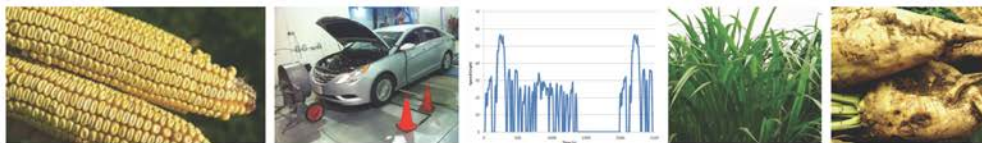


Annex 35-2



A Report from the IEA Advanced Motor Fuels Implementing Agreement

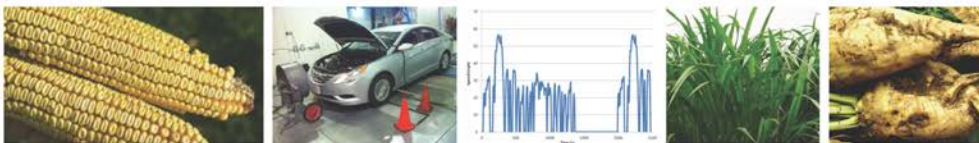
Particulate Measurements: Ethanol and Isobutanol in Direct Injection Spark Ignited Engines

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Abbreviations

AMF	Advanced Motor Fuels
ANL	Argonne National Laboratory, United States
CATARC	China Automotive Technology and Research Center
CFR	U.S. Code of Federal Regulations
CFV	Critical flow venturi
CO	Carbon monoxide
CO ₂	Carbon dioxide
CO ₂ e	Carbon dioxide equivalent
CPC	Condensation particle counter
CPMA	Centrifugal Particle Mass Analyzer
CVS	Constant volume sampling
DISI	Direct injection spark ignited
DMA	Differential mobility analyzer
EC	Environment Canada
EEPS	Engine exhaust particle sizer
EGR	Exhaust gas recirculation
ELPI	Electrical Low Pressure Impactor
ERMS	Emissions Research and Measurement Section, Canada
EPA	United States Environmental Protection Agency
FTP	Federal Test Procedure
GDI	Gasoline direct injection
GHG	Greenhouse gas
GPF	Gasoline particulate filter
HEPA	High Efficiency Particulate Air
IEA	International Energy Agency
LDV	Light duty vehicle
MECA	Manufacturers of Emission Controls Association
NEDC	New European Driving Cycle
NMHC	Non-methane hydrocarbons
NO _x	Oxides of nitrogen

PFI	Port fuel injection
PM	Particulate matter
PMP	Particle measurement programme
SMPS	Scanning mobility particle sizer
TEM	Transmission electron microscopy
UCPC	Ultrafine condensation particle counter
US06	Supplemental Federal Test Procedure
VPR	Volatile particle remover
VTT	Technical Research Centre of Finland

Executive Summary

The global emissions in 2005 of carbon dioxide equivalent (CO₂e) were approximately 27 Gt (giga-tonnes) of which 17% were attributable to road transport.¹ Light-duty vehicles account for the bulk of road transport emissions. Road sector carbon dioxide (CO₂) emissions in 2007 were 127.25 Mt (mega-tonnes) in Canada, 280.47 Mt in China, 12.32 Mt in Finland and 1527.58 Mt in the United States.

Government mandates worldwide are promoting renewable fuels as part of the global energy mix, with the renewable content of transportation fuels being an important part of this strategy. As a result of more stringent Light Duty Vehicle (LDV) exhaust emissions regulations and energy efficiency standards in North America, Europe, Asia and elsewhere, the number of auto manufacturers that are offering downsized turbocharged gasoline direct injection (GDI) engine technologies is growing. Understanding how renewable fuels and new engine technologies impact exhaust particulate matter emissions will be important for setting future emissions regulations.

Environment Canada, as the Operating Agent for *Annex 35: Ethanol as Motor Fuel – Sub-task 2: Particulate Measurements: Ethanol and Butanol in DISI Engines*, has prepared this final sub-task report detailing an international collaborative research initiative to study particulate emissions from direct injection spark ignited (DISI), or alternately termed GDI, engines operated on renewable fuels. Research tasks, organized into European and North American Test Programs, were shared collaboratively between Canada, Finland and the United States in an effort to obtain results that are relevant to multiple regions worldwide. The main goal of this program was to investigate the effects of low, mid and high level blends of ethanol and isobutanol in gasoline on particulate emission rates from GDI engines and passenger vehicles.

This report summarizes particulate data from two different GDI engines and three different GDI vehicles tested in four separate facilities in three countries. These engines and vehicles were tested under different operating modes, driving conditions, and at different ambient test conditions to assess the impacts of alcohol fuel blends and varying fuel blend levels on particulate emissions.

The use of low- to mid-level alcohol blends (E10, E15, E20, iB16) with these GDI engines/vehicles gave mixed results; with some studies noting decreases in particles with alcohol blends and some studies showing increases. These test alcohol fuels were splash blended with gasoline and an investigation of the impacts of other fuel parameters, besides alcohol content, on emissions was not undertaken.

¹ OECD/ITF, 2010.

In contrast to the low level ethanol blends the E85 studies did yield consistent results indicating the potential to mitigate particulate emissions from GDI engines. Research conducted with two test engines under the North American program and one vehicle under the European test program using E85 as a test fuel showed that reductions in particle emissions from GDI engines can be achieved under varying operating conditions and ambient temperatures. In some cases the number of particles was roughly an order of magnitude lower with E85 as compared to E10 and resulted in reductions in the range of 70–90% between E85 and E0. Along with a reduction in particle number, the shape of the particle number distribution curve was also impacted with the distribution peak occurring at a smaller particle size with E85 compared to E10 and E0.

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1. Introduction

Environment Canada, as the Operating Agent for *Annex 35: Ethanol as Motor Fuel – Sub-task 2: Particulate Measurements: Ethanol and Butanol in DISI Engines*, has prepared this final sub-task report on an international collaborative research initiative to study particulate matter emissions from gasoline direct injection (GDI) engines operated on renewable fuels. Research tasks, organized into European and North American Programs, were shared collaboratively between Canada, Finland and the United States in an effort to obtain results that are relevant to multiple regions worldwide.

1.1 Structure of Report

The Introduction to this report provides context and background information relevant to the research. The Research Methodology section is organized according to the North American and European programs that make up this Annex sub-task. Section 3 presents the research results and the final section of the report discusses the implications of the results in light of the research objectives. Section 4 describes how this research is aligned with other Annexes under the International Energy Agency Implementing Agreement for Advanced Motor Fuels.

1.2 Research Context

Worldwide the use of renewable fuels is being promoted as part of the global energy mix. Coupled with more stringent Light Duty Vehicle (LDV) exhaust emissions regulations and energy efficiency standards in North America, Europe, Asia and elsewhere, the number of auto manufacturers that are offering downsized turbocharged GDI vehicle engine technologies is growing. Understanding how renewable fuels and new engine technologies impact exhaust particulate matter emissions will be important for setting future emissions regulations. Additional context for undertaking this research can be found in the International Energy Agency (IEA)– Advanced Motor Fuels (AMF) Implementation Agreement 2012 Annual Report.¹

1.3 Research Objectives

Initial research comparing GDI engines to port fuel injected (PFI) engines has shown that GDI engines, while providing fuel economy benefits, may under some conditions produce greater particle number concentrations and higher particulate mass emission rates. It is anticipated that the North American LDV fleet may consist of up to 60% GDI vehicles by 2016.² Because research has shown that low-level ethanol blends may decrease particulate matter (PM) formation, confirmation that a similar result is observed with GDI vehicles operated under variable ambient temperatures and real world driving conditions is needed.

¹ http://www.iea-amf.org/content/publications/annual_reports.

² State of California Air Resources Board, 2010.

The main goal of this research program was to investigate the effects of blends of ethanol and isobutanol in gasoline on particle emissions rates from GDI engines and passenger vehicles.

Recent research on the use of butanol as an advanced fuel has demonstrated its versatility and suitability for providing greenhouse gas (GHG) emissions reductions.³ Butanol is a four-carbon alcohol with four isomeric structures: n-butanol, sec-butanol, tert-butanol and isobutanol. Different butanol isomers may produce a differing emissions profile. At the initiation of this Annex, knowledge of how butanol blends in GDI engines affect particulate formation was limited. Reliable baseline emissions data for emerging biofuels and energy efficient vehicle engine technologies could help inform the process of certifying performance parameters of next generation renewable fuels. For this report, the discussion is limited to isobutanol.

As an international collaborative study, vehicles for this Annex were tested over numerous drive cycles including: U.S. Federal Test Procedure 75 (FTP-75), US06 Supplemental Federal Test Procedure (US06) and the New European Drive Cycle (NEDC). Also, tests were conducted at various ambient temperatures, to assess particle emissions from GDI vehicles using renewable fuels under simulated real-world driving conditions. Engine testing over various steady-state cycles was used to assess the impacts of renewable fuels on particulates and also to visually characterize particle structure.

1.4 Renewable Fuels

Globally, the transportation sector accounts for 13% of GHG emissions.⁴ In 2009, the transportation sector accounted for approximately 28% of Canada's overall GHG emissions.⁵ The Environmental Protection Agency (EPA) has reported a similar result for the United States.⁶

Renewable fuels have the potential to reduce targeted emissions, in addition to contributing to energy independence.⁷ Use of first generation renewables such as corn ethanol, and advanced biofuels such as butanol and bio butanol, is an important step towards investigating particle emission reductions. Sub-task 1 of Annex 35 looked at "Ethanol as a Fuel in Road Vehicles". Research for Sub-task 1 was conducted by the Technical University of Denmark, Department of Mechanical Engineering, and the results have been reported in a final IEA-AMF publication.⁸ It was concluded that the greatest benefits, in terms of engine efficiencies and emissions reduction, were realized from using high level ethanol blended fuels. Annex 35, Sub-task 2 builds on Sub-task 1 results by

³ Wasil *et al.*, 2012.

⁴ IPCC, 2007.

⁵ <http://www.ec.gc.ca/ges-ghg/default.asp?lang=En&n=72E6D4E2-1#X-201103220714032>.

⁶ <http://www.epa.gov/otaq/climate/documents/420f12012.pdf>.

⁷ Wasil *et al.*, 2012.

⁸ http://www.iea-amf.org/content/publications/annual_reports.

exploring the relationship between alcohol-blended fuels and energy efficient technologies operated under real world driving conditions.

In addition to reducing GHG emissions, regulators are also advocating greater energy efficiency to help reduce dependence on fossil fuels. Vehicle manufacturers are responding with new engine technologies that reduce fuel consumption.⁹ While a primary goal of increased energy efficiency and renewable fuel technologies is to contribute to climate change mitigation, these changes may give rise to negative outcomes such as the increase in particulate matter emissions observed with GDI engine technologies. Knowing that renewable fuels, when used at high blend levels, can decrease PM emissions, understanding the interactions between renewable fuel and energy efficient technologies will help to inform regulatory decision-making while addressing energy supply and efficiency mandates. Furthermore, since a significant proportion of the existing and near future LDV mix is not designed for high level ethanol blends, there is still a need to understand how low- and mid-level renewable fuels perform in gasoline engines.

