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Very low particle matter mass measurements from light-duty vehicles

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ABSTRACT

Recently, there are discussions about whether current sampling and measurement practices for the regulated gravimetric PM measurement are sufficiently accurate in quantifying PM at the proposed 3 and 1 mg/mi emission standards for light-duty vehicles. In this study, a series of modifications were made to the existing gravimetric PM measurement method, aiming to preserve the integrity of the method while increasing the robustness and decreasing the testing variability. The experiments were conducted with a Higher (~2 mg/mile) and a Lower (0.1–0.2 mg/mile) PM Source Vehicle over the Federal Test Procedure (FTP) and US06 cycles, providing PM emissions with various solid/semi-volatile compositions and size distributions. The results showed the suggested modifications, i.e., increased filter face velocities (from 100 to 150 cm/s) and combined filters (single filter vs. 3/4 filters), could increase the collected filter mass without introducing statistically significant differences in the measured PM mass emission rates. No statistically significant improvements were seen in the measurement variability with the Higher PM Source Vehicle. For the Lower PM Source Vehicle; however, the 4-phase cumulative filter showed a statistically significant reduction in PM mass measurement variability, while not impacting the measured PM mass emissions, but these improvements must be weighed against the increased testing costs/time required for the longer test time.

1. Introduction

Present motor vehicle particulate matter (PM) mass emission regulations (Code of Federal Regulations [CFR] Title 40 Parts 1065 and 1066) require gravimetric determination of PM collected onto filter media from a diluted exhaust stream (CFR, 2001,2002). These regulations were put in place to address issues in making PM measurements for the 2007 PM standard for heavy-duty engines (HDEs), when diesel particulate filters were essentially implemented in the U.S (Khale, 2005a, 2005b, 2005; Swanson, Kittelson, & Dikken, 2009).

Reductions to PM mass emission standards are now also being implemented for light-duty vehicles (LDVs). These regulations have been of particular interest since reductions in corporate average fuel economy levels have led to the introduction of gasoline direct injection (GDI) vehicles, which generally have higher PM mass emission rates relative to more traditional port fuel injection (PFI) gasoline vehicles. The PM emission standards for LDVs will be lowered from 10 to 3 mg/mi for the United States Environmental

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Protection Agency (EPA) Tier 3 and the California Lower Emission Vehicle (LEV) III standards by 2017, with an additional reduction to 1 mg/mi expected in 2025 as part of the California LEV III requirements (CARB, 2012; USEPA, 2013). While the 2007 changes to the PM measurement methodology considerably improved measurement practices, there is a remaining need to improve the understanding of and confidence in mass measurements for LDVs, given the implementation of these new standards.

Currently, the protocol for light-duty chassis dynamometer testing (CFR Part 1066) refers to the heavy-duty engine testing regulations (CFR Part 1065) for most PM measurement parameters, even though the equipment and methods used to determine PM emissions in light-duty chassis dynamometer laboratories are generally quite different from those used in heavy-duty engine dynamometer laboratories. For example, most heavy-duty laboratories have secondary dilution tunnels to manage the dilution factor (DF), filter face temperature, and condensation for PM measurements, but for LDVs, secondary dilution tunnels are not typically used. The HDV Federal Test Procedure (FTP) is also a single phase test with a single filter, while the LDV FTP test is composed of three phases utilizing one filter per phase along with different weighting factors to determine cycle averaged PM emissions.

Recently, there has been discussion about whether current sampling and measurement practices are sufficiently accurate in quantifying PM at the proposed 3 mg/mi standards, and even more so at the 1 mg/mi PM emissions standards for LEV III LDVs (CFR, 2011, 2012). Studies by Hu et al. (2014) have suggested that the gravimetric method is sufficiently accurate to measure PM emissions at the levels for both the 3 and 1 mg/mile standard. There is uncertainty, however, if these methods can be more widely implemented under the conditions seen for certification testing, and a number of strategies are still being investigated to further improve the understanding of and confidence in mass measurements for LDVs at low levels. Increasing the filter face velocity (FFV) is one of them. Currently, the FFV specified in CFR Part 1065 is up to 140 cm per second (cm/s), which represents approximately 84 l per minute (lpm) through a 47 mm-diameter filter. Increasing FFV has been shown to reduce filter efficiency based on model predictions from the University of Minnesota, but it is unknown what the real filter efficiency will be (see Section S1 in Supporting Information (SI)). Zhang and McMurry (1987, 1991) examined the dependence of the PM collected on a filter on the FFV and found that higher FFVs could minimize sampling artifacts associated with gas phase adsorption, but can also increase volatilization of the PM, although this was for ambient particles that are largely semi-volatile. Similar results were reached in the CRC E-66 study (Khale, 2005a, 2005b, 2005)..

Combining filters is another method to increase the filter mass collected that can be utilized for the LDV FTP test. A single cumulative filter approach as opposed to using one filter per phase was examined by Andersson et al. (2007), but using different filter media of TX40. They found that a relatively low coefficient of variation of 15% could be obtained for exhaust PM levels of ~1 mg/mile using a cumulative filter method. Maricq, M. M., Szente, J., Loos, M., & Vogt., R. (2011) also suggested that use of a single filter can reduce both the magnitude of artifacts and uncertainty.

