



Effects of engine load and dilution conditions on gas-particle partitioning of primary organic aerosol emitted from a light-duty diesel engine



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ABSTRACT

The effects of engine load and dilution conditions on the gas-particle partitioning of primary organic aerosol (POA) emitted from a light-duty diesel engine were investigated in a constant volume container (CVC). A new method was adopted to determine the POA concentration by using the combination of an OC/EC analyzer and a scanning mobility particle sizer (SMPS). A thermodenuder (TD) combined with a gas-chromatography/mass-spectrometry (GC/MS) was further used to study the temperature effect on the POA phase partitioning. Our results indicate that the engine load significantly affects the total organic carbon (OC) emissions while the dilution ratio (DR) influences the phase partitioning. Both sorption and evaporation are observed during the isothermal dilution process. Phase partitioning of the organic aerosol (OA) is dominated by sorption at low dilution condition and by evaporation of the adsorbed/absorbed OA at high dilution condition. A further analysis on the TD experimental data obtained at different engine loads suggests that the OA from the medium load is more sensitive to the temperature while the OA from the high load is hard to evaporate. Two TD kinetic models based on volatility distribution are evaluated and the results are compared with the experimental data. The present study demonstrates the importance of dilution conditions and engine loads on the phase partitioning of diesel POA emitted from light-duty diesel engines at the near tailpipe environment.

1. Introduction

The carbonaceous particulate matter emitted from a diesel engine includes primary organic aerosol (POA) and elemental carbon (EC) (Watson, Chow, & Lowenthal, 1994; Lu, Huang, Cheung, & Ma, 2012). A diesel engine emits organic compounds which exist both in the particle phase and gas phase. POA refers to the organic compounds which exist in the particle phase. POA contains volatile substances which condense on the elemental carbon upon cooling. The volatility of a volatile substance can be defined by considering its effective saturation concentration, C^* , which is related to the subcooled liquid saturation vapor pressure of the substance (May et al., 2013a). Based on the value of C^* , organic compounds can be classified as intermediate volatility organic compounds (IVOCs, $10^3 < C^* < 10^6 \mu\text{g m}^{-3}$), semivolatile organic compounds (SVOCs, $10^{-1} < C^* < 10^3 \mu\text{g m}^{-3}$) and low-volatility organic compounds (LVOCs, $C^* < 10^{-1} \mu\text{g m}^{-3}$). Previous research have demonstrated that the majority of POA are semivolatile (Risse & Kettrup 2000; Sihabut, Ray, Northcross, & McDow, 2005; Lipsky and Robinson, 2006; Robinson et al., 2007; Grieshop,

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Miracolo, Donahue, & Robinson, 2009; Robinson, Grieshop, Donahue, & Hunt, 2010) and POA proceeds gas-particle partitioning during the dilution process by changing from the gas phase to the particle phase through adsorption onto the particle surfaces or through absorption into the organics solution, and changing from the particle phase to the gas phase through evaporation. The process which dominates the gas-particle partition is related to the OC/EC ratio and the dilution conditions.

Previous investigations demonstrated the existence of sorption process in the exhaust pipe or in tunnel conditions (Dachs & Eisenreich, 2000; Roth, Goss, & Schwarzenbach, 2005). For sources with a significant amount of OA, the fine particle mass emission rates were higher when measured with dilution samplers compared to those measured with filters collected at exhaust temperatures (Frisch, Johnson, & Leddy, 1980; Hildemann, Cass, & Markowski, 1989), indicating the sorption of the OA. Schneider et al. (2005) confirmed that organic compounds preferred to condense on the particle surfaces during the cooling and dilution process. Thus, in a small constant volume near the outlet of an exhaust pipe or in a dilution sampler, the OA will go through a sorption process, including both absorption and adsorption. However, their investigations are focused on the sorption process while the evaporation process has not been analyzed.

In the atmosphere, gas-particle partitioning of POA is considered as a desorption (or evaporation) process which could be explained by the absorption mechanism (Pankow, 1994a; Odum et al., 1996) by taking into consideration the influence of local temperature, saturation pressure and organic compounds concentrations. Recently, Robinson and his co-workers conducted experiments to verify the absorption mechanism by comparing the results obtained from a smog chamber with that obtained from a dilution sampler (Presto, Hennigan, Nguyen, & Robinson, 2012; Ranjan, Presto, May, & Robinson, 2012; May et al., 2013b). Their results showed that when the OA mass concentrations was diluted from 1000 to $1 \mu\text{g m}^{-3}$, the fraction of total OC in the particle phase was shifted from ~ 0.8 to ~ 0.1 , indicating the majority of the SVOCs resided in the gas phase under atmospheric conditions, which was in agreement with the absorption mechanism. However, their experiments were conducted in a smog chamber having a large volume of about 7 m^3 , and thus the results reflected changes in the ambient atmosphere rather than changes in the region close to the engine exhaust pipe, which should have a much smaller volume with a different dilution condition.

The above literature survey shows that phase partitioning of diesel POA at atmospheric conditions have been investigated in recent years, however, more independent studies are still necessary to confirm and extent the prior work, especially focusing on the engine sources testing. In this study, we focus on a systematic investigation on the effects of engine load and dilution conditions on the phase partitioning of the organic aerosols in a region close to the engine exhaust, where the majority of the measured organic aerosols could be considered as diesel POA. The dilution conditions near the engine tailpipe is close to the condition of tunnel and canyon, which are both highly polluted by vehicles under a relative lower dilution condition, and is totally different from the ambient conditions. Meanwhile, the engine load would determine the initial OC/EC ratio of diesel carbonaceous compounds that might also influence the POA phase partitioning. Therefore, in the present study, the effects of dilution conditions, including the dilution ratio and temperature, as well as engine load (low, medium and high) on the gas-particle partitioning of POA emissions emitted from a direct-injection diesel engine were investigated in a constant volume container (CVC). The CVC was designed to simulate the condition near the outlet of the engine exhaust pipe, where the exhaust gas is diluted by the atmospheric air to room temperature. Diesel POA emitted at various engine loads would proceed the gas-particle phase partitioning in the CVC under different dilution conditions. A new method was adopted to determine the POA gas-particle partitioning through analyzing the results obtained from a scanning mobility particle sizer (SMPS). Furthermore, temperature effect on POA gas-particle partitioning was studied by using a thermodenuder (TD) and the volatility distribution derived from GC/MS was used to fit the TD models by using kinetic equations.

2. Materials and methods

2.1. Test engine and fuel properties

The experiments were conducted on a naturally aspirated, water-cooled, 4-cylinder, direct-injection light-duty diesel engine. The specifications of the engine are shown in Table 1. The engine was connected to an eddy-current dynamometer and a controller was used for adjusting the engine speed and engine torque. The fuel used in this study was a commercial diesel fuel (sulfur, $< 10 \text{ ppm-wt}$) that meets the fuel sulfur limit of China V emissions regulation for diesel vehicles. The major properties of the fuel are shown in Table 2.

Table 1
Engine specifications.

Model	Isuzu 4HF1
Type	In-line 4-cylinder
Maximum power	88 kW/3200 rev/min
Maximum torque	285 N m/1800 rev/min
Bore \times stroke	112 mm \times 110 mm
Displacement	4334/cc
Compression ratio	19.0:1
Fuel injection timing (BTDC)	8°
Injection pump type	Bosch in-line type
Injection nozzle	Hole type (with 5 orifices)

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