

Novel Index for Evaluation of Particle Formation Tendencies of Fuels with Different Chemical Compositions

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ABSTRACT

Current regulatory developments aim for stricter emission limits, increased environmental protection and purification of air on a local and global scale. In order to find solutions for a cleaner combustion process, it is necessary to identify the critical components and parameters responsible for the formation of emissions. This work provides an evaluation process for particle formation during combustion of a modern direct injection engine, which can help to create new aftertreatment techniques, such as a gasoline particle filter (GPF) system, that are fit for purpose. With the advent of “real driving emission” (RDE) regulations, which include market fuels for the particulate number testing procedure, the chemical composition and overall quality of the fuel cannot be neglected in order to yield a comparable emission test within the EU and worldwide. Even within the legislative boundaries such as the DIN EN 228 in Europe, critical gasoline parameters can vary greatly and influence the measured particle number. This work provides a straightforward approach to evaluate any gasoline worldwide considering particle emission tendencies via comprehensible chemical analyses and transparent interpretation. The Menger/Wittmann model (MW index) presented in this work consists of fourteen fundamental physico-chemical parameters (density, vapor pressure, distillation range, volumetric fraction of aromatics, olefins, etc.), measured with standardized and reproducible techniques. These macroscopic quantities incorporate the complex interactions of the chemical single components and directly represent the matrix effects responsible for the magnitude of particle formation during the combustion cycle. An exponential dependence of the PN emissions on the index model and its chemical foundation was found, supporting the notion of cumulative matrix effects of critical parameters applied in this model. In order to validate the correlation and to understand the fundamental parameters, which are necessary to evaluate fuels worldwide, the PN emissions of several reference, market and customized fuels were measured on an engine test bed under laboratory conditions with a customer-related RDE cycle. Thus a versatile method to comparatively evaluate fuels considering their potential particle forming tendency was developed.

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INTRODUCTION

The increasingly strict emission regulations worldwide ensure the proper integration of personal mobility into an environmentally conscious age. Aside from the various technical advancements of the modern DI engine and exhaust aftertreatment (e.g. the GPF), the focus was shifted towards the fuel quality itself, especially since the introduction of “real driving emissions” [1]. The motion to apply local fuel qualities in the emission tests necessitates the evaluation of influences by fuel aspects in order to adapt and improve the engine and fuel landscape efficiently. The first approach to a correlation of fuel quality to particle emission by Aikawa et al. in 2010 [2] provided a facile method, which was based on the double bonding equivalents DBE, the vapor pressure $V.P.$ (443K) and mass fraction Wt of single components, analyzed via gas chromatography-mass spectrometry (GC-MS). The index was developed on a vehicle test bed with the new European driving cycle (NEDC) and a steady-state cycle at 120 km/h. The index was verified for different Japanese market fuels,

custom gasolines and a EU5 reference fuel, which provided a linear correlation between the index and the measured particle emissions. In 2013, Leach, Stone and Richardson [3, 4] published a PN-index based on the dry vapor pressure equivalent of the fuel mixture $DVPE$, the volumetric fraction V of paraffins, olefins, aromatics and ethanol multiplied by a weighting factor. Thus, the model can be interpreted as a modification of the Honda index from 2010, which also presents a linear correlation between the calculated index based on a reduced analytical effort. The model presented by Moriya [5] from 2016 provides a reduced approach to the calculation of the particle number via two distillation range parameters, $E130$ and $E170$, which describe the volumetric fraction of evaporated fuel at $T = 130$ and 170 °C respectively. Additionally, Moriya presented a more simplified version with just one variable from the distillation range at $T = 150$ °C, which resulted from a weighted combination of the initial two-parameter equation (see Table 2). Two similar approaches were published by Chapman et al. [6, 7], which relied on the $E170$ value

and the carbon to hydrogen ratio (C/H) of the fuel or the aromatic groups from the PIONA analysis to provide a correlation to the index by Aikawa et al.

Table 2. Equations for the different PN models found in literature.

