Fuel Property Impact on Soot Emissions in SI-Engines: A Comprehensive Analysis of Gasoline Blends in RDE-Testing

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Abstract

In this study, the influence of fuel properties on the soot emissions of a spark ignition engine with direct injection was investigated. Eighteen fuel blends, including certification and acrylate fuel, as well as blends with different oxygenate and aromatic content and different boiling curves were compiled for this purpose. RDE tests were carried out on the highly dynamic test bench with cold and warm engine and the PN and PM emissions were measured in the raw exhaust gas using a particle spectrometer. The aim of the study is to analyze the relationship between known comparative indices such as the Yield Sooting Index (YSI), selected fuel properties and soot emissions in high-transient engine operation. For each fuel, 5 cycles were carried out with the engine cold and 5 cycles with the engine at operating temperature and the results were averaged to compensate for the scatter. The soot emission was then analyzed with regard to fuel properties and composition.

The experiments show that both the number of particles and the mass tendentially correlate with the comparative indices. While the addition of oxygenates such as ethanol and methanol to the reference fuel leads to a lower YSI, this can only be transferred to the test bench tests to a limited extent. This suggests that the influence of vaporization properties of a fuel, which are of great importance for mixture formation and therefore soot formation, are not sufficiently considered if only common indices are used for fuel characterization.

Notation

d	*Nozzle exit diameter.*
D_{10}	*Arithmetic mean diameter of droplets.*
DF	Density Factor
D_P	Particle Diameter in nm
DVPE	*Dry Vapor Pressure Equivalent*
FBP	*Final Boiling Point*
m_p	*Particle Mass*
PM	*Particulate Matter*
PM2.5	*Particulate Matter with diameter < 2.5 µm*
PN	*Particle Number*
RON	*Research Octane Number*
YSI	*Yield Sooting Index.*

1. Introduction

While electric drives are becoming increasingly important for newly registered vehicles in Germany, gasoline engines are still the most common drive type for passenger cars at over 62% [1]. The focus on reducing CO_2 emissions while simultaneously increasing performance led to the introduction of turbocharged engines with direct injection. While these engines have advantages over units with intake manifold injection in terms of fuel consumption, the internal mixture formation leads to an increased formation of harmful particles. The mass of particulate matter (PM) emitted has therefore been limited by law since the introduction of the Euro 5 emissions standard for gasoline engines with direct injection [2,3]. Although the amount of particulate matter emitted has been decreasing since the introduction of particle

filters, around 21% of PM2.5 near roads can be attributed to engine exhaust fumes [4]. As fine particles are considered to have a higher hazard potential, limit values for the number of particles in spark-ignition engines were introduced for the first time with the introduction of Euro 6 [5]. A further reduction of the particle size to be taken into account to 10 nm is being planned [6]. As particle filtration in the exhaust gas is always accompanied by more or less increased fuel consumption, the optimization of fuels to reduce the formation of particles is increasingly becoming the focus of development. Reduced raw emissions can minimize the increase in exhaust back pressure when the particulate filter is loaded as well as the frequency of regeneration.

1.1 Fundamentals of Particles (Characterization & Formation Mechanism)

Particles can be divided into volatile and non-volatile fractions. Sulphates, nitrates and organic compounds form the volatile particles, carbonaceous compounds and ash the non-volatile particles, whereby the carbonaceous fraction is generally referred to as soot. Due to the complex formation mechanisms, the composition depends on numerous influencing factors such as the combustion process, operating point, oil and fuel composition as well as the ambient conditions. [7] As ash is an incombustible component, a reduction can be achieved by avoiding such components in the fuel. The formation of soot, on the other hand, follows a complex formation path, so that these particles cannot be traced back directly to individual components, but instead require consideration of the fuel as a whole.

During the combustion of hydrocarbons, acetylene and other smaller hydrocarbon chains are formed through pyrolysis. Due to their structure, these species can combine to form aromatics. By combining with other aromatics or alkyls, larger, polyaromatic hydrocarbons (PAHs) are formed, which can form larger soot particles through agglomeration [8]. Due to the complexity of the formation mechanism, the formation of particles during the combustion of hydrocarbons cannot be completely avoided, but the formation of soot precursors can be reduced by selecting a suitable fuel.

The following fuel properties can be derived from this to reduce soot formation:
- Optimization of mixture formation through favorable evaporation behavior
- Avoiding the formation of soot precursors at the molecular level:
  - Avoidance of double bonds and ring structures

The influence of fuel on soot formation has been a subject of scientific interest for years. Aikawa et al. translated the observation that early-boiling fuel components as well as those with a low double bond equivalent (DBE) have a low soot tendency into a PM index and were able to show that this correlates with both PN and PM emissions [9]. In contrast, Dageförde et al. compared toluene with RON 95 and showed that the soot emission of the pure aromatic is above or below that of the reference fuel depending on the operating point. This is attributed to differences in mixture formation due to the lower calorific value and the associated longer injection times [10]. Although aromatics generally have the potential to form soot precursors due to their ring structure, the thesis is supported that only late-boiling aromatics favor an increase in PN emission. [10,11].

