Quantitative Laser-Induced Incandescence Measurements of Particulate Matter from the Exhaust of a Direct Injection Spark Ignition Automobile

Sub-task 4.4A

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ABSTRACT
Diesel and gasoline 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 soot 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 information on the increasingly desirable size and morphology information. Quantitative LII is shown to provide a sensitive, precise, and repeatable measure of the soot concentration over a wide measurement range. The current research determined the tailpipe particulate emissions characteristics from a DISI (direct injection spark ignition) vehicle, including identifying the relative contributions of various engine modes to the total particulate emissions. The volume concentration measurements were obtained in the undilute exhaust with laser-induced incandescence (LII). Particulate measurements were also performed with ELPI instrumentation, sampling from a mini-diluter. Gravimetric filter sampling was performed to measure mass emission rate, organic/elemental carbon, and sulphates/nitrates/trace elements.

INTRODUCTION
From an environmental perspective, there is an urgent need to decrease the total emissions from transportation engines. The undesirable exhaust emissions include CO, CO_2, NO_x, HC, 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 direct injection spark ignition (DISI) engines offer substantially improved efficiency for gasoline-fueled vehicles. The concession is that the emissions reduction systems for other pollutants are not as well developed for these lean burn stratified combustion engines as they are for conventional port fuel injected homogeneous charge engines. Although PM is currently regulated only for diesel vehicles, future regulations will limit PM for all light duty vehicles. Especially deleterious are the small sized respirable aerosols and particulate matter that are known to produce adverse health effects and are suspected of producing high altitude clouds which adversely affect the earth's climatology [1, 2].

To develop processes and techniques for limiting the emission of soot, we must first possess suitable means for reliably measuring various soot-related parameters. These methods must have adequate measurement range in order to be able to monitor and characterize the pollutant emissions over a very wide range of concentrations and must operate under a range of environmental conditions from *in situ* exhaust to atmospheric monitoring. In the case of particulate matter, information on the particle mass, size, and volume fraction is needed. Laser-induced incandescence (LII ge) has emerged as a technique for measuring soot concentration and size, as non-soot matter is evaporated and/or does not contribute to the signal.

LII Background
With the LII method, the soot within the laser beam path is heated rapidly using a pulsed laser source with duration typically less than 20 ns (FWHM). The soot is conventionally heated from the local ambient soot temperature to the soot vaporization temperature (approximately 4000 to 4500 K). The incandescence from the soot particles is measured using collection optics and photodetectors. Using appropriate calibration and analysis of the incandescence signal, information on the soot volume fraction and primary soot particle size may be obtained. Laser energy absorption by the soot particles and the subsequent cooling processes involve complex analysis of the nano-

scale heat and mass transfer in time and space [3, 4]. The method is essentially nonintrusive and is capable of making *in situ* measurements over a very large range of soot concentrations in both flames and under ambient conditions.

Recently, a novel technique for performing absolute light intensity measurements in LII has been presented, thus avoiding the need for a calibration in a source of soot particulates with a known concentration [5, 6], and thus extending the capabilities of LII for making practical quantitative measurements of soot. Using this *in situ* absolute intensity self-calibration technique, LII has been applied to measure soot particle volume fraction in laminar diffusion flames [6], carbon black [6, 7] and in diesel engine exhaust streams [6, 8, 9].

EXPERIMENTAL

Full details on the experiments have been reported elsewhere [10].

Vehicle

A state-of-the-art production direct injection spark ignition engined vehicle from the Japanese market was used as the evaluation platform for this research. This vehicle uses a lean-burn stratified charge combustion concept for most of its operating modes. The vehicle was unmodified, and all emissions were measured from the tailpipe. The fuel for this work was indolene reference fuel (4 ppm sulfur).

