# Effect of alcohol blending on real driving emissions of particulate matter from ordinary gasoline automobile engines: A comparison among ethanol, n-butanol and isobutanol

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### Abstract

This paper summarizes the recent and ongoing work on real driving emissions of several automobiles with ordinary, non-flexible-fuel spark ignition engines, powered by alcohol-gasoline blends with higher concentrations of ethanol, n-butanol and isobutanol. On a Ford Focus automobile with a direct injection EcoBoost engine, powered by gasoline and its blends with 15% ethanol, 25% n-butanol and 25% isobutanol, particle size distribution were measured with an on-board fast mobility particle sizer along a 55 km route. Particle emissions were moderately reduced by ethanol and considerably by both butanol blends. On a Škoda Fabia and a Škoda Felicia cars with indirect injection engines, powered by blends with higher concentrations of ethanol, n-butanol and isobutanol, particle emissions measured by a miniature on-board system were examined over a 13 km route. Blends of 30% and 50% of butanol had no or slightly positive effect on particle emissions. Blends of 70% ethanol and 85% n-butanol and 85% isobutanol, used with an auxiliary engine control unit, had no to slightly positive effect on particle length (roughly corresponding to lung deposited surface area) by about one half.

## Introduction

This study evaluates the emissions performance of ordinary in-use gasoline engines when operated on higher concentrations of ethanol, n-butanol and isobutanol, with focus on real-world particulate matter emissions (real driving emissions).

Replacement of fossil automotive fuels with renewable, low carbon footprint, domestically produced fuels and reducing exhaust emissions of primarily particulate matter and secondarily nitrogen oxides are among the main challenges automobile engines are currently facing. A large variety of fuels have been examined, out of which several have obtained larger market penetration: natural gas in gaseous and liquid form, liquified petroleum gas, ethanol, and biodiesel. Of these, ethanol and biodiesel are produced from renewable resources, with ethanol being used primarily in spark ignition engines, and biodiesel virtually exclusively in compression ignition engines.

Ethanol is an oxygenated compound with 35% of oxygen by weight. For this reason, more ethanol (both by weight and by volume) is needed, compared to gasoline, to form a

stoichiometric mixture with a given amount of air. Therefore, on any engine calibrated to run on gasoline, the quantity of the fuel delivered must be increased when running on ethanol. There are therefore two strategies to use ethanol: either blended in small concentrations (up to around 10%) with gasoline for the general use, or in high concentrations in designated engines. The current practice in the Czech Republic, where E85 (spark ignition engine fuel containing 70-85% of ethanol) is widely available at filling stations, while the number of flexible fuel vehicles certified to run on this fuel is rather small, suggests that ethanol is used in higher concentrations in the existing vehicle fleet. Assuming that the fuel does not lead to adverse performance (otherwise it would not be used by the public), the remaining question is the effect of such practice on exhaust emissions. The effects observed during laboratory studies are reviewed in [1-3] and in the previous works by the authors [4-6]. It has been, however, known that the emissions under realistic driving conditions are often higher than during standardized type-approval laboratory tests. Therefore, the question of the effects of higher concentrations ethanol blends on real driving emissions was sought to be answered by real driving emissions tests.

Also, as ethanol is known to be hygroscopic and aggressive to many elastomers and other materials found in the fuel systems [7,8]. For this reason, additional alcohols which could also be produced from biomass were considered. Two isomers of butanol, n-butanol (1-butanol) and isobutanol (2-methyl-propan-1-ol), have the potential to be commercially produced from biomass [9-12] at costs and fossil energy inputs comparable to ethanol [9]. Compared to ethanol, both n-butanol and iso-butanol have higher energy density, lower hygroscopicity, higher viscosity, better lubricity, lower vapor pressure [13], and are less aggressive towards many materials commonly used in vehicle fuel systems. Both isomers of butanol have been used in spark ignition engines, both port fuel injection [1-5,14-19] and direct injection type [6,20-22], with encouraging results, yet without a universal consistent conclusion as to the effect on the emissions.

