Application of multi-criteria decision making methods to compression ignition engine efficiency and gaseous, particulate and greenhouse gas emissions

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Abstract

Compression ignition (CI) engine design is subject to many constraints which presents a multi-criteria optimisation problem that the engine researcher must solve. In particular, the modern CI engine must not only be efficient, but must also deliver low gaseous, particulate and life cycle greenhouse gas emissions so that its impact on urban air quality, human health, and global warming are minimised. Consequently, this study undertakes a multi-criteria analysis which seeks to identify alternative fuels, injection technologies and combustion strategies that could potentially satisfy these CI engine design constraints. Three datasets are analysed with the Preference Ranking Organization Method for Enrichment Evaluations and Geometrical Analysis for Interactive Aid (PROMETHEE-GAIA) algorithm to explore the impact of 1): an ethanol fumigation system, 2): alternative fuels (20 % biodiesel and synthetic diesel) and alternative injection technologies (mechanical direct injection and common rail injection), and 3): various biodiesel fuels made from 3 feedstocks (i.e. soy, tallow, and canola) tested at several blend percentages (20-100 %) on the resulting emissions and efficiency profile of the various test engines. The results show that moderate ethanol substitutions (~20 % by energy) at moderate load, high percentage soy blends (60-100 %), and alternative fuels (biodiesel and synthetic diesel) provide an efficiency and emissions profile that yields the most "preferred" solutions to this multi-criteria engine design problem. Further research is, however, required to reduce Reactive Oxygen Species (ROS) emissions with alternative fuels, and to deliver technologies that do not significantly reduce the median diameter of particle emissions.

1. Introduction

The CI, or diesel, engine is both a reliable and durable internal combustion engine type that is used ubiquitously for a range of on-road and off-road transportation purposes. The CI engine offers many design advantages compared to its spark ignition (SI), or petrol, engine counterpart (1). Since the combustion process is initiated by compression, rather than from an electrical discharge from a spark plug, CI engines can be operated with a higher compression ratio relative to SI engines. The benefit of a higher compression ratio is that CI engines have a higher thermal efficiency compared to SI engines. In addition, since CI engines are not throttled, they typically operate under lean air-fuel ratios; which reduces emissions of carbon monoxide (CO) and hydrocarbons (HCs). In direct comparison, SI engines typically operate under near stoichiometric air-fuel ratios which exacerbates CO and HC emissions. Despite these design advantages, CI engines are noisy, have higher nitrogen oxide (NOx) emissions than SI engines, and without after-treatment, emit significantly more particulate matter from the tailpipe. As a result, modern CI engine design is confronted with many challenges that aim to minimise its impact on human health, urban air quality, and global climate (2).

From the noted list of CI engine disadvantages, the issue of Diesel Particulate Matter (DPM) emissions has been the subject of considerable research and development, and at present remains an unresolved problem (3). Whilst alternative fuels, injection technologies, and combustion strategies can be used as a tool to reduce DPM emissions, investigating these technologies could improve other aspects related to the impact of CI engines, such as: reducing gaseous emissions, improving engine efficiency, and in the case of alternative fuels, their implementation could significantly reduce their environmental footprint (or life cycle greenhouse gas emissions). Whilst advanced after-treatment such as Diesel Particulate Filters and Diesel Oxidation Catalysts have a profound impact on the particulate emissions profile from a test engine (4, 5), this study considers "raw exhaust" emissions properties only. Thus we leave consideration of this optimisation problem with after-treatment as a topic for future investigation.

Life cycle analysis (LCA) of a transport fuel (also known as wells-to-wheels emissions) takes into account the total greenhouse gas emissions associated with a given transportation task. A critical point with LCA is that it considers not only tailpipe greenhouse gas emissions, but it also accounts for the pre-combustion greenhouse gas emissions (also known as upstream emissions) associated with extraction, production, transport, processing, conversion and distribution of a given fuel (6). The regulated gaseous emissions discussed above (CO, HCs, and NO) primarily affect human health; however, CI engines are also responsible for emitting greenhouse gases that play a role in global warming. The three greenhouse gases included in the life-cycle assessment were carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O). Whilst technically, N_2O is a nitrogen oxide and CH_4 a hydrocarbon, these compounds are treated separately due to their strong greenhouse forcing potential. Relative to carbon dioxide (global warming potential of 1), N_2O has a 100 year global warming potential of 298 and CH_4 has a global warming potential of 25 (7). To minimise the environmental footprint of transportation fuels, clearly, reductions in both tailpipe and upstream greenhouse gas emissions are desired.

To assess whether alternative fuels, injection technologies and combustion strategies can improve the gaseous, particulate and life-cycle greenhouse gas emissions profile from a CI engine, whilst simultaneously maintaining or improving engine efficiency, offers a multi-faceted, multi-criteria optimisation problem that the engine researcher must solve. The Operations Research (OR) literature is populated with techniques that are able to provide "optimal" solutions to problems characterised by a uni-variate (or multi-variate) objective function which requires optimisation subject to several constraints (8). However, in multi-criteria problems the notion of an "optimal" solution breaks down, meaning that the decision maker is not able to use the standard techniques available in the OR literature (such as the simplex method and derivative based optimisation techniques) (9). A way around this problem is to utilise techniques based on preference rather than "optimal" solutions.

There are a variety of multi-criteria decision analysis (MCDA) algorithms which vary in complexity from the elementary (e.g. weighted sum method) to methods which include the notion of outranking (10); such as ELECTRE (ELimination Et Choix Traduisant la REalité), PROMETHEE and REGIME. A review of the MCDA literature revealed that the PROMETHEE-GAIA approach proved quite useful in environmental applications (11). A significant advantage of the PROMETHEE-GAIA algorithm (compared to other MCDA methods) is that it facilitates a rational decision making process. This is achieved by virtue of a decision vector that directs the decision maker towards "preferred" solutions (12).

This study applies the PROMETHEE-GAIA algorithm to 3 experimental datasets that explored alternative fuels, injection technologies, and combustion strategies to assess the impact of these techniques on the gaseous, particulate and life-cycle greenhouse gas emissions from CI engines. Engine efficiency was monitored during all experiments, enabling the impact of these techniques on brake thermal efficiency to be assessed. Consequently, this study aims to identify potentially viable engine technologies that may simultaneously enable various design constraints (i.e. gaseous, particulate, and life-cycle greenhouse gas emissions, and engine efficiency) to be satisfied.

