Final Report

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Title: Assessment of an Advanced Method for Measurement of the Solid Carbonaceous (Soot) Component of Mobile Source Particulate Matter

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Abstract

Laser induced incandescence represents a promising means for measuring soot particulate from diesels, gas turbine engines as well as other sources. Under the current program, the measurement sensitivity has been enhanced to also allow ambient soot particulate concentration measurements. The purpose of this program was to develop the technology into a useful portable instrument that will provide immediate time-resolved measurements of particulate emissions for a broad range of applications including diesel and gas turbine engine development, evaluation of particulate control systems, and help evaluate compliance to particulate emissions regulations. During this effort, a NIST traceable calibration method was developed and refined to improve the reliability and to better define the instrument response to soot. At the same time, optical systems were developed to produce a more compact instrument which was thoroughly tested for operational integrity under typical industrial environments. Software was developed that included algorithms to fully automate the instrument set up functions and to automatically track the soot concentrations over a range of one million to one. The earlier LII 200 version was evaluated and tested at our industrial partners sites. As a result of these tests, several improvements were defined and implemented. From these results, a next generation LII 300 instrument evolved which is much more compact, has greater sensitivity and is easier to operate. Measurement comparisons to made with gravimetric results showed excellent agreement.

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1. Introduction

Laser-induced incandescence (LII) for soot (elemental carbon) measurements is an emerging technology that promises a reliable means for spatially and temporally measuring the soot volume fraction and primary soot particle size in engine exhausts. The LII is a real-time measurement technique that can be used for in-situ measurement of particulate emission in engine exhausts and other areas of interest. The first product developed by Artium Technologies, Inc. using this technology is the LII 200. The objective of this project was to work closely with the technical experts from industry and have them evaluate the LII 200 in real test environments to identify areas that needed improvement. Field experience will also be used as a mechanism for gaining widespread recognition and acceptance of the instrument. This grant funding has allowed us to extensively evaluate the capabilities of the LII 200, and subsequently, develop and evaluate the next generation LII 300, at Cummins' facilities. The instrument was also used for road testing. The results from the field testing have been used to develop the next generation instrument, namely the LII 300, which is a compact instrument that provides turn-key operation, ease-of-use, portability, and high measurement sensitivity. This report is the final element of the ICAT grant and summarizes the technology, R&D efforts, and test results leading to the development of the LII 300 instrument for the characterization of particulate emissions, specifically, soot..

2. Innovative Technology

1.1 The Artium LII 200

With laser-induced incandescence, the soot within the laser beam path is heated rapidly using a pulsed laser source with duration typically less than 20ns. The soot is heated from the local ambient soot temperature to a temperature to just slightly less than the soot vaporization point or, more accurately, the sublimation temperature (approximately 4000 to 4500 K). The incandescence from the soot particles is measured using collection optics and photodetectors. With appropriate calibration and analysis of the incandescence signal, information on the soot volume fraction and primary soot particle size may be obtained. The method is largely non-intrusive and is capable of making in situ measurements over a very large range of soot concentrations in both flames and under ambient conditions.

With the support of prior Small Business Innovative Research (SBIR) program funding from NASA, EPA, and NIST, Artium Technologies, Inc. had been able to develop a fully-integrated, rack-mountable LII system for soot particulate characterization in various applications, Fig 1. This instrument was called the LII 200.

The Artium LII 200 consists of a self-contained rugged optics enclosure which includes the laser and all components needed for operating the instrument. The optical system, Fig 2, consists of a computer-controlled automated laser beam energy detection and adjustment system that maintains the laser light fluence at the sampling volume at optimum conditions. The system automatically maintains constant laser fluence over a wide range of environmental conditions. Incandescence signal is made at 90 degrees to the transmitted beam which helps form a well-defined measurement volume. The incandescence signal is detected by a pair of detectors that use light filters centered at wavelengths of approximately 400 nm and 780 nm. The innovative two-color pyrometry technique permits accurate measurement of soot particle temperature and leads to an auto-compensating approach for soot concentration measurements. Measurement of the soot temperature allows the use of low fluence heating of the soot. As a result, the sublimation of soot is insignificant.

A custom 2-channel 12-bit, 100 MHz PCI A/D digital signal acquisition card is used for optimized high-speed data acquisition and transfer to the computer. A

USB interface between the LII 200 optics package and the system computer is used for the control functions. Besides measuring the soot volume fraction, the

Figure 1: Photograph of the Artium LII 200 system including computer, laser controller, and dilution flow controller. This is how the instrument looked before the ICAT project started.

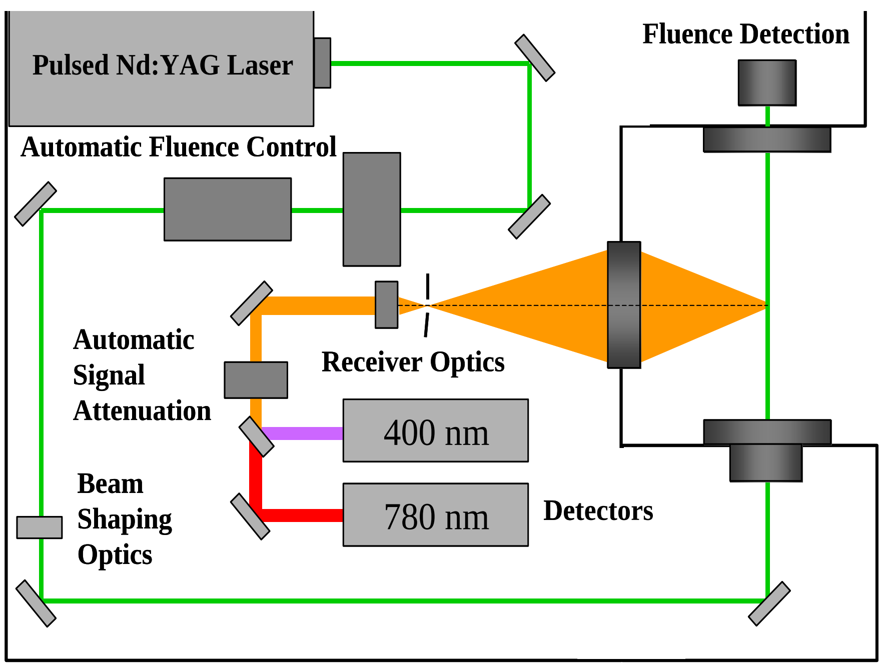


Figure 2: Optical schematic of the LII 200.

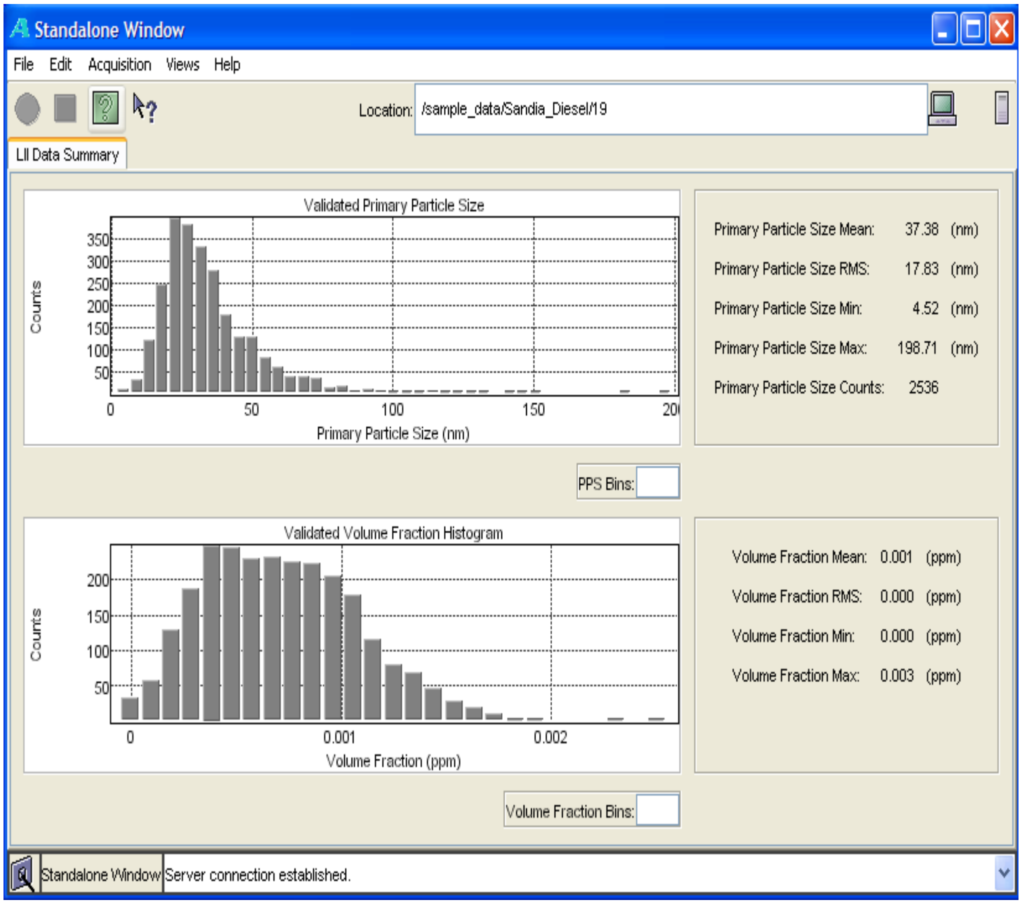


Figure 3: Typical primary particle size and soot concentration histograms displayed by the Artium LII 200 software.

LII signal decay characteristics are also processed to infer the primary particle size.

The Automated Instrument Management System (AIMS) software controls all aspects of the instrument setup and operation. It can run either on Windows (2000/XP) or LINUX (2.4 kernel) operating systems. The system software is designed around a client/server model that allows remote operation via the intranet or the Internet and is multi-user accessible. Dedicated algorithms are included for automated setup of the instrument functions and online adjustment to the prevailing measurement conditions. An external input feature allows tagging of the LII signals with external events such as engine RPM or throttle. The software has the capability of accepting, processing, displaying, and storing data for extended periods of time without interruption. During long acquisition periods, the data can be broken into multiple files for greater manageability. The data analysis and display are easily extensible and may be modified as needed. The data can be easily exported to Microsoft Excel for analysis and plotting. Figure 3 shows typical histograms plotted by the software. The LII 200 has been tested thoroughly over the past year and the fundamental capabilities demonstrated under a variety of applications including carbon black monitoring and process control, diesel engine exhaust emission and vehicular on-board road tests.

1.2 Comparison of PM Measurement Techniques

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 are needed. The lack of availability of suitable diagnostics has resulted in a degree of uncertainty in the correlation of the particulate loading with health effects. Improvements in the instrumentation are needed to help in developing the test protocols, standards and regulations that will preserve the environment and limit risks to health. The following section provides an overview of some of the currently available measurement methods for PM measurement and monitoring.

<u>Gravimetric Method (EPA Title 40 of the Code of Federal Regulations (CFR) parts 86 through 94 and part 1065)</u>

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^{O}C. This includes the organic compounds that condense at lower temperatures, but excludes 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 engine technologies improve, the quantity of PM generated will be 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 vehicles, engines and fuels. Gravimetric techniques are considered to be adequate for characterizing the total particle loading. However, the method does not provide information on the PM size distribution, requires significant sampling times at low particle concentrations, is very time consuming, and is often unreliable because filters do not discriminate particulate matter from other condensed material.

<u>Differential Mobility Particle Sizers (DMPS) and Scanning Mobility Particle Sizers (SMPS)</u>

In these devices (e.g. TSI Series 3936 SMPS), the particles are charged using an electric field and separated by size based on the electrical mobility. The particles that are separated into classes are then counted using a condensation nuclei counter (CNC) to produce a number-weighted size distribution. The charging characteristics of soot particles are not well-understood and can be a significant source of error.

<u>Electrical Low Pressure Impactor</u>

Another approach for measuring particles in the size range of 0.05 to 10 μm is the electrical low-pressure impactor (e.g. DEKATI ELPI). With this instrument, the particles are first charged and then passed through a cascade impactor to segregate them into bins or size classes. The current deposited on each stage of the impactor may be related to the particle concentrations in the various size classes. This method is useful but leaves a significant degree of uncertainty due to the sampling process.

Photo-Acoustic Sensor

The photoacoustic (PA) instruments (e.g. AVL 483 Micro Soot Sensor) detect the absorption of a laser beam by aerosol placed in its acoustic resonator. The laser beam power is modulated at the acoustic resonant frequency. Light absorbing aerosol components, e.g., elemental carbon, convert laser beam power to an acoustic pressure wave through heating, accompanied by gas expansion. An acoustic resonator amplifies this pressure wave by its Q factor. A microphone detects the acoustic signal that provides a measure of light absorption.

The PA method has some shortcomings. First, the instrument needs to be calibrated with the use of a soot source and gravimetric technique. To ensure accurate measurement, the technique requires calibration with the type of soot to be measured. This is difficult to perform so the soot source is derived from a quenched diffusion flame and this tends to be expensive. Heating of the soot by the laser in the PA device does not raise the temperature of the soot by more than a few degrees. Therefore, unlike the LII technique, the condensed material may not be evaporated and the measurements can be affected. Another disadvantage of the PA device is that the optical windows need to be cleaned regularly. Unfortunately, the window contamination is a continuous process and it is possible that the measurement accuracy can degrade continuously between cleanings.

Tapered Element Oscillating Microbalance (TEOM)

A continuous measurement of the inertial mass of aerosol deposited on a filter substrate has been implemented using tapered element oscillating microbalance technology (e.g. Rupprecht and Patashnick Co. Inc.). This technology utilizes a hollow tube with the wide end of the tapered tube fixed. The narrow end of the of the tube holds a filter cartridge, and a sample is passed through the filter and tube to a flow controller. The tube-filter unit acts as simple harmonic oscillator with its oscillating frequency being a function of filter mass loading. The system can be calibrated by placing a calibration mass on the filter and recording the frequency change due to this mass. As with many other methods, the instrument samples all particulate material, including condensed liquids. The instrument requires relatively frequent maintenance which can be problematic in industrial environments. The method also requires a “correction factor” to achieve agreement with gravimetric. This factor has been found to introduce additional systematic measurement uncertainty.

Nephelometer

Nephelometers are fairly simple and compact instruments with good sensitivity and temporal resolution (e.g TSI DustTrak 8520). They measure light scattered by aerosol introduced into their sample chamber. However, scattering per unit mass is a strong function of particle size, shape, and refractive index. If particle size distributions and refractive indices in diesel exhaust strongly depend on the particular engine and operating condition, this may not be an effective way to measure exhaust particle.

1.3 Key Advantages of the LII Technique Over the Other Technologies

The LII 200 has several advantages over the other PM measurement techniques described above.

- The LII performs direct measurement of the exhaust. It does not require any dilution such as the PA technique. As a result, the sensitivity gain is a factor of 10 or more.
- Since the LII 200 does not require dilution of the sample, the high capital cost for a dilution tunnel is eliminated.
- The LII 200 calibration is based on a NIST traceable incandescence source and does not require gravimetric calibration and an artificial soot source.
- LII measurements are based on first principles that include the physical

properties of the soot, the optical absorption, and the incandescence characteristics.

