Comparison of the emission potential of renewable fuels in mono- and bi-fuel systems from the point of view of a car fleet in an incoming circular economy

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Abstract
The aim of the study is to investigate the most effective approach to reduce the emissions of a SI-engine while using a limited amount of renewable fuel. In this study, the renewable fuels ethanol, methanol, 2-ethoxy-2-methylpropane (ETBE), acetone, and dimethylformamide (DMF) were investigated with various fixed admixture rates and with a fully variable on-board fuel mixture (Smart-Fuel concept). One result of the study is that for a Smart-Fuel concept using methanol a reduction in CO₂ emissions of approx. 12.5% and a reduction in particulate emissions of approx. 60% can be achieved, when considering an entire car fleet. In terms of engine efficiency, as well as particulate emissions, the pure substances, except DMF, achieved significant improvements compared to standard gasoline. Compared with the pure substances, the Smart-Fuel concept achieved lower advantages; however, it used significantly less scarcely available renewable fuel in the process. Based on the limited availability of renewable fuels within the first stages of a circular economy, the Smart-Fuel concept proves to be a very efficient transition technology to achieve the CO₂ reduction targets. The Smart-Fuel concept only uses renewable fuel when it is worthwhile in terms of efficiency or emissions. Predefined fuel blends in a mono-fuel concept offer much less reduction potential in terms of emissions than the Smart-Fuel concept. However, with respect to particulate raw emissions, especially for moderate mixing rates significantly increased particle emissions are sometimes observed, despite the overall very good performance of the pure substances.

Keywords Dual fuel · e-fuel · Renewable fuel · Smart-Fuel · Circular economy · Fleet emissions · SI-engine · Ethanol · Methanol · ETBE · Acetone · Dimethylformamide

1 Introduction
Defossilisation of fuels for internal combustion engines is a necessary condition for a functioning circular economy. According to current studies [1–3], a worldwide purely electrically oriented mobility scenario would mean a significant failure of the targets set in the climate protection agreement and a depletion of the currently available raw materials. Therefore, internal combustion engines must also be operated CO₂-neutrally in the future. However, in the transition to a fully CO₂-neutral circular economy, it is not possible in the short term to substitute the entire demand for fuel with 100% renewable alternatives. The share of sustainable fuels must be steadily increased until fossil fuels are no longer needed.

This leads to the question of how the limited amount of available renewable fuel can best be used to provide the most significant overall CO₂ reduction. It is known that the engine efficiency of the gasoline engine can be significantly increased with knock resistant renewable fuels. The level of engine efficiency increase depends on the blending rate of the knock resistant renewable fuel component. The question to be considered is whether it is advantageous from a fleet perspective to generate only a low potential with respect to a single vehicle through a low blending rate for all vehicles or to realize a large specific potential for a fraction of the vehicles. This can only be assessed on the basis of the entire vehicle fleet with a
certain predetermined fuel availability. Here, a promising solution approach is to deviate from fixed fuel blends at the gas stations and to use variable on-board mixing systems by means of a multi-tank concept. One such concept is, for example, the Smart-Fuel concept \cite{4,5}, which allows a very efficient usage of the limited available knock resistant renewable fuels, using it only when it is necessary or when it offers a noticeable advantage. In addition to the peak engine efficiency, lower pollutant emissions can also be achieved. Particularly noteworthy are the particulate raw emissions, where synthetic fuels usually have a significantly lower soot tendency than an average gasoline based on fossil resources.

In this study, we compare the potential engine efficiency of an exemplarily designed Smart-Fuel concept with those of different fixed blending rates. Following this single-vehicle analysis, the impact in the entire vehicle fleet is considered under the boundary condition of limited availability of renewable fuels. Furthermore, the potential, as well as challenges, of fuel blends and especially of the Smart-Fuel concept are examined with respect to particulate raw emissions. The renewable fuels considered (Table 1) were ethanol, methanol, 2-ethoxy-2-methylpropane (ETBE), acetone, and dimethylformamide (DMF). The base fuel was RON95 E10, to which the renewable fuels were also blended volumetrically. This means that a fuel with a 25 vol% admixture then contains 25 vol% of the admixed fuel and 7.5 vol% ethanol due to the composition of the base gasoline fuel.

The experiments were carried out on an engine test bench with a specially adapted turbocharged BMW four-cylinder test engine, which is based on the BMW series engine kit. In addition, it should be mentioned that the dual injection approach used here, a combination of direct injection (DI) and port injection (MPI), would in principle also allow fuel-type stratification (inhomogeneous fuel-type distribution in the combustion chamber) in the case of a Smart-Fuel concept. For reasons of the scope of the test, a fuel-type stratification was not investigated in this first step.