Lowering the DF, or the ratio of the volumetric flow rate of the dilution air to that of the raw engine/vehicle exhaust, is another strategy to increase the mass collected on a filter. Although it is desirable to reduce DFs, this approach raises the possibility of increasing water condensation and other uncertainties (Hood & Silvis, 1998; Kittelson, Arnold, & Watts, 1999; Maricq, Chase, Podsiadlik, & Vogt, 1999). Other strategies for improving PM mass emission measurements include using more advanced weighing strategies, such as using an automated robotic weighing system, and utilizing real-time or other measurement methods or methodologies (Park, Kittelson, McMurry & P. H, 2003; Swanson & Kittelson, 2008; Bushkuhl, Silvis, Szente, & Maricq, 2013; Xue et al., 2015, 2016, 2017; Quiros & Zhang, 2015; Quiros, Hu et al., 2015; Li et al., 2014).

The goal of this study was to investigate PM sampling methodologies that extend beyond the recommendations of CFR Part 1066 and 1065. The objective was to evaluate how differences in sampling methodology could improve accuracy, reduce variability, and increase the signal-to-noise ratio of the measurement. The modifications investigated were limited to varying FFV and using cumulative filters, with some testing also to examine DF effects. The results provide the most detailed information available to date to characterize the fundamental limitations of the current gravimetric PM method and its application for LDVs. This is becoming increasingly important given the introduction of a wider range of vehicle technologies designed to achieve increased fuel economies that may have a wider range of PM emission rates. This information may also provide value for low level PM mass sampling for ambient or other low PM emitting sources.

2. Experimental section

2.1. Test vehicles, test cycles, and study design

Vehicles were tested over two regulated driving cycles, the FTP cycle and the US06 cycle. The FTP is the primary emission certification test for all LDVs in the U.S., and the cycle to which the 3 mg/mi and 1 mg/mi standards will be applied. The FTP includes 3 phases designed to represent different types of driving. This includes a “cold start” phase 1, which represents operation when the vehicle is first started for the day, a “stabilized phase” 2, which represents driving after the vehicle is warmed up, and “hot start” phase 3, which is a repeat of phase 1 conducted after the engine has been turned off for 10 min. The different phases are then weighted using the factors of 0.43 for the cold start phase, 1.0 for the stabilized phase and 0.57 for the hot start phase to obtain a composite emission rate for the full cycle. The FTP can also be run as a 4 phase test by repeating the phase 2 stabilized driving cycle immediately after phase 3. The 4-bag FTP was introduced in the 1970s, but was subsequently modified to allow 3 phase testing as well. For the 4 phase FTP test, weighting factors of 0.75 are used for phase 1 and phase 2, and 1.0 for phases 3 and 4 to obtain a composite emission rate for the full cycle that is mathematically the same as for the 3-bag FTP, assuming that phase 2 and phase 4 are essentially equivalent. The 3-bag and 4-bag FTPs have been shown to be equivalent with the use of these appropriate weighting factors Danielson (1979). During preliminary testing for this study, we also found that PM emission rates for 3-bag and 4-bag FTPs approaches showed good agreement well within the experimental uncertainty for the Lower PM Source Vehicle, as discussed in more detail in section S2 in the SI. The US06 cycle is a single phase test that was incorporated into the regulatory procedures as part of the Supplemental Federal Test Procedure (SFTP) beginning in 2000, and is designed to represent more aggressive driving behavior. The driving schedules for the FTP and US06 are provided in the Supplemental information (Section S3). For both the FTP and US06, the driving schedules are followed to within ± 2 mph on a second by second basis, except in cases where the vehicle cannot achieve the desired acceleration rates.

Since the FTP is a 3 or 4 phase test, individual PM filters are typically used to obtain PM mass emissions for each individual phase. PM emissions can also be obtained using a single composite filter that collects PM cumulatively over the entire FTP. When a single cumulative filter is utilized as opposed to individual filters, the flow rates for the different phases must be adjusted to ensure that the cumulative filter provides the appropriate weighting between the different phases for the composite emissions factor, as described in CFR Part 1066.815(b). Thus, the FFV is adjusted to 43% of the nominal phase 2 FFV for phase 1 and 57% of the nominal phase 2 flow rate for phase 3 for the 3 phase test. For the 4 phase FTP, the FFVs for phase 1 and 2 are adjusted to 75% of the nominal FFVs for phases 3 and 4. The use of lower FFVs for phase 1 is a particularly important consideration as gravimetric filter measurements by phase show that between 62% and 100% (in cases where the PM levels for bags 2 and 3 are below background levels) of the PM mass is emitted during bag 1 for the test vehicles in this study. As such, in comparing cumulative filters for the 3- and 4-phase FTPs, the 4-phase FTP provides more PM mass gain with the cumulative filter approach because it includes an additional phase, and more importantly because it utilizes a 74% higher flow rate during the cold start phase 1 where most of the PM mass is emitted. A 4-phase FTP test does take ~35% longer test time compared with a 3-phase FTP test, however, which is a disadvantage in a high production testing environment.