- LII measurements have very high temporal sensitivity and are not significantly affected by changes in the environmental conditions. The gas temperature, pressure, humidity, and other factors do not affect the LII soot volume fraction measurements.
- With our designed window purge system, the LII 200 may be operated for extended periods of time (weeks to months) without maintenance or cleaning.
- The LII 200 offers turn-key operation and can be easily operated by trained operators.

1.4 The Artium LII 300

Based on the ICAT project, we have developed the next generation LII instrument, namely the LII 300, Fig. 4. The LII 300 has some key attributes that differentiate it significantly from the LII 200:

- Enclosed sampling cell
- Built-in computer and display with touchscreen control
- Built-in pneumatics controller and sampling system
- Includes real-time pressure and temperature measurements to reduce data to STP
- Fail safe valve prevents sample from entering cell if purge air or power are off
- Compact, rugged, and portable instrument; the footprint is significantly reduced compared to the LII 200
- Very high sensitivity ; the lower detection limit is < 0.5 pptv (< 1 $\mu g/m^3$ when using a soot density of 2g/cc) and the upper limit is < 10 ppm. This is approx 10x better sensitivity compared to the LII 200.
- High dynamic range (>1,000,000:1)

Figure 4: Photograph of the Artium LII 300. The footprint is significantly reduced compared to the LII 200.

3. ICAT Project

The objective of the proposed project was to provide LII instruments to industrial users and technical experts from the industry, and the government and have them evaluate and critique the LII 200 and identify areas needing improvement. Field experience was also used as a mechanism for gaining widespread recognition and acceptance of the instrument. The results of the testing were used to develop an improved version of the instrument, namely the LII 300. The proposed efforts fell into three major categories:

1. Ease-of-use design improvements: Although the prototype instrument performed well, there were some requirements for simplifying the operation of the hardware and software for general use by less than expert users. This included:

- automatic self-testing to ensure that the calibration is always accurate,
- rugged optical systems and enclosures to ensure safe, maintenance free operation,
- user-friendly software to automatically setup and monitor the instrument operation.

Design improvements implemented prior to embarking upon an extensive field test plan.

2. Demonstrated performance in the facilities of major Diesel engine manufacturers: The LII 200 instrument was applied to the measurement of exhaust from stationary diesel engines to ascertain how well or how accurately the soot PM emissions could estimated. The emphasis was on establishing the minimum detection sensitivity and comparison to other techniques. This testing took place at the R&D facilities of a major engine manufacturer.

3. On-board road testing: The instrument was extensively tested to define the capabilities of the LII instrument via on-board measurements. The successful completion of these tests will has led to greater acceptance of the LII instrument as a suitable method for monitoring and enforcing the strict PM emission regulations.

TASK 1*: Developed automatic self-testing methods to ensure calibration is accurate*

This task was focused on verifying the robustness of the instrument calibration procedure and ensuring that the LII 200 did not drift over time. Every LII 200 system is calibrated in the factory using a NIST traceable calibration lamp source. During this program, we conducted tests to verify the accuracy of data provided by two different instruments that were calibrated in the factory. Experiments were conducted using two identical LII instruments (LII#1 and LII#2) and a sampling hose between the sample cells so that the same soot laden gas would be sampled by both instruments simultaneously. The set-up is shown in the photograph of Figure 5. Although there was an approximate 2m length of sampling hose between the two instruments, the data indicated that this was not a problem for the sampling process. In our original tests, we found an approximate 10% difference between the two instruments that was systematic with one instrument continually reading too high. After some further investigation and experimentation, we determined that the flow through the sample cell was intermittent. Hence, the two instruments were not sampling the same conditions at all times. Increasing the sample flow rate eliminated this difference. We also determined that the neutral density filters used in these instruments differed in their spectral characteristics; they did not meet the manufacturer's specification. The differences in their spectral characteristics were sufficient to cause measurement differences between the two instruments. As a result, we have now set up an incoming inspection process for every ND filter. Each filter will be characterized prior to installation.

Comparison of data between the two instruments, LII#1 & LII#2, which were of identical design and construction are shown in Figure 6 and the expanded results are shown in Figure 7 for the low end of the soot volume fraction. These data were obtained at 5 Hz sampling frequency for a period of 1000 samples at each point. The soot was generated by a small kerosene lamp and was allowed to fill the small room shown in the photograph. As can be seen in the plot, the majority of the data were in agreement within 2% between the two instruments.

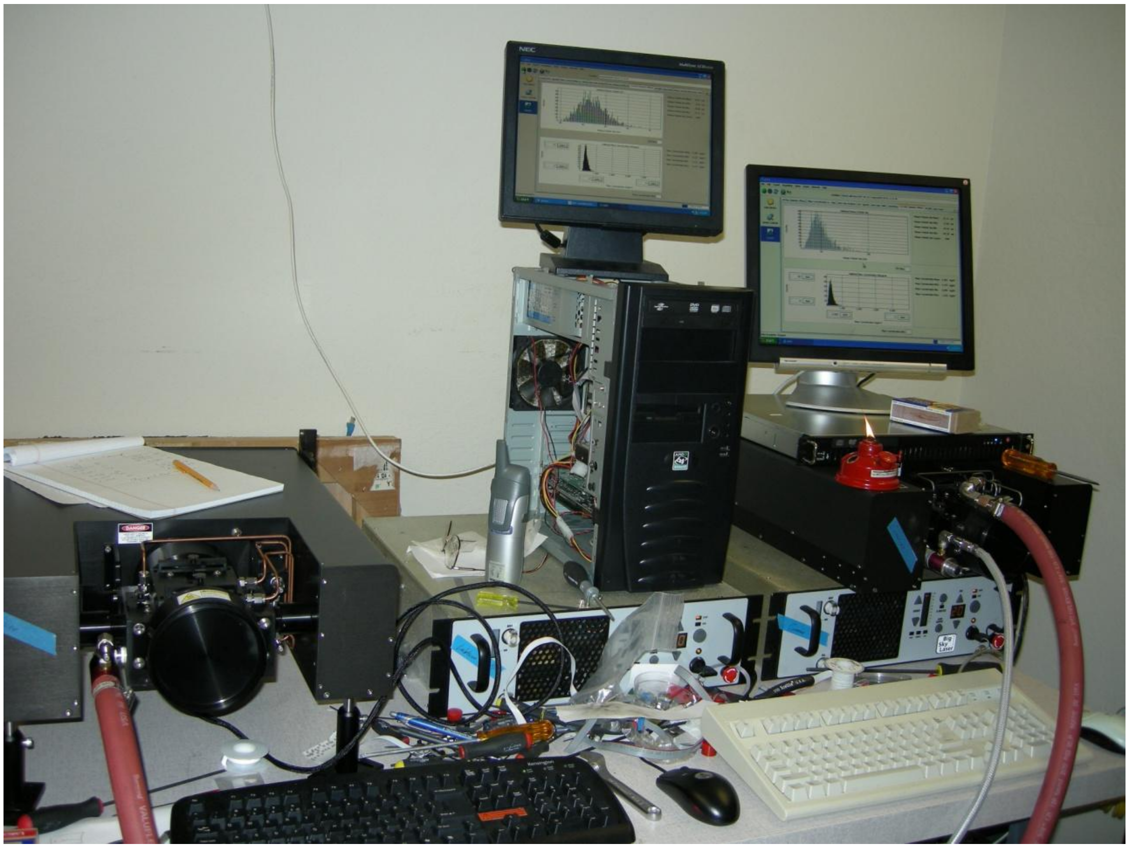


Figure 5: Photograph of the two LII systems used for the comparison of the results obtained over a wide range of such concentrations.

The laser induced incandescence instrument (LII) is known to have very high precision and resolution; these data helped to verify that observation. We have now established that our factory calibration process is sufficient for ensuring instrument-to-instrument matching. The next question that we were interested in answering was whether this factory calibration would drift over time. This question was also satisfactorily addressed by our industrial partner, Cummins, who tested the instrument over an extended period of time.

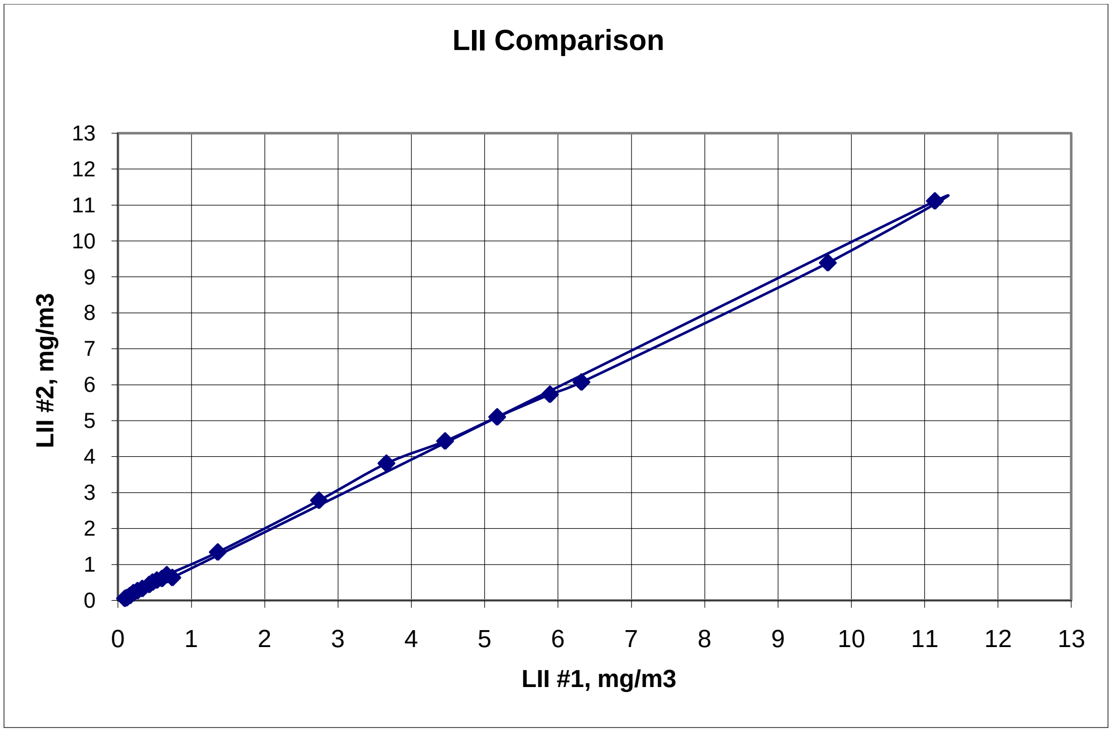


Figure 6: Comparison of the data obtained between the two identical LII instruments measuring a wide range of soot concentration.

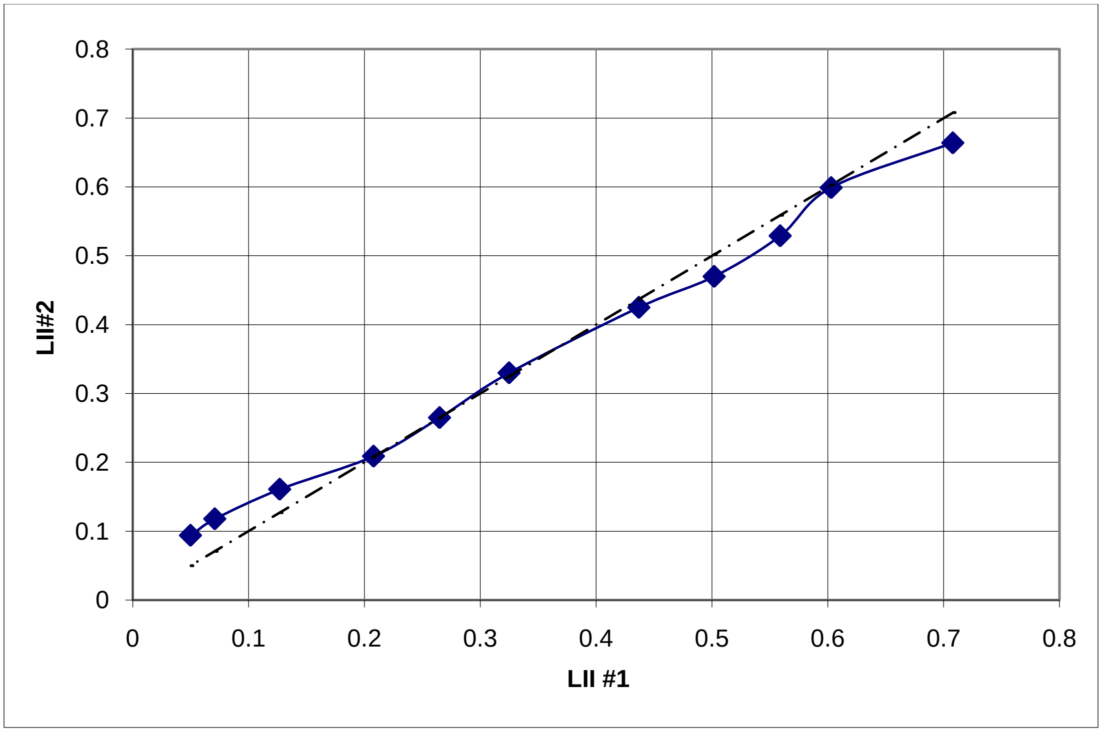


Figure 7: Comparative data expanded around the low end of the soot volume fraction end of the distribution.

TASK 2: *Ruggedize the design*

This task was focused on the development a rugged, safe, and maintenance-free instrument and which was tested to verify the appropriateness of the opto-mechanical design approach.

During this program, a new sample cell was designed fabricated, Fig. 8. Other improvements to the LII 200 system included the incorporation of light-tight enclosures around the photodetectors to prevent stray light from reaching the detectors. As the instrument will be used by operators with a wide range of skill levels, the optics had to be designed so that no operator intervention is required in the normal operation of the system. The optical components have been designed and configured such that normal vibration, shocks, and thermal cycling do not affect the operation. Control of the climate and environmental conditions within the instrument enclosure serve to eliminate thermal expansion effects and contamination.

