

Particle Emissions from Light-Duty Vehicles during Cold-Cold Start

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ABSTRACT

To ensure reliable starting under cold weather conditions ($< 0^{\circ}\text{C}$ ambient), gasoline engines use fuel enrichment, leading to higher soot formation and greater tailpipe particle number (PN) emissions. In gasoline direct injection (GDI) engines, PN emissions are higher due to liquid fuel impingement on cold surfaces of the combustion chamber and piston. This study characterizes solid (mostly elemental carbon) and semi-volatile (organic) particle number, mass, and size distributions during cold-cold engine start-up from light duty vehicles. Particle emissions were sampled from vehicles upon engine start-up after an overnight soak, with an average ambient temperature of $-8 \pm 7^{\circ}\text{C}$. The average PN emitted during 180 seconds by GDI and PFI vehicles were $3.09\text{E}+13$ and $2.12\text{E}+13$ particles respectively. Comparing to 2017 Euro 6 New European Driving Cycle (NEDC) limit on cumulative particles emitted over the entire test cycle, most PFI and GDI vehicles exceeded this limit in 6-12 seconds after a cold-cold start. In addition, EPA Tier 3 particle mass requirements are exceeded for tested GDI vehicles due to their characteristically high concentration of accumulation mode particles. In comparison, diesel vehicles with DPF's were the cleanest, with particle concentrations close to background levels. Cold-cold starts were followed by a five-minute soak and a second engine start. GDI vehicles' total emitted PN was almost two to three times that of PFI vehicles during restart. This study highlights the need for better particle control strategies to reduce particle emissions during engine start-up in cold ambient temperatures. Our work also confirms that under cold-cold start conditions, most of the total exhaust particles are solid soot and only a small fraction are semi-volatile.

CITATION: Badshah, H., Kittelson, D., and Northrop, W., "Particle Emissions from Light-Duty Vehicles during Cold-Cold Start," *SAE Int. J. Engines* 9(3):2016, doi:10.4271/2016-01-0997.

INTRODUCTION

A cold start is defined as when an engine is started after allowing the engine and its components to reach a temperature of 23°C or below. Likewise, an engine started after reaching a much colder temperature, of 0°C and below is referred to, in this work, as a cold-cold start. Both, cold start and cold-cold engine start-up events are known to produce high particle emissions. Several laboratory studies have been conducted either on a chassis dynamometers or engine test cells outlining higher particle emissions at start-up [1, 2, 3, 4, 5]. The problem largely arises from lower fuel volatility at low temperatures, thereby forcing the engines to run in fuel-rich conditions [6, 7, 8]. A rich mixture ensures that sufficient fraction of the fuel is evaporated at the time of ignition so that a combustible equivalence ratio of the evaporated fuel air mixture is reached (i.e. $\phi \geq 0.5$). At low subzero temperatures, fuel atomization is also impeded, thus liquid fuel can accumulate on cold surfaces of the combustion chamber. The surface to volume ratio of the fuel droplets is also reduced, thereby making the air/fuel mixture less combustible [6]. Fuel condensation on cold combustion chamber surfaces and higher viscosity of the lubricating oil are additional sources of particles during cold start. Enrichment also leads to reduced flame speed, which increases the time needed for complete combustion, thereby leading to incomplete fuel conversion and drastically increasing gaseous and particulate emissions. Furthermore, all modern gasoline engines are equipped with three-way catalysts that take much longer to warm up during

cold ambient temperatures and thus do not control semi-volatile hydrocarbons in vehicle exhaust until they reach their "light-off" temperature [7,9]. Particle emissions can vary among gasoline engines depending on the type of injection methodology. Port fuel injected (PFI) is still most commonly used in gasoline engines, but transition to gasoline direct injection (GDI) engines has accelerated in recent years.

Light-duty passenger vehicles have undergone significant changes in response to emissions regulations for NO_x, hydrocarbons, and particulate matter (PM), as well as greenhouse gases like CO₂ [10,11]. These regulations combined with those for fuel economy, have prompted engine manufacturers to shift from traditional PFI engines to GDI. GDI engines are capable of increasing fuel economy, thus lowering CO₂ emissions, and of producing more power compared to PFI engines [12, 13, 14]. Due to these benefits, GDI market penetration is progressively increasing, expected to reach about 50% in 2016 and surpass PFI engines to about 65% by 2021 [15]. Compared to PFI engines however, GDI engines have much higher particle emissions due to differences in injection methodologies and modes of operation.