Leach et al. [12] developed a PN index which, in contrast to the PM index, uses the Dry Vapor Pressure Equivalent (DVPE) of the fuel for the calculation. Thus, a calculation from a standardized fuel analysis is possible. It was shown that the PN index correlates with the PN emission of a test engine with direct injection.

Ratcliff et al. [13] investigated the interaction between aromatics and ethanol in fuel. They were able to show that adding ethanol to the fuel can reduce PM formation by diluting and thus reducing the aromatics content. At the same time, the evaporation enthalpy of the alcohol can slow down the evaporation of low-boiling aromatics and thus increase their concentration in the last incompletely evaporated parts of the fuel jet. An admixture of alcohol can therefore have the opposite effect on soot formation than the oxygen content in the fuel would suggest. Similar effects were also described by Vuk [11] and Burke [14].

2. Experimental Setup

2.1 Engine and test bench setup

The engine tests were carried out on a turbocharged four-cylinder unit with direct injection. The exhaust gas aftertreatment and the control unit's configuration correspond to the standard Euro 6 specification. Ancillary units were also adopted from the standard version, as was the entire intake and exhaust gas section up to the catalytic converter and particulate filter. In order to ensure realistic heating behavior, additional oil conditioning was dispensed with and the coolant circuit was limited to a volume close to series production. The technical data of the unit is summarized in **Table 1**.

The number and size distribution of particles was recorded between the turbocharger and the gasoline particle filter using a DMS500 particle spectrometer from the manufacturer Cambustion. An external cooling water conditioning unit, cell ventilation and an additional air circulation fan enable the engine and cell to be conditioned to 20 °C between tests.

As there is no direct measurement of the particle mass, this is calculated from the particle number concentration and the mobility diameter. The calculation is carried out analogously to the manufacturer's specifications [15] according to **Equation 1**, assuming a spherical geometry for GDI engines. A power factor (PF) of 3 and a density factor (DF) of 5.2×10^{-16} are therefore recommended.

$$m_p(\mu g) = DF \; x \; D_p^{\,PF}$$

Equation 1: Calculation of the particle mass

Table 1: Technical data of the test engine

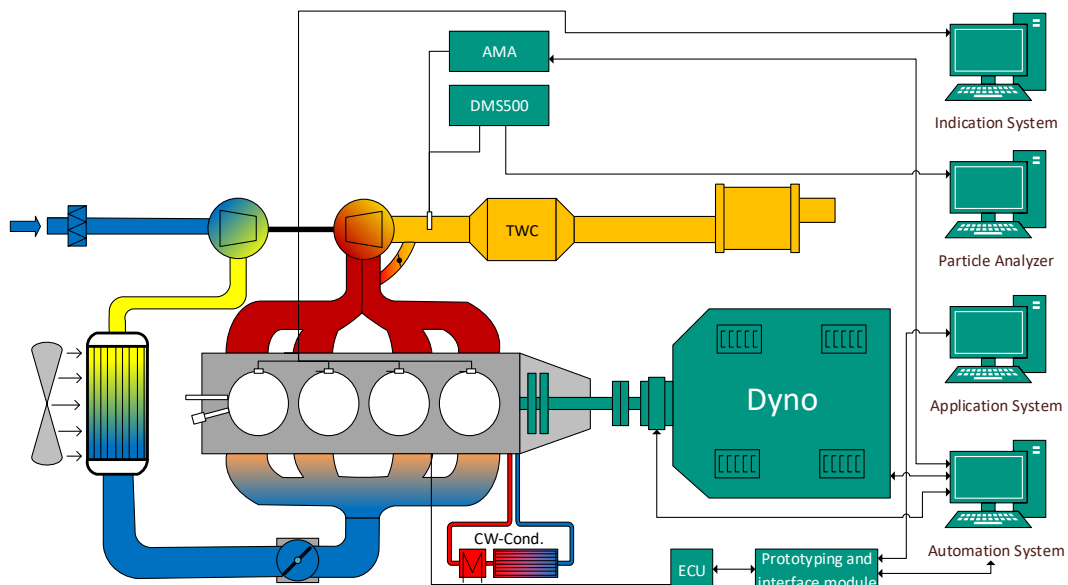| Number of cylinders	4	-
Effective capacity	1998	cm³
Bore	82.0	mm
Stroke	94.6	mm
Compression ratio	9.5 : 1	-
Number of valves	4	-