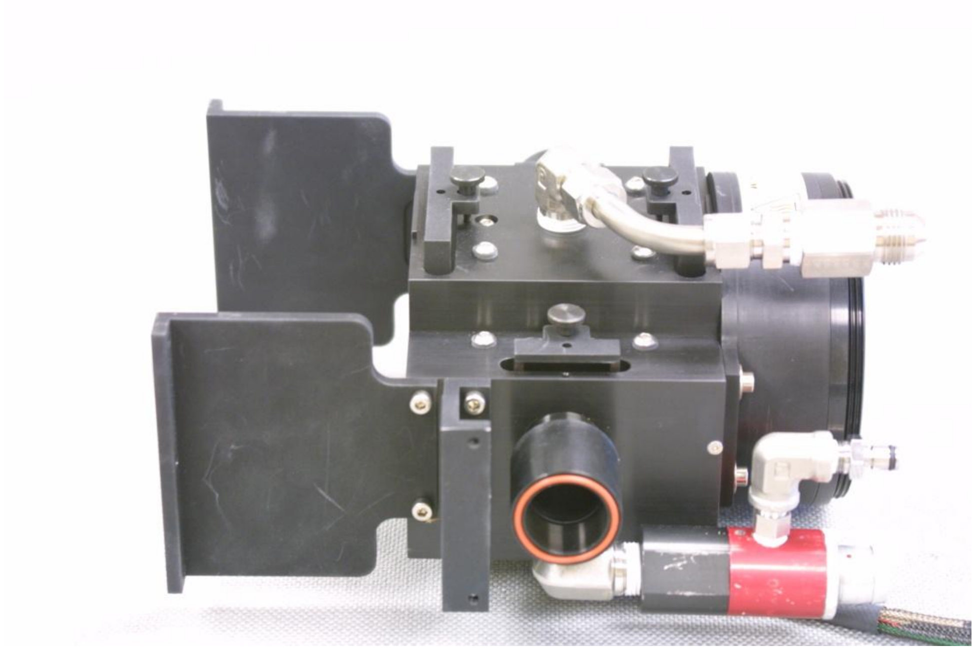


Figure 8: Photograph of the sample cell that incorporates the retro-reflecting spherical mirror for increased signal strength.

We performed a series of vibration tests to check the ruggedness of the prototype instrument package. We initially set the test condition at ~15 G (1 in/sec at 1000 Hz). We tested this condition but found that the vibration was not very severe so we turned to the Mil Spec requirement for flight-based instruments.

MIL-STD-810F Method 514.5 - Vibration

Vibration testing was performed to determine the resistance of equipment to vibrational stresses expected in its shipment and application environments. Vibration can cause:

- Optical misalignment
- Wire chafing.
- Loosening of fasteners.
- Intermittent electrical contacts.
- Touching and shorting of electrical parts.
- Seal deformation.

- Component fatigue.
- Display / Touch Panel misalignment.
- Cracking and rupturing.
- Excessive electrical noise.

Shock and Vibration tests were performed on the prototype LII 200. The minimum integrity (general) test was at a Power Spectral Density = $0.04G^2/Hz$, 20 to 1000Hz, descending 6dB/oct to 2000Hz. Three axes were tested for up to 1/2-hour per axis.

MIL-STD-810F Method 516.5 – Shock

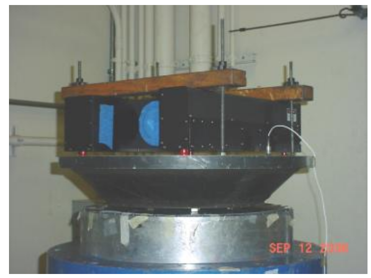
In general, shock tests are performed to assure that materiel can withstand the relatively infrequent, non-repetitive shocks or transient vibrations encountered in handling, transportation, and service environments. Shock tests are also used to measure an item's fragility, so that packaging may be designed to protect it, if necessary. Mechanical shocks will excite an equipment item to respond at both forced and natural frequencies. This response, among other things, can cause:

- Failures due to increased or decreased friction, or interference between parts.
- Changes in dielectric strength, loss of insulation resistance, variations in magnetic and electrostatic field strength.
- Permanent deformation due to overstress.
- More rapid fatiguing of materials.

Shock Tests on the LII 200

Although the MIL-STD suggests three shocks from each direction, we ran 50 for each axes (plus vertical, negative vertical, plus X, negative X, plus Y, and negative Y). The following are the plots of the shock profiles used in the test. The three setup configurations are shown in Figures 9 & 10.





X Axis **Y Axis** **Z Axis**

Figure 9: Setup used for the shock studies.

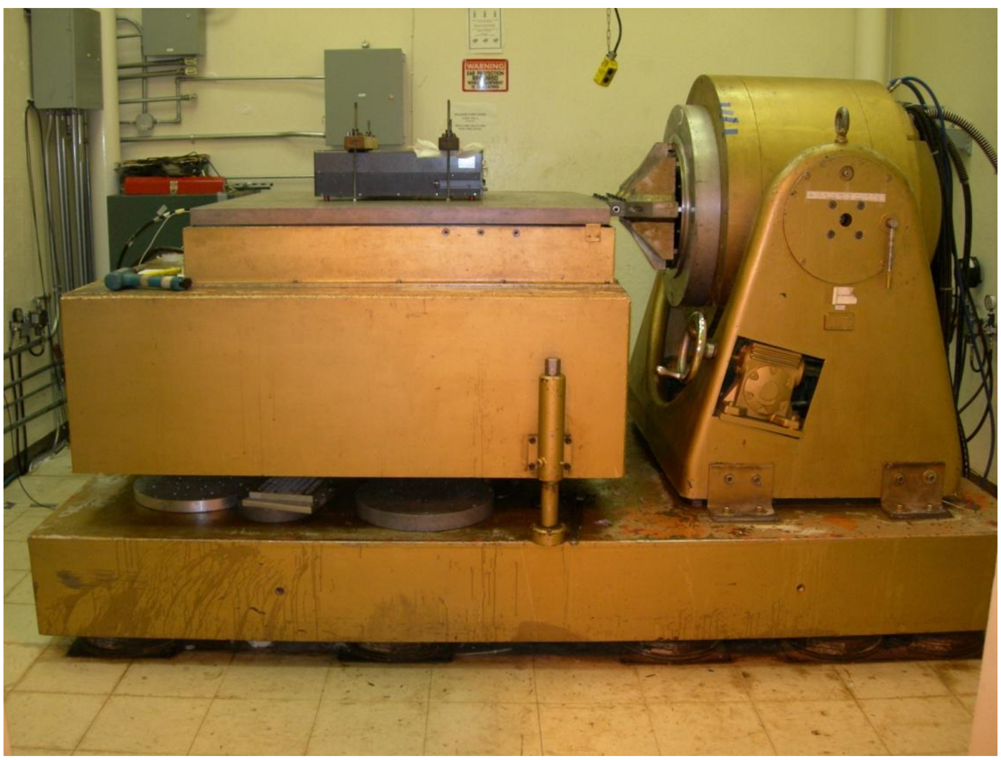
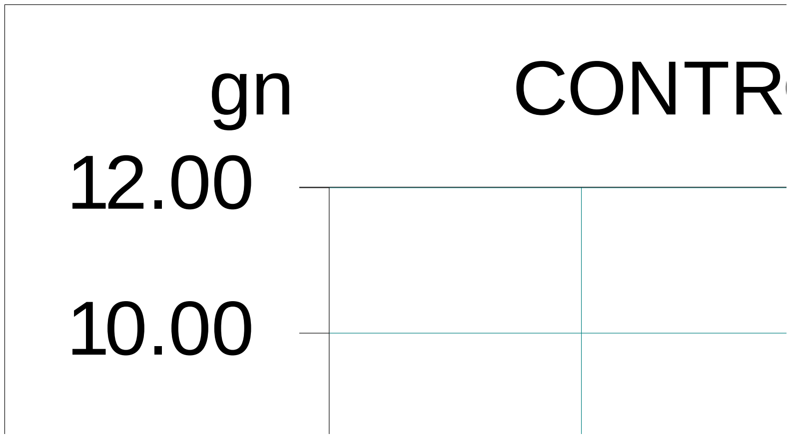
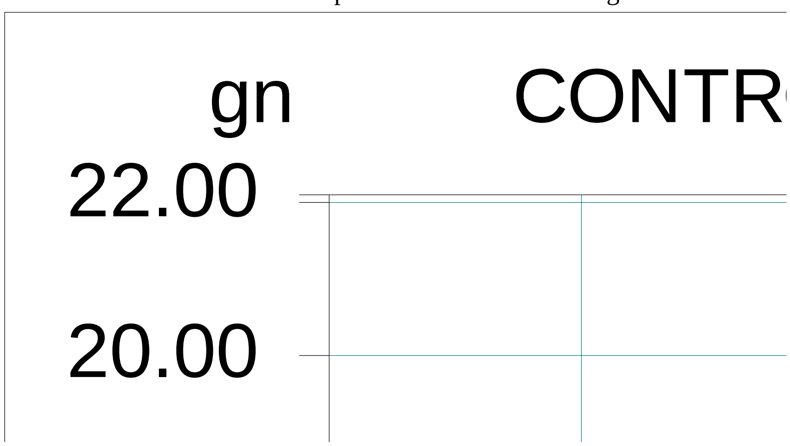


Figure 10: LII Instrument on the Electrodynamic Shaker System.

The following are the examples of measured shock wave forms that were used on each axis. Both positive and negative shocks were used on each axis.



Level: 100 % Block Size: 1024 Elapsed Pulses: 59
Frame Time: 0.200000 Seconds Control Peak: 17.704582 gn Control RMS: 2.474191 gn Full Level Elapsed Pulses: 50
dT: 0.000195 Seconds Demand Peak: 14.999999 gn Demand RMS: 2.454969 gn Remaining Pulses: 0
Pulse Type: Forward Sawtooth Amplitude: 14.999999 gn



Level: 100 % Block Size: 1024 Elapsed Pulses: 59
Frame Time: 0.200000 Seconds Control Peak: 15.558984 gn Control RMS: 2.461471 gn Full Level Elapsed Pulses: 50
dT: 0.000195 Seconds Demand Peak: 14.999999 gn Demand RMS: 2.454969 gn Remaining Pulses: 0
Pulse Type: Forward Sawtooth Amplitude: 14.999999 gn

After the shock tests were completed, the instrument was checked out for alignment. Close observation of the laser beam revealed that it was slightly out of alignment. This was determined to have resulted from a slight movement in the last 2 mirror mounts in the laser beam path. As a result, we located a new mirror mount having a locking mechanism on the adjustments. This innovation delivers much more rigid performance than the existing mirror mounts. We replaced all of the mirror mounts and conducted additional tests using vibration.

Vibration Testing

After the mirror mounts were replaced and the LII 200 system was realigned, we re-tested using the standard for a Category 9 -- Aircraft -- Helicopter Testing, MIL- STD – 810F, January 2000.

The instrument was installed on the shaker table and tested in two axes. The instrument was shaken on two horizontal axes for one half hour each. The attached film strips provide an indication of the intensity of the vibration but do not represent the most intense segments. For example, the bolts holding the instrument down were actually flexing under the vibration/acceleration loads.

At the end of the tests, the instrument was returned to our laboratory and the alignment was thoroughly checked. We found no evidence of any misalignment after these tests. Thus, the new mirror mounts with the locking mechanisms solved our previous problems with misalignment. Furthermore, our concerns with the laser being misaligned or damaged under such severe loading proved to be unwarranted. The vibration testing clearly demonstrated the excellent robustness and ruggedness of the LII 200 optical design.

Besides ruggedness, we also wanted a system that would be compact. Therefore, the design principles employed in the LII 200 were also rolled into the LII 300 which, in addition, also incorporates other changes such as replacing the PC, monitor, ADC I/O card and microcontroller used in the LII 200 with a small “single-board” computer (SBC) and a small high resolution touch-screen LCD display that is integrated directly into the LII box. The implementation of a SBC eliminates several points of failures (hard-drive, interface cables and cards) and also eliminates the need for the user to install any software. The SBC handles all aspects of LII operation and acquisition. More details on the LII300 design are provided later.

TASK 3: *Develop user-friendly software*

This task was aimed at simplifying the software GUI, based on feedback from our industrial project partners. A key software task was to develop an auto-setup method. Auto-setup is a process for automatically (without user input) determining and using the best acquisition parameters for controlling the signal intensity. The LII signal intensity follows a consistent pattern of sudden increase to maximum intensity in the first few nanoseconds (during the laser light pulse), followed by an exponential decay that is observable for typically 1 – 2 microseconds. There are three basic acquisition parameters for LII:

1. Photodetector Gain
2. Filters
3. Laser Fluence

The ADC (Analog-to-Digital Converter) card used for LII has a fixed input range of 2V. In order to maximize the resolution, the input signals should occupy as much of the entire 2V range as possible. The previous version of the AIMS software required the user to make the choice of photodetector gain and neutral density filters (if any) to use. Incorrect settings would lead to lower resolution on the signals acquired by the ADC card, which led to a decreased signal-to-noise ratio, and less precision in the resulting analysis. A lower peak signal would also cause the decaying signal dropping to the noise floor sooner, providing a shorter time base for determining the primary particle size from the decay rate, thus affecting the quality of the measurement.

The LII laser energy is set to a constant value, but the LII photonics package includes a ½-wave plate for controlling the amount of energy reaching the probe volume. This laser fluence needed to be adjusted so the nominal peak temperature of the particulate (as heated by the laser) would be about 3700 to 4000 degrees K, so that significant sublimation of the particles does not occur.

In the LII instruments, filters and photodetector gain are both used to optimize the signal intensity, the combination of which provides over six orders of magnitude of measurement range for particulate concentration. Since the photodetector gain can be adjusted much more rapidly than the motorized filter holder can physically change the filters in the signal path, the selection of filters are determined such that for the mean anticipated concentration, the photodetectors are in the middle of their gain range. This minimizes collection delays due to filter changes.

By automating the selection of these parameters, the software now ensures a correct and consistent setup of the instrument. This process is completed more rapidly in an our newly automated systems than can be done by human operators, even an experienced one. This process is now aso performed for each acquisition run.

Under new auto-setup algorithm, the software samples the incoming LII signals, analyzes the data, and selects the optimum parameters for data acquisition. Under this task, the algorithm for LII auto-setup was developed, implemented, and tested extensively

TASK 4: *Build and test LII instruments for field demonstration*

Under this program, we built a two fully functional, rack –mounted, high sensitivity LII 200 systems for testing at our industrial partner sites, specifically Cummins and John Deere, Figs 11-14,

Figure 11: Prototype high-sensitivity LII 200 system.