1.5 Gasoline Direct Injection Engines

Downsized turbocharged GDI engines offer fuel economy benefits over PFI vehicles and have begun to enter the global vehicle fleet mix as auto manufacturers search for ways to meet increasingly stringent LDV emissions regulations. With conventional multi-port fuel injection systems, the fuel is injected into the engine intake ports where it mixes with air to compose a homogeneous mixture before entering the cylinder. With a GDI fuel system, the highly pressurized fuel¹⁰ is injected directly into the cylinder in a manner similar to a diesel engine. GDI engines offer improvements in thermal efficiency over PFI engines through the combination of homogeneous and heterogeneous air fuel mixtures. By eliminating the air/fuel mixing step in the intake port and by incorporating a relatively high compression ratio, GDI engines can more accurately control injection timing to meet vehicle load requirements. Note that in practice, GDI vehicles use an operating strategy that is based on the use of stoichiometric mixtures throughout the entire load and speed range to ensure that the after-treatment system remains effective.

Over the past decade, different auto manufacturers have released various versions of GDI vehicles and it is expected that the number of GDI vehicles on the road will continue to increase in the near future. GDI, coupled with turbocharging technology, is a viable option for gasoline engines. As a result of this trend, the GDI engines are expected to account for over 35% of the European gasoline engine market by 2013,¹¹ and are projected to reach up to 60% of the new LDV fleet in North America by 2016.¹² Compared to traditional PFI engines, GDI engines may have higher particle emissions due to various factors such as differences in fuel injection and mixture preparation, as well as lean and stratified

⁹ State of California Air Resources Board, 2010.

¹⁰ Zhao *et al.*, 1999.

¹¹ http://www.enginetechnologyinternational.com/market_forecast.php.

¹² State of California Air Resources Board, 2010.

operation. As these vehicles form a greater part of the LDV mix there is a need to better understand the environmental impact of GDI technologies.

With the increased use of renewable transportation fuels, the identification of potential synergies between these energy efficient engines and energy dense renewable fuels like ethanol and isobutanol may be advantageous. Since particle number concentrations could be higher from GDI engines than from PFI engines, it is important to understand the impact of using alcohol-gasoline blends on particulate matter emissions and on the design and effectiveness of after-treatment systems.

1.6 Particulate Matter Emissions

1.6.1 Formation and Composition

Particulate matter (PM) is a complex mixture of compounds with varying chemical composition and physical properties. Particles in vehicle engine exhaust emissions are formed from incomplete combustion and may be volatile, soluble or solid in nature.¹³ Typically, motor vehicle particulate contains various components:

- A carbonaceous fraction composed of elemental carbon (or sometimes referred to as black carbon or refractory carbon) and organic carbon, which includes alkanes, alkenes, alcohols, esters, ketones, acids, and aromatics;
- Sulfates and sulphuric acid;
- Nitrates that are water soluble found chiefly in the form of nitric acid; and
- An ash fraction containing inorganic compounds or elements such as trace metals.

A variety of factors contribute to the formation of PM including fuel consumption, fuel composition, engine operation, fuel injection method, mixture preparation and wall wetting in the cylinder. In vehicular emissions, carbonaceous materials make up the majority of the total PM mass.¹⁴ When emitted into the atmosphere, these particles can undergo various atmospheric processes, such as coagulation or providing a surface area for other less volatile atmospheric compounds to adsorb onto.

Scientific literature has emerged that provides evidence of associations between exposures to ambient PM and increased mortality and cardiovascular and respiratory diseases.¹⁵ Epidemiological studies have shown a link between automotive combustion emissions and adverse effects on the reproductive, nervous, immune and respiratory and cardiovascular¹⁶ systems, as well as being able to induce cancer.¹⁷ The size of the particles is linked to their

¹³ Eastwood, 2008.

¹⁴ Ibid.

¹⁵ HEI, 2010.

¹⁶ Hoffman *et al.* 2007.

¹⁷ Chellam *et al.*, 2005, Schauer, J.J., 2006 and Hung *et al.*, 2012.

potential for health concerns. When the fine particles are inhaled, they may be deposited deep into the lungs and may be transferred to the circulatory system and spread to the organs of the body.¹⁸

Direct emissions of particles from vehicles and the photochemically generated ozone (due to the reaction between NO_x and volatile organic compounds) are a major cause of visual impairment in the form of smog.¹⁹ Depending on the diameter of the atmospheric particles, they could have an atmospheric lifetime up to weeks and therefore can be transported and deposited on surfaces away from the source. Depending on the composition of these particles, they could cause damage to structures and vegetation.

1.6.2 Measurement and Analysis

In North America, the use of standard gravimetric (mass) analysis is the most common method of measuring PM for regulatory purposes. Typically in gravimetric analysis, sample collection filters are preconditioned in a temperature and humidity controlled environment prior to static discharge and weighing on a high precision balance.²⁰ The PM emission rate is then determined based on sample volume and dilution tunnel flow rate. PM emission rates, such as those from vehicles with ultra-low emission engines or those equipped with particulate traps or filters, can be low enough to cause issues in accurately determining PM mass by the gravimetric method. In North America the gravimetric method is currently the method used for regulating PM emissions limits.

In contrast to filter collection of PM, particles in engine exhaust emissions can be monitored by real-time analyzers. Typically, particles are size selected based on either the aerodynamic or electrical mobility diameter in these analyzers. Then the particles can be detected by light scattering or in the case for refractory carbon, the mass information can be inferred based on the light absorption properties of the particles. Some commonly used real-time or semi real-time analyzers include:

- Condensation Particle Counter (CPC), which is often used to provide particle number concentration. In the CPC, individual particles are grown to sub-micrometer diameter through condensation growth and then detected by a laser.
- Electrical Low Pressure Impactor (ELPI) separates particles based on the aerodynamic diameter of the particles and provides real-time particle number size distribution information. Particles are charged inside the charger and the aerodynamic size classification is done inside the impactor by measuring the current values from each stage and transforming them into the number of particles using complex calculations.

¹⁸ <http://www.epa.gov/airquality/particulatematter/health.html>.

¹⁹ <http://www.epa.gov/region7/air/quality/pmhealth.htm>.

²⁰ Code of Federal Regulations, 2004d.

- Scanning Mobility Particle Sizer (SMPS) can also be used to provide particle number concentration. Incoming particles are first charged by a bipolar charger and then size selected based on their electrical mobility diameter. As it takes a finite amount of time to separate the particles, SMPS cannot be used to provide real-time information.
- Engine Exhaust Particle Sizer (EEPS) detects particles based on a similar principle as a SMPS system except particles are charged by a corona. Unlike the SMPS, the EEPS is capable of providing real-time particle number size distributions.
- Transmission electron microscopy (TEM) is an offline method that can be used to investigate the morphology of the exhaust particle. In this method, particles are collected on a copper grid, and then the image is magnified under a microscope using electrons as a light source.
- Differential Mobility Analyzer (DMA) measures the size distribution and concentration of particles is based on the physical principle that the ability of a particle to traverse an electric field is related to particle size. In DMA, an electric field is created, and the airborne particles drift in the DMA according to their electrical mobility. Particle size is then calculated from the mobility distribution.
- Centrifugal Particle Mass Analyzer (CPMA) is an aerosol classifier which selects particles according to their mass to charge ratio. The CPMA uses opposing electrical and centrifugal force fields to classify aerosol particles.²¹

1.6.3 Regulations

The health and environmental impacts of particulate matter are well known and regulations limiting particulates in air are becoming increasingly stringent worldwide. For example, in China emission standards for passenger cars and light duty commercial vehicles, equivalent to the Euro 6 standards, were introduced in Beijing in 2013 and will be expanded nationwide in 2018.²²

In Europe, the Euro 5/6 regulation stipulates a particle mass limit of 4.5 mg/km (7.2 mg/mile), over the NEDC, that is equal to the diesel engine limits. The Euro 5/6 regulations also introduce a particle number limit for GDI light-duty vehicles of 6.0×10^{11} particles/km that would not come into effect until three years after the introduction of Euro 6 emission standards in Europe (starting in September 2017).²³ Several recent studies have suggested that the stoichiometric GDI and lean stratified GDI engines typically have particle number and mass emissions higher than and close to the proposed Euro 6 limits, respectively.²⁴

²¹ Olfert and Collings, 2005.

²² <http://english.cri.cn/6909/2013/09/18/2561s788137.htm>.

²³ http://europa.eu/legislation_summaries/environment/air_pollution/l28186_en.htm.

²⁴ Piock *et al.*, 2011.

The tailpipe emissions standards for cars and light duty trucks in Canada and the United States are aligned. In North America the current Tier 2/LEV II PM limit is 10 mg/mile (6.2 mg/km) over the FTP-75 test cycle.²⁵ California recently approved their LEV III light-duty vehicle emissions limits, which includes the phase-in of a 3 mg/mile (1.9 mg/km) PM limit starting in 2017 and a 1 mg/mile (0.6 mg/km) PM limit starting in 2025 over the FTP-75 test cycle. The US Tier 3 emission regulations are closely aligned with these limits.²⁶

Since 2009, Japan has limited PM emissions from new vehicles produced domestically to 5 mg/km. Russia introduced the Euro 5 standard (PM₅ ≤ 5 mg/km) effective 2014 for new vehicles and 2016 for all sales and registrations. Similarly, Argentina, Brazil and Chile have adopted or will be adopting Euro 5 standards or equivalent in the near term.²⁷

In light of the on-going changes to the global LDV fleet mix and because many countries are beginning to introduce PM limits, it is important and necessary to understand and address the particle emissions from GDI vehicles in order to meet current and future emissions regulations.

1.7 Annex 35 Sub-Task 2 Research

GDI vehicles are currently receiving increased attention by auto manufacturers because of their fuel economy as compared to traditional PFI gasoline vehicles.²⁸ With the use of turbocharging on GDI vehicles, the same driving performance can be maintained with downsized engines, which further reduces vehicle fuel consumption. According to the U.S. Environmental Protection Agency (EPA), among other technologies such as a diesel engine, turbocharging, and vehicle downsizing, the GDI engine was considered as one of the technology paths for meeting future emission regulations for reducing greenhouse gases.²⁹ Based on current data and estimation of compliance with federal requirements in North America, the light-duty vehicle fleet could comprise up to 60% GDI vehicles by 2016.³⁰ Despite the benefits of reduced fuel consumption and CO₂ emissions, GDI vehicles have relatively higher particle emissions because they often run lean and stratified as opposed to stoichiometric and homogeneous like PFI engines.³¹

Similar to diesel engines, exhaust particles from GDI vehicles contain a large fraction of soot with typical mean particle diameters ranging from 60 to 100 nm.³² Particles emitted from GDI engines can, however, vary significantly depending on operating conditions. For

²⁵ <http://www.epa.gov/otaq/standards/light-duty/tier2stds.htm>.

²⁶ <http://www.epa.gov/otaq/tier3.htm>.

²⁷ <http://transportpolicy.net/>.

²⁸ Zhao *et al.*, 1999 and Saito *et al.*, 2011.

²⁹ US Environmental Protection Agency, 2010.

³⁰ State of California Air Resources Board, 2010.

³¹ Zhao *et al.*, 1999, Hall and Dickens, 1999 and Graskow *et al.*, 1999.

³² Hall and Dickens, 1999, Graskow *et al.*, 1999, Maricq *et al.*, 2011, Maricq *et al.*, 2012 and He *et al.*, 2012.

example, during the homogeneous charge mode, the better fuel mixture yields soot particles with a much smaller primary particle size, similar to those observed from PFI vehicles.³³ During stratified charge mode, the air/fuel ratio may vary from very rich near the spark plug, where the soot particles have a much larger primary particle size similar to those emitted from diesel engines, to very lean near the cylinder wall. Potentially, a reduction in particle emissions from GDI vehicles could be achieved by improving engine combustion parameters, by using a particle emissions control device such as a gasoline particulate filter (GPF)³⁴ or by using renewable fuels.