2. Methods

2.1 The PROMETHEE-GAIA algorithm

The theory of the PROMETHEE-GAIA algorithm is well described in the literature, and the interested reader may consult the works conducted by the developers (such as Brans and Vincke (9), Brans et al. (13), Mareschal and Brans (14), Brans and Mareschal (12)) for a detailed explanation of its capabilities. A thorough discussion of the theoretical underpinnings of the PROMETHEE-GAIA algorithm is also provided in the supporting information of this manuscript. The Decision Lab Software (Decison Lab, 2000) was used to perform all the quantitative analysis undertaken in this manuscript.

2.2 The datasets analysed

The gaseous and particulate emissions datasets investigated in this study were the subject of several recent publications (15-18). In this above set of studies, gaseous emissions, engine efficiency and the physico-chemistry of DPM emissions were investigated in 3 distinctly different experimental study designs. Note that all datasets were collected using a sub-set of test modes from the ECE R49 test cycle. Thus, the optimisation described in this manuscript applies to steady-state test modes only. The authors therefore leave the optimisation of emissions and performance for real-world transient test cycles for future studies.

In the first study, an ethanol fumigation system was equipped to a direct injection diesel engine (17, 18). The objective of that study was to investigate the impact of an alternative fuel (ethanol) and an alternative combustion strategy (fumigation - a type of pre-mixed compression ignition) on the resulting emissions profile from the test engine.

In the second study, an engine was tested with 2 injection configurations (mechanical direct injection, and common rail injection) and 3 fuels (ultra-low sulphur diesel (ULSD), 20% biodiesel blend, and a synthetic diesel) to explore the role of fuel type and injection configuration on the emission profile from the test engine (16).

The third study investigated the role of different biodiesel fuels made from different feedstocks (i.e. soy, tallow, and canola) tested at a range of different blend percentages (20-100 %), using the results from ULSD as baseline for comparison on the emissions profile from the test engine (15).

Table 1 provides an overview of the experimental data which formed the basis for the application of the PROMETHEE-GAIA algorithm. The following pollutants were measured in the 3 studies listed above (15-18); namely: regulated emissions (CO, HCs, NO, and PM), unregulated emissions (particle number emissions, particle surface area emissions, Count Median Diameter (CMD) (all derived from a Scanning Mobility Particle Sizer distribution), ROS concentrations, and the organic volume percentage of particles which is defined as the percentage of particle volume that evaporates upon heating with a thermodenuder set to 300 ^{o}C. Other unregulated emissions that were measured included measuring: the percentage of volatile particles (PVP) for the ethanol fumigation study, and measuring either total polycyclic aromatic hydrocarbon emission (mg/kWh) (alternative fuels and injection technologies study), or measuring particle phase and vapour phase polycyclic aromatic hydrocarbons separately (alternative fuels study with 3 biodiesel feedstocks).

Table 1: An overview of the measurements conducted in the 3 datasets analysed with the PROMETHEE-GAIA algorithm. Each column refers to a different study, and each row refers to a different CI engine parameter that was measured or calculated. "X" denotes that a given parameter was measured or calculated in a particular study, whereas "-" denotes that this parameter was not measured or calculated. Note that V-TDMA stands for Volatilisation-Tandem Differential Mobility Analyser.

	Study number		
	1	2	3
Techniques investigated	**Alternative fuels** (ethanol) **Alternative combustion strategy** (fumigation)	**Alternative fuels** (biodiesel, synthetic diesel) **Alternative injection**	**Alternative fuels** (biodiesel fuels made from soy, tallow and

			technologies (mechanical direct injection, common rail injection)	canola tested at different blend percentages 20-100 %)
Regulated emissions	Carbon monoxide (CO) (g/kWh)	X	X	X
	Hydrocarbons (HCs) (g/kWh)	X	X	X
	Nitric oxide (NO) (g/kWh)	X	X	X
	Particulate Matter (PM) (g/kWh)	X	X	X
Unregulated emissions	Particle number emissions (#/kWh)	X	X	X
	Count median diameter (CMD) (nm)	X	X	X
	Particle surface area (nm^2/cm^3)	X	X	X
	Reactive oxygen species (ROS) (nmol/mg)	X	X	X
	Particle organic volume percentage (%)*	X (Only determined at 80 nm with a V-TDMA system)	X	X
	Percentage of volatile particles (%) (PVP)	X	-	-
	Total polycyclic Aromatic Hydrocarbons (PAHs) (mg/kWh)	-	X	X
	Particle phase polycyclic Aromatic Hydrocarbons (PAHs) (mg/kWh)	-	-	X
	Vapour phase polycyclic Aromatic Hydrocarbons (PAHs) (mg/kWh)	-	-	X
Engine performance	Brake thermal efficiency (-)	X	X	X
	Brake specific energy consumption (MJ/kWh)	X	X	X
	Pre-mixed combustion percentage (%)	X	-	-
	Diffusion flame combustion	X	-	-

	percentage (%)			
	Indicated power (kW)	X	-	-
	Indicated mean effective pressure (kPa)	X	-	-
	Maximum rate of pressure rise (kPa/deg)	X	-	-
	Maximum pressure (kPa)	X	-	-
Sustainability measure	Life-cycle greenhouse gas emissions (g/km)	X	X	X

Engine performance measures were also recorded during all 3 experiments. The two engine performance measures calculated were brake thermal efficiency and Brake Specific Energy consumption (BSEC).

The brake thermal efficiency was calculated according to (1):

$$\eta_f = \frac{P_b}{\dot{m}_f Q_{LHV}}, \qquad [1]$$

where P_b is the brake power output of the engine (measured with a dynamometer), $\dot{m}_f$ is the fuel mass flow rate, and Q_{LHV} is the lower heating value of the fuel.

