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Particle speciation of emissions from iso-butanol and ethanol blended gasoline in light-duty vehicles

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ABSTRACT

Vehicular emissions of Particulate Matter (PM) will change under different driving conditions and as higher levels of alternative fuels are blended with gasoline. In particular, the chemical composition and water-soluble components of PM below 2.5 μm can be modified. In this study, three light-duty Port-Fuel Injection (PFI) and two Gasoline Direct Injection (GDI) vehicles were tested. All vehicles were tested over the Federal Test Procedure (FTP), Unified Cycle (UC), and at steady-state speeds (30, 50, and 70 mph) while operating on various ethanol and iso-butanol gasoline blends. The chemical and physical properties of the aerosol composition were measured via online/offline methods. Black carbon (BC), water-soluble organic carbon (WSOC), and droplet surface tension were measured. Water-Insoluble Mass (WIM) fractions were estimated from online size distribution measurements. Results show that the PFI vehicles, fuel composition and testing conditions (transient versus steady-state) impact PM emissions. GDI vehicles emit more PM and twice as much BC compared to PFI vehicles. Older PFI vehicles also produce more particles and BC emissions than newer vehicles. For three of the five vehicles, as speed increased, the WIM fraction increased. The results show vehicle operating conditions (steady-state or transient) can greatly impact the average composition of particles regardless of fuel composition.

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1. Introduction

Port-Fuel Injection (PFI) vehicles have represented the vast majority of the U.S. Light-Duty Vehicle (LDV) market for several decades. Gasoline Direct Injection (GDI) vehicles have recently garnered attention due to their increased fuel economy and reduced carbon dioxide (CO_2) emissions. These vehicles are predicted to dominate the U.S. market in the next few years (Graham, 2005; Zhao e al., 2006). For PFI systems, the fuel is injected into the intake ports outside the combustion chamber during the intake stroke as the air moves into the combustion chamber. GDI engines, on the other hand, use an injection system similar to that used in diesel engines, but at a much lower pressure (Alkidas, 2007). For the GDI engines, the fuel is injected directly into the combustion chamber during the intake stroke, which provides higher thermodynamic efficiency and lower greenhouse gas emissions. The drawback is that the direct injection of fuel can lead to liquid fuel

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wetting on the piston bowl and cylinder surfaces and fuel-rich zones during combustion, which will likely result in higher emissions of particulate matter (PM) (Stevens & Steeper, 2001; Piock et al., 2011). A number of studies have shown that GDI vehicles produce higher PM and Particle Number (PN) emissions compared to their PFI counterparts (Liang et al., 2013; Aakko & Nylund, 2003; Ristimaki et al., 2005; Mamakos et al., 2013; Szybist et al., 2011; and references therein). PM can affect regional air quality, human health, and climate (IPCC, 2007; Nauss, 1995; Andreae & Rosenfeld (2008), Andreae & Crutzen, 1997; Baumer et al., 2008; Avol et al., 1979; Twomey, 1977; and references therein). Particles below 2.5 μm, ($PM_{2.5}$) can be inhaled deep within the lungs and can be deposited in the nose and subsequent access to the brain (Nauss, 1995; Yang et al., 2008; Oberdorster, 2001; Oberdorster et al., 2004). Long-term exposure to $PM_{2.5}$ has been linked to cardiovascular and pulmonary diseases (Pope et al., 2004; Mills et al., 2009). Hence, the number and size distributions of ultrafine particles from modern GDI and PFI vehicular emissions need to be characterized.

$PM_{2.5}$ that contains water-soluble material has the ability to form droplets. Particles that promote water vapor condensation and form droplets are termed Cloud Condensation Nuclei (CCN). By behaving as CCN, inhaled hygroscopic particles can grow to droplet sizes and enhance harmful PM deposition rates (Longest et al., 2010). In addition to affecting health, CCN emissions of water-soluble particles affect climate by altering cloud droplet formation and cloud properties (Ervens et al., 2005). Water-soluble PM can be toxic when inhaled (Ramgolam et al., 2009; Gutiérrez-Castillo et al., 2006; Valavandidis et al., 2008), and water-soluble particles have the ability to induce DNA damage (Gutiérrez-Castillo et al., 2006). Water-soluble matter can cause pro-inflammatory response, thus promoting pulmonary and cardiovascular diseases (Ramgolan et al., 2009). Furthermore, water-soluble and water-insoluble organic materials (WSOC and WIOC) and metals from vehicular emissions are highly correlated with the cellular production of reactive oxygenated species (ROS) linked to toxicity and detrimental health effects (Cheung et al., 2009; Geller et al., 2006; Verma et al., 2010; Biswas et al., 2009).

