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## Investigation of the Impact of Fuel Properties on Particulate Number Emission of a Modern Gasoline Direct Injection Engine

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### Abstract

Gasoline Direct Injection (GDI) has become the preferred technology for spark-ignition engines resulting in greater specific power output and lower fuel consumption, and consequently reduction in CO<sub>2</sub> emission. However, GDI engines face a substantial challenge in meeting new and future emission limits, especially the stringent particle number (PN) emissions recently introduced in Europe and China. Studies have shown that the fuel used by a vehicle has a significant impact on engine out emissions. In this study, nine fuels with varying chemical composition and physical properties were tested on a modern turbo-charged side-mounted GDI engine with design changes to reduce particulate emissions. The fuels tested included four fuels meeting US certification requirements; two fuels meeting

European certification requirements; and one fuel meeting China 6 certification requirements being proposed at the time of this work. Two risk safeguard fuels (RSG), representing the properties of worst case market fuels in Europe and China, were also included. The particle number concentration of the solid particulates was measured in the engine-out exhaust flow at steady state engine operations with load and speed sweeps, and semi-transient load steps. The test results showed a factor of 6 PN emission difference among all certification fuels tested. Combined with detailed fuel analyses, this study evaluated important factors (such as oxygenates, carbon chain length and thermo-physical properties) that cause PN emissions which were not included in PMI index. A linear regression was performed to develop a PN predictive model which showed improved fitting quality than using PMI.

### Introduction

Introduction of the GDI engines into the light-duty gasoline fleet has been an enabler to achieve the reduction targets of CO<sub>2</sub> emissions. One of the significant challenges for the new internal combustion engines has been the particulate emissions. Locally rich combustion leads to the formation of soot, resulting in high particulate emissions, especially at the most efficient engine operation points in terms of fuel consumption, i.e. low engine speeds and medium to high loads. Improved engine component designs, updated controls and optimized calibration have resulted in significant improvements in particle emissions reduction; nevertheless, the impact of the fuel and its chemical composition and physical properties must also be considered. Many researchers have been assessing the impact of the fuel properties on engine out emissions, in addition to trying to determine an appropriate correlation that would provide some predictive method of the fuels physical and chemical properties to the engine out emissions [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11].

Aikawa et al. introduced Particulate Matter Index (PMI) as a model to predict particle emissions from fuel composition (measured by detailed hydrocarbon analysis), including all

individual components of the fuel [1]. There are many other researchers who have developed methods to determine a numerical relationship for fuel properties and the propensity of the fuel to produce PN or PM [5, 7, 8]. As Chapman and coworkers have shown, all provide a correlation to PMI number with some having a more direct relationship than others, and some having a stronger relationship to vehicle out emissions [5]. Recent work by Leach and co-workers, and Wittmann and Menger show stronger correlations of the PMI to engine out PM and PN, as well as demonstrating that a model tool can be developed to help translate fuel properties to predicted engine out emissions [7]. Wittmann and Menger have compared their approach to many others in the industry and shown a stronger correlation, even to those correlations that try to use simple and single fuel properties [7]. They showed that correlation would be based on 14 properties, and developed a model around a single engine. Leach showed that a detailed analysis could be avoided by using composition classes and the bulk fuel dry vapor pressure equivalent (DVPE) in a new particle number index (PNI), essentially improving on the work of Aikawa and co-workers [8]. These papers seem to indicate that in addition to the fuel properties, some

combination of engine hardware, fuel preparation, engine calibration settings and their effect on the engine out emissions explain why the PMI number does not always show a strong correlation to the PN or particulate mass (PM) emissions.

Additionally, researchers have been looking at the impact of oxygenates on the PM/PN emissions. Leach et al. tested three different alcohols, and found different effects on PN emissions, with methanol being the highest [11]. N-butanol performed similar to the model base fuel gasoline. Overall, PN increased for each of the oxygenated fuels. Conversely, work by Sakai and Rothamer showed that the addition of ethanol caused a decrease in the engine out particulate in proportion to the ethanol content [9]. Yinhui et al. showed that a fuel with 10% ethanol produced limited improvement on particulate emission compared to reducing the aromatic and olefin content in the gasoline [6]. They also showed that the 10% ethanol increased the PN emissions at low load conditions. Mohd Murad and coworkers studied the impact of methanol and showed that the methanol does not lead to low levels of particulate number emissions, specifically across a range of fuel injection timing [10]. They also showed that the addition of heavier components to a fuel does not alter the spray structure under flash boiling conditions. Also, they demonstrated that a higher initial boiling point does not necessarily lead to higher particulate emissions, and that the light end of the gasoline range did not influence a rich mixture or a liquid film that promoted an increase in the particulates [10]. Ratcliff and coworkers examined a broad range of oxygenates and found that in certain cases oxygenates could produce higher PM than hydrocarbons with equivalent double bond equivalent and vapor pressure at 443K - the fuel parameters used in calculating PMI [12].

Various regions around the world have specific local emission regulations that require the use of specific certification fuels which must be considered in the engine design. This presents a very challenging task for Automotive OEMs, and so continued effort is devoted to determine an appropriate correlation or predictive model relating the fuels physical and chemical properties to engine out emissions. This work focuses on the investigation of the influence of chemical and physical fuel properties relating to particle number emissions on an GDI engine with design improvements to significantly reduce engine-out PN emissions to levels approaching what is required by Euro6 PN emission legislation [13]. It is necessary to reassess published particulates vs. fuel properties correlations which were generated on older engine designs. Furthermore, understanding the PN performance differences among certification and worse case fuels in different markets will allow Automotive OEMs to avoid engine hardware proliferation and reduce calibration effort.

## Experimental Setup

### Test Engine and Dyno Setup

The engine used in this study was a 2.0 L turbocharged four cylinder direct injection spark ignition engine. The side mounted fuel injector and the centrally located spark plug were in a longitudinal arrangement. The Bosch 6-hole

**TABLE 1** Test engine specification

Engine type		4-stroke, 4-valve
Displacement	[cm <sup>3</sup> ]	1998
Bore x stroke	[mm]	86 x 86
Compression ratio	[-]	9.5:1
Aspiration		Turbo-charged
Fuel delivery		DI, side mounted
Injector		Bosch 6-hole
Maximum rail pressure	[bar]	350

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HDEV5.2 injectors were mounted between the two intake valves in each of the four cylinders, with a spray targeting that was spreading diagonally downwards into the combustion chamber. The engine specifications are detailed in [Table 1](#).

The engine design improvements to reduce PN emissions included updated combustion chamber and piston designs that enhanced air/fuel mixing and reduced fuel rich pockets. The engine piston cooling was optimized to avoid unnecessary cold piston surfaces that contributed to increased fuel film on the piston top. The injector spray target and injector seat design went through multiple iterations, and achieved low PN emissions and reduced soot deposits both inside and outside the spray holes on the injector tip.

The test engine was setup on an engine dynamometer with well controlled coolant, oil, fuel, ambient air and intercooler outlet temperatures. In the majority of the engine testing, coolant and oil temperatures were set at 90 (±3) °C, simulating hot engine operations. The ambient air temperature was set at 23(±1) °C, the intercooler outlet temperature at 31(±2) °C, and the fuel temperature at 21(±1) °C, all values simulating the typical engine operation in vehicles. The engine oil used for this test was a 5W-30 available in the market. The in-cylinder pressure was measured using piezoelectric pressure transducers from AVL, through a series of machined ports in the cylinder head. The automated test procedure for the engine dynamometer was controlled with iTest and INCA was used for the engine control unit command, both communicating with IndiCom data acquisition and evaluation software. Engine parameters were resolved to crank angle and the emission data were acquired at 1Hz frequency.