Studies have suggested that the additional oxygen in ethanol can lower PM emissions from PFI engines,³⁵ but may have a different impact on GDI engines because of the different operating principle. Ethanol blended fuel has a lower heat of combustion necessitating 1.5 to 1.8 times the volume of fuel to produce the same energy as gasoline. In addition, the higher heat of evaporation coupled with the injection timing could lead to wall wetting and contribute to PM formation because fuel cannot evaporate completely.³⁶ Chen³⁷ and others have suggested that as GDI engines use late injection during specific operations, the leaning effect of E10 could potentially limit particulate emissions. Other researchers have suggested that repeat variability makes reduction measurements not statistically significant³⁸ and that this could obscure increases in ultrafine particle emissions.³⁹ Higher reductions than seen with E10 have been observed with mid-level blends (E20) and US06 reductions have exceeded FTP-75 reductions.⁴⁰ The splash blending of ethanol blends also plays an important factor in these studies. Splash blending of ethanol with gasoline may impact fuel properties such as vapor pressure, distillation profile and aromatic content. Changes in these properties can impact particle formation during combustion as well as vehicle operability in varying temperature conditions resulting in mixed observations of the impact of ethanol blends on particle emissions.⁴¹

The Annex 35 Sub-Task 2 project compares the particle size distributions and number emission rates from GDI vehicles run on various alcohol-blended fuels over a variety of drive cycles and at different ambient temperatures. Canada is the Operating Agent for this international collaboration with Finland and the United States. Research was conducted over the period of November 2010 to May 2014 under the following test programs:

³³ Barone *et al.*, 2012 and Chan *et al.*, 2012a.

³⁴ Saito *et al.*, 2011, Chan *et al.*, 2012a, Chan *et al.*, 2013 and Chan *et al.*, 2014.

³⁵ Hseih *et al.*, 2002 and Pouloupoulos *et al.*, 2001.

³⁶ Hseih *et al.*, 2002, Serras-Pereira *et al.*, 2008, Price *et al.*, 2007 and He *et al.*, 2012.

³⁷ Chen *et al.*, 2010 and He *et al.*, 2012.

³⁸ Maricq *et al.*, 2012.

³⁹ He *et al.*, 2010.

⁴⁰ Storey *et al.*, 2010.

⁴¹ Chan *et al.*, 2014.

North American Test Program:

Canada – the Emissions Research and Measurement Section (ERMS) of Environment Canada performed chassis dynamometer emissions testing of a flex-fuel GDI vehicle and a standard GDI vehicle over a variety of fuel, drive cycle and temperature configurations. Real-time continuous monitoring using an EEPS determined particle size and number emission rates.

Through the Auto21 Network (Universities of Alberta, British Columbia and Toronto) SMPS scans were performed using a combination of a 3081 series DMA and a 3776 series CPC to assess particle size and number emission rates from a GDI engine tested on an engine dynamometer. Primary particle morphology was investigated with a TEM.

United States – Argonne National Laboratory (ANL) used engine dynamometer testing to study particulate emissions from a General Motors GDI engine operating on alcohol-blended fuels at different load levels. Particle size and number were determined with a SMPS and soot morphology was analyzed by TEM.

European Test Program:

Finland – the VTT Technical Research Centre of Finland conducted chassis dynamometer emissions testing on a turbocharged, direct injection, spark ignited engine run on 85% ethanol fuel using the NEDC. Particle size distributions were assessed by an ELPI.

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2. Research Methodology

2.1 North American Test Program

Canada

General Test Description

At Environment Canada, two separate test programs were conducted to evaluate the impact of renewable fuels on emissions from GDI vehicles. The first project involved the characterization of tailpipe emissions from a model year 2013 flex fuel GDI, operated on E85, E10 and E0. Tests were conducted at 22°C, -7°C and -18°C using the FTP-75.

The second project investigated the impacts of E10, E15, E20 and iB16 on emissions as compared to neat gasoline (E0/iB0) and the impacts of a gasoline particulate filter (GPF) in combination with the use of low-level blend ethanol fuels. A 2011 GDI engine light-duty vehicle was operated over two test cycles (FTP-75 and US06) at standard and cold temperatures (22°C, -7°C and -18°C) with certification gasoline and three different ethanol splash blends: 10% ethanol by volume (E10), 15% ethanol by volume (E15) and 20% ethanol by volume (E20), as well as a isobutanol-16 (iB16) blend. See Appendix A for test fuel analyses.

Laboratory tests were conducted in accordance with the procedures specified in the *Canadian Environmental Protection Act 1999* and are equivalent to the U.S. Code of Federal Regulations Title 40 Part 86 (40 CFR 86). The vehicles were tested at Environment Canada’s climate controlled vehicle chassis dynamometer test laboratory using a 122 cm diameter single roll electric dynamometer with the capability of simulating both the road load power and inertia weight that light-duty vehicles are subjected to during on-road operation.

Driving Cycles

In order to complete a test plan representative of real world driving conditions, emissions from both test vehicles were determined over different transient chassis dynamometer cycles simulating city and highway driving, under standard and cold temperature conditions. The drive cycle parameters for all test cycles used in the Annex 35 Sub-Task 2 research are presented in Table 1. See Appendix A for drive cycle speed-time traces.

Table 1. Test Cycle Parameters

Cycle	Average Speed (mph/kph)	Maximum Speed (mph/kph)	Total Time (s)	Total Distance (miles/km)	Number of Stops
FTP-75	21.2/34.1	56.7/91.3	1874	11/17.7	18
SFTP US06	48.4/77.9	80.3/129.2	596	8/12.9	5
NEDC	21.1/34.1	93.1/120	1180	11/17.7	12

The Federal Test Procedure (FTP-75) replicates a typical city driving pattern and consists of three distinct segments: a cold start phase (first 505 seconds) that is used to determine cold-start emissions, a stabilized phase (i.e., the vehicle is warmed up) and a hot start phase. The US06 is a Supplemental Federal Test Procedure used to simulate aggressive highway driving. Prior to the test cycle, a warm-up US06 cycle is driven so that this test is performed with a hot engine.

The New European Driving Cycle (NEDC) consists of four repeated ECE-15 driving cycles of 195 seconds each and one extra-urban driving cycle (EUDC) of 400 seconds. The four ECE-15 cycles represent urban driving conditions that are characterized by low vehicle speed, low engine load and low exhaust gas temperature. By contrast, the EUDC in the second part of the NEDC accounts for extra-urban and high speed driving modes.

Exhaust Particle Sampling

The exhaust sampling system collected gaseous emissions using a critical flow venturi – constant volume sampling (CFV-CVS) system. The equipment and instrumentation used in this study met the criteria set forth in the U.S. Code of Federal Regulations (CFR) Title 40, Part 86.¹ The particle-free dilution air used in the CVS was HEPA filtered test cell air.

Particle number size distributions (5.6-560 nm) were monitored in real time using a TSI 3090 EEPS. Exhaust particles, sampled directly from the CVS tunnel, were first conditioned using a Dekati thermodenuder (ELA-11). The thermodenuder was used to remove the volatile component on the particles. The thermodenuder consists of the heater and absorber sections. The heater section was operated at 300°C to evaporate a majority of the volatile fraction on the solid particles as well as the liquid particles. In the absorber section, the gaseous phase compounds were removed from the airstream by activated charcoal to prevent the gaseous material from re-condensing onto any pre-existing particles exiting the thermodenuder.

2.1.1 United States

Argonne National Laboratory (ANL) tested a stock spark-ignited Opel 2.2L 4- cylinder Ecotec Engine (GM L850) with direct fuel injection, with the cylinder head modified for cylinder pressure transducers. The engine also had an exhaust gas recirculation system with no cooling, maximum output power of 114 kW at 5600 rpm, and maximum torque of 220 Nm at 3800 rpm. For this research, the engine control unit was modified by disabling some of the original functions that are unavailable on a dynamometer setup, to allow the engine to operate on the dynamometer. The stock automotive ignition system that was supplied with the engine was used without modifications. The engine operating conditions were not optimized for the combustion of blended fuels. Base fuels used in this study were gasoline, ethanol and isobutanol, and the alcohol blended fuels (E10, E85 and iB16) were splash blended at ANL. See Appendix A for fuel analysis.

¹ Code of Federal Regulations, a b c.

The engine was equipped with a high-pressure-gasoline direct injection fuel system that operated at pressures ranging from 40 to 120 bar. The injector parameters, such as cone angle (52°), maximum fuel flow (8.86 g/s), and droplet size (SMD < 16 μm), were optimized for gasoline operation. The stock calibration of the injection parameters (fuel pressure and injection timing), as well as the position of the swirl control valve and the exhaust gas recirculation (EGR) rate, were not changed when the engine was operated on the different alcohol/gasoline blends. The EGR levels used were those prescribed by the manufacturer for operation using E10 and gasoline: for the idling case EGR was approximately 14%; and for all other loads it was approximately 4%.

The thermophoretic sampling technique used in this research was first developed at Argonne National Laboratory to sample particulates from internal combustion engines. The sampling system consisted of a sampling chamber connected to the exhaust manifold, an air cylinder, a solenoid valve and an electronic timing/trigger unit controlling the residence time of the probe. Particulates were collected on the sampling grid by the thermophoresis effect driven by the temperature gradient between the hot exhaust stream and the near room-temperature grid surface. The residence time for sampling was optimized to ensure the collection of a sufficient number of particles, as well as to avoid the artificial agglomeration of particles. Further details for this sampling system are available elsewhere.²

The collected particulate samples were examined by using a Philips CM30 and a Jeol JEM-2100F TEM. The TEM digital images of particulates were analyzed for particle size and morphology by a custom image processing/data acquisition system. Further detailed examination of particulate nanostructures was performed at a high-resolution TEM mode over 600,000 times, which enables examination at an angstrom-resolution level.

2.2 European Test Program

2.2.1 Finland

All vehicle emissions tests were conducted in VTT's temperature controlled light-duty vehicles emission measurement laboratory in Espoo Finland using a flexi-fuel vehicle equipped with a turbo-charged direct injection spark ignited (DISI) engine that met the Euro 5a emissions standard.³ The commercially available fuels used in this research program were 95 octane gasoline with 10% ethanol content (95 E10) and high concentration ethanol fuel with 85% ethanol content (E85). See Appendix A for test fuel analysis.

² Seong *et al.*, 2012.

³ Nuottimäki and Murtonen *et al.*, 2013.

The vehicle was tested on a chassis dynamometer (Froude Consine) at 23°C and -7°C using the NEDC, see Table 1 and Appendix A, and emissions were collected with a venturi-type constant volume sampler (AVL CVS i60 LD). All equipment used for the measurement of regulated gaseous emissions (exhaust dilution and collection, concentration analysis, etc.) conforms to the specifications of European Test Directive 70/220/EEC and its amendments. The tests were completed in parallel with the measurements taken for IEA-AMF Annex 43: Performance Evaluation of Passenger Car, Fuel and Power Plant Options.

Particle number size distributions were measured with the ELPI. For this research a 10 lpm low pressure impactor was used with a filter stage giving a lowest cut point of 8 nm. The sample was taken from raw exhaust gas and diluted in two stages: a porous tube diluter (diluter1) was used as primary diluter and an ejector type diluter (diluter2) as secondary diluter. Dilution air was filtered and dried with an absorber dryer. The total dilution ratio was set to 93 (primary 12.5 and secondary 7.4). The dilution air flow to the porous tube diluter was controlled with a mass flow controller and the total dilution ratio was measured over the test cycles via CO₂. The measured dilution ratio was used for calculating the results. Figure 1 presents a block diagram of the particle number size distribution measurement system used.

