



# The effects of biodiesels on semivolatile and nonvolatile particulate matter emissions from a light-duty diesel engine<sup>☆</sup>



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## ARTICLE INFO

### Article history:

Received 13 March 2017

Received in revised form

1 June 2017

Accepted 5 June 2017

Available online 22 June 2017

### Keywords:

Biodiesel

Particulate matter emission

SVOCs

Dilution

Black carbon

## ABSTRACT

Semivolatile organic compounds (SVOCs) represent a dominant category of secondary organic aerosol precursors that are increasingly included in air quality models. In the present study, an experimental system was developed and applied to a light-duty diesel engine to determine the emission factors of particulate SVOCs (pSVOCs) and nonvolatile particulate matter (PM) components at dilution ratios representative of ambient conditions. The engine was tested under three steady-state operation modes, using ultra-low-sulfur diesel (ULSD), three types of pure biodiesels and their blends with ULSD. For ULSD, the contribution of pSVOCs to total particulate organic matter (POM) mass in the engine exhaust ranged between 21 and 85%. Evaporation of pSVOCs from the diesel particles during dilution led to decreases in the hydrogen to carbon ratio of POM and the PM number emission factor of the particles. Substituting biodiesels for ULSD could increase pSVOCs emissions but brought on large reductions in black carbon (BC) emissions. Among the biodiesels tested, tallow/used cooking oil (UCO) biodiesel showed advantages over soybean and canola biodiesels in terms of both pSVOCs and nonvolatile PM emissions. It is noteworthy that PM properties, such as particle size and BC mass fraction, differed substantially between emissions from conventional diesel and biodiesels.

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## 1. Introduction

Driven by energy security concerns and greenhouse gases (GHGs) emission mitigation targets, more than 50 countries have set goals to replace a substantial fraction of transport fuel with biofuels (mainly biodiesels and bioethanol), e.g., 10% in the European Union (EU) by 2020 (Directive 2009/28/EC, 2009) and 30% in the U.S. by 2025 (Ragauskas et al., 2006). Early life-cycle studies typically found that substituting biofuels for gasoline or diesel will reduce GHGs emissions because biofuels sequester atmospheric carbon dioxide through the growth of the feedstock (Farrell et al., 2006; Hill et al., 2006). These studies compared petroleum and biofuels for emissions during the separate steps of mining or growing the feedstocks, refining them into fuel and burning the fuel in vehicles; but they did not account for the changes in GHGs emissions from land-use change. If emission perturbations from

land-use change are considered, production and use of many biofuels such as corn-based ethanol and soybean biodiesel are likely to emit more GHGs than fossil fuels (Fargione et al., 2008; Searchinger et al., 2008; Melillo et al., 2009; Lapola et al., 2010). Therefore, it has been argued that a biofuel cannot be considered as “beneficial” unless (1) it offers advantages in reducing GHGs, and (2) it neither competes with food production nor causes land-use change directly or indirectly (Tilman et al., 2009).

The analyses of biofuel policy have appropriately focused on GHGs emissions. However, these analyses to some extent overlooked other important impacts of biofuel use. One impact that can benefit from further investigations is particulate matter (PM) which is co-emitted with GHGs, especially in the case of biodiesel use. PM exerts important effects both on climate (Bond et al., 2013; Ravishankara et al., 2015; von Schneidmesser et al., 2015), and air quality, and hence human health (Samet et al., 2000; Pope and Dockery, 2006; Burnett et al., 2014). Inventory studies (e.g., Bond et al., 2004; Wang et al., 2014; Huang et al., 2015) have identified the transportation sector as a large source of primary particulate organic matter (POM) and black carbon (BC). The contribution of

<sup>☆</sup> This paper has been recommended for acceptance by David Carpenter.

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transportation to BC can be dominant in many regions such as North America and Europe (Wang et al., 2014). The use of biofuel likely has an impact on the emissions of PM from the transportation sector.

Estimation of POM emissions from vehicles is more complex than BC, mainly due to the presence of particulate semivolatile organic compounds (pSVOCs) which can evaporate from the particle-phase during atmospheric dilution (Lipsky and Robinson, 2006; Fujitani et al., 2012; Ranjan et al., 2012; May et al., 2013). A recent chamber study found that primary POM emitted from diesel engines could even evaporate completely once gas-phase organic compounds were consumed due to photochemical oxidation (Presto et al., 2014). Evaporation of pSVOCs not only decreases the POM emission factor with increasing dilution but also creates substantial amounts of gaseous semivolatile organic compound (gSVOC) precursors of secondary organic aerosol (SOA). Such gSVOCs have been demonstrated to represent a dominant category of SOA precursors that are missing in most air quality models (Robinson et al., 2007; Liggió et al., 2016). Until recently, however, direct and quantitative measurement of both pSVOCs and gSVOCs emissions in vehicular exhaust has not been achieved under ambient-relevant conditions (i.e., with a dilution ratio of  $10^3$ – $10^4$  relative to the tailpipe), regardless of fuel types.