Most GDI engines today employ wall-guided injection due to its lower implementation cost, but this has an adverse effect on particle emissions. In wall-guided injection, fuel comes directly into contact with the cold cylinder walls and piston, limiting vaporization as well

as contributing to flame quenching, resulting in incomplete combustion[15,16]. These effects are more prominent in extremely low temperature engine starts, thereby increasing the particle concentrations even more.

The goal of this study was to investigate particle number (PN), mass, and size distributions from PFI, GDI, and light-duty diesel vehicles with diesel particulate filters (DPF's) during and after cold-cold starts. One focus of the study was to analyze GDI powered vehicle emissions compared to the PFI powered vehicles they are replacing in cold ambient conditions. To date, a limited number of studies have been conducted under real-world, in-use conditions reporting gaseous and/or particle emissions during a cold-cold start [17, 18, 19, 20]. This study quantifies the magnitude of particle number/mass emissions during cold-cold starting and differentiates between different engine technologies with the goal to guide future particle emissions control solutions from GDI powered vehicles.

EXPERIMENTAL SETUP

Due to harsh testing conditions in subzero temperatures, a specially designed system for sampling, diluting, and measuring exhaust particles was designed and built. Tailpipe particle emissions were expected to be very high during the cold-cold starts, thus a comprehensive exhaust dilution system had to be implemented. Particle instruments are limited to operating temperatures of above 0°C, thus an added challenge was to transport the exhaust from the tailpipe to an indoor location for particle measurement. This involved using a long (15 m) heated sample line. Furthermore, the system was designed to allow measurement of both total (solid plus semi-volatile) and solid particle number, size, and mass. Semi-volatile particles are formed mainly during sampling and are very sensitive to dilution conditions [21]. Since semi-volatile material was to be sent through the sample line, it was critical that the raw exhaust sample be diluted immediately after exiting the tailpipe of the vehicle in order to mimic atmospheric dilution. The system was designed to allow formation of semi-volatile particles in a stable and repeatable way without excessive coagulation and wall losses. It is known however that a lower sample line temperature would have promoted higher semi-volatile particle formation. The intention of this study was not to study the fate of the particles once they exit the tailpipe, but rather only examine what a cold engine generates in terms of particle emissions. In the actual atmosphere, it is not clear what happens to these particles once they exit the tailpipe, a topic that could be included in future research.

Two, TSI Engine Exhaust Particle Sizers (EEPS) were used to measure PN for this study, one for total PM and another for measurement downstream of a catalytic stripper (CS) for solid PM. The EEPS instruments reported particle size distributions and concentrations at a 10 Hz frequency, allowing for transient particle emissions to be captured during engine start-up. An EEPS responds differently to spherical particles and to soot agglomerates and uses a different internal inversion matrix for the two different particle types. A fully updated soot inversion matrix was used for both EEPS instruments to better reflect the particle size and concentrations associated with engine exhaust particles [22]. A Scanning Mobility

Particle Sizer (SMPS) and Condensation Particle Counters (CPC) were also used to perform loss check experiments on the sample line used for this study.

Dilution System

The sampling/dilution system used in this study was split into three sections: primary tailpipe dilution, sample line transport, and secondary dilution as shown in Figure 1. The sample line consisted of two concentric 15 m long lines, both enclosed together in an insulated outer cover. The core that transported the exhaust was composed of carbon impregnated Teflon and the outer Teflon line was used for dilution air delivery to the tailpipe location. The sample line was maintained at a temperature of 47 °C. CFR regulations for PM mass determination using filtration require a dilution temperature of 47 °C ± 5 °C (CFR § 86.1310-2007). Although the application and objective of this study differs from emissions testing in the laboratory, 47 °C was selected here to avoid water condensation and prevent freezing. It should be noted that dilution conditions like temperature and residence time can have a significant impact on particle size and concentration, especially of semi-volatile particles [23].

