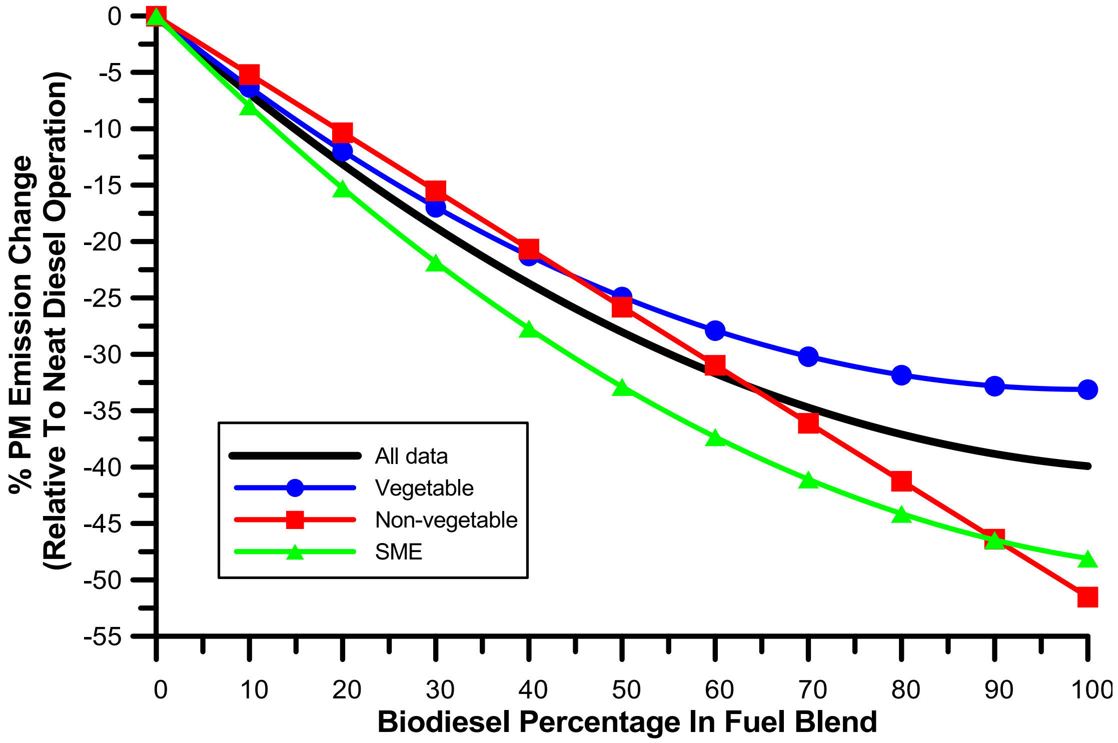

Fig. 19 – Comparative PM emission change best-fit curves for various biodiesel feedstocks.