Two test vehicles were used for this study, a Lower PM Source Vehicle (a PFI vehicle) with an FTP emissions rate of ~0.1 mg/mi and a Higher PM Source Vehicle (a GDI vehicle) with an FTP emission rate just below 2 mg/mi. These vehicles were targeted to be representative of the PM emissions levels that new vehicles meeting the upcoming 3 mg/mi and 1 mg/mi standards will have going into the foreseeable future. Looking forward, the range of light-duty powertrains will include, gasoline PFI, GDI, GDI with particle filter, diesel with particle filter, and CNG. The Lower PM Source Vehicle used here is representative of PFI and GDI with particle filter vehicles, while the Higher PM Source Vehicle is more representative of GDI vehicles without a particle filter. The PM (mass) emissions from CNG and diesel with filter are likely to be extremely low, comparable to the levels seen for Lower PM Source Vehicle.

Testing with two vehicles with different PM emission levels and over two cycles provided PM emissions with different solid/semi-volatile compositions and particle size distributions (PSDs) (Section 3.3), which are important characteristics in understanding the impacts of the alternative parameters investigated in this study. All tests were conducted in CE-CERT's Vehicle Emissions Research Laboratory (VERL) equipped with a Burke E. Porter 48-inch single-roll electric dynamometer. The dynamometer room is maintained within temperature limits of 20–30 °C specified in the regulations, with testing typically conducted at a room temperature of 21–24 °C. Detailed specifications of the vehicles and the test cycles are provided in Table S4 and Section S3 in SI, respectively.

The Higher PM Source Vehicle was tested using a 3-phase FTP, while the Lower PM Source Vehicle was tested using a 4-phase FTP. Overall, six FTP tests were completed for both vehicles and six US06 tests were completed for the Lower PM Source Vehicle. Only two US06 tests were completed with the Higher PM Source Vehicle, however, because the engine overheated and was unable to maintain the US06 trace after two tests. Some preliminary testing was conducted at the start of the program to evaluate a broader range of test conditions in terms of FFV, DF, 3 vs. 4 phase FTPs, and cumulative filters and to identify conditions under which stable and repeatable PM emissions are obtained while providing the most PM mass on the filters. Results from the preliminary tests are presented in Johnson et al. (2015). It should be noted that during the preliminary testing, the PM mass variability for the US06 testing was found to be two times higher compared to the FTP for both vehicles, which suggested that the preparation cycle for the US06 may be more critical for PM emissions than gaseous emissions. As such, the US06 cycles for the final testing were run following a single preparation FTP cycle, followed by a preconditioning US06, a 2 min soak, and then the actual US06 emissions test was run. For more details about the impact of the precondition of the vehicles, please refer to Johnson et al. (2015).

2.2. PM sampling

A schematic of the laboratory setup is shown in the Abstract graph and Fig. S9 in the SI. Two unique PM samplers were utilized in this study: a multi-filter PM sampler in a constant volume sampler (CVS) system and a single filter partial flow dilution system (PFD).

2.2.1. CVS

A multi-filter sampler was utilized that simultaneously collected PM on four different gravimetric filter samplers from the dilute CVS to evaluate parameter changes in parallel. This sampler utilized a single heated sample probe, a particle impactor, a heated and temperature controlled chamber for the various filter holders, a residence chamber designed to provide a residence time of 2.5 s from the point at which the exhaust is first diluted to the point where the PM sampling media is located, CFR Part 1065 compliant filter holders with filter cassettes, solenoid bypass valves, and four mass flow controllers (MFCs). The heated chamber is maintained at 47 ± 5 °C, and contains four filter samplers. Samplers A, B and C were designed to collect a cumulative PM filter at different nominal FFVs during FTP or US06 tests, while sampler D represented the traditional CVS filter sampling system, which collected filters for the individual phases of the FTP. The nominal FFVs for samplers A, B, C, and D were 150, 125, 100 and 100 cm/s, respectively. Note that 100 cm/s was used as the base case for this study, as this was the FFV specified in the CFR at the time this study was conducted. The actual FFVs for the individual FTP phases could be different for samplers A, B and C, due to different flow weightings per 1066.815(b). Preliminary test results showed the MFCs could not maintain a FFV of 175 cm/s for the Higher PM Source Vehicle

Table 1
Test matrix design for confirmation testing (n = 6 except for GDI US06).

Cycle[a]	Vehicle	Test	CVS sample (DF = 7)[b]		PFD Sample[e]	
			FFV(cm/s)			
		No.	1 Filter/phase[c]	1 Filter/FTP[d]	1 Filter/FTP	
			"D"	"C", "B", "A"	FFV	DF
4-phase FTP	Lower	6	100	100, 125, 150	150	5
3-phase FTP	Higher	6	100	100, 125, 150	150	5
US06	Lower	6	100	100, 125, 150	150	5
US06	Higher	2	100	100, 125, 150	150	5

[a] FTP is performed as a cold start FTP. The US06 is performed as a hot start test. The US06 preparation is a FTP, followed by a US06 (prep) followed by a US06 (test) where the time between the US06 (prep) and US06 (test) is between 1 and 2 min. Also the next repeat will be performed on the following day. The US06 test with the Higher PM Source Vehicle (GDI vehicle) was not able to maintain the speed trace due to possible cooling issues; thus only two tests were completely.