BSEC (MJ/kWh) was calculated according to:

$$BSEC = \frac{Q_{LHV} \times \dot{m}_f \times 3600}{P_b}. \qquad [2]$$

In the first study (ethanol fumigation study), access to in-cylinder pressure versus crank angle data was available. By undertaking a combustion analysis with the AVL Boost program (19), heat release versus crank angle could be computed using the first law of thermodynamics. This enabled the percentage of heat released in the pre-mixed, and diffusion flame phases of combustion to be computed. These two engine performance measures were included in the first study to assess their impact on the overall emissions profile.

Another quantity that was computed in all 3 experiments was life-cycle greenhouse gas emissions for each transportation fuel type investigated. Upstream greenhouse gas emissions (i.e. pre-combustion) estimates for N_2O, CH_4 and CO_2 associated with extraction, production, transport, processing, conversion and distribution of various Australian alternative fuels were taken from published data by Beer et al. (20) and Beer et al. (21). The same information source was used to obtain tailpipe (i.e. post combustion) emissions for N_2O and CH_4. Tailpipe CO_2 emissions factors were calculated using the data collected as part of the 3 studies described above. The total life cycle greenhouse gas emissions (g/km) were obtained by multiplying the raw emissions factor by its IPCC Global Warming Potential factor (7) and then summing the results for the pre-combustion (i.e. upstream) and post-combustion (i.e. tailpipe) emissions contributions.

Parameters listed in Table 1 that were maximised (i.e. higher values were preferred) mainly relate to engine performance measures such as: brake thermal efficiency, the pre-mixed combustion percentage, indicated power, indicated mean effective pressure, the maximum rate of pressure rise, and the maximum pressure. However, the count median diameter of particles was also maximised to prevent giving preference to engine technologies which could lead to problems of smaller particles (typically with a high organic content) depositing in the human respiratory tract, since these types of particles are heavily implicated in respiratory and cardiovascular health effects (22). All other criteria were minimised (i.e. lower values were preferred) as they relate to gaseous, particulate and life-cycle greenhouse emissions which the engine designer wants to eliminate. The decision made about how to treat all the different criteria (i.e. maximise or minimise) gives an indication of the overall "metric" that needs to be optimised to satisfy CI engine design requirements. In the absence of any information suggesting that one criterion is quantitatively more important than another; all criteria were weighted equally. However, a sensitivity analysis was also performed to test how the outcomes were influenced by the choice of different preference functions (see Table S4) and different weights for criteria (see Figures S3-S5).

Overall, the sensitivity analysis tests for the sensitivity of our results with respect to:

I. The choice of preference function, and
II. The relative weighting given to different criteria.

Part I employs 3 preference functions, namely: the V-shaped, linear, and "usual" preference functions. This set of 3 preference functions were selected as they are available for modelling quantitative variables. The other 3 preference functions in the PROMETHEE-GAIA software are for modelling qualitative variables which do not feature in this analysis.

As can be seen in Table S4, regardless of the preference function used, the outranking order is similar (i.e. the most preferred and least preferred alternatives are almost always the same) for a particular study and preference function-by-weighting regime. This implies that the rank orders and decision outcomes of these studies are robust.

Part II applies 3 different weightings for each preference function selected. The 3 different weightings are:

a. Equal weighting for all criteria, which is the default setting. As explained in the methods section, in the absence quantitative information driving the relative weighting of criteria, this is a logical weighting to apply. This scenario could be applicable for a Chemical Engineer who wishes to make sure that alternative engine technologies satisfy all criteria.

b. Double weighting for emissions criteria, with all other criteria weighted as per equal weights. This scenario could be applicable for an Environmental Engineer/Scientist who gives greater weighting to emissions due to pollution concerns.
c. Default weighting for emissions, double the weighting for all other criteria. This scenario could be applicable for a Mechanical Engineer concerned with making sure that CI engines are fuel efficient and economical.

In total, 9 (i.e. 3 x 3) sensitivity results are computed for each study. As the manuscript features 3 different studies, the supplementary information has 27 sensitivity analysis runs performed. Overall, for a particular preference function and study, application of the 3 different weighting schemes had limited impact on the rank order, which again reinforced the robustness of the decision outcomes.

3. Results and discussion

Figure 1 shows the PROMETHEE II outranking for all 3 datasets, which involves ranking the alternatives (for all 3 studies) from most preferred to least preferred based on the value of their net outranking flow (ϕ). Note that Tables S1 and S2 (in the supporting information) provide a full listing of the abbreviations used for alternatives and criteria, along with information on how each criterion is treated by the PROMETHEE-GAIA algorithm (i.e. maximised or minimised).

For the first, second and third studies, the range of ϕ is 0.63, 0.13, and 0.26 and respectively. The range of ϕ gives an indication of how well the PROMETHEE II outranking can distinguish amongst alternatives and indicate preferred engine technologies. Whilst it is noted that the alternatives are very closely grouped for the second study, some clear differences in ranking are observed for the first, and especially the third study.

Investigation of the PROMETHEE II outrankings for each study generally shows a weak preference for alternative fuels, injection technologies, and combustion strategies. In the first study (despite a large ϕ range), a mixed trend is exhibited for preferences involving ethanol fumigation with respect to load. Ethanol fumigation is preferred at idle for the E10 test, and for E40 at full load; however, the strength of these preferences (based on the outranking flow differential) are quite small, being 0.02 and 0.05 respectively. Conversely, neat diesel operation is preferred at half load, with a net outranking flow differential of 0.11 relative to E40 fumigation at this load setting. Alternatively, the net outranking flows are numerically equal (to 3 significant figures) at the quarter load setting. Based on the net outranking flows, neither strong preferences nor strong non-preferences emerge from the analysis.

For the second study, no strong preferences for alternative fuels or injection technologies emerged from the analysis. It is interesting to note; however, that if the net outranking flows are compared for each speed and load setting individually, that neat diesel operation (for both mechanical direct injection and common rail injection) occupies the lowest 2 ranks (out of 6) for 3 of the 4 test settings. An exception to this trend is the intermediate speed and half load setting, where the neat diesel setting occupies the 2nd (mechanical direct injection) and 6th rank (common rail injection). Whilst the net outranking flow differentials are small, there appears to be a slight preference for the implementation of alternative fuels in the second study, with the fuels being ordered in the following fashion: biodiesel > synthetic diesel > ultra low sulphur diesel (ULSD). As for the effect of injection technology, no clear trends emerge from the analysis over the 4 point test cycle investigated.