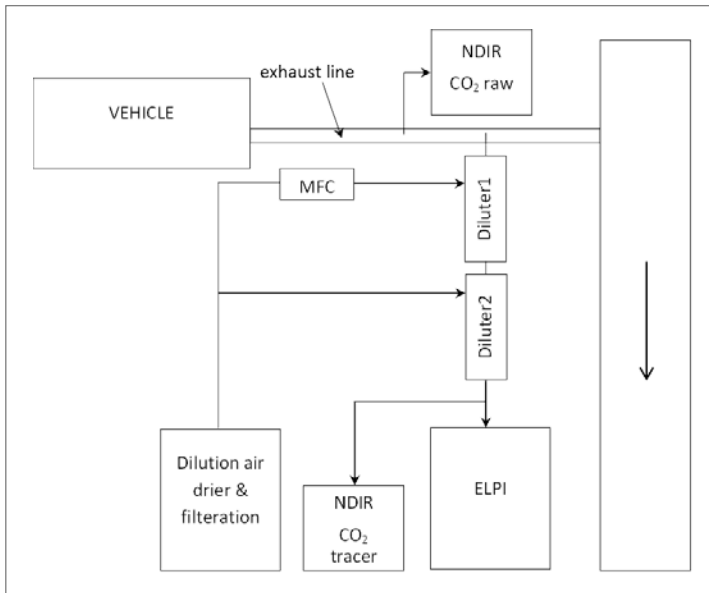


Figure 1. Block diagram of the particle measurement system at VTT Finland, European Test Program.

3. Research Results

3.1 Effect of E85 on Particle Emissions

3.1.1 Engine Test Results

As part of the North American Test Program, Argonne National Laboratory (ANL) analyzed total particulate number, size and morphology for several biofuel blends using an engine dynamometer and a 'dyno-ready' GM L850 Opel 2.2L 4 cylinder Ecotec engine.¹ Test fuels included gasoline (E0), E10 and E85 ethanol blends and isobutanol-16 (iB16). A Scanning Mobility Particle Sizer (SMPS) was used to measure engine particulate matter emissions at 25%, 50%, 75% load and at idle. ANL found that, regardless of fuel used, particulate numbers increased from idle to 50% load then decreased under the high load (75%) condition.

Particle number emission rates varied depending on the fuel used. Under all engine load conditions, the number emission rate on E85 was an order of magnitude lower when compared to the other test fuels. This is in keeping with sub-task 1 results that found the greatest emissions reduction benefits were with high level ethanol blends, as well as with the results from VTT Finland, and Environment Canada, as reported in the next sub-section.

ANL also found that E10 and iB16 produced higher total particulate numbers than did neat gasoline; a finding that ERMS has also observed in their Annex 35 Sub-task 2 research. This result was attributed to the properties of low level alcohol fuel blends, i.e., the higher viscosity and latent heat of vaporization and the reduced lower heating value. These properties affect fuel atomization and mixing, and result in additional fuel use to maintain engine torque. Total particulate number for E85 was 10-fold lower than for the other fuels, probably due to its significantly higher oxygen content.

¹ Lee *et al.*, 2012.

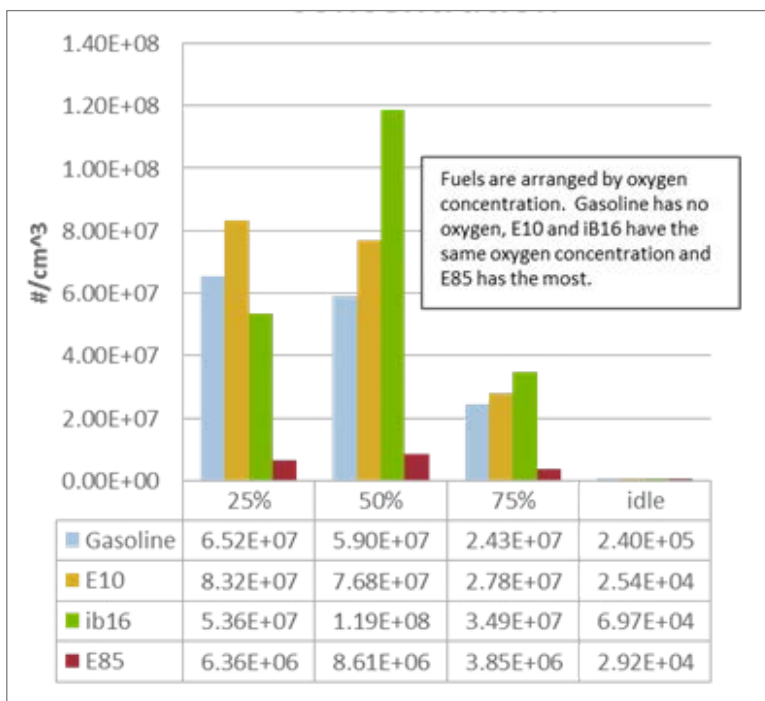


Figure 2. Total particulate concentration measured by SMPS analysis of emissions from a General Motors Ecotec engine run on gasoline and alcohol blended fuels. ANL, North American Test Program.

Figure 3 shows the particle size distributions for each fuel and engine loading condition. For the 25% and 50% load cases, E10 and iB16 produced particle size distributions peaking at a larger particle size compared to gasoline. At 75% load, size distributions peaked at approximately 25nm for all fuels.

Figure 4 summarizes the histogram for the primary particle diameter observed by transmission electron microscopy (TEM) and illustrates that the number of relatively larger particles increased in the sequence of gasoline, E10 and iB16. As a result, the average primary particle diameter increased from 31.0 nm for gasoline to 36.2 nm and 38.8 nm for E10 and iB16, respectively. It is speculated that the increased size of individual primary particles contributed to the formation of larger aggregate particles with those two biofuels. Primary particle diameter information for E85 are unavailable from the TEM analysis due to difficulty in measuring sizes from the sample at 75% load, because of the low total particle number density and dense agglomeration of particles.

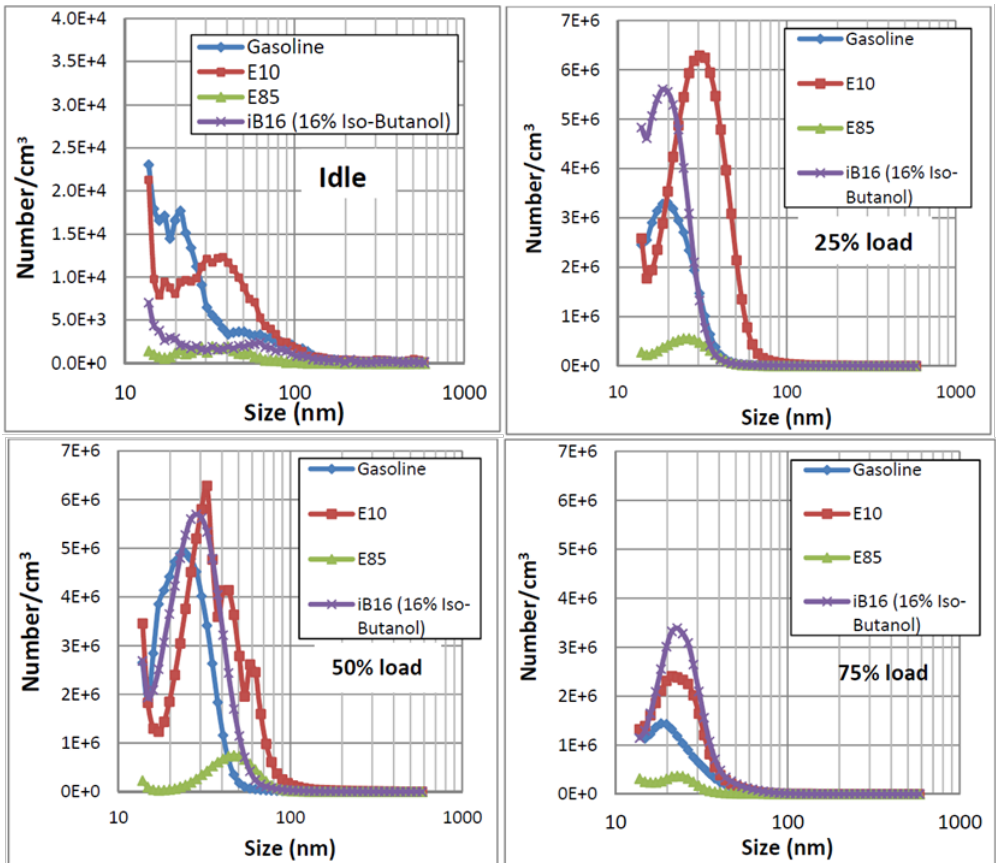


Figure 3. Particle number size distributions for an Ecotec engine operated on gasoline and alcohol blended fuels at idle and 25%, 50% and 75% engine load. ANL, North American Test Program.

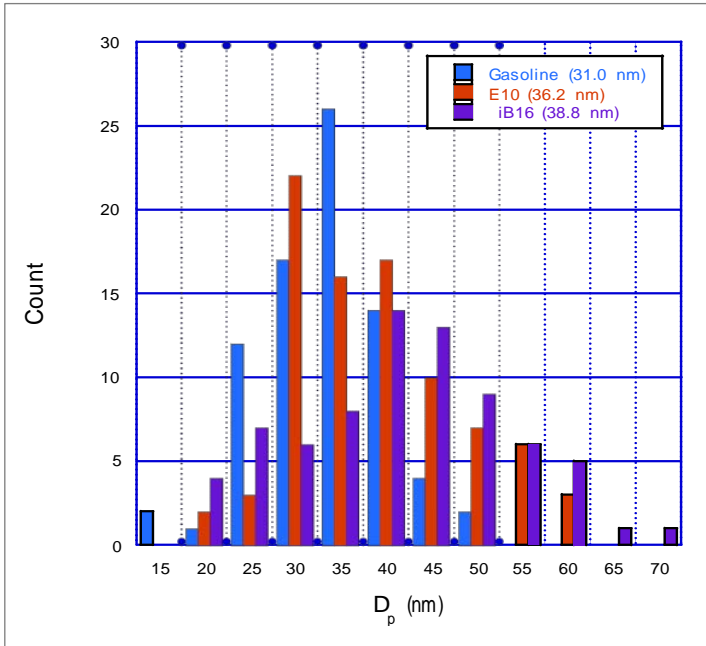


Figure 4. Histogram showing the primary particles diameter of the emitted particulate matter from an Ecotec engine operated on gasoline and alcohol blended fuels at 75% engine load. ANL, North American Test Program.

3.1.2 Chassis Test Results

To determine how different fuels and test temperatures affect particulate emissions from a flexible fuel vehicle operated on E10 and E85 fuels, VTT in Finland tested a typical light duty European GDI vehicle using the (NEDC) at 23°C and -7°C. Particle size distributions were measured using an ELPI and, as illustrated in Figure 5, the test results show that at both temperatures E85 produced a significantly lower number of particles in each size class. At 23°C the number of particles was roughly an order of magnitude lower with E85 as compared to E10. At -7°C the difference between fuels is smaller but still significant.

The shape of the distribution curve also differs between the fuels at both temperatures with the distribution peak occurring at a smaller particle size with E85 than with E10. When temperature decreases the peak of the distribution shifts slightly to a larger particle diameter for both fuels. With E10 the peak of the distribution shifts from approximately 40 nm to 80 nm and with E85 from approximately 20 to 40 nm.

