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Variations of the Particulate Carbon Distribution from a Nonroad Diesel Generator

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The emissions of diesel particulate matter (DPM) from diesel engines are causing increasing health concerns due to their suspected carcinogenicity, especially the carbonaceous fractions. The total DPM emissions and the organic and elemental carbon (OC and EC) distributions of the DPM depend on many operating factors, such as load, engine design parameters, fuel sulfur content, fuel usage rate, and sampling conditions. Results of previous studies on the OC/EC variations with load for heavy-duty vehicles have been reported, but information is scarce for nonroad diesel generators. There is a clear need to better characterize nonroad DPM emissions, as studies have indicated that DPM emissions from nonroad diesel engines are significantly higher than those from on-road sources. The objective of the study is to provide a detailed account of the OC/EC distributions for a nonroad diesel generator operated with high and low sulfur fuels under different load conditions. DPM emissions were collected using an EPA Method 5 (Determination of Particulate Matter Emissions from Stationary Sources) sampling train. The OC and EC concentrations were quantified by NIOSH Method 5040. DPM concentrations and the relative contributions of OC, EC, and noncarbonaceous materials vary significantly with engine load, fuel sulfur content, and sample collection temperature. The fractions of EC over DPM increase with increasing load from 21% at 0kW to 84% at 75 kW for the low sulfur fuel, while those of OC decrease from 62% to 9%. This is consistent with other studies, and the same trends exist regardless of the sulfur content and DPM collection temperature. The fractions of organic compounds range from 77% to 19% for the high sulfur fuel. Noncarbonaceous materials are from 27% to 18% in fraction from high sulfur DPM as opposed to the 17% to 7% in the low sulfur diesel emissions. At lower collection temperatures, more OC and noncarbonaceous materials are observed.

Introduction

Nonroad diesel engines are widely used in construction, mining, agriculture, and as power equipment at various capacities and operating conditions. However, the emissions from nonroad sources are not as well characterized as those from on-road diesel vehicles. Studies by the U.S. Environmental Protection Agency (EPA) indicate that emissions of diesel particulate matter (DPM) from nonroad diesel machines are significantly higher than those from on-road sources due to lower fuel quality, inadequate engine maintenance, and older engine age, etc. Nonroad diesel machines currently account for approximately 44% of total DPM and 12% of NO_x emissions from mobile sources nationwide (*1*). Although the sales volume of nonroad diesel equipment is much lower than that of on-road equipment, the total petroleum consumption rates of nonroad sources and on-road sources are relatively close, 9% and 11%, respectively (*2*). The emission characteristics of nonroad sources can be more complex than on-road sources, as there are more than 1700 manufacturers of nonroad diesel equipment producing hundreds of diesel engine types, while there are only about a dozen heavy-duty truck manufactures (*2*).

DPM consists mostly of carbonaceous materials, which are often classified as elemental carbon (EC) and organic carbon (OC). EC and OC affect the environment in different ways due to their optical, physical, chemical, and toxicological properties. EC, the graphite portion of DPM, is a byproduct of incomplete combustion. The OC fraction of DPM is a complex mixture of unburned fuel, engine oil, and numerous soot-associated organics including PAHs (polycyclic aromatic hydrocarbons) formed during combustion. EC has been linked to dysrhythmia and cardiovascular diseases (*3*), while the PAHs in the OC fraction are reasonably anticipated to be human carcinogens (*4*, *5*). These PAHs, when adsorbed on particle surfaces, can be detrimental to human health, as 90% of the DPM is in the size range of less than 1.0 μm which can be inhaled deep into the human lungs (*6*). The National Institute for Occupational Safety and Health (NIOSH) considers diesel exhaust a potential occupational carcinogen (*7*). Other organizations, including the International Agency for Research on Cancer (IARC) (*8*), the World Health Organization (WHO) (*9*), the California Environmental Protection Agency (*10*), the U.S. EPA (*11*), and the National Toxicology Program (*12*), have reviewed the animal and human exposure evidence, and each has classified diesel exhaust as a probable human carcinogen or similar designation.

EC and OC distributions in DPM may vary, depending upon a number of factors, such as load, the fuel type, engine type, duty cycle, engine maintenance, individual operators, the use of emission control devices, and the composition of the lubricant oil, etc. There have been some previous studies on the OC/EC variation with load conditions for new heavy-duty diesel vehicles (*13*) and military vehicles (*14*), but information is scarce for nonroad diesel generators. In addition, the uniqueness and high complexity of nonroad diesel equipment also calls for detailed characterization of the DPM emissions.