The primary stage of tailpipe dilution consisted of a short probe section followed by an ejector diluter. An ejector diluter is a well-established method of simultaneously diluting raw exhaust with minimal losses and establishing good mixing [24]. 80 slpm of HEPA filtered dried compressed air, controlled using a mass flow controller, and was delivered to the tailpipe location via the outer Teflon line. The airflow entering the ejector created a vacuum at the probe location. A 1.14 mm orifice was placed upstream of the ejector pump to limit the flow from the probe to about 8.5 slpm. This gave a primary dilution ratio, DR1, of ~10.5:1. The dilution ratio (DR) is defined as:

$$DR = \frac{\text{air flow} + \text{sample flow}}{\text{sample flow}} \quad (1)$$

A thermocouple was placed just upstream of the pressurized air inlet and the vacuum/orifice side to ensure that these locations were also at 47 °C. The entire test section from the sample probe to the sample line was maintained at this same temperature. The 8.5 slpm and 80 slpm of compressed air mixed in the ejector and were dispersed in two ways. A bypass was placed just upstream of the sample line, thus causing most of the diluted sample to exit at this point. A part of the diluted flow entered the sample line, being drawn by another ejector located downstream of the sample line. At the downstream location, 75 slpm of air was delivered to the 2nd ejector, creating a vacuum through a 1.37 mm orifice that drew 13 slpm through the 15 m sample line. A secondary dilution ratio (DR2) of 7:1 was achieved at this location, resulting in a total DR of ~ 75:1. An EEPS instrument was located just downstream of this ejector to measure the total PM. The rest of the diluted flow was either exhausted or entered the catalytic stripper (CS) followed by another EEPS instrument for solid particle measurement. The oxidation catalyst in the CS was maintained at 300°C and was followed by a copper cooling coil to cool the sample exiting the CS down to ambient temperature suitable for the EEPS instrument. The main purpose of the CS was to remove semi-volatile particles and only pass solid particles [25].