The results of the Smart-Fuel concept were derived computationally from the multi-dimensional test result space, which is defined by speed, load, engine application, and boundary parameters as well as several fuel components with different fixed blending rates. Furthermore, due to the readily available efficient exhaust gas cleaning by means of a three-way catalytic converter, the investigations were limited to a purely stoichiometric combustion air ratio with largely homogeneously premixed fuel–air mixture without external exhaust gas recirculation.

### 2 Comparison between Smart-Fuel concept and fixed fuel blends

The Smart-Fuel concept was designed in such a way that the median heat release (HR50) was close to its optimal position in the entire operating field. A compression ratio of 11 was used as a basis, which is why knock resistant secondary fuel was only added at high loads. The resulting admixture requirement was significantly lower for the strongly evaporating cooling fuels (ethanol and methanol) than for ETBE and acetone, despite the significantly better chemical knock resistance of ETBE and acetone. This is due to the temperature reduction achieved by the evaporation cooling of the fuels. Because of the gasoline engine technology used, the evaporation cooling has a greater effect on the engine knock resistance than on the conventionally knock resistance value (ROZ/MOZ), which is determined in the standard test procedure.

The different variants of fixed fuel blends and the Smart-Fuel concept were evaluated for various secondary fuel types using the WLTP and an exemplary RDE test cycle. The selected RDE test cycle has significantly more dynamic accelerations and velocities of the vehicle than the WLTP. In the RDE test cycle, full-load accelerations up to over 200 km/h occur. According to the current legal requirements, this is not an officially valid test drive, but it represents a highly challenging drive. Steady-state operation maps were used to calculate the fuel consumption in the test cycles, these maps were determined on the engine test bench for warm operating conditions.

| Table 1 Fuel properties \cite{6–15} | RON95 E10 | Ethanol | Methanol | 2-Ethoxy-2-methyl propane | Propanone | N,N-Dimethylformamide |
|----------------------------------|-----------|---------|----------|--------------------------|-----------|----------------------|
| Molecular formula                | C\textsubscript{7.2}H\textsubscript{13.9}O\textsubscript{0.225} | C\textsubscript{2}H\textsubscript{6}O | CH\textsubscript{3}O | C\textsubscript{4}H\textsubscript{8}O | C\textsubscript{3}H\textsubscript{7}NO |
| Boiling point \(^\circ\text{C}\)  | 302.10    | 78.3    | 64.7     | 72.5                     | 56        | 153                  |
| Lower heating value \(\text{MJ/kg}\) | 41.1      | 26.6    | 19.5     | 35.8                     | 28.3      | 25                   |
| Stoichiometric air–fuel ratio \(\text{kg/kg}\) | 14        | 9       | 6.4      | 12.1                     | 9.5       | 8                    |
| CO\textsubscript{2} emission \(\text{g/MJ}\) | 74.2      | 71.8    | 71       | 72.4                     | 80.4      | 72.3                 |
| Enthalpy of vaporization \(\text{kJ/MJ}\) | 8         | 30      | 56       | 8                        | 18        | 17                   |
2.1 Engine efficiency

While using the knock resistant fuels ethanol, methanol, ETBE, and acetone in the fuel blends of 50 vol% up to 100 vol% and in the Smart-Fuel concept, the HR50 was close to the optimum position throughout the entire engine operating map. This results in consumption maps, where the best points are at the highest load. However, restricting optimization solely to full load is not a suitable engine design for a low fuel consumption passenger car, because full load is only approached during short time periods. Improvements exclusively in full-load consumption contributes very minimally to an improvement in average fuel consumption. Therefore, it is useful to increase the compression ratio to improve the part load efficiency. Engines with a variable valve train, as implemented for instance with BMW-VANOS and BMW-Valvetronic, offer further potential with regard to the fuel consumption in the lower part load range, since the residual gas tolerance rises due to the increased combustion stability and the limits of the strategy of early inlet closing (EIC) are extended. This is because the engine is de-throttled by extended EIC and with a higher residual gas content, the engine efficiency then increases, as the thermodynamic properties are improved, and the temperature level is lowered. In addition, further potentials and challenges in hardware optimization arise from the fuel properties deviating from RON95 E10.