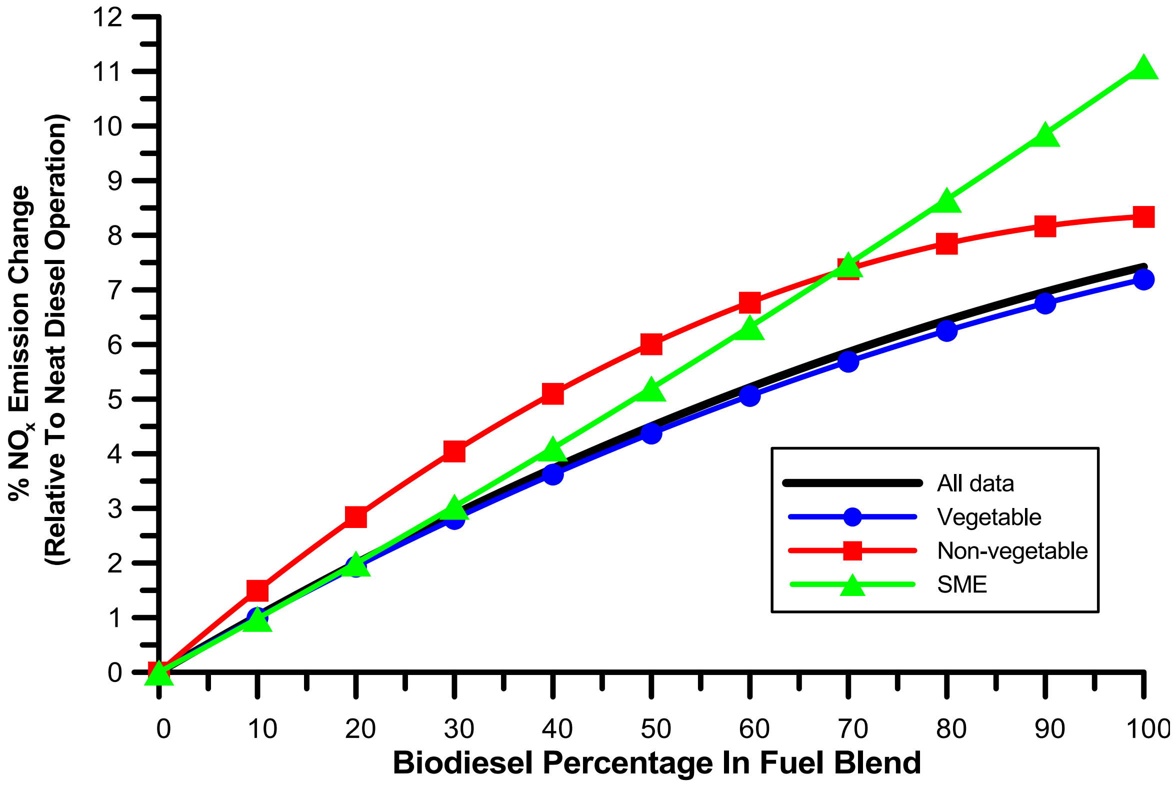

Fig. 20 – Comparative NO_x emission change best-fit curves for various biodiesel feedstocks.

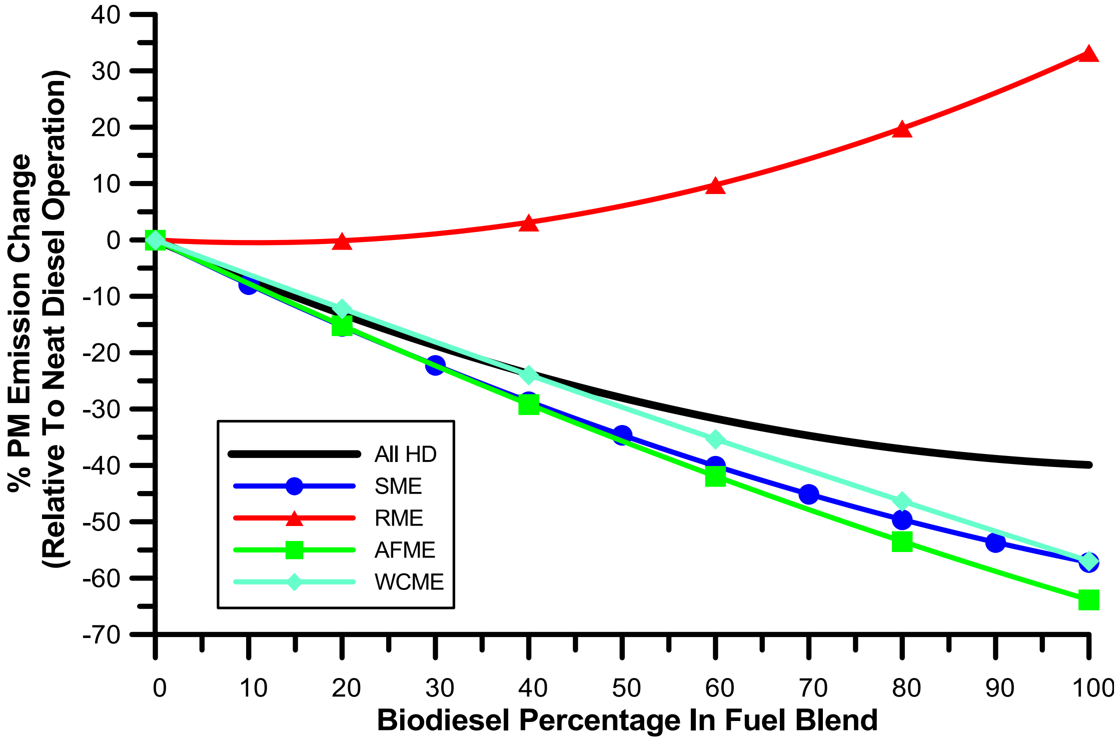

Fig. 21 – Comparative PM emission change best-fit curves for HD engines and for various biodiesel feedstocks.