Diesel generators are readily transportable and widely used to provide power for many applications that require on-site electricity generation. Diesel generators run at a fixed

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rpm and voltage, unlike on-road or off-road diesel vehicles (*15*). The difference in operating mode can potentially result in different emission characteristics. Studies presented in this paper were performed on a Generac diesel generator (model SD080) rated at 80 kW and 1800 rpm. A load simulator (model Merlin 100 by Simplex) was used to simulate various load conditions from 0 to 75 kW. The objective of the study is to provide a detailed account of the OC/EC distributions for a nonroad diesel generator operated with high and low sulfur fuels under different load conditions. Studies on nonroad engines based on their load capacities are necessary due to the more varieties of nonroad diesel sources than on-road sources, in addition to higher DPM emissions. The findings of this study may prove useful to the control and regulation of nonroad diesel generators in this capacity range.

Experimental Methods

Detailed information on the experimental methods used for this work has been published elsewhere (*16*) and is briefly summarized here. DPM was collected on quartz fiber filters (Millipore) with an EPA Method 5 sampling train (Figure S1 in the Supporting Information) (*17*). The applied engine load ranged from 0 to 75 kW with 25 kW increments. The DPM mass concentrations were determined by gravimetric analysis. Before being weighed, filters were desiccated at 20 °C ± 5.6 °C and ambient pressure for at least 24 h. The OC/EC loadings on the filters were determined by NIOSH Method 5040, a thermal-optical transmittance method commonly used in OC/EC measurements (*18*). Two diesel fuels with different sulfur contents (0.05 and 0.37 wt %) were used in the study to evaluate the effects of fuel sulfur content on DPM OC/EC distributions. The sulfur contents were measured by a commercial source (OKI laboratories). The OC concentrations from blank quartz filters were measured and subtracted from DPM results.

In EPA Method 5, the engine exhaust is directly sampled through a heated sampling probe with a stainless steel liner. Particulate matter is withdrawn isokinetically from the source and collected on a glass fiber filter maintained at 120 °C ± 14 °C. The particulate mass, which includes any material that condenses at or above the collection/filtration temperature, is determined gravimetrically after the removal of uncombined water. Uncombined water refers to free water or water that is not chemically combined, which can be easily removed by Method 5 operating procedures, such as heating and/or desiccation. Unlike dilution methods used by Shi et al. (*13*) and Rogers et al. (*19*), Method 5 collects DPM emissions directly from the source without dilution, and both the sampling line and the filter are heated to approximately 120 °C. The sampling probe and transfer line are washed with acetone after each sampling, and the wash liquid is dried and weighed to account for the DPM deposited at these locations.

Two modifications were made to Method 5 to meet the specific requirements of the OC/EC analysis: (1) DPM was collected on quartz fiber filters instead of glass fiber filters, as quartz filters are required by NIOSH Method 5040 for thermal-optical analysis; (2) due to the maximum loading limitation of the OC/EC analyzer, the sampling duration for DPM collection ranged from 8 to 15 min, which is much shorter than the typical Method 5 sampling period of approximately 60 min.

Samples were collected at two different temperatures to determine the temperature effects on the OC/EC distributions and total DPM, as the gas-particle partitioning of the condensable fractions of DPM can be sensitive to temperature and pressure. As already mentioned, in EPA Method 5, the DPM collection temperature is maintained at 120 °C ± 14 °C to prevent water condensation, which can also result in the evaporation of semi-volatile organic compounds (SVOC). On the other hand, most DPM collection systems employing dilution methods operate under ambient conditions. To better understand the influence of collection temperature, a comparison group of samples was collected with the filter unheated, and the collection temperature was maintained at 25 °C ± 3 °C.

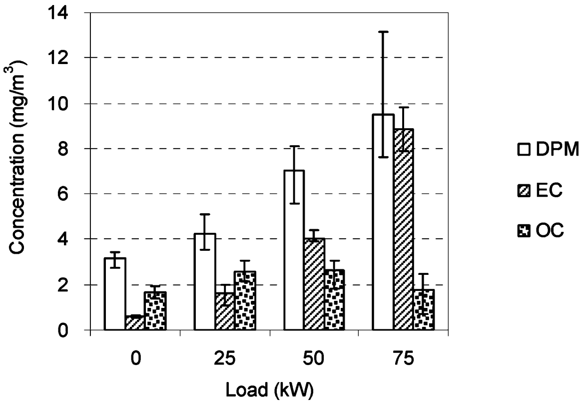

FIGURE 1. Concentration of OC, EC, and DPM versus engine load (sulfur content = 0.05%; collection temperature = 120 °C ± 14 °C; error bars correspond to actual ranges of concentration measurements).