Figure 1: Test bench setup

2.2 Test Procedure

The test cycle used was derived from real driving data of a compact SUV with the same engine. Three sections of equal length were combined in the order of urban, interurban and highway driving. As the engine is preconditioned to 20 °C before each test, the start with a cold engine, catalytic converter heating phase and warm-up behavior are also taken into account. The test cycle is shown in **Figure 2**. Each test run was repeated 5 times under constant conditions and the average PN and PM emissions were calculated from the test runs with the same fuel. As there is no gravimetric measurement of the particle mass, this was calculated on the basis of the size distribution.
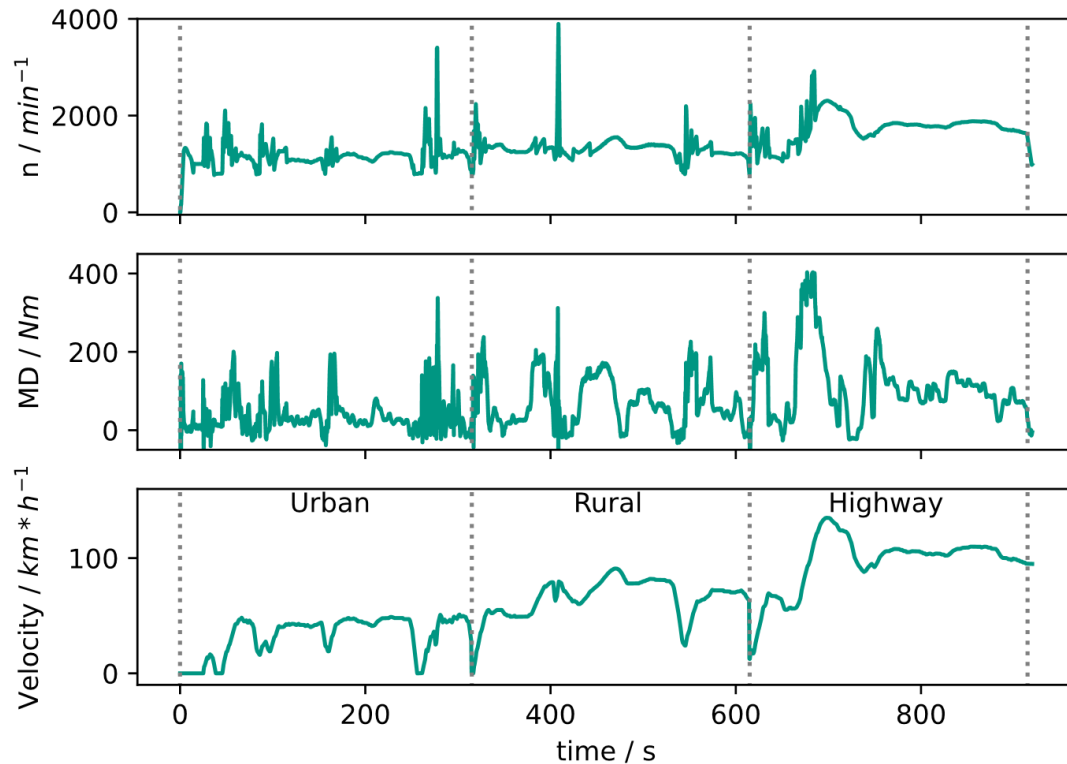

Figure 2: Test cycle sequence and segmentation

The measurement of the YSI was carried out by OWI Science for Fuels gGmbH. A steady flame co-flow burner according to Das et al. [16] was used. The unified YSI and the maximum soot volume fraction were determined for all fuels based on color pyrometry using the "peak-region" method described by Das et al. [17], where the mean soot volume fraction is determined by averaging over the sootiest parts of a doped methane flame. The robustness of this method proven by Montgomery et. al for various fuels and air to fuel ratios [18].

2.3 Tested Fuels

For the tests, 18 different fuels were selected to investigate the influence of the composition on soot emissions in the burner and engine tests. The aim is to vary the fuel properties relevant to soot formation as widely as possible. On the one hand, this includes a variation of the boiling behavior through different concentrations of low- and low-boiling components as well as a targeted adjustment of the chemical composition. For example, the proportion of aromatics overall, oxygenates and olefins was specifically changed. Since the number of carbon atoms within the aromatics group also has an influence on the soot tendency, the composition of the aromatics was also varied. A paraffinic base fuel (base low) with a low aromatics and olefin content was selected as the lower reference. The upper reference is a second base fuel (base high), which in contrast has a high content of aromatics greater than C7, olefins and a late end of boiling point. Based on these reference fuels, blends were produced with the alcohols methanol and ethanol as well as the aromatic toluene. In order to investigate the influence of boiling behavior and composition of the aromatic fraction, 5 additional fuels with variable boiling end and aromatic size, but comparable total aromatic, olefin and alcohol content were added. In addition, two regenerative fuel

blends with 33 % and 85 % regenerative content and a Euro 5 certification fuel were investigated. The key fuel parameters are shown in **Table 2**.