Aikawa et al. 2010	$\text{PN index} = \sum_i^n \left(\frac{DBE_i + 1}{V.P.(443K)_i} \times Wt_i \right) \quad (1)$
Leach et al. 2013	$\text{PN index} = \frac{(V_{\text{Paraffins}} + V_{\text{EtOH}} + 2 \times V_{\text{Olefins}} + 5 \times V_{\text{Aromatics}})}{DVPE} \quad (2)$
Moriya 2016	$\text{PN index} = -0.0647 \times E170 - 0.0324 \times E130 + 9.92405 \quad (3)$
Moriya (E150) 2016	$\text{PN index} = -0.0757 \times E150 + 7.8511 \quad (4)$

DEVELOPMENT AND VERIFICATION OF THE MW INDEX

Theory and Calculation

The Menger/Wittmann (MW) index relies on fourteen physicochemical parameters, which are analyzable via standardized methods (Table 1).

Table 1. Physico-chemical parameters for the evaluation of fuel particle formation tendency.

Parameter	Unit	Analytical method
Density (15 °C)	kg m ⁻³	ASTM D 4052 / EN ISO 12185
DVPE (37.8 °C)	kPa	ASTM D 5191 / EN 13016/1
AVP (100 °C)	kPa	EN 13016/2
10% v/v evap.	°C	ASTM D 86 / EN ISO 3405
90% v/v evap.	°C	ASTM D 86 / EN ISO 3405
95% v/v evap.	°C	ASTM D 86 / EN ISO 3405
Final boiling point	°C	ASTM D 86 / EN ISO 3405
Total aromatics	% v/v	ASTM D 6839 / EN ISO 22854
Total olefines	% v/v	ASTM D 6839 / EN ISO 22854
Aromatics C10	% v/v	ASTM D 6839 / EN ISO 22854
Aromatics >C10	% v/v	ASTM D 6839 / EN ISO 22854
Naphthenes C10	% v/v	ASTM D 6839 / EN ISO 22854
Paraffins >C11	% v/v	ASTM D 6839 / EN ISO 22854
High boilings (α-Methyl-Naphthalene to n-C26)	% v/v	VDA 265

The parameters were chosen after an initial consideration of 200 parameters on 358 fuel samples across Europe. After sorting (ascending) the resulting indices, the trend was cross-examined in every single parameter (Fig. 1). Most of the parameters proved to have no significant influence on the overall trend of the total index and were therefore omitted from the final model.

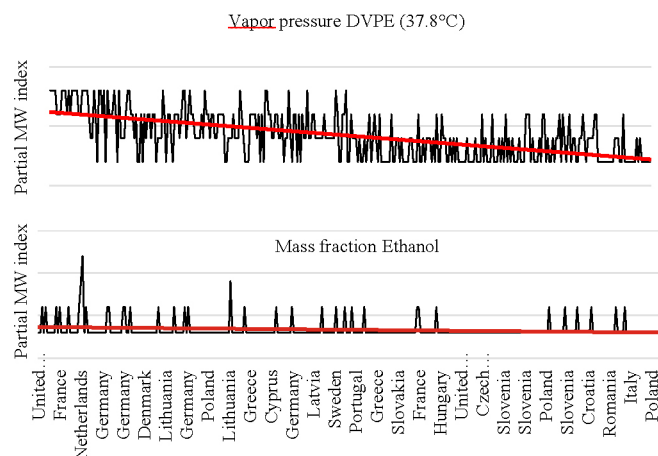


Figure 1. Choice of parameters based on the influence on the total index of the fuel. The vapor pressure DVPE decreases from the sample of highest overall index to lowest, whereas the mass fraction of ethanol does not exhibit a specific trend (red line).

Considering the database of reference fuels analyzed for this work, which covered over 770 fuel samples from Europe, China and the USA, a specific range from the minimum to maximum value is found for each parameter over the whole dataset. Each fuel is evaluated based upon where its value is positioned with respect to the range given by all the other fuel samples. This approach results in a realistic consideration of the field values. Thus, the range is split in 12.5 % intervals, which are assigned with a range factor *Ra* from -3.5 to 3.5 (density, DVPE, AVP, boiling curve, total aromatics and total olefins). This categorization is based on the theorem, that for example a higher vapor pressure value exhibits a decreasing influence on the particle formation tendency (increasing the raw index for up to 3.5). This assumption is based on an enhanced evaporation of the fuel spray, which even under low temperatures leads to an improved combustion, even including an increased conversion of components with a high boiling point. Specific parameters are only deemed to increase the particle formation tendencies of a fuel and therefore have an index range from -1 to -8 (aromatics C10, aromatics >C10, naphthenes C10, paraffins >C11, high boilings). Summing the factors over all parameters yields a final value for the fuel, which is either at the negative end (high tendency to form particulate emissions) or positive end of the spectrum (low tendency).