In the third study, some moderately strong preferences emerge for alternative fuels made from different biodiesel feedstocks. Out of the 14 fuel types investigated, 9 of the 13 biodiesel fuels are preferred to ULSD, and 4 of the biodiesel fuel types are less preferred to ULSD. Furthermore, the net outranking flow differential between the most preferred fuel

(B100 soy) and ULSD is 0.21, which indicates the strength of this preference. A feedstock dependency is also evident in the preference structure, as 4 of the 5 soy blends are preferred to ULSD, whilst only 3 tallow blends, and 2 canola blends are preferred to ULSD. Thus, there appears to be a preference for soy blends, and furthermore, the strength of the preference increases as the blend percentage is increased.

Overall, the PROMETHEE II outranking results only show moderately strong preference for the implementation of alternative fuels for the third study involving biodiesel. Conversely though, the results show that strong non-preference does not result from the implementation of alternative engine technologies (i.e. fuels, injection technologies, and combustion strategies); which is a promising result. The next step in the analysis is to analyse in more detail the test settings (e.g. speed, load, fuel etc) that are associated with strong preferences or non-preferences; a topic which is discussed subsequently.

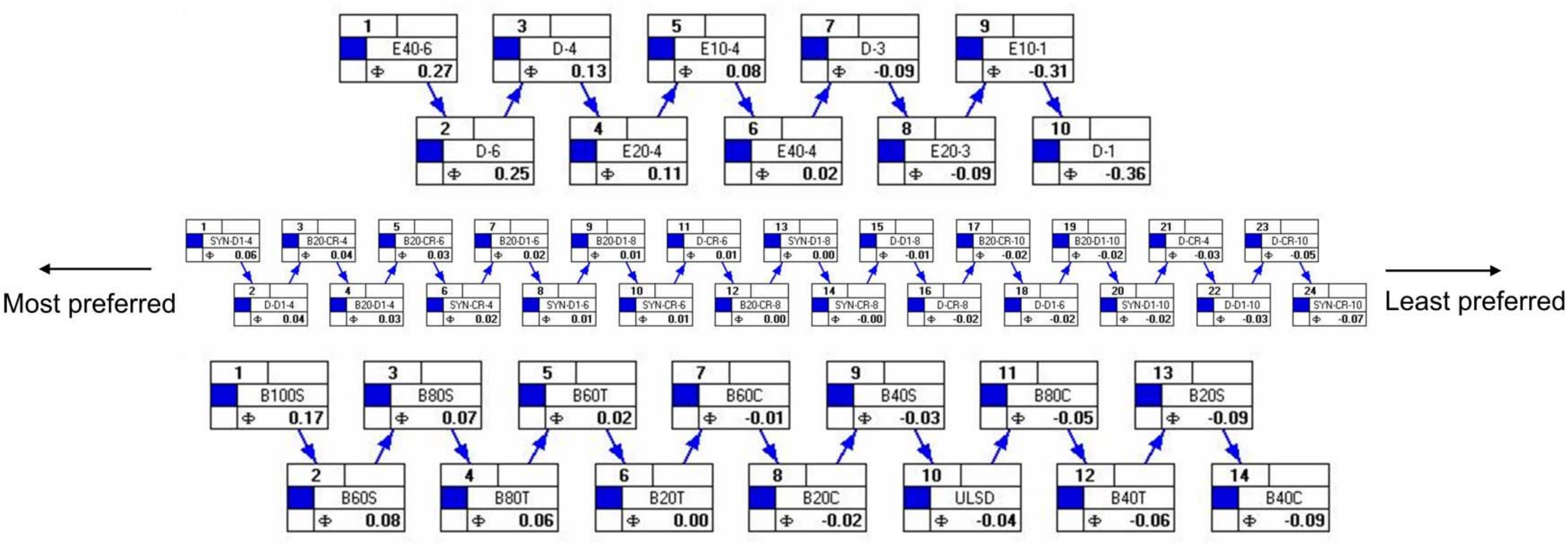
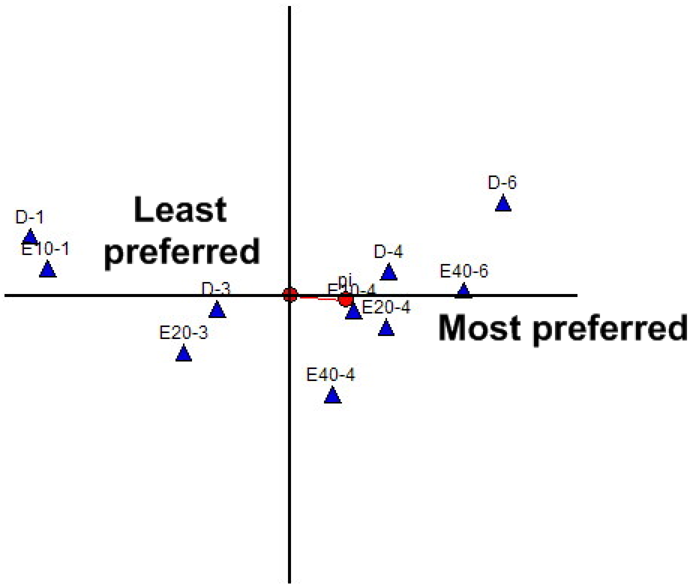