## Fuels and Fuel Analysis

A total of nine fuels were tested in this study. The data for the fuels is found in the Appendix. The fuel test matrix includes the certification fuels for Europe, China, and the United States and additionally two Risk Safeguard fuels (RSG) which are representing the worst-case fuels for Europe and China, namely RSG E10 and RSG M15. The RSG fuel blends were defined as the most aggressive fuels to simulate the worst-case situations that engines could face. Certification fuels are used to check the compliance of the region-specific emission standards by the respective emission regulation authorities. The North American fuels in this test are represented by CARB LEV II, CARB LEV III, EPA Tier 2, and the EPA Tier 3 premium blends. A special China 6 Premium fuel blend, based on the draft of the China 6 Premium fuel specifications, was also formulated by GM for this study. The European certification fuels, Euro 5 and Euro

6, complete the test fuel matrix. These fuels covered a wide range of fuel properties that are important to PN emissions.

All the certification fuels and the RSG E10 blend were sourced from Gage Products Company. RSG M15 fuel was blended by Haltermann Carless. All test fuels have been subjected to Detailed Hydrocarbon Analysis (DHA), by a modified ASTM D6730 method, measuring individual components in the fuels. The oxygenated hydrocarbon content was analyzed using the procedure described in reference [14] and ASTM D4815, and the distillation curves for the fuels were measured with the ASTM D86 method at the GM Fuels laboratory. The PMI number was calculated using the standard calculator as shared from Honda, Aikawa and co-workers. In this study, the oxygenates for the DHA were corrected using the data produced by the ASTM D4815 method. This provides a more representative PMI number, by adjusting all the components in the DHA and thus the PMI final number.

Additionally, the heat of vaporization ( $\Delta H_{\text{vap}}$ ) of the fuels were measured at the National Renewable Energy Laboratory (NREL), using the methods described by Chupka et al. [15]. Briefly,  $\Delta H_{\text{vap}}$  was measured using a TA Instruments (Newark, DE) simultaneous DSC/TGA (SDT) Q600 instrument. The instrument was calibrated per manufacturer's specifications. The cell constant was further calibrated using deionized water. Three replicate analyses of water were run and a ratio of the measured value to that reported in the literature was calculated to generate a calibration factor. All  $\Delta H_{\text{vap}}$  measurements were then multiplied by this factor. Samples were introduced via a gas tight syringe into a 90  $\mu\text{L}$  platinum pan which was equipped with an aluminum lid with a laser drilled pinhole, 75  $\mu\text{m}$  in diameter. Samples were held at ambient lab temperature for the duration of the test and a nitrogen purge flow of 50 mL/min was used to aid in sample evaporation. In some cases where the test run time was over 3 hours, the pinhole lid was not used to avoid baseline drift (these samples are noted). To calculate the  $\Delta H_{\text{vap}}$ , the heat flow signal was corrected for the heat flow associated with the empty pans by subtracting this value from the total measured heat flow. The area under the curve was calculated and divided by the sample mass to obtain the  $\Delta H_{\text{vap}}$  of the sample. The samples were run in duplicate in a randomized order for DSC/TGA analysis.

## Particle Counter

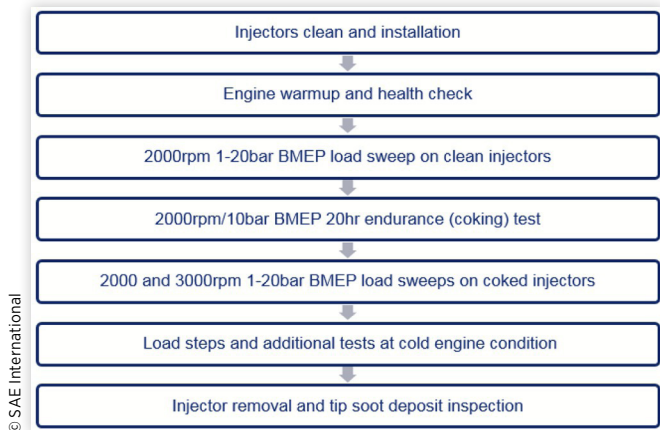
Particulate number emissions were measured in the engine-out exhaust flow using a Horiba MEXA-2100SPCS. The measurement method is based on the requirements described in Revision 4 of the UN/ECE Regulation No.83, which is a uniform provision concerning the approval of new vehicles regarding the emission of pollutants. To achieve sufficient repeatability of the measurement, this regulation enforces the count of only solid particles in the engine exhaust which are larger than 23 nm. The exclusion of volatile particles stems from the fact that the dilution conditions significantly affect the formation of Nucleation Mode particles due to condensation of volatile substances such as sulfates and soluble organic fractions. This is being achieved by having heated sample lines (to 150°C) and Volatile Particle Remover (VPR) at 300-400°C after the first dilution stage. The total dilution ratio of 2500 was set during the tests in two stages.

## Test Procedure and PN Measurement

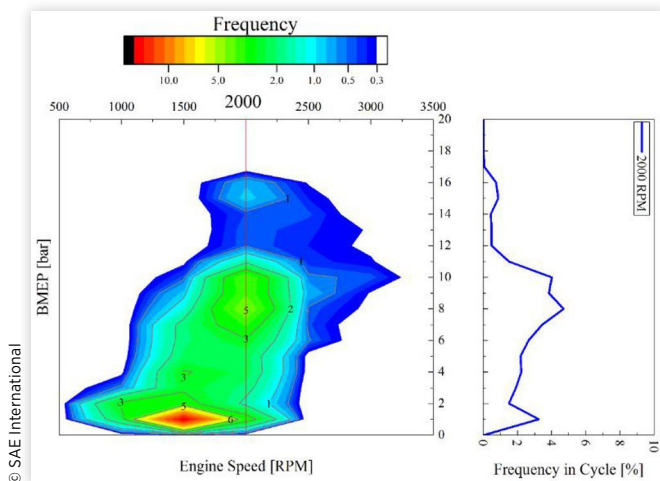
All measurements are performed at the engine test laboratory at the Robert Bosch LLC facility in Farmington Hills, Michigan. An automated test procedure, shown in [Figure 1](#), was implemented to assess the PN emission of the fuels. Tier 3 premium fuel was chosen as the baseline fuel for this study and the engine calibration was performed to optimize the fuel injection timing, targeting to minimize the PN emission and specific fuel consumption. Single homogenous injection strategy was used for the fuel injection timing calibration to reduce the complexity of the test matrix. The same engine calibration settings were used for all test fuels.

To address the concern whether the PN emission performance at a single steady state point of 2000rpm/10bar BMEP would sufficiently represent real world engine operations, the visitation frequency of the engine speed and load during a typical drive cycle was calculated and shown in [Figure 2](#). The distribution of the engine load at 2000 RPM is shown in the right panel. It is clear that the engine spent a substantial amount of time at 2000rpm and 8-10 bar BMEP.