Exhaust Emission Measurements

The research was performed on a chassis dynamometer normally used for regulatory compliance testing, equipped for EPA FTP emissions measurements. A constant volume dilution tunnel was employed for regulated gaseous emissions and filter paper particulate collection. The gaseous emissions were bag collected and sampled for CO, CO_2, NO_x, and THC. Gravimetric filter sampling was performed to measure $PM_{2.5}$ mass emission rate and particle phase organic carbon/elemental carbon content, and to collect samples for sulphate / nitrate / trace elements content. Particulate size distributions were measured after sampling with a fixed dilution ratio Dekati mini-diluter. The samples were drawn from the heated transfer hose at a point within 1 m of the tailpipe exit, through a heated sample line (100ºC), to the diluter. The stream was then split, and sent to a Dekati ELPI for the size distribution measurements. The ELPI provides data at 1 s intervals, but for a limited number (12 stages) of size bins. As LII only measures the elemental carbon component of the particulates, dilution is not necessary. Other condensed materials, such as the organic fraction of the particulates and water, do not contribute to the signal measured by LII, and are typically evaporated by the high energy laser well before the significant portion of the LII signal is detected.

LII Measurement System

The use of the absolute intensity approach [6] provides for continuous self-calibration of the LII technique, and allows use of lower laser fluences and lower maximum soot temperatures. Thus, issues associated with evaporating a significant portion of the soot are avoided. The absolute intensity method applies two-color pyrometry principles to determine the particle temperatures, relating the measured signals to the absolute sensitivity of the system as determined with a strip filament lamp.

RESULTS

Data for two complete trials (cold start LA4, hot start LA4, and HWFET transient driving cycles) was collected on consecutive days.

Gaseous / Particulate Filter Measurements

The CO_2, NO_x, and THC remained consistent from the first trial to the second. However, the CO concentration showed a marked decrease of 27% for the EPA FTP cycle between trials. The $PM_{2.5}$ mass emission rate as measured by the gravimetric technique also showed a marked decrease between trials. For the cold start, hot start, and HWFET tests, the OC had a marked decrease between trials, while the EC had little variation. The only ions found in measurable quantities were nitrate and sulphate ions, which were both near the minimum quantitation limit for all trials.

LII Particulate Measurements

The LII measurement of soot mass emission rates for two cold start LA-4 trials on consecutive days is shown in Figure 1. Concentrations below 5 ppt (0.01 mg/ m^3) were measured frequently with sufficient precision during these experiments. Sensitivity for LII is only limited by the probe volume dimensions and the available laser energy. The greatest mass emission rates occur during the first 250 s of this cycle, during which the engine and emissions systems are warming up. The large gap in the data for Trial 1 between 140 and 280 s was due to the issues with the data acquisition software. Throughout the cycle, the increased mass emission rate episodes for this vehicle appear to correspond to the acceleration phases. After the warm-up period, deceleration and cruising produce minimal levels of soot, and even under acceleration, the levels do not reach 1 mg/m^3. Although there is significant variation in the soot emissions on a trial-to-trial basis, the results clearly track each other and the transients in the driving cycle. Similar results were found for the hot start LA-4 driving cycle, with the exception that the high soot levels during the first 250 s were not present, as the vehicle was already warmed up. This is shown in Figure 2, with an expanded vertical axis as compared to Figure 1. At no point do the soot levels reach 1 mg/m^3.

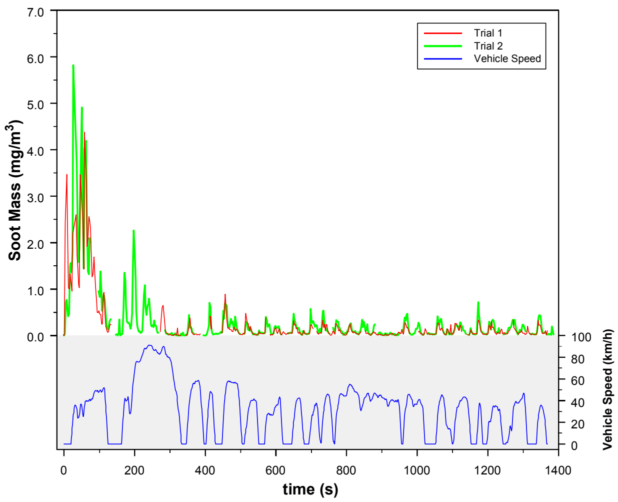

Figure 1: Comparison of cold start LA-4 transient cycle soot mass emissions as determined by LII for two separate trials (top) and vehicle speed during LA-4 transient cycle (bottom).