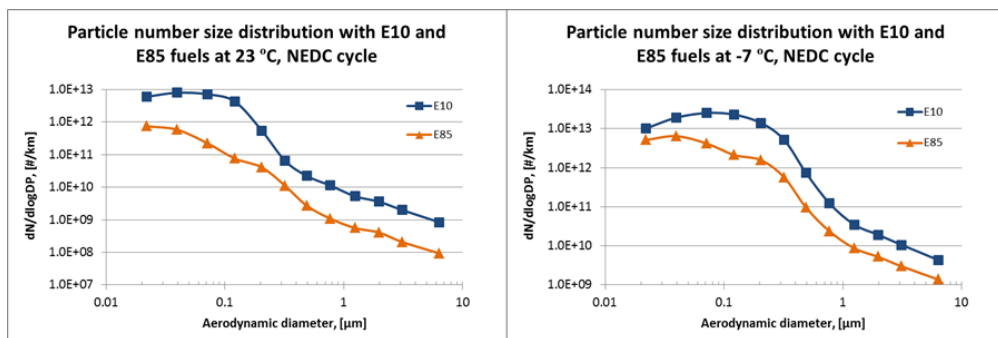


Figure 5. Particle number size distributions at 23°C and at -7°C for a European Turbocharged GDI flexible fuel vehicle operated on E10 and E85 fuels over the NEDC. VTT, European Test Program.

At Environment Canada, particle number size distributions from a GDI flex-fuel vehicle were measured using an EEPS. Figure 6 illustrates the FTP-75 results from the vehicle tested at 22°C and -18°C. Results from the NEDC are also provided for E0 and E85 at -18°C. Note the particle number is presented in particles per mile for the three FTP-75 graphs and is presented in particles per kilometre for the NEDC graph.

As indicated in Figure 6, the use of E85 significantly reduced particle number emissions as compared to E0 and E10. Along with the reduction in particle number, for the FTP-75, there was a shift to a lower primary peak diameter, from 70-80 nm to as low as 34 nm at 22°C. With the NEDC test there was also a shift from 80 nm with E0 to 65 nm with E85. The smaller particle size observed with use of E85 corresponds to the VTT results described above. This reduction in particle number emissions with the use of mid to high level ethanol blends has been reported elsewhere and is thought to be owing in part to the lower sooting tendency of ethanol due to increased oxidation from higher levels of oxygen and to a reduced quantity of aromatics present in the blended fuel.²

Over the FTP-75, for both fuels, the cold temperature generally resulted in a shift to a slightly larger particle diameter, i.e., when temperature was reduced from 22°C to -18°C peak diameter increased from 70 nm to 80 nm for E0 and from 35 nm to 50 nm for E85.

² Mamakos and Manfredi, 2012, Szybist *et al.*, 2011 and Storey *et al.*, 2010.

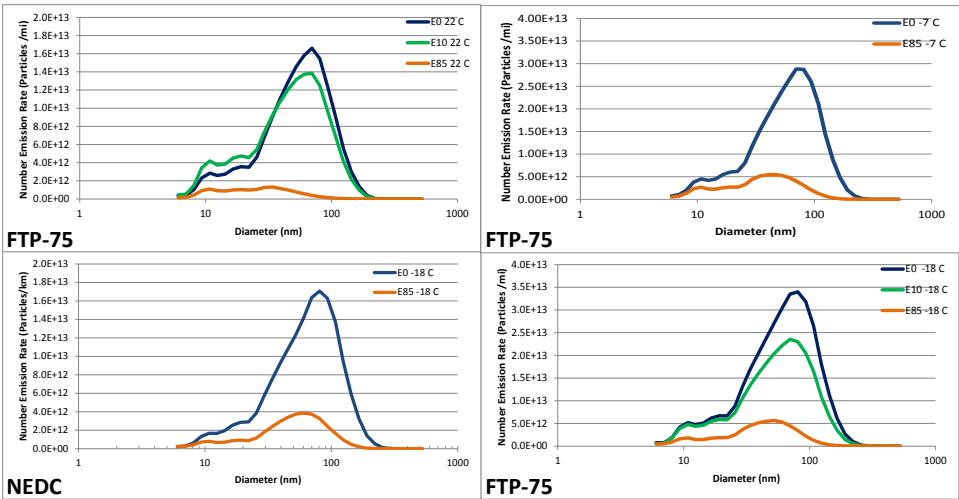


Figure 6. Average particle number size distributions for a GDI engine flex fuel vehicle operated using E0, E10 and E85 fuels over the FTP-75 and NEDC drive cycles at different ambient temperatures. ERMS, North American Test Program.

Figure 7 provides a typical time series of particle number concentration in the exhaust stream recorded by VTT over the NEDC drive cycle at 23°C and -7°C. As expected, particle concentrations were highest at the beginning of the cycle due to the cold-start. After the engine and catalyst had warmed up, the particle number concentration decreased. Particle concentration with E85 was lower at each point of the test cycle as compared to E10. At a lower temperature of -7°C, the particle concentrations were much higher at the beginning of the test with both fuels. By the end of the driving cycle the ambient temperature makes no difference for E10. The E85 particle concentrations are higher at the lower temperature even once the vehicle is well warmed up.

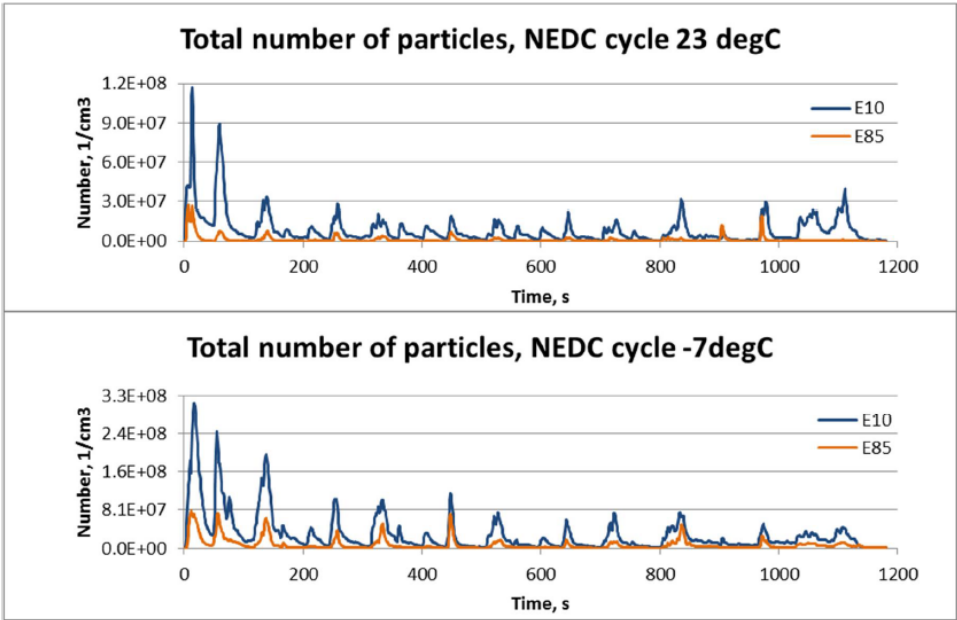


Figure 7. Particle number concentration measured by VTT at different temperature conditions for the GDI vehicle driven over the NEDC cycle. European Test Program.

Figure 8 provides the time series of the particle number concentration measured in a dilution tunnel by the EEPS recorded over the FTP-75 and NEDC drive cycles at -18°C . Each individual data point on the time series presented in the figure was obtained by integrating all individual size bins in the particle number size distribution at that specific time interval. Consistent with the VTT results, the EC data also indicates that a significant number of particles were emitted during the first phase of the test cycle. Once the engine and catalyst had warmed up, the particle emissions during the following three ECE-15 cycles became repeatable. With the EC data, during the cold start of the NEDC the E85 concentration was similar to that of E0, however, the particle concentrations with E85 were markedly lower compared to E0 following the cold start. During the initial cold start phase of the FTP-75 reductions in particles with the use of E85 were noted.

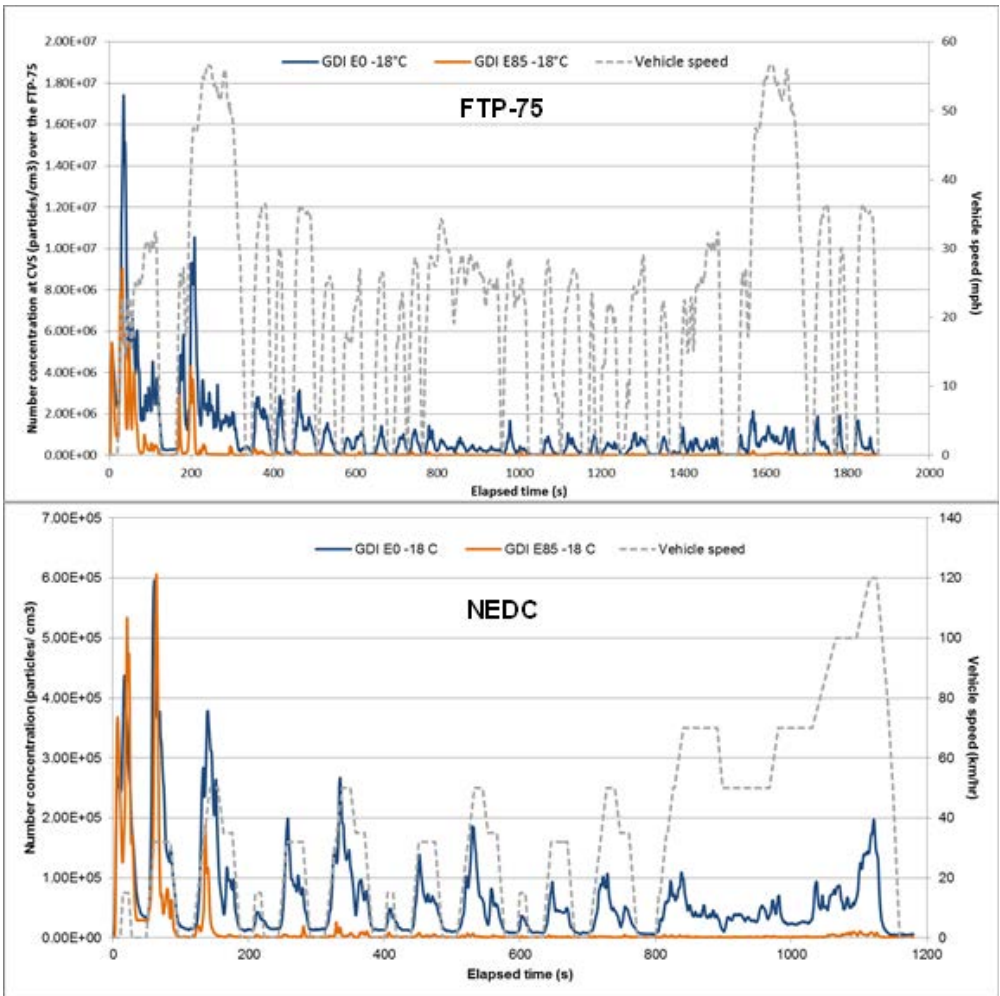


Figure 8. Particle number concentration time series measured in diluted exhaust by EC at -18°C from a GDI vehicle over the FTP-75 and NEDC drive cycles. ERMS, North American Test Program.

3.2 Effect of Low- to Mid-Level Ethanol Blends on Particle Emissions

Figure 9 shows the average particle number size distributions for a GDI vehicle tested at Environment Canada over the FTP-75 and the US06 drive cycles. The vehicle was fueled with splash blends of E10, E15 and E20, as well as E0.

The effect of low- to mid-level ethanol blends on particle emissions is variable for the two drive cycles. On the FTP-75 drive cycle, increasing ethanol content increased particle emissions for the E10 and E15 cases and decreased emissions for the E20 fuel. In the case of the US06 drive cycle, emissions increased with increasing ethanol content, however, fewer particles were emitted over the US06 drive cycle compared to the FTP-75 drive cycle.

