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Measurement of diesel engine emitted nanoparticles

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The European Particle Measurement Programme (PMP) has introduced particle number regulation for Euro 5 and Euro 6 standard to meet more stringent Particulate Matter mass regulation. The PMP protocol specifies measurement of accumulation mode solid particles larger than 23 nm, to exclude the possible measurement artifact caused by volatile particles present in nucleation mode. However, exclusion of sub-23 nm particles may have some potential issues. This study presents chassis dynamometer tests of diesel particle penetration/formation under PMP protocol using both a PMP system and a catalytic stripper, which is another type of volatile particle remover. It was found that particles below 23 nm were present downstream of the PMP and CS. The PMP always measured higher particle number emissions than the CS.

1. Introduction

As regulation of diesel Particulate Matter (PM) mass gets more stringent, the current gravimetric methods for the legal determination of emissions will have difficulty accurately quantifying PM mass emissions. Although the United States (US) Environmental Protection Agency (EPA) issued an improved protocol for the gravimetric method [1], accuracy will continue to be an issue at the very low emission levels of new diesel vehicles equipped with after treatment.

Progress in regulating diesel particle emissions by non-gravimetric means has been made in Europe. The United Nations Economic Commission for Europe-Group of Experts on Pollution and Energy (UNECE-GRPE) initiated the Particle Measurement Programme (PMP) working group to develop new particle measurement techniques to supplement or replace the current gravimetric method. The PMP has recommended a particle number-based method to complement the gravimetric method. The PMP protocol specifies measuring solid particles larger than 23 nm (nanometer). By the operational definition of the PMP, solid particles are particles that can survive after passing through an evaporation tube (ET) that has a wall temperature of 300 – 400°C. The PMP only measures solid particles larger than 23 nm to avoid issues with poor repeatability caused by volatile particles present in the nucleation mode of diesel exhaust [2]. Exclusion of sub-23 nm particles may have some potential issues; however, since not all sub-23 nm or nucleation mode size range particles are volatile. Some studies have found solid particles in the nucleation mode from heavy-duty diesel vehicles operating at idle or low loads [3, 4]. Even at high load operating conditions, solid particles in the nucleation mode have been observed for heavy-duty diesel vehicles [5-7]. By excluding these sub-23 nm solid particles, the full human health of solid particles is not characterized by the PMP standard [2]. An alternative system commonly used by researchers to remove volatile particles is a Catalytic Stripper (CS) [8-12]. In contrast to the PMP system, the CS uses a different approach to remove volatile particles. It removes all volatile hydrocarbon components and sulfur components by catalytic reactions at an elevated temperature. Therefore, re-nucleation will not occur downstream the CS. A study comparing the volatile removal efficiency of a CS with a thermal denuder, which is another type of volatile particle remover, showed that the CS had a better performance than the thermodenuder [11]. However, no studies have been conducted to compare the PMP system with the CS in terms of volatile particle removing efficiency.

This study presents vehicle experiments of diesel particle penetration/formation using the PMP system and CS. This study investigated and compared the effectiveness of the European PMP system and CS in removing volatile aerosols using exhaust from a heavy-duty diesel vehicle operating under different conditions.

2. Experimental

A schematic of the chassis dynamometer test is shown in Figure 1. The setup can be divided into two parallel systems, the CS system and the APC system. Both the CS and APC systems took samples from the same inlet. A cyclone was used on this inlet to remove particles bigger than 2.5 μm, in accordance to the PMP protocol. After the cyclone, tubes leading to the CS and APC were heated up to 150°C, which is the same temperature as the heating temperature of the primary diluter of the APC. On the CS side, an ejector pump (*model, vendor*) was used to pull exhaust through the CS. The pressure of the filtered compressed air used for the ejector pump was 55 psi, making the flow rate through the CS to be 10 Lpm. The exhaust was further diluted by a venturi pump after the ejector pump to avoid saturating instruments.

An Engine Exhaust Particle Sizer (EEPS, cut off diameter is 5.6 nm) spectrometer (TSI, 3936) and a 3022A CPC (labeled as CPC 3022A_CVS, D50 = 7 nm) were used to sample directly from the CVS to measure the source aerosol particle size distributions and particle number concentrations. A 3790 CPC (labeled as CPC 3790_APC, D50 = 23 nm) is built into the APC by the manufacturer and it always sampled from the APC side. Similarly, a 3772 CPC (labeled as CPC 3772_CS, D50 = 11 nm) and CE-CERT's fast-SMPS (labeled as fast-SMPS) were fixed to always sample from the CS side. One nano-SMPS and three CPCs with different cut off sizes were switched alternatively between the CS side and the APC side to measure particle size distributions and particle number concentrations. For the purpose of this manuscript, this set of instruments is called the alternate set. The nano-SMPS consisted of a TSI 3085 nanoDMA and a TSI 3776 CPC. The three CPCs were a TSI 3025A CPC (D50 = 3 nm), a TSI 3776 CPC (D50 = 3 nm), a TSI 3772 CPC (D50 = 11 nm). The specifications of all these instruments are also summarized in table 1, including cut off sizes, maximum concentrations, and sample locations.