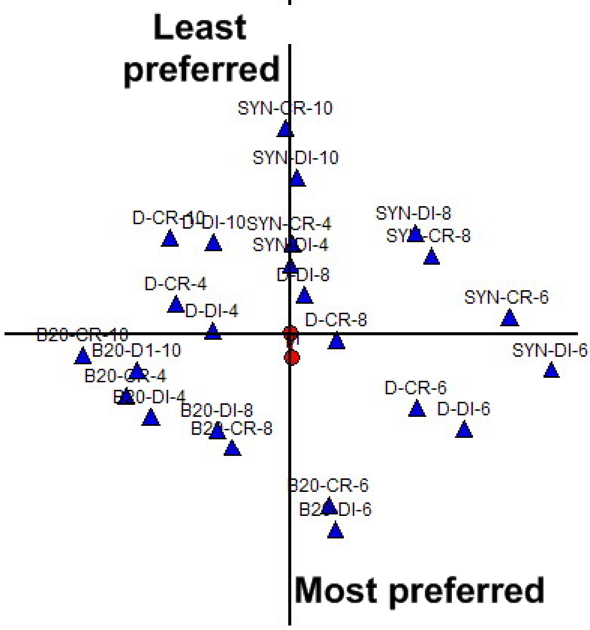
Figure 1: PROMETHEE II outranking for all 3 datasets. Top panel: alternative fuels and combustion strategy study, middle panel: alternative fuels and injection technologies study, bottom panel: alternative fuels study. Note well that the top number in each box indicates the rank of the alternative, where 1 is most preferred and $\boldsymbol{n}$ is least preferred, the middle box indicates the alternative, and the lower number gives the value of the net outranking flow $\boldsymbol{\phi}$ for each alternative.

Figure 2 shows an alternatives plot for each of the 3 studies. Note that a 2 dimensional view of alternatives (Figure 2) and criteria (Figure 3) for a multi-dimensional problem is obtained using Principal Component Analysis, along with a decision vector which is a weighted combination of the alternatives of the problem. Figure S2 in the supplementary information provides extra detail on how to interpret results geometrically in the GAIA plane.

In interpreting these results, the length of the decision vector (pi) is critical, as a longer decision vector indicates greater decision making power (23). From Figure 2, we observe that the decision vector is short for the second study, but a longer decision vector is observed with the first, and especially the third study.

Inspection of the alternatives plot for the first study (top panel) clearly shows the load-dependent nature of operating a CI engine, both with, and without, ethanol fumigation. Tests conducted at idle and quarter load are located on the left hand side of the alternatives plot, in a direction roughly opposite (i.e. 135-225 ^{o}) to that of the decision vector. Alternatively, tests conducted at half and full load are located on the right hand side of the alternatives plot, in a direction roughly parallel (i.e. ± 45 ^{o}) to the decision vector. Furthermore, it can be noted that the E20 test at half load, and the E40 test at full load, both lie roughly in the direction of the decision vector far from the origin. Thus, the PROMETHEE GAIA algorithm suggests that these settings are preferable for the implementation of ethanol fumigation.


Least preferred
Most preferred
D-6
D-1
E10-1
D-4
E40-6
D-3
pi
E20-4
E20-3
E40-4


Least preferred
SYN-CR-10
SYN-DI-10
SYN-DI-8
D-CR-10
D-DI-10
SYN-CR-4
SYN-CR-8
SYN-DI-4
D-DI-8
D-CR-4
SYN-CR-6
D-DI-4
D-CR-8
B20-CR-10
B20-D1-10
SYN-DI-6
B20-CR-4
B20-DI-4
D-CR-6
D-DI-6
B20-DI-8
B20-CR-8
B20-CR-6
B20-DI-6
Most preferred

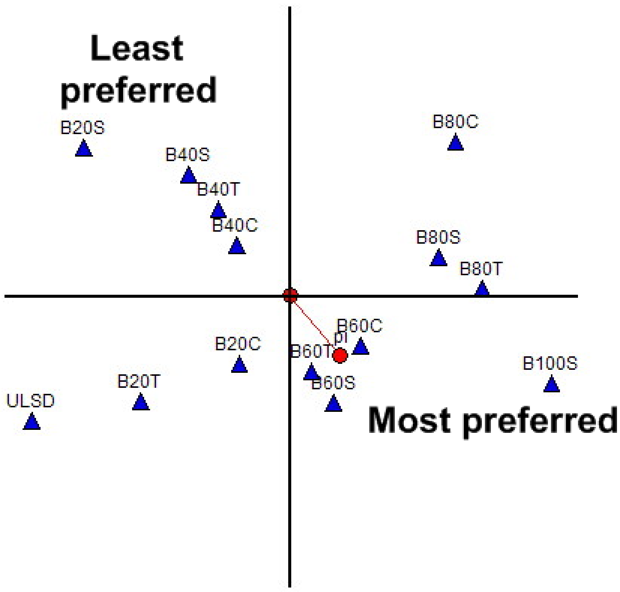

Least preferred
B20S
B80C
B40S
B40T
B40C
B80S
B80T
B60C
B20C
B60T
pi
B100S
B20T
B60S
ULSD
Most preferred

Figure 2: GAIA plot of alternatives for all 3 datasets. Top panel: alternative fuels and combustion strategy study, middle panel: alternative fuels and injection technologies study, bottom panel: alternative fuels study. Principal components 1 and 2 explained 83.9 % (top panel), 64.9 % (middle panel), and 72.4 % (bottom panel) of the variance in the 3 datasets respectively.

The results from the alternatives plot for ethanol fumigation are valuable for two main reasons. Previous research has suggested that ethanol fumigation is generally more successful under moderate load (i.e. not full or low load) conditions (24), and that when implemented, a 20 % ethanol substitution (by energy) is close to the optimal level of secondary fuel substitution (25). Whilst the E40 test at full load is also located in close proximity to the decision vector, it is difficult to recommend such a high level of ethanol substitution at this load, especially when the corresponding ULSD test at full load (D-6) is also located roughly in the direction of the decision vector much further from the origin.

The analysis also identifies ethanol substitutions that should not be undertaken, such as the 20 % substitution at quarter load (E20-4), and also the 10 % substitution at idle (E10-1). This decision is reached, as these two alternatives are located roughly in the opposite direction of the decision vector (135-225 ^{o}) far from the origin - meaning that these alternatives are in conflict with the proposed engine design “metric” which drives the multi-criteria analysis. From a mechanical perspective, previous research explains this result (24, 25), as the problem of flame quenching is encountered with ethanol fumigation under low load conditions. Overall, the results from this analysis and previous research suggest that ethanol fumigation is best implemented under partial load conditions. The practical significance of this finding is that ethanol fumigation may be a suitable system to implement with engines operating during transient gear changing operations.