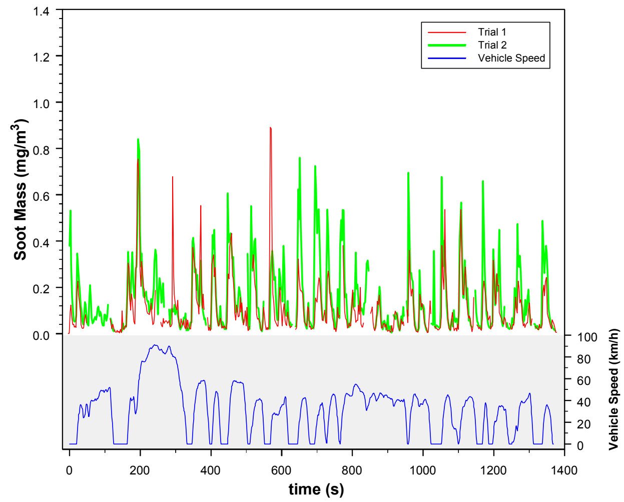

Figure 2: Comparison of hot start LA-4 transient cycle soot mass emissions as determined by LII for two separate trials (top) and vehicle speed during LA-4 transient cycle (bottom).

For the highway fuel economy test, Figure 3, moderate soot levels of an average 0.2 mg/m3 were recorded by LII, with occasional peaks due to acceleration. Again, at no point do the soot levels reach 1 mg/m3. During the second trial, 100 pulse averages were recorded after 125 s, providing reduced temporal resolution of 5 s. This can be observed in Figure 3 , where the second trial data displays far less structure than the first trial data at times after 125 s. If more temporal resolution is desired than the 2 s shown for the first trial, the number of pulses averaged could be reduced at the expense of increased noise in the signal.

In general, the second trials produced higher levels of soot, which could be due to a number of factors, such as changes in the environmental conditions or variations in the driver performance from one day to the next. The mean concentration measured by LII was 31% less for the hot start Trial 1 than for Trial 2. The differences were substantially less for the cold start trials (14% less) and slightly less for the HWFET trials (25% less). These differences are opposite to the $PM_{2.5}$ results, which decreased from Trial 1 to Trial 2. This is likely due to the different nature of the particles being measured. Note that this is a time average of the LII data, not a volume or mass average, so direct comparison to the filter measurements is not advised.

ELPI Measurements

Due to cooling of the stream sampled by the ELPI instrument during the dilution process, the measured particulates are composed of condensable species, such as organics, water, and sulphuric acid, as well as the soot and trace elements present in an undilute stream. The ELPI instrument has a 1 s data acquisition rate, yet appears to have less time response than the LII system with 2 s intervals. This is likely due to the fact that the time taken to sweep the volume of the impactor is greater than 1 second.

The volume concentration of particulates at each measurement time is shown in Figure 4. The larger diameter particles, although fewer in number, represent proportionately more volume, and thus dominate the total volume measurement. The ELPI results showed similar behavior in two consecutive trials. However, in contrast to the data obtained with LII, the volume concentration data from ELPI for the second day appeared less than the data for the first day. The difference from the LII data may be due to the fact that the ELPI instrument is measuring a different property than the LII system. The implication is that there were significantly more condensed organics in the first trial to more than offset the slight increase in elemental carbon matter observed in the second trial. The total volume measured by ELPI was 27% greater for the hot start Trial 1 than for Trial 2. The differences were substantially less for the cold start trials (2 % less) and the HWFET trials (11 % greater).

