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Effects of the Particulate Matter Index and Particulate Evaluation Index of the Primary Reference Fuel on Particulate Emissions from Gasoline Direct Injection Vehicles

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Abstract: The purpose of this experimental study was to evaluate the range of particulate mass (PM) and particulate number (PN) results from gasoline direct injection (GDI) vehicles by using four test fuels with a range of particulate matter index (PMI) from 1.38 to 2.39 and particulate evaluation index (PEI) from 0.89 to 1.92. The properties of four test fuels were analyzed with detailed hydrocarbon analysis (DHA). Two passenger cars with a GDI engine were tested with four test fuels by conducting the China 6 test procedure, which is equivalent to the worldwide harmonized light-duty vehicle test procedure (WLTP). When the fuels could meet the China 6 primary reference fuel standard with PMI from 1.38 to 2.04 and PEI from 0.89 to 1.59, the PM variation of Vehicle B was from 1.94 mg/km to 3.32 mg/km and of Vehicle A was from 2.55 mg/km to 4.15 mg/km, respectively. In addition, the PN variation of Vehicle B was from 1.57×10^{12} #/km to 3.38×10^{12} #/km and of Vehicle A was from 3.02×10^{12} #/km to 4.80×10^{12} #/km. It was noted that the two different cars had a unique response and sensitivity by using the different fuels, but PMI and PEI did trend with both the PM and the PN response. All PM and PN results from the two cars had an excellent correlation $R^2 > 0.94$ with PMI and $R^2 > 0.90$ with PEI. Therefore, PMI/PEI would be the appropriate specification for sooting tendency in reference fuel standards of emission regulations.

Keywords: particulate mass; particulate number; primary reference fuel; particulate matter index; particulate evaluation index; worldwide harmonized light-duty vehicle test procedure

1. Introduction

Reference fuel standards are defined by a relatively narrow range of specifications, in contrast to market fuel standards in vehicle emissions and fuel economy regulations, which aim to improve the repeatability and reproducibility of vehicle type approval (TA), conformity of production (COP), and in-used conformity (IUC) [1,2]. The specifications of reference fuel standards are part of vehicle emission/fuel economy regulations and are revised along with regulation upgrades. The reproducibility of TA, COP and IUC needs to be well managed, otherwise it would cause a potential risk to emission compliance. Previous studies showed that the framework of reference fuel could present very good repeatability and reproducibility with conventional gaseous pollutants such as carbon oxide (CO), hydro carbon (HC), and nitrogen oxide (NOx) [3]. Some key parameters which can be attributed to particulate emission have been strictly controlled in China 6 emission standards and would have a limited impact, e.g. sulfur content <10 ppm [4] and Mn content $\leq$2 ppm [5,6]. However, the China 6 reference fuel framework has carried over the previous one for more than a decade lacking an appropriate specification that has a strong correlation with PM and PN results.

The new vehicle emission regulations around the world urged more stringent particulate emission. The EU introduced a significant reduction in PN from 6×10^{12} #/km by Euro 6b to 6×10^{11} #/km by Euro 6c [7]. $PM_{2.5}$ is a major health concern in China and motor vehicles contribute to 10%–50% of $PM_{2.5}$ in megacities of the Chinese east coast [8]. Particulate emissions from gasoline engines are comprised of carbon soot, sulfates, ash, and soluble organic fraction (SOF) of unburned fuel [9]. As one of the counter measures, China developed the China 6 emission regulation with a phase-in approach and requested that all light duty vehicles be compliant with PN at 6×10^{11} #/km and PM at 4.5 mg/km for China 6a and 3.0 mg/km for China 6b. Before July, 2020, a transition limit of 6×10^{12} #/km should be applied for PN [10]. Furthermore, the EU and China also introduced the real driving emission (RDE) requirement for NOx and PN. U.S. emission regulations historically did not regulate PN, but Tier 3 requested very stringent emission of PM at 3 mg/mile phase-in, starting from 2017 model year nationwide and 1 mg/mile phase-in starting in 2025 in some states [11]. All of these upcoming regulations present big challenges to manufacturers for emission compliance, and have also increased the awareness of all factors contributing to vehicle tailpipe emission, especially the influence of fuel.

Various studies indicated that there are some other indices linking the measures of aromatic content and vapor pressure and having an excellent correlation between fuel composition and PM and PN emissions [12]. PMI was developed by Aikawa et al. from Honda as a predictive model of particulate emissions related to fuel properties and correlated with actual engine-out emission results [13]. PMI is given by the following equation as:

$$\mathrm{PMI} = \sum_{\mathrm{i}=1}^{\mathrm{n}} \mathrm{I}_{[443\mathrm{K}]} = \sum_{\mathrm{i}=1}^{\mathrm{n}} \left(\frac{DBE_{\mathrm{i}}+1}{\mathrm{V.P}(443\mathrm{K})_{\mathrm{i}}} \times Wt_{\mathrm{i}} \right) \quad (1)$$

where Wt is the weight fraction of an identified constituent molecule within a fuel sample, DBE is double bond equivalence and achieved by detailed hydrocarbon analysis (DHA) from American society for testing and materials (ASTM) standard D6729, n is the total number of compounds in the fuel, and V.P (443 K) is the vapor pressure at 443 K. The significant paper took into account V.P at 443 K of the gasoline range and DBE of all compounds in gasoline. The result was obtained by the DHA method [14–16]. PEI was developed by Chapman et al. from General Motors and has an excellent correlation $R^2 = 0.93$ with PMI, which focuses more on just aromatics instead of the whole spectrum of molecules [17]. The calculation involved the use of a regression equation based on the aromatics from the fuel, and was derived based on various fuel data sets in comparison with the PMI number. Both indices provide an indication of sooting tendency and are widely used by the industry as a metric to understand the impact of fuel on sooting.