**FIGURE 1** Automated Test Procedure.



**FIGURE 2** Engine load and speed visitation points during a drive cycle.

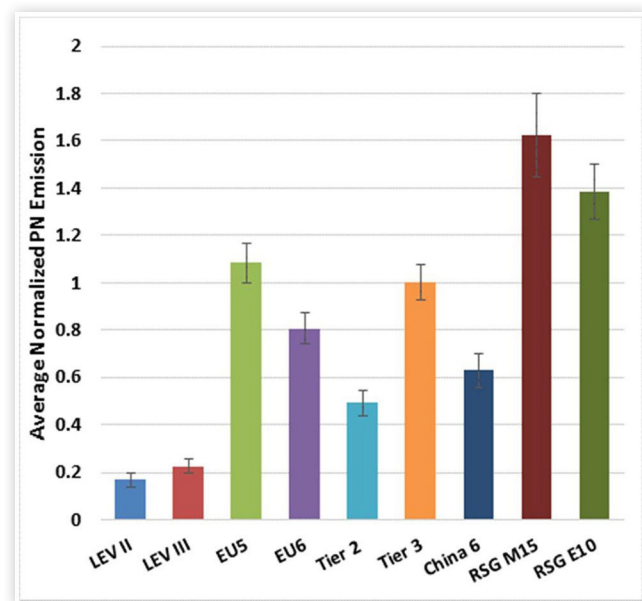


It is well known that PN measurements have a high uncertainty and it is not easy to ensure adequate PN measurement repeatability and accuracy so that the impact of fuel properties can be determined with sufficient confidence. A few measures were taken in this study to reduce the impact of engine hardware performance shift and improve PN measurement repeatability and accuracy:

1. Before testing each fuel, the injectors were cleaned in an ultrasonic bath for an hour to remove all the soot deposits from the injectors and reset the engine PN performance to that of clean injectors.
2. A 20 hour endurance test was performed for each fuel to ensure that the level of soot deposit on the injectors and other parts of the engine and measurement equipment has achieved equilibrium. The PN emissions were measured every 5 minutes during this test.
3. The average of the last 4 hours of the endurance test data at 2000rpm and 10bar BMEP were used to assess the impact of the fuels. This not only ensured that the engine had sufficient time to reach desired operating conditions and stable PN performance, but also allowed a large data ensemble of PN measurements. The average values and the standard deviations are shown in [Figure 3](#).
4. The test order for different fuels were randomized and each fuel was tested more than once non-consecutively. The Tier-3 premium were tested periodically throughout the entire study and repeatable results were observed.

For the different test fuels, the same valve timings and injection strategies were used that were optimized for Tier-3 premium fuel. The engine was operated at the stoichiometric condition by injecting the same energy content into the

**FIGURE 3** PN emission average values, normalized to emissions of Tier 3 certification fuel, during the last 4 hours of endurance tests



combustion chamber. The closed loop lambda control of the ECU controlled the stoichiometry during the test based on the oxygen concentration in the exhaust. Due to differences in the carbon, hydrogen and oxygen composition of the fuels, the lower heating values are different among the studied fuels. The ignition timing was adjusted to achieve the maximum brake torque (MBT), such that the CA50, where 50% of the cumulative heat release has been converted, is approximately 8 °CA aTDC with the baseline fuel, Tier-3 premium and kept constant for the other tested fuels. A summary of the engine parameters for the three types of testing is shown in [Table 2](#).

## Results and Discussions

Although the main focus of this study was to assess the impact of certification fuels and worst case fuels in different markets on PN emissions, it is desirable to establish a correlation between fuel properties and PN emissions that provides a tool to project the PN performance of a certain hardware for different markets.

As shown in [Figure 3](#), among the certification fuels, LEV II and LEV III produced the lowest PN results and were not significantly different from one another. As expected, the highest PN emissions were from the RSG fuels with RSG M15 a factor of roughly 2.5 higher compared to China 6 Premium. The difference in the heat of combustion between the fuels, based on various levels of oxygenates, as well as the fuel densities led to different injection durations. This difference resulted in 10% longer duration of injection for RSG M15 compared to Tier 2 premium fuel. From injector tip inspection after the test and measured injector energizing time during the 20hr test, none of the fuels had produced major soot deposit inside the spray holes that caused fuel flow shifts. The maximum injector energizing time shift of the worst PN performing fuel, the RSG M15, is around 1%.

## Comparison of the PM Index

The PM Index, developed by Aikawa et al. [1], is the most established particulate matter and number predictor for gasoline fuels. Regardless of engine type or test cycle, the PMI predicts the relative tendency of a specific gasoline blend to produce PM. Detailed compositional information about the fuel with the volatility and structural characteristics of its constituents

**TABLE 2** Test parameters

		Load sweep	20 hour test	Transient load step
Lambda	[-]	1	1	1
RPM	rpm	2000, 3000	2000	1500
BMEP	[bar]	1-20	10	1 and 8
SOI	[°CA bTDC]	290 1 bar: 300	290	260-300
CA50	[°CA aTDC]	8	8	8
Coolant temperature	[°C]	90	90	90

are combined in this method. As seen in the formula below, PMI is calculated based on the double bond equivalent (DBE), the vapor pressure at 443 Kelvin (V.P. (443K)) and the weight fraction ( $W_i$ ) of every single hydrocarbon and oxygenate component of the fuel. In addition, Aikawa et al. also concluded that the PMI can be correlated to either mass or number based particle emission.

$$PM\ Index = \sum_{i=1}^n \left[ \frac{DBE_i + 1}{V.P(443K)_i} \times W_{t_i} \right] \quad (1)$$

Detailed hydrocarbon analysis is required to get the necessary data for the PMI calculation. The fuel analysis and PMI calculation was performed by the GM Fuel Lab and at NREL and the results are shown in Table 3. The calculated PMI for the LEV certification fuels are the lowest in this comparison. The calculated PMI for RSG E10 is significantly higher than the rest of the tested fuels, which is approximately twice as high as the calculated index for Euro 6 fuel. The PMI for the worst-case scenario fuel for China, RSG M15, is 13 to 16% higher than that of China 6 Premium certification fuel. Because of differences in the implementation of the DHA analysis, including differences in the approach to calculation of the vapor pressure at 443K, the NREL PMI values are slightly different, but exhibit identical trends.

Figure 4 shows the PN emissions as a function of PM index. The coefficient of determination ( $R^2$ ) for the correlation is low, indicating that PMI is not adequate to predict the variation in PN emissions for these fuels. This may not be surprising in that all but one of the fuels contain oxygenates, which are not explicitly accounted for in the calculation of PMI. There may also be other factors that are not captured by PMI such as density, viscosity, and surface tension and their effect on fuels spray penetration, spray angle and breakup, as well as  $\Delta H_{vap}$ . Removal of the two highest PM emitting fuels (the RSG fuels) improves the  $R^2$  to 0.74 supporting the idea that PMI is not capturing all the important fuel related factors for this group of fuels. In particular, the high PN emissions for RSG M15 relative to other fuels with similar PMI values (such as Tier 2 and Tier 3) is of interest. To aid in subsequent discussion of these results, additional fuel properties are presented in Table 4.

TABLE 3 PM Index of the tested fuels

Fuel type	PM Index		Oxygenate Type	Oxygenate % D4815
	GM Fuel Lab	PM Index NREL		
LEV II	1.12	1.24	MTBE	10.88
LEV III Premium	1.17	1.30	Ethanol	10
Tier 2	1.66	1.66	None	0
Tier 3 Premium	1.68	1.76	Ethanol	9.71
Euro 5	1.89	1.75	Ethanol	5.07
Euro 6	1.28	1.64	Ethanol	9.87
China 6 Premium	1.33	1.49	MTBE	8.17
RSG E10	2.77	3.07	Ethanol	9.87
RSG M15	1.5	1.73	Methanol	15.3
			MTBE	7.1

FIGURE 4 Normalized PN emissions at 2000rpm and 10bar BMEP as a function of average PMI