Table 2: Properties of tested fuels

No.	Name	T10	T50	T90	FBP	Alcohols	Aromatics	Aromatics >C7	Olefins	Oxygen
-	-	°C	°C	°C	°C	vol.-%	vol.-%	vol.-%	vol.-%	m.-%
1	Base High	62.3	101.7	174.4	230.5	0.0	37.4	28.7	21.0	0.03
2	Base Low	55.9	82.5	111.9	182.9	0.3	0.7	0.7	0.3	2.51
3	E10 High	56.2	90.0	171.2	223.1	10.0	33.7	22.3	19.1	3.64
4	E20 High	57.9	71.9	168.6	216.4	19.7	29.9	19.8	17.1	7.12
5	E30 High	58.4	72.9	165.0	215.6	29.4	26.2	17.3	15.2	10.55
6	M5 High	47.7	103.9	172.2	223.1	5.2	34.6	30.9	18.5	2.71
7	M15 High	49.6	86.9	171.7	226.2	14.8	33.2	29.6	15.8	7.69
8	T5 High	64.0	102.7	172.5	225.3	0.0	40.1	23.7	20.3	0.03
9	T10 High	65.3	103.6	171.2	220.5	0.0	43.5	22.3	19.2	0.04
10	T10 Low	59.6	87.0	111.1	183.5	0.3	8.3	0.6	0.3	2.27
11	FBP Var. 1	55.0	101.1	174.1	209.0	4.4	32.8	24.7	10.5	1.59
12	FBP Var. 2	55.6	104.6	166.3	208.9	4.3	36.9	28.7	10.2	1.57
13	FBP Var. 3	56.2	105.9	174.8	209.9	4.9	33.5	9.9	10.8	1.79
14	FBP Var. 4	43.4	80.1	146.2	165.4	5.0	31.7	18.0	10.8	1.86
15	FBP Var. 5	42.2	77.5	132.5	175.2	5.0	32.3	11.3	11.0	1.87
16	G33	56.7	97.4	153.7	184.0	8.5	24.6	17.4	7.1	3.80
17	G85	54.1	101.1	178.3	196.9	0.0	34.6	19.1	5.8	2.78
18	EU5	66.7	105.6	145.3	165.3	5.0	32.4	19.3	1.3	1.82

3. Results

3.1 RDE-Results

The ambient and component temperature has a significant influence on particle formation, so that the particle concentration in the exhaust gas is significantly increased immediately after starting the engine [19,20]. **Figure 3:** Influence of oxygenate and toluene addition on cold start emissions shows the cumulative average PM emissions of various blends in comparison with their base fuels. All the investigated blends based on base-high lead to a significant reduction in soot formation compared to the base fuel. In the case of ethanol, an admixture of up to 20 percent leads to a 55% reduction in the cumulative particulate mass. A further increase in the ethanol content to 30%, however, leads to a flattening of the effect. The effect is more pronounced with methanol. While 5% methanol reduces PM emissions by about the same amount as adding 10% ethanol, a further increase in the methanol content leads to a further reduction in particle mass, but the effect also flattens out. Here, the mixture formation can be negatively influenced both by the lower density of the oxygenates and by an increased enthalpy of vaporization, so that a reduced soot tendency on a molecular basis of the blends is partially compensated. Toluene as a C7 aromatic, shows a comparatively high soot tendency in burner tests. As a result, in contrast to the alcohols, the calculated YSI of the toluene blends is above the base-high fuel. Nevertheless, a reduction in particulate mass is observed for both toluene blends when the engine is started. The T10-Low, on the other hand, shows a slight increase in particulate emissions compared to its base fuel. From this it can be concluded that the effect of small aromatics has a minor influence on a low-soot, aromatics-free fuel. In the case of the high-boiling aromatics base high fuel, on the other hand, dilution with smaller aromatics appears to be advantageous.