This study further investigated the impact of PM/PN emissions by PMI/PEI variations in the reference fuel over a legislative drive cycle, which is also a research hotspot for academia, the industry and the government. In this experimental setup, four test fuels were synthetized with different blending compounds to have a linear PMI range from 1.38 to 2.39 and PEI from 0.89 to 1.92. One sport utility vehicle (SUV) and one multi-purpose vehicle (MPV) with a GDI engine were tested by conducting the China 6 type I test procedure, which is equivalent to the worldwide harmonized light-duty vehicle test procedure (WLTP), to evaluate the range of PM and PN results. Moreover, four test fuels were analyzed by DHA and PIONA (basic data from hydrocarbon analysis of paraffins, iso-paraffins, olefins, naphthenes, and aromatics). The compounds in the fuels which would significantly contribute to the PEI/PMI value were investigated. Afterwards, the PM and PN results were presented and correlated with fuel properties.

2. Experimental Setup and Test Procedure

2.1. Fuels

The four test fuels were synthetized by local refiners and filled into a 5 Liter container. To ensure that PMI and PEI can represent the range of market fuel, Fuels A, B, C and D were well designed to blend with different blending compounds in order to have a linear PMI and PEI distribution [18]. Before shipping them to the test lab, the fuels were sampled and tested. Before the vehicle emission test was conducted, the fuels were analyzed by DHA and were compared with the primary reference fuel standard of the China 6 emission standards, which was defined by Annex K of the Chinese national standard GB 18352.6-2016 [10]. The key specifications of the test fuels and the China 6 primary reference fuel and market fuel requirements are in Table 1. It can be noted that Fuels A, B and C could meet the China 6 primary reference standards and market fuel standards but Fuel D had a higher final boiling point (FBP).

Table 1. Key specifications for test fuels, China 6 primary reference fuel and market fuel standards.

Specification	Unit	Fuel A	Fuel B	Fuel C	Fuel D	China 6 Primary Reference Fuel Standard	China 6 Market Fuel Standard
Density	kg/m^3	740	740	745	744	735–755 (RON92–94) 745–760 (RON95–98)	720–775
Research Octane Number (RON)	Unit	93.47	93.65	93.73	90.54	RON92–94 RON95–98	≥89 (#89) ≥92 (#92) ≥95 (#95)
Reid Vapor Pressure (RVP)	kPa	57.5	56.0	57.0	56.0	56–60	45–85 (1st Nov.–30th Apr.) 40–65 (1st May–31st Oct.)
Sulfur	ppm	1	<1	<1	1	≤10	≤10
Mn	mg/L	<1	<1	<1	<1	≤2	≤2
Pb	mg/L	<1	<1	<1	<1	≤5	≤5
Fe	mg/L	<1	<1	<1	<1	≤10	≤10
T10	°C	58	58	57	57	50–65	≤70
T50	°C	92	93	92	95	90–105	≤110
T90	°C	153	160	162	175	150–165	≤190
FBP	°C	191	198	200	216	190–200	≤205
Aromatic	Vol%	28.3	28.8	30.8	31.2	27–32 (RON92–94) 30–35 (RON95–98)	≤35

2.2. Test Vehicle

One SUV with a 1.5 turbo charged GDI engine and one MPV with a 2.0 turbo charged GDI engine were selected because this type of engine would emit more particulate emission than a port fuel injection (PFI) engine [19–21]. These vehicles were engineered and type-approved to comply with the China 5 emission standards which are equivalent to the Euro 5 ones. The technical specifications are listed in Table 2.

Table 2. Vehicle specifications.

Technical Specifications	Vehicle A (SUV)	Vehicle B (MPV)
Displacement	1490 cc	1998 cc
Fuel Injection	Direct Injection	Direct Injection
Boost	Turbocharger	Turbocharger
Power	124 kW	191 kW
Cylinder/Valves	4/16	4/16
Transmission	7 DCT	6AT
Curb mass	1575 kg	1878 kg
Fuel consumption at New European Driving Cycle (NEDC)	6.6 L/100 km	8.8 L/100 km

2.3. Test Apparatus

Testing was performed in a China6/Euro 6 capable climate-controlled test lab on a 44-inch single-roll chassis dynamometer. A Horiba MEXA-7400LE gas analyzer system was used to measure the exhaust gas composition collected through a constant volume sampling (CVS) system. PN was measured through a particulate measurement programme (PMP) system in accordance with the China 6 emission standard. A HORIBA MEXA-1000SPCS was used to count PN. Before the HORIBA MEXA-1000SPCS counted PN, volatile particles were removed by a volatile particle remover (VPR), and the solid particle counting system (SPCS) only counted the number of solid particles. PM was measured by the filter gravimetric method: Four 47 mm diameter fiberglass filters were applied in a WLTP test. PM measurement was not made through the VPR, so both volatile and solid particle mass was captured by the filters and measured by analytical balance later on. A schematic of the chassis dynamometer with measurement devices is in Figure 1.