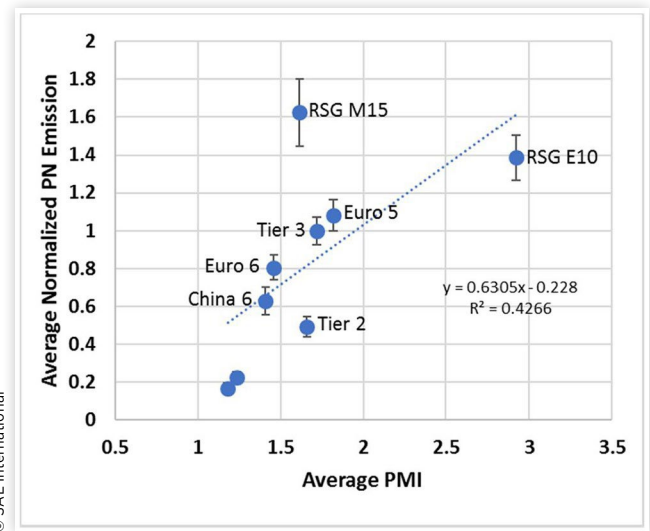


TABLE 4 Heat of vaporization and lower heating value measurement results

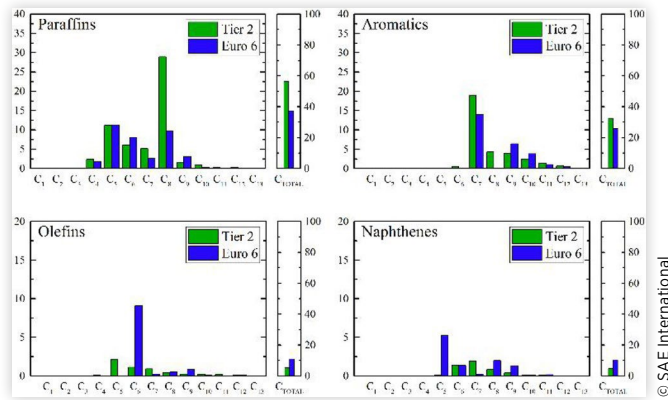
Fuel type	Lower Heating Value [MJ/kg] <sup>a</sup>	HOV DSC/TGA [kJ/kg]	Density at 15°C
LEV II	42.54	381	0.743
LEV III Premium	41.78	425	0.749
Tier 2	43.18	372	0.740
Tier 3 Premium	41.69	423	0.749
Euro 5	41.91	421	0.753
Euro 6	41.73	461	0.752
China 6 Premium	42.51	385	0.759
RSG E10	-	426	0.740
RSG M15	-	513	0.739

<sup>a</sup> ASTM D240 from Supplier Certificate of Analysis

## Tier 2 vs Euro 6

In order to dissect the discrepancy in the PN prediction of the PMI, the parameters affecting the PN levels for Tier 2 and Euro 6 fuels are investigated in more details by examining the factors that make up PMI, as well as those that are not directly included in the PMI calculation. The Tier 2 and Euro 6 fuels had fairly similar PMI values (1.66 and 1.28), but PN emissions are almost twice as high for Euro 6. The chemical group composition of the two fuels with respect to the carbon number are shown in Figure 5. The peak of the overall hydrocarbon composition of Tier 2 is at the carbon number of 8, whereas Euro 6 exhibit a flat composition with the maximum at C<sub>6</sub>. The overall aromatic content of Euro 6 is less than Tier 2, contributing to the higher PMI of the later fuel. However, the level of aromatic hydrocarbons with more than 8 carbon atoms (C<sub>9</sub>/C<sub>9+</sub> aromatics) is higher in Euro 6 which contains 11.7 % C<sub>9</sub>/C<sub>9+</sub> aromatics compared to only 8.51 % in Tier 2.

**FIGURE 5** Volume fraction distribution of chemical structures as a function of carbon number for of Euro 6 and Tier 2 fuels.

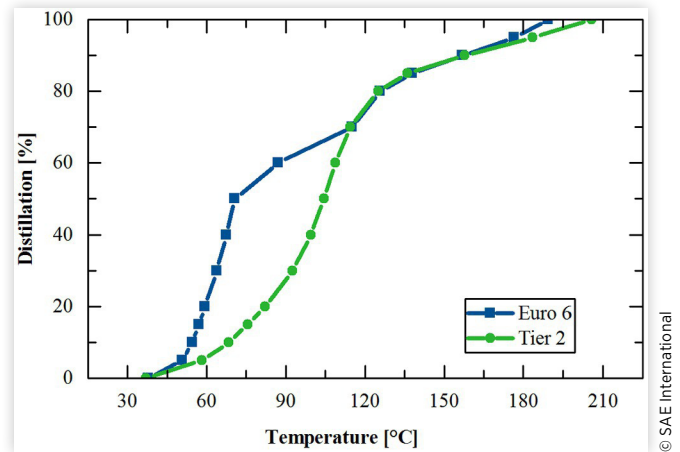


Another reason for the lower PMI of Euro 6 is due to the ethanol content. In PMI calculation, each component is linearly weighted in the blend, which neglects the synergist or antagonistic behavior of individual components in the fuel blend. Low boiling and thus high vapor pressure components in the fuel are under predicted for near-azeotropic ethanol-gasoline blend. Although several studies have confirmed a reduction of PM emissions using ethanol instead of gasoline, there are investigations showing different results [16, 17, 18, 19]; Vuk et al. presented a considerable reduction in PM emissions with E10 (10% ethanol in gasoline) compared with the E0 fuel but increasing with E30 and E50 [20].

Considering the physical effects, ethanol has a higher density and viscosity. This results in poor spray atomization and deeper spray penetration [18]. The lower heat of combustion of ethanol further influences the spray penetration of Euro 6, due to a higher amount of fuel that is required at equal engine operation points. The injector energizing times (as a proxy for injection duration) of Euro 6 was around 2 % longer than Tier 2 which could result in deeper spray penetration and thereby more piston wetting at the same SOI. This change of the spray is not included in the PMI although it can significantly affect PN emissions. Another reason for the opposite trend in the PMI and the PN can be the  $\Delta H_{vap}$  of the fuels, seen in Table 4 Heat of Vaporization measurement results. The  $\Delta H_{vap}$  of Euro 6 is around 24 % higher than that of Tier 2. In order to vaporize, the fuel needs a certain amount of thermal energy to transform from the liquid into the gaseous phase. This means that Euro 6 needs more thermal energy to fully vaporize and thereby causes colder conditions in the combustion chamber. Due to these colder conditions, the evaporation of the heavy end of the fuel would be impeded. [4, 21].

Due to the correlation with the vapor pressure and linear weighting, PMI doesn't capture the entire evaporation behavior - especially the low distillate end. The distillation curves of Tier 2 and Euro 6 fuels are presented in Figure 6. The figure is intentionally plotted as a function of temperature, since during the engine operation the fuel temperature would be directly affected by the coolant temperature. The evaporation of the last 30 % of the fuels is similar but the final boiling point of Tier 2 is higher. The higher the boiling point and the larger the percentage of fuel concentrated in its heavy end,

**FIGURE 6** The effect of ethanol on the distillation curve for Euro 6 and Tier 2 fuels.



the higher is the probability for the presence of liquid fuel due to non-timely evaporation. Due to the large proportion of ethanol in Euro 6, the initial vaporization vary significantly. At 90 °C, equivalent to the engine operation temperature, almost 60 % of Euro 6 is evaporated. Flash boiling might occur under these conditions. Heat transfer in the cylinder head and pressure increase in high-pressure pumps increase the temperature of the liquid fuel before the injection. In homogeneous injection mode, the injection takes place during the intake stroke and therefore the fuel is injected into approximately the manifold pressure which can be lower than the saturation pressure, especially under throttled conditions. High volatility components of the fuel become superheated under these conditions and flash vaporization would occur. This effect is known to greatly influence the atomization and vaporization process [22, 23]. However, flash boiling can also adversely affect the fuel vaporization. During the injection process, the temperature of the fuel assimilate to the intake air temperature and the saturation pressure rises above the ambient pressure. Flash boiling components cause spray plume collapse and therefore lead to a deeper spray penetration and thereby to wetting of the piston and the cylinder walls [24, 25].

Based on the discussion above, the higher PN emissions observed for Euro 6 relative to Tier 2, in spite of their similar PMI values, are caused by several factors that are not included in the PMI model. These likely include the combined effects of reduced lower heating value in the Euro 6 requiring longer injection duration, increased  $\Delta H_{vap}$  causing reduced temperature during evaporation and thus hindering evaporation of heavy aromatics, and the potential for flash boiling to occur for the ethanol blend.