Results and Discussion

Overall Trends of OC/EC Distribution with Load. Figure 1 presents the OC, EC, and DPM concentrations for low sulfur diesel (fuel sulfur content at 0.05%) emissions collected at 120 °C ± 14 °C under various loads. Multiple measurements were made at each load condition; that is, 5, 7, 5, and 4 measurements were taken at 0, 25, 50, and 75 kW, respectively, and sufficient reproducibility is evidenced by the figure. The data indicate that the EC concentrations increase by a factor of about 15 when the load is increased from 0 to 75 kW. The increase in EC concentrations with load is consistent with the higher fuel usage, lower air-to-fuel ratio, and higher temperature at higher loads (observed in another study of ours (*20*)), as EC is a product of incomplete combustion. In contrast, the OC concentrations exhibit much less variation with load. Therefore, OC/EC ratios decrease with increasing load.

DPM has been classified into three portions by this study: organic compounds, EC, and noncarbonaceous materials. The mass concentration of organic compounds can be estimated from OC by a multiplicative factor of 1.2 to account for other elements, (e.g., hydrogen, oxygen, nitrogen, sulfur) in addition to carbon (*21*). The noncarbonaceous materials may include sulfates, nitrates, metals, and ash, which were indirectly measured by the difference between DPM and the carbonaceous mass fractions (i.e., the total mass of EC and the organic compounds).

The fractions of organic compounds, EC, and noncarbonaceous materials in DPM at various loads for the low sulfur diesel fuel and a collection temperature of 120 °C ± 14 °C are shown in Figure 2. It is seen that the EC fraction increases from 21% at 0 kW to 84% at 75 kW, while the fraction of organic compounds decreases from 62% at 0 kW to 9% at 75 kW. These results are consistent with studies by Shi et al. (*13*) and Burtscher et al. (*22*). Shi et al. reported that EC fractions increase by a factor of about 1.5 and OC fractions decrease by a factor of about 2 when increasing the load on a diesel engine from 25% to 100%. Burtscher et al. (*22*) reported that the volatile fraction decreased significantly with increasing load while the black-carbon concentrations increased with increasing load. The total mass of the organic compounds plus EC ranged from 83% to 93% for this study.

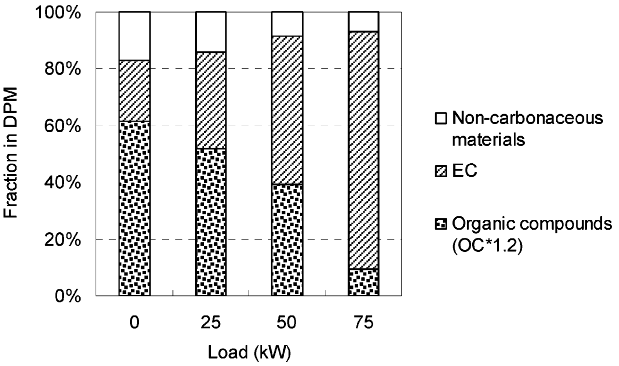

FIGURE 2. Fractions of organic compounds, EC, and noncarbonaceous materials in DPM versus load (sulfur content = 0.05%, collection temperature = 120 °C ± 14 °C).

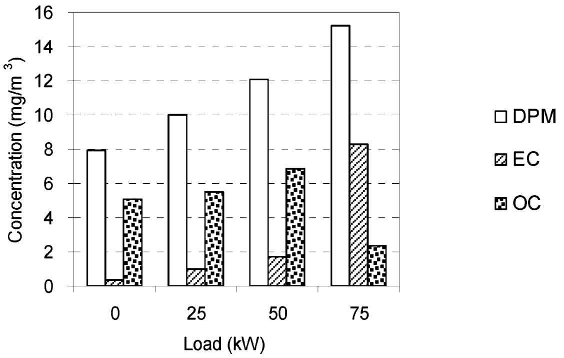

FIGURE 3. Concentration of OC, EC, and DPM versus engine load (sulfur content = 0.37%, collection temperature = 120 °C ± 14 °C).