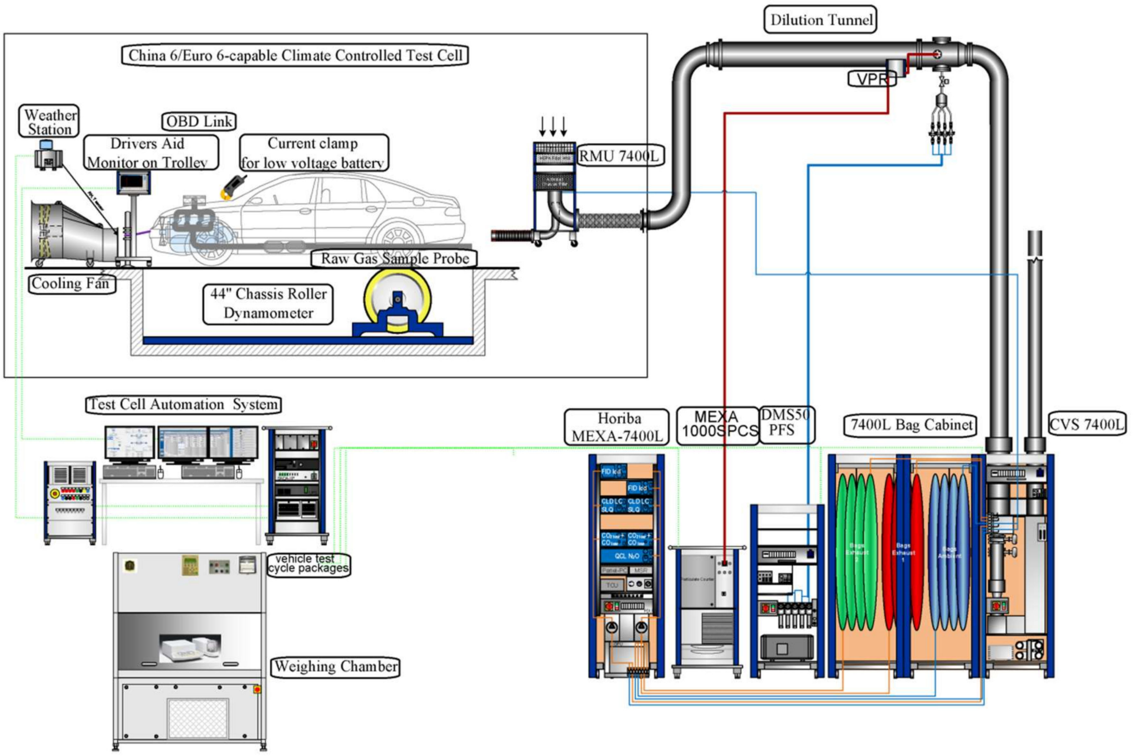

Figure 1. Chassis dynamometer and exhaust gas measurement device.

2.4. Test Procedure

The experimental test was initiated following the standardized China 6 test procedure. During soaking, the test vehicle was charged and tire pressure was adjusted accordingly. What is more, the same driver was assigned to conduct the experiments, to reduce other effects to the results.

The key phase was the replacement of experimental fuel. The vehicle was driven until all existing fuel in the tank was drained. Afterwards, the vehicle was refueled with 5 Liters of test fuel and then the fuel was drained on the dynamometer. This fuel changing cycle was repeated twice. Before conducting the type I test of China 6, the vehicle was filled with 15 Liters of test fuel, which was estimated to be adequate for at least three complete WLTP tests. Each fuel was used for dyno tests at least two times, with a standardized soaking in between. The China 6 test procedure with low, medium, high, and extra high phases was used for test setting. The complete test setting flowchart is in Figure 2. In the next section, the test result for each fuel will be documented.

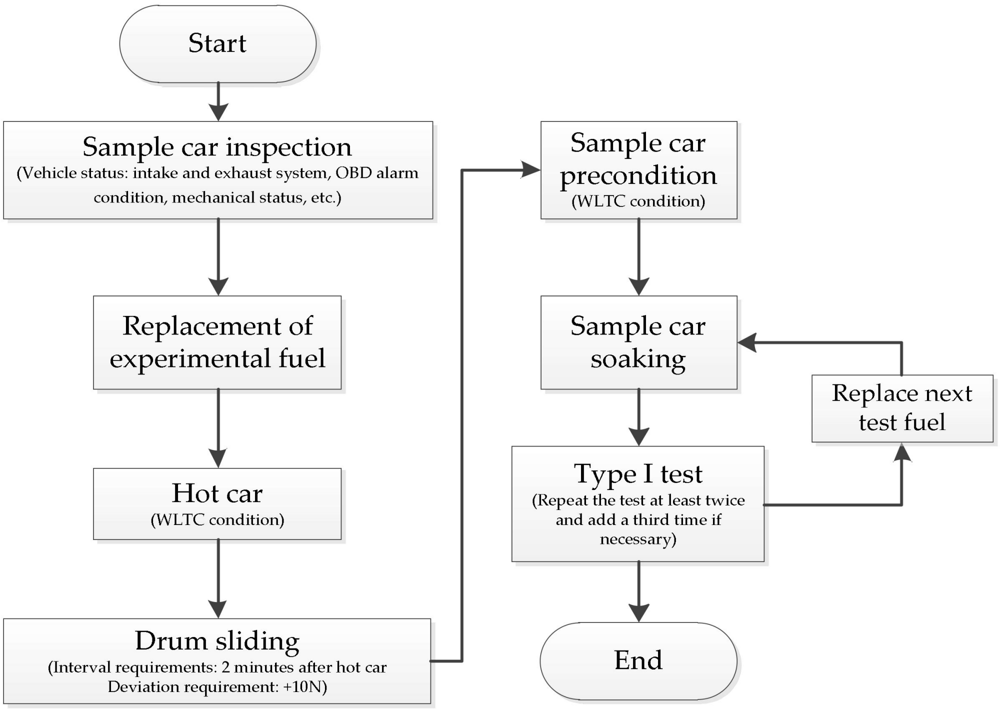

Figure 2. Flowchart for the test procedure.

3. Results and Discussion

3.1. Fuel Properties

Table 3 and Figures 3–5 present the particulate indices from DHA analysis, PIONA analysis and the distillation curve on the four test fuels. The PMI and PEI values were calculated based on DHA analysis (see Table 3).

Table 3. Particulate matter index (PMI) and particulate evaluation index (PEI) values from the sample sets.

	Fuel A	Fuel B	Fuel C	Fuel D
PMI	1.38	1.60	2.04	2.39
PEI	0.89	1.22	1.59	1.92

PIONA (paraffins, iso-paraffins, olefins, naphthenes, and aromatics) separates the fuels into individual compounds and identifies them by carbon number, except when only one species is present (e.g., benzene, etc.). The compound group amounts per carbon number were calculated by addition of the single compound of one group.