The vehicle and aftertreatment system used for the chassis dynamometer testing was the same as that used for the on-road test in CARB's previous study [13]. It was a 14.6 liter, 2000 Caterpillar C-15 engine equipped, Freightliner class 8 truck. A Johnson Matthey Continuously Regenerating Trap (CRT^{TM}) was installed on the vehicle. The CRT^{TM} is a passive DPF system that had previously been shown to provide sufficient levels of particles over driving conditions similar to those used in this experiment [13]. The MEL trailer and truck combined have a weight of approximately 65,000 lbs, including all emission instruments. The truck had a mileage of 41,442 miles. CARB Ultra Low Sulfur Diesel (ULSD) fuel (S < 15 ppmw) and standard lubricating oil with sulfur level ranging from 0.3 to 0.6% were used. Two cruise cycles with extremely different nucleation mode particle number concentrations were tested. The two cycles both had a constant speed of 56 mph, one with a 26% engine load and the other with a 74% engine load.

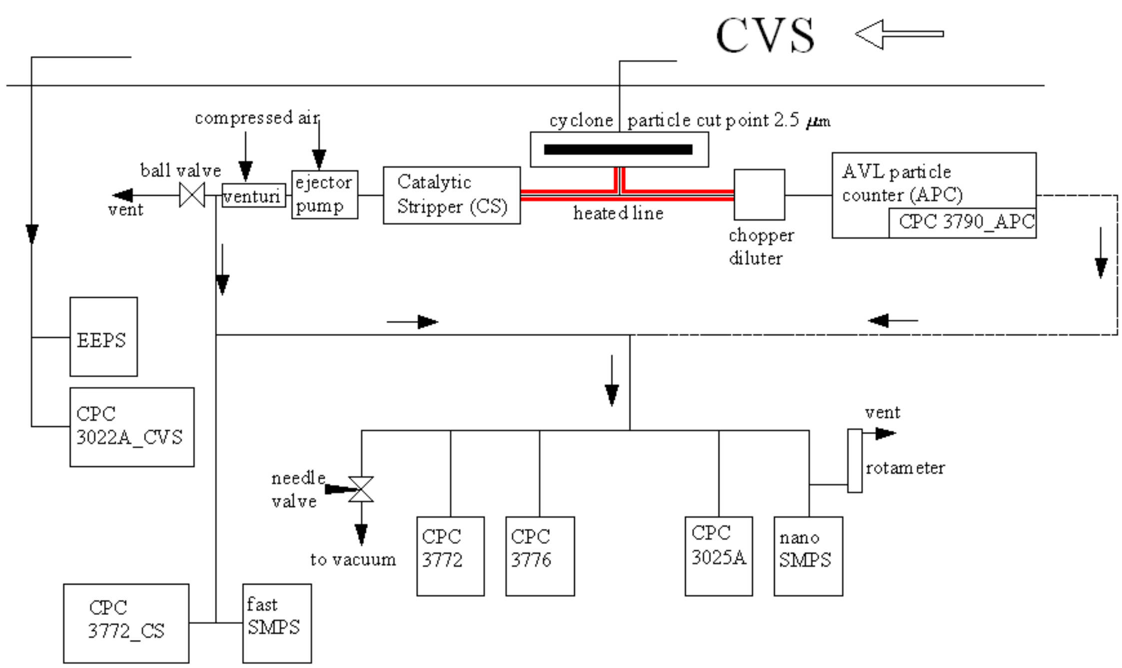
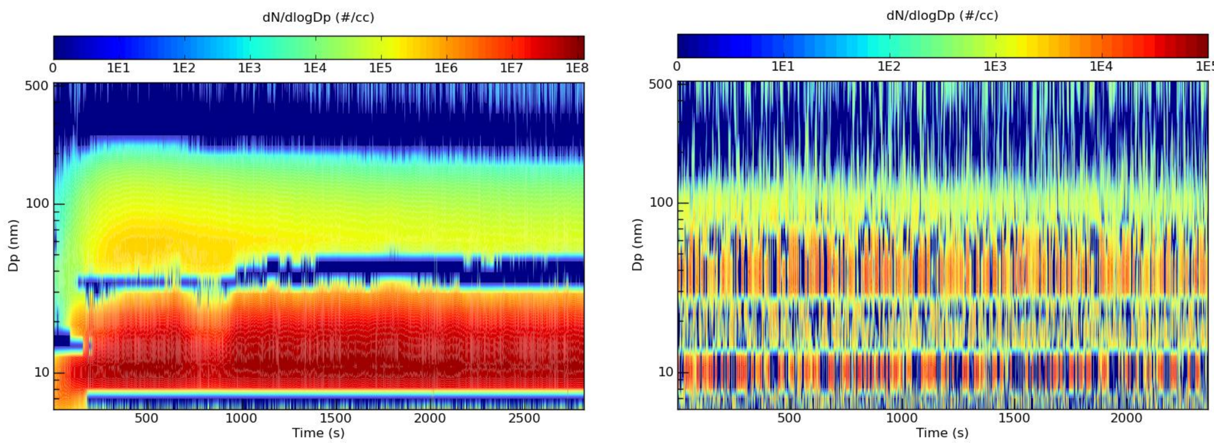