In the recent past, the performance of butanol blends has been investigated by the authors on several engines, including throttle body injection, port injection and direct injection automobile gasoline engines, and several small carbureted engines used in garden equipment and an electric generator. Of these, three automobiles have been tested under real driving conditions with a portable on-board monitoring system, during which the emissions of particulate matter were also measured. These measurements are summarized in this paper.

## Experimental

## Portable on-board monitoring system

The vehicles were fitted with a portable, on-board exhaust emissions monitoring system designed by the first author [23,24]. The system samples raw, undiluted exhaust gases via a 6 mm diameter stainless steel tube inserted into the tailpipe, and a 6 mm internal diameter, 5 m long conductive fuel line used as a sample line. The sample passes through condensation bowl where condensate is trapped and periodically removed. The sample is then reheated to approximately 60 C by passing through a resistance-heated copper coil. Concentrations of nitrogen monoxide (NO), carbon monoxide (CO) and carbon dioxide (CO<sub>2</sub>) were measured online with a pair of modified, optimized and tuned BAR-97 grade analyzers, utilizing non-dispersive infra-red analyzers (HC, CO, CO<sub>2</sub>) and electrochemical cells (NO and NO<sub>2</sub>). The response of the NDIR sensor used in this study to ethanol and to hydrocarbon mix during the operation on E85 has not been determined. Specifications for an analogous detector [25] show that the sensitivity to ethanol lies between the sensitivities of propane and hexane, both of which

are commonly used to calibrate the automotive NDIR analyzer. Traditionally used flame ionization detector (FID) was not determined to be a reliable reference, as it has cross-sensitivity to oxygenated compounds, resulting in understatement of the concentration of oxygencontaining hydrocarbons [26]. Also, as the sample system is not heated, and portion of water vapor in the sample is removed by the condensate, it can be presumed that ethanol, which is water-soluble, is lost to condensate. Ethanol has been found to be one of the major constituents of organic species on ethanol-fueled vehicles [27]. The CO and CO<sub>2</sub> measurements using the NDIR method are rather straightforward and no adverse issues were anticipated. While the instrument measures both NO and NO<sub>2</sub> using electrochemical cells, only the NO measurement is sufficiently dynamic for transient tests, and was evaluated here quantitatively. The volumetric concentrations of total nitrogen oxides (NO<sub>x</sub>) were assumed to be identical to those of NO in most cases during this study. This overall assumption has been verified by extensive comparison tests of the on-board system, and is also in agreement with analogous sensors being used, in many regions, in periodic emissions inspections of spark ignition vehicles nominally operating at stoichiometric ratio. This is also in agreement with general experience that for engines with no catalytic devices and for engines operating mostly at stoichiometric conditions, the concentrations of nitrogen dioxide (NO<sub>2</sub>) are several percent of the total nitrogen oxides (NO<sub>x</sub>); the only engines known to produce relatively high emissions of NO<sub>2</sub> are those equipped with a highly doped oxidation catalyst and operating lean (with excess air). This is also in agreement with the observed range of response of the NO<sub>2</sub> cell, based on which it is not apparent that larger quantities of NO<sub>2</sub> (tens of percent of total NO<sub>x</sub>) were produced.

Concentrations of particulate matter were measured online with a forward scattering integrating nephelometer, which, for a given engine and a given setup, tends to provide output proportional to particle mass concentration [4,23]. This measurement is believed to be possibly affected both by the low light scattering efficiency of smaller particles, and by the effects of fuel on the particle composition and morphology (mean size, fractal dimension, ...) and hence on the ratio of the light scattering efficiency to particle mass.