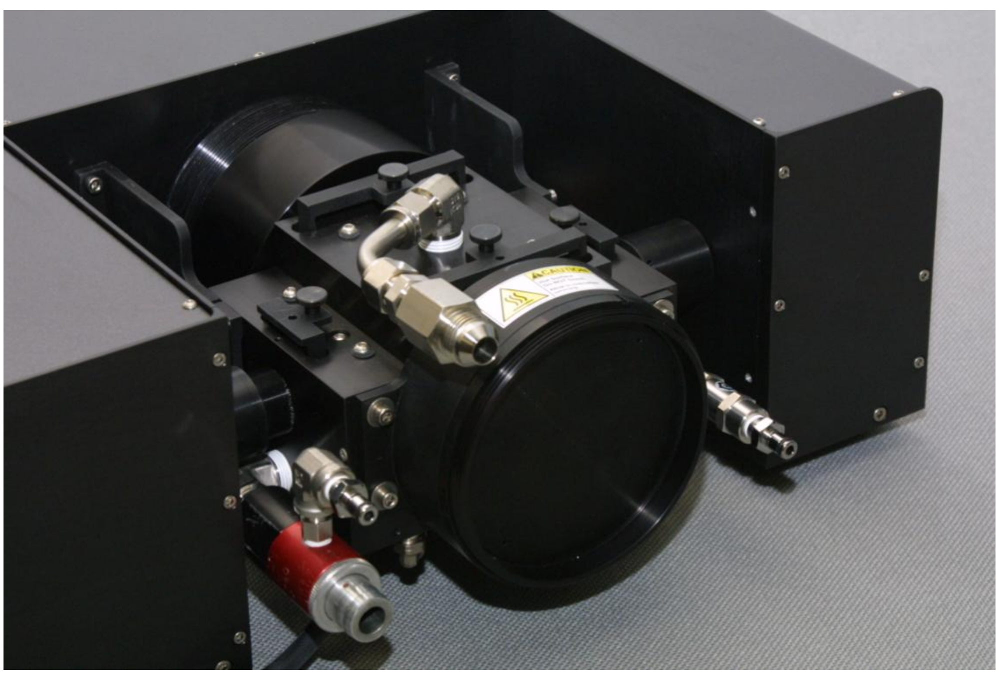


Figure 12: Close up shot of the sampling cell used in the LII 200.

Figure 13: Front side view of the rack-mounted LII 200 system; computer and monitor shown on the right.

Figure 14: Rear view of the rack mounted LII 200 system.

The prototype units were extensively tested in the factory and then shipped to our industrial partners, namely Cummins and John-Deere, for extended testing in their test cells.

Based on the test results and the feedback received, we also proceeded to design, develop and completed a more compact system, namely the LII 300. As mentioned earlier, this instrument integrates the pneumatics, optics, and sample cell into one rack-mountable box, Fig. 15.

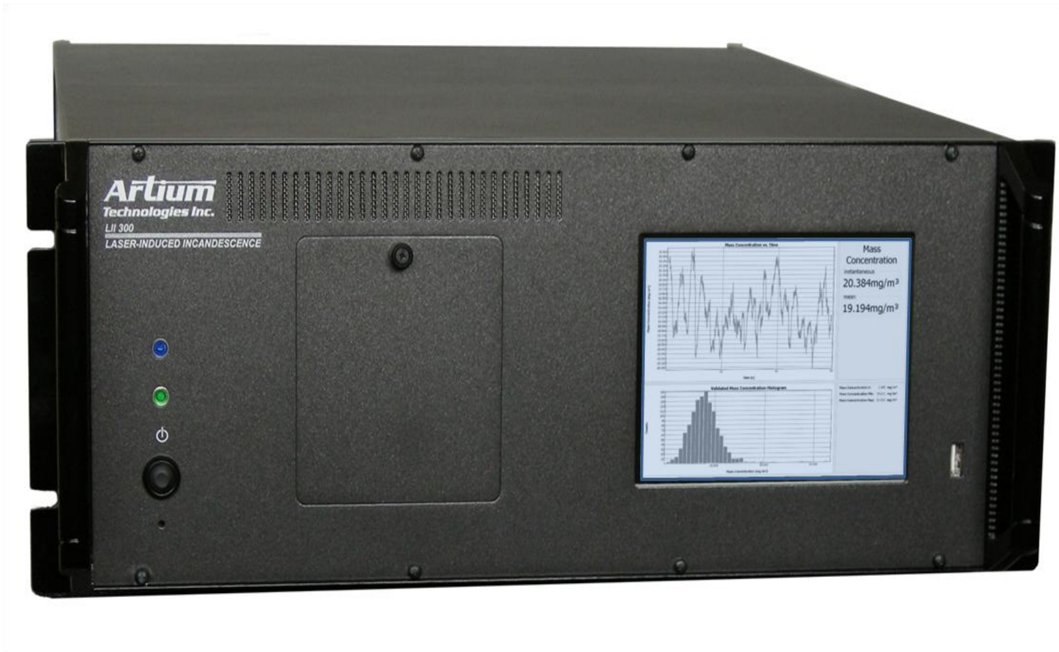


Figure 15: The LII 300 system with integrated pneumatics, optics, sample cell, control computer and touch-screen monitor.

TASK 5: *Extended tests at Cummins and/or other engine manufacturer*

The purpose of this task was to conduct extensive chassis dynamometer tests at the facilities of leading diesel engine manufacturers such as Cummins and John Deere to evaluate the performance of the LII instrument. The scope of the evaluation includes:

- verification of the instrument ease-of-use functionality
- suitability for long term testing in engine tests cells,
- data recording and analyses requirements,
- measurement accuracy,
- measurement precision,
- reliability of the data,
- sensitivity requirements, and
- other operational characteristics required of a robust PM measurement instrument.

The first phase of testing at Cummins was based on the LII 200. The results from this testing was presented at the CRC Meeting (March 2008). Also, successful independent validation of the LII 200 performance was performed by Southwest Research. The second phase of testing at Cummins was with our improved, compact system, namely the LII 300.

The LII 200 instrument was evaluated extensively at the Constant Volume Sampling (CVS) emission laboratory at the Cummins Technical Center, where a Cummins' standard gravimetric PM measurement system was available as a comparative standard. Preliminary testing was conducted in a raw exhaust test cell. The raw exhaust was directly sampled and measured. The results are shown in Figure 16. The same results are presented in Figure 17 where the data from three different cycles have been overlaid to show measurement repeatability. The results presented are for the raw exhaust. A joint paper was presented at the 18th CRC On-Road Vehicle Emissions Workshop in San Diego (March 31-April 2, 2008).

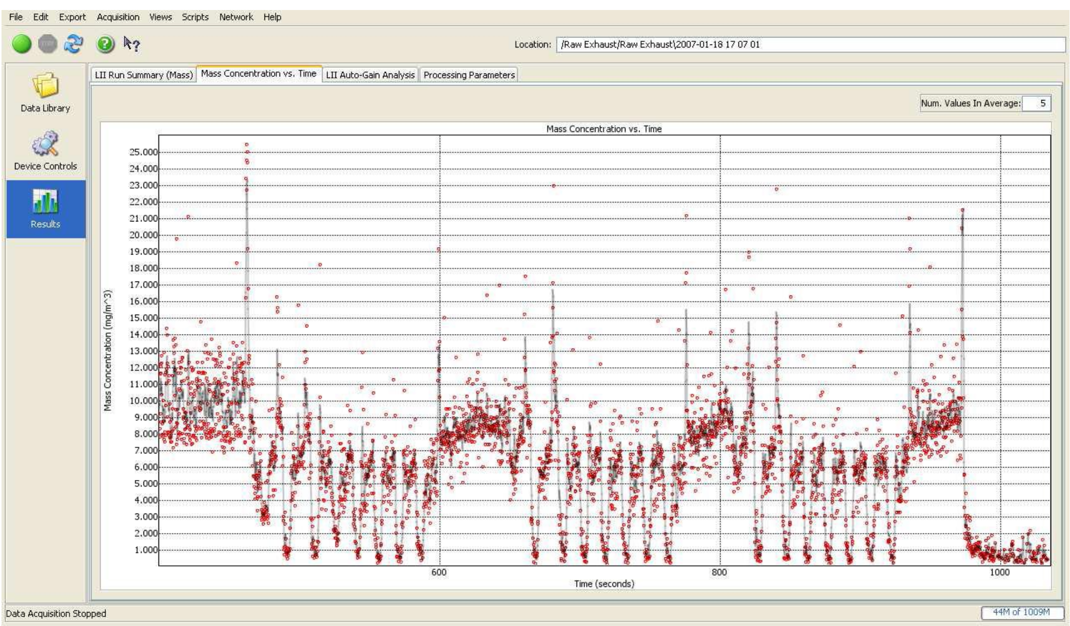


Figure 16: Preliminary data obtained with the LII 200 HS by sampling raw exhaust in a Cummins test cell.

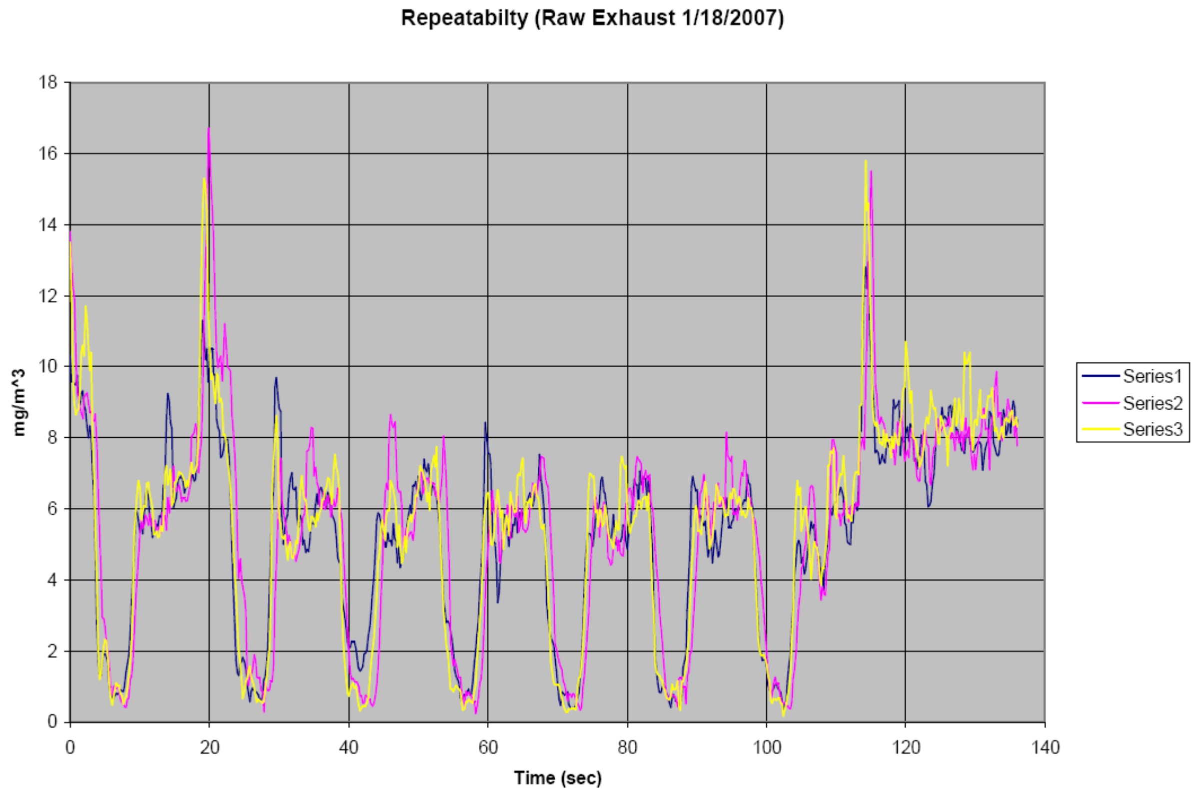


Figure 17: Data from three cycles overlaid to show excellent repeatability in the measurement.

The LII 200 was tested in the dilution tunnel as well as in the raw exhaust. The Artium AIMS results (two runs of transient dilution tunnel at 20 Hz, two runs of transient dilution tunnel at 10 Hz, and two runs of transient raw exhaust at 10 Hz) were exported to Excel. A running average in Excel was calculated at 5 Hz and then the data was extracted at 0.2 s intervals and plotted. The time base for each run was shifted so that the transient events approximately coincided. The results are presented separately for the data from the dilution tunnel, Figures 18-19, and the data from the raw exhaust, Figures 20-23, as these two sampling locations were not directly comparable.

The repeatability of the LII-200 instrument was demonstrated and shown in Figures 18-23 with four runs from the dilution tunnel, Figure 18-19, showing excellent agreement for the timing of the transient events in the off-road cycle that the engine was performing.

At low to moderate concentrations, the four runs also showed excellent agreement in terms of the magnitude of the soot mass concentration. However, the high spikes were somewhat less repeatable. In some cases, this may have been due to an issue of outliers (i.e. single point high values without a rising and falling slope before and after the maximum) or large particles as is discussed later in this section.

The different time segment data shown in figures 18 – 24 (expanded segments of the overall run time for that test) demonstrated the excellent agreement in terms of timing and magnitude for the low-to-moderate concentrations, and somewhat better agreement for some of the high spikes in concentration.

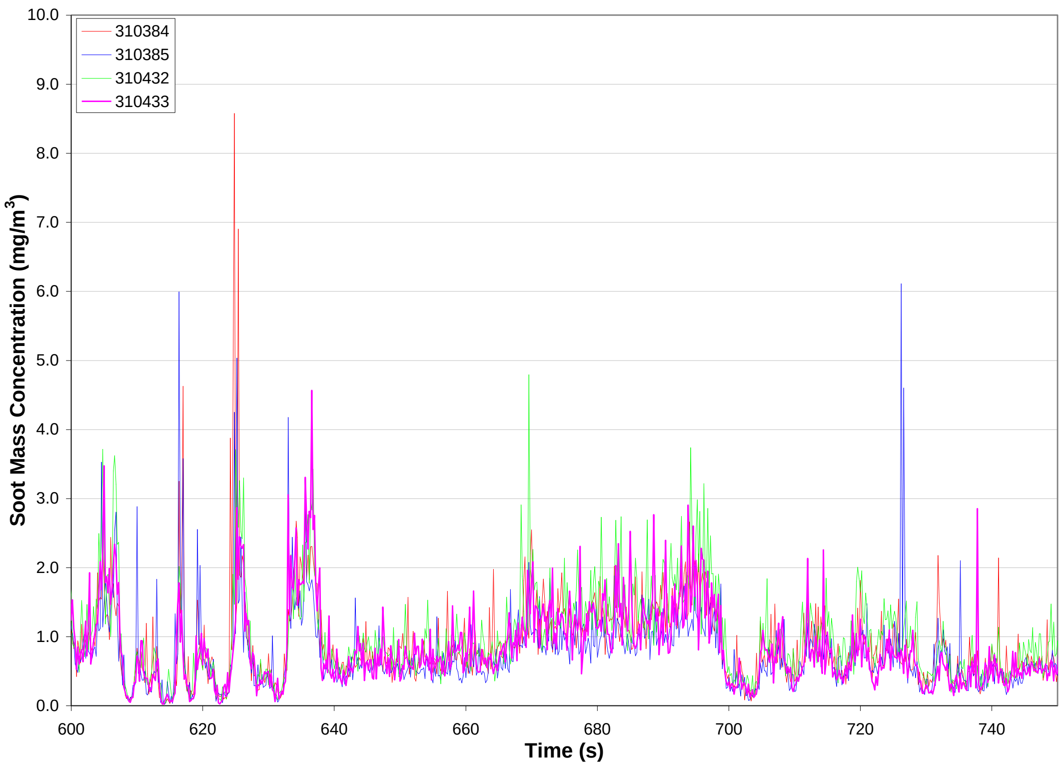


Figure 18: Dilution Tunnel (time segment from 600-750s – four repeats).