In the second study (middle panel), the decision vector is quite short which limits the conclusions that can be drawn. It is interesting to note that distinct "banding" occurs in the alternatives plane, whereby 3 separate lines (or bands) run from the top-left to the bottom right of the alternative planes for each fuel type. Furthermore, it can be noticed how the "band" for biodiesel leads to alternatives located near the bottom of the alternatives plane that lie in the direction of the decision vector far from the origin. Given the nature of the results, the bands for ULSD and synthetic diesel do not coincide with the preferred solution to the problem (i.e. in the direction of the decision vector far from the origin). Overall, only 8 alternatives (out of 24) are located within close proximity to the decision vector; however, it is interesting to note that 5 of these alternatives are for alternative fuels (4 for biodiesel and 1 for synthetic diesel). The 8 alternatives are equally split between direct injection (4) and common rail injection (4). This fact combined with the limited $\boldsymbol{\phi}$ range for the analysis does not enable clear recommendations to be drawn from the second study. However, it can be stated that the engine design "metric" is not considerably worsened by the implementation of alternative fuels and injection technologies – which is a promising result.

In the third study (bottom panel), the decision vector is long and a clear separation of alternatives can be seen in the alternatives plot. Five alternatives are located roughly in the direction of the decision vector ($\pm 45^{o}$) pi. These 5 highly preferred alternatives are: the neat and 60% soy blends, the 60 and 80% tallow blends, and the 60% canola blend. Additionally, all three 40% blends, and the 20% soy blend are located in roughly the opposite direction of the decision vector emerging as less preferred options. All other alternative fuel types are roughly orthogonal to the decision vector; which indicates that these alternatives are not correlated with the criteria against which alternatives are optimised. Overall, a reasonably clear conclusion to emerge from the third study is that high percentage soy blends are preferred at full load for the test engine investigated.

Thus far, different alternative engine technologies (i.e. fuels, injection technologies, and combustion strategies) have been ranked from most-to-least preferred, and the location of different alternatives (with respect to the decision vector) have been analysed. The next aspect to consider in the analysis is to identify which engine parameters (e.g. gaseous/particulate emissions or engine efficiency etc) influence decision making preference. This information is provided by the GAIA plot of the criteria which appears in Figure 3. The first major observation to make is that in the first and third study, the decision vector lies roughly in the direction ($\pm 45^{o}$) that maximises brake thermal efficiency, or conversely minimises BSEC. For the last third study, maximising brake thermal efficiency has the added benefit of reducing the number of particles emitted, and also the number of particles that are likely to deposit in various regions of the human respiratory tract. In the first study, whilst maximising brake thermal efficiency (or minimising BSEC) lies roughly in the direction of the decision vector, a concomitant reduction in particle number emissions did not occur. Particle number increases (with tests involving an efficiency increase) occurred due to nucleation modes being present in the size distributions; therefore, factors other than merely engine efficiency influence the formation of this pollutant.