$$\text{MW index} = \sum_{i=1}^{14} Ra \cdot Ge \quad (1)$$

The weighting factor *Ge* can either be -1 or +1 depending on the fact if a large value (*Ge* = -1) or low value (*Ge* = +1) of the respective parameter results in a high tendency of particle formation (Fig. 2). The final MW index for the fuel ranges between -71.5 and 26.5, which can be reversed and rescaled with the appropriate reference fuels to yield a scale from zero to one (low to high particulate formation tendency) for direct comparison with other indices.

$$MW \text{ index} = \sum_{i=1}^{14} Ra_i \cdot Ge_i$$

Parameter i	Unit	Range		Weighting									
		min	max	-3.5	-2.5	-1.5	-0.5	0.5	1.5	2.5	3.5		
Vapour pressure DVPE (37.8°C)	kPa	36.4	107.7	45.3	54.2	63.1	72.1	81.0	89.9	98.8	107.7	1	
Density	kg/m³	690.8	782.2	702.2	713.7	725.1	736.5	747.9	759.4	770.8	782.2	-1	
Aromatics	%v/v	0.5	51.3	6.9	13.2	19.6	25.9	32.3	38.6	45.0	51.3	-1	
Olefines	%v/v	0.3	37.2	4.9	9.5	14.1	18.8	23.4	28.0	32.6	37.2	-1	
10%v/v evap	°C	32.2	73.8	37.4	42.6	47.8	53.0	58.2	63.4	68.6	73.8	-1	
90%v/v evap	°C	110.1	186.7	119.7	129.3	138.8	148.4	158.0	167.6	177.1	186.7	-1	
95%v/v evap	°C	115.2	197.3	125.5	135.7	146.0	156.3	166.5	176.8	187.0	197.3	-1	
Final boiling point	°C	142.0	225.1	152.4	162.8	173.2	183.6	193.9	204.3	214.7	225.1	-1	
Vapour pressure AVP (100°C)	kPa	168.4	583.6	220.3	272.2	324.1	376.0	427.9	479.8	531.7	583.6	1	
		Range		1	2	3	4	5	6	7	8		
Aromatics C10	%v/v	0	11.0	1.4	2.7	4.1	5.5	6.9	8.2	9.6	11.0	-1	
Aromatics >C10	%v/v	0	2.9	0.4	0.7	1.1	1.4	1.8	2.1	2.5	2.9	-1	
Naphthenes C10	%v/v	0	1.5	0.2	0.4	0.6	0.8	0.9	1.1	1.3	1.5	-1	
Paraffins >C11	%v/v	0	9.2	1.2	2.3	3.5	4.6	5.8	6.9	8.1	9.2	-1	
High boilings	%m/m	0	1.7	0.2	0.4	0.6	0.9	1.1	1.3	1.5	1.7	-1	

Figure 2. Exemplary matrix of parameters and the respective range distribution dependent on the database of fuel samples. Expanded version available in the [Appendix](#) section (Figure A1).

The parameter matrix and the incorporated analytical values are often found to be dependent on each other. For example a high fraction of large aromatics is likely to result in an increased final boiling point (FBP), which can be compared with FBP of a higher density fuel. Thus, the critical parameters are emphasized on via physico-chemical interactions without applying purely mathematical weighting factors. The general idea behind all of the chosen variables of the index is the effectiveness of combustion. Large, branched, structured hydrocarbon components or aromatic systems are known to combust incompletely. The incomplete radical combustion process leads to large recombination products (polyaromatic hydrocarbons (PAH)). Even in otherwise state-of-the-art engine setups and under perfect conditions (temperature, compression ratio, stoichiometric composition etc.), these precursors (PAH) lead to the formation of initial nuclei, which can coagulate and coalesce to larger particles, measurable in the exhaust stream [8, 9, 10, 11].