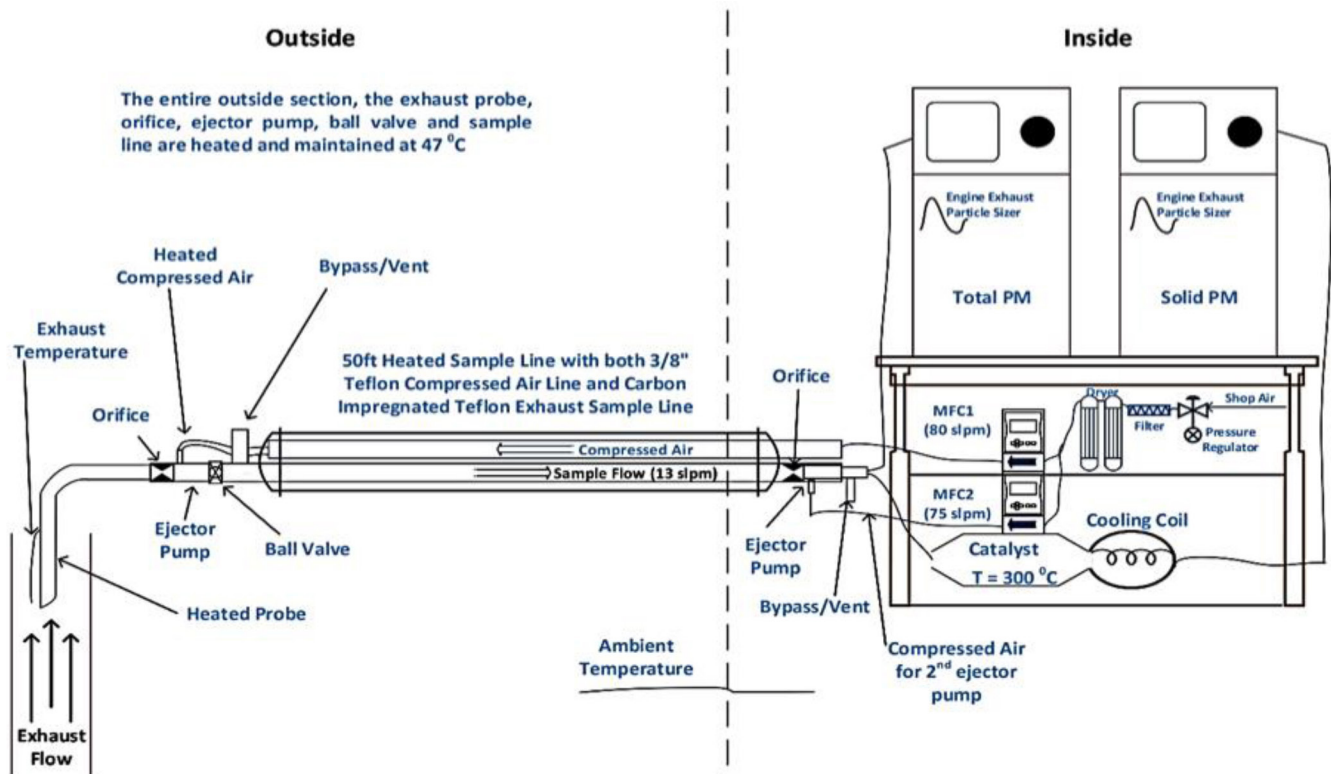


Figure 1. Schematic of vehicle particle sampling apparatus

Loss Checks

Long sample lines lead to significant losses of ultrafine particles (<100 nm), therefore necessitating a proper particle loss evaluation of the system being used for this study [26]. A theoretical model was used to predict particle losses in the 15 m sample line. Among the considered loss mechanisms were: diffusion, thermophoresis, inertial, and electrostatic [27, 28, 29]. Except for cooling between the probe tip and the first dilution stage, the sample stream was kept at the same temperature so that thermophoresis effects were minimal, and most of the losses were associated only due to diffusion. Although the model should have provided a good basis for loss corrections, it was crucial to verify the findings via experimental efforts. As discussed below, doing this revealed critical information that allowed for better loss corrections in the sample line.

Solid particles used in the loss check experiments were generated using a miniCAST soot generator. A catalytic converter was used downstream of the miniCAST to remove the volatile fraction of particles. Different fuel, air, and quench gas settings were used on the miniCAST to vary the particle size distributions [30]. The dilution system, composed of the sample probe, ejectors, and the 15 m sample line were tested as one entire system to obtain the net particle penetration. A model 3081 long DMA was used to size select particles at 100 nm, 70 nm, 50 nm, 30 nm, and 15nm electrical mobility diameters. The polydisperse source's mean particle size entering the SMPS was always maintained below the selected SMPS size for loss checks. This was done to avoid effects of doubly charged particles that could pass from the DMA [31]. The polydisperse

distribution entering the DMA was measured using an EEPS to ensure it was within the expected size range. Two calibrated CPC 3025's were used, one upstream and the other downstream of the dilution system. The CPC's were calibrated to ensure comparability.

The dilution system was maintained at the same conditions (sample flows, pressures, and temperatures) as those used during the actual cold start experiments. The size-selected solid particles from the DMA were mixed with HEPA filtered dilution air (to provide sufficient flow to enter the dilution system ~ 8.5 slpm and the 1.5 lpm required by the upstream CPC. The mixed flow was transported to the inlet of the dilution system where the upstream CPC was located. The sample then entered the dilution system and followed the same dilution process as previously described. After secondary dilution, the diluted monodisperse flow of particles was sampled by the downstream CPC, and the remainder of the flow was exhausted. The upstream and downstream CPC were monitored for at least five minutes to allow for a stable concentration to be reached. Once this occurred, the upstream and downstream CPC concentrations were recorded for five to ten minutes. This procedure was repeated for all five size set points mentioned above. Figure 2 shows the particle penetration in fraction form as determined by the equation below.

$$\text{penetration} = \frac{(C_a \cdot DR)}{C_b} \quad (2)$$

Where, C_a is the concentration of particles measured after dilution, C_b is the concentration of particles measured before dilution and DR is the dilution ratio.

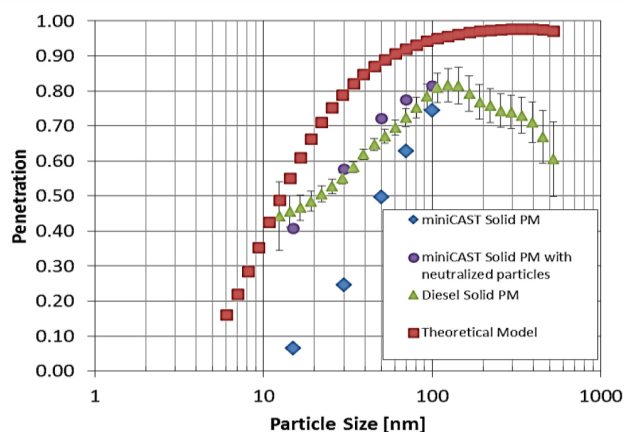


Figure 2. Particle penetration for entire dilution system - does not include losses for solid particles through the catalytic stripper