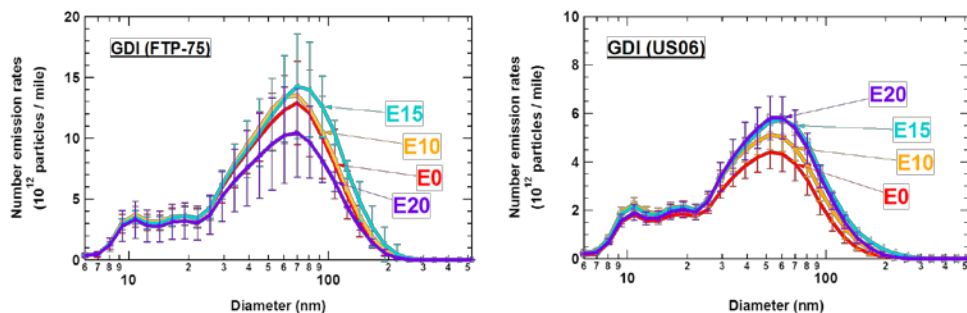


Figure 9. Average particle number size distributions for a GDI vehicle running the FTP-75 and US06 test cycles at 22°C. ERMS, North American Test Program.

Also under the North American collaborative program, the Canadian Auto 21 Network funded a study conducted by the Universities of Alberta, British Columbia and Toronto, to examine exhaust from a 2.0 Litre, 4 cylinder, GDI engine fueled with gasoline (E0) and ethanol blended fuel (E10 and E30). Exhaust was passed through a two-stage diluter (TSI 379020A) and a thermodenuder heated to 200°C. SMPS scans were conducted with a 3081 series DMA and a 3776 series CPC. Additionally, a CPMA was placed between a DMA and CPC and CPMA scans were performed with the DMA set to constant particle diameter over a particle size range of 30 nm to 130 nm. Test conditions included cold and hot (120°C) starts, a simulated highway cruise and a high-speed low-torque operating condition. As illustrated in Figure 10, no notable change in particle effective density was noted from with the use of the ethanol blends.³

³ Graves *et al.*, 2013.

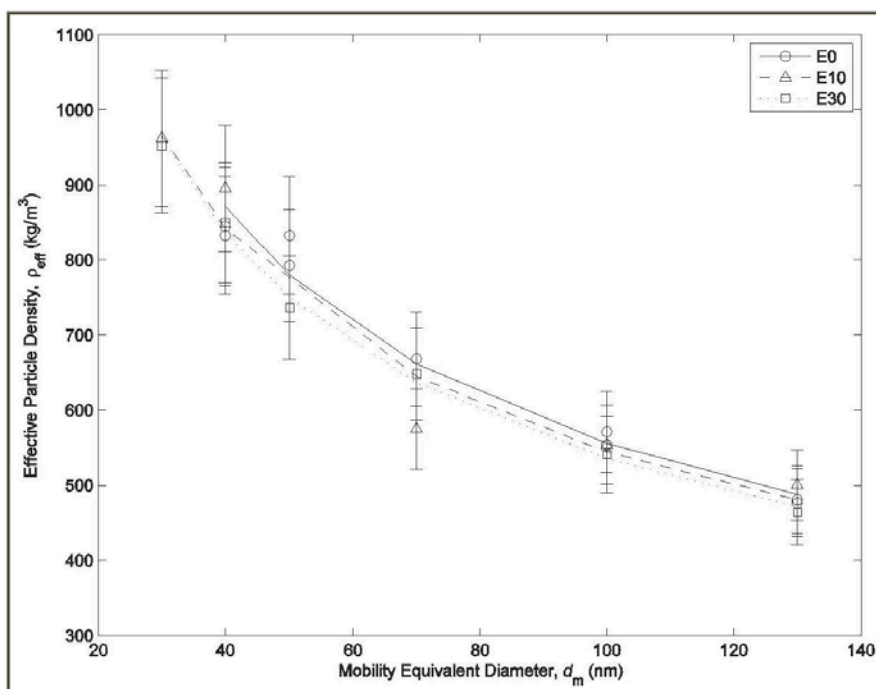


Figure 10. Comparison of effective particle density as a function of mobility equivalent diameter for E0, E10, and E30 at a high speed, low torque operating condition, with denuding. [Graves et al., 2013].

3.3 Effect of Isobutanol on Particle Emissions

Figure 11 illustrates the average particle number size distributions measured for the GDI vehicle using splash blended isobutanol fuel at 16% (iB16) and gasoline (iB0). Particle number emissions over the US06 drive cycle were lower than what was emitted over the FTP-75 drive cycle due to the absence of a cold start as illustrated by the different scales used in Figure 11.

The average particle diameter for the FTP-75, iB0, standard temperature condition appeared to be slightly smaller (by 10 nm) than the average particle diameter for iB16 at standard temperature, which was comparable to the observed average particle diameters for both fuels at -18°C. Over the FTP-75 drive cycle, iB16 had limited impact on particle emissions from the GDI vehicle at standard temperature and led to slightly lower particle emissions at -18°C.

For the US06, iB16 was observed to reduce particle emissions at both test temperatures. The US06 also exhibited a more obvious bi-modal distribution and relatively more nucleation mode particles.

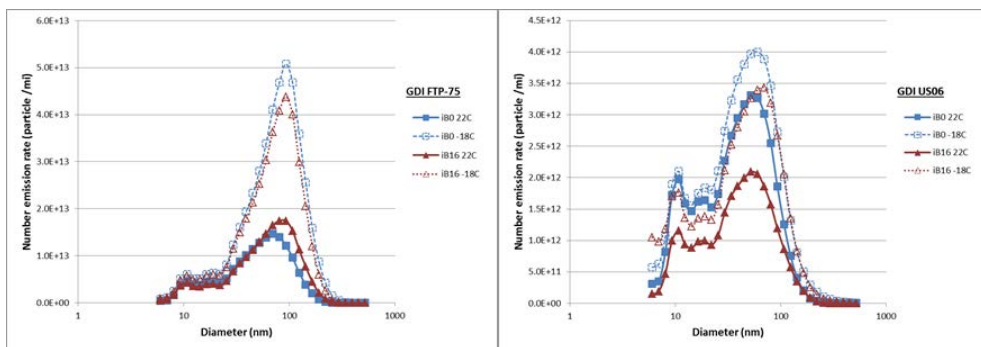


Figure 11. Average particle number size distribution. ERMS, North American Test Program.

3.4 Particle Morphology

Under the North American collaborative program, the Canadian Auto 21 Network group consisting of the Universities of Alberta, British Columbia and Toronto, conducted a study of soot particle morphology using a Transmission Electron Microscope (TEM) and exhaust from a 2.0 Litre, 4 cylinder, GDI engine fueled with gasoline (E0) and ethanol blended fuel. Test conditions included both cold and hot starts, a simulated highway cruise condition and a high-speed low-torque operating condition.

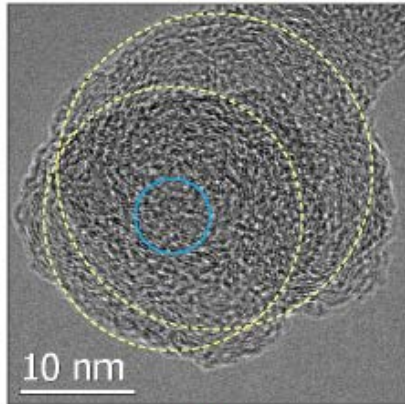
Table 2 summarizes the TEM imaging processing results. Numbers in parenthesis are one standard deviation of the measured parameter. In the table, dP, primary particle diameter, is the diameter of the nearly spherical monomers that make up the soot particles. Soot properties such as light scattering and absorption are directly related to dP (note that light absorption and scattering show additional dependence on the 3rd and 6th powers of dP respectively). Soot mass and surface area and soot mass are proportional to the 2nd and 3rd powers of dP respectively. The projected area equivalent diameter, dA, of the aggregate is the diameter of a sphere with the same projected area as the aggregate. It is equal to the mobility diameter of particles in the free molecular flow regime. Np is number of the primary particles in each aggregate, i.e., the number of monomers from which the aggregates are formed. Finally, Rg is the radius of gyration of the aggregate. In three dimensions it is defined by the means square of the distances between the center of the primary particle and the center of mass of the aggregate. In two dimensions it is defined by the mean square of the differential elements of the projected area and the center of area of the projected images. Rg is an important parameter in determination of the fractal structure of the aggregates.

The results in Table 2 show that for the Highway cruise mode, the primary particle diameter is smaller for the E30 fuel compared to the E0 fuel. No such fuel effect was seen for the cold start samples.

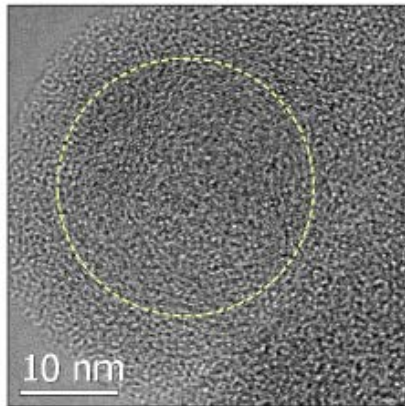
Table 2. TEM Imaging Processing Results [Graves et al., 2013]

	Mean d_p (nm)	Median d_p (nm)	Mean d_A (nm) $N_{p \geq 1}$	Median d_A (nm) $N_{p \geq 1}$	R_g (nm) $N_{p > 1}$
E0-Highway Cruise-Denuded	25 (± 10)	23	107 (± 59)	88	57 (± 37)
E30-Highway Cruise-Denuded	18 (± 5)	17	78 (± 57)	67	44 (± 35)
E0-Cold Start-1 st 15 s-Denuded	28 (± 11)	27	118 (± 89)	102	79 (± 60)
E30-Cold Start-1 st 15 s-Denuded	32 (± 12)	31	98 (± 99)	77	75 (± 64)

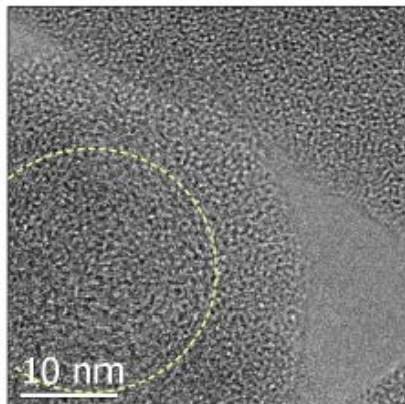
Also under the North American Test Program, Argonne National Laboratory (ANL) visually analyzed particles emitted from E0, E10 and iB16 fuel using a TEM and a magnification of 600,000 times. Based on TEM observation, the number concentrations of particulates were extremely low at 25% and 50% engine loads, particularly with the biofuel blends, therefore, analysis was performed only for the 75% engine load. With the engine operating at a 2000 rpm and 75% load condition, the gasoline fuel produced well defined particle nanostructures or graphite structures where the concentric fringes were clearly observed (Figure 12a). The yellow dotted circles represent the approximate outer bounds of individual primary particles, in which numerous circular fringes are arranged concentrically. For each primary particle, a nucleus, indicated by the blue solid circle in Figure 12a, usually locates near the center of the particle. In this TEM image, several primary particles seem to be overlapped with interference from each other. In contrast, soot particles from iB16 and E10 exhibited a lower degree of graphitic structures (Figures 12b and 12c). Because the particles emitted from the two biofuel blends exhibited surface growth with the input of thermal energy from the TEM electron beams, it was inferred that these particles contained a higher concentration of volatile organic compounds, as compared to those from E0 fuel.



(a)



(b)



(c)

Figure 12. Particle nanostructures from combustion of (a) E0, (b) E10 and (c) iB16 at a 75% engine load. ANL, North American Test Program.