From the data shown in Figure 3, it can be seen that Fuels A and C had the lowest mass content of paraffins. There was no obvious difference in the content of olefins among the four fuels. The content of iso-paraffins decreased progressively from Fuel A to D. It should also be noted that the content of naphthenes decreased significantly from Fuel A to D. In addition, the content of aromatics increased progressively from Fuel A to D. It is clear that the aromatic content was positively correlated with the PMI/PEI index.

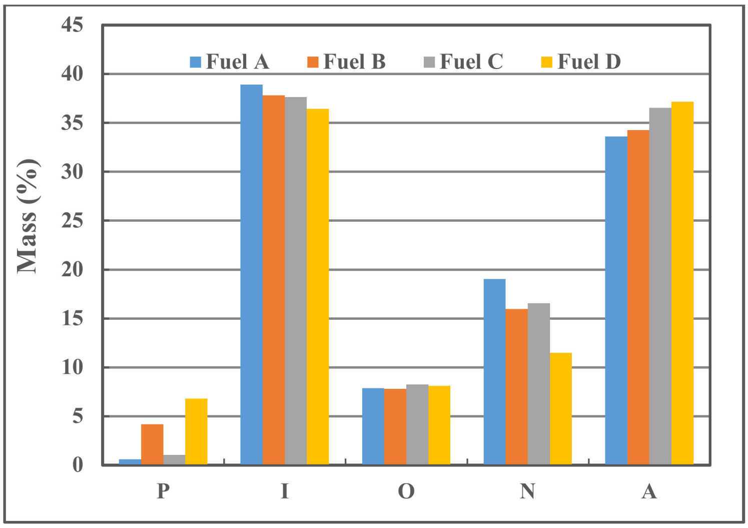

Figure 3. PIONA (paraffins, iso-paraffins, olefins, naphthenes, and aromatics) analysis results.

More detailed aromatic mass content data is shown in Figure 4. The content of total aromatic hydrocarbons increased from Fuel A to D. It has to be noted that the most significant among the four fuels was the content decreasing from carbon number 7 compound (C7) to C11. When looking further into each carbon number group of C9, C10 and C11, the content progressively increased form Fuel A to Fuel D. The content of the C8 aromatic remained the same level among the four fuels, but toluene (C7) significantly decreased from Fuel A to D. The experimental results showed that heavy aromatic (C9 and above) content was significantly positively correlated with the PMI/PEI value. This was consistent with a referenced study that stated that heavy aromatics are major contributors in PEI [17].

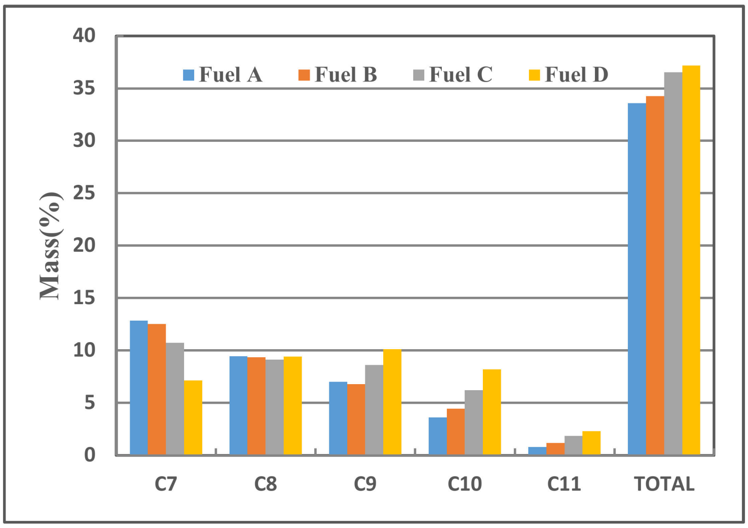

Figure 4. Aromatics content with different carbon numbers and total aromatic hydrocarbons.

The difference in aromatics is further illustrated in the following distillation curve in Figure 5. As can be observed, the differences in distillation among the four test fuels appeared after T50 (the temperature of 50% of distilled volume). T70 and T90 of Fuel A had the lowest temperature and Fuel D had the highest one after T50. This was observed from the above DHA data that Fuel D presented the highest content of heavy aromatics and fuel A had the lowest one. It was also seen by the PEI value which was more focused on DBE in aromatics.

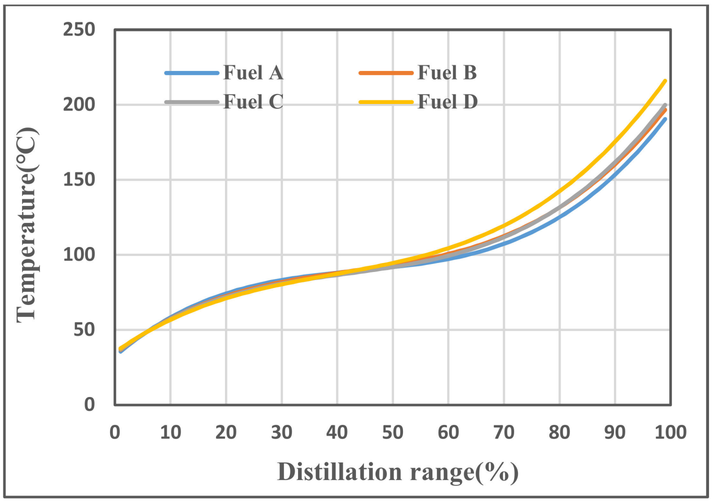

Figure 5. Distillation curve.