[b] The CVS will remain at DF = 7 where the DF is based on Phases.1 for the FTP.

[c] CVS Sampler “D” is a complete sampler and configured for one filter per phase.

[d] CVS Samplers “C”, “B”, and “A” are three separate samplers. Flow- weighting is as per 1066.815(b)(4) [0.43, 1.0, 0.57] and (b)(5) [0.75, 0.75, 1.0, 1.0].

[e] The PFD will operate at DF = 5 where the DF is based on phase 1 for the FTP.

during the 4-phase FTP test due to increased pressure differential of around 10 in Hg across the filter and limitations with the MFCs. This suggested that 150 cm/s used with sampler A represented the approximate upper flow limit achievable with current flow controller technology in this application. The CVS flow was set to provide a mean DF for phase 1 of the FTP and for the US06 of 7, based on the carbon balance calculation in CFR Part 1066.610. This provided average DFs from 8 to 12 for the other phases of the FTP, depending on the vehicle. These DFs are sufficiently high to prevent condensation, but allow for higher amounts of PM to be collected on the filter. The actual DFs will oscillate above and below the mean value during a transient test, but predicted relative humidity at the filter surface will never reach a level leading to condensation at 47 °C (see Johnson et al., 2015). An AVL Micro Soot Sensor (MSS, Model 483 AVL Inc.) and an Engine Exhaust Particle Sizer (EEPS, Model 3090, TSI Inc.) sampled directly from the CVS to provide real-time measurements of soot concentration and PM PSDs. It should be noted that the transfer line between the vehicles and the CVS was insulated and kept as short as possible to minimize any potential sampling artifacts. Table 1 lists the test conditions for the FTP and US06 cycles

2.2.2. PFD

The PFD used for this study was a commercially available CFR Part 1065 compliant system (AVL Smart Sampler_478). The PFD was mounted upstream of the CVS and directly sampled from the raw exhaust. A PFD can help reduce interference from artifacts by allowing lower DFs and avoiding the need for a long transfer hose for the full exhaust, which can cause storage/release effects (Foote et al., 2013). The PFD used a sample flow proportional to the total exhaust flow, mimicking the varying dilution in the CVS as a function of exhaust flow rate. In this study, the PFD extracted approximately 1–4% of the total exhaust volume, depending on the driving condition and FFV, and the CVS PM mass emission rates were corrected to account for the fraction of exhaust extracted by the PFD. The DF for FTP phase 1 was set to be 5 for the PFD. Our preliminary tests showed a lower DF (< 5) will cause overloading of the filter and prevent the PFD from maintaining an FFV of 150 cm/s. A DF of 5 could not be applied to the CVS, on the other hand, due to the greater potential for condensation that was prevented in the PFD with a heated line at 47 ± 5 °C. Methods to calculate dilution factors for CVS and PFD systems were introduced in Section 4 in SI.

2.3. PM mass emission measurements

PM sampling was conducted following the procedures in CFR Part 1066.110 and associated references in CFR Part 1065, with the exception of changes to the protocol designed to test experimental extremes in variables. Total PM mass samples for both the CVS and PFD were collected using Whatman 47 mm polytetrafluoroethylene (PTFE) filters. They were weighed using a CFR Part 1065-compliant microbalance with a neutralizer in a conditioned room meeting CFR Part 1065 requirements. Filters were weighed at least twice both pre- and post-test until two measurements within 3 μg were obtained. The stability of the weighing conditions was also monitored with 5 Teflon reference filters, that were weighed at least daily and sometimes hourly, which showed standard deviations of between 1.0 and 2.0 μg.

The PM emission results were background corrected based on average tunnel blank filter masses collected periodically over the course of the program. Tunnel blanks were collected over a test with the same duration as the cumulative or individual FTP phases or US06 tests, but without exhaust flow. The average tunnel blank equaled to 5.0 ± 4.0 μg (average ± standard deviation, No. = 20) for the CVS probes and 0.8 ± 0.6 μg for the PFD sampler (No. = 2). In this study, the same tunnel blanks were applied to all CVS filter probes over both FTP and US06 cycles. In some cases, the background correction resulted in negative filter weights, but only for individual bag 2, 3, and 4 results for probe D. Since the objectives of this project were to understand biases, variability, and mean differences, these negative filter weights were allowed to be negative, and no truncation was performed, for the evaluations relating