In the present study, an experimental system was developed which is capable of measuring emission factors of pSVOCs and other PM components for vehicle engine emissions at dilution ratios representative of ambient air conditions. The system was applied to investigate semivolatile and nonvolatile PM emissions from a light-duty diesel engine using ultra-low-sulfur diesel (ULSD), pure biodiesels and biodiesel blends. Feedstocks for the biodiesels used in this study included tallow/used cooking oil (tallow/UCO), soybean and canola. Biodiesels derived from tallow and UCO are considered “beneficial” according to the criteria suggested by Tilman et al. (2009), with some assessments suggesting that they can reduce life-cycle GHGs emissions by more than 80% (Directive 2009/28/EC, 2009). This study investigated the tallow/UCO biodiesel in detail to determine whether it has benefits in reducing PM emissions (both pSVOCs and nonvolatile components) and whether it shows comparative advantages over soybean and canola derived biodiesels. The effects of pSVOCs evaporation on the physical and chemical properties of the diesel particles were examined to provide insights into the evolution of primary PM emissions from diesel vehicles during dilution.

## 2. Methods

### 2.1. Engine, fuels and operation modes

A light-duty diesel engine (1.9 L, four-stroke, four-cylinder, turbocharged and direct fuel injection), recovered from a 2001 Volkswagen Jetta passenger car and connected to an engine dynamometer, was used for the experiments. The engine was equipped with an exhaust gas recirculation (EGR) system and a diesel oxidation catalyst (DOC). It was tested in three steady-state operation modes, namely aggressive, highway and city average which represent the average driving conditions of three commonly-used Dynamometer Driving Schedules (DDS) also known as the US 06 Supplemental Federal Test Procedure (SFTP), the Highway Fuel Economy Test (HWFET) and the Federal Test Procedure (FTP-75), respectively (<https://www.epa.gov/vehicle-and-fuel-emissions-testing/dynamometer-drive-schedules>). For the aggressive, highway and city average modes, engine speeds were set at 2000, 1800 and 1600 rpm (rotation per minute), engine torques were set at 97, 82 and 55 N m, and engine powers were 20, 16 and 9 kW, respectively.

Test fuels included the baseline ULSD and pure biodiesels reformulated from tallow/UCO, soybean and canola (B100), as well as their blends with ULSD at levels of 5, 20 and 50% by volume (also referred to as B5, B20, and B50, respectively). Fuel properties are summarized in Table S1 for the various test fuels.

### 2.2. PM emission measurements

An illustration of the experimental setup for the PM emission measurements is shown in Fig. 1. The engine was installed on a dynamometer and enclosed in a controlled-access laboratory. The engine exhaust was directly vented into a constant volume sampling (CVS) system in the next room where it underwent primary dilution with air filtered through charcoal and high-efficiency particulate arresting (HEPA) filters. Primary dilute exhaust from the CVS, with an ambient temperature of 23.5 °C, was drawn into a Dekati dilutor (Dekati Ltd., Tampere, Finland) for the second-stage dilution, again with air filtered through charcoal and HEPA filters. Subsequently, secondary dilute exhaust from the Dekati was drawn into a specially designed flow tube for the third-stage dilution with zero air provided by an AADCO pure-air generator (AADCO Instruments, Cleves, OH, USA). Particle residence time in the flow tube was 3.2 min. At each dilution stage, mass concentration of non-refractory components of particles (mainly POM and sulfate) was measured using a high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS; Aerodyne Research Inc., Billerica, MA, USA), refractory black carbon (rBC; Petzold et al., 2013) mass concentration was derived from a single particle soot photometer (SP2; Droplet Measurement Technologies Inc., Boulder, CO, USA), and particle number-size distribution was determined by a scanning mobility particle sizer (SMPS; TSI Inc., Shoreview, MN, USA). The engine exhaust was continuously drawn into the dilution sections of this study. Therefore, repeated measurements could be made for each test, including by the SMPS which was operated at a relatively low time resolution (i.e., 3 min, with a detection range of 14.6–685.4 nm in terms of mobility diameter) compared to the SP2 and HR-ToF-AMS. Operation of these instruments has been described in detail elsewhere (Li et al., 2011). Terminologies involved in this study (e.g., POM and pSVOCs) and their relationships are summarized in Appendix A.

In the CVS, carbon monoxide (CO) and carbon dioxide (CO<sub>2</sub>) were measured by non-dispersive infrared (NDIR) analyzers while a flame ionization detector (FID) was used to quantify total hydrocarbon (THC). Primary dilution ratios in the CVS ( $DR_{CVS}$ ) were calculated based on the measured concentrations of CO, CO<sub>2</sub> and THC (Table S2), and then used together with BC concentrations (sulfate was used when BC was not available) to determine the dilution ratios in the subsequent Dekati and flow tube (Tables S3–S4). The dilution ratios achieved in the CVS, Dekati and flow tube were on the order of  $10^1$ ,  $10^2$  and  $10^3$ – $10^4$ , respectively (relative to the tailpipe, similarly hereinafter; Tables S2–S4).

Emission factors were estimated using a mass balance approach

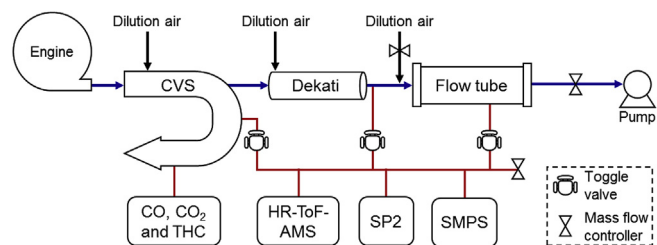


Fig. 1. Experimental setup for the measurement of semivolatile and nonvolatile PM emissions.

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