## Tier 2 vs RSG M15

Tier 2 and RSG M15 are predicted to emit similar PN emissions. However, based on the PN results in Figure 4, there is a big difference between these. Many of the same considerations discussed for the Tier 2 - Euro 6 comparison apply here as well. While energy density of RSG M15 was not measured, given this fuels high oxygenate content its lower heating value will be significantly lower than that of Tier 2, requiring longer injection

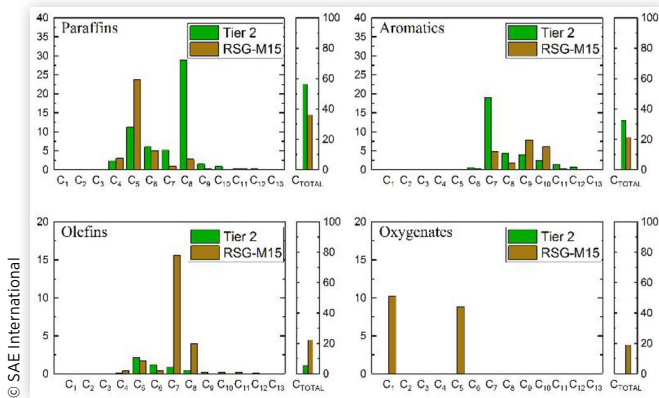
duration at the same BMEP. RSG M15 also has the highest  $\Delta H_{vap}$  of the fuels studied. Figure 7 shows the carbon histogram of Tier 2 and RSG M15. M15 fuel blend contains higher content of larger aromatics but also 15 % methanol and 7% MTBE, mirroring the higher heavy aromatics content and high alcohol content of Euro 6. The high heavy aromatics combined with the high  $\Delta H_{vap}$  may be a cause of the much higher PN of RSG M15.

Regarding the distillation behavior, similar evaporation behavior can be found between RSG M15 and Euro 6. The distillation curves of Tier 2 and RSG M15 are shown in Figure 8. The study by Qin et al. showed a significant reduction of PN emissions with the addition of methanol [13]. The low boiling oxygenate components of the fuels cause a high percentage of evaporated fuel at low temperatures. Due to the lower boiling point of methanol versus ethanol, approximately 65 % of the RSG M15 is evaporated at 90 °C.

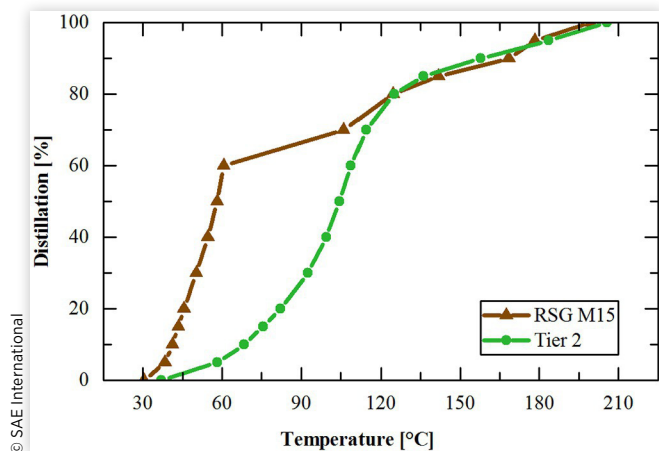
### RSG M15 vs RSG E10

The two risk safeguard fuels RSG M15 and E10 show the largest discrepancies in the PMI and PN measurement results. While PM emissions are similar, the E10 has a PMI that is more than 1.2 PMI units higher. Figure 9 exhibits the results

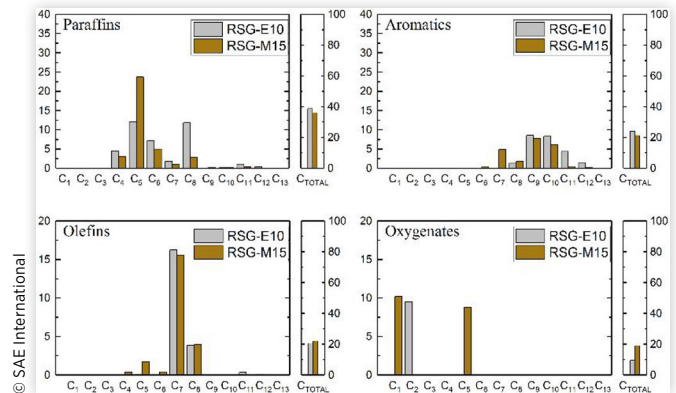
**FIGURE 7** Volume fraction distribution of chemical structures as a function of carbon number for of RGS M15 and Tier 2 fuels.



**FIGURE 8** Distillation curves of Tier 2 and RSG M15.



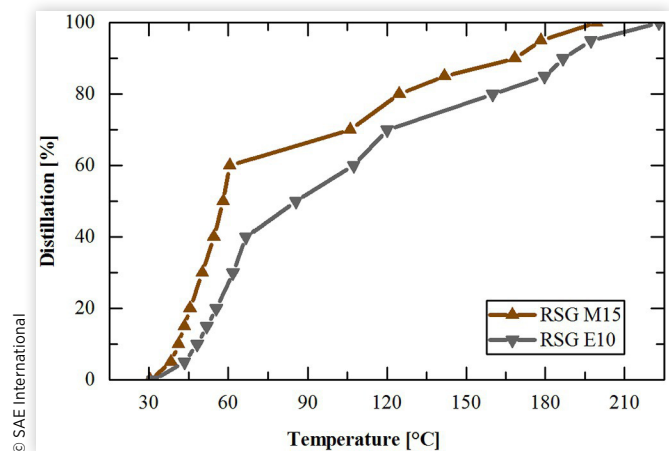
**FIGURE 9** Volume fraction distribution of chemical structures as a function of carbon number for RSG E10 and RSG M15 fuels.



of the hydrocarbon analysis of RSG M15 and E10. The overall hydrocarbon composition seems to be identical at first glance, but particularly the distribution of the higher molecular weight hydrocarbons diverges. RSG E10 includes approximately 23%  $C_9/C_{9+}$  aromatics whereas RSG M15 only consist of 14%. Within the tested fuels, the amount of  $C_9/C_{9+}$  aromatics is highest with E10. When comparing the  $C_{11}/C_{11+}$  aromatics, an even bigger difference is shown. The percentage of  $C_{11}/C_{11+}$  aromatics of the E10 fuel is approximately 6 % whereas the percentage of M15 is only 0.5 %. Particularly the high amount of heavy aromatic hydrocarbons leads to the highest PMI for RSG E10. RSG M15 has the highest  $\Delta H_{vap}$  which is significantly higher than the value for RSG E10. This may contribute to the similar level of PN observed for these fuels, in spite of the higher heavy aromatic content of RSG E10.

The distillation curves of RSG M15 and RSG E10 are shown in Figure 10. Except the bigger difference between 50 and 70 % distillation, which can be attributed to the higher percentage of low boiling oxygenate components of M15, the two fuels similarly evaporate with a small offset. Due to the bigger proportion of high boiling point components of E10, the respective percentages vaporize at higher temperatures over the whole range. Based on the distillation curves, a clear statement on whether the higher final boiling point of E10 or

**FIGURE 10** Distillation curves for RSG M15 and RSG E10



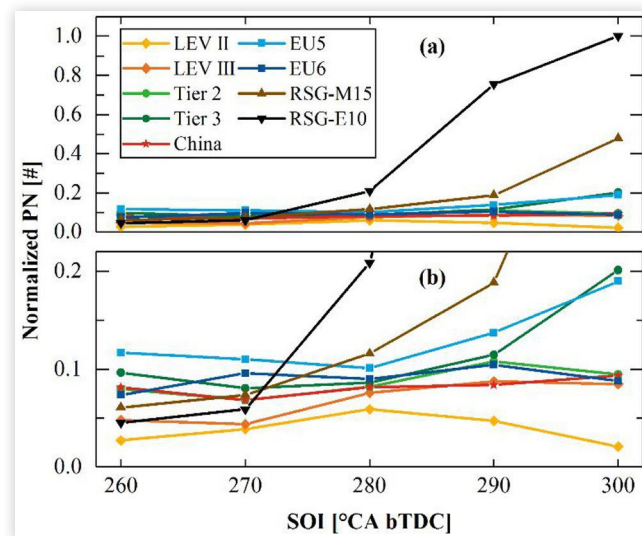
the higher probability of flash boiling of M15 has a stronger impact on PN emissions cannot be given.