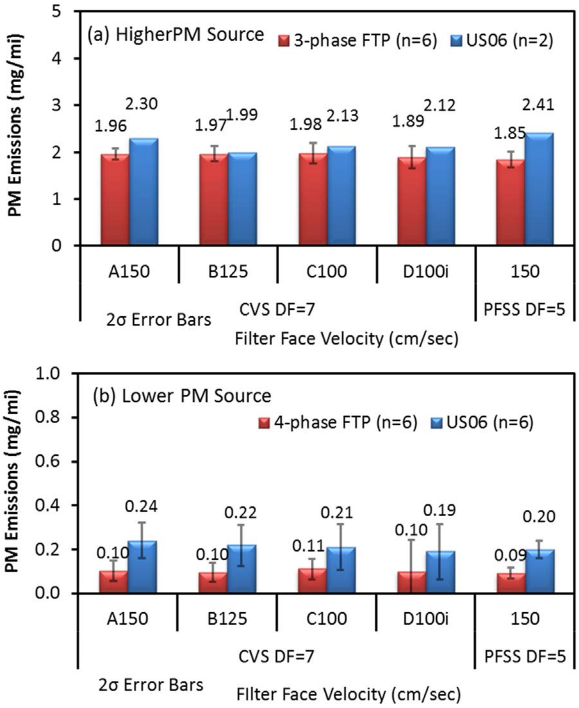

Fig. 1. PM emission rates for the CVS and PFD samplers (a) Higher, (b) Lower PM Source Vehicle. [1] Data are corrected for tunnel blanks. Error bars represent two standard deviations (2σ). The labels A150, B125, C100, and D100i, denote different sampling samplers (i.e., A, B, C, and D) along with the associated FFV (i.e., 100, 125, and 150). The D100i represents the Sampler D system with individual filters per phase; the others represent cumulative filters. The error bars of the Higher PM Source Vehicle with US06 cycles was not presented because the GDI vehicle was unable to maintain the US06 trace, reducing the sample size to n = 2, which means that the data variability may be higher due to the operational difficulties.

to the total weight of PM mass collected on a filter (e.g., Fig. 2). However, negative filter weights were zeroed out prior to calculating emission factors (e.g., Fig. 1), following CFR 1066."

3. Results

3.1. PM mass emissions (mg/mi)

The PM emissions for both vehicles are shown in Fig. 1 for the FTP and US06 test cycles. The PM emissions were in close agreement for the Higher PM Source Vehicle and averaged between 1.85 and 1.98 mg/mi for different PM samplers (with individual tests ranging from 1.73 to 2.12 mg/mi) over the FTP cycle and between 1.99 and 2.41 mg/mi for different PM samplers (with individual test ranging from 1.92 to 2.64 mg/mi) over the US06 test cycle.

The Lower PM Source Vehicle had much lower overall PM emission levels. Average emission rates measured with the different samplers ranged from 0.09 and 0.11 mg/mi (with individual tests ranging from 0.04 to 0.24 mg/mi) over the FTP test and 0.19–0.24 mg/mi (with individual tests ranging from 0.10 to 0.30 mg/mi) over the US06 test. The differences between individual samplers and the overall average for all the samplers were only 0.02 and 0.05 mg/mi for the FTP and US06 cycle, respectively. For the FTP, this represents less than 1% of the 3 mg/mi standard and 2% of the 1 mg/mi standard.

The above results suggest good consistency among samplers. However, although individual samplers tended to agree with one another, there was significant test-to-test variability, with maximum differences of 0.72 and 0.20 mg/mi for the Higher PM Source Vehicle and Lower PM Source Vehicle, respectively, suggesting significant test-to-test variability of the vehicle emission rates. This is consistent with the results of other studies of low level PM measurements from LDVs (Kittelson et al., 1999).

3.2. PM filter weights (μg)

The amount of mass collected on the PM filters is an important consideration in terms of understanding the levels of PM being sampled, and the applicability of these results to other types of PM measurements. Fig. 2 shows the accumulated PM filter weights for all the PM samplers. The filter weights increased with increased FFVs and reduced DFs for both test cycles and both vehicles, indicating that strategies can be used to increase the signal to noise ratios for PM mass measurements. For the FTP tests the Higher PM Source filter weights varied from 205 to 417 μg for the C100 samplers and PFD, respectively, while the Lower PM Source filter weights varied from 26 to 46 μg for the 4-phase FTP test for the C100 and PFD, respectively. The US06 test cycle showed lower filter