Particle Number Measurements of Various Fuels

The correlation of the MW index to PN data is based on measurements on an engine test bed with a two-liter BMW four-cylinder direct injection engine (Tab. 2) under laboratory conditions in a RDE [1] conform driving cycle (Fig. 3).

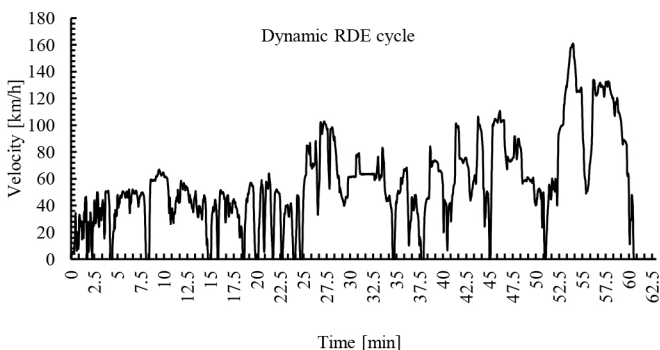


Figure 3. Dynamic RDE cycle for PN emission tests applied in this work.

The test cycle was repeated until a stable particle number emission was measured. After this equilibrium was achieved, five to seven PN measurements were recorded per fuel. The particle emission was analyzed with a Horiba MEXA 2100 SPCS condensation particle counter and the chemical analysis of the fuels was provided by an external accredited analytical institute [12] according to standardized techniques (see Table 1). The range of fuels covers low emission, purely paraffinic gasoline (PPG), premium fuels available on the market (RON102E0), Euro5 and Euro6 reference gasoline, custom EU worst-case (EU-WC) fuel adapted from survey data and several critical specifications systematically focusing on high aromatics components found mainly in Russian and Chinese gasoline. The choice of fuels for the supporting database lead to a coverage of a large range for each parameter as pointed out in Fig. 2 and Tab. A1.

Table 2. Test engine properties.

Engine property	Unit	Test engine
Cylinder configuration		In-line four-cylinder
Aspiration/fuel injection		Turbocharged / direct injection
Nr. of valves / cylinder		4 (Valvetronic, VVT)
Injector system		Bosch multi-hole nozzle
Bore	mm	82
Stroke	mm	94.6
Displacement	cm ³	1998
Injection pressure	bar	250

RESULTS AND DISCUSSION

In Fig. 4, the results of the PN measurements and the MW index calculated from the fourteen analyzed parameters exhibit an exponential correlation. The exponential growth of PN emissions was expected, due to the cumulative nature of the parameters, where an increase in one parameter can have a direct effect on another, thus emphasizing on the critical values deemed responsible for the particle formation at combustion.

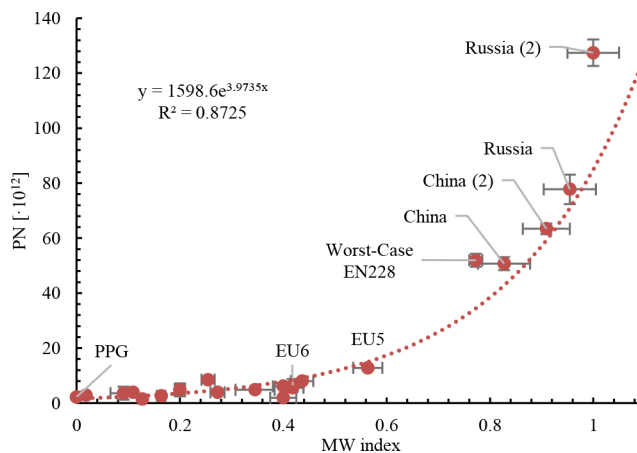


Figure 4. Measured particle number plotted against the MW index calculated from physico-chemical parameters for several market, reference and customized fuels. Error bars stem from standard deviations of multiple measurements. Only specific data labels are shown for visual clarity. The index (2) denotes a different fuel batch, which was analyzed separately.