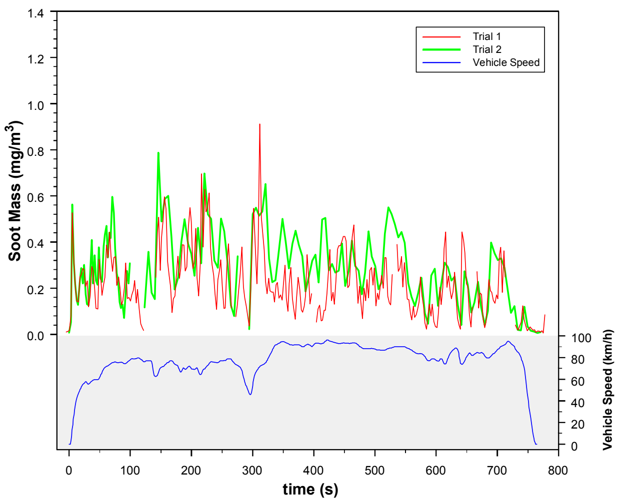

Figure 3: Comparison of HWFET transient cycle soot mass emissions as determined by LII for two separate trials (top) and vehicle speed during HWFET transient cycle (bottom). Note that after 125 s, data for Trial 2 is based on 5 s averages.

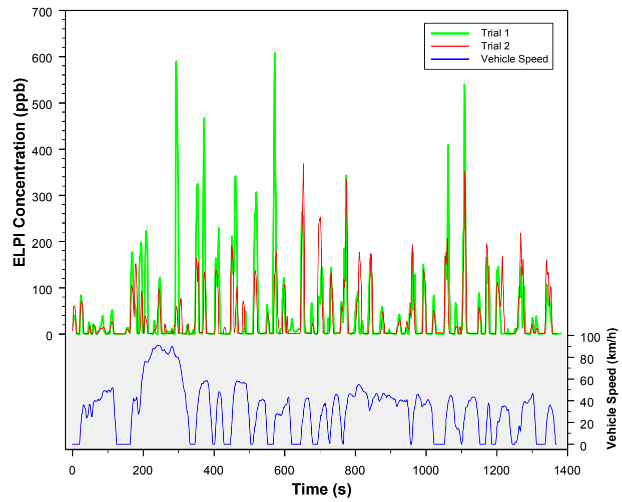

Figure 4: Comparison of hot start LA-4 transient cycle particulate volume concentration emissions as determined by ELPI for two separate hot start LA-4 transient cycle trials. Data is for all twelve stages combined.

LII / ELPI Comparisons

Direct comparison of the soot concentration as determined by LII and the total volume of particles as determined by ELPI is presented in Figure 5. In a qualitative sense, these two properties appear to correlate well, as both follow the driving cycle transients with similar relative magnitudes. A detailed view of a portion of the data from Figure 5 is shown in Figure 6. The ELPI has a sensitivity threshold below which it reports zero signal, whereas LII is demonstrating its superior sensitivity, showing response at very low concentration levels. The greater temporal response of LII is also evident, as there is much more structure in the temporal profile of the LII concentration data. Although the two signals do show mostly similar features on a gross scale, there is a significant difference in response to the event at 730 s, with the ELPI showing far less response in comparison to some of the other peaks, whereas the LII indicates a much more significant response. The peaks in particulate concentration are clearly aligned with the acceleration phases of the transient driving cycle, with minimal levels of particulate produced during steady-state and deceleration. Although the peaks for the two methods are aligned, LII shows considerably more detail in regions where ELPI has dropped below its sensitivity threshold.