The results of the overall cycle are shown in **Figure 4**. It can be seen that the results of the start phase can differ significantly from a dynamic driving scenario depending on the fuel. While the same trends can be seen with toluene as with the starting phase, the effect achieved by adding alcohols is reduced and, in the case of methanol, can even lead to an increase in particle emissions. Since methanol has a low boiling point and at the same time a high enthalpy of vaporization, this can impair the vaporization of the late-boiling components. This phenomenon, already described by Ratcliff [13], appears to be further intensified in the case of the base high fuel, which is rich in late-boiling aromatics. A comparison with ethanol, on the other hand, shows that the enthalpy of vaporization alone does not adequately explain the effect. Although the mass-related enthalpy of vaporization of methanol is around 33% higher than that of ethanol, in the case of E10 this is compensated for by the double mass fraction compared to M5. In the case of ethanol, there is an increase in particle mass over the cycle of 3 %, which can possibly be attributed to increased enthalpy. However, the negative effect is limited exclusively to phases of higher load at the end of the cycle. In the urban and rural segment, the PM is reduced by 14 % and 2 %. With an increase to 20 or 30 % the effect of dilution appears to be dominant, so that there is again a decrease in particle output, even at higher loads. Overall, it can be seen for all oxygenate fuels that a significant reduction in particles can be achieved, particularly in the start phase and in the first 5 minutes of the cycle. The normalized, cumulative emissions are thus approximately at the level of the cold start. (48% for E30, 61% for E20 and 86% for E10). The differences decrease over the course of the cycle. On the one hand, this can be favored by the rising engine temperature and the associated better mixture formation for all fuels; on the other hand, errors in mixture formation, for example due to changes in the density of the fuel, are more significant at higher loads and due to longer injection times.
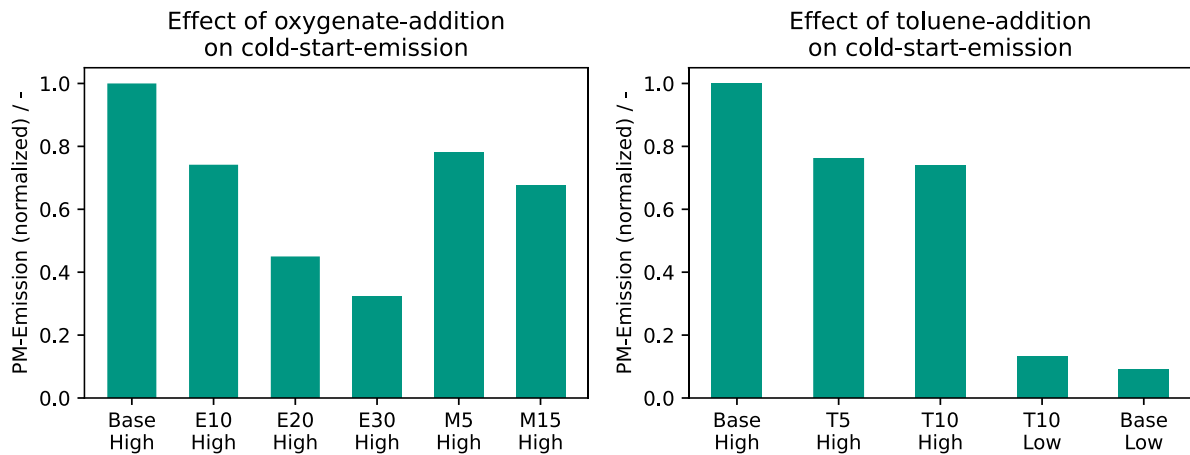
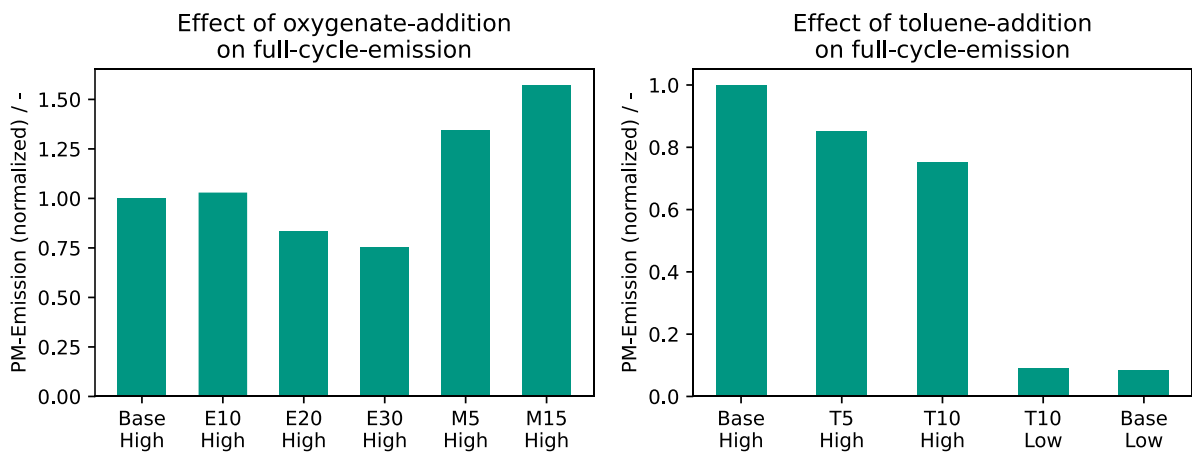**Figure 3:** Influence of oxygenate and toluene addition on cold start emissions