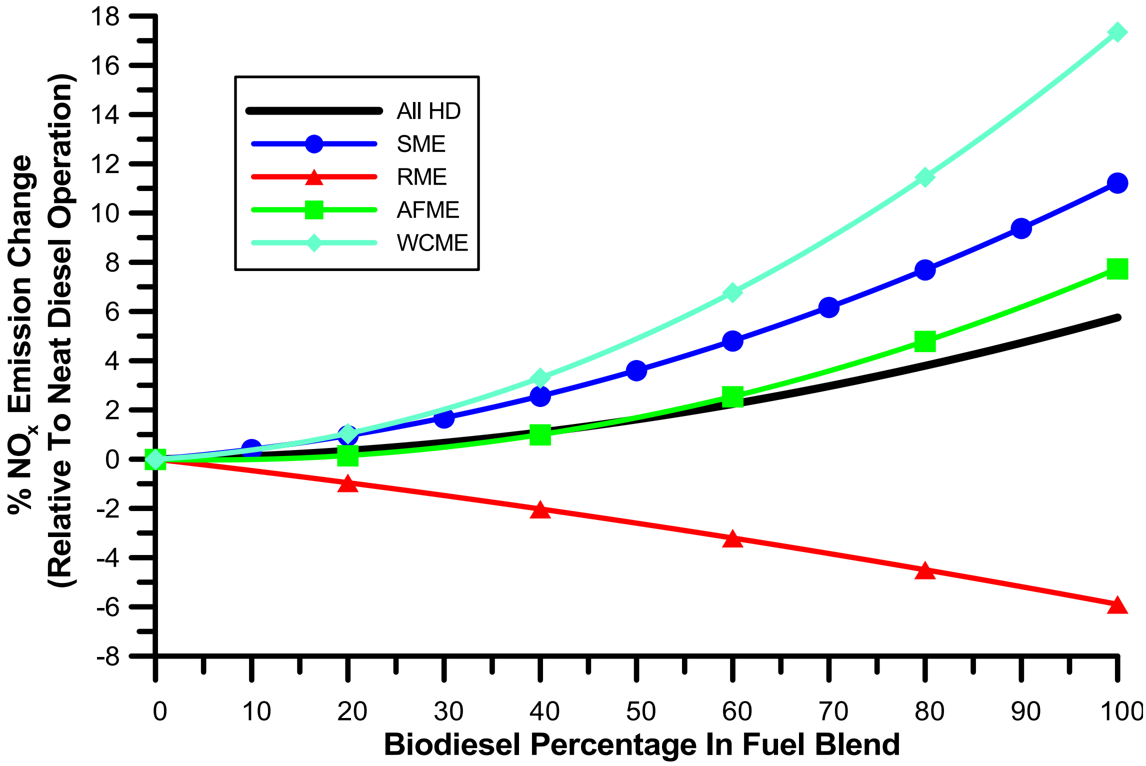

Fig. 22 – Comparative NO_x emission change best-fit curves for HD engines and for various biodiesel feedstocks.