The differences in the fuels in terms of vaporization and flash boiling would reflect significantly in terms of sensitivity to the injection timing. More advanced injection timing leads to higher chances of spray impingement with the piston and consecutively diffusion flames with high PN emission. A sweep of injection timing was performed as the part of the semi-transient portion of the automated test at 1500 RPM and 8 bar BMEP. Normalized PN emissions for each fuel as a function of injection timing is plotted in Figure 11. The values for all fuels are normalized to the highest emission of the RSG E10 fuel at the most advanced injection timing and shown in the top panel (a). The bottom panel (b) is scaled to highlight the certification fuels more clearly. Most of the certification fuels did not show significant sensitivity to the start of injection for the studied range (from 300 °CA bTDC to 260 °CA bTDC), with Euro 5 fuel exhibiting the largest reduction which was in the order of 2.

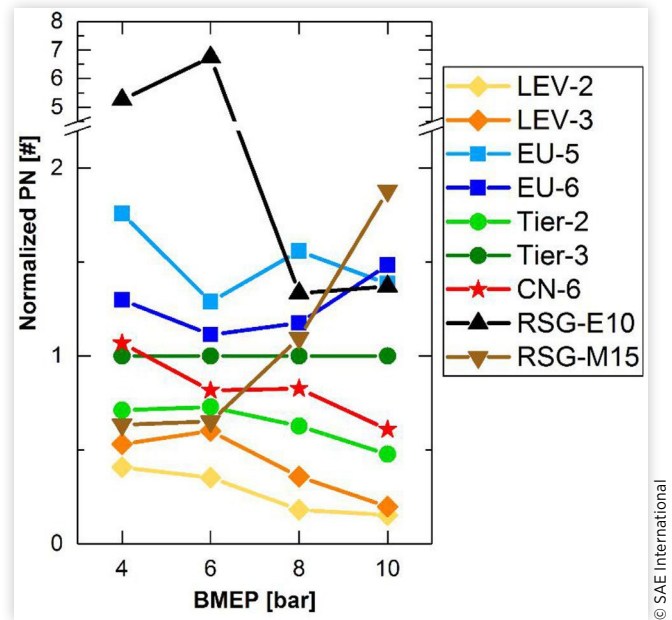
RSG E10 exhibits the highest sensitivity to the start of injection, which can be attributed to the highest final boiling point, high heavy aromatics content, and high heat of vaporization. The results indicate that with a retarded start of injection and avoiding the piston impingement, the PN emission could be reduced by an order of magnitude. This reduction comes at the cost of charge inhomogeneity and increases the specific fuel consumption by 1 %.

As highlighted in Figure 2, the engine resides at mid-load points for a significant time at 2000 RPM; therefore engine load sweep was performed to investigate the sensitivity of the fuels to different engine load operations. The result of the PN emissions for the load sweep between 4-10 bar are shown in Figure 12. The results are normalized to the PN emission of the Tier-3 premium fuel as the baseline. All of the certification fuels exhibit similar sensitivity to the engine load at this speed and the PN emission ranking of them doesn't change. The injection timing during these loads were kept constant at 290

**FIGURE 11** Sensitivity of PN emission to the start of injection timing. The PN values are normalized to RSG E10 at 300 °CA bTDC. Panel b is scaled for clarity.



**FIGURE 12** Normalized PN emission at mid-load conditions at 2000 RPM. The data is normalized to the PN emission of Tier-3 fuel.



°bTDC and as shown in Figure 11, the certification fuels didn't exhibit a significant sensitivity to the start of injection timing. The RSG-M15 and RSG-E10 fuels don't follow the same trend. This range of load sweep represents the transition between the throttled operations to the boosted conditions which would affect the flash boiling characteristic of the fuels. Significant calibration effort would be required to develop a robust SOI for these fuels at different loads.

## Regressions Analysis

Besides the investigation of the impact of chemical and physical fuel properties on particulate number emissions in gasoline direct injection engines, the development of a PN predictive model was the main goal of this work. Aikawa et al. introduced the PMI and it became the established PN and PM predictive model [1]. However, complex calculation and insufficient PN and PM prediction for oxygenated fuels, which have been confirmed in this study, leave potential for improvement.

The measured PN was set as the dependent variable and the chemical components and the physical properties as the independent ones. Due to the small number of dependent variables and a huge selection of the independent ones, it was impossible to calculate a general model to predict the PN emissions. The regression analysis provided a wide variety of possible models which perfectly correlate to the PN. In order to counteract, the selection of the independent variables was based on previous research and results of this study and thereby reduced to a small number, which resulted in the coefficient of determination of 0.92. The values of the parameters of the final model are rounded for clarity. The loss in the coefficient of determination due to rounding of the parameters is minor and does not significantly affect the final result. The final model and the selected variables are shown below and



explained subsequently. (PNR is referring to the PN Regression Equation)

$$\text{PNR} = 0.03 * A - 0.055 * O + 0.001 * V + 0.004 * B + E * [0.01 + 0.001 * O - 0.00005 * B] \quad (2)$$

A = Aromatics C9/C9+ [Vol. %]

O = Total percentage of oxygenates [Vol. %]

V = Heat of vaporization [kJ/kg]

B = Final boiling point [°C]

E = Vol. % of fuel evaporated @ 90 °C [%]

**C9/C9+ Aromatic Hydrocarbons** The most significant correlation to the PN was found with the C<sub>9</sub> and C<sub>9+</sub> aromatic hydrocarbons. In the fundamental research by Kobayashi et al., PM from gasoline and hydrocarbons of various chemical structures were measured and it was found that the aromatic flame showed the highest concentration [4]. The reason for this is that the aromatic compounds are harder to evaporate and relatively slower to decompose than other hydrocarbons. Furthermore, in the case of decomposition, aromatics may disintegrate into unsaturated alkyl compounds such as acetylenes which serve again as precursors for the formation of a benzene ring. These results were also shown in several studies with GDI engines [20, 26]. Detailed investigations of the effects of the aromatic contribution on PM resulted in a higher propensity to produce PM with higher aromatic species, although the tested fuels consist of a wide range between 0 and 20 % of C<sub>7</sub> aromatics, the low molecular weight aromatics showed no effect on PN, which could indicate that the heavy aromatics do not disintegrate into smaller ones but directly take part in the PAH and thereby the soot formation mechanisms. Furthermore, it was found that only the percentage of the high molecular weight aromatics markedly affect the PN emissions.

**Total Percentage of Oxygenated Hydrocarbons** A majority of studies have supported the significant effects of oxygenate hydrocarbons on PN emissions. Gasoline fuel with oxygenated compounds tend to produce less PN emissions, but negative effects on PN were also shown with higher percentage of oxygenates [20]. The highest percentage of oxygenates of the tested fuel is RSG M15 with 19 Vol. %. This high percentage could be an explanation for the worse PN emissions of RSG M15. Although oxygenates are included in the PMI, they merely play a minor role in the final value. Due to the lack of double bonds and the high vapor pressure compared to other components, the oxygenates are roughly counted as paraffins and the potential of positive effects on PN is not included. That is why the oxygen containing hydrocarbons are selected as a variable for the regression analysis.