Figure 4: Influence of oxygenate and toluene addition on full cycle emission

Due to the lack of a carbon-carbon bond, methanol as a pure substance has hardly any potential to form soot, which is also reflected in a low YSI of 6.6 [17]. The molecular structure alone cannot therefore explain why the particle mass increases with increasing methanol content in the cycle. While a 32 % reduction in particle mass is achieved with M15 in the cold start (the measured YSI is 28 % lower than with base high), the particle mass emitted over the entire cycle is 57 % higher than with the reference fuel. Analysis of the data over time shows that these differences can only be attributed to a few cycle segments in the 50 to 75 % engine load range. Here, the particulate mass concentration is briefly 2 to 3 times higher than with the reference fuel. Due to the simultaneously occurring high exhaust gas mass flows, these segments are strongly taken into account in the overall result of the cycle. The cause of the short peaks cannot be identified with the measurement technology used, but optical recordings are planned to investigate possible anomalies.
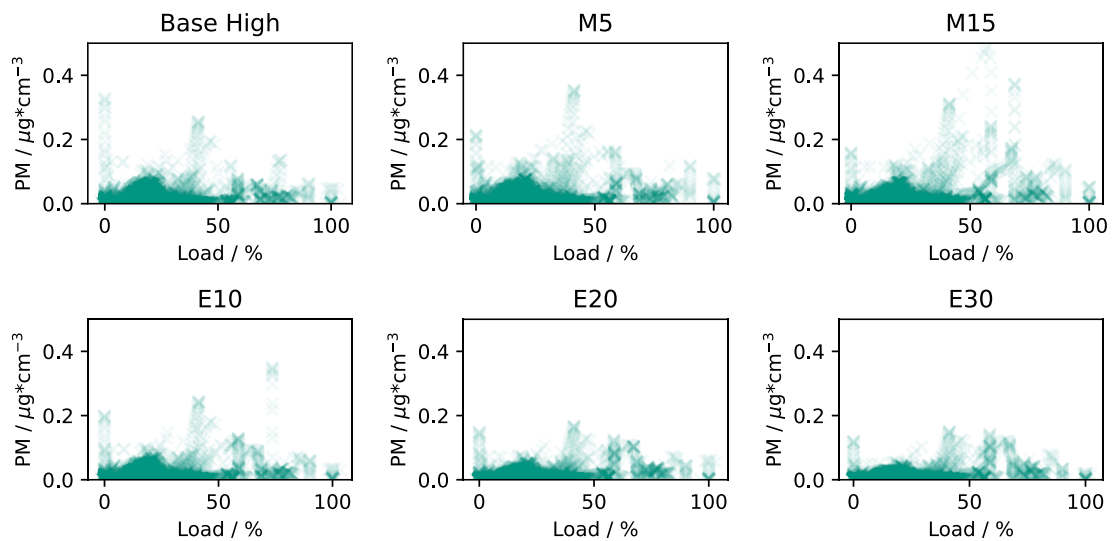
Figure 5: Distribution of PM concentrations by engine load for oxygenated fuels

The results from the YSI determination on the burner were compared with the measured particulate emissions below. For this purpose, the PN and PM emissions were cumulated for each segment of the test cycle and normalized to the respective result of the base-high fuel by dividing the cumulated PN or PM emission of the fuel by the cumulated result of the base high fuel. As the two methanol blends M5 and M15 lie outside the results of the other fuels across all segments, they are considered outliers and are not included in the calculation of the coefficient of determination. A linear relationship between the measured YSI and the PM emission can already be seen at engine start with subsequent idling phase in **Figure 6**. Although this decreases further as the cycle progresses, the correlation in the end-of-cycle results remains at a comparable level as the particulate emissions continue to decrease overall as the engine reaches operating temperature.

The PN10 emission, on the other hand, shows a reduced dispersion compared to the PM emission and therefore a stronger correlation in the urban to highway sections and therefore also in the end-of-cycle emissions. At engine start, on the other hand, there is no recognizable correlation between the two variables. However, this is due to the high number of particles in the ultrafine particle range. Here, the measurement stop of the strain gauge is reached shortly, so that no valid results can be obtained. As the particle mass is not measured directly, but is calculated from the particle size distribution, there are different effects at engine start and highway section. In principle, the particle mass is significantly influenced by the occurrence of larger particles. As the size ranges relevant for the particle mass are still within the measuring range when the engine starts, a comparison with the YSI is still possible here in contrast to the PN10 emission. In the further course, however, the particle number concentration in the exhaust gas tends to decrease. The signal to noise ratio therefore deteriorates for the already rare accumulation and coarse mode particles. At the same time, other factors such as the engine oil can influence the particle mass, particularly at high loads and with a warm engine. A clear separation is not possible based on the data.