The reduction in fuel consumption by increasing the compression ratio from 11 to 13 was determined using GT power and amounts to approximately 2% in the RDE test cycle. The knock-sensitive areas were influenced negatively at high loads, but these represents only a relatively small time portion of the test cycle. The further increase in efficiency due to the resulting higher residual gas compatibility and the extended limits of the EIC could be estimated to be about 2%, derived from the experience of existing engine development measurements. The increase in efficiency due to the possible hardware optimization of the engine (e.g., charging system, exhaust system) is assumed to be 1%. These are examples of areas, which can be optimized as a result of different fuel properties. Figure 1 shows the efficiency equivalent, which represents the resulting volumetric heating value corrected fuel consumption of the various fuel variants for the RDE test cycle. The hardware improvements described above have been accounted for in the 50 vol% mixtures, the pure substances, and the Smart-Fuel concept for the fuels ethanol, methanol, ETBE, and acetone. The measurement has been executed on the standard engine and is estimating the engine hardware optimization potentials. The other fuel blends represent directly the results, gained with a compression ratio of 11 and without any further hardware optimization.

The diagram shows the efficiency equivalent for the RDE test cycle for the different fuel variants relative to the corresponding value of RON95 E10 as 100%. The pure fuels of acetone and DMF were not part of the experimental matrix. The knock-resistant fuels ethanol, methanol, ETBE, and acetone steadily reduce the efficiency equivalent as the blending rate increases. The blending rates 25 and 50 vol% show a clear difference, since for the 50 vol% blending rate, significant hardware optimization is enabled and considered. Particularly with methanol, there is a clear advantage of pure methanol compared to the 50 vol% blend, although both cases are based on the identical HR50 phases and hardware optimization. The reason for this is the high evaporation cooling of the pure methanol. The increased evaporation cooling during compression reduces the compression work. A lowered temperature level due to the high evaporation cooling results in a more favorable thermodynamic process, which increases the efficiency. The C/H-ratio of methanol is the reason why significantly more water and thus more three-atom molecules are formed by the combustion. This significantly lowers the isentropic coefficient. This leads to a lower pressure decrease during expansion, which is why expansion work increases [15]. The DMF shows a slightly lower knock resistance than RON95 E10 but a slightly faster and more complete burning process. This, in general, does not lead to a significant difference in the engine efficiency.
compared to the base fuel RON95 E10 except for a small improvement for the 10 vol% admixture.

In contrast, the simplified Smart-Fuel concept reduces the cycle-related efficiency equivalent for the highly knock-resistant fuels ethanol, methanol, ETBE, and acetone almost to the extent, that the 50 vol% fixed blending rate can. This is due to the fact that the efficient use of the secondary fuel enables hardware optimization analogous to the usage of fixed fuel admixtures of 50 vol%. Figure 2 shows the average blending rate in the Smart-Fuel concept for the WLTP test cycle and the very dynamic RDE test cycle. It shows impressively that for a Smart-Fuel concept, the efficiency advantages, described above, are already achieved at much smaller average blending rates than compared to the fixed admixtures.

In the RDE test cycle, the fuel consumption is significantly higher than in the WLTP. This is due to the dynamic driving profile with strong accelerations and high speeds. The large difference between the fuels with high evaporation cooling, ethanol as well as methanol, and the chemically very knock-resistant fuels ETBE and acetone results from their higher effectiveness in the reduction of the knocking tendency due to the evaporation cooling. In both cases, however, the average admixture rate is significantly less than 50 vol%, while providing a comparable improvement in efficiency. The reason for this is that in the Smart-Fuel concept, the knock-resistant secondary fuel is only used when necessary, but in a fixed fuel blending, the secondary fuel is used permanently. In addition, the Smart-Fuel concept enables a higher compression ratio than would be possible with a fixed blending with the equivalent average blending rate. This is because critical boundary conditions with respect to the limitation of the compression ratio, such as a high load or high ambient temperatures, can be specifically transferred to non-critical operation with a map-specific high porting rate of the secondary fuel.