Concentrations of particulate matter expressed as total particle length were measured with a modified industrial building smoke detector equipped with a measuring ionization chamber utilizing a small radioactive source (241Am, 30 kBq) to ionize the air. When voltage is applied to the electrodes in the chamber, a small ionization current flows through the chamber. Particles entering the chamber absorb the ions and decrease the ionization current. The detector was modified so that ionization current can be sensed directly and recorded by a data acquisition system. Laboratory comparison tests carried on engine exhaust by the first author [28] have shown that the system provides a response proportional to total particle length concentration (i.e., ft of particles per cu.in., or m.cm-3), that is, the sum of electric mobility diameters of all particles in a unit of volume.

On the DISI engine, where particulate emissions were anticipated to be the primary issue, particle size distributions and concentrations were measured online with a fast mobility particle sizer (EEPS, Model 3090, TSI), preceded by a secondary dilution by a rotating disc diluter (MD-19, Matter Engineering) set to 180:1 dilution ratio; the diluter head was heated to 150 C.

On the throttle body and MPI engines, where particulate emissions were anticipated to be low, while unregulated gaseous compounds were of concern, measurements of gaseous emissions were also done with a prototype miniature portable FTIR (Fourier Transform Infra Red) analyzer with liquid nitrogen cooled MCT detector with a 6-meter path length cell running at 121 C and a resolution of  $0.5 \text{ cm}^{-1}$ .

#### **DISI engine tests**

A typical European small family car (C-segment production passenger car), 2013 Ford Focus station wagon, with downsized three-cylinder 1.0 liter turbocharged gasoline direct injection EcoBoost engine (parameters of the engine are given in Table 1), 6-speed manual transmission, tire size 205/55 R16, 1242 kg curb weight, has been tested at the Czech Technical University in Prague on a 55 km route used for real driving emissions measurements. The vehicle was certified to Euro 6 standards, with rated fuel consumption of 5,8/4,2/4,8 l/100 km, rated CO<sub>2</sub> emmissions 114 g/km, designed to run on 95-octane (RON) gasoline (EN228). The vehicle mileage was 7962 km (4948 mi) at the beginning and 10 130 km (6296 mi) at the end of the study. The test route, overlaid on the map in Fig. 1, is located northwest of Prague in central region of Czech Republic with total altitude difference 165 m and includes approximately one third of urban, one third of suburban, and one third of freeway travel. The motorway part contains one single ascent of 75 m. The elevation profile of the route and typical speeds are given in Fig. 2. The speed in urban part is limited to 50 km/h, suburban to 90 km/h, motorway to 130 km/h. All runs were driven with the same driver in an attempt to compensate influence of different drivers.

Non-oxygenated gasoline with a nominal research octane number of 95, meeting ČSN EN228 specifications, has been obtained at the local fueling station (EuroOil, Buštehrad, Hrebečská 695, 27343), and used as the baseline fuel for the testing. Commercially available E85 fuel, also obtained from a local fueling station (LPG-AUTO s.r.o., Michelská 4/11, Prague 14000) and analyzed to contain 70% of ethanol, was mixed with the base fuel to produce a blend containing 15% of ethanol by volume (E15). Technical grade n-butanol (Chemlogistic, Pardubice) and isobutanol (Chemap, Dašice) were also mixed with the baseline fuel to obtain a blend of 25% of n-butanol with gasoline (nBu25) and a blend of 25% of isobutanol with gasoline (iBu25). The fuels were metered on mass basis using their actual (measured) densities into 20-liter (five-gallon) canisters and splash-blended.

## Throttle body and port fuel injection engine tests

Two cars representative of significant share of cars with naturally aspirated spark ignition engines were used for experimental runs. Both cars are equipped with five gear manual transmission. For high alcohol share an additional control unit for fuel injection pulse width increase has been used. Additional unit producer instructions were followed, so for mixtures containing 85% by volume of butanol settings equivalent to concentration of ethanol up to 50% and for E85 settings for more than 50% ethanol mixture were applied. No other modifications of tested cars and their engines were carried out.