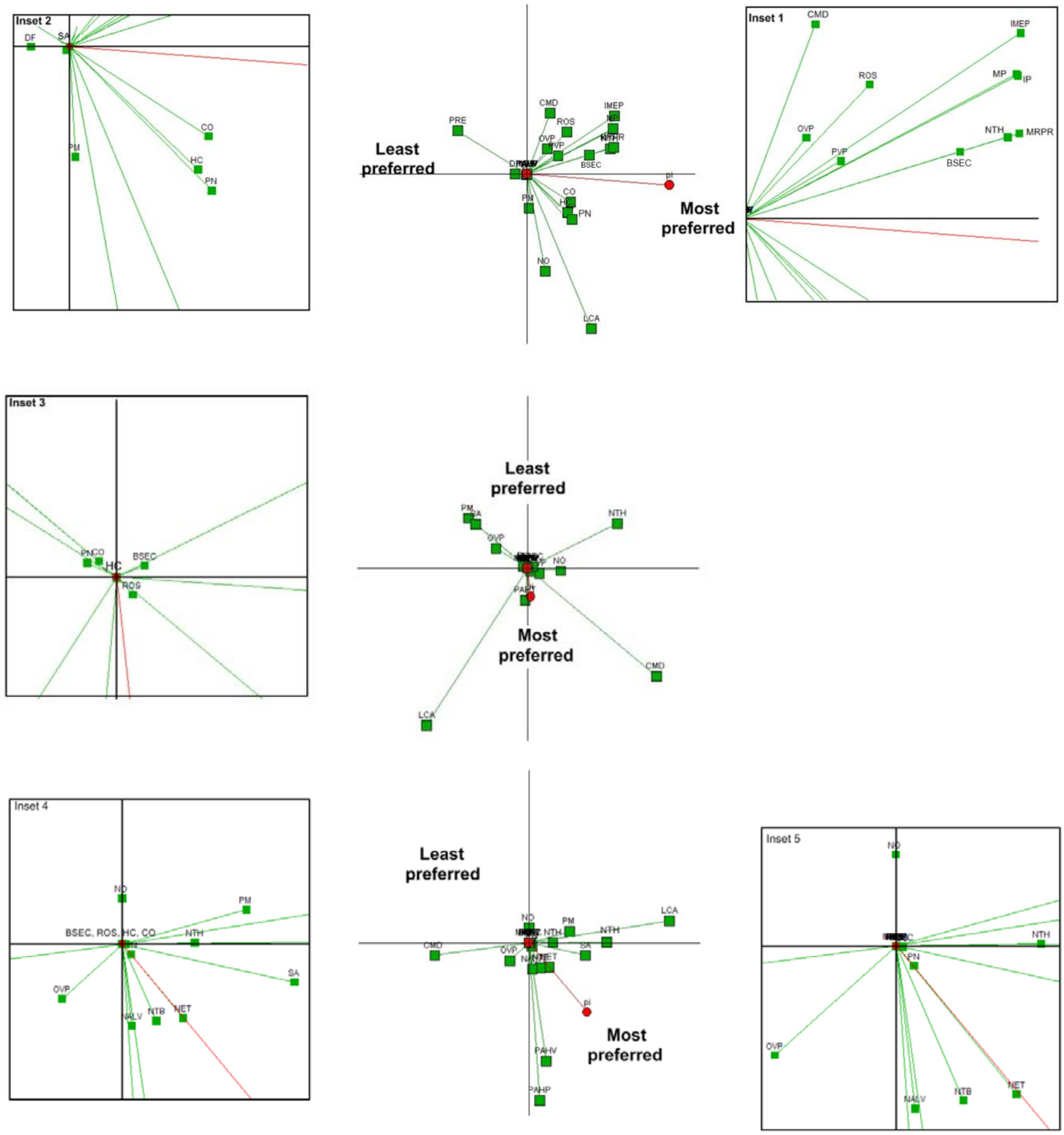


Figure 3: GAIA plot of criteria of the 3 datasets. Top panel: alternative fuels and combustion strategy study, middle panel: alternative fuels and injection technologies study, bottom panel: alternative fuels study. Insets 1 and 2 provide a zoomed in view of the GAIA criteria plot for the top panel, Inset 3 is for the middle panel, and insets 4 and 5 are for the bottom panel.

In the second study, the well-known NOx-PM trade-off can be observed in the GAIA plot for criteria, as NO and PM emissions are roughly in opposing directions in the GAIA plane (135-225 ^{o}). As suggested by Majewski and Khair (2), measures taken to reduce one pollutant (e.g. PM) leads to increases in NO, with the converse case holding true. Another interesting result to emerge from the first and third studies is the strong correlation between ROS concentrations and the organic volume percentage of particles. This result is consistent with recent research that has implicated the organic fraction of DPM in the formation of ROS present on the particle surface (26, 27). Furthermore, in the first study, the organic volume percentage of particles is correlated with the percentage of particles that are purely volatile. As a result, the PROMETHEE-GAIA analysis is consistent with the conclusions drawn in Surawski et al. (17) which stated that the nucleation modes observed with ethanol fumigation were composed of largely organic material.

Several results are evident from the GAIA plot which indicate criteria that are orthogonal (i.e. are un-correlated or independent) with respect to the decision vector for the problem. For example, finding techniques for maximising the CMD of particles (subject to the other constraints) was not possible; especially for the first and third study. This is evident because the CMD vector is always roughly orthogonal to the decision vector. Another parameter that was roughly orthogonal to the decision vector was life-cycle greenhouse gas emissions. This result suggests that the selection of a sustainable fuel is usually independent of the other criteria (i.e. emissions and efficiency) against which alternative engine technologies are assessed.

The GAIA plot for criteria shows also that alternative engine technologies are generally capable of delivering improvement in the physical characteristics of particles (such as reduced particle mass, number, and surface area emissions, but not increased CMD) especially for the third study. However, the improvements in the physical characteristics of particles were achieved by making the unregulated chemistry of the particles worse. This can be observed due to the roughly orthogonal nature of the particle physical and chemical characteristics in the GAIA criteria plot. For example, the organic volume percentage of particles was roughly orthogonal (95-135 ^{o}) to the decision vector in all 3 studies. Thus, for the engine designer who contemplates maximising brake thermal efficiency as a *modus operandi* for improving the DPM emissions profile from a CI engine may be able to achieve reductions in the physical components of DPM (e.g. mass, number etc); however, this is generally achieved at the expense of making the unregulated chemistry of DPM worse. Therefore, eliminating the semi-volatile fraction of DPM emerges as a critical need for further research, especially when one considers the strong correlation between the organic fraction of DPM and the presence of ROS.

A comparison dataset was also analysed to "validate" the results arising from the PROMETHEE-GAIA analysis undertaken in this study (see Figures S6-S8). The comparison dataset is based on ethanol fumigation emissions testing performed by Zhang et al. (28). Note that the PROMETHEE II outrankings, the correlation between criteria, and the location and length of the decision vector are similar to the results presented previously in this manuscript for ethanol fumigation.

In conclusion, this study has investigated common alternative engine technologies (i.e. fuels, combustion strategies, and injection technologies) that are applied in CI engines in an attempt to reduce their impact on urban air quality, human health and global warming. The results show that alternative combustion strategies (such as ethanol fumigation) are recommended at moderate load with ethanol substitutions of around 20 %. The third study shows a clear preference for high percentage soy blends 60-100 % at full load. The second study does not exhibit strong preferences; however, biodiesel and synthetic diesel are slightly preferred to ULSD; whereas, no clear preferences emerged for either injection technology. Whilst the results show that alternative engine technologies are capable of improving the nature of the CI engines emissions profile, further research is required to deliver technologies that minimise the impact of CI engines. Specifically, improving the unregulated chemistry of DPM emissions with alternative fuels has emerged as one problem requiring further research attention, along with finding alternative engine technologies that do not significantly reduce the CMD of particles for the purpose of protecting the respiratory health of those exposed to DPM.

Supporting information available

Figures S1-S8 and Tables S1-S4. This information is available free of charge via the Internet at http://pubs.acs.org.

References

(1) Heywood, J. B., *Internal combustion engine fundamentals*. McGraw-Hill, Inc: New York, 1988; pp 1-930.

(2) Majewski, W. A.; Khair, M. K., *Diesel emissions and their control*. SAE International: Warrendale, 2006; pp 1-561.

(3) Eastwood, P., *Particulate emissions from vehicles*. John Wiley & Sons, Ltd: Chichester, 2008; pp 1-493.

(4) Khalek, I. A.; Bougher, T. L.; Merritt, P. M.; Zielinska, B., Regulated and Unregulated Emissions from Highway Heavy-Duty Diesel Engines Complying with US Environmental Protection Agency 2007 Emissions Standards. *Journal Air Waste Manage.* **2011,** *61*, (4), 427-442.

(5) Hesterberg, T. W.; Long, C. M.; Sax, S. N.; Lapin, C. A.; McClellan, R. O.; Bunn, W. B.; Valberg, P. A., Particulate Matter in New Technology Diesel Exhaust (NTDE) is Quantitatively and Qualitatively Very Different from that Found in Traditional Diesel Exhaust (TDE). *Journal Air Waste Manage.* **2011,** *61*, (9), 894-913.