3.5 Gasoline Particulate Filters and Particle Emissions

Future GDI vehicles could adopt multiple strategies (e.g., with the use of both oxygenated fuel and an emission control device) to control ultrafine particle emissions to meet the more stringent future particulate matter emission standard. Through a separate program the Emissions Research and Measurement Section of Environment Canada conducted a program to evaluate the particle emissions from two GDI vehicles with the use of both gasoline particulate filters (GPF) and 10% volume blended ethanol (E10).⁴

In this program, two custom designed, wall-flow, passive regenerating GPFs were tested. The catalyzed GPF was installed on a compact GDI vehicle while the non-catalyzed filter was installed on a mid-size GDI sedan. Both GPFs were optimized according to the specification of the vehicle and did not increase back pressure over the FTP-75 and US06 drive cycles. E10 had limited impact on particle emissions from the two GDI vehicles except during the cold start phase of the FTP-75 drive cycle when considerable reduction in black carbon (BC) mass by 52-66% from both vehicles at standard temperature were observed. When the two vehicles were equipped with the GPF, further reductions in BC mass and particle number emissions were observed. Gasoline engines typically have much lower soot emissions than diesel engines, this turns out to be a key factor on the overall particle filtration efficiency of the GPF. Typical engine exhaust temperatures of both GDI vehicles over the FTP-75 drive cycle were not high enough to trigger continued soot regeneration. This permits the soot cake to develop over the course of the FTP-75 drive cycle even starting with an empty filter. Overall particle filtration efficiency for solid particles for both the catalyzed and non-catalyzed GPF were typically in the 80% range at standard temperature. Over the more aggressive US06 drive cycle, exhaust temperature was high enough to trigger multiple soot regeneration in the non-catalyzed GPF even at standard temperature. The emission of ultrafine particles during the soot regeneration reduced the filtration efficiency for particle number but less of an effect on BC mass filtration efficiency. In comparison, the soot regeneration from the catalyzed GPF over the US06 drive cycle was much more severe and continued for an extended period of time, during which a significant number of ultrafine particles, almost completely volatile in nature, were emitted downstream of the GPF.

The similar and different observations from these tests revealed the usefulness of a GPF as one strategy for reducing fine particle emissions in congested urban area. At the same time, observations from this study also suggested the need for continued investigation and improvement for filter technology deployment in gasoline vehicles.

⁴ Chan *et al.*, 2012, Chan *et al.*, 2013 and Chan *et al.*, 2014.

4. Summary and Links to Other Annexes

4.1 Summary

Alcohol blended fuels offer a means to reduce emissions of ultrafine particles and particulate matter mass while meeting increasingly stringent renewable fuel mandates. Gasoline direct fuel injection (GDI) technology provides an alternative means to power gasoline vehicles and improve fuel economy, but has been linked to increased emissions of particulate matter. In this international collaborative research program, dynamometer testing was used to conduct measurements of particle number emissions and size distributions in exhaust from GDI vehicles and engines. The vehicles and engines were fuelled with 10%, 15%, 20%, 30% and 85% ethanol as well as a 16% isobutanol blend. Effects of cold ambient temperature, driving cycle, and engine speed/load were investigated. Transmission electron microscopy was used to conduct an assessment of particle morphology. The findings included in this report are from the North American and European Test Programs. Related research conducted under the Asian Test Program will be available through the final reporting for Annex 44.

The results of this research indicate the use of E85 as a transportation fuel as a means to mitigate PM increases from GDI vehicles. Research conducted under the North American and European Test Programs (ANL-U.S.A., EC-Canada, VTT-Finland) shows that a significant reduction (up to 90%) in particulate emissions from GDI vehicles and engines can be realized with the use of E85. This trend was observed over both FTP-75 and NEDC drive cycles (vehicle testing), and at several different engine loading conditions (engine testing). There was also a shift to a lower primary peak diameter, from 70-80 nm to ~ 30-40 nm at 22°C, due to the use of E85. A shift to a smaller particle size diameter with E85 was also observed at cold ambient temperatures, and over both FTP-75 and NEDC drive cycles. Although the two drive cycles were designed to meet regionally specific driving conditions, the trends in particle emission changes with E85 compared to E10 are similar with both the NEDC and the FTP-75 drive cycle.

The use of low- to mid-level ethanol blends in GDI vehicles and engines gave mixed results. The ANL-U.S.A. study showed increased particle number concentrations in GDI engine exhaust with the use of E10, while EC-Canada observed lower emissions with E20 and higher emissions with E10 and E15 for a GDI vehicle operating on the FTP-75 cycle. Over the US06 cycle, increased PM emissions with increasing ethanol content (up to E20) were consistently observed. In the EC-Canada study, the use of various ethanol blend levels (E10 and E15) resulted in limited changes on the shape of the particle number size distributions.

The focus of this study was the impacts of alcohol blends on particulate emissions, however, it should be acknowledged that alcohol fuels may also impact gaseous emissions. In a study of 2000 to 2003 model year vehicles fuelled with E10, Graham showed decreases in carbon monoxide (CO), and increases in emissions of non-methane hydrocarbons (NMHC), acetaldehyde and benzene and no statistically significant changes in oxides of nitrogen (NO_x), compared to E0.¹ With two model year 2012 GDI vehicles, Karavalakis² showed that when comparing E15 and E20 to E10 there were some changes in CO, NMHC and NO_x emissions however, emissions were found to be relatively low for these newer vehicles.

Graham et al. also showed with a 2004 flex fuelled vehicle, the use of E85 significantly decreased emissions of NO_x, NMHC, 1,3-butadiene and benzene, with no statistical changes to CO and CO₂ and NMOG. Nevertheless, increases in emissions of formaldehyde and acetaldehyde were noted with E85.³

In this limited study the use of isobutanol blend fuel (iB16) also produced mixed results. EC-Canada observed reductions in particle number emissions for a GDI vehicle with the use of iB16 on the FTP-75 cycle at -18°C and on the US06 cycle at both standard and cold temperature. Conversely, in the study by ANL-U.S.A., the total particle number emission rate increased for a GDI engine with the use of iB16 relative to neat gasoline. When this research was started, little work had been undertaken on assessing the impact of using isobutanol as an alternative renewable fuel for on-road transportation. Since that time, the investigation of particle and gaseous emissions from isobutanol blends has been ongoing in several different countries. Recent examples of this research include work done in the Aakko-Saksa and Karavalakis laboratories.⁴ It should be noted that the use of butanol blended fuel may also change the carbonyl compound profile compared to gasoline without alcohol and to gasoline blended with ethanol fuel.⁵

Through steady state engine testing, the particle number concentrations were extremely low with E85 compared to E0, E10 and iB16. This agrees well with the low particle number counts observed with E85 from the chassis vehicle emissions testing conducted in both Canada and Finland. In the United States program, TEM analysis and size measurement of E85 particulate was difficult due to low total numbers. However, visual observation showed structure changes with E10 and iB16 compared to E0.

Lastly, the use of prototype gasoline particulate filters (GPF) was shown to substantially reduce particle number emissions from GDI vehicle exhaust regardless of the ethanol fuel blend level.

¹ Graham *et al.*, 2008.

² Karavalakis *et al.*, 2014.

³ Graham *et al.*, 2008.

⁴ Karavalakis *et al.*, 2013 and 2014 and Aakko-Saksa *et al.*, 2011.

⁵ Karavalakis *et al.*, 2014.

4.2 Links to Other Annexes

In addition to supporting Annex 35 Sub-task 2, the sub-task consortium also contributes research results to Annex 43 and Annex 44 initiatives.

4.2.1 Annex 43

Annex 43: Performance Evaluation of Passenger Car, Fuel, and Power Plant Options is an international collaboration between Canada, China, Finland, Japan, Sweden and the United States. The VTT Technical Research Centre in Finland is Operating Agent for the Annex. The objective of the research is to develop benchmark data on a variety of vehicle makes and models regarding fuel efficiency, engine efficiency and tailpipe emissions, with an emphasis on the differences between alternative engine technologies, to enable the comparison and development of different fuel options. Fuels used within this Annex include low- and mid-level ethanol blends.

4.2.2 Annex 44

The China Automotive Technology and Research Center (CATARC) is the Operating Agent for Annex 44: Research on Unregulated Pollutant Emissions of Vehicles Fueled with Alcohol Alternative Fuels. Participating countries are Canada, China, Finland, Israel, Sweden and Switzerland. The objective of this research is to examine how unregulated pollutants from vehicles are influenced by measurement methods, automotive technology, alcohol content, ambient temperature, test cycle and other test parameters in order to establish measurement methods and limits of unregulated pollutants. Fuels used include low- to mid-level ethanol and E85.

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Appendix A: Supplementary Technical Information

Table A-1. ERMS Test Vehicles Technical Data. North American Test Program.

Vehicle Description	GDI Engine	Flex-fuel Vehicle (FFV) GDI Engine
Model Year	2011	2012
Engine	2.4L Wall Guided GDI I4 DOHC	2.0L Ti-VCT Wall Guided Spark Ignition Direct Injection I4
Power Output (HP)	198	160
Transmission Type and # of Gears	6-Speed Automatic	5-Speed Manual
Starting Odometer Reading (km)	3200	n/a
Emission Standard	Tier 2 Bin 5, ULEV	Tier 2 Bin 4 LDV, LEV-II ULEV
Emission Control	DFI/HO2S(2)/TWC	TWC/HO2S/HAFS/DFI
Test Weight	1590 kg (3500 lbs)	1474 kg (3250 lbs)

Table A-2. VTT Finland Test Vehicles Technical Data. European Test Program.

Model Year	2011
Engine	Turbocharged direct-injection spark ignition engine
Engine Displacement	1390 cm ³
Engine Power (kW)	155
Transmission Type and # of Gears	7-gear dual-clutch automatic transmission
Starting Odometer Reading (km)	6523
Emission Standard	Euro 5a
Inertia Weight	1615 kg

Test Fuels and Analysis

For Canada's contribution to the North American Test Program, the following test fuels were selected and splash blended for the low-to mid-level ethanol testing:

- E0/iB0 Certification gasoline (Tier2);
- E10 10% ethanol by volume (E10-Tier2);
- E15 15% ethanol by volume (E15-Tier2); and
- E20 20% ethanol by volume (E20-Tier2).

For the isobutanol testing, Canada splash blended by volume a reagent grade isobutanol with Tier 2 (iB0) fuel. See Tables A-3 to A-5 for test fuel analyses.

Table A-3. Test Fuel Analysis. ERMS, North American Test Program.

Fuel Component	Method	E0	E10	E15	E20
Carbon, %wt	ASTM D5291	86.21	83.05	81.47	79.89
Hydrogen, %wt	ASTM D5291	13.61	13.58	13.56	13.55
Nitrogen, %wt	ASTM D5291	<0.15	<0.15	<0.15	<0.15
Density, kg/m ³ @ 15 °C	ASTM D4052	743.0	748.0	750.5	753.0
Oxygen	In-house Method	<0.1/0	3.04	4.56	6.08
Specific Gravity 60/60F GRAV	ASTM D4052	0.7440	0.7489	0.7515	0.7540
Sulphur, mg/kg	ASTM D7039	32.	29.19	27.79	26.38

The fuels specifications used for the emissions testing by ANL under the North American Test Program are provided in Tables A-6 and A-7.

Table A-4. Test Fuel Analysis of gasoline and isobutanol used for preparing iB16 a 16% by volume splash blend of Tier 2 gasoline and butanol (iB16-Tier2). ERMS, North American Test Program.