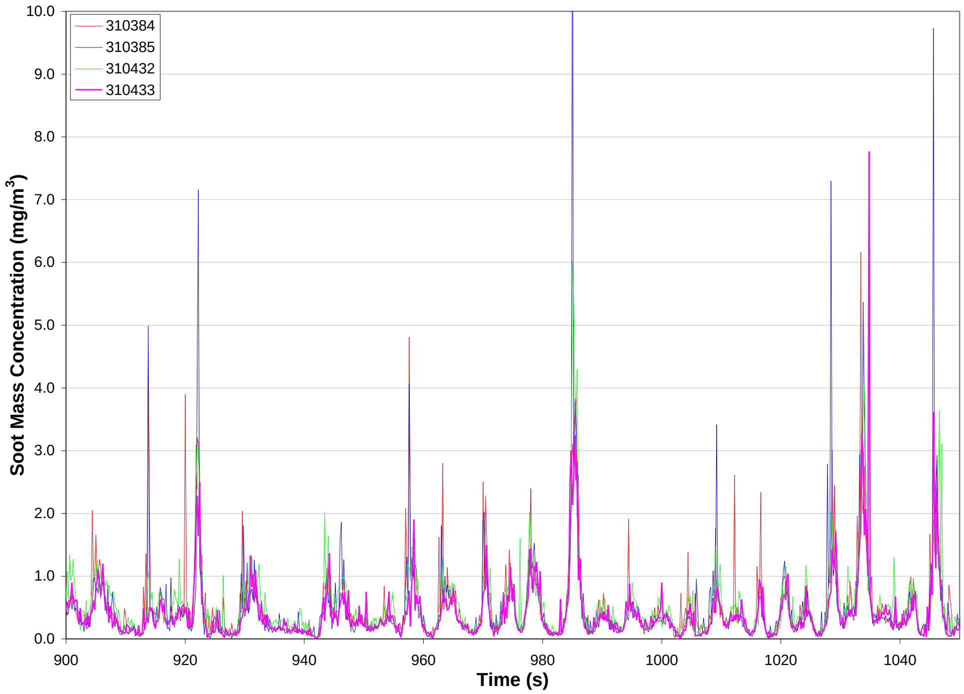


Figure 19: Dilution Tunnel (time segment from 900-1050s – four repeats).

The two runs from the raw exhaust appeared to present even better agreement, with similar magnitudes reported for the two runs on many of the high spikes in concentration, Figures 20-22.

The raw exhaust data shown in figures 20 – 22 seemed to have fewer outliers, possibly due to the higher concentrations and correspondingly higher signals for the Artium AIMS software to process.

To demonstrate the ability of the LII-200 instrument to follow rapid transients, the data for a time segment from 790 to 800 seconds is presented in Figure 23. This followed a sharp reduction from a high soot mass concentration of ~25 mg/m3 to a low soot mass concentration of ~0.2 mg/m^3 in 1.2 seconds. This change was greater than a two-order-of-magnitude reduction in concentration documented with 12 discrete measurement points at 100 ms intervals. Note that for the two repeats, between 791.16s and 791.75s, the LII-200 instrument tracked an increase from ~10 mg/m^3 to ~25 mg/m^3 and then a decrease to ~3 mg/m^3, all in less than 0.6 s.

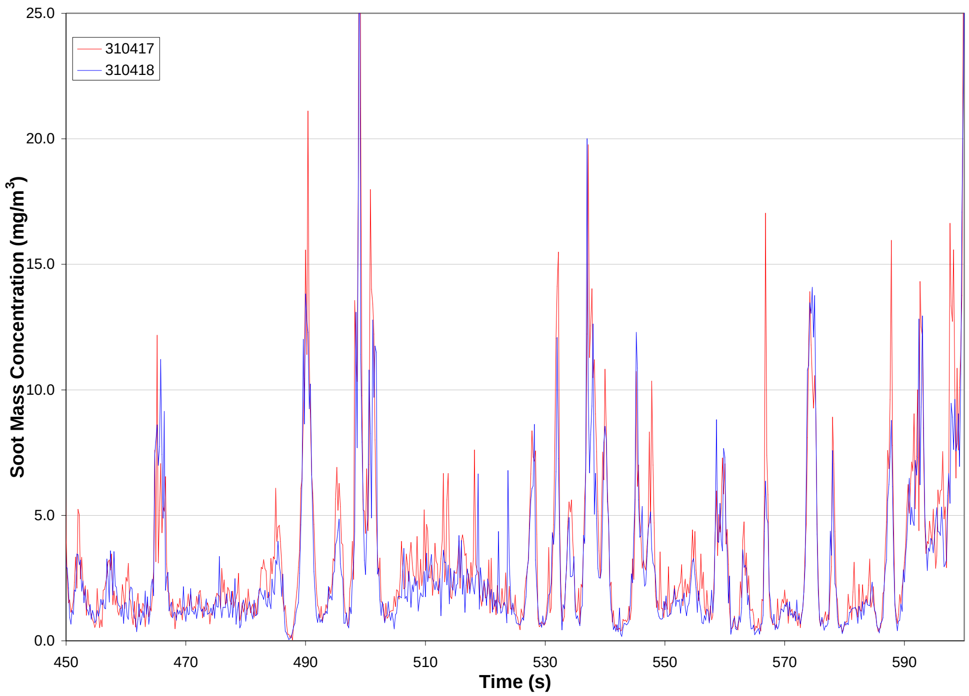


Figure 20: Raw Exhaust (time segment from 450-600s – two repeats).

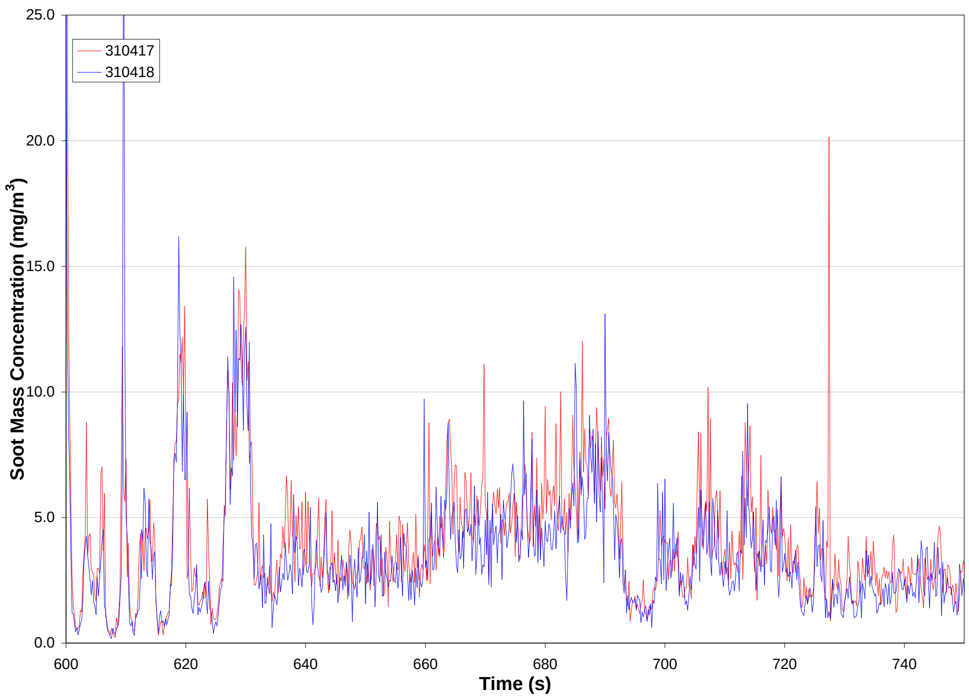


Figure 21: Raw Exhaust (600-750s – two repeats).

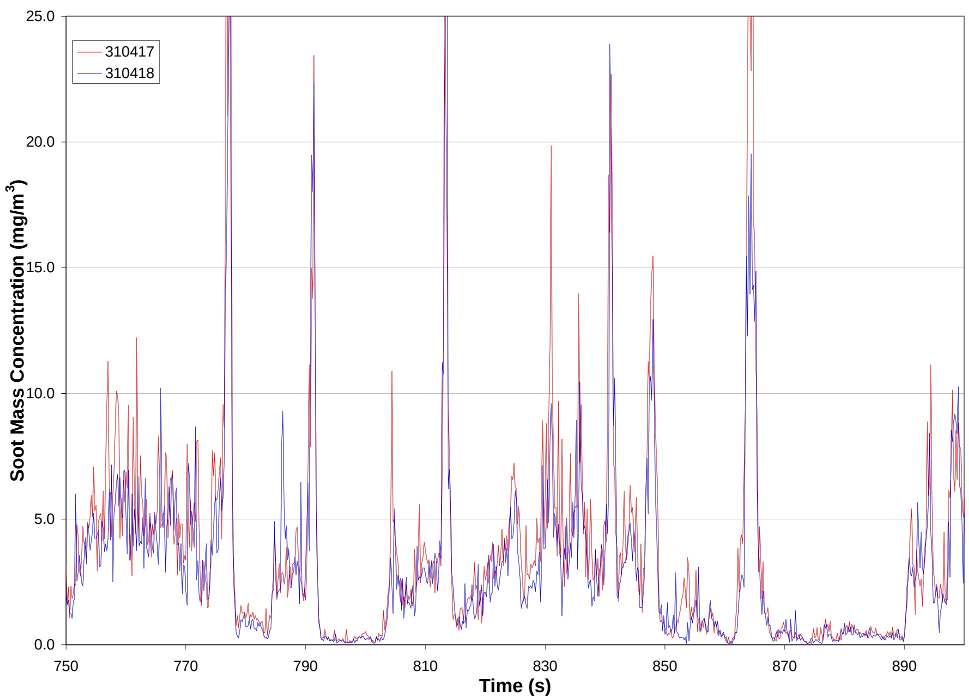


Figure 22: Raw Exhaust (750-900s – two repeats).

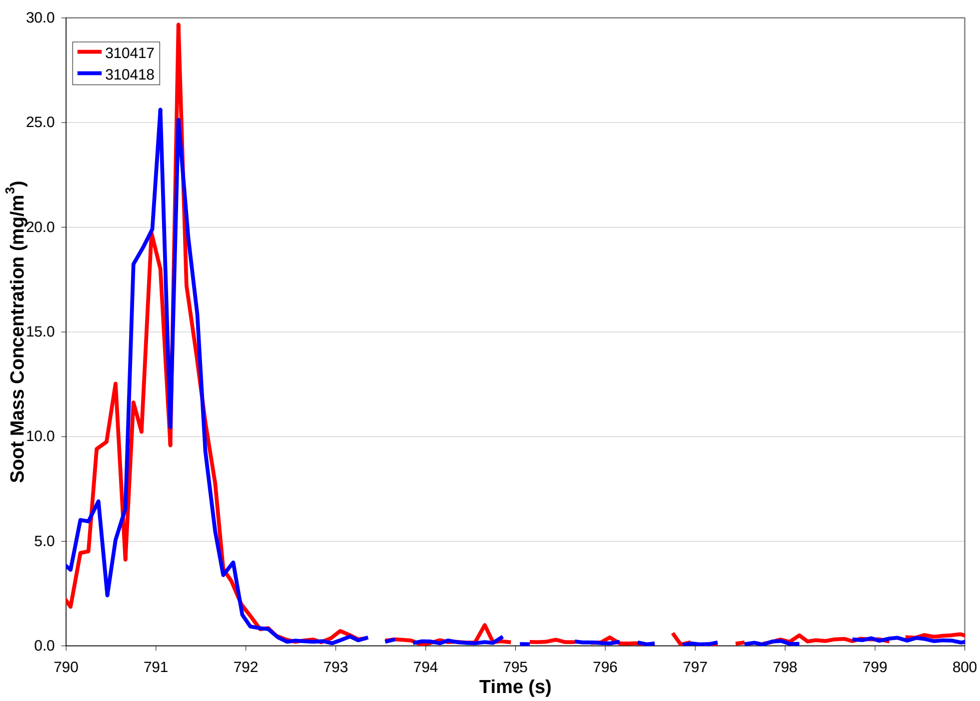


Figure 23: Raw Exhaust (790-800s – two repeats).

Preliminary tests were also conducted successfully at John-Deere where the measurements were made in the raw exhaust with after-treatment. Excellent repeatability for these measurements is shown in Figure 24.

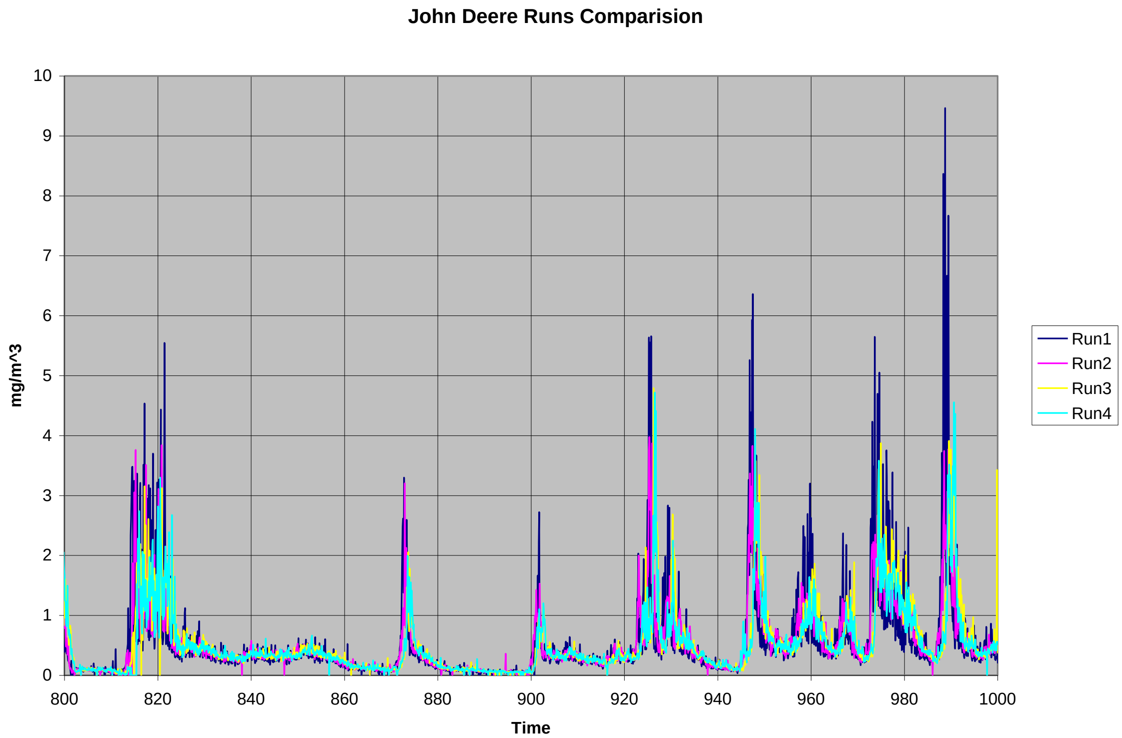


Figure 24: Raw exhaust data comparison.