First of tested vehicles was Škoda Felicia equipped with a four cylinder in-line single point fuel injection (SPI, or more accurately, throttle body injection - TBI) spark ignition engine manufactured in 1996 with mileage about 150 thousand km. This vehicle remains popular in the Czech Republic and represents large share of the vehicle fleet despite obsolete air-fuel mixture technology (SPI). Selected parameters of this engine are listed in Table 1. This engine is equipped with three way catalyst, utilizes air to fuel ratio control circuit and is designed to meet emission standard Euro 2.

The second car Škoda Fabia was chosen to represent broadly used type of cars with naturally aspirated downsized engines. This car has been manufactured in 2006, exhibits mileage about 150 thousands km and is propelled by a three cylinder port fuel injection (PFI) engine with

selected parameters listed in Table 1. This engine is also equipped with three way catalyst, utilizes air to fuel ratio control circuit and is designed to meet emission standard Euro 4.

The base fuels were identical to the fuels for the DISI engine, however, neat E85 (71% ethanol) was used, and both isomers of butanol were blended with gasoline at 30%, 50% and 85% volume. For 85% butanol concentration a significant decrease of available torque made ordinary driving through the steep part of the testing route impossible on both cars, and an auxiliary control unit for fuel injection pulse width prolongation (Europecon Flex) has been used for E85 and for 85% butanol blends.

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Engine	Ford Ecoboost 10	Škoda 136B	Škoda 1.2 HTP (VW code BME)
Number of cylinders	3	4	3
Displacement [dm <sup>3</sup> ]	0.999	1.289	1.2
Bore [mm]	71.9	75.5	76.5
Stroke [mm]	82	72	86.9
Compression ratio [-]	10	10	10.5
Brake power [kW]	92 at 6000 rpm	50 at 5500 rpm	47 at 5400 rpm
Maximum torque [Nm]	170 Nm at 1400 – 4500 rpm	100 at 3750 rpm	112 at 3000 rpm
Firing order		1 - 3 - 4 - 2	1 - 2 - 3

Table 1: Selected parameters of the tested engines

The local test route selected for this testing is a 13-km route featuring urban and hilly rural driving, typical for the region, and typical for the operation of this type of vehicle. The route and its elevation profile are given in Fig. 2. The route starts at the university campus. Traversing through residential neighborhood in the first part, the road ascends 292 m through forest into a pass in Rudolfov (at 5 km), from where it descends along a creek (until 9.2 km), continuing through residential and mixed-use neighborhoods. With a range of driving styles, the inclines and numerous curves on this route allow for the engine to be operated at points throughout its operating range. A conservative, leisurely driving style was practiced, as the most representative style for this type of vehicles, during all tests described here. On each fuel, typically five to six runs of the test cycle were made, with the first run considered a "preconditioning" run, with the expectation that of the remaining 4-5 runs, at least three will produce valid data with a reasonable variance in total emissions per route among the runs.



Figure 1: Test routes: Prague 55 km DISI route (left) and Liberec 14 km MPI route (right)



Figure 2: Speed (top) and elevation (bottom) profiles of the routes: Prague (left) and Liberec (right)

#### **Results and Discussion**

The cumulative (left) and instantaneous (right) emissions of particulate matter expressed on mass (top) and number (bottom) basis for the DISI engine are plotted in Figure 3.



Figure 3: Cumulative (left) and instantaneous (right) emissions of particulate matter expressed on mass (top) and number (bottom) concentration basis for the DISI engine: comparison of non-oxygenated gasoline (Gas) and its blends with 15% ethanol (E15), 25% n-butanol (nBu25) and 25% isobutanol (iBu25).