### 2.2 Gaseous emissions

With regard to the gaseous emissions downstream of the catalytic converter, hardly any differences could be observed with both the fixed fuel blends and the simplified Smart-Fuel concept without fuel-type stratification for the homogeneously premixed case. This is because a warm three-way catalyst in the optimal conversion range reduces emissions to a very low level. Upstream of the catalyst, however, there were significant differences in gaseous raw emissions depending upon the fuel properties. The catalyst heating revealed minor differences, which could be compensated by adjusting the engine calibration, so there was even in this phase no significant potential for improvement of gaseous emissions for the simplified homogeneous premixed case without fuel-type stratification. The nitrogen-containing fuel DMF behaved differently, because in addition to the Zeldovich mechanism, the fuel-NO is gaining importance. With the increased nitrogen content, the fuel-NO dominates for operating points with a moderate combustion temperature, because the fuel-NO mechanism is significantly less temperature dependent than the Zeldovich mechanism. At part load, this means an increase in NOx raw emissions. However, these increased nitrogen raw emissions are converted without any loss due to the warm three-way catalyst. Also, during the catalyst warm-up phase, the NOx emissions are clearly increased. Furthermore, during catalyst heating, nitrous oxide and ammonia are also significantly produced in the catalyst [15]. The precise quantification and analysis of gaseous raw emissions in both catalyst heating and normal operation are complex. For details, the reader is referred to paper [15]. With a fuel-type stratification, i.e., an inhomogeneous distribution of the two types of fuel engaged, or a global or even only locally non-stoichiometric combustion air–fuel ratio, however, potentials with alternative fuels at high admixture rates or in the Smart-Fuel concept are possible regarding gaseous emissions.

![Fig. 2 Average blending rate in the Smart-Fuel concept within different test cycles](image)
2.3 Particulate emissions

In this chapter, only a short summary of particulate emissions along with the main differences is provided. For details, including the results for the warm stationary and cold nonstationary conditions with the investigation of the particle-size distribution spectrum, the reader is referred to the paper [15]. The emphasis of this study is put upon the effects for a vehicle fleet, also including the Smart-Fuel concept. This publication focuses primarily on the resulting emissions for a vehicle.

The dynamic particulate emissions cannot be derived in a representative manner from the stationary test bench measurements. This is because the largest proportion of particulate emissions occurs under dynamic and cold boundary conditions. Calculating the particle emission level of the steady-state warm map also does not appear to be expedient, since the particle emission level in large relevant map range is very low and the generally large dispersions in particle formation do not allow a sufficiently precise statement. Therefore, the particle emissions are evaluated exemplarily by means of a dynamic load–speed ramp to a target operating point under cold boundary conditions. The ramp proceeded from an idle operating point to a speed of 4500 rpm and an indicated load of 2.1 MPa within 2 s to get reproducible results. The selected coolant and collector temperature was 20 °C. Figure 3 shows the integral particulate emissions (i.e., the particulate number) of the described load–speed ramp for the different fuel variants.

As previously explained, raw particulate emissions are presented relative to the value of the RON95 E10 base fuel (as 100%). The pure substances ETBE, acetone and DMF were not considered in these studies. The admixture of ethanol and especially methanol leads to a significant increase in particle formation, displaying a local maximum for low-to-moderate blending rates. This is due to the significantly increased evaporation cooling in the mixture composite, because the low boiling point and the chemical soot tendency should have the opposite effect on the particle emissions. However, the high evaporation cooling seems to lead to a more difficult evaporation in the case of wall wetting and thus to an increased soot formation tendency of the remaining proportion of base fuel in the mixture. An increased penetration depth due to the lower volumetric heat value, which means more fuel mass for the same amount of energy, and partially due to a hampered evaporation due to the local evaporation cooling effects might also contribute to this phenomenon. At higher blending rates, the lower boiling point and the significantly reduced proportion of base fuel (with its greater tendency to soot formation) predominate. The raw particulate emissions with 50 vol% ethanol are therefore significantly reduced and those of the 50 vol% methanol blend is approximately at the level of RON95 E10. The pure ethanol and methanol fuels, on the other hand, display only very low soot emissions.

The fuels ETBE and acetone lead to a significant increase in raw particulate emissions only at the 10 vol% blending rates, but lead to a significant reduction at higher rates. This effect is assumed to be due to the very high volatility and the low viscosity of these fuels, which causes the fuel spray to break down very quickly into very fine droplets. As a result, an increased wetting of the injector dome and possibly the combustion chamber roof can occur via an increased interaction with the charge motion [15]. Visual inspection of the injectors after the test program showed a very distinct soot layer on the injector tip when 50 vol% acetone was used, although a relatively clean injector tip would have been expected for acetone as a strong solvent. In contrast, after a run with 50 vol% DMF, which has a high boiling point and a high viscosity, the injector tip was found to be comparatively clean despite a clear increase in raw particle emission. This suggests an increased injector wetting in the case of volatile fuels, which could explain the maximum at their low admixture rates. This is because as the admixture rate increases, the wall wetting presumably increases, but at the same time, the volatility of the fuel mixture also increases, which in turn causes the wall film to evaporate more quickly [15]. DMF, with its high boiling point, generally leads to an

![Fig. 3 Integral particle raw emissions (particulate number) at a dynamic speed–load ramp from near idle to a speed of 4500 rpm and an indicated load of 2.1 MPa at 20 °C coolant and collector temperature for different fuel variants (relative to the value of the RON95 E10 base fuel as 100%)](image-url)
increase in raw particle emission. Nevertheless, there is a local minimum at 25 vol% admixture. The reason for this is assumed to be the analogous mechanisms of action, since DMF with its high boiling point as well as its high viscosity presumably behaves opposite with respect to rapid fine droplet formation in the initial spray break up.