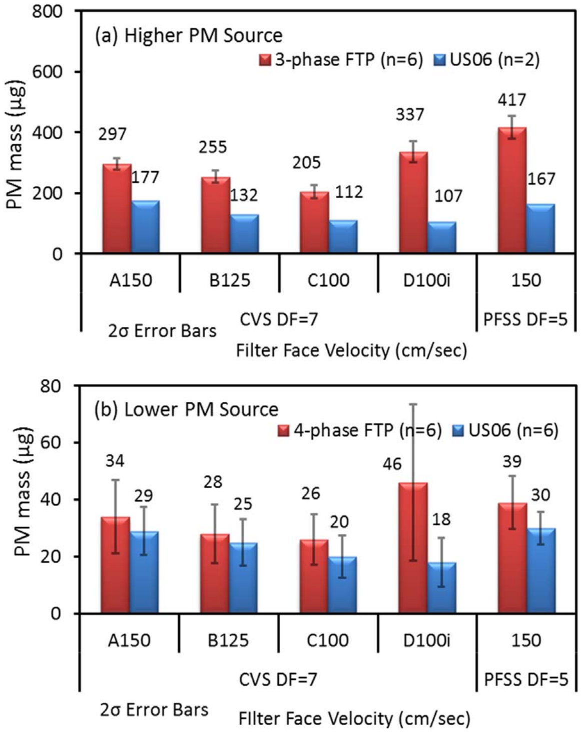

Fig. 2. PM filter mass gain for the CVS and PFD samplers (a) Higher, (b) Lower PM Source Vehicle. [1] Error bars represent two standard deviations (2σ). The labels A150, B125, C100, and D100i, denote different sampling samplers (i.e., A, B, C, and D) along with the associated FFV (i.e., 100, 125, and 150). The D100i represents the sampler D system with individual filters per phase; the others represent cumulative filters. The Higher PM Source Vehicle was unable to maintain the US06 trace, which reduced the sample size to n = 2, which means that the data variability may be higher due to operational difficulties.

weights compared to the FTP test cycle, despite the fact that the US06 is a more severe high speed cycle with typically higher emission rates. This is because it does not include a cold start, and also because it is much shorter in time, 596 vs 1877 s, and somewhat shorter in distance, 8.01 vs 11.04 miles, compared to the 3 phase FTP with even greater differences compared to the 4 phase.

3.3. Solid/semi-volatile composition of PM and particle size distribution

Particle composition was not measured directly, but MSS and size distribution results are suggestive. The Higher PM Source Vehicle showed a nearly linear relationship between black carbon (BC) mass measured with MSS and filter mass during FTP tests as shown in Fig. 3. The slope was 0.97 suggesting that nearly all the mass was BC. On the other hand, for US06 there were only 2 successful tests and the ratios of BC to filter mass ranged from 0.5 to 0.6 suggesting the particles were nearly half semi-volatile. During FTP testing, the Lower PM Source gave nearly constant BC emissions but variable filter mass emissions with the ratio of BC to filter mass ranging from 0.7 to 1.2. This suggests that a variable amount of semi-volatile material that is the source of most of the variation of filter mass in this case. For the US06 tests, there is much more semi-volatile material present with the ratio of BC to filter mass varying from about 0.4–0.9. Again, most of the variation in filter mass is due to semi-volatile material. Xue et al. (2017) also showed a bigger contribution of adsorbed semi-volatile PM in their study using a variety of late model light duty vehicles.

The size distributions shown in Fig. 4 also give indications of composition. For the Lower PM Source Vehicle the mass in the accumulation mode (area under the curve), where most BC is typically found, is nearly the same for the FTP and US06 tests, consistent with the MSS data. The nucleation mode, usually composed mainly of semi-volatile material, is very small for the FTP tests and much larger for the US06 tests, again consistent with the relationship between MSS and filter mass data. For the Higher PM Source Vehicle mass in the accumulation mode is nearly twice as large for the FTP compared to the US06 test, again consistent with MSS data. Much more material is found in the nucleation mode range for US06 than for FTP tests, consistent with the higher semi-volatile to BC ratio, but the mode is not as distinct as that of the Lower PM Source Vehicle. The comparisons of PM mass determined by real-time instruments, gravimetric filter, and emission of soot determined by MSS were described in details in other papers (Xue et al., 2016, 2017).

3.4. Comparisons between sampling parameters

This section provides a detailed discussion of the impacts of the changes in FFV and the cumulative filter approach in improving

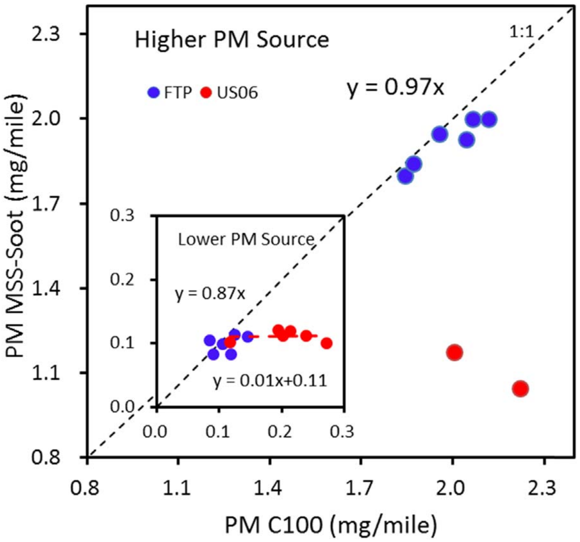

Fig. 3. MSS and PM mass correlations for the Higher and Lower PM Source Vehicles.