DISCUSSION

The results presented above indicate that the greatest level of soot is emitted during the acceleration phases of the driving cycles, with substantially lower levels emitted during steady speed driving and deceleration. Further investigation of the LII data supports these findings. To investigate the effect of acceleration, it is interesting to relate the soot mass to the acceleration data acquired during the LA-4 cycles, which are dominated by stop-start driving. The data for the cold start LA-4 cycle is presented in two parts: that which was significantly different from the hot start data, which was the first 250 s, and that which was similar to the hot start data, indicating a fully warmed engine and emissions system, for the period after 250 s. This data is presented in Figure 7, where the data for the first 250 s is generally much higher than the data after warm-up has occurred, and also shows little correlation with acceleration. However, Figure 7 also shows that for the cold start LA-4 cycle, after 250 s, there is a distinct positive correlation between positive acceleration and soot mass emission rate, with little or no correlation for deceleration. This observation is even more apparent for the hot start LA-4 cycle, which was not split into two time periods. Figure 8 shows a similar, slightly more definitive, result for the relationship between acceleration and soot mass emission rate for the hot start LA-4 cycle.

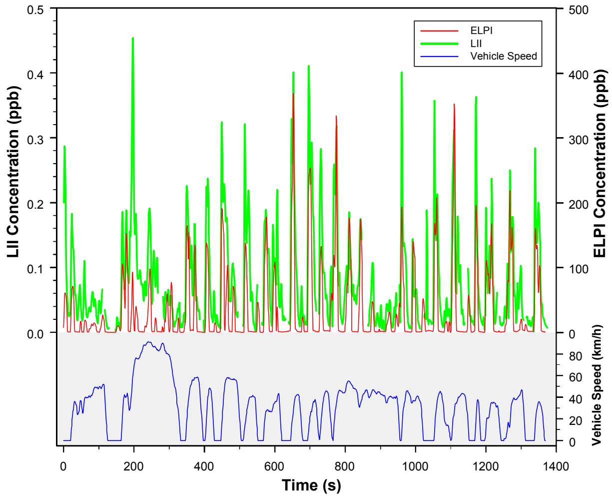

Figure 5: Comparison of levels of soot determined by LII and particulate determined by ELPI for hot start LA-4 transient cycle.

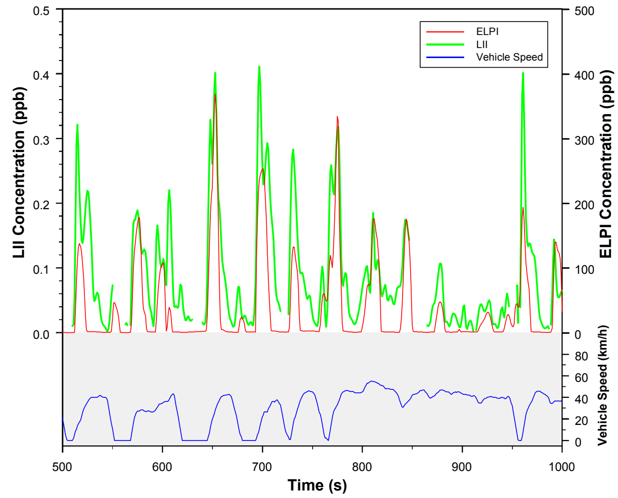

Figure 6: Detail from Figure 5 illustrating relative temporal response and sensitivity of LII and ELPI to vehicle speed, acceleration, and deceleration.