For the throttle body injection car, the cumulative and instantaneous particulate matter emissions expressed as particulate mass are shown in Figure 4 for each run on gasoline, E85 and blends of 85% n-butanol and isobutanol, and in Figure 5 for each run on blends containing 30% and 50% of butanol. The cumulative and instantaneous particulate matter emissions expressed as total particle length (a value rougly corresponding to the total surface of particles deposited in the lungs, or lung deposited surface area) are shown in Figure 7 for each run on gasoline, E85 and blends of 85% n-butanol and isobutanol, and in Figure 7 for each run on blends containing 30% and 50% of butanol. The particle emissions on the MPI engine were relatively low, however, strong effects of what appears to be lubricating oil consumption during engine motoring were observed. For this reason, data from the MPI engine are not presented, as they do not offer a meaningful comparison of fuels.

For the DISI engine, the overall emissions over the test route, in the range of approximately  $1 \times 10^{12}$ -8x10<sup>12</sup> particles >23 nm per km, correspond to the  $2 \times 10^{12}$ -4x10<sup>12</sup> particles per km range observed during the laboratory tests. The differences among the fuels are larger, and the general repeatability lower, compared to the laboratory tests (described in [6]). Furthermore, it is apparent that large portion of total particle emissions from gasoline and E15 runs originates from high-power operation, notably acceleration on an uphill stretch of a freeway. The concentrations during such spikes are highest for gasoline, lower for E15, lower for nBu25, and lowest for iBu25. What appears to come out as the most potent take-home message is the observation that when we choose to blend either ethanol or butanol with gasoline to reach about 5% of oxygen by weight in the fuel, both n-butanol and iso-butanol, at 25% by volume in gasoline, appear to yield substantially higher reduction in particle mass and number emissions compared to 15% ethanol. There is no conclusive evidence as to which butanol isomer is better, leaving both n-butanol and iso-butanol as suitable candidates for consideration.

For the throttle body injection engine, particulate matter mass emissions were in the range of 2 to 2.5 mg/km. While some decrease was observed on all alcohol blends, and given the generally good test-to-test repeatability, they could be considered statistically significant, the method itself – light scattering – is a surrogate method for mass measurement, and it is the opinion of the authors that a 20% difference is too small to be reliably attributed to the fuel. Likewise, while a small reduciton in total particle length is apparent for 30% and 50% blends of both butanol isomers, such difference is too small to be conclusively attributed to the fuel effects. The difference in particle length emissions for 85% blends of both n-butanol and isobutanol and for E85, all being approximately one half of gasoline values, are, however, substantial, and given the small variances among individual measurements, are statistically significant. It can therefore be concluded that a) intermediate concentrations of 30% and 50% of butanol, used in unmodified engine, had no or slightly positive effect on both particle mass and particle length emissions, and b) that high concentrations of 85% butanol, as well as E85, when used with an auxiliary control unit, had no or slightly positive effect on particle mass emissions, and reduced particle length emissions by approximately one half compared to gasoline.

The particle mass emissions for the DISI and TBI engine were comparable, in the range of one to several mg/km. This is consistent with the cumulative effects of DISI engine particulate matter emissions being considerably higher, and of emissions of newer cars being substantially smaller compared to older vehicles.



Figure 4: Instantaneous and cumulative particulate matter emissions measured by laser scattering expressed as particulate mass for (top to bottom) gasoline, E85 and blends of 85% n-butanol and isobutanol with gasoline.



Figure 5: Instantaneous and cumulative particulate matter emissions measured by laser scattering expressed as particulate mass for (top to bottom) blends of 30% isobutanol, 30% n-butanol, 50% isobutanol and 50% n-butanol with gasoline.



Figure 6: Instantaneous and cumulative particulate matter emissions measured by ionization chamber expressed as total particle length (a value corresponding to lung deposited surafce area) for (top to bottom) gasoline, E85 and blends of 85% n-butanol and isobutanol with gasoline.



Figure 7: Instantaneous and cumulative particulate matter emissions measured by ionization chamber expressed as total particle length (a value corresponding to lung deposited surafce area) for (top to bottom) ) blends of 30% isobutanol, 30% n-butanol, 50% isobutanol and 50% n-butanol with gasoline.

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