**Final Boiling Point** The physical properties with the significant effects on the PN emissions are figured out to be the final boiling point, the percentage of fuel evaporated at 90 °C, and the heat of vaporization. In the PMI calculation, the boiling point is indirectly accounted for each component of the fuel through a logarithmic correlation with the vapor pressure. However, the evaporation behavior of the fuel blend is different than the evaporation of each single component added. As can be seen with Tier 2 compared to Euro 6 and also RSG M15, the

distillation of the fuel blend behaves in a different way. Due to the respective higher long-chain aromatic content of Euro 6 and RSG M15, an assumption could be made that this leads to a higher boiling point. However, in both cases the final boiling point of Tier 2 is higher and the effects can be explained due to the near-azeotropic behavior of the fuel mixture.

**Vol. % of Fuel Evaporated @ 90 °C** The value for the percentage of fuel evaporated at 90 °C is a characteristic value, which can be found from the distillation curve of the fuel. This value is suitable to quantify the effects of flash boiling and therefore was set as an independent variable. The disparity between the C9/C9+ aromatics and the PN of RSG M15 and E10 can be explained due to flash boiling.

**Heat of Vaporization** A certain amount of thermal energy is required to vaporize the fuel, which is defined as the heat of vaporization or enthalpy of vaporization. In a direct injection engine, the required thermal energy is extracted from the charge air. The effect of charge cooling enables achieving higher efficiency through increased compression ratio by mitigating knock. Thereby, the CA50 be adjusted closer to the optimum of 8 °CA aTDC. However, the cooling effect would cause undesirable conditions regarding the formation of soot in the cases of spray impingement with the piston surface or the cylinder liner. The higher the heat of vaporization, the worse the evaporation of the fuel which leads to a higher probability of diffusion flames on the engine surfaces.

## Conclusions

The effects of chemical and physical properties of gasoline fuel on particulate number emissions have been studied in a modern GDI engine. The certification fuels of Europe, China, the US, and additionally two Risk Safe Guard fuels were used for this investigation. There are a few conclusions that can be drawn from this study:

- A factor of 6 in PN difference was observed during the steady state operation point at 2000 RPM and 10 Bar BMEP among all certification fuels tested. The same trend was observed at different engine operating points. The risk safeguard fuels exhibited the highest PN emission and highest sensitivity to the injection timing calibration.
- The long-chain aromatic hydrocarbons, more precisely the C<sub>9</sub>/C<sub>9+</sub> aromatics, showed the biggest impact on particulate number emissions. This finding is consistent with previously published investigations and the C<sub>9</sub>/C<sub>9+</sub> aromatics. Despite the emphasis in the PM Index calculation, the contribution of the smaller-chain aromatics on PN emission were insignificant at the tested condition.
- The oxygenated compounds blended in the fuels have non-linear effects. The fuel bound oxygen lowers the sooting tendency of the fuel but the effect of ethanol addition in terms of affecting the spray development and vaporization of the fuel can result in higher PN emission.
- Based on the regression analysis performed on the various physical parameters of the fuel blends, final

boiling point, percentage of fuel evaporated at 90 °C, and heat of vaporization indicated to be the best suitable variables to characterize the evaporation behavior. Higher final boiling point and higher heat of vaporization values result in worse evaporation and therefore lead to higher particulate emissions due to diffusion flames of liquid film on the engine surfaces.

Based on the findings of this study, a PN predictive model was successfully developed. Including the aromatic and oxygenated hydrocarbons as two fuel components and the three physical properties, final boiling point, the percentage of fuel evaporated at 90 °C, and the heat of vaporization. It is nevertheless necessary to validate the PNR with different engines and fuels to further improve this model.

In summary, it is clear that the gasoline fuel chemical and physical properties impact the PN emissions from an engine. The variation in certification fuels and market fuels (which the certification fuels in some cases represent) present a significant challenge for OEMs to meet future emissions regulations. The wide range of PMI number with those fuels and the impacts of the oxygenates present challenges not understood before. Since the certification fuels are representations of market fuels, an improvement in market fuel quality variation can substantially reduce the risk of vehicle in-use PN emission compliance.

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## Definitions/Abbreviations

**DVPE** - Dry Vapor Pressure Equivalent  
**ECU** - Engine Control Unit  
**GDI** - Gasoline Direct Injection  
**HDEV** - Name of a Bosch type of Fuel Injector  
**HOV** - Heat of Vaporization  
**PM** - Particulate Matter  
**PMI** - Particulate Matter Index  
**PN** - Particulate Number  
**PNR** - PN Regression Equation  
**RSG** - Risk Safeguard Fuel  
**RSG E10** - Risk Safeguard Fuel 10% Ethanol  
**RSG E15** - Risk Safeguard Fuel 15% Methanol  
**VP** - Vapor Pressure

## Appendix: Fuel Properties

**TABLE A1** Fuel Properties: Heat of Vaporization and Detailed Hydrocarbon Analysis with Carbon Number by Aromatics shown with PMI number

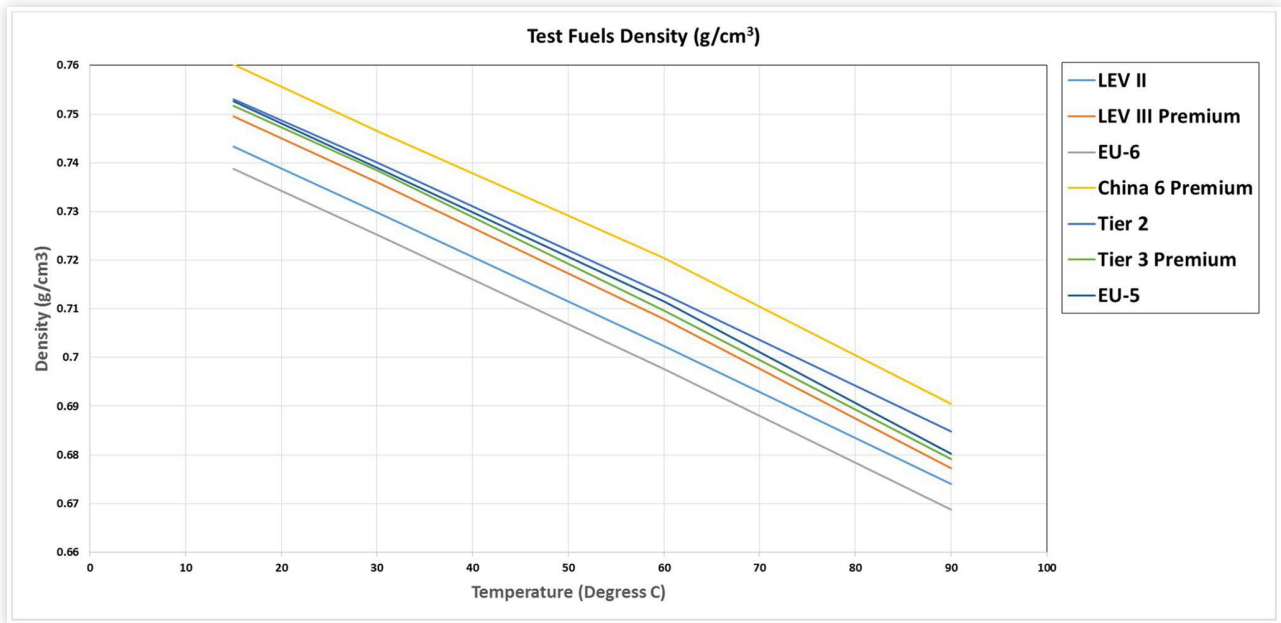
	Euro 5	LEV III Prem	Tier 3 Prem	LEV II	Euro 6	Tier 2	China 6 Premium	RSG E10	RSG M15
<b>Oxygenate Type</b>	Ethanol	Ethanol	Ethanol	MTBE	Ethanol	None	MTBE	Ethanol	Methanol MTBE
<b>Heat of Vaporization DSC/TGA (KJ/kg)</b>	421	425	423	381	461	372	385	426	513
<b>CARBON# ( Vol %)</b>	<b>Aromatics</b>	<b>Aromatics</b>	<b>Aromatics</b>	<b>Aromatics</b>	<b>Aromatics</b>	<b>Aromatics</b>	<b>Aromatics</b>	<b>Aromatics</b>	<b>Aromatics</b>
<b>1</b>	--	--	--	--	--	--	--	--	--
<b>2</b>	--	--	--	--	--	--	--	--	--
<b>3</b>	--	--	--	--	--	--	--	--	--
<b>4</b>	--	--	--	--	--	--	--	--	--
<b>5</b>	--	--	--	--	--	--	--	--	--
<b>6</b>	0.01	0.70	0.58	0.83	0.01	0.58	--	--	0.27
<b>7</b>	19.00	5.74	6.42	6.42	14.06	18.95	17.05	--	4.85
<b>8</b>	0.06	6.71	6.15	9.30	0.11	4.31	6.48	1.28	1.74
<b>9</b>	12.36	6.09	5.68	6.04	6.39	3.94	9.48	8.59	7.82
<b>10</b>	2.36	2.85	4.52	2.05	3.79	2.46	0.93	8.26	6.12
<b>11</b>	0.85	0.67	1.35	0.47	1.00	1.38	0.79	4.36	0.27
<b>12</b>	0.41	0.33	0.66	0.26	0.52	0.73	0.04	1.50	0.16
<b>13</b>	--	0.00	0.00	--	0.00	0.00	0.01	--	0.03
<b>TOTAL</b>	35.04	23.10	25.36	25.36	25.88	32.34	34.79	23.99	21.27
<b>C9 + Aromatics</b>	15.98	9.95	12.21	8.81	11.70	8.51	11.24	22.71	14.38
<b>C10 + Aromatics</b>	3.62	3.85	6.53	2.77	5.31	4.57	1.76	14.12	6.56
<b>PMI Number</b>	1.89	1.17	1.68	1.12	1.28	1.66	1.33	2.77	1.5