For the design of a Smart-Fuel concept, it is therefore desirable to find an optimum of consumption-optimized and particle-optimized application. DMF shows no advantages in both particulate emission and engine efficiency at different blending rates, so no advantages are expected with variable blending either. For the highly evaporative-cooling fuels ethanol and especially methanol, consumption-optimized mixing would significantly increase particulate emissions at low-to-moderate mixing rates due to the disadvantages identified. For a particle-optimized strategy, the proportioning rate would therefore have to be significantly increased in the dynamic range and especially in the cold range. For a particle-optimized strategy, especially in the cold dynamics, the admixture of ethanol and methanol, for example, could be increased by a factor of two to three depending on the operating point. However, since this dynamic situation exists only in small time portions of the engine operation, since the engine heats up quickly during operation, especially under a high load, and the operating mode also occurs infrequently, such an emission-optimized application adaptation can be regarded as consumption-neutral in a first order of approximation. The consumption-optimized application of the volatile ETBE and acetone appears to also be very advantageous in terms of particulate emissions.

3 Accounting for the entire vehicle fleet

It has been shown that with the exemplary design of the Smart-Fuel concept used in this study (without fuel–air ratio and fuel-type stratification in the combustion chamber) and the predefined fuel blends (also in homogeneously premixed operation), there are neither significant advantages nor disadvantages with regard to gaseous emissions at the engine downstream of the catalytic converter, provided that an adequately dimensioned three-way catalytic converter at operating temperature ensures the conversion of the raw pollutant emissions. Early combustion phasing or nitrogen-containing fuel types produce a higher concentration of NOx in the exhaust gas and can thus cause the need for a larger catalyst volume. There are significant differences in gaseous raw emissions depending upon the fuel. With nitrogen-containing fuels, an exhaust gas aftertreatment approach must be used, that differs from the current state of the art catalyst heating, such as an electrically preheated catalyst. However, the gaseous emissions can therefore be assessed as neutral for the point of view of the entire vehicle fleet, assuming that the exhaust gas aftertreatment system is adapted accordingly.

In contrast, regarding engine efficiency and particulate emissions, there were significant differences between the various variants. Now, the task was to determine how these effects would play out with a different demand for renewable fuel in the entire vehicle fleet with limited fuel availability in the early phases of a circular economy. To do this, a computational example was applied with a given specific amount of energy for renewable fuel synthesis, and then, the amount of produced fuel was used across the fleet with the different operating options based on this fuel availability. For all fuels in Table 1, a theoretical synthesis route based on renewable electricity (e-fuels) was used. Unlike biomass fuels (first-generation biofuels), these e-fuels do not compete with food production. In addition, e-fuels have a significantly higher emission reduction potential when considering all secondary emissions during production. Fuels from recycled residues were not considered due to their foreseeable limited availability. Table 2 shows how much fuel volume can be produced for which fuel grade from a defined amount of available energy according to manufacturing efficiencies.

The amount of renewable electrical energy available for sustainable fuel synthesis in Germany was assumed to be in the range of 100–200 petajoules based on the energy data on renewable energy from the German Federal Ministry for Economic Affairs and Energy for 2019 [16]. With the corresponding synthesis efficiency (based on energy), which was estimated based on the literature [17–22], the heating value according to Boie [6] (pure gasoline without ethanol and ethanol with 5 vol% water), and the density [9–14] of renewable fuels, the resulting available renewable fuel volume can be calculated. The produced volume of fuels with a low energy density, such as methanol, is significantly higher compared to synthetic gasoline. It should be pointed out that the synthesis efficiencies’ values stated in the available literature vary in part significantly for a given fuel type. However, the complexity of the molecular structure also has an impact on the efficiency. Therefore, based on the literature