(6) Beer, T.; Grant, T.; Williams, D.; Watson, H., Fuel-cycle greenhouse gas emissions from alternative fuels in Australian heavy vehicles. *Atmos. Environ.* **2002,** *36*, (4), 753-763.

(7) Intergovernmental Panel on Climate Change *Climate change 2007: the physical science basis, contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, IPCC Fourth Assessment Report (AR4)*; IPCC: Cambridge, United Kingdom, 2007; pp 1-996.

(8) Winston, W. L., *Operations research: applications and algorithms, fourth edition*. Brooks/Cole - Thomson Learning: Toronto, 2004; pp 1-1418.

(9) Brans, J. P.; Vincke, P. H., A preference ranking organization method - (The PROMETHEE method for multiple criteria decision-making) *Manage. Sci.* **1985,** *31*, (6), 647-656.

(10) Guitouni, A.; Martel, J. M., Tentative guidelines to help choosing an appropriate MCDA method. *Eur. J. Oper. Res.* **1998,** *109*, (2), 501-521.

(11) Behzadian, M.; Kazemadeh, R. B.; Albadvi, A.; Aghdasi, M., PROMETHEE: a comprehensive literature review on methodologies and applications. *Eur. J. Oper. Res.* **2010,** *200*, (1), 198-215.

(12) Brans, J. P.; Mareschal, B., The PROMCALC & GAIA decision-support system for multicriteria decision aid. *Decis. Support Syst.* **1994,** *12*, (4-5), 297-310.

(13) Brans, J. P.; Vincke, P. H.; Mareschal, B., How to select and how to rank projects - the PROMETHEE method. *Eur. J. Oper. Res.* **1986,** *24*, (2), 228-238.

(14) Mareschal, B.; Brans, J. P., Geometrical representations for MCDA. *Eur. J. Oper. Res.* **1988,** *34*, (1), 69-77.

(15) Surawski, N. C.; Miljevic, B.; Ayoko, G. A.; Elbagir, S.; Stevanovic, S.; Fairfull-Smith, K. E.; Bottle, S. E.; Ristovski, Z. D., A physico-chemical characterisation of particulate emissions from a compression ignition engine: the influence of biodiesel feedstock. *Environ. Sci. Technol.* **2011,** *45*, (24), 10337-10343.

(16) Surawski, N. C.; Miljevic, B.; Ayoko, G. A.; Roberts, B. A.; Elbagir, S.; Fairfull-Smith, K. E.; Bottle, S. E.; Ristovski, Z. D., Physicochemical Characterization of Particulate Emissions from a Compression Ignition Engine Employing Two Injection Technologies and Three Fuels. *Environ. Sci. Technol.* **2011,** *45*, (13), 5498-5505.

(17) Surawski, N. C.; Miljevic, B.; Roberts, B. A.; Modini, R. L.; Situ, R.; Brown, R. J.; Bottle, S. E.; Ristovski, Z. D., Particle emissions, volatility, and toxicity from an ethanol fumigated compression ignition engine. *Environ. Sci. Technol.* **2010,** *44*, (1), 229-235.

(18) Surawski, N. C.; Ristovski, Z. D.; Brown, R. J.; Situ, R., Gaseous and particle emissions from an ethanol fumigated compression ignition engine. *Energ. Convers. Manage.* **2012,** *54*, (1), 145-151.

(19) AVL List GmbH *AVL BOOST v5.1*, Graz, Austria, 2008.

(20) Beer, T.; Grant, T.; Morgan, G.; Lapszewicz, J.; Anyon, P.; Edwards, J.; Nelson, P.; Watson, H.; Williams, D. *Comparison of transport fuels, Final Report (EV45A/2/F3C), to the Australian Greenhouse Office, on the Stage 2 study of Life-cycle Emissions Analysis of Alternative Fuels for Heavy Vehicles*; CSIRO: 2001; pp 1-463.

(21) Beer, T.; Grant, T.; Campbell, P. K. *The greenhouse and air quality emissions of biodiesel blends in Australia. CSIRO Report Number KS54C/1/F2.27*; 2007; pp 1-126.

(22) Ristovski, Z. D.; Miljevic, B.; Surawski, N. C.; Morawska, L.; Fong, K. M.; Goh, F.; Yang, I. A., Respiratory health effects of diesel particulate matter. *Respirology* **2012,** *17*, (2), 201-212.

(23) Espinasse, B.; Picolet, G.; Chouraqui, E., Negotiation support systems: a multi-criteria and multi-agent approach. *Eur. J. Oper. Res.* **1997,** *103*, (2), 389-409.

(24) Ecklund, E. E.; Bechtold, R. L.; Timbario, T. J.; McCallum, P. W., State-of-the-art report on the use of alcohols in diesel engines. *SAE Tech. Pap. Ser.* 1984, *840118*.

(25) Abu-Qudais, M.; Haddad, O.; Qudaisat, M., The effect of alcohol fumigation on diesel engine performance and emissions. *Energ. Convers. Manage.* **2000,** *41*, (4), 389-399.

(26) Biswas, S.; Verma, V.; Schauer, J. J.; Cassee, F. R.; Cho, A. K.; Sioutas, C., Oxidative Potential of Semi-Volatile and Non Volatile Particulate Matter (PM) from Heavy-Duty Vehicles Retrofitted with Emission Control Technologies. *Environ. Sci. Technol.* **2009,** *43*, (10), 3905-3912.

(27) Miljevic, B.; Heringa, M. F.; Keller, A.; Meyer, N. K.; Good, J.; Lauber, A.; Decarlo, P. F.; Fairfull-Smith, K. E.; Nussbaumer, T.; Burtscher, H.; Prevot, A. S. H.; Baltensperger, U.; Bottle, S. E.; Ristovski, Z. D., Oxidative potential of logwood and pellet burning particles assessed by a novel profluorescent nitroxide probe. *Environ. Sci. Technol.* **2010,** *44*, (17), 6601-6607.

(28) Zhang, Z. H.; Tsang, K. S.; Cheung, C. S.; Chan, T. L.; Yao, C. D., Effect of fumigation methanol and ethanol on the gaseous and particulate emissions of a direct-injection diesel engine. *Atmos. Environ.* **2011,** *45*, (11), 2001-2008.

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