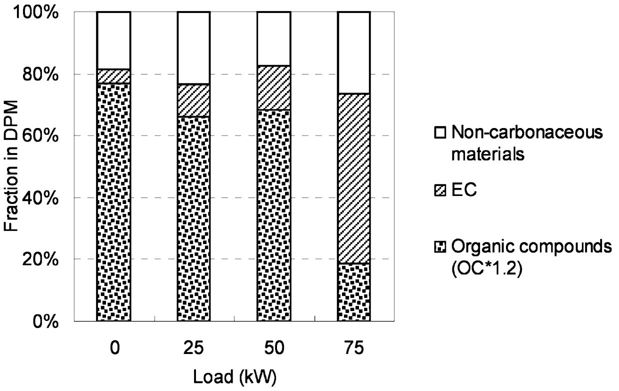

FIGURE 4. Fractions of organic compounds, EC, and noncarbonaceous materials in DPM versus load (sulfur content = 0.37%, collection temperature = 120 °C ± 14 °C).

Effects of Diesel Fuel Sulfur Content. Figures 3 and 4 present concentrations and fractions of organic compounds, EC, and noncarbonaceous materials in DPM, respectively, at various loads for the high sulfur diesel (fuel sulfur content at 0.37%) and a collection temperature of 120 °C ± 14 °C. The increase in EC fractions and decrease in organic compound fractions with increasing load is consistent with the low sulfur diesel. Consistent with a previous study, more DPM was collected at higher fuel sulfur content (*20*). However, the organic compounds and the noncarbonaceous materials contribute more to DPM in the high sulfur diesel emissions than in the low sulfur diesel emissions. In the high sulfur diesel emissions, the organic compounds account for 19–77% as opposed to 9–62% in the low sulfur diesel emissions, and the noncarbonaceous materials account for 18–27% as opposed to 7–17% in the low sulfur diesel emissions.

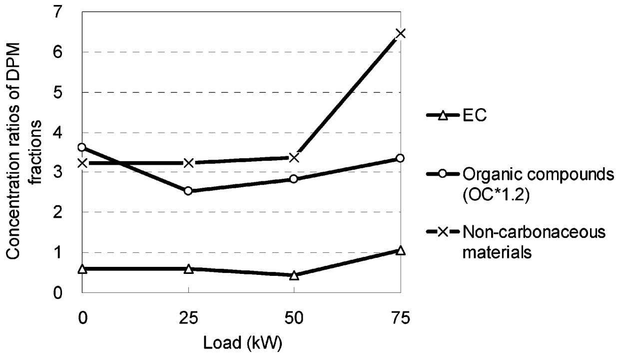

FIGURE 5. Concentration ratios of DPM fractions (organic compounds, EC, and noncarbonaceous materials) from high sulfur diesel fuel (sulfur content = 0.37%) over those from low sulfur diesel (sulfur content = 0.05%) emissions. The collection temperature is 120 °C ± 14 °C for both scenarios.

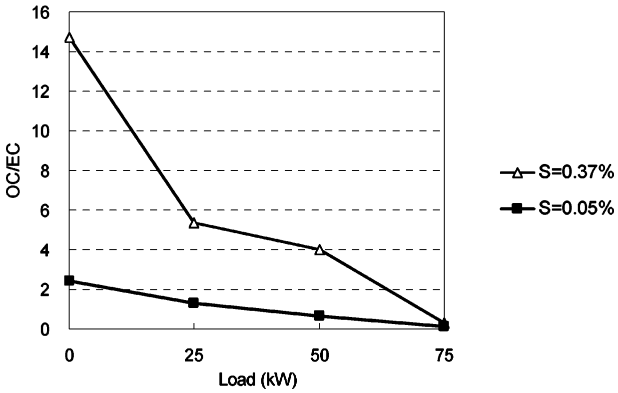

FIGURE 6. OC/EC versus load at collection temperatures of 120 °C ± 14 °C (S: sulfur content).

As illustrated in Figure 5, in the high sulfur diesel emissions, the organic compounds measured are 2.5–3.6 times higher than in the low sulfur diesel emissions, and the noncarbonaceous materials are 3.2–6.4 times higher than in the low sulfur diesel emissions. The increase of organic compounds in the high sulfur diesel emissions is consistent with what has been presented by Wall and Hoekman (*23*), who reported that the soluble organic fraction (SOF) in DPM increases with increasing sulfur content in diesel. The higher sulfur content results in higher concentrations of sulfur acid, which promotes the nucleation of organic compounds (*24*). The increase of noncarbonaceous materials may partially be the result of the increase of sulfate fractions. Research is underway to study the compositions of the noncarbonaceous materials with load variations.