The experimental penetration differed significantly from the theoretical model calculations. At 100 nm the particle penetration was only 0.75 and dropped to 0.25 at 30 nm, but the model predicted penetration was 0.94 and 0.80 at 100 nm and 30 nm respectively. Due to the large difference in particle penetration, another round of loss check experiments was conducted with a slightly different configuration. A Krypton 85 neutralizer was added downstream of the DMA so that particles exiting the DMA would be neutralized and consist of a Boltzmann distribution of charge [31]. Without the added neutralizer, all particles exiting the DMA are singly charged, but a room temperature Boltzmann distribution only consists of approximately 20% charged particles. Repeating the same loss check experiments with the added neutralizer resulted in significant improvements in the particle penetration. This exercise revealed that the charge on the particles had an enormous effect on the losses through the sample line, indicating the possibility of a nonconductive core used to transport the particles.

After communication with the manufacturer of the sample line about this issue, it was discovered that although the sample core used for particle transport was made of “carbon impregnated Teflon,” its level of carbon content was very low and thus had little impact on its conductivity. Particle sampling is always conducted with conductive tubing because materials such as plain PTFE (Teflon) develop local electric fields and act as an electrostatic precipitator and remove charged particles [32]. This information about the sample line’s conductivity was unfortunately discovered after the cold-start experiments had been conducted.

Due to the nonconductive nature of the sample line used for this study, it became imperative to investigate the effect of charged particles on particle penetration through the sample line. Particle penetration was no longer simply a function of diffusion, but also electrostatic effects. A change in the charge distribution of the entering particles would also change the particle penetration. Additional testing was conducted in an engine test cell with a John Deere diesel engine exhaust that would approximate the charge on the particles of gasoline engines. Kittelson et al. investigated the charge on particles from a diesel engine exhaust and found that only a small percent of particles below 50nm exist with a charge [33]. It is predicted that gasoline engines, especially GDI vehicles would emit largely neutral particles and thus would provide

similarly charged particles as those from the John Deere diesel engine. The Deere engine exhaust was diluted with a two-stage ejector diluter system along with a catalytic stripper for volatile particle removal to generate a steady source of solid particles. Two different testing modes were used: 1400 rpm with 100 Nm torque and 1400 rpm with 250 Nm torque. The two testing modes provided particles that spanned most of the EEPS size range. These tests were different from the miniCAST tests in that polydisperse rather than size classified particles were used and upstream and downstream measurements were made by switching back and forth with a single EEPS. Despite these differences, it is seen in Figure 2 that the results from diesel particles are consistent with those from neutralized miniCAST particles.

From the various loss check experiments described above, the penetration values from that of the diesel particles was used to make actual corrections to the cold start data. Below 12 nm, due to a lack of a high concentration of particles from either the miniCAST or the diesel engine, the theoretical model’s penetration values were used. It is important to note that the particle penetration fractions used for correcting concentration data are a best estimate of the authors based on the discussion above. An accurate penetration factor could only be obtained if the actual charge distribution of the particles from each vehicle during the start-up tests were known. Due to the uncertainties in the charge distribution expected from the actual starts, it is estimated that the particle penetration values used to make the official corrections are within $\pm 20\%$ of actual values.

Catalytic Strippers have significant losses of solid particles, mainly due to diffusion and thermophoresis. The hot sample stream leaving the catalyst is cooled in a cooling coil to ambient temperature to allow measurements with standard aerosol instruments. This cooling effect leads to particle losses via thermophoresis. Penetration values used here were taken from a detailed study of losses through the same catalyst plus cooling coil configuration that was conducted earlier [25]. The total penetration correction for solid particles was then a simple multiplication of the penetration through the dilution system times the penetration through the catalytic stripper.

Vehicle Types

All vehicles used in this study were rental vehicles that had below 50k miles, with most being below 25k. Only the diesel w/DPF vehicle tested had more than 100k miles. Additionally, the pool of tested vehicles comprised of only model year 2010 and newer. A total of 11 PFI, 10 GDI, and 2 diesel w/ DPF vehicles were tested.

Cold-Cold Start Procedures

Every vehicle to be tested was parked overnight (duration of at least 12 hours) in overnight temperatures that dropped down to -20 ± 10 C. To simulate the real-world operation of vehicles, in the evening before the test day, each vehicle was driven on highway speeds for approximately 5-10 miles for preconditioning. This might approximate urban driving with the vehicle driven on a highway near the end of the day when individuals are returning from their daily activities and then parked overnight. Additionally, the vehicles obtained in this study were rental vehicles with no previous driving history. Because of this, in addition to avoid any lingering