3.2. Particulate Mass and Particulate Number Emissions

Each paired PM and PN result was calculated based on the average value from at least two China 6 type I test paired results by using the same test fuel. Table 4 shows these test results and the corresponding uncertainties in measurement, which are calculated based on a reference study [22,23]:

Table 4. Test results and uncertainties in measurement.

		Test Fuel	Test 1	Test 2	Average	Combined Standard Uncertainty
Vehicle A	PM mg/km	A	2.58	2.53	2.55	0.02
		B	3.58	3.49	3.54	0.04
		C	4.12	4.18	4.15	0.03
		D	5.70	5.58	5.64	0.06
	PN #/km	A	2.79×10^{12}	3.26×10^{12}	3.02×10^{12}	1.67×10^{11}
		B	4.08×10^{12}	3.87×10^{12}	3.98×10^{12}	7.84×10^{10}
		C	4.55×10^{12}	5.06×10^{12}	4.80×10^{12}	1.82×10^{11}
		D	5.59×10^{12}	5.29×10^{12}	5.44×10^{12}	1.12×10^{11}
Vehicle B	PM mg/km	A	1.92	1.95	1.94	0.02
		B	2.53	2.24	2.39	0.10
		C	3.36	3.29	3.32	0.03
		D	5.54	4.82	5.18	0.26
	PN #/km	A	1.45×10^{12}	1.69×10^{12}	1.57×10^{12}	8.30×10^{10}
		B	1.75×10^{12}	1.77×10^{12}	1.76×10^{12}	1.33×10^{10}
		C	3.48×10^{12}	3.28×10^{12}	3.38×10^{12}	7.54×10^{10}
		D	3.94×10^{12}	3.79×10^{12}	3.87×10^{12}	5.97×10^{10}

Figures 6–9 present the correlation between particulate indices and the particulate emission results by using 4 test fuels over WLTP. In Figure 6, the x-axis is the value of PMI corresponding to four test fuels from Vehicle B with a 2 Liter turbocharged GDI engine. The left y-axis is the PM result from the China 6 type I emission test, and the right y-axis is the PN result. The limits of China 6b PM and China 6 transition PN from the China 6 emission standard have been illustrated as the reference. It was noted that PM results from Vehicle B varied from 1.94 mg/km to 5.18 mg/km. Looking further into the results from Fuels A, B and C, which met the China 6 primary reference standards, PM results were from 1.94 mg/km to 3.32 mg/km and the ratio of maximum and minimum value was 1.71. Comparing these with the China 6 limit, it can be concluded that PM results, by using Fuels A and B, can meet the China 6b limit of 3 mg/km. It was also noticed that PN results varied from 1.57×10^{12} #/km

to 3.87 × 10^{12} #/km. Looking further into the results from Fuels A, B and C, the PN results were from 1.57 × 10^{12} #/km to 3.38 × 10^{12} #/km and the ratio of maximum and minimum value was 2.15. Comparing these with the China 6 limit, it can be concluded that PN results, by using four test fuels, can meet the China 6 transition limit (before July 2020) of 6 × 10^{12} #/km. Additionally, PMI offered an excellent correlation R^2 = 0.9434 with PM and R^2 = 0.9594 with PN, respectively.

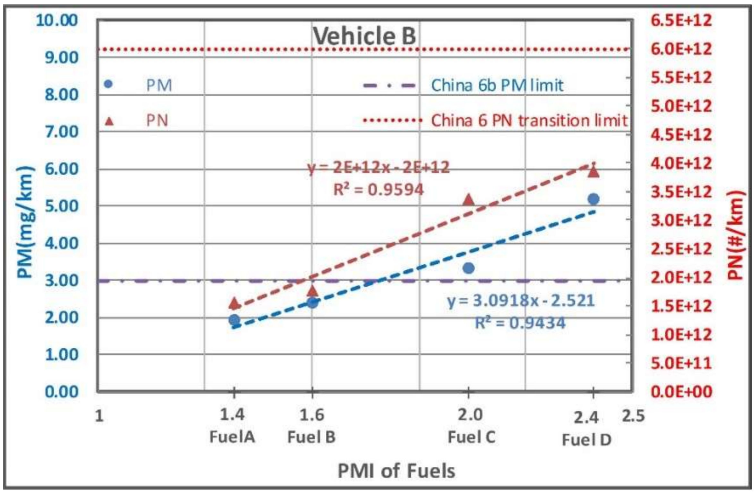

Figure 6. PMI vs. particulate mass (PM) and particulate number (PN) emissions from Vehicle B.

In Figure 7, the x-axis is the value of PMI corresponding to four test fuels from Vehicle A with a 1.5 Liter turbocharged GDI engine. The left y-axis is the PM result from the China 6 type I emission test, and the right y-axis is the PN result. It was noted that PM results from Vehicle A varied from 2.55 mg/km to 5.64 mg/km. Looking further into the PM results from Fuels A, B and C, which met the China 6 primary reference standard, the PM results were from 2.55 mg /km to 4.15 mg/km and the ratio of maximum and minimum value was 1.62. Comparing these with the China 6 limits, it can be concluded that the PM result, by using Fuel A, can meet the China 6b limit of 3 mg/km. It was also noticed that PN results varied from 3.02 × 10^{12} #/km to 5.44 × 10^{12} #/km. Looking further into the results from Fuels A, B and C, which could meet the China 6 primary reference standard, the PN results were from 3.02 × 10^{12} #/km to 4.80 × 10^{12} #/km and the ratio of maximum and minimum value was 1.59. Comparing these with the China 6 emission standard, it can be concluded that PN results, by using four test fuels, can meet the China 6 transition limit (before July 2020) of 6 × 10^{12} #/km. Additionally, PMI offered an excellent correlation R^2 = 0.9553 with PM and R^2 = 0.9656 with PN, respectively.