In Figure 6, the OC/EC ratios for the high sulfur diesel are compared to those for the low sulfur diesel at various loads. For the low sulfur diesel, the OC/EC ratios decreased from 2.41 at 0 kW to 0.09 at 75 kW. For the high sulfur diesel, the OC/EC ratios decreased from 14.71 at 0 kW to 0.29 at 75 kW. Significantly more OC was obtained from the high sulfur fuel at lower loads, and the OC/EC ratio rapidly decreased (Figure 6) with load increase. The OC/EC ratios under the same loads should mainly be affected by fuel sulfur content. In addition, the OC/EC ratios over the load range may be affected by the fuel usage rate and the amount of excess air in the system (*20*).

Effects on DPM Collection Temperature. The fractions of organic compounds, EC, and carbonaceous materials in DPM at various loads for the low sulfur diesel (diesel sulfur content = 0.05%) and a collection temperature of 25 °C ± 3 °C are shown in Figure 7, and their trends with load are consistent with those in Figures 2 and 4. At the collection

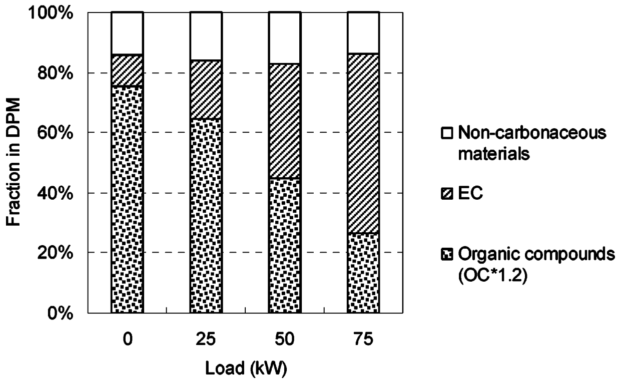

FIGURE 7. Fractions of organic compounds, EC, and noncarbonaceous materials in DPM versus load (sulfur content = 0.05%, collection temperature = 25 °C ± 3 °C).

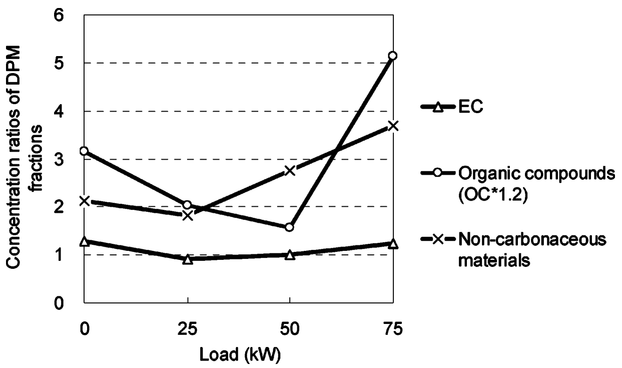

FIGURE 8. Concentration ratios of DPM fractions (organic compounds, EC, and noncarbonaceous materials) at the collection temperature of 25 °C ± 3 °C over those at 120 °C ± 14 °C (sulfur content = 0.05%).

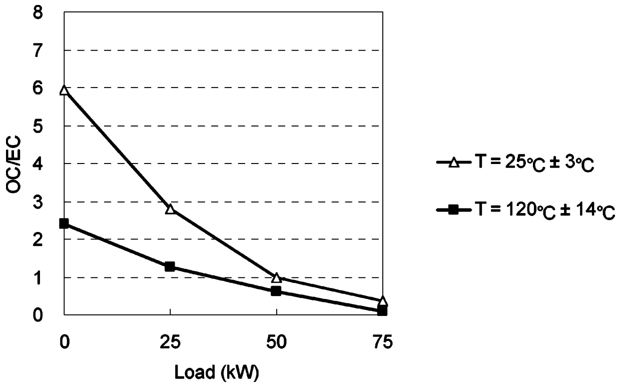

FIGURE 9. OC/EC versus load (sulfur content = 0.05%, *T* = temperature).