Nevertheless, across all phases of the cycle, it is evident that the YSI is suitable for evaluating a fuel's soot generation. A linear relationship is recognizable for both the particle mass and the number of particles across all cycle phases.
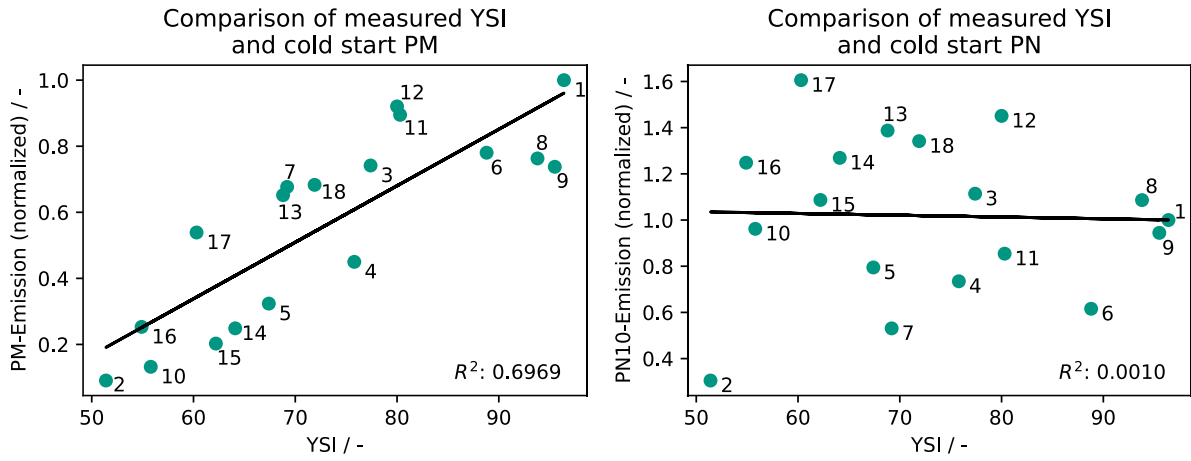
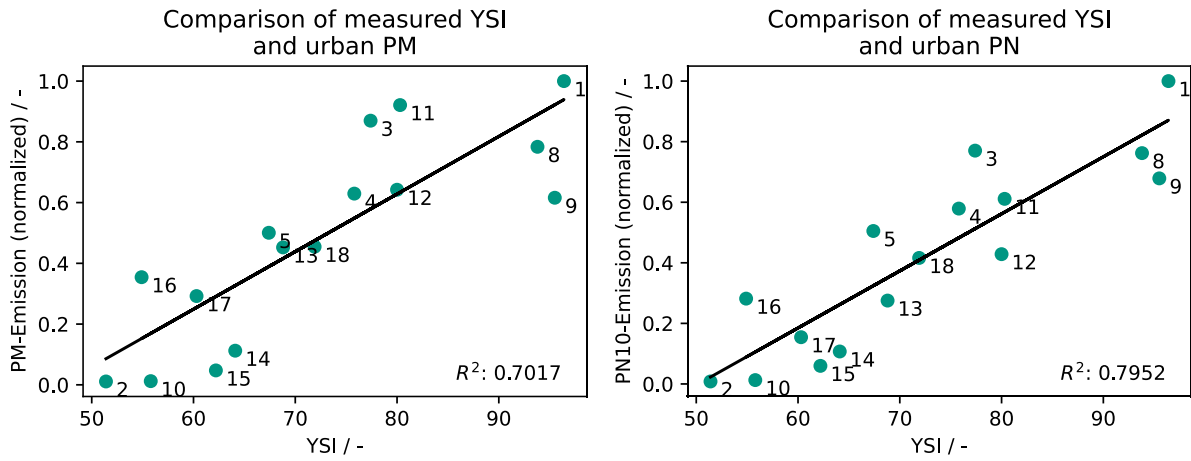**Figure 6:** Correlation between cold-start PM-Emission and measured YSI

Figure 7: Correlation between urban PM-Emission and measured YSI
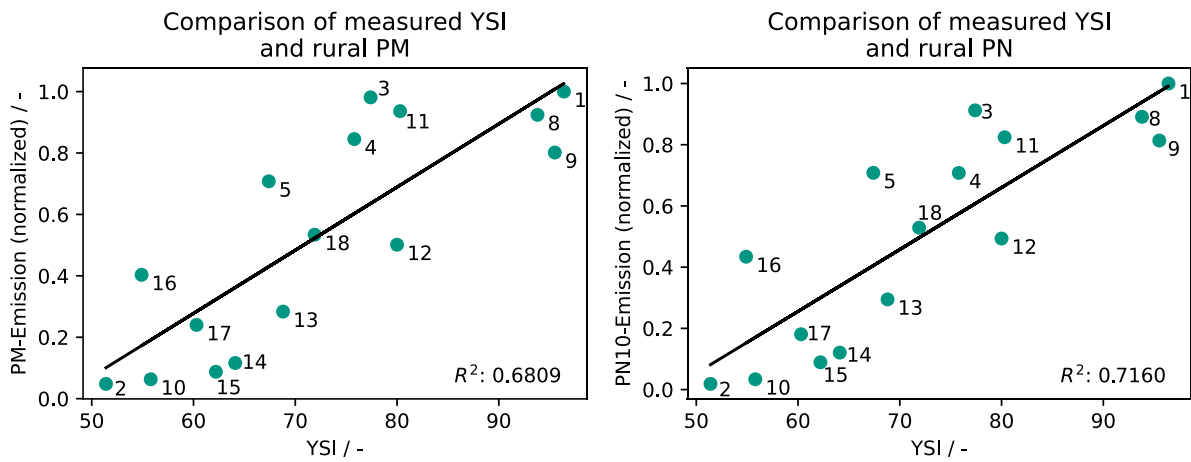