| Table 2 | Producible fuel volume (relative to the corresponding value of gasoline as 100%) of different fuels from a defined amount of energy |
|---------|------------------------------------------------------------------------------------------|
|          | Synthesis efficiency | Lower heating value [6] | Density kg/l | Fuel volume % |
| Gasoline | 40                  | 42.03                     | 0.75 [9]     | 100          |
| Ethanol  | 50                  | 25.22                     | 0.80 [10]    | 194          |
| Methanol | 60                  | 19.46                     | 0.79 [11]    | 306          |
| ETBE     | 45                  | 35.75                     | 0.74 [12]    | 134          |
| Acetone  | 50                  | 28.32                     | 0.78 [13]    | 177          |
| DMF      | 50                  | 24.99                     | 0.94 [14]    | 166          |
[17–22], methanol has been assumed to have a high synthesis efficiency and gasoline a correspondingly lower synthesis efficiency. For the other fuels, synthesis efficiencies between methanol and synthetic gasoline were assumed, correlating with their complexity of the molecular structure. In the final analysis, the exact degree of synthesis efficiency is not decisive, as an estimate carried out for the worst-case scenario displayed. In this scenario, the synthesis efficiency was set to 40% across the board for all fuels, and the benefit of the various fuel types and in particular the Smart-Fuel concept was retained in principle.

Therefore, depending upon the fuel type, a significantly different volume of generated renewable fuel is available based on the same amount of regenerative energy input for synthesis. In addition, the different concept variants have significantly different renewable fuel requirements, so it is necessary to determine how many passenger cars in the entire fleet can be operated with each variant. For this purpose, the fuel consumption-optimized application in the Smart-Fuel concept is used. As explained on the basis of the time shares when operating the vehicle, this application is dominating with respect to fuel consumption aspects compared to the particle-optimized fuel mixing and can therefore be used as a good approximation for the overall fuel consumption. According to information from the Federal Motor Transport Authority, the passenger car fleet in Germany in the year 2019 consisted of approximately 46 million vehicles with an average mileage of 13,600 km per year [23]. Based on the fuel consumption determined in Sect. 2.1, this information can be used to calculate the number of operable vehicles per fuel variant for a given fuel synthesis energy input with a resulting available renewable fuel volume by Table 2. The results are displayed in Fig. 4 for the case of a dynamic RDE test cycle.

Relative values of the plot were adjusted to a fixed admixture of 10 vol% methanol (to 100%). The additional bar at 25 vol% RON95 E10 represents synthetic gasoline production with the assumption that the fuel characteristics are identical to those of the RON95 E10. This assumption does not result in a difference in the fleet rating depending upon the chosen allocation of synthetic gasoline to the fleet. It is generally true that more vehicles are reached with a lower blending rate and more vehicles are also reached with fuels with a lower energy density, such as methanol, because the fuels were mixed by volume and not by energy content. Blending 10% methanol by volume can reach a large portion of the passenger car fleet and is in the range of several tens of millions of passenger cars. The Smart-Fuel concept also reaches many vehicles, because the average blending rates are relatively low. The fuels ethanol and methanol have an advantage over ETBE and acetone due to the lower secondary fuel consumption.

3.1 Carbon dioxide emissions

As part of this research, the tailpipe emissions of the vehicle are adjusted for the CO₂ reduction potential of the e-fuels used. This means that an internal combustion engine operating on fully renewable fuel can be evaluated as CO₂-free in a net consideration. Depending upon the variant of the blending rate or the Smart-Fuel concept, a significantly different proportion of the passenger car fleet can be operated with the renewable variant (see Fig. 4). However, since the entire fleet must always be used, this leads to a different fleet share of vehicles in each case, which must be operated completely with the fossil fuel. Figure 5 shows the CO₂ emissions of the entire car fleet, taking into account the previously used limited availability of the renewable fuel with the aforementioned deduction of CO₂ from the circular economy. The calculation is based on the previously mentioned RDE test cycle.

Relative CO₂ emissions of the entire passenger car fleet were set to the value of RON95 E10 as 100%. The additional bar at 25 vol% RON95 E10 represents a synthetic gasoline substitution with the assumption that the fuel characteristics remain unchanged. Because of the identical fuel properties, there is no difference due to the blending rate and the proportion of how many vehicles run on a blend,
because there is no change in engine efficiency. The only difference is that part of the emitted CO₂ originates from a circular economy. It results that the substitution of the fossil RON95 E10 with renewable gasoline, assuming identical fuel properties, amounts to about 3.5% CO₂ reduction in the passenger car fleet. This is exactly the share of fuel substituted by renewables.