Limited studies have characterized the water-insoluble/soluble composition of gasoline vehicle emissions. Cheung et al. (2009) reported Water-Soluble Organic Carbon (WSOC) and Water-Insoluble Organic Carbon (WIOC) emissions from a PFI vehicle fueled with gasoline. The WSOC and WIOC were each found to compose ~20% of the total PM mass. Small amounts of elemental carbon (EC) were also emitted in their study (Cheung et al., 2009). To the best of our knowledge, little to no studies have characterized the water-insoluble/soluble composition from light-duty PFI and GDI vehicles with different alcohol fuel formulations. The addition of oxygenated fuels, such as ethanol and iso-butanol, can alter the resulting particle chemical composition and hygroscopicity. Thus, the potential influence of emissions of gasoline blends with different alcohols is not completely understood, especially with respect to regional air quality, health, and climate.

Black carbon (BC) is another important component of the vehicular PM composition. Black carbonaceous materials (i.e., soot or EC) are considered a significant source of insoluble material. Limited studies have investigated the contribution of BC to particle emissions from light-duty vehicles, especially as a function of different fuel compositions (Forestieri et al., 2013). Dutcher et al. (2011) found a decreasing trend of BC concentrations using an aetholometer with increasing ethanol content in gasoline in a PFI engine on an engine dynamometer. Giechaskiel et al. (2010) reported a ~90% increase in BC concentrations with increased vehicle speed from 50 to 140 km/h.

The objectives of this study are to evaluate the emissions of PFI and GDI vehicles over a range of ethanol and iso-butanol blends with an emphasis on characterizing particle hygroscopcity. Five vehicles were tested on seven fuels over the Federal Test Procedure (FTP), the Unified Cycle (UC), and at steady-state speeds of 30, 50, and 70 mph. This study investigates how factors, such as fuel type and driving cycle influence particle hygroscopicity, BC, WSOC, surface tension, and water-insoluble fractions.

2. Experimental

2.1. Test vehicles, fuels, and driving cycles

Three light-duty PFI vehicles and two GDI vehicles were tested for this study. The PFI vehicles tested were a 2007 Honda Civic (PFI 1), 2007 Dodge Ram (PFI 2), and 2012 Toyota Camry (PFI 3). The GDI vehicles tested include the 2012 Kia Optima (GDI 1) and 2012 Chevrolet Impala (GDI 2). Vehicle specifications can be found in Karavalakis et al. (2014).

A total of seven fuels were employed in this study, including ethanol and iso-butanol blends. Each vehicle was tested on E10 (10% ethanol and 90% gasoline), E15, and E20 blends. In addition to the ethanol blends, iso-butanol blends were used including B16 (16% iso-butanol and 84% gasoline), B24, and B32. The butanol blends of B16, B24, and B32 were the oxygenated equivalent of E10, E15, and E20, respectively. An alcohol mixture comprising of 10% ethanol and 8% iso-butanol (E10/B8) was also used in this study. It should be noted that only the PFI 3 and the GDI 1 were tested on the E10/B8 fuel blend. The fuels were custom made to maintain Reid Vapor Pressure (RVP), oxygen content, and fuel volatility and other properties. The main fuel properties can be found elsewhere (Karavalakis et al., 2014).

Each vehicle was tested on each fuel over three FTPs and three UC tests. The speed versus time plots for each cycle is shown in Fig. S1. The 6 tests on a particular fuel were conducted sequentially once the vehicle was changed to operate on that fuel, and the fuel was not changed to another fuel during this time. A fuel change with multiple drain and fills was conducted between the testing on each fuel to condition the vehicle and ensure no carryover effects. Detailed information on the driving cycles employed in this study and the testing protocol can be found elsewhere (Karavalakis et al., 2013). After transient operation, the vehicles were driven at steady-state speeds of 30, 50, and 70 mph for each fuel. Thus, the

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