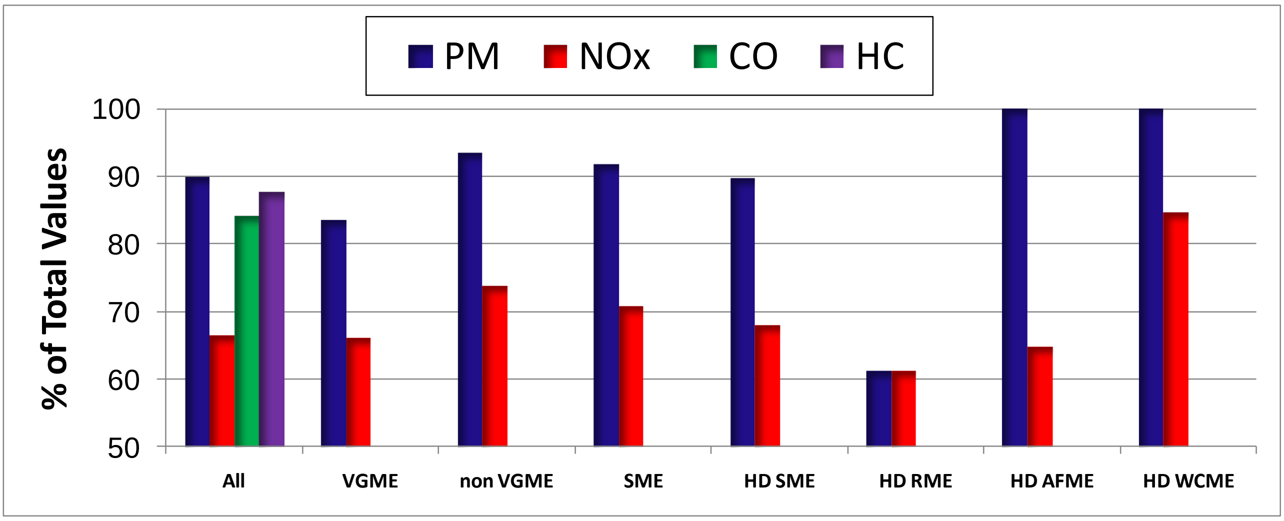

Fig. 23 – Percentage of total negative (PM, CO, HC) or positive (NO_x) measurement values for each biodiesel feedstock.