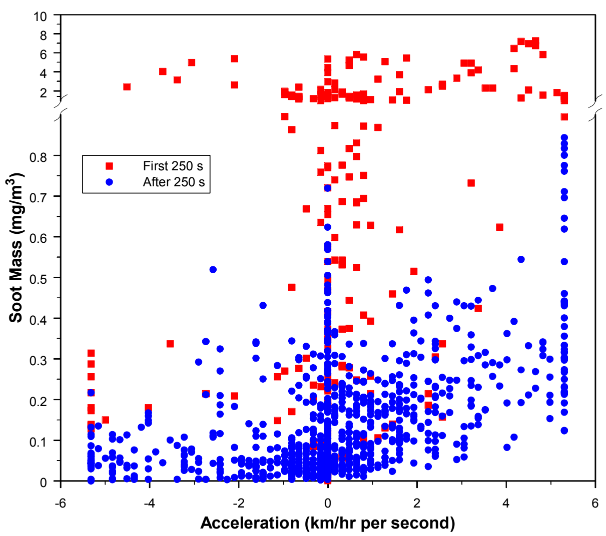

Figure 7: Soot mass concentration determined by LII versus acceleration during cold start LA-4 transient cycle.

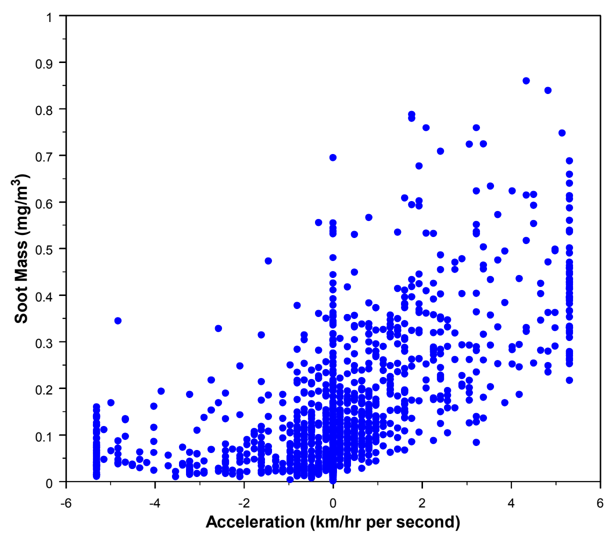

Figure 8: Soot mass concentration determined by LII versus acceleration during hot start LA-4 transient cycle.

SUMMARY AND CONCLUSIONS

The tailpipe particulate emissions from a state-of-the-art production direct injection spark ignition vehicle were measured with a variety of techniques, including laser-induced incandescence. Whereas most of the techniques measure both the soot and condensed aerosols, which contribute significantly to the lack of repeatability, LII measures only the concentration of soot. The large differences in the gravimetric measurements are attributable to the poor repeatability of this method and to variation in the levels of condensed particulates. The elemental carbon measurements have been demonstrated to be the most consistent measure of particulates. From the LII results, there was a measurable increase in the soot content from Trial 1 to Trial 2. By comparing the ELPI and LII results, it is apparent that there was a significant decrease in the concentration of the organic carbon content from Trial 1 to Trial 2. This is consistent with the results obtained from the OC/EC analysis.

The soot measurements reported a new minimum threshold for LII of 5 ppt (0.01 mg/m^3), demonstrating the sensitivity of this technique. As expected, the highest levels of soot emissions for this DISI vehicle were recorded during the cold start phase. After the cold start period, average soot emissions were 0.2 mg/m^3, with peak levels all below 1 mg/m^3.

Both LII and ELPI demonstrated that maximum emission rates occur during acceleration transients. Steady–state driving and deceleration produce much lower levels of particulates. From the LII results, there is a clear correlation between acceleration and soot emission rates. LII demonstrated superior sensitivity to soot at the low levels observed with this vehicle, and also provided greater temporal response to variations in the soot levels.

The LII technique is capable of real-time soot measurements over all vehicle transient operations, making it a valuable tool in tuning gasoline engine soot emissions performance. The wide measurement range and low detection limit of LII make it a preferred standard instrument for soot measurements. Further development of the LII technique has the potential to give information about extensive aspects of the morphology of the particulate matter. LII also provides a significant time advantage over the gravimetric procedure, providing real-time results.

ACKNOWLEDGMENTS

Partial funding for this work has been provided by the AFTER POL and the Particulates POL of the Canadian Government's PERD Program (Advanced Transportation Task).

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