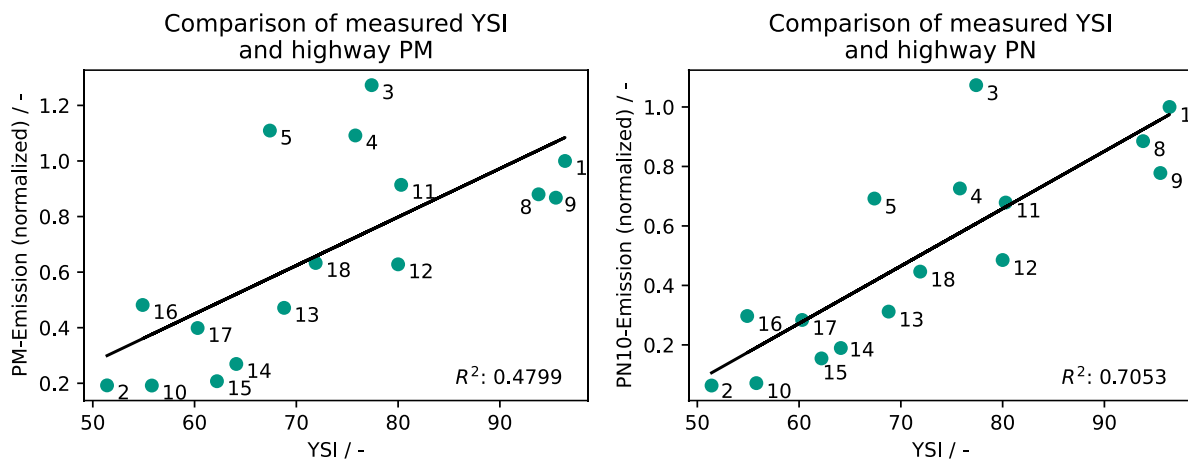**Figure 8:** Correlation between rural PM-Emission and measured YSI

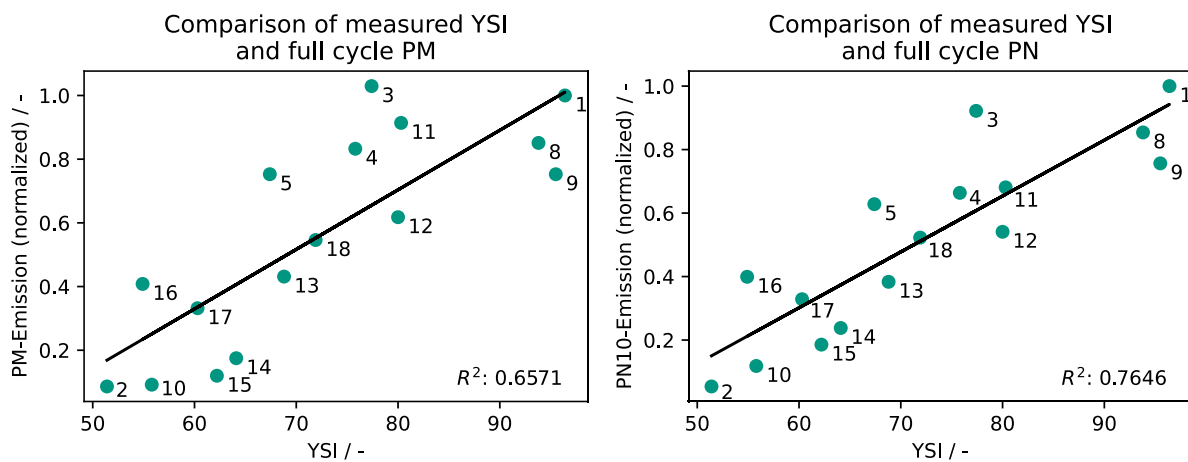**Figure 9:** Correlation between highway PM-Emission and measured YSI

Figure 10: Correlation between full cycle PM-Emission and measured YSI

4. Conclusion

18 fuels were selected and examined on the basis of their properties. A fuel analysis was carried out for each fuel and the yield sooting index was determined on a laminar burner. The fuels were tested on the engine test bench with a production unit and the effects on PM and PN emissions in the raw exhaust gas were determined. It was shown that the YSI is suitable for a first suggestion of both the number of particles and the mass. The fuel with the lowest YSI compared to the base high fuel shows a 95% reduction in particle count and a 91% reduction in particle mass over the entire cycle. This shows that the selection of a suitable fuel also has a high potential for reducing pollutants in the existing fleet. While the YSI is by and large suitable for selecting such a fuel, there are nevertheless differences with regard to various fuels of comparable YSIs. For example, in the case of the blends investigated, an admixture of toluene leads to a higher YSI regardless of the fuel, while in the case of the base high fuel a reduction in particulate emissions is nevertheless achieved. Such opposing effects may occur in particular due to the interaction of late-boiling aromatics and early-boiling alcohols. Diluting an aromatic-rich fuel with ethanol or methanol leads to a reduction in YSI, but depending on the operating point, particle emissions can be increased. Particularly in the case of E10 and the methanol blends investigated, the PN emission are reduced when the engine is cold and under low load, but in the overall cycle the emission is higher than the base fuel, although the measured YSI indicates otherwise. This leads to the conclusion that for an evaluation of the soot potential of a fuel, further characteristic values such as the evaporation

behavior should be taken into account. Since the multifactorial nature of the test setup makes it difficult to draw conclusions about individual causes, additional investigations are planned using 2d in-cylinder two-color pyrometry and an extended fuel matrix to consider the influence of individual fuel properties in particular.

Acknowledgement

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