Cummins wanted an independent, third-party, validation of the LII system performance. Therefore, they contracted with Southwest Research to evaluate the LII system. The tests by Southwest Research included a comparison of the LII with an established EC/OC measurement technique. Excellent linearity was demonstrated between the two techniques over a large range, Figure 25. This test also clearly demonstrated the validity of the factory calibration and its ability to not drift over time or be impacted by instrument shipment. The difference in slope (approximately 30%) may be attributed to a number of reasons including differences in response of the two methods to diffusion flame soot, a problem with the calibration of one or the other (or both) instruments, differences in the sampling location, effects of soot morphology, soot density, etc.. If one method can be trusted to provide soot volume fraction (SVF, defined as the volume of soot primary particles in a volume of space) with low measurement uncertainty, then the response slope could be corrected and called a “calibration factor”. Unfortunately, we have not reached a point where we can conclude that any instrument is reliably reporting the “true value” or SVF. Note that the vertical bars on the graph are not “error bars” but rather the rms fluctuations of the measured data.

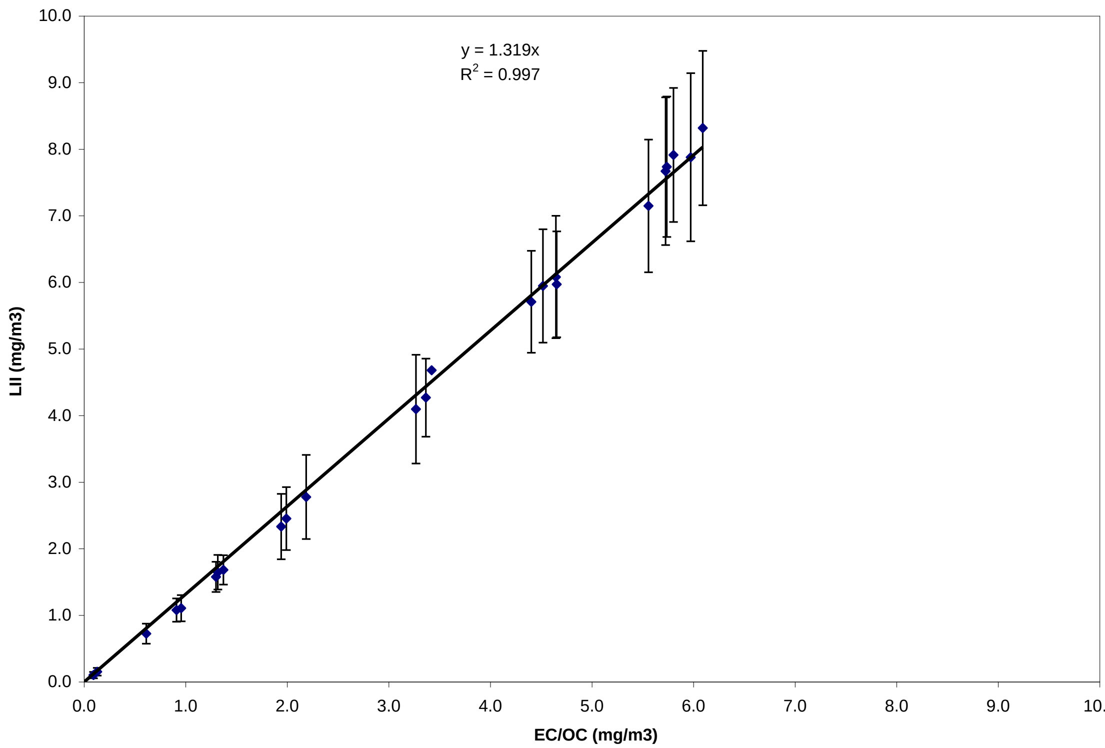


Figure 25: Comparison of LII and EC/OC soot concentration measurements.

A final round of testing was also conducted at our partner site, Cummins with the LII 200. The LII instrument was thoroughly evaluated and calibrated before being returned to the Cummins Technical Center (CTC). Subsequent tests were conducted from April 28 to April 30, 2009 and were designed to evaluate instrument performance for a range of sampling conditions including engine with DPF, engine out using CVS, and sampling raw exhaust.

During this program, we have spent some considerable effort trying to determine why we were achieving exceptionally good agreement when measuring soot produced by gaseous diffusion flames (data obtained at NASA GRC using CAST burners, as shown in Figures 26 & 27) but could not achieve such good agreement measuring diesel and gas turbine soot particulate. Note that in both cases, figure 26 and 27, the LII measurements are lower than the filter based method and the SMPS since these methods measure all of the particulate material (PM) whereas the LII isstrument only responds to the elemental carbon (soot). When measuring diesel particulate emissions and simulated gas turbine particulate emissions, LII has typically reported values that are too low. At low concentrations, values have been as much as an order of magnitude lower than the filter-based techniques were reporting. There was also some evidence that when the SVF was higher than 2 to 5 mg/m^3, the LII instrument would report values that were too high.

It was crucial that we resolve these differences and apparent uncertainties in the measurements if LII instruments are to be accepted as a superior method for characterizing soot emissions in real time. LII has the significant potential of measuring soot emissions where there are no CVS systems. The major attraction of LII is its ability to measure raw exhaust in real time. This unique capability represents major advantages of reducing facility costs and significantly reducing engine development time.

Our hypothesis was that the LII instrument was producing soot volume fraction measurements that did not agree with filter techniques due to the presence of very large particles in the samples. Soot accumulation on combustor surfaces, exhaust pipes and within sampling tubes was assumed to subsequently shed as large particles (1 to 50 µm in diameter). These large particles were observed on smoke meter and gravimetric filters. An objective of the tests conducted at CTC was to determine whether large particles formed, shed, and collected in the gravimetric samples. We would also estimate if the elimination of these large particles could result in better agreement between gravimetric and LII measurements.

Prior to the measurements, we checked all connections on the sampling system and all connections were determined to be airtight. The test cell windows were cleaned even though they only had slight contamination. We connected the sampling cell to the heated hose, checked suction, and determined that the sampling system was working properly. Room air was measured and showed usable signals although they were noisy. Room air was measured as having a soot concentration of 4 μg/m3. We demonstrated that the instrument had sufficient sensitivity to measure down to 1 or 2 μg/m^3.

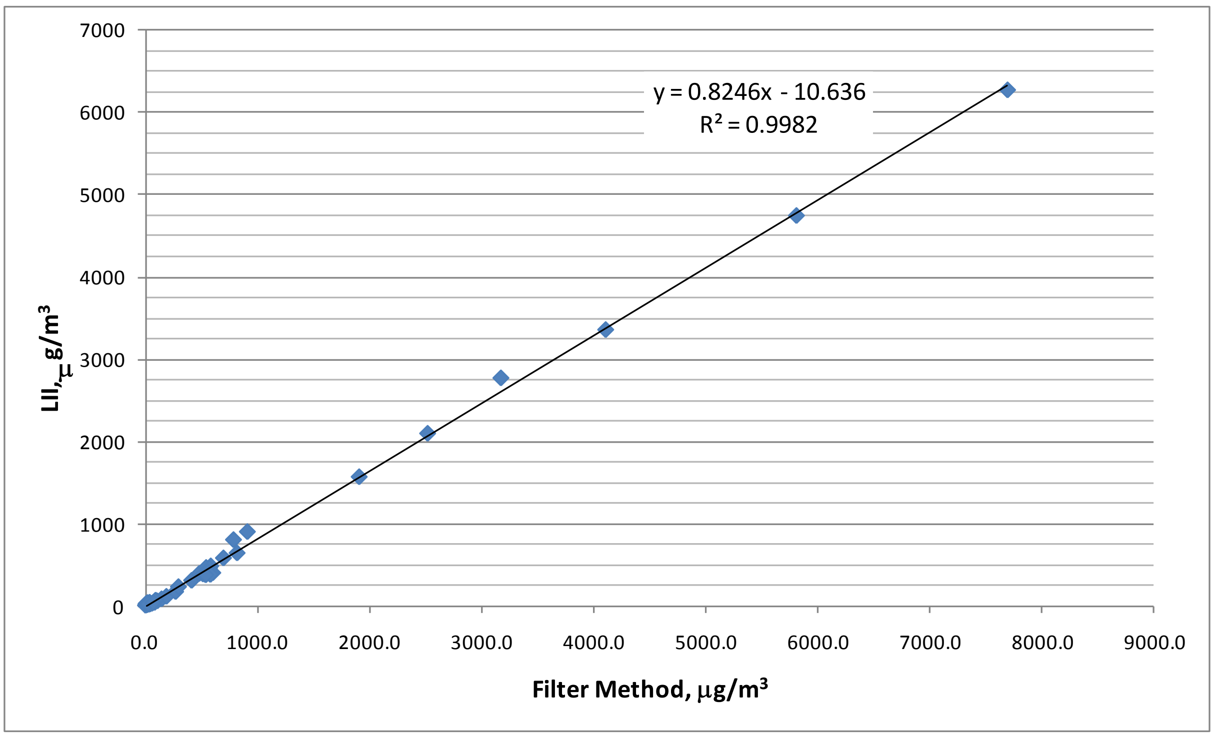


Figure 26: NASA Glenn Research Center tests conducted on soot from a mini-CAST burner in December 2009 prior to measuring gas turbine particulate emissions.

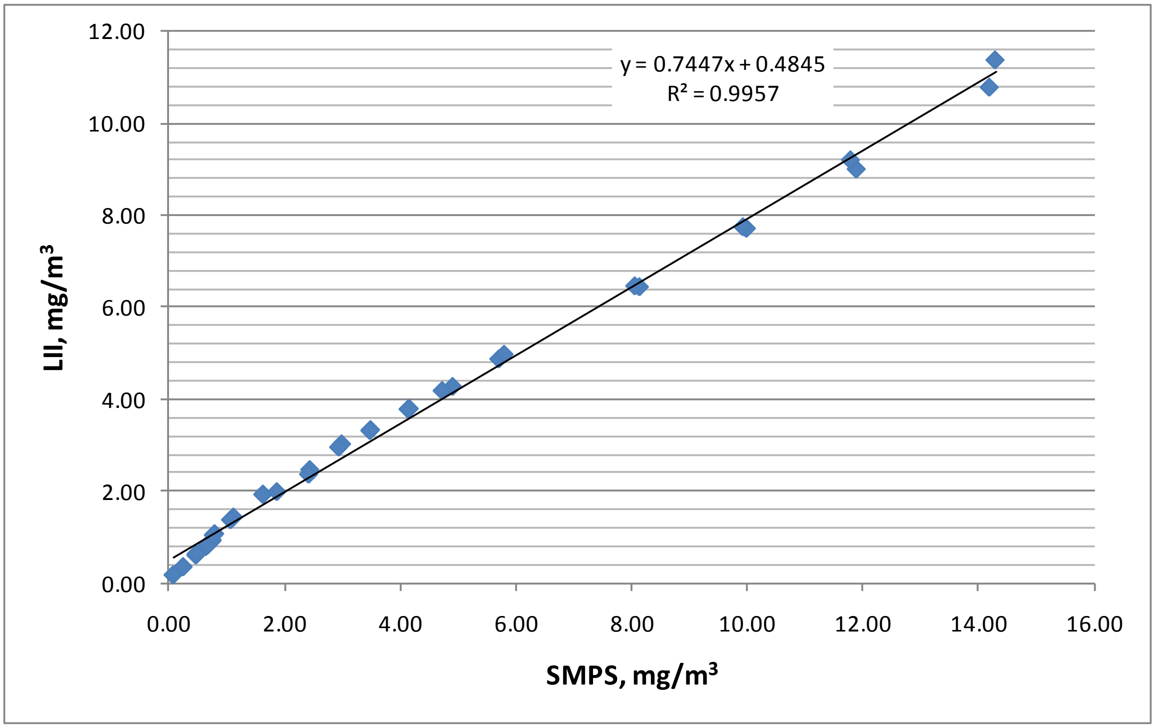


Figure 27: NASA Glenn measurements of soot generated by the mini-CAST burner and compared to the TSI SMPS measurements.

About 50 filters from various previous tests at CTC were obtained and studied under an optical microscope, see Figs. 28 and 29.. Most filters revealed the presence of big particles (1 to 50 μm), all with similar morphology and size distributions. CTC personnel also observed the filters and confirmed the presence of these large particles. We were assured that these filter results were collected after 2.5 μm cyclone separators which are built into the gravimetric sampling systems. For example, filters indicating a weight of approximately 40 μg of soot had a relatively large number of large particles (roughly estimated as 10,000 from a microscope count) as well as many more smaller particles trapped in the filter mesh. The lighter loaded filters (10 to 20μg) had fewer particles but they were present and small particles (estimated at <1 μm) could also be observed. The questions were, where did this agglomeration of particles occur? Did they happen in the engine or in the sampling system? Did these particles represent the actual PM emissions?

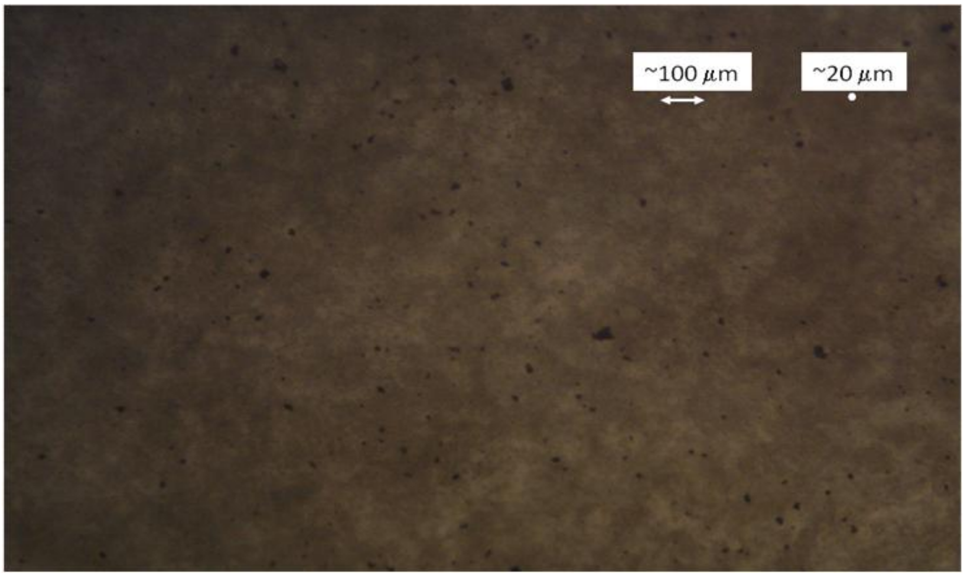


Figure 28: Optical microscope images of soot filters obtained April 28, 2009. Filter data was obtained downstream of a cyclone separator supposedly able to cut off particles larger than 2.5 µm. The scale shown in the image is approximate and is provided for a rough reference for particle size estimation.