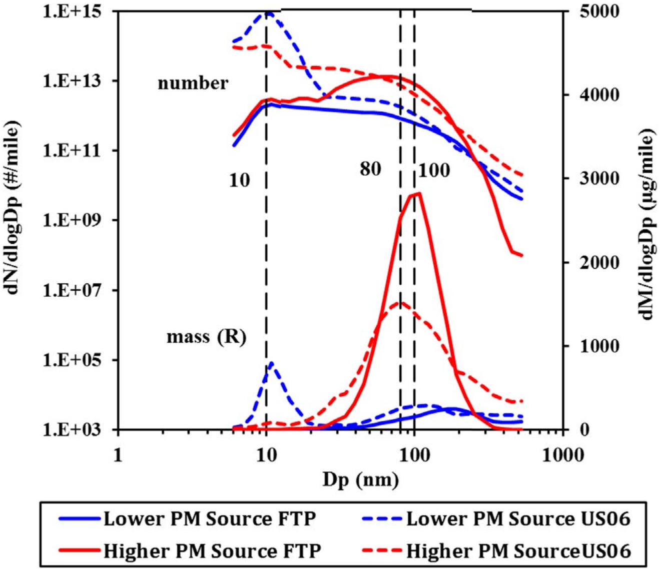

Fig. 4. Averaged number (solid lines log scale) and mass (dashed lines linear scale) PSD for the Higher and Lower PM Source Vehicles.

the effectiveness of gravimetric PM mass measurements. Different dilution ratios were also utilized between the CVS and PFD tests, but comparisons in the impact of DF on improving the effectiveness of PM emission measurements are inappropriate because the sampling systems are different. The DF results are presented in Section S5 in SI. We have also recently conducted a more extensive comparison of PM sampling between a CVS and different PFD systems, and these results will be presented in a separate paper.

In this section, we first investigate if the alternative values altered the measured PM emission rate. Then, we evaluate whether the alternative parameters reduced the measurement variability. The results for paired t-tests for the means differences and f-tests for variability differences are shown in Table 2, with f-test results shown in parentheses. Note that a paired t-test was used to evaluate the differences between filter samplers within each test, and therefore is independent of the test-to-test variability that is represented by the error bars for the average emission test values presented in Fig. 1. P-values are shown in red for values below 0.05 and in blue for values greater than 0.05 but less than 0.1, indicating that the differences between the samplers are statistically significant at a confidence level of 95% or more or 95% > 90%, respectively.

3.4.1. *Filter face velocity (FFV)*

In this subsection, differences in the PM mass emissions between CVS samplers A, B, and C were evaluated. These samplers were all operated with the same DF and flow-weighting conditions, and sampled simultaneously from the same CVS system.

Table 2
Paired two tail t-test and f-test (numbers in parentheses) for the evaluation of FFV: p-values (mg/mi).

Cycle	Vehicle	n	Part 1: FFV Comparison p-value			Part 2: Cumulative Filters p-value
			A150 vs. B125	A150 vs. C100	B125 vs. C100	C100 vs. D100i
4-phase FTP	Lower	6	0.280(0.874)	0.450(0.986)	0.068(0.019)	0.840(0.027)
3-phase FTP	Higher	6	0.614(0.582)	0.339(0.211)	0.553(0.377)	0.085(0.869)
US06US06	Lower	6	0.460(0.757)	0.331(0.590)	0.638(0.527)	–
	Higher	3[a]	0.473(0.242)	0.615(0.389)	0.187(0.398)	–

[a] One test from the preliminary test was included for the calculation.

3.4.1.1. Mean differences. Table 2 part 1 shows the paired two tailed t-test results for the selected FFVs evaluated from the CVS PM samplers. Each column shows the p-values for a statistical comparison between two different samplers operating at different FFVs. Out of the 12 combinations evaluated, none of the means were statistically different at a 95% confidence level, and only one of the means was statistically different at the $90 < x < 95\%$ confident level. These results in general suggest that increasing the FFV (from 100 to 150, flow-weighted, and at a DF of 7) does not alter the mean measured emission rates for either the Higher or Lower PM Source Vehicles.

3.4.1.2. Variability. Variability was evaluated using an f-test to compare the variance among the different FFV cases utilized for samplers A, B, and C (Table 2 part 1). The p-values for all the tested combinations were greater than 0.05 except for the B125/C100 combination. The low p-value suggests the FFV of 125 compared to the FFV of 100 cm/s for the 4-phase Lower PM Source Vehicle showed a statistically significant difference in the variance at a 95% confidence level. The remaining f-tests showed p-values higher than 0.05, suggesting that there were not significant differences in the variance for the different FFVs evaluated.

3.4.2. Cumulative filters

The D100i and C100 samplers in the CVS system provide the most direct comparison of the effects of combining filters, where the FFV is the same and the only difference was combining or not combining filters. So these samplers are the focus of the comparisons for this section.

3.4.2.1. Mean differences. Table 2 part 1 shows the statistical results of the paired t-tests performed between the D100i sampler and sampler C100 for both vehicles over the FTP cycle. Similar comparisons were not done for the US06 cycle, as it contains only a single phase. Neither of the vehicles showed statistical differences in means for an FFV of 100 cm/s for a cumulative filter as compared to the individually utilized D100i conditions. The Higher PM Source Vehicle FTP tests showed a relatively small increase of 0.09 mg/mi for probe sampler C100 compared to D100i that was statistically significant at a 90% confidence level.