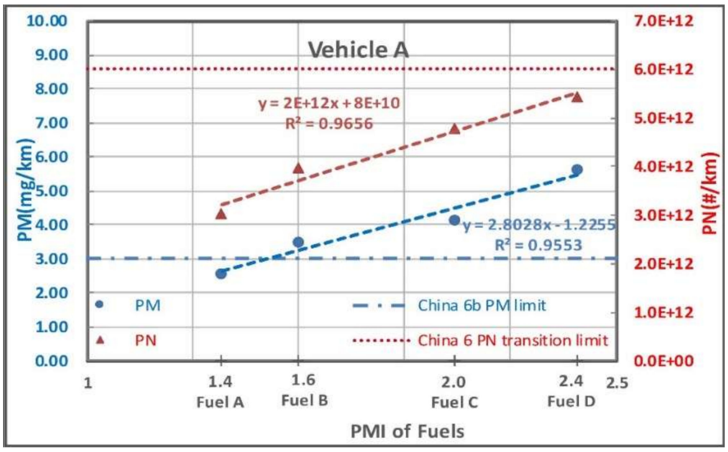

Figure 7. PMI vs. PM and PN emissions from Vehicle A.

The data shown in Figure 8 concern the correlation between PEI and the PM/PN results from Vehicle B. PEI offered a good correlation R^2 = 0.918 with PM and R^2 = 0.9229 with PN, respectively.

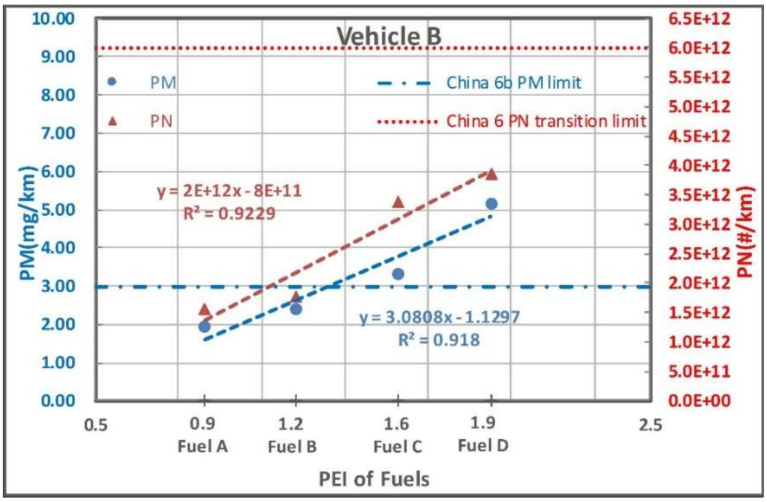

Figure 8. PEI vs. PM and PN emissions from Vehicle B.

The data shown in Figure 9 concern the correlation between PEI and PM/PN emission results from Vehicle A. PEI offered an excellent correlation R^2 = 0.9670 with PM and R^2 = 0.9920 with PN, respectively.

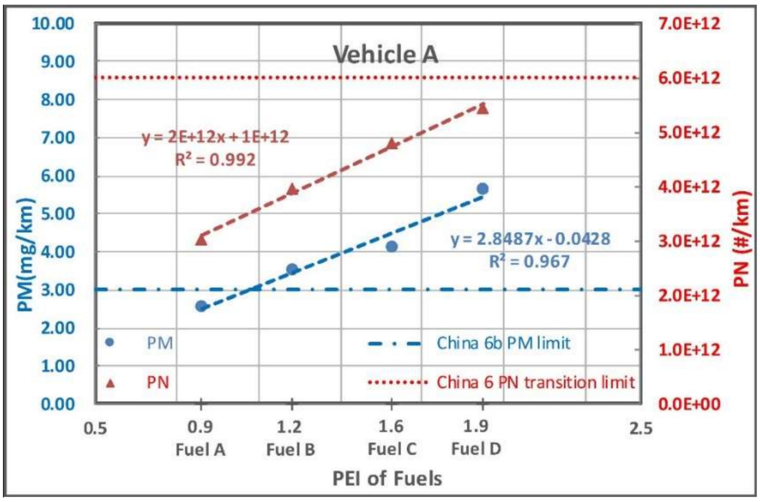

Figure 9. PEI vs. PM and PN emissions from Vehicle A.

When Fuels A, B and C could meet the China 6 primary reference fuel standard, all PN results could meet the China 6 emission limit of 6×10^{12} #/km during the transition period (before July 2020) but could not meet the final stage (after July 2020) of 6×10^{11} #/km for both cars. In terms of PM emissions, only the result by using Fuel A could ensure both cars meeting the China 6b limit of 3 mg/km. PM variations were up to 1.71 from Vehicle B and up to 1.62 from Vehicle A, while PN variations were up to 2.15 from Vehicle B and up to 1.59 from Vehicle A. Based on these test results, the risk for emission incompliance could be forecasted if this specific vehicle model was tested using a primary reference fuel with low sooting tendency (e.g., Fuel A) for TA, but later on retested by using the primary reference fuel with higher sooting tendency (e.g., Fuel C) for COP and IUC.

It is noted that two cars with a different curb mass and powertrain combination have a unique response and sensitivity. When comparing the PM and PN results from the two test vehicles using the same test fuel, the variation of PM and PN in Vehicle A was relatively lower than that in Vehicle B.

It can be concluded that Vehicle A has a relatively lower sensitivity than Vehicle B on sooting tendency with test fuels. However, there are various other factors influencing the fuel burning process and after treatment efficiency. Nevertheless, PMI and PEI did trend with both the PM and the PN response in this study.

Both PM and PN data sets from Vehicles A and B were excellently R-squared with PEI and PMI. This was consistent with previous studies that suggested that the existence of an aromatic ring occurs in the early stage of the fundamental particulate formation process. Moreover, heavy aromatics such as hydrocarbon C9, C10 and C11 should have a high boiling point and more DBE [20,24,25]. This was also proven by the distribution curve in Figure 5. It may be attributed to the fact that fuels with low heavy aromatic content should have less sooting tendency [26–28].