Figure 1 Schematic of the testing arrangement for the chassis dynamometer test

3. Results and Discussions

As the aerosol in the CVS was the source aerosol being fed into the APC and CS, it is important to characterize the CVS aerosol. Contour plots of particle size distributions in the CVS, as shown in Figure 2, showed distinctive patterns of accumulation mode and nucleation mode particles for the two tested cycles. The number concentrations of both nucleation mode and accumulation mode particles were higher and more stable at the 74% engine load than at the 26% engine load. For accumulation mode particles, the number concentration was about one order of magnitude higher at the 74% engine load than at the 26% engine load. For nucleation mode particles, it was about four orders of magnitude higher at the 74% engine load than at the 26% engine load. At the lower engine load, the number concentration of nucleation mode particles oscillated from 0 to 6×10^4 particles/cm^3. This is near the lower detection limit of the EEPS for particles in the nucleation mode size range, which is about 2×10^3 particles/cm^3. The observation of higher number concentrations of nucleation mode particles at higher engine loads is consistent with previous studies that have shown the formation of nucleation mode particles for vehicles with aftertreatment is a strong function of temperature. Specifically, once a certain temperature is reached, which varies depending on the system configuration, there is a significant increase in the conversion efficiency of SO_2 to SO_3, leading to sulfate based nucleation mode particles [14].

Figure 3 (a) and (b) show real-time particle number concentrations measured by the CPCs that were alternated between the APC and CS sides during the 74% and 26% engine load, respectively, at four different sampling locations and conditions. The locations and conditions were the APC side with a dilution ratio (DR) of 500 (labeled as APC500), the APC side with a DR of 100 (labeled as APC100), the CS side (labeled as CS), and the CVS directly (labeled as CVS). For both engine loads, the tests were performed in the following sequence, the APC with a DR of 500, the APC with a DR of 100, and the CS. At the 26% engine load, this test sequence was conducted twice and an additional CVS test was conducted following the second sequence. At the 74% engine load, this test sequence was conducted three times, and no CVS test was conducted.



(a) (b)

Figure 2 Contour plots of real-time particle size distributions measured by the EEPS in the CVS (a) at the 74% engine load, (b) at the 26% engine load.

Particle number emissions for the two tested cruise cycles were calculated to provide comparisons with emission regulation standards. At the 74% engine load, particle number emissions in the CVS averaged 4.91×10^{13} and 5.29×10^{13} particles/kWh on the APC and CS sides, respectively, as measured by the CPC 3022A (D50 = 7 nm). Both the APC and CS reduced the emission levels significantly. Particle number emissions were 1.15×10^{12} particles/kWh for the APC, as measured by the CPC 3790_APC (D50 = 23 nm) and 8.36×10^{11} particles/kWh for the CS, as measured by the CPC_3772_CS (D50 = 11 nm). These particle number emissions were higher than the proposed Euro VI particle number emission limit for heavy-duty (HD) diesel vehicles, which is 6×10^{11} particles/kWh for the stationary test cycle, World Harmonized Stationary Cycle (WHSC). At the 26% engine load, particle number emissions under the APC and CS were 1.23×10^{11} and 5.58×10^{10} particles/kWh, respectively, which were both below the proposed Euro VI HD limit. It should be noted that the 74% engine load cycle is more aggressive than the WHSC and the 26% engine load cycle is less aggressive than the WHSC.