The knock-resistant fuels ethanol, methanol, ETBE, and acetone generate a further advantage over the engine efficiency or a greater reduction of CO₂ emissions due to the increased knock resistance. In addition, the highly evaporative-cooling fuels ethanol and especially methanol not only increase knock resistance more efficiently due to evaporative cooling, but also have other thermodynamic advantages in terms of engine efficiency. In general, the trend shows that in the entire passenger car fleet, it is more effective for a mono-fuel concept with fixed proportion of admixture to cause a small improvement on many vehicles (red bars in each case in Fig. 5) than to cause a large improvement on a few vehicles (purple bars in each case in Fig. 5). The major advantage of the blending rate of 50 vol% compared to 25 vol% is due to the hardware optimization of the unit to the fuel properties.

The Smart-Fuel concept shows that for the knock-resistant fuels ethanol, methanol, ETBE, and acetone that the reduction of CO₂ emissions due to the variable on-board mixing is significantly greater than with fixed fuel mixtures. This is due to the combination of large number of vehicles reached, the increase in compression ratio made possible, and the use of the second fuel only at necessary operating points. It is noticeable that the benefit is significantly greater for ethanol and especially methanol than for ETBE and acetone. This is due to the greater increase in engine efficiency as a result of the strongly evaporation-cooling fuels and to the larger fuel volume available as a result of the low energy density.

Methanol can thus produce the same efficiency improvement with a lower input of renewable primary energy on a vehicle as the other renewable fuels used, each with higher renewable primary energy inputs. In contrast to the blending of synthetic gasoline with a CO₂ reduction of about 3.5%, a reduction of about 12.5% in CO₂ emissions can be achieved in the Smart-Fuel concept with higher volumetric amounts of methanol but based on the same amount of renewable electricity.

### 3.2 Particle emissions

Even with modern particle filters, particle emissions are relevant. This is because it is possible to compensate for different levels of particulate raw emissions using different sized or complex particulate filters, but this has a negative impact on exhaust gas backpressure and thus on engine efficiency. Due to the very strict legal limits, it is necessary to consider the particulate emissions even after the particulate filter, despite their already very low level. Particulate emissions are mainly generated in dynamic and especially in cold operating conditions. This was assessed, as described in Sect. 2.2, using a dynamic load–speed ramp on the engine test bench. Since these operating conditions only occur in very small proportions during a test cycle, the admixture in the Smart-Fuel concept can be increased significantly in favor of lower particulate emissions without any significant disadvantage in terms of consumption. In the case of the fuels ETBE and acetone, the additions of the fuel-optimized calibration were sufficient for a significant reduction in raw particulate emissions, but for the fuels ethanol and methanol, the admixture rate had to be increased by a factor of two to three. This is due to the increased evaporation cooling of these fuels and the resulting behavior under cold boundary conditions. In the following calculation, this adjustment is assumed to be consumption-neutral in the first approximation due to the
low temporal share relevance within a driving cycle. This means that, in a first-order approximation, as many vehicles can be operated in a particle emission-optimized application for each variant as they can be operated in an optimized fuel consumption application. Figure 6 shows the particulate raw emissions of the entire vehicle fleet as assessed based on a dynamic load–speed ramp taking into account the limited availability of renewable fuels.

The particle raw emission of the entire passenger car fleet is displayed relative to the basis of RON95 E10 as 100%. The additional bar at 25 vol% RON95 E10 represents synthetic gasoline production with the assumption that the fuel characteristics are identical to those of RON95 E10. Therefore, synthetic gasoline does not differ from fossil fuels. DMF shows disadvantages regarding the raw particulate emissions and therefore has disadvantages in all fixed blending variants. In the Smart-Fuel concept, DMF can be approximately neutral regarding the particle raw emissions, if only RON95 E10 is used in the critical driving situations. In the case of fixed admixtures, the advantages and disadvantages of Sect. 2.2 continued in scale via the number of vehicles that can be operated. Ethanol and especially methanol lead to an increase in particulate raw emissions at low-to-moderate blending rates due to the high evaporation cooling. The fuels ETBE and acetone lead to an increase in raw particulate emissions at blending rates of 10 vol% due to the presumed stronger fuel spray breakup and the resulting stronger wall film formation. Here, the high rates are clearly positive due to the rapid evaporation of the wall film despite the strong jet breakup. Due to the great variability in the Smart-Fuel concept, the main influence factor for the generally low-soot fuels ethanol, methanol, ETBE, and acetone depends mainly upon the number of vehicles that can be operated. Because of the low time shares of critical operating conditions, in which a large part of the integral particulate emissions occur in the driving cycle, the admixture rate can be adjusted in an emission-optimal manner, and thus, a very large advantage can be achieved. The fuels ethanol and especially methanol are therefore significantly more advantageous compared to ETBE and acetone. The Smart-Fuel concept with methanol can reduce the particulate raw emissions of the passenger car fleet by approximately 60%. It should be noted that an unadapted blending of renewable fuels can also lead to an increase in particulate raw emissions, as was found for the low-to-partially medium blending rates.