3.4.2.2. Variability. Table 2 part 2 also shows the statistical results of the f-tests performed on the D100i and C100 samplers for both vehicles over the FTP cycle. The low p-value for the 4-phase FTP tests suggests that the differences in variabilities are statistically significant at the 95% confidence level for the Lower PM Source Vehicle. The results are different from the results in Sardar et al. (2016), which showed no improvement or reduction of the measurement variability when comparing a cumulative single filter with conventional individual filter sampling for vehicles with emission rates from ~0.1 to 2 mg/mi. Sardar et al. (2016) used two different PFD systems to compare the difference in filter collection strategies, which may have added additional uncertainties. The variability for the Higher PM Source Vehicle did not show statistically significant differences possibly due to the much higher filter weights.

It is also useful to look at the absolute differences in test variability between different samplers, and compare these with theoretical estimates. The measurement variabilities, as characterized by two times the standard deviation for the flow-weighted CVS, i.e., A150, B125 and C100, and the PFD sampler, were 0.05, 0.04, 0.05 and 0.03 mg/mi, respectively, while it was 0.14 mg/mi for the individual filter by phase PM sampler (D100i). This result is consistent with a theoretical estimation based on propagation of error analyses that indicates the measurement uncertainty could be reduced by a factor of four between traditional sampling conditions (one filter per phase, an FFV of 100 and a DF of 7), in comparison to a composite 4-phase FTP test, with an FFV of 150 and a DF of 5. These theoretical estimations are discussed in greater detail in Johnson et al. (2015).

4. Discussion

For this study, different modifications have been applied to the current regulatory gravimetric PM measurement method (i.e., the use of cumulative PM filters and increasing filter face velocities (FFVs) (from 100 to 150 cm/s)) in an effort to increase the robustness of the PM measurements, while decreasing the testing variability. The evaluations were designed to the question “Does changing the sampling parameter improve the signal-to-noise ratio and decrease variability, while not altering the measured PM mass emission rate?” The results showed that PM mass emission rates were in close agreement over the range of sampling parameters evaluated. Average FTP PM emission rates for different samplers for the Higher PM Source Vehicle ranged from 1.85 to 1.98 mg/mi. Average FTP PM emission rates for different samplers for the Lower PM Source Vehicle ranged from 0.09 and 0.11 mg/mi. These differences represent less than 5% of the 3 mg/mi and 1 mg/mi standards, respectively. The differences for individual tests for various PM samplers were slightly larger, ranging from 1.73 to 2.12 mg/mi for the Higher PM Source Vehicle and from 0.04 to 0.24 mg/mi for the Lower PM Source Vehicle. Statistical analysis results suggested that neither increasing the FFV (from 100 to 150, flow-weighted, at a DF of 7) nor using a cumulative vs. individual filters significantly altered the mean emission rates for either the Higher or Lower PM Source Vehicles at a 95% confidence level.

The results showed these suggested modifications could increase the collected filter mass/signal-to-noise ratio without introducing statistically significant differences in the PM mass emission rates. For a given mass emission rate, the signal (i.e., filter mass) could be increased with either higher FFVs or cumulative filters, thus the overall signal-to-noise ratio was increased. A statistically significant reduction in measurement variability was not found when the Higher PM Source Vehicle was tested, indicating the modifications investigated in this study may have limited benefit to lower the measurement uncertainty when testing a vehicle targeting the EPA Tier 3 and 2017 California LEV III PM standard of 3 mg/mile. When the Lower PM Source Vehicle was tested, the 4-phase cumulative filter for both the CVS (with a DF of 7 and FFV of 100 cm/s) and the PFD (with a DF of 5 and FFV of 150 cm/s) showed a statistically significant reduction in the PM mass measurement variability while not impacting the PM mass measurements, when compared with the traditional individual bag filter PM collection method. Therefore, these modifications could lower the measurements uncertainty when testing a vehicle that targets California LEV III requirements in 2025 (1 mg/mi). The 4-phase FTP does; however, require increasing testing time and associated increases in testing costs, so the tradeoffs between improved PM measurement and increased testing effort would need to be more carefully evaluated.

It is important to note that the results of this study represent carefully constrained conditions designed to reduce variability in PM measurements, including the use of dedicated vehicles, the same fuel (E10), a single test site at one facility, the use of one driver, low dilution factors of 7:1, dedicated PM sampler hardware, the same environmental conditions, no intermittent contamination from testing high emitting vehicles, the same PM filter weigh room/filter handling procedures, etc. The variability of PM measurements in a commercial setting would likely be higher than the variability found in this study, where multiple facilities, different vehicle architectures, different drivers, different PM samplers, sites exposed to different fuels (gas, diesel, CNG, E85, etc.), carry over from higher emitting vehicles, higher DF's up to 20 or more (hybrids), different environments, varying blank test (or tunnel blank) PM corrections, etc. would be encountered. Therefore, variability of PM measurements in a commercial environment needs to be further investigated and considered.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.jaerosci.2017.12.006.

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