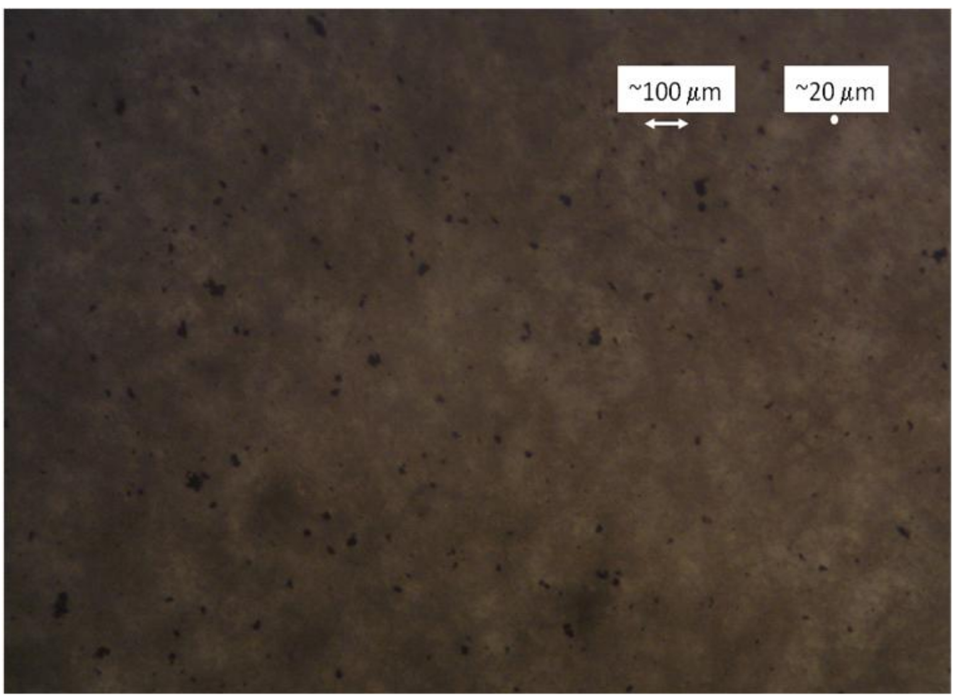


Figure 29: Optical microscope images of soot filters obtained April 28, 2009. Filter data was obtained downstream of a cyclone separator supposedly able to cut off particles larger than 2.5 µm. Note that there are particles as large as 50 µm in these images. The filters were contained in the Petri dishes and were not contaminated during the observation process.

Clearly, the cyclone separators which were supposed to remove particles larger than 2.5 µm were not completely effective in removing large particles. These data provide irrefutable evidence that the filters were collecting large particles. The LII instrument was not designed to measure such large particles reliably. Furthermore, the regulations specify that particles larger than 2.5 µm should be removed before gravimetric analysis. If these large particles are present, other means will be required to characterize them. However, such large particles have not been considered as a part of the soot mass emissions of interest.

Subsequently, tests were conducted with the LII instrument on the Cummins CVS. Three cycles were measured to determine repeatability and to evaluate the quantitative results as compared to gravimetric. Typical results for the LII soot volume fraction versus time are shown in Figs. 30 and 31.. These results show very low variations for the shot to shot measurements. Clearly, this suggests that there were few, if any, large particles in these samples. The large excursions and soot volume fraction were due to the changing engine conditions of this FTP cycle, and not due to individual large particles. A closer examination of the time record shows that the LII readings systematically track the rise and fall of the soot volume fraction with time. That is, the individual readings consistently increase and then consistently decrease rather than showing very large changes for individual LII readings.

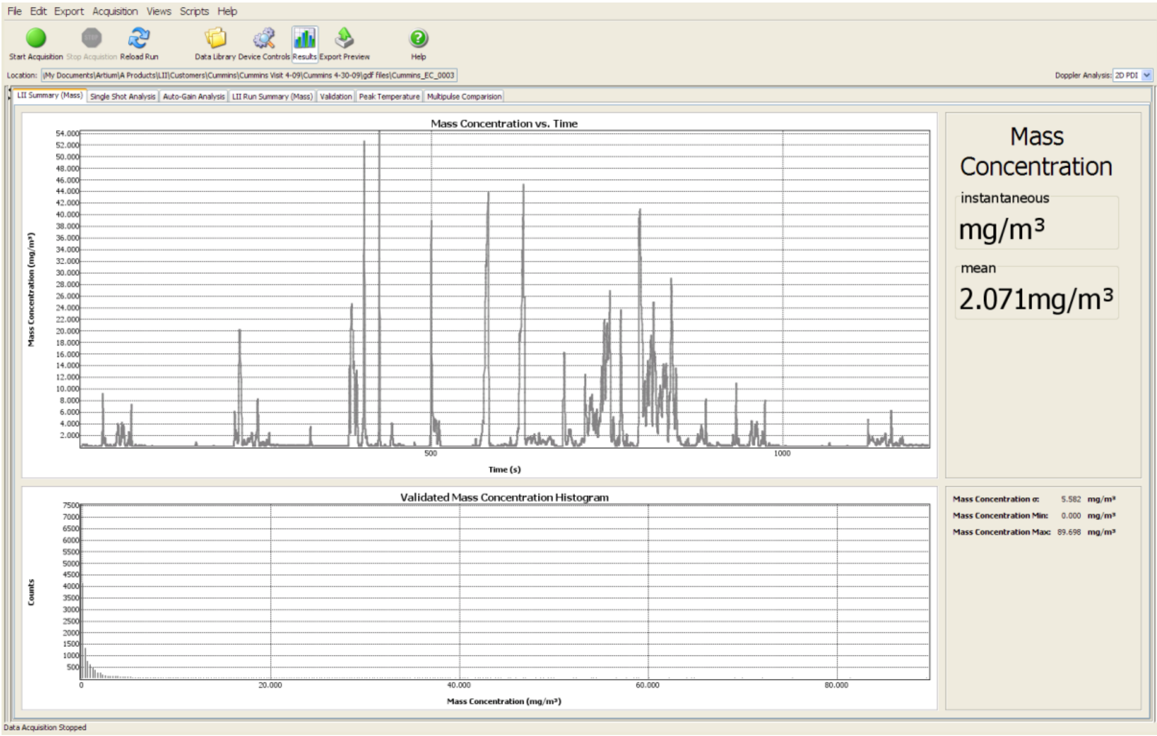


Figure 30: In this test, the sampling was essentially ideal in the sense that there were no large spikes indicating the presence of large particles. The CVS and sampling system appeared to be clean and the LII signals were very consistent with very low variations shot to shot except for changes in engine cycles.

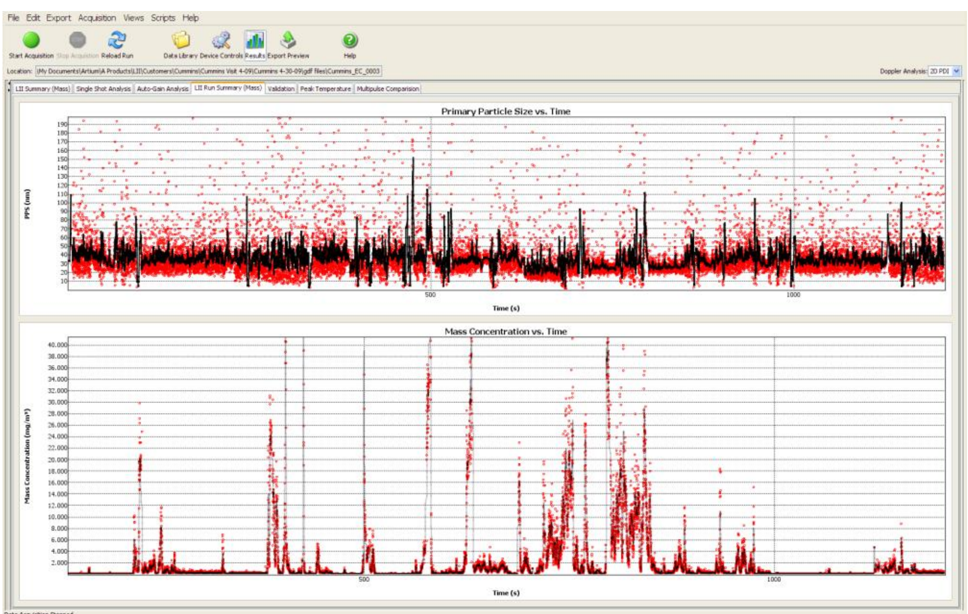


Figure 31: A closer observation of a section of the FTP cycle shows that the LII measurements consistently rise and fall shot to shot indicating that there were no individual spikes in the data. This implies that there are no large particles in the sample and so the results should agree with gravimetric.

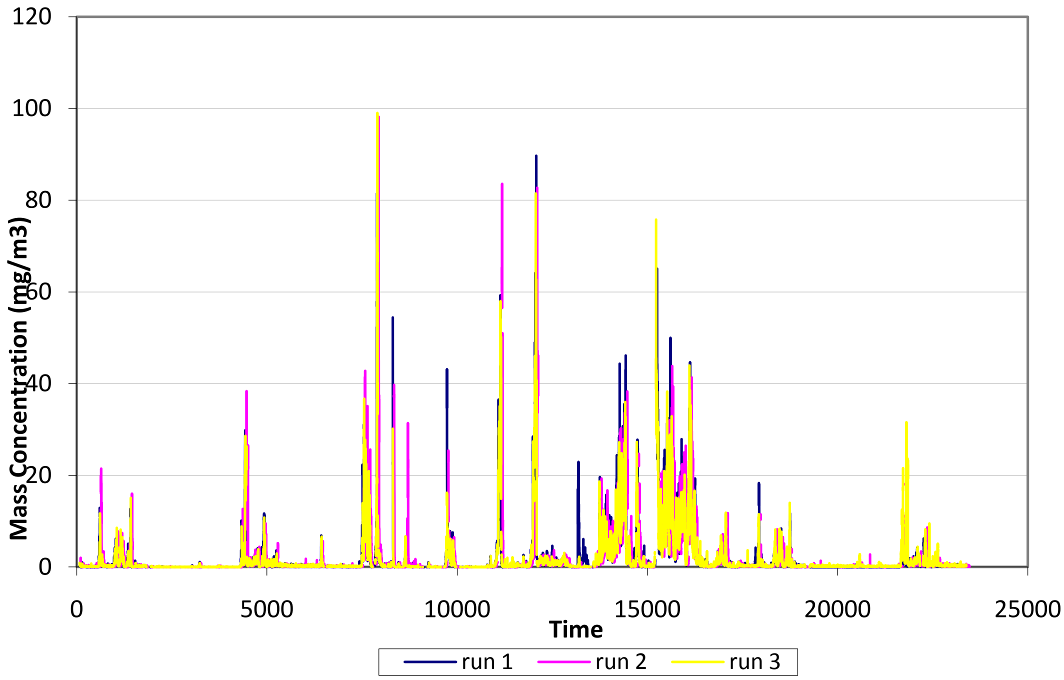


Figure 32: Three repeated FTP cycles are overlaid in this figure showing the excellent agreement from test to test.

Figure 32 consists of three overlaid FTP runs emphasizing the excellent agreement from run to run. The LII instrument was able to track very fast changes in soot volume fraction, especially when sampling at 20 Hz. The sampling rate was more than adequate for tracking the large excursions in soot volume fraction as engine conditions were changed abruptly.

Without large particles present, the LII measurements agreed relatively well with the gravimetric results. The gravimetric values were approximately 30% higher than the LII measurements but this was expected since the gravimetric filters collect both the elemental carbon (EC) as well as the SOF material. We expect that comparison with extracted filter data would be much better.

run #	cycle	work (bhp)	cvs flow (scfm)	gravimetric PM (g)	cvs flow (m3/sec)	LII conc. (mg/m3)	LII soot (g)
a438445	FTP	29.99	2117.0	3.045	0.999	38908.109	1.944
a438446	FTP	29.99	2113.0	3.008	0.997	30866.534	1.539
a438447	FTP	30.00	2111.4	3.025	0.996	37997.471	1.893

From these observations one must conclude that the sampling was problematic and must be corrected. That is, large particles were being generated by soot accumulating on surfaces and subsequently re-entrained into the flow. These large particles cannot be measured reliably by the LII method alone. Furthermore, these large particles produce ambiguity to the gravimetric results since it is difficult to predict when the particles are accumulated on the wall and when they were released.

To obtain reliable measurements, cyclones that perform as specified need to be added to both the gravimetric sampling line and the LII sampling line so that instruments are measuring under the same conditions. Gravimetric filters need to be systematically examined using an optical microscope to determine whether or not large particles are present. These large particles need to be accounted for in the results since they can be a significant contributors to the weight of the collected soot mass.

It appears that the LII instrument generally has been measuring soot properly and that the gravimetric method was flawed. It was not only measuring normal soot particulate but also large agglomerates of soot which were artifacts in the sampling process, namely, particles that were entrained from the engine and sampling system surfaces. Consistent agreement between LII and other instruments will not be achieved unless the sampling system is operating properly wherein the sample consists of soot aggregates and not large particles.

The LII is very robust in that it will not detect or measure condensed material or material other than elemental carbon. LII can also measure directly from the engine exhaust which may be a very important requirement since that would limit the amount of agglomeration of soot particulate into large particles (greater than 1 μm). Furthermore, the need for costly CVS systems can be eliminated.

The LII 300 has also been tested at Cummins and the results compared extremely well with the gravimetric technique and were also very repeatable, Fig 33.

run #	cycle	PM (g/bhp)	work (bhp)	cvs flow (scfm)	gravimetric PM (g)	cvs flow (m3/sec)	LII soot (g)	LII (g/bhp-hr)	LII/Grav PM
A448733	EPA 2010	0.1072	31.46	2035	3.373	0.96	2.520	0.0801	**0.7473**
A448734	EPA 2010	0.1065	31.45	2035	3.349	0.96	2.504	0.0796	**0.7477**

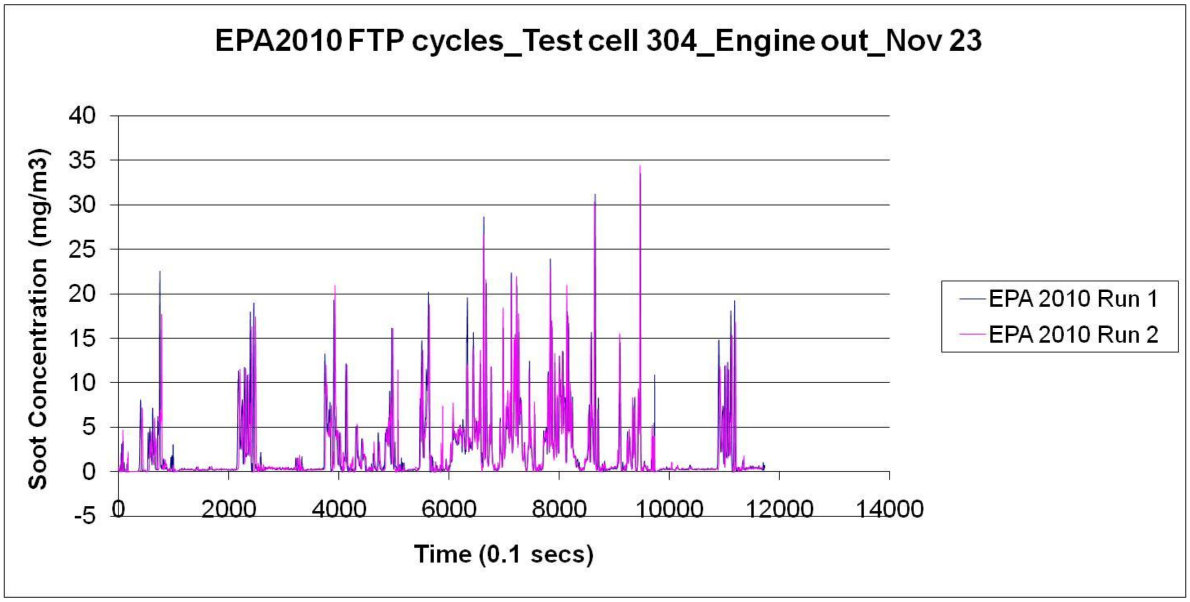


Figure 33: Repeatability of LII 300 runs at Cummins.