As shown in Figure 3, particle number concentrations downstream of the APC were always higher than those measured downstream of the CS at both engine loads. Although the CPC 3790_APC (D50 = 23 nm) has a larger cut off size than the CPC 3772_CS (D50 = 11 nm), it reported higher number concentrations than the CPC 3772_CS. The average CPC 3790_APC concentrations were 28% and 44% greater at the higher and lower engine loads, respectively, than the CPC 3772_CS. The differences in particle number concentrations downstream of the APC and CS were more dramatic for those CPCs with smaller cut off sizes, such as the CPC 3025A and CPC 3776. The CPC 3776 concentration was 46% higher with a standard deviation of 5%, on average, on the APC side than on the CS side at the 74% engine load. At the 26% engine load, it was 78% higher with a standard deviation of 13% on the APC side on average. It should be noted that both the APC and CS have particle losses in the system and the data shown here was not corrected for particle losses. Similar particle losses for 50 nm particles of 27% and 30%, respectively, for the APC and CS, have been previously reported [11, 15].

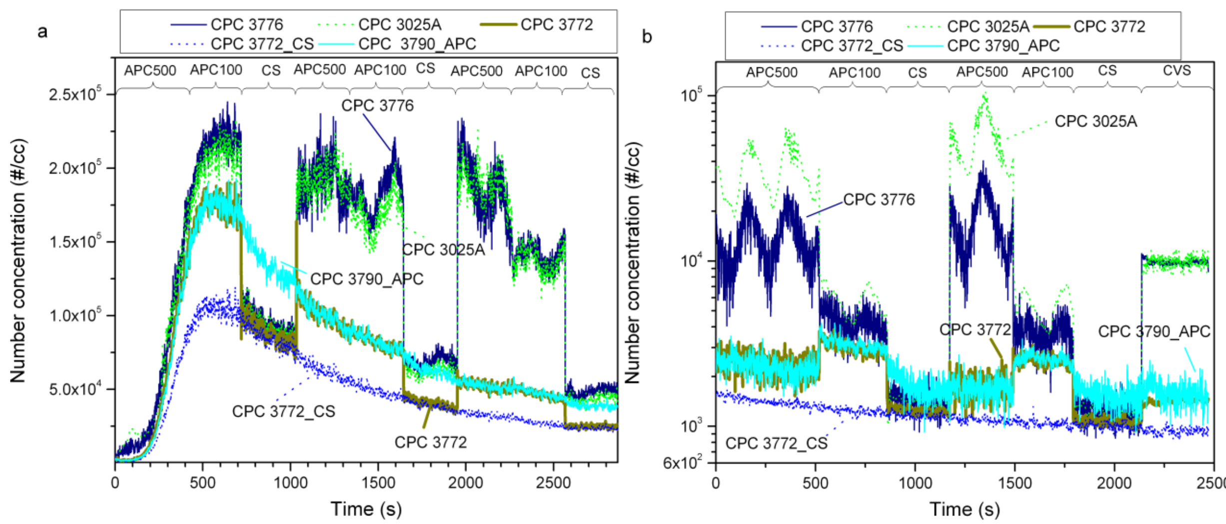


Figure 3 Real-time particle number concentrations measured by different CPCs (a) at the 74% engine load, (b) at the 26% engine load.

To evaluate the size spectrum of sub-23 nm particles downstream of the APC and CS, several CPCs with different cut off sizes in parallel, as shown in Figure 3. On the APC side, the CPC 3772 (D50 = 11 nm) and the CPC 3790_APC (D50 = 23 nm) always agreed to within 5% at both engine loads, indicating a negligible number of particles in the size range of 11-23 nm. The CPCs with the lowest cut off sizes (CPC 3025A and CPC 3776, D50 = 3 nm) showed higher number concentrations than those of the higher cut point CPCs (CPC 3772 and CPC 3790_APC), however, with differences varying from 28% to 90% at the 26% engine load and from 20% to 72% at the 74% engine load. These differences suggested that particles below 11 nm were present downstream of the APC. On the CS side, the CPCs with the lowest cut off sizes also showed higher number concentrations than those of the higher cut point CPCs, but the magnitudes of the differences were less. The differences between the low cut point CPCs and the high cut point CPCs for the CS were ~22% at the 26% engine load and from 11% to 51% at the 74% engine load. Both 3772 CPCs agreed to within 6% when sampling from the CS side, as expected.

4. Conclusion

The comparison study between a European PMP system and a CS showed that the PMP system always measured higher particle number emissions than the CS, regardless of the cut off sizes and engine loads. Both the APC and CS reported particle number emissions above the proposed regulation limits at the higher engine load and below the regulation limits at the lower engine load. Particle below 23 nm were found under the APC and CS.

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