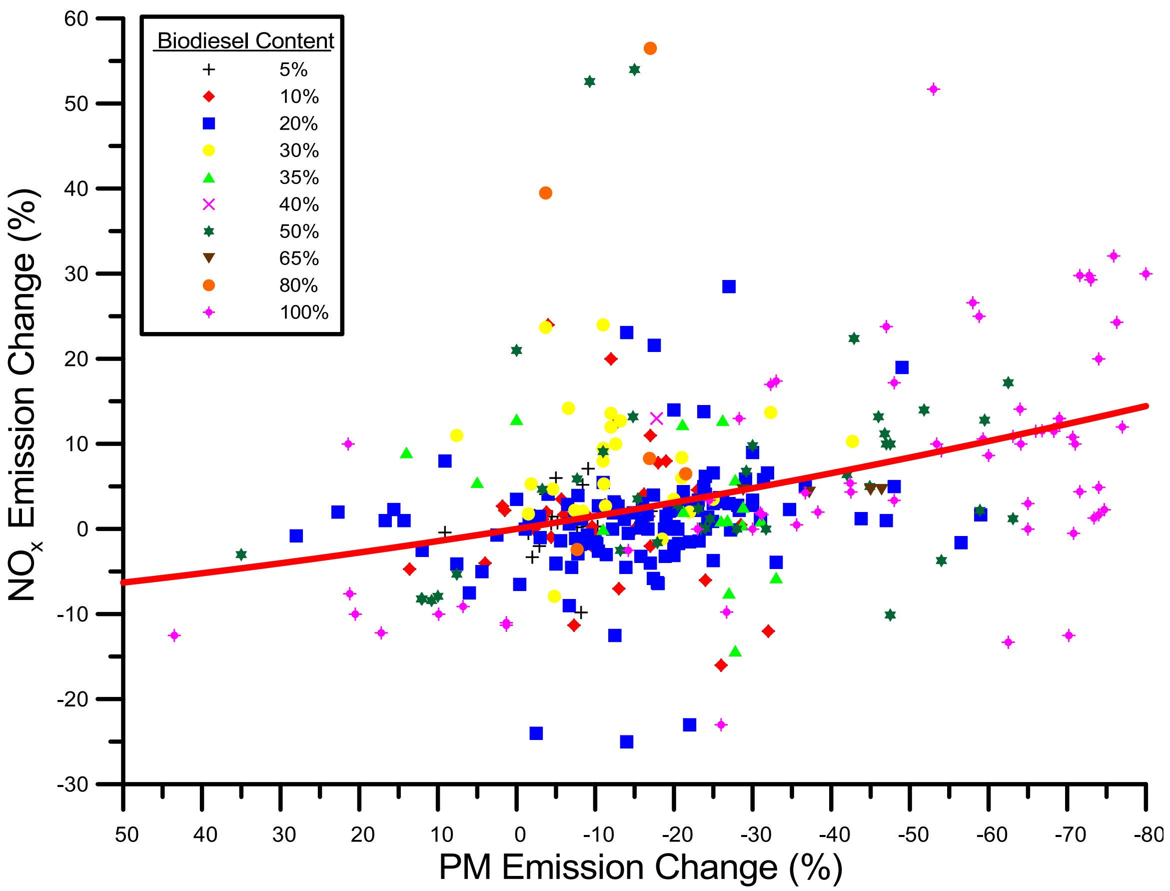

Fig. 24 – Collective PM/NO_x trade-off from all biodiesel blend ratios and transient cycles.

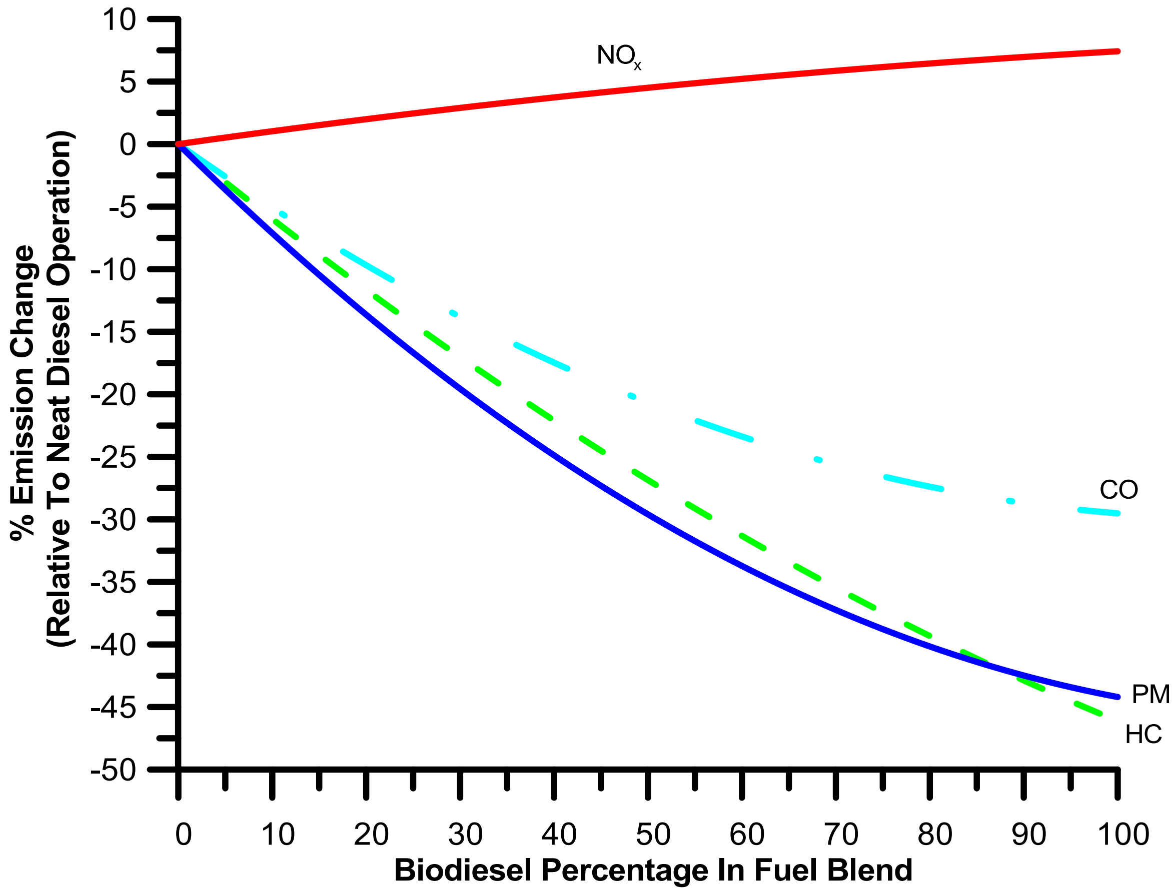

Fig. 25 – Collective results of biodiesel effects on regulated pollutants from all transient cycles up to 2011.