**TABLE A2** Fuel Properties as listed on the Certificate of Analysis from the supplier

Fuel Property	Euro 5	LEV III Prem	Tier 3 Prem	LEV II	Euro 6	Tier 2	China 6 Premium	RSG E10	RSG M15
RON D2699	96.9	99.8	99.5	100.5	96.3	96.5	96.6	>101	104
MON D2700	87.1	88.8	88	88.5	85.6	86.8	85.9	88.6	87.7
RVP @100F D5191 (psi)	8.52	7.2	9.2	6.71	8.27	8.83	8.5	10.6	13.9
Aromatic % D1319/5769	32.2		23.4	23.1	25.1	26	31.3	24.83	22.1
Olefin % D1319/6550	5.1		6.5	4.8	10.1	8.4	12.5	18.2	22.8
Oxygenate % D4815	5.07	10	9.71	10.88	9.87	0	8.17	9.87	15.3 7.1
Oxygenate Type	Ethanol	Ethanol	Ethanol	MTBE	Ethanol	None	MTBE	Ethanol	Methanol MTBE
Specific Gravity @15.56 D4052	0.7532	0.7494		0.7433		0.7401	0.7588		
Density @15 C (g/cc)	0.7529		0.7491		0.752			0.74	0.7393
T10 ( C ) D86		58.3	52.4	59.7	53.3	57.2	51.9	48.7	39.4
T90 ( C ) D86		155.9	160.7	144.3	160.7	149.4	160.8	185.9	166.6
T95 ( C ) D86		168.9	177.5		177.6	186.1	171.6	196.1	
FBP ( C ) D86		185.2	201.9	182.8	190	212.8	195.5	221.7	197.7
% Evaporated at 70C	34.1				44.9			45.6	63.2
% Evaporated at 100C	52				57.4			55.2	67.3
% Evaporated at 150C	84				87.5			79.2	87

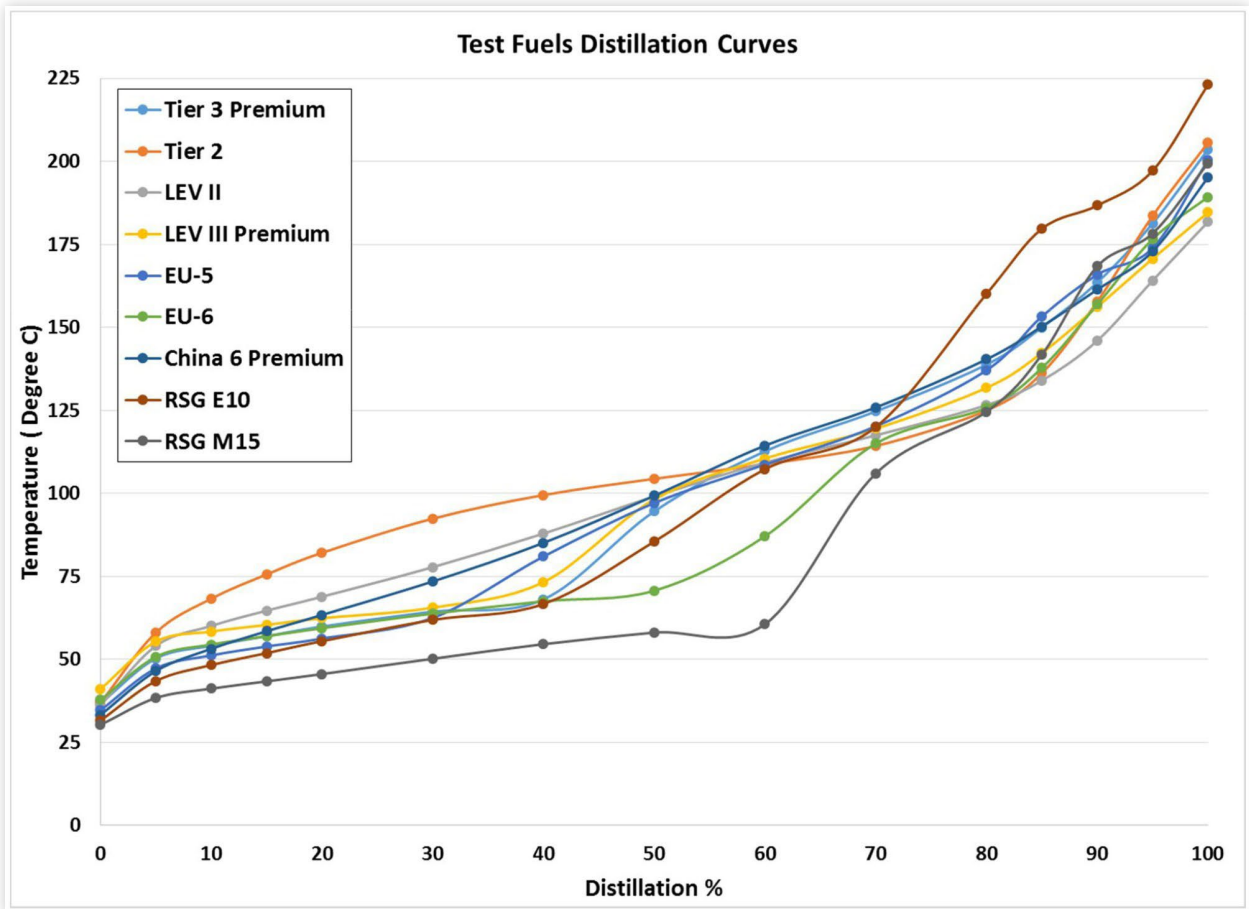
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**FIGURE A1** Chart of the test fuel densities (ASTM 4052).



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**FIGURE A2** Distillation Curve (ASTM D86) Chart for all fuels.



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