4 Summary

The fuels ethanol, methanol, ETBE, acetone, and DMF were investigated in various fixed admixture rates as well as in a simplified Smart-Fuel concept. In terms of engine efficiency in an exemplary RDE cycle, DMF was found to be minimally disadvantageous, ETBE as well as acetone advantageous, and ethanol as well as methanol very advantageous. The reason why ethanol and especially methanol have a clear advantage over ETBE and acetone is due to the high evaporative cooling of the alcohols, although the chemical anti-knock properties of ETBE and acetone can be classified as significantly higher. High evaporative cooling more effectively reduces the tendency to knock on modern spark ignited direct-injection internal combustion engines and has other significant thermodynamic advantages, such as reduction of compression work, increase of expansion work, and lower temperature level with accompanying more favorable isentropic exponent. The pure substances of the knock resistant fuels have the greatest absolute benefit, but the Smart-Fuel concept can achieve much of these benefits with a smaller volume input from the renewable fuels.

When looking at the entire fleet in a start-up circular economy, it was found that the number of vehicles reached
in the fleet is crucial. In a mono-fuel concept with fixed predetermined fuel blends, it is also more effective to distribute the limited available fuel evenly to all vehicles with a small advantage than to distribute it to a few vehicles with a large advantage. The Smart-Fuel concept with the efficient use of the limited available fuels is significantly better than the fixed fuel blends. The advantage of the Smart-Fuel concept can be explained by the fact that the limited available fuel is only used when it is profitable in terms of efficiency and emissions. As a result, a very large number of vehicles can be reached by this efficiency measure. However, the Smart-Fuel concept requires a technical modification to the vehicle. In contrast, this is not necessary for admixtures of fuels that are compatible with materials, combustion processes, and engine processes (e.g., crankcase ventilation, …), and thus, the entire existing fleet could be reached.

During a chosen representative driving cycle, particulate emissions mainly occur during dynamic operation and especially under cold boundary conditions. Therefore, this potential was investigated using a dynamic load–speed ramp with cold temperature boundary conditions. It was found that a low-to-moderate addition of ethanol and especially methanol due to the high evaporation cooling can lead to a significant increase in the particle raw emissions. In the case of ETBE and acetone, a low admixing rate also leads to increased raw particulate emissions, because the volatility and low viscosity presumably influenced the direct-injection spray breakup, and this can lead to more wall film formation. Toward even higher proportioning rates, however, this effect is overcompensated by the improved evaporation behavior and the lower proportion of base fuel. DMF, with its high boiling point, is generally considered to be disadvantageous for the raw particle emissions.

Since the time portions of the dynamic cold boundary conditions in an RDE test cycle are rather small, the admixture rate can be increased in the Smart-Fuel concept in favor of particulate emissions in dynamic or cold operation, without significant additional consumption of secondary fuel. Throughout the vehicle fleet, emissions for the fixed fuel admixtures are scaled over the number of vehicles accessible. For the Smart-Fuel concept, it only depends upon how many vehicles can be reached with the fuel-optimized application, since the particle-critical events can be bypassed with an excessive admixture rate.

The fuels ethanol and especially methanol have an advantage over ETBE and acetone in the Smart-Fuel concept due to the more effective increase in engine efficiency and the larger volume produced due to the lower energy density. The Smart-Fuel concept with methanol can reduce the CO₂ emissions of the entire vehicle fleet by about 12.5% and the raw particulate emissions of the entire vehicle fleet by about 60%. To achieve this, a secondary fuel tank size of about 15% of the main tank volume is sufficient in the worst-case, provided that the secondary tank is refilled with each filling of the main tank.

In this study, no fuel-type stratification in the combustion chamber was considered, although this would be possible in principle with the Smart-Fuel concept. In addition, primarily homogeneously premixed conditions were considered. By including fuel type stratification and local stratification of the combustion air ratio in the operating strategy, the potential for increasing engine efficiency and reducing pollutant emissions (particulate and gaseous) can presumably be significantly improved [24].

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