Applied Energy 102 (2013) 44–54

Contents lists available at SciVerse ScienceDirect

Applied Energy

journal homepage: www.elsevier.com/locate/apenergy

Effects of low temperature on the cold start gaseous emissions from light duty vehicles fuelled by ethanol-blended gasoline

M. Clairotte [a], T.W. Adam [a], A.A. Zardini [a], U. Manfredi [a], G. Martini [a], A. Krasenbrink [a], A. Vicet [b], E. Tournié [b], C. Astorga [a,*]

[a] European Commission Joint Research Centre Ispra, Institute for Energy and Transport, Sustainable Transport Unit, 21027 Ispra (VA), Italy
[b] IES, UMR CNRS 5214, CC067, Université Montpellier 2, Place Eugène Bataillon, 34095 Montpellier Cedex 05, France

HIGHLIGHTS

- Most of the pollutants studied were emitted during the cold start of the vehicle.
- More carbonyls were associated with oxygenated fuel (E85–E75) than with E5.
- Acetaldehyde emissions were found particularly enhanced at −7 °C with E75.
- Elevated methane and ozone precursor emissions were measured at −7 °C with E75.
- Ammonia and toluene emissions associated to E75–E85 were lower than with E5.

ARTICLE INFO

Article history:
Received 1 February 2012
Received in revised form 6 August 2012
Accepted 10 August 2012
Available online 9 October 2012

Keywords:
Unregulated emissions
Alcohol fuel
Vehicle cold start
Ozone formation potential
Ethanol
Ammonia

ABSTRACT

According to directives 2003/30/EC and 2009/28/EC of the European Parliament and the Council, Member States should promote the use of biofuel. Consequently, since 2011 all fuels on the market used for transport purpose must contain a fraction of 5.75% renewable energy sources. Ethanol in gasoline is a promising solution to reach this objective. In addition to decrease the dependence on fossil fuel, ethanol contributes to reducing air pollutant emissions during combustion (carbon monoxide and total hydrocarbons), and has a positive effect on greenhouse gas emissions. These considerations rely on numerous emission studies performed in standard conditions (20–30 °C), however, very few emission data are available for cold ambient temperatures, as they prevail in winter times in e.g., Northern Europe. This paper presents a chassis dynamometer study examining the effect of ethanol (E75–E85) versus gasoline (E5) at standard and low ambient temperatures (22 °C and −7 °C, respectively). Emissions of modern passenger cars complying with the latest European standards (Euro4 and Euro5a) were recorded over the New European Driving Cycle (NEDC) and the Common Artemis Driving Cycle (CADC). Unregulated compounds such as methane, ammonia, and small chain hydrocarbons were monitored by an online Fourier Transformed Infra-Red spectrometer. In addition, a number of ozone precursors (carbonyls and volatile organic hydrocarbons) were collected and analyzed offline by liquid and gas chromatography in order to evaluate the ozone formation potential (OFP) of the exhaust. Results showed higher unregulated emissions at −7 °C than at 22 °C, regardless of the ethanol content in the fuel blend. More carbonyls were associated with oxygenated fuel, and acetaldehyde emissions were found particularly enhanced at −7 °C with E75. In addition, elevated methane emission was measured at low ambient temperature when ethanol fuel was used. Moreover, the OFP of the exhaust gas at −7 °C increased with the amount of ethanol in gasoline when the cold start excess emissions were included. However, regardless of the ambient temperature, the ammonia and toluene emissions associated to E75–E85 were lower than with E5.

1. Introduction

Nowadays, the transport sector contributes significantly to air pollution and climate change [1,2] and on-road transportation is considered as a key issue for the next decade [3,4]. At the same time, the European Union (EU) attempts to promote the share of energy from renewable sources in transport with an objective of 10% by 2020 [5,6], and to decrease fuel life cycle greenhouse gas (GHG) emissions [7]. This tendency has also been followed in the United States since 2005 by the Environmental Protection Agency (EPA). In the last version of the Renewable Fuel Standard (RFS II),

* Corresponding author.
E-mail address: covadonga.astorga-llorens@jrc.ec.europa.eu (C. Astorga).

0306-2619/$ - see front matter © 2012 Published by Elsevier Ltd.
http://dx.doi.org/10.1016/j.apenergy.2012.08.010

EPA has set a target of 36 billion gallons of renewable fuel to be blended with gasoline by 2022 [8]. Ethanol in gasoline may be considered a promising alternative which reduces some regulated gaseous emissions such as carbon monoxide (CO) and total hydrocarbons (THC) [9,10] as well as sulfur dioxide (SO_2) [11]. It also contributes to decrease the emission of GHGs by increasing the plant-based carbon content in the fuel, and thus, by closing the carbon cycle. In addition, depending on the feedstock used, ethanol in gasoline can be recognized as sustainable [12]. Therefore, the EU certified recently several programs e.g., the "Greenergy Brazilian Bioethanol verification programme", for the biofuel target under the renewable energy directive [6,13]. Despite the favorable effects on some primary regulated emissions and GHGs, ambient air measurements taken in several areas where high ethanol content in gasoline was implemented highlight the deterioration of air quality. The presence of significant amounts of health-related compounds such as potentially carcinogenic carbonyls and ozone precursors was found [14–16]. Indeed, chassis-dynamometer studies on primary emissions demonstrated such side effects of ethanol-blended fuels a decade ago [17–20]. On the contrary, a recent study in Rio de Janeiro states that modern engine technology and after-treatment together with a higher fuel quality lead toward an improvement of the air quality [21]. However, all these investigations are based on measurements carried out at ambient temperature (about 20 °C) and limited information is available on the emissions of vehicles running with ethanol blended fuel at lower temperature [22,23]. Only the type approval for light-duty (LD) vehicles fuelled with reformulated gasoline (RFG) is currently implemented in EU at low temperature (−7 °C, the so-called type 6 test). This is not the case for fuel with ethanol content higher than 5% [24]. However, such assessment is necessary for emission characterization and modelling before considering the widespread use of ethanol fuel in large scale. Therefore, this study aimed to investigate the impact of fuels with high ethanol content on the regulated and unregulated gases emitted by modern LD vehicles at low temperature. The ammonia (NH_3) was evaluated more in detail because of its impact on human health and vegetation [25]. Moreover, it can form ammonium aerosols which have a direct and indirect effect on climate, and contribute to local visibility problems [26]. The transport sector increasingly contributes to the total atmospheric NH_3 emission especially during winter when agricultural sources decrease [27]. Thereby, the motor vehicle fraction can reach up to 73% in urban areas [28]. Finally, the emissions of ozone precursors and the influence of the cold engine start were evaluated in more detail.