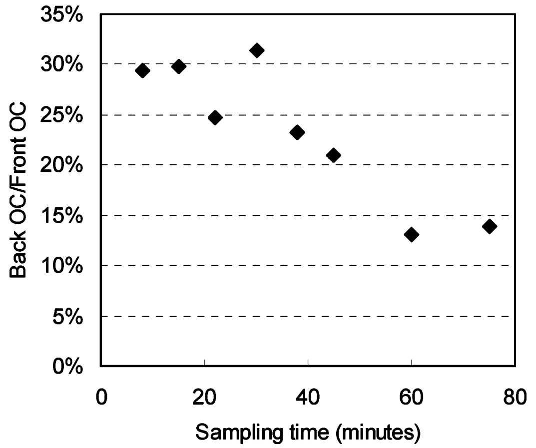

FIGURE 10. The OC ratios of the backup filter to the front filter over time.

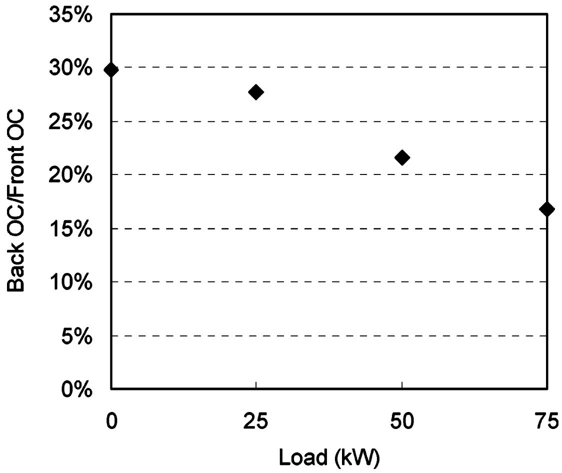

FIGURE 11. The OC ratios of the backup filter to the front filter under various loads at the sampling time of 15 min.

temperature of 25 °C ± 3 °C, the organic compounds account for 27–75% of the DPM mass, as opposed to 9–62% at the collection temperature of 120 °C ± 14 °C.

In Figure 8, the ratios of organic compounds, EC, and noncarbonaceous materials concentrations collected at 25 °C ± 3 °C and 120 °C ± 14 °C are presented. At the collection temperature of 25 °C ± 3 °C, both organic compounds and noncarbonaceous materials concentrations are much higher than in those collected at 120 °C ± 14 °C, an indication of more condensation of both of these components from vapor to particle phase at lower temperatures. As expected for EC (which is nonvolatile), almost no differences were observed at the two different collection temperatures, which indicates that the EC concentrations were not much influenced by the collection temperature. The decreased OC ratios (OC at 25 °C relative to that at 120 °C) at 25 and 50 kW are consistent with the findings of Ning et al. (*25*), who reported higher SOFs at low and high loads than at medium load due to increased condensation and nucleation of organic compounds under low and high load conditions.

In Figure 9, the OC/EC ratios for a collection temperature of 25 °C ± 3 °C are compared to those found at 120 °C ± 14 °C. At a collection temperature of 25 °C ± 3 °C, the OC/EC ratios decrease from 5.95 at 0 kW to 0.37 at 75 kW. The OC/EC ratios at 25 °C ± 3 °C are higher than those obtained at 120 °C ± 14 °C. The higher OC fractions found at the lower temperature reflect the collection of a larger amount of condensable organic compounds (semi-volatile fraction).

Sampling Artifacts. The sampling artifacts of the system were investigated using a quartz-quartz tandem filter method (*26*). Figure 10 shows the variations of the ratios of back filter OC to front filter OC from 8 to 80 min under the same load (0 kW). The 0 kW load was examined because it produces the most OC. The percentage of OC on the back filter relative to the front filter ranges from 29.3% at 8 min to 13.2% at 80 min and decreases with sampling time. Figure 11 shows the percentages of back filter OC to front filter OC as a function of engine load for the sampling time of 15 min, and the ratio decreases as engine load increases. For the 8–15 min sampling time that we used to study OC/EC variations, the OC on the backup filter can range from 29.3% to 16.8% of that on the front filter. These results are consistent with Shah et al. (*26*) and Cadle et al. (*27*). However, sampling artifact is a complex issue, where both adsorption and volatilization of DPM can occur. Thus, the actual correction may require further study, which is beyond the scope of this paper.

Acknowledgments

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Supporting Information Available

EPA Method 5 sampling train, the actual picture, and the schematic diagram. This material is available free of charge via the Internet at http://pubs.acs.org.

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