Fuel Parameter	Method	iB0	iB16	iB100
Carbon, %wt	ASTM D5291	86.5	83.1	64.6
Hydrogen, %wt	ASTM D5291	13.4	13.4	13.4
Isobutanol	CAN/CGSB-3.0 No. 14.3	-	15.77	99.86
Density, kg/m ³ @ 15 °C	ASTM D4052	745.3	754.3	805.5
Oxygen, %wt		0	3.63	21.57
Vapour Pressure psi (kPa)	ASTM D5191	8.6 (59.3)	7.9 (54.5)	<1.02 (<6.9)
Gross Heat of Combustion @ 25°C MJ/kg	ASTM D4809	47.6	46.3	38.3
Net Heat of Combustion, MJ/kg		45.0	43.7	35.6
Aromatics, Volume %	ASTM D1319	28.8	27.3	-
Olefins, Volume %		0.8	0.8	-
Saturates, Volume %		70.4	56.2	-

Table A-5. Test Fuel Analysis of gasoline and ethanol blends used in the flex fuelled vehicle. ERMS, North American Test Program.

Fuel Identification	Method	E0	E10	E85 [†]	E85 ^{††}
Carbon, %wt	ASTM D5291	86.31	82.40	57.60	59.49
Hydrogen, %wt	ASTM D5291	13.34	12.99	13.14	13.29
Density, kg/m ³ @ 15 °C	ASTM D4052	743.0	748.3	779.3	769.9
Oxygen, %wt		0	3.03	29.26	27.22
Specific Gravity °API	ASTM D4052	58.69	57.42	49.90	52.13
Specific Gravity 60/60F GRAV		0.744	0.749	0.780	0.771
Reid vapour pressure kPa	ASTM D5191	62.7	68.7	45.0	78.0
Ethanol volume %	ASTM D5501	0	9.8	82	75

[†] E85 used for 22°C tests

^{††} E85 used for -7°C and -18°C tests

Table A-6. Base Fuel Analysis. ANL, North American Test Program.

Base Fuel Features	Gasoline	Ethanol	Isobutanol
Composition (C, H, O) (%)	86, 14, 0	52, 13, 35	65, 13.5, 21.5
Lower heating value(MJ/kg) (LHV)	42.7	26.8	33.1
Density (kg/m ³)	715 – 765	790	801.8
Octane number ((R+M)/2)	86 – 90	100	103.5
Boiling temperature (°C)	25 – 215	78	108
Latent heat of vaporization (25°C) (kJ/kg)	380 – 500	904	716
Viscosity (cSt at 20°C)	0.4 – 0.8	1.52	3.64
Surface Tension (dynes/cm at 20°C)	20	22.27	23
Self-ignition temperature (°C)	~300	420	343
Stoichiometric air/fuel ratio	14.7	9	11.2
Laminar flame speed (cm/s) I, II	~33	~39	
Mixture calorific value (MJ/m ³) II	3.75	3.85	3.82
Ignition limits in air (Vol-%), Lower limit – Upper limit	0.6 – 8	3.5 – 15	1.2 – 10.9

Table A-7. Test Fuel Analysis. ANL, North American Test Program.

Fuel Specific Features	Gasoline	E10	iB16	E85
Composition (C, H, O) (%)	86, 14, 0	82, 13.5, 4.5	83.3, 13.5, 4.5	56.9, 13.1, 30.0
Lower heating value (MJ/kg)	42.7	41.2	41.8	29.1
Density (kg/m ³)	742	747	749	783
Energy Density (MJ/l)	31.8	30.7	31.3	22.8
Stoichiometric air/fuel ratio	14.7	14.1	14.3	9.8

In Finland (European program), 95 octane (RON) gasoline with 10 % ethanol content (E10) is the most commonly used gasoline in spark ignition passenger cars. High concentration ethanol fuel with 85 % ethanol content (E85) has been on the Finnish market since 2009 and was also tested. Basic information on test fuels used in VTT Finland’s research is as follows:

Table A-8. Test Fuel Analysis. VTT Finland, European Test Program.

Fuel Specific Features	E10	E85
Density (kg/m ³)	746.3	784.9
Ethanol content (vol-%)	9.29	85.7
Vapor pressure (kPa)	83.2	50.7
Net calorific value (MJ/l)	31.081	22.715

Drive Cycles

The New European Driving Cycle (NEDC) is designed to assess vehicle emissions and fuel economy of cars driven under typical European driving conditions. Consisting of four repeated Urban Driving Cycles (ECE-15) and an Extra-Urban Driving Cycle (EUDC), the 1200-second NEDC is performed with a cold vehicle and ancillary loads (fan, lights, etc.) turned off.¹ The ECE-15 portion of the test takes 780 seconds (13 minutes) to complete and is characterized by low engine load, low exhaust gas temperature and a maximum speed of 50 km/hr. It is followed by a 20-second stop and the EUDC, which takes 400 seconds (6 minutes, 40 seconds) to cover theoretical 6956 meters at an average speed of 62.6 km/hr.

¹ http://www.unece.org/trans/main/wp29/meeting_docs_wp29.html.

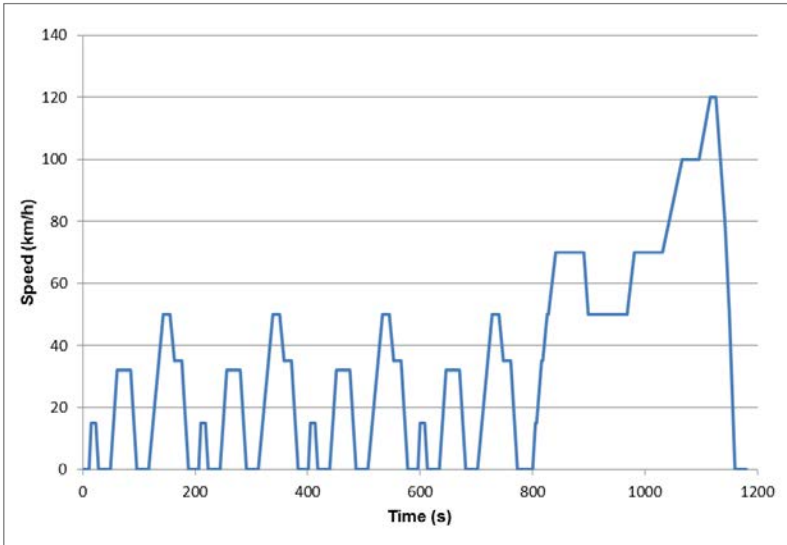


Figure 13. Time trace for New European Drive Cycle (NEDC). North American and European Test Programs.

The City Test, known as the Federal Test Procedure (FTP) or FTP-75 replicates a typical city driving pattern and consists of three distinct segments: a cold start phase, a stabilized phase (i.e., the vehicle is warmed up) and a hot start phase.

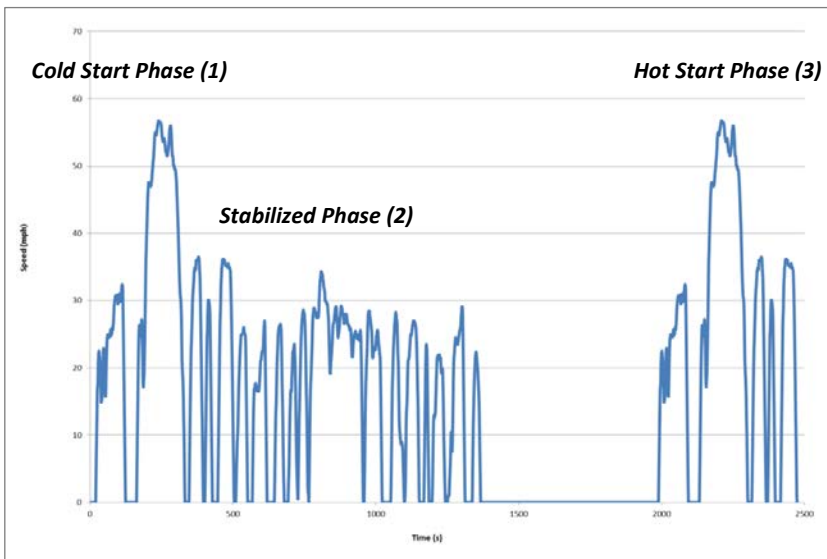


Figure 14. Time trace for Federal Test Procedure (FTP-75). North American Test Program.

The first two phases of the test are known as the LA4; a cycle that was developed by the U.S. EPA in the late 1960s and early 1970s based on the “Los Angeles Route Four”, a trip to and from the then headquarters of the Air Resources Board in Downtown Los Angeles. This portion of the test is also referred to as the Urban Dynamometer Driving Schedule (UDDS) for light-duty vehicles and light-duty trucks, as described in Appendix I (a) of 40 CFR 86. The LA4 driving schedule simulates a 12-kilometre (7.45 mile) stop-and-go trip with an average speed of 32 km/h (20 mph) and a top speed of 91 km/h (57 mph). It runs for 23 minutes and includes 18 stops. Approximately 4 minutes of the driving schedule are spent idling to represent waiting at traffic lights. This portion of the test begins from a cold engine start, which is similar to starting a vehicle after it has been parked overnight. After a 10-minute soaking period, the first 8 minutes of the driving schedule are repeated (phase 3), only this time with a hot engine start. This simulates restarting a vehicle after it has been warmed up, driven, and then stopped for a short time.

The US06 Supplemental Federal Test Procedure (SFTP) was designed to simulate aggressive highway driving for vehicles certified over the FTP test-cycle. It consists of a 10-minute, 13-kilometre (8 mile) route with an average speed of 78 km/h (48 mph) and a maximum speed of 129 km/h (80 mph). The US06 driving schedule for light-duty vehicles and light-duty trucks is described in Appendix I (g) of 40 CFR 86. Prior to the test cycle, a warm-up US06 test cycle is driven so that this test is performed with a hot engine.

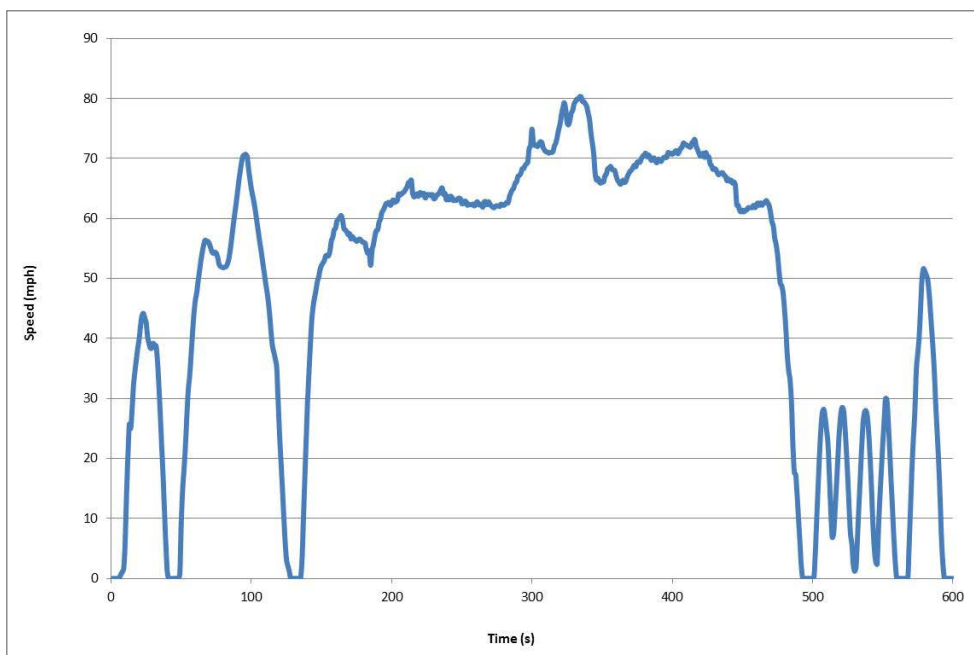


Figure 15. Time trace for Supplemental Federal Test Procedure (SFTP). North American Test Program.