2. Experimental section

2.1. Chassis dynamometer experiments

This study was conducted at the European Commission Joint Research Centre (EC-JRC) Ispra, Italy, in the Vehicle Emission Laboratory (VELA). The facility includes a test cell with controlled temperature and relative humidity to mimic the typical conditions in Europe (temperature range: [−10; 35] °C; relative humidity: 50%). The tests were performed on chassis dynamometer (inertia range: [454–4500] kg), designed for two-wheel and four-wheel drive light duty (LD) vehicles (two 1.22 m roller benchs – Maha GmbH, Germany). The emissions fed to a Constant Volume Sampler (CVS, Horiba, Japan) using a critical Venturi nozzle to regulate the flow (CVS flow range: [3; 30] m^3/min). A series of thermocouples monitored the temperature of the oil, cooling water, pre and post-catalyst exhaust, and ambient conditions.

Three LD gasoline vehicles (Car 1–3) and two LD flexi-fuel vehicles (Car 4 and 5) were tested. Car 1–4 complied with the Euro4 emission standard; whereas Car 5, equipped with a modern direct injection engine, complied with the Euro5a emission standard [24].

Two driving cycles have been applied, the official New European Driving Cycle (NEDC) which is used for type approval of LD vehicles in Europe, and the Common ARTEMIS Driving Cycles (called hereinafter CADC). The NEDC is a cold-start driving cycle (i.e., performed with a cold engine at the beginning of the cycle), which includes a first urban phase of 780 s (UDC) followed by an extra-urban phase of 400 s (EUDC). The CADC has been developed to better simulate typical driving behavior in Europe [29]. Experiments with this cycle are performed after a pre-conditioning of the vehicle i.e., with a warm engine. The CADC consists of three phases, representative of urban (920 s), rural (1081 s) and motorway (1067 s) driving. Fig. 1 depicts the speed profiles of both studied cycles. The tests were conducted at two temperatures, 22 °C and −7 °C, and 50% relative humidity.

A certified reference fuel E5 (called hereinafter RFG) was used in all the vehicles. Non-flexi vehicles were also fuelled with a 10% ethanol blend with gasoline (E10), whereas flexi-fuel vehicles were also fuelled with E85 and E75 while running at 22 °C and −7 °C, respectively. A detailed description of the fuel characteristics is available in Table 1.

Table 2 gives an overview of the combination of fuels, cycles, and ambient temperatures studied.

2.2. Analytical instrumentation

The regulated emissions from all vehicles were measured in accordance with directive 70/220/EEC and its following amendments [30], with an integrated setup which uses following techniques: non-dispersive infrared (for CO/CO_2), a chemiluminiscence (for NO_x) and a heated (191 °C) flame ionization detector (FID for THC) (Horiba, Japan).

A selection of unregulated gaseous emission, including (i) small hydrocarbons like methane (CH_4), ethylene (C_2H_4), and toluene (C_7H_8), (ii) nitrogen species like nitric oxide (NO), nitrogen dioxide (NO_2), nitrous oxide (N_2O), NH_3, and hydrogen cyanide (HCN), and (iii) oxygenated compounds like formaldehyde, acetaldehyde, and ethanol, were monitored at 1 Hz acquisition frequency by a High Resolution Fourier Transform Infrared spectrometer (HR-FTIR – MKS Multigas analyzer 2030, Wilmington, MA, USA). The method is described in detail in the literature [31], therefore, only a brief description is given here. The device is made up of a multipath cell (optical length: 5.11 m), a Michelson interferometer (spectral resolution: 0.5 cm^{-1}, spectral range: 600–3500 cm^{-1}) and a liquid nitrogen cooled mercury cadmium telluride detector (MCT). The raw exhaust was sampled directly from the tailpipe of the vehicles with a heated PTFE (politetrafluoroetilene) line and a pumping system (flow: ca. 10 L min^{-1}, T: 191 °C) in order to avoid the absorption of hydrophilic compounds (i.e., NH_3, NO_2, carbonyls, or ethanol) in condensed water. The residence time of the undiluted exhaust gas in the heated line before the HR-FTIR measurement cell was less than 2 s. The pressure during the measurement was 1013 hPa (±20), and the temperature was set to 191 °C. The calibration of the instrument was based on a factory developed multivariate model. CO, CO_2 and NO_x measurements from the previously described analyzers were used to check the HR-FTIR calibration model, and to synchronize the time-resolved signal.

Recent studies pointed out the lack of selectivity of the heated FID towards oxygenated hydrocarbon compounds emitted in the exhaust from engines fuelled with high ethanol blended mixtures [23,32]. Therefore, we corrected the time-resolved THC volumetric concentration measured with the FID with the concentration of selected aldehydes and alcohols measured with the FTIR as follows: