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ABSTRACT

Diesel engines face tightening particulate matter emissions regulations due to the environmental and health effects attributed to these emissions. There is increasing demand for measuring not only the concentration, but also the size distribution of the particulates. Laser-induced incandescence has emerged as a promising technique for measuring spatially and temporally resolved particulate volume fraction and size. Laser-induced incandescence has orders of magnitude more sensitivity than the gravimetric technique, and thus offers the promise of real-time measurements and adds the increasingly desirable size and morphology information.

The usefulness of LII as a diagnostic instrument for the precise measurement of particulate concentration and primary particle size has been demonstrated. Measurements have been performed in the exhaust of a single cylinder DI research diesel engine. Simultaneous gravimetric filter measurements were made for direct comparison with the LII technique. Quantitative LII is shown to provide a sensitive, precise, and repeatable measure of the particulate concentration over a wide dynamic range. LII and gravimetric measurements are shown to correlate well over a wide range of operating conditions. A novel method for determining the primary particle size is shown to be precise enough to distinguish particle sizes for different engine operating conditions, and subsequently the number density of primary particles was determined. LII has also been shown to be sensitive in differentiating the PM performance between four different fuels.

The LII technique is capable of real-time particulate matter measurements over any engine transient operation. The wide dynamic range and lower detection limit of LII make it a potentially preferred standard instrument for particulate matter measurements.

INTRODUCTION

From an environmental perspective, there is an urgent need to decrease the total emissions from transportation engines. The undesirable exhaust emissions include CO_2, NO_x, and particulate matter (PM). CO_2 is a recognized greenhouse gas, and as a result of the Kyoto Protocol, industrialized countries have committed to reducing emissions of CO_2. This can be primarily achieved by reductions in fuel consumption, and diesel engines offer the highest efficiency for road-going vehicles. The concession is that the emissions reduction systems for other pollutants are not as well developed for diesel engines as they are for spark-ignited engines.

Demand for improved environmental performance has led to increasingly restrictive emission regulations for diesel-powered vehicles throughout Europe, North America, and Japan. Proposed regulations indicate that this trend to lower emissions levels will continue for the foreseeable future. Although PM is regulated for environmental reasons, from an operational point of view, particulate formation is not desirable.

A significant portion of atmospheric particulates arises from combustion of fuels in various engines and furnaces. In urban areas, mobile sources are major contributors to ambient PM concentrations. The particulate emissions from diesel engines are in the form of complex aerosols consisting primarily of soot and volatile organics. For regulatory purposes, particulate matter emissions are defined as the mass of the matter that can be collected from a diluted exhaust stream on a filter kept at 52°C. This includes the organic compounds that condense at lower temperatures, but excludes the condensed water. This measurement provides the time-averaged PM emissions over the period during which the particulates are collected on the filter, making measurements of the transient behavior of PM emissions impractical. Since the collected PM and other condensed material on the filter agglomerate, it is also impractical to determine the particulate size and size distribution. As diesel engines improve, the quantity of PM generated is reduced, pushing the gravimetric technique nearer to its sensitivity and reproducibility limits. In spite of its drawbacks and limitations, the gravimetric filter technique is the EPA certified test method for diesel vehicles, engines and fuels.

The detrimental effect of PM on human health is a current concern. Recent indications are that not only the mass of particulates, but the size and number of particulates have a role in the health effects upon humans. The size of the aggregates affects the respirability of the particulates. It is well known that atmospheric particulates are implicated in human respiratory distress. The size of the primary particles forming the aggregates affects the surface area of the particulates, and thus affects the active area for transfer of toxic compounds (organic and inorganic) from the surface of the particulates, and for secondary particulate formation in the atmosphere. Medical research is continuing in an effort to determine if there are linkages between diesel produced particulates and cancer and to understand the mechanisms for them. There is limited information currently available on the size and morphology of diesel particulates; most measurements have been for concentration only. As the particulate concentrations in the exhaust of modern diesel engines are reduced, the aggregate sizes are also being decreased.

Particulate size and number emissions from diesel engines are not currently regulated. This may be traced to two compelling factors: lack of medical research evidence as to acceptable levels of PM as a function of aggregate size; and lack of instrumentation to adequately characterize the size and number of particulates. Recent developments in the medical and instrumentation fields [1] are opening the opportunity to provide the data for future regulations based upon particulate size.

Much of the current research in diesel combustion is driven by the need to meet near- and longer-term reductions in the regulated levels of PM and NO_X. The mechanisms for formation and elimination of PM are poorly understood in comparison to NO_X. In order to develop processes and techniques for limiting the emission of PM, we must first possess suitable means for reliably measuring various particulate-related parameters. Laser-induced incandescence [2-12] has emerged as a useful diagnostic for making spatially and temporally resolved quantitative measurements of diesel PM concentration [13 and references therein]. LII has recently been successfully compared with gravimetric sampling [1], and has been shown to have the potential for primary particle sizing [9, 14].

Particulate matter emissions have been simultaneously measured by LII and the standard gravimetric procedure in a mini dilution tunnel connected to the exhaust of a single-cylinder DI research diesel engine. The engine used in this study incorporates features of contemporary medium- to heavy-duty diesel engines and is tuned to meet the U.S. EPA 1994 emission standards. The engine experiments have been run using the AVL 8-mode steady-state simulation of the U.S. EPA heavy-duty transient test procedure. Results of the PM concentrations measured using the two methods are compared, the primary particle sizes are determined on a mode-by-mode basis, and the use of LII for comparing the PM emissions from four different fuels is demonstrated.

THEORY

In LII, a short duration laser pulse is used to heat the particulates. With sufficiently high laser energies, the particulates reach peak temperatures in excess of the carbon evaporation temperature (> 4000 K). The resultant incandescence, while of short duration, can be readily detected and processed to yield concentration and size information. LII typically has a temporal resolution of 10 ns and can be used to perform both quantitative point measurements and 2-D planar visualization. LII has orders of magnitude more sensitivity than the gravimetric technique, and thus offers the promise of real-time measurements and the potential to add the increasingly desirable size and morphology information.

The approach to the numerical modeling of the transient heating and subsequent radiation and cooling of particulates exposed to short duration laser pulses has been previously described [1, 12]. The approach is similar to that used by several authors [3, 7, 8, 11]. The model, as most recent efforts are, is based upon more realistic soot morphology, assuming the soot particles to be aggregates of just touching primary particles, as opposed to the previous models' assumption that the particles were spheres of equivalent volume to the aggregate. This allows use of the applicable Rayleigh theory for absorption instead of the clearly inappropriate Mie theory, provided that the primary particle is within the Rayleigh limit (significantly smaller than the wavelength of the light source).

The methodology to determine the PM concentration and the primary particle size from the LII experiments is described below.

PARTICULATE CONCENTRATION – Typically, quantitative LII measurements of particulate concentration are made by calibrating the system in a medium of known concentration, and then scaling the exhaust results by that calibration factor. A different approach was followed in the current research. It is based upon knowledge of the particulate surface temperature, determined by three-wavelength pyrometry. A single point calibration is made in a known source at a known temperature, which results in an absolute sensitivity (in $W/m^3 \cdot ster$). By recording the time-resolved exhaust data at two wavelengths (two or more are required) the temperature of the particulate can be determined at any point in time, by solving

$$\frac{I_{\lambda_1}}{I_{\lambda_2}} = \frac{\lambda_2^6}{\lambda_1^6} \frac{\left(e^{\frac{hc}{k\lambda_2 T}} - 1\right)}{\left(e^{\frac{hc}{k\lambda_1 T}} - 1\right)} \frac{E(m)_{\lambda_1}}{E(m)_{\lambda_2}} \qquad (1)$$

where T is the particle surface temperature, I_λ is the LII intensity, λ is the detection wavelength, $E(m)$ is a refractive index dependent function, h and k are Planck and Boltzmann constants, respectively, and c is the velocity of light.

$E(m)$ has been assumed to be 0.26 at all wavelengths, based upon the commonly accepted refractive index of m=1.57+0.56i [15]. The temperature is insensitive to the variation of $E(m)$ with wavelength, increasing or decreasing only 2.5% for a corresponding 10% increase or decrease in the value of $E(m)$ with wavelength.

The radiation, P_p, from a single primary particle of diameter d_p and known temperature T can be determined as

$$P_p(\lambda) = \frac{8\pi^3 c^2 h}{\lambda^6 \left(e^{\frac{hc}{k\lambda T}} - 1\right)} d_p^3 E(m) . \qquad (2)$$

The number of primary particles, N_p, is then determined from the ratio of the experimental intensity to P_p. The particulate volume fraction (PVF) can then be determined from

$$PVF = \frac{\pi d_p^3}{6} \cdot \frac{N_p}{V} , \qquad (3)$$

where V is the sample volume determined by the product of the cross-sectional area of the laser sheet viewed and the sheet thickness. The calculated particulate volume fraction is independent of the assumed primary particle size since N_p depends inversely on d_p^3. To determine d_p an additional measurement is required and then both d_p and N_p can be determined, as discussed below.

PARTICULATE SIZE – At some time after the laser excitation, the dominant cooling mechanism for the particle becomes conduction to the surrounding gas. During the conduction phase, the difference between the particle surface temperature and the ambient gas temperature decays steadily in an exponential manner. An equation of the form

$$\Delta T = A \cdot e^{-\tau \Delta t} , \qquad (4)$$

where A is a constant, is fit to the temperature data to determine τ, the time constant of the exponential decay. This method, as do all approaches dependent upon signal decay, requires *a priori* knowledge of the ambient gas temperature, which is 52°C in the dilution tunnel, and may be determined by thermocouple or other means for other measurement locations.

The particle diameter is then determined from the relation [16]

$$d_p = \frac{12 k_g \alpha}{G \lambda_{MFP} c_p \rho_p \tau} \qquad (5)$$

where k_g is the thermal conductivity of the ambient gas, α is the accommodation coefficient, G is a geometry dependent heat transfer coefficient, λ_{MFP} is the mean free path in the ambient gas, c_p is the specific heat of the particle and ρ_p is the density of the particle. The relationship between particle size and the time constant of the exponential decay is shown in Figure 1 for a range of sizes expected for primary particles in a diesel exhaust. This method provides a sensitive and reliable measure of the primary particle size.

EXPERIMENTAL

Details of the engine, conventional emissions measurement system, and LII system have been provided previously [1, 17], and are summarized below.

RESEARCH ENGINE AND EXHAUST EMISSION MEASUREMENTS – The engine used in this work is a single-cylinder research version (Ricardo Proteus) of a Volvo TD123 heavy-duty truck engine. The engine is a direct-injection type and had a displacement volume of 2 liters. The significant engine parameters are summarized in Table 1. The research engine incorporates many features of contemporary medium- to heavy-duty diesel engines. It is tuned to meet the U.S. EPA 1994 emission standards.

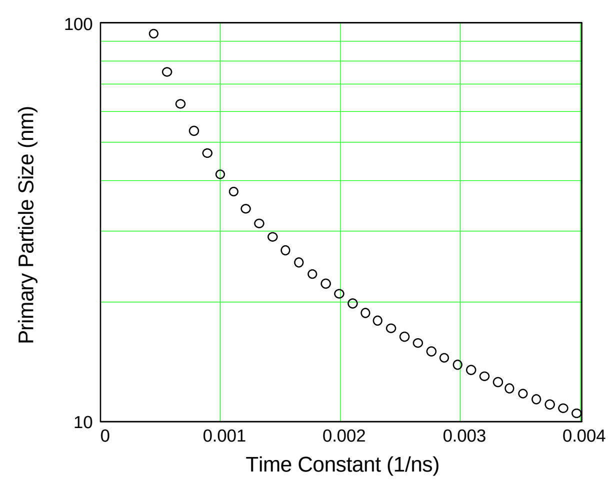

Figure 1. Primary particle size as determined from the exponential decay of the temperature difference between the particle surface and the ambient gas.

The speed and the load of the research engine were controlled independently by a dynamometer and a fuel control system. To simulate a turbocharger, externally compressed and dried air, controllable for temperature and pressure, was supplied to the engine. The exhaust system was fitted with an orifice downstream of the exhaust port, which, together with the exhaust back pressure valve, provided a cylinder pressure pumping loop similar to that of the multi-cylinder parent engine. A mixing tank in the exhaust line reduced the pressure/flow pulsations and provided complete mixing of the exhaust gases before sampling.

Table 1. Engine Specifications

Model	Ricardo Proteus (replicates one cylinder of Volvo D123)
Number of Cylinders	1
Bore	130.2 mm
Stroke	150.0 mm
Displacement	2.0 litres
Combustion Chamber Type	Toroidal Bowl, 2 Valves/ Cylinder
Compression Ratio	17:1
Injection Type	Direct Injection
Fuel Injection Pump	Bosch PE6P 120A 320RS8011
Fuel Injection Nozzle	Bosch DLLA 152 P 285
Injection Pressure	120 MPa (typical)
Maximum Power	45 kW @ 1900 rpm
Maximum Torque	260 N·m @ 1640 rpm

A heated probe was mounted after the exhaust surge tank to sample the gaseous emissions. The emissions instrumentation (Rosemount, model NGA 2000) consisted of a chemiluminescent oxides of nitrogen (NO_X) analyzer, a flame ionization total hydrocarbon (HC) analyzer, a non-dispersive infrared carbon monoxide (CO) analyzer, a paramagnetic oxygen (O_2) analyzer, and a non-dispersive infrared carbon dioxide (CO_2) analyzer for measuring the CO_2 concentration in the dilution tunnel. A non-dispersive infrared CO_2 analyzer (Horiba, model MEXA-211GE) was used to measure the CO_2 concentration in the engine exhaust.

A separate probe was used to sample the particulate emissions. The temperature of the particulate probe was maintained at 190°C to prevent condensation of the heavy hydrocarbons. The exhaust sample was diluted in a mini-dilution tunnel using filtered and dried air. The flow rate of the dilution air was regulated to maintain a temperature of 52°C. A particulate sampling line was installed in the dilution tunnel and connected to a 47 mm particulate filter and a volume meter. A computer running a data acquisition software recorded engine control parameters and emission values. A total of 60 data points were recorded in duration of 5 minutes. Averaged values of speed, power, fuel consumption rate, temperatures, pressures and exhaust emission concentrations were used in the calculation of composite emissions.

To establish a link of the results from this work to those obtained with the EPA transient test procedure, the AVL 8-mode steady-state simulation test procedure was adopted [18]. The engine operating conditions and the weighting factors of this test procedure are shown in Figure 2. The engine speed in this test procedure varies widely, from low idle speed (600 rpm) to rated speed (1900 rpm). The load also varies widely from 0% to 95%. The low idle condition is weighted heavily in the test procedure. In calculating the composite brake specific emissions, the weighting factor (WF) at each mode is used to determine the brake specific emissions (BSE) as follows:

$$BSE = \frac{\sum (Emission\ Rate)_i \times WF_i}{\sum (Brake\ Power)_i \times WF_i} \qquad (6)$$

The emission rates in the equation are calculated from measured emission concentrations and fuel consumption rates.

To make the engine experimental results relevant to multi-cylinder production engines, an effort was made to run the research engine at operating conditions as closely as possible to the "parent" production engine. The research engine manufacturer supplied the speed and load mapping of the multi-cylinder "parent" engine.

Using this information, engine speed, torque, intake manifold temperature, intake manifold pressure, intake airflow, engine brake torque and exhaust backpressure were determined. Further details of the engine and the conventional particulate measurements are given in Refs. 19-21.

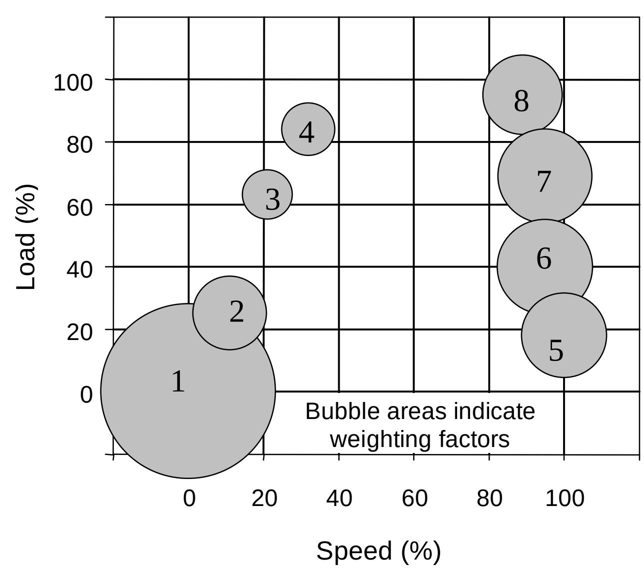

Figure 2. Engine operating conditions for AVL 8-Mode steady-state simulation of EPA transient test procedure. The area of each circle represents the relative influence of that mode's weighting factor.

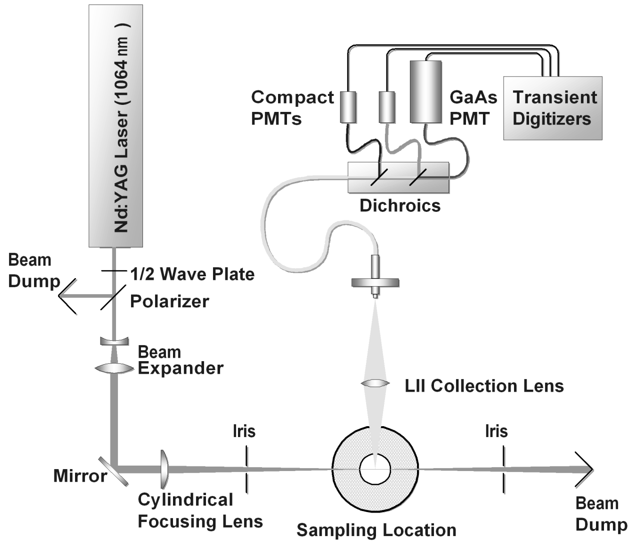

Figure 3. Optical layout of three-channel LII excitation and collection system.

LII MEASUREMENT SYSTEM – The LII system that was described previously [1, 12], Figure 3, has been modified somewhat for these diesel exhaust particulate matter measurements. Briefly, a pulsed Nd:YAG laser, operating with 60 mJ/pulse at 20 Hz and 1064 nm, was used as the excitation source. A half-wave plate (to rotate the plane of polarization) in combination with a thin film polarizer (angle-tuned to transmit horizontally polarized radiation) was used to limit the laser energy to 15 mJ/pulse.

The beam was then focused with a cylindrical lens to form a sheet through the probe volume. The beam intensity profiles in the probe volume were measured with a Coherent BeamView system. These profiles were used to ensure that the laser fluence was just beyond the saturation threshold for raising most of the soot particles to the evaporation temperature.

The LII signal from the center of the laser sheet was imaged at 0.5:1 magnification with an achromatic 31.5 mm diameter lens of 80 mm focal length onto an optical fibre of 600 mm diameter, which transmitted the incandescence signal to the photomultipliers. The imaging system was arranged such that the imaging axis was at an angle of 35° to the plane of the laser sheet. Thus the sample volume in the flame was a slanted cylinder of diameter 1.2 mm whose mean length was 2.1 mm.

The LII signal was recorded by three photomultipliers, equipped with narrowband interference filters centered at 403 nm (36 nm FWHM), 552 nm (18 nm FWHM), and 781 nm (19 nm FWHM), respectively. Transient signals from the photomultipliers were recorded and subsequently transferred to a computer for further analysis. Multipulse averages were acquired, with 400 samples per average, depending upon the particulate load. 5 averages were collected during each trial, and three trials were performed for each of the eight engine modes.

A sampling cell for producing and acquiring the LII signal was inserted between the dilution tunnel and the filters used for gravimetric sampling. This cell (Figure 4) provided a window for introducing the laser beam and for signal collection, a second window for passing the laser beam to a beam dump, and a third window orthogonal to the laser beam for viewing and alignment. The laser sheet was centered 2 mm from the open end of the tube carrying the exhaust from the dilution tunnel. The LII data was recorded simultaneously with the gravimetric sampling, to provide a direct relationship between the two measurements of PM.

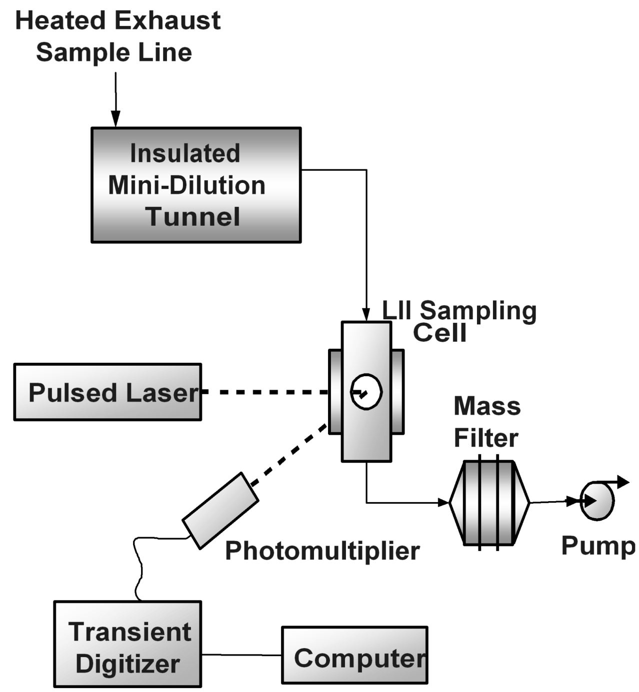

Figure 4. General layout of the LII exhaust particulate matter measurement system.

RESULTS AND DISCUSSION

The LII theory and analysis method described above was applied to measure the particulate concentration and primary particle size. Unless otherwise noted, all measurements were performed with the reference fuel, Diesel No. 2-D.

PARTICULATE CONCENTRATION – The LII signal was recorded as a function of time, capturing not only the peak signal but also the decay of the signal as the particles cool to the ambient temperature. An example is shown in Figure 5. The three curves represent the data simultaneously acquired at the three wavelengths. The signal has been corrected for any background offset. The timebase for the three wavelengths has been adjusted to compensate for variation in the lengths of cable and fiber, and the time constants of the photomultipliers have been found to be similar, so that the relative time between the signals is correct, although the Q-switch of the laser may not be exactly at an indicated time of 0 ns. The calibration allows conversion of the photomultiplier signals into absolute spectral intensity, with the results shown in Figure 6. The data has been time-averaged to reduce the number of data points at long times, where the signal is changing slowly.

As would be expected, the longest wavelength (781 nm) channel is the first to rise following the beginning of the laser pulse, and is the slowest to decay. It is followed by the middle (552 nm) and shortest wavelength (403 nm) channels. This is due to the shift towards blue wavelengths as black-body emitters increase in temperature. Further investigation of Figure 6 shows that after 600 ns, the signals have dropped 3 to 4 orders of magnitude from the peak values, and the noise level is becoming significant.

The optical pyrometry technique was applied to determine the particulate surface temperature throughout the rapid heating due to laser irradiation and subsequent cooling. Peak temperatures of 4400 – 4500 K were typically observed, as shown in Figure 7. As indicated by the 95% confidence limits, the LII technique provides a precise measure of the particulate surface temperature from shortly before the peak to when the signal has dropped more than two orders of magnitude. As the confidence limits indicate, the maximum error is typically ± 4%. The concentration of particulates for the data shown in Figure 6 and Figure 7, determined from the maximum absolute intensity and peak particulate surface temperature, is 3.37 ppb. The results of the conversion from absolute intensity to particulate concentration are shown in Figure 8. The linearity of the calibration can be observed, as the data is nearly within a single standard deviation of the best-fit line over almost two orders of magnitude.

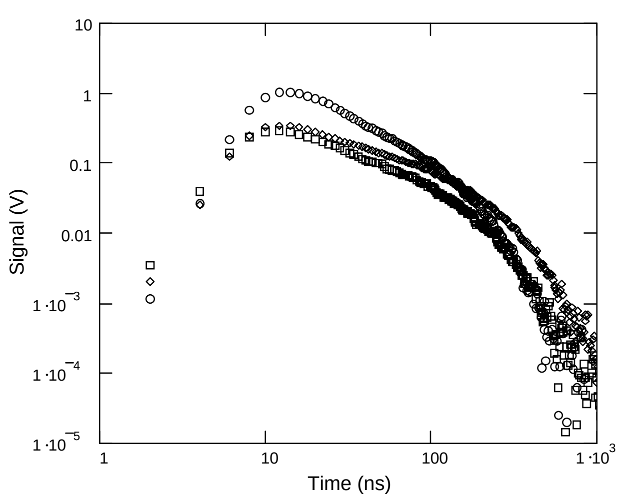

Figure 5. LII photomultiplier voltage for the three wavelengths. Data was recorded during Mode 3 operation. (circles – 403 nm; squares – 552 nm; diamonds – 781 nm)

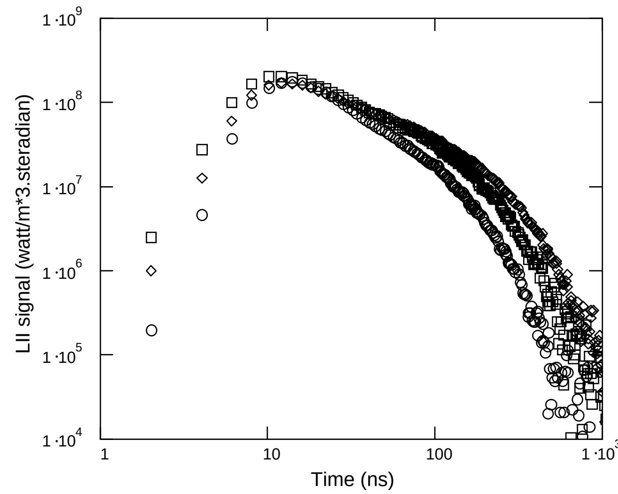

Figure 6. LII absolute signals for the three wavelengths. Data was recorded during Mode 3 operation. (circles – 403 nm; squares – 552 nm; diamonds – 781 nm)

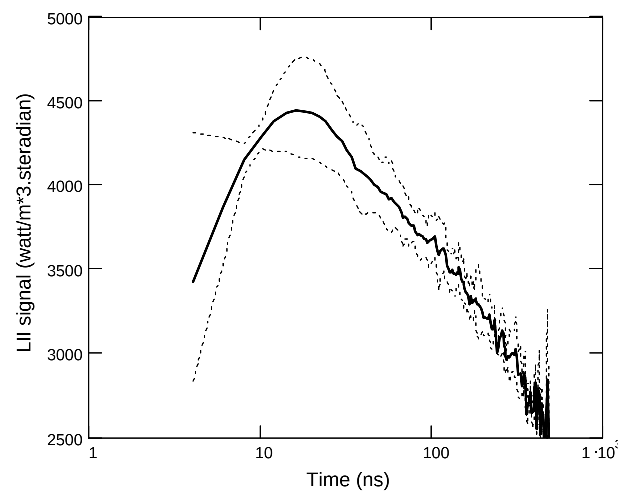

Figure 7. Particulate surface temperature as determined by LII. The dotted lines represent 95% confidence limits for the temperatures. Data was recorded during Mode 3 operation.

The particulate concentration data presented in Figure 8 is shown by engine operating mode in Figure 9. The particulate volume fractions reported are for the dilute exhaust, as measured in the sampling cell. As expected, the idle condition (Mode 1) produces the lowest levels of particulates, the low speed, high load conditions (Modes 3 and 4) produce the highest levels of particulates, and the high speed modes (5 – 8) produce moderate levels of particulates. All trials for a given mode produce results that agree within a single standard deviation, and that the greatest variability occurs in Mode 4. Mode 4 also produces the greatest amount of particulates and is the most unstable of the engine modes. The high variability was also observed in the gravimetric results, and is attributed to instability in the engine emissions output.

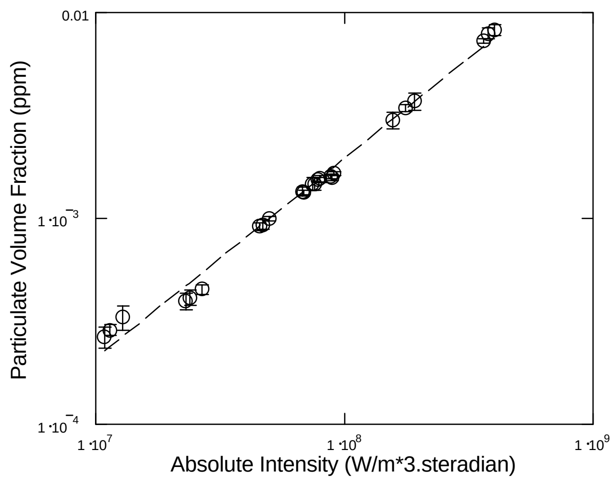

Figure 8. Particulate volume fraction as determined from absolute intensity of LII signal in diesel exhaust for three trials each of eight engine modes. Error bars indicate standard deviation for each of the trials.

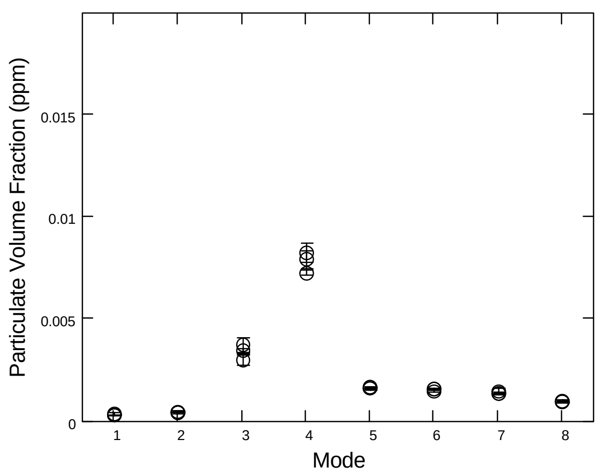

Figure 9. Particulate volume fraction by engine operating mode. Error bars indicate standard deviation for each of the three trials per mode.

Comparison of the particulate concentration data obtained from LII via the absolute calibration method is in reasonable agreement with gravimetric data acquired simultaneously, as shown in Figure 10. The largest discrepancies are at the two extremes. In general, the gravimetric method tends to produce higher results for the modes with the lowest particulate concentrations. These are also the modes with the highest fraction of soluble organic fraction (SOF), and thus much of this discrepancy may be due to components other than dry soot.

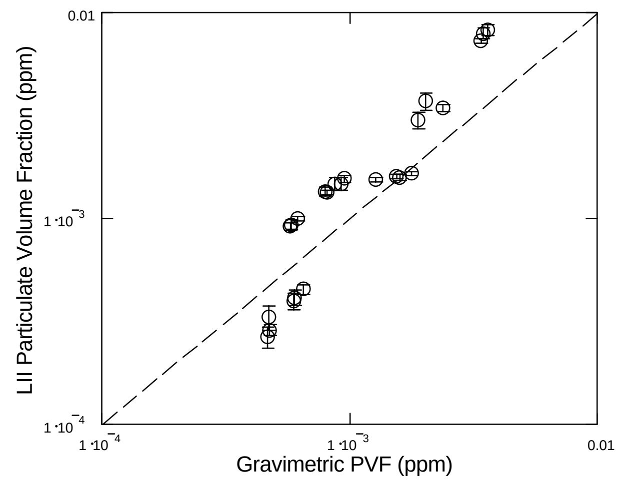

Figure 10. Particulate concentration as determined by the LII and gravimetric methods.

It should be noted that in general, detailed analysis is not performed on particulates measurements on a trial-by-trial, or even a mode-by-mode, basis. Due to the uncertainties in the acquisition of gravimetric data from the dilution tunnel, often it is only the composite emissions from the full cycle that are reported. For comparison, the engine-out (undiluted) brake specific emissions (BSE) of particulate matter (PM) are 0.112 g/hp·hr for the LII results and 0.074 g/hp·hr for the gravimetric results.

There are many possible reasons for this discrepancy, which primarily derive from the fact that the two techniques are measuring significantly different aspects of the particulate emissions. Given the uncertainties in the gravimetric and LII measurements this agreement is very satisfactory. Gravimetric sampling includes an organic fraction that does not contribute to the signal measured by LII. The density of the particulates is required to convert the mass determined by the gravimetric filter method to a volume fraction for comparison with LII. A density for dry soot is used, which does not account for the organic fraction, producing another source of error. The repeatability and accuracy of our gravimetric data is compromised by the use of an older dilution tunnel and related instrumentation. We are in the process of upgrading to a modern commercial mini-dilution tunnel, which is anticipated to provide more reliable results. The calculated LII particle intensity is sensitive to errors in the soot surface temperature. The total black body radiation scales as T^4, so that the maximum 4% error in temperature results in a 16% error in intensity. Finally, the particulate volume fraction is inversely proportional to $E(m)$, and thus a 30% increase in $E(m)$ would result in a 30% decrease in the measured PVF. Recent results have indicated that the value of $E(m)$ is higher than previously thought [22]. The LII uncertainty and the known large errors in gravimetric sampling cover the apparent differences between the two techniques.

PRIMARY PARTICLE SIZE – As described above, the primary particle size may be determined from the decay of the LII signals. For each of the engine operating modes, three 1000-pulse averages were acquired. The temperature during the steady phase of the exponential decay was analyzed to determine the primary particle size. A typical example, recorded during Mode 5 operation, is shown in Figure 11. The agreement between the best-fit exponential decay and the data is exceptional over a time period of several hundred nanoseconds, until the signals have become so weak that the measured temperatures are starting to become unreliable. For this example the diameter determined by the fit to the experimental data was 42.9 nm.

The results for primary particle size acquired for all modes are shown in Figure 12. From this data it appears that the primary particle size can be reliably determined. Further experiments are required to establish the precision of the technique. The mean particle sizes ranged from 36 nm to 87 nm, with a trend for the larger particles to appear in the low speed, low load modes (Modes 1 and 2) and the smaller particles to appear in the high speed, low load modes (Modes 5 and 6). At high load, the engine speed appeared to have little effect, as the sizes recorded in Modes 3, 4, 7 and 8 were all similar. These sizes are consistent with primary particle size determination from photomicrographs, which indicate diameters from 10 to 80 nm [23]. The primary particle size determined by LII is linearly proportional to the accommodation coefficient [24], which is 0.26 for the reported results.

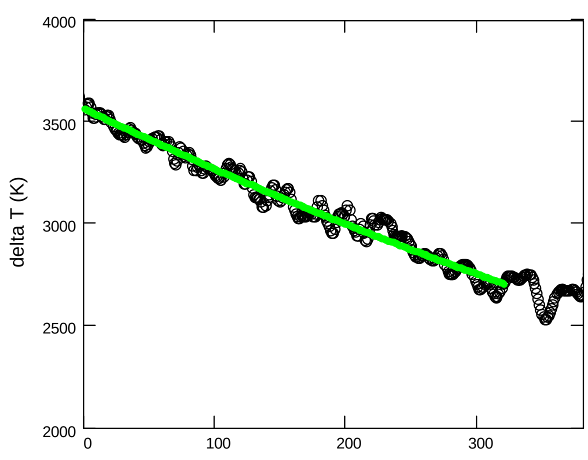

Figure 11. Differential between particle surface temperature and ambient temperature during conductive cooling period. Time scale has an arbitrary origin during the steady exponential decay. Circles represent experimentally derived data, line is best-fit to data.

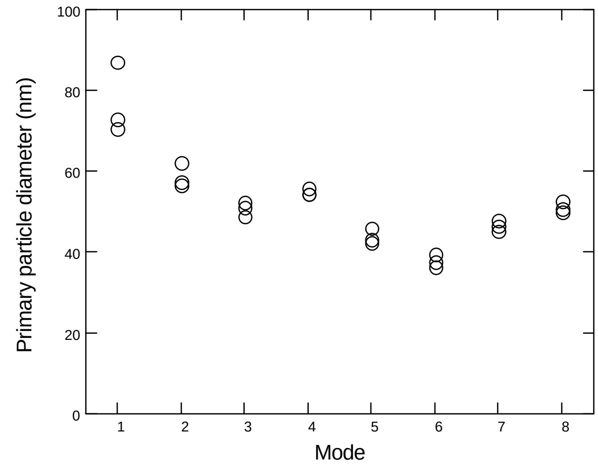

Figure 12. Primary particle diameter measured for each of the eight engine operating modes.

Combining the primary particle size data (Figure 12) with the measured particulate concentrations in the dilution tunnel (Figure 9), the number density of primary particulates in the dilute exhaust is shown by engine mode in Figure 13. There are two orders of magnitude variation in the number density of primary particles, from Mode 1 (lowest) to Mode 4 (highest). The number of aggregates would be significantly lower, as there are typically tens of primary particles per aggregate.

It must be emphasized that the reported primary particle diameters represent an assumed monosized average over the ensemble of particles in the sample volume and over the number of single-shot measurements recorded. Ultimately, it is the aggregate size that is of the greatest interest from the health, environment, and regulation perspectives. Further work is planned to determine the aggregate sizes of particulates in diesel exhaust.

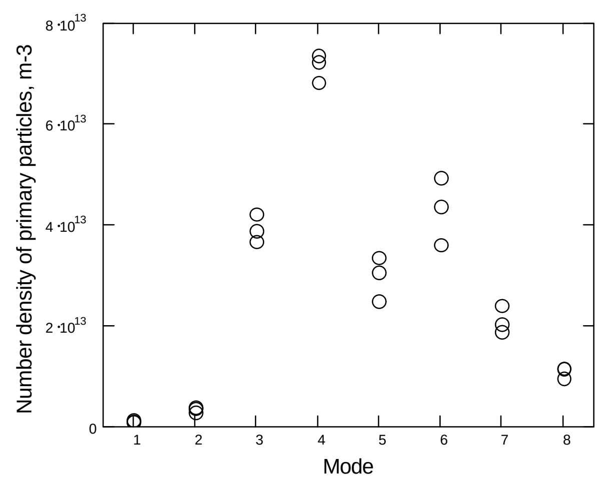

Figure 13. Number density of primary particles measured in the diluted exhaust for each of the eight engine operating modes.

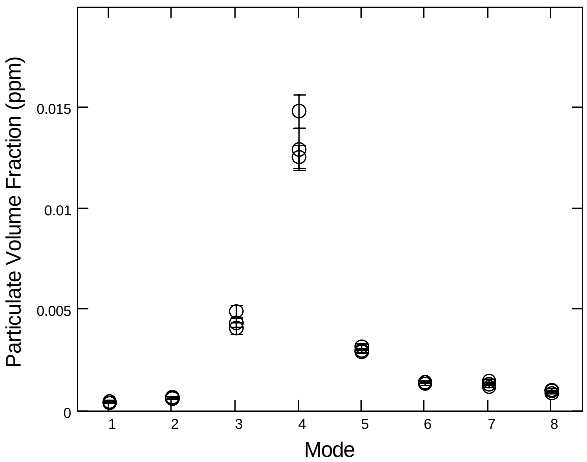

Figure 14. Particulate volume fraction by mode for Fuel C. Error bars indicate standard deviation for each of the three trials per mode.

EFFECT OF FUEL TYPE – In addition to the reference fuel (Diesel No. 2-D), three different fuels were used to assist in evaluating the sensitivity of the LII technique. The significant properties of all four fuels are summarized in Table 2. The fuels are grouped in two pairs, with the reference fuel and Fuel C having moderately high levels of sulfur, whereas Fuels D and E have relatively low levels of sulfur.

The concentrations of particulates as measured in the sampling chamber after the dilution tunnel are shown in Figure 14, Figure 15, and Figure 16 for Fuels C, D, and E, respectively. From these figures it can be observed that, as with the reference fuel, all trials for a given mode produce results that agree within a single standard deviation, and that the greatest variability occurs in Mode 4.

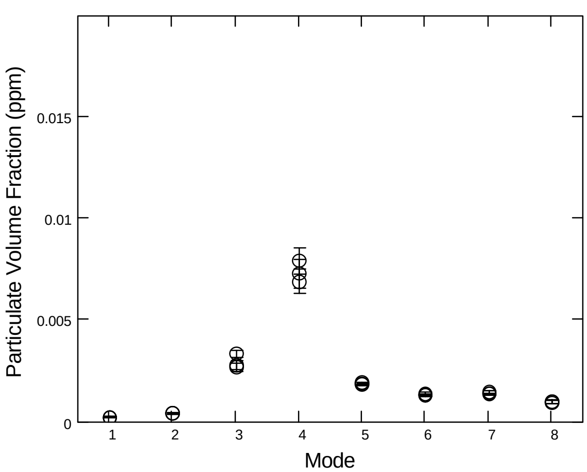

Figure 15. Particulate volume fraction by mode for Fuel D. Error bars indicate standard deviation for each of the three trials per mode.

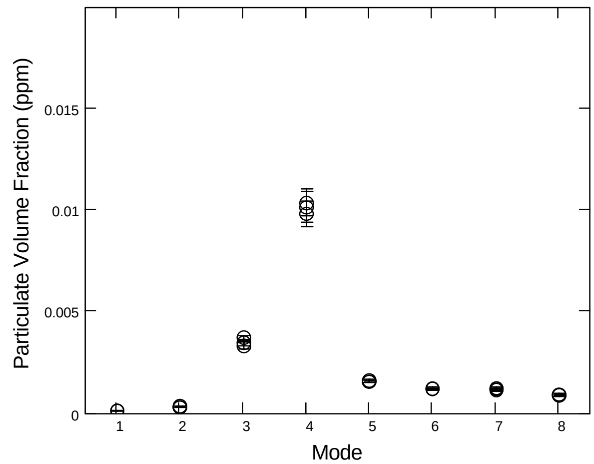

Figure 16. Particulate volume fraction by mode for Fuel E. Error bars indicate standard deviation for each of the three trials per mode.

The engine-out (pre-dilution) concentrations of particulates, as measured by the LII and gravimetric techniques, are shown for the three additional fuels in Figure 17, Figure 18, and Figure 19. The symbols each represent the average of the 5 multipulse LII data acquisitions recorded during each engine trial and the corresponding gravimetric data. There were three trials for each engine mode, and each mode is represented by a different symbol. As shown with the reference fuel, the gravimetric method tends to produce higher results for the modes with the lowest particulate concentrations. These are also the modes with the highest fraction of soluble organic fraction (SOF), and thus much of this discrepancy may be due to components other than dry soot.

Table 2. Fuel Specifications

Fuel	Ref	Fuel C	Fuel D	Fuel E
Density (kg/m^3, 15°C)	838.7	846.3	845.3	826.2
Viscosity (cSt, 40°C)	2.08	2.86	2.95	1.61
T10 (°C)	198	215	218	179
T50 (°C)	255	267	274	215
T90 (°C)	310	340	322	275
Cetane No.	40.7	43.7	45.6	41.5
Hydrogen content (%)	13.20	13.26	13.44	13.76
Sulfur content (ppm)	192	184	46	32

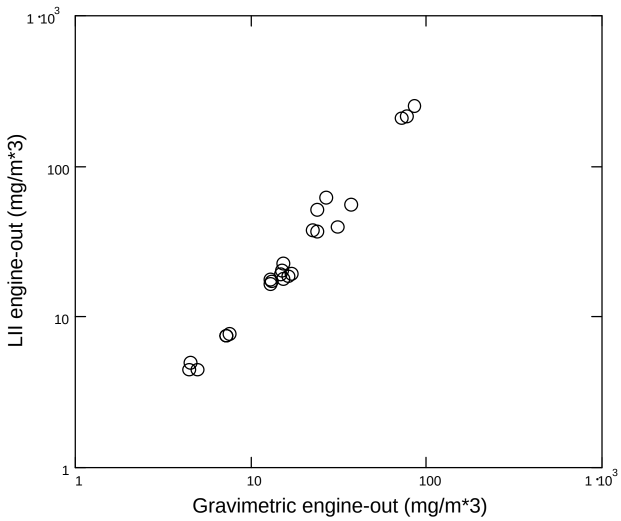

Figure 17. Particulate concentration for Fuel C.

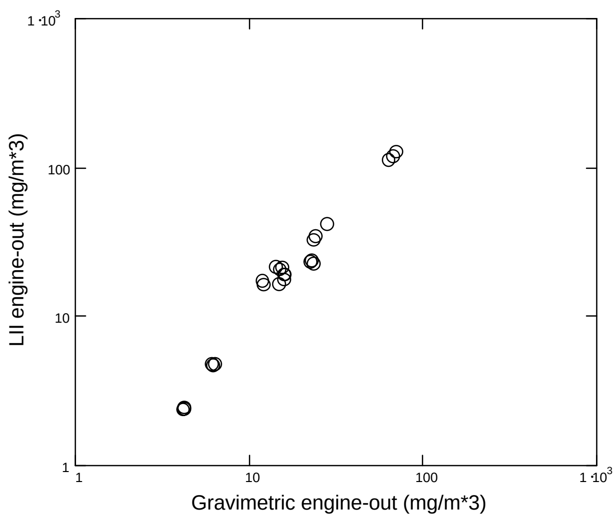

Figure 18. Particulate concentration for Fuel D.

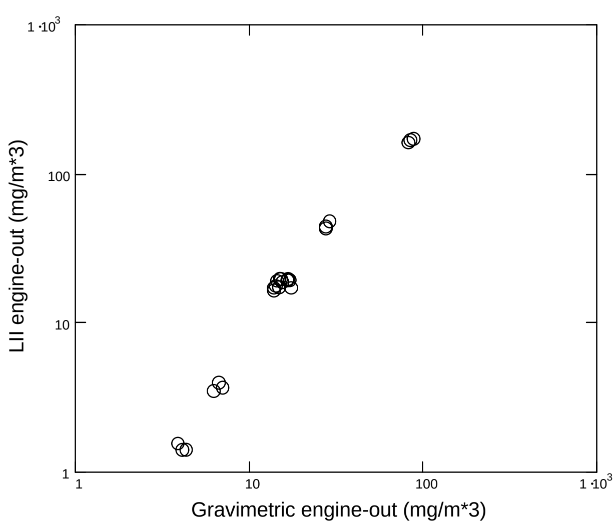

Figure 19. Particulate concentration for Fuel E.

The results are summarized in Table 3. For each of the pairs of fuels, the LII results show the same trend as the gravimetric results, with the particulate BSE increasing from the reference fuel to Fuel C, and also increasing from Fuel D to Fuel E. However, for the higher sulfur fuels the LII technique measures a significantly higher quantity for the particulate BSE as compared to the gravimetric technique. The reasons for this discrepancy are unknown. In contrast, the results for the lower sulfur fuels show closer agreement.

One possible explanation may be that the assumed density for the particulates is too high in the case of the higher sulfur fuels, as the same density was used in all calculations.

Table 3. Brake Specific Emissions by Fuel Type

Fuel	**Gravimetric PM BSE (g/hp·hr)**	**LII PM BSE (g/hp·hr)**
Reference	0.074	0.112
Fuel C	0.087	0.140
Fuel D	0.082	0.106
Fuel E	0.084	0.112

It must be emphasized that LII and gravimetric sampling do not measure the same quantities. LII measures the dry soot volume, whereas gravimetric sampling measures the mass of dry soot and the adsorbed SOF as a bulk quantity. LII provides time-dependent data if required, and has been shown to be capable of determining the primary particle size. It is well known that the number and size of particles, as well as the chemical composition of the adsorbed SOF, are the primary health risks. Thus LII performs better than gravimetric sampling at measuring the quantities of interest from a health risk perspective. It is also the number and size of particulates that affect urban air quality and visibility.

All of the reported measurements were made downstream of the dilution tunnel, specifically to demonstrate the capabilities and advantages of LII in comparison to the gravimetric technique. The particulate concentrations measured ranged from below 100 parts-per-trillion (ppt) to over 10 parts-per-billion (ppb), demonstrating a fraction of the technique's dynamic range, and also demonstrating its sensitivity in low concentration environments. Measurements can also be performed in the exhaust manifold, where the signal levels will be 5 to 10 times higher, due to the fact that the exhaust stream will not be diluted. Furthermore, measurements can be performed in-cylinder in an optically accessed engine, provided that excitation and signal beam extinction can be accounted for and compensated for.

CONCLUSIONS

The usefulness of LII as a diagnostic instrument for the precise measurement of particulate concentration and primary particle size has been demonstrated. Measurements have been performed in the exhaust of a single cylinder DI research diesel engine. Simultaneous gravimetric filter measurements were made for direct comparison with the LII technique. Results have shown that:

1. The use of three wavelength detection to determine particle surface temperature, combined with absolute sensitivity calibration, provides a sensitive, precise, and repeatable measure of the particulate concentration over a wide dynamic range
2. The LII technique produces good correlation with the gravimetric filter method measurements on a mode-by-mode basis over a wide range of operating conditions.
3. The primary particle size can be determined from the LII signals, and that this method is precise enough to distinguish particle sizes for different operating conditions.
4. Once the particulate concentration and primary particle size are known, it is possible determine the number density of primary particles.

LII has also been shown to be sensitive in differentiating the PM performance between four different fuels, predicting the same trends in brake specific PM emissions as the gravimetric filter method.

The LII technique is capable of real-time particulate matter measurements over any engine transient operation, making it a valuable tool in tuning diesel engine PM emissions performance. The wide dynamic range and lower detection limit of LII make it a potentially preferred standard instrument for PM measurements. Further development of the LII technique has the potential to give information about extensive aspects of the morphology of the particulate matter. Use of LII also provides a significant time advantage over the gravimetric procedure, both in the collection and processing of data.

ACKNOWLEDGMENTS

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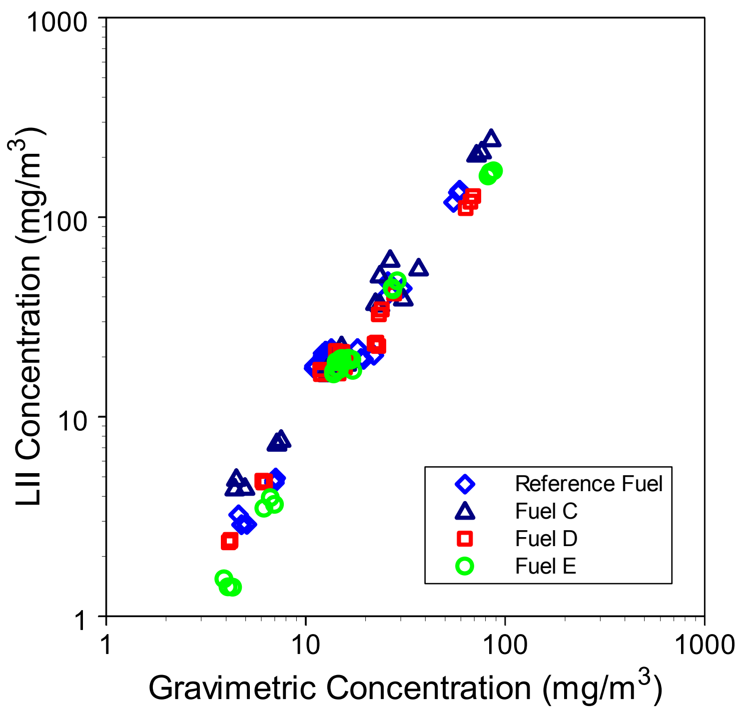
With further refinement of the algorithms and improved knowledge of the probe volume characteristics, we have revisited the data that was published in SAE Paper No. 2000-01-1994, "*In-Situ Real-Time Characterization of Particulate Emissions from a Diesel Engine Exhaust by Laser-Induced Incandescence*," by David R. Snelling, Gregory J. Smallwood, Robert A. Sawchuk, W. Stuart Neill, Daniel Gareau, Daniel J. Clavel, Wallace L. Chippior, Fengshan Liu, Ömer L. Gülder and William D. Bachalo.

The engine out (undilute) emissions are shown to the right for the four fuels reported in this paper, replacing the data shown in Figs.17-19 of the paper (the data was not presented in this format for the reference fuel in the paper). As can be seen in the new figure, the correlation between LII and the standard gravimetric technique is excellent.



The brake specific particulate emissions (PM BSE) for the EPA transient test procedure are summarized in the table below. This revised data replaces that shown in Table 3 of the paper.

As before, the LII results show same trends as the gravimetric results, with the PM BSE increasing from the reference fuel to Fuel C for the higher sulphur fuels, and from Fuel D to Fuel E for the lower sulphur fuels. The agreement between the two methods is now much closer.

However, some differences do remain. The fact that LII measures lower quantities for PM BSE than gravimetric technique for fuels D and E emphasizes that LII and the gravimetric techniques are measuring different properties of the particulate matter.

Brake Specific Emissions by Fuel Type

Fuel	Sulphur (ppm)	Gravimetric PM BSE (g/hp·hr)	LII PM BSE (g/hp·hr)
Reference	192	0.074	0.074
Fuel C	184	0.087	0.092
Fuel D	46	0.082	0.070
Fuel E	32	0.084	0.074