4. Summary and Recommendations

The goal of this study was to evaluate the range of PM and PN results from cars with a GDI engine using test fuels with linear PMI and PEI. Four test fuels were blended and analyzed with DHA and PIONA. Two passenger cars with a GDI engine were instrumented to be tested with four test fuels using WLTP of the China 6 emission standards.

Based on the conducted experiments and associated analyses and evaluations, the following conclusions are drawn:

These four test fuels had a wide PMI distribution from 1.38 to 2.40 and PEI from 0.89 to 1.92. When Fuels A, B and C could meet the China 6 reference fuel standards, the PMI distribution was from 1.38 to 2.04 and PEI was from 0.89 to 1.59.

With the increasing of PMI and PEI, the content of aromatics in Fuels A, B, C and D progressively increased. The distillation curve and the content in each carbon number showed a heavy aromatic increase from Fuel A to D. It can be concluded that heavy aromatics are major contributors to the PMI and PEI value.

Wide PM and PN emission variations could be observed from the two vehicles' engine-out emissions. When Fuels A, B and C could meet the China 6 reference fuel standard, the PM variation of Vehicle B was from 1.94 mg/km to 3.32 mg/km and of Vehicle A was from 2.55 mg/km to 4.15 mg/km, respectively. Additionally, the PN variation of Vehicle B was from 1.57×10^{12} #/km to 3.38×10^{12} #/km and of Vehicle A was from 3.02×10^{12} #/km to 4.80×10^{12} #/km, respectively.

The two GDI cars had a unique response and sensitivity. However, the PM and PN results from the two GDI cars were exactly linear with PMI and PEI. All data sets showed that when the PMI and PEI value increased, the PM and PN response increased. PM and PN results had a very high correlation $R^2 > 0.94$ with PMI and $R^2 > 0.9$ with PEI.

This study indicated that the current framework of the reference fuel standard should be further improved in order to improve the reproducibility of PM and PN emissions. Considering the excellent correlation with sooting tendency, PMI and PEI would be the appropriate specification to be introduced into the reference fuel standard.

A downside to this study is the small data samples with two cars and four test fuels; despite how good the correlations were, PMI and PEI could only be considered as comparative rather than predictive for sooting tendency. It would make sense to produce expanded fuel samples and fleet tests in order to verify and establish predictive modelling for sooting tendency in the reference fuel standard.

As a short term measure, recording the PMI/PEI value of the reference fuel should be considered when the TA test has been conducted on a specific vehicle model. If there is a dispute or failure while following COP and IUC testing for this specific model, it should be allowed to verify the PMI/PEI value of the reference fuel within a reasonable offset by comparing it with the value during TA.

Previous studies showed that burning of different levels of ethanol fuel in PFI engines and GDI engines would result in both an increase and decrease in PM and PN emissions [29–31]. Considering the Chinese government will have implemented E10 gasoline (10% ethanol blends) fuel nationwide by

2020, it is also important to understand the impact of ethanol blending fuel if the reference fuel in the emission standard changed to E10 accordingly.

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Conflicts of Interest: All authors declare that there is no conflict of interest.

References

1. McCormick, R.L.; Alvarez, J.R.; Graboski, M.S.; Tyson, K.S.; Vertin, K. *Fuel Additive and Blending Approaches to Reducing NOx Emissions from Biodiesel*; SAE Technical Paper 2002-01-1658; SAE International: Warrendale, PA, USA, 2002.
2. McNutt, B.; Pirkey, D.; Dulla, R.; Miller, C. *A Comparison of Fuel Economy Results from EPA Tests and Actual In-Use Experience, 1974–1977 Model Year Cars*; SAE Technical Paper 780037; SAE International: Warrendale, PA, USA, 1978.
3. Zhai, Q.; Yang, Z.; Gao, J. Research on trends of liquid reference fuels in automobile emission standards. *Auto Ind. Res.* **2016**, 17–23. [CrossRef]
4. Wall, J.C.; Shimpi, S.A.; Yu, M.L. *Fuel Sulfur Reduction for Control of Diesel Particulate Emissions*; SAE Technical Paper 872139; SAE International: Warrendale, PA, USA, 1987.
5. McCabe, R.W.; DiCicco, D.M.; Guo, G.; Hubbard, C.P. *Effects of MMT®Fuel Additive on Emission System Components: Comparison of Clear- and MMT®-fueled Escort Vehicles from the Alliance Study*; SAE Technical Paper 2004-01-1084; SAE International: Warrendale, PA, USA, 2004.
6. Peng, G.; Yao, C. Corrigendum to "Combustion and emission characteristics of a direct-injection gasoline engine using the MMT fuel additive gasoline" [Fuel 144 (2015) 380–387]. *Fuel* **2016**, *165*, 554. [CrossRef]
7. Alessandro, M.; Jelica, P.; Biagio, C.; Simone, S.; Georgios, F. Gaseous emissions from light-duty vehicles: Moving from NEDC to the new WLTP test procedure. *Environ. Sci. Technol.* **2015**, *49*, 8315–8322. [CrossRef]
8. Ministry of Ecology and Environment of the People's Republic of China China Vehicle Environmental Management Annual Report. Available online: http://dqhj.mee.gov.cn/jdchjgl/zhgldt/201806/P020180604354753261746.pdf (accessed on 15 January 2019).
9. Arvind, T.; Besch, M.C.; Seungju, Y.; John, C.; Hemanth, K.; Carder, D.K.; Alberto, A.; Jorn, H.; Mridul, G. Characterization of particulate matter emissions from a current technology natural gas engine. *Environ. Sci. Technol.* **2014**, *48*, 8235. [CrossRef]
10. Ministry of Ecology and Environment of the People's Republic of China. GB 18352.6-2016 Limits and Measurement Method for the Emission of Light Duty Vehicle (China 6). Available online: http://www.vecc.org.cn/180514/1-1P514104206.pdf (accessed on 15 January 2019).
11. Sellnau, M.; Foster, M.; Moore, W.; Sinnamon, J.; Hoyer, K.; Klemm, W. Second generation GDCI multi-cylinder engine for high fuel efficiency and US Tier 3 emissions. *SAE Int. J. Engines* **2016**, *9*, 1002–1020. [CrossRef]
12. Myung, C.L.; Ko, A.; Park, S. Review on characterization of nano-particle emissions and PM morphology from internal combustion engines: Part 1. *Int. J. Autom. Technol.* **2014**, *15*, 203–218. [CrossRef]
13. Aikawa, K.; Sakurai, T.; Jetter, J.J. Development of a predictive model for gasoline vehicle particulate matter emissions. *SAE Int. J. Fuels Lubr.* **2010**, *3*, 610–622. [CrossRef]
14. Aikawa, K.; Jetter, J.J. Impact of gasoline composition on particulate matter emissions from a direct-injection gasoline engine: Applicability of the particulate matter index. *Int. J. Engine Res.* **2014**, *15*, 298–306. [CrossRef]
15. Khalek, I.A.; Bougher, T.; Jetter, J.J. Particle emissions from a 2009 gasoline direct injection engine using different commercially available fuels. *SAE Int. J. Fuels Lubr.* **2010**, *3*, 623–637. [CrossRef]

16. Leach, F.; Stone, R.; Richardson, D. *The Influence of Fuel Properties on Particulate Number Emissions from a Direct Injection Spark Ignition Engine*; SAE Technical Paper 2013-01-1558; SAE International: Warrendale, PA, USA, 2013.
17. Chapman, E.; Winston-Galant, M.; Geng, P.; Latigo, R.; Boehman, A. *Alternative Fuel Property Correlations to the Honda Particulate Matter Index (PMI)*; SAE Technical Paper 2016-01-2250; SAE International: Warrendale, PA, USA, 2016.
18. Chapman, E.; Winston-Galant, M.; Geng, P.; Konzack, A. *Global Market Gasoline Range Fuel Review using Fuel Particulate Emission Correlation Indices*; SAE Technical Paper 2016-01-2251; SAE International: Warrendale, PA, USA, 2016.
19. Chen, L.; Liang, Z.; Xin, Z.; Shuai, S. Characterizing particulate matter emissions from GDI and PFI vehicles under transient and cold start conditions. *Fuel* **2017**, *189*, 131–140. [CrossRef]
20. Chen, L.; Stone, R.; Richardson, D. A study of mixture preparation and PM emissions using a direct injection engine fuelled with stoichiometric gasoline/ethanol blends. *Fuel* **2011**, *90*, 120–130. [CrossRef]
21. He, X.; Ratcliff, M.A.; Zigler, B.T. Effects of gasoline direct injection engine operating parameters on particle number emissions. *Energy Fuels* **2012**, *26*, 2014–2027. [CrossRef]
22. Ye, S.; Li, L.; Jing, S.; Shi, Z. Uncertainty estimation in light-duty vehicle type I test. *Energy Conserv. Environ. Prot. Transp.* **2017**, *13*, 9–13. [CrossRef]
23. JCGM: 200 Evaluation of Measurement Data—Guide to the Expression of Uncertainty in Measurement. Available online: https://www.bipm.org/utils/common/documents/jcgm/JCGM_100_2008_E.pdf (accessed on 15 January 2019).
24. Kelly, F.J.; Fussell, J.C. Size, source and chemical composition as determinants of toxicity attributable to ambient particulate matter. *Atmos. Environ.* **2012**, *60*, 504–526. [CrossRef]
25. Überall, A.; Otte, R.; Eilts, P.; Krahl, J. A literature research about particle emissions from engines with direct gasoline injection and the potential to reduce these emissions. *Fuel* **2015**, *147*, 203–207. [CrossRef]
26. Mwangi, J.K.; Lee, W.-J.; Tsai, J.-H.; Wu, T.S. Emission reductions of nitrogen oxides, particulate matter and polycyclic aromatic hydrocarbons by using microalgae biodiesel, butanol and water in diesel engine. *Aerosol Air Qual. Res.* **2015**, *15*, 901–914. [CrossRef]
27. Georgios, K.; Daniel, S.; Diep, V.; Robert, R.; Maryam, H.; Akua, A.A.; Durbin, T.D. Evaluating the effects of aromatics content in gasoline on gaseous and particulate matter emissions from SI-PFI and SIDI vehicles. *Environ. Sci. Technol.* **2015**, *49*, 7021–7031. [CrossRef]
28. Bielaczyc, P.; Szczotka, A.; Woodburn, J. Regulated and unregulated exhaust emissions from CNG fueled vehicles in light of Euro 6 regulations and the new WLTP/GTR 15 test procedure. *SAE Int. J. Engines* **2015**, *8*, 1300–1312. [CrossRef]
29. Whitaker, P.; Kapus, P.; Ogris, M.; Hollerer, P. Measures to reduce particulate emissions from gasoline DI engines. *SAE Int. J. Engines* **2011**, *4*, 1498–1512. [CrossRef]
30. Storey, J.M.E.; Barone, T.L.; Norman, K.M.; Lewis, S.A. Ethanol blend effects on direct injection spark-ignition gasoline vehicle particulate matter emissions. *SAE Int. J. Fuels Lubr.* **2010**, *3*, 650–659. [CrossRef]
31. Sarathy, S.M.; Oßwald, P.; Hansen, N.; Kohsehöinghaus, K. Alcohol combustion chemistry. *Prog. Energy Combust. Sci.* **2014**, *44*, 40–102. [CrossRef]