TASK 6: *On-board road testing*

This task involved the on-board road testing of the LII 300. This testing provided basic information on any further operational requirements for a Portable Emissions Measurement System (PEMS). The LII 300, including the laser controller and a portable generator, were carried on-board an SUV and driven to the vicinity of the Port of Oakland. The vehicle was parked on the street, and the air was sniffed continuously for soot emissions which were correlated to the passing of diesel trucks. Figure 33-36 shows the picture of the setup. Typical LII 300 results are presented in Figs 38-40. The overall soot concentration measured was very low which could have been attributed to the generally clean air on the day of the measurement and the reduced particulate generation by the trucks.

Figure 34: LII 300 along with the portable generator in the back of an SUV.

Figure 35: SUV with the LII 300 parked in the vicinity of the Port of Oakland.

Figure 36: Sampling line to sniff the air for soot particles when trucks pass by.

Figure 37: A line of trucks waiting to load and unload goods at the Port of Oakland.

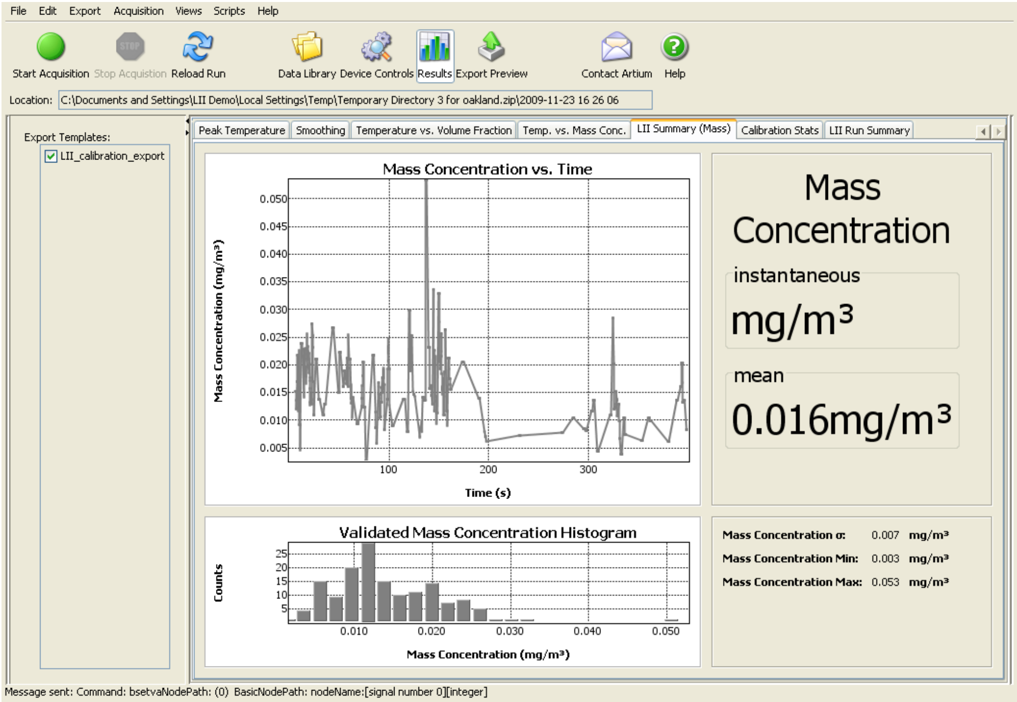


Figure 38: Plot showing measurement of soot mass concentration in air.

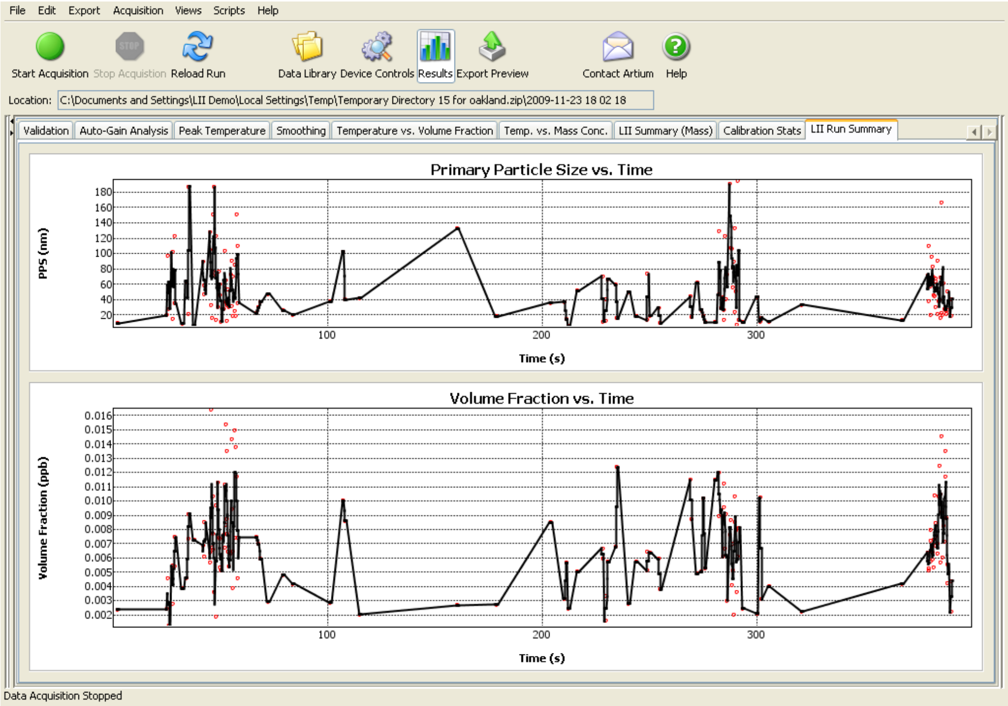


Figure 39: Plot showing variation of primary particle size and soot volume fraction with time.

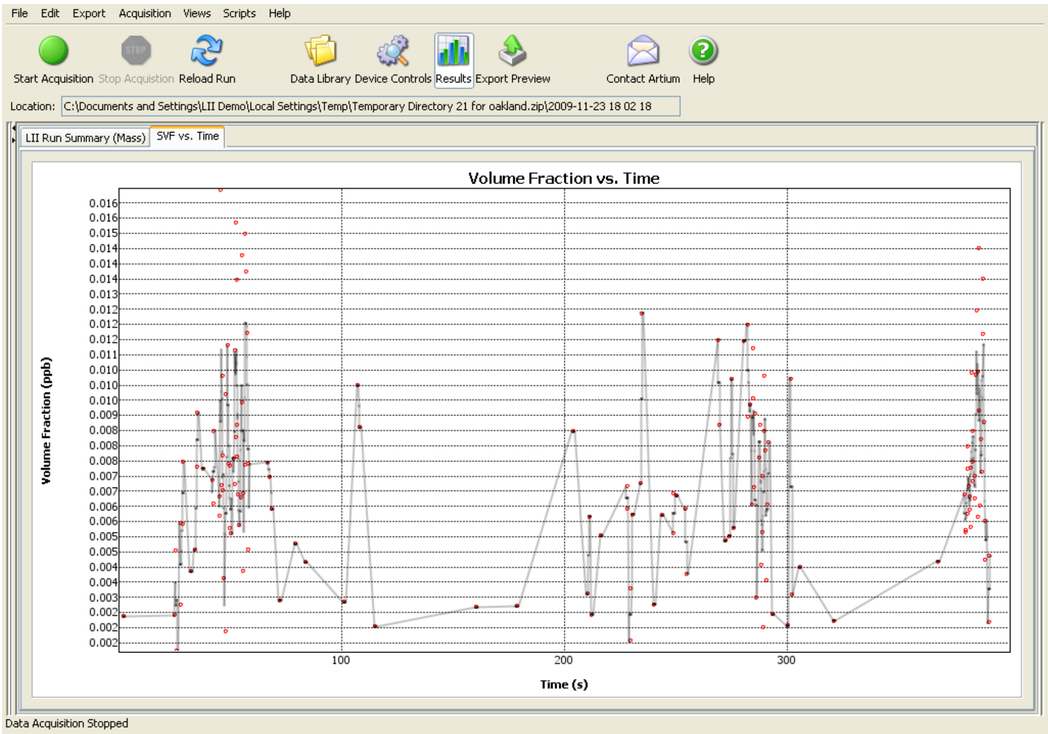


Figure 40: Plot showing variation of soot volume fraction with time.

4. Status of the Technology

The current development program had a significant impact on the commercial readiness of this technology. The LII instrument was carefully reviewed and evaluated under realistic industrial measurement conditions. Given the opportunity to work directly with industrial users was a very important component of this development effort. We were able to learn about the application requirements, gain a better understanding of the level of expertise of the users, and get a firsthand appreciation of the measurement environments in which the instruments are expected to operate. These findings were incorporated in the evolving design of the new generation LII 300 instrument.

Based on the qualifications of the potential users, we revised and enhanced our instrument automation algorithms making the instrument easier-to-use and requiring less maintenance and servicing during extensive operation. Our sampling cell and purge systems were refined to guarantee that there would be little or no window contamination over months of operation. As such, no maintenance is necessary except for the occasional simple cleaning of the windows after a month or more of continuous operation. Our current instrument has evolved with a built-in computer, touchscreen control with easy commands and a data display format that is direct and provides only the information needed to determine that the instrument is working properly. Much of the complexity of the earlier LII 200 instrument was removed from the current generation system. The current instrument is very portable while providing a complete set of measurement information. We included the real-time measurement of temperature and pressure in the sampling cell so that the soot volume fraction measurements can be reduced to STP or any other standard.

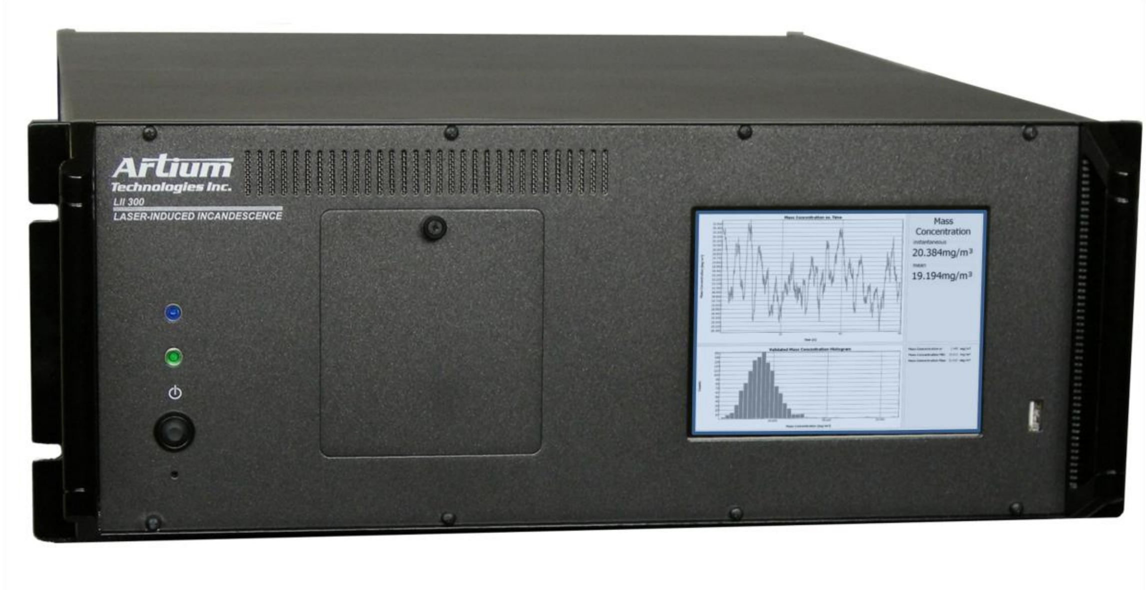
Extensive measurements conducted by our industrial partner have shown that the current LII 300 performs very well in the field, is easy to use, and provides measurements that are in very good agreement to gravimetric. The first LII 300 version had improved sensitivity to low SVF measurements but was still not able to measure DPF soot levels from the CVS dilution chamber reliably. A revised LII 300 has been developed with a new innovation allowing from 5 to 10 times higher sensitivity to low SVF values. This unit will be delivered to our industrial partner for testing. We expect that it will provide more than adequate sensitivity to the very low soot concentrations produced by the latest generation diesel engines. We have also shown that this instrument will be able to measure ambient soot concentrations in areas where there are diesel truck or other diesel engine operations.

Thus, we are convinced that are instrument is ready for the commercial market. The new LII 300 offers ease-of-use, can measure engine exhaust directly without the use of expensive dilution tunnels, provides excellent repeatability and precision, and the measurements agree with gravimetric. Furthermore, we have been able to reduce the costs of production of these latest generation instruments. Our instruments have much greater dynamic range, much better sensitivity to low soot concentrations, and are cost competitive with other instruments in the field. Furthermore, we provide real-time measurements of only soot or elemental carbon and the measurements are not affected by any condensed material or other particulate.

ARTIUM TECHNOLOGIES, INC.
150 West Iowa Avenue, Unit 202
Sunnyvale, California, 94086

LII 300 LASER-INDUCED INCANDESCENCE
Instrument for Soot Characterization

Artium Technologies, Inc. introduces the **LII 300** system, the most advanced laser-induced incandescence instrument available in the market today. Laser-induced incandescence is an optical technique for accurate, non-intrusive, and temporally resolved measurement of soot concentration, specific surface area, and primary particle diameter.



Measures Soot Concentration (mass or volume basis), Specific Surface Area, and Primary Particle Diameter in Real-Time

- ***Fast, convenient, reliable and easy to use***
- ***Measures raw exhaust or from CVS***
- ***No dilution required***
- ***Measures soot (free of interference from other material)***
- ***Measures elemental carbon (EC) independent of condensed volatile or organic material (high selectivity)***
- ***Proprietary NIST Traceable Calibration method***
- ***Rugged system capable of extended operation without maintenance***
- ***Dynamic Range 1: 1,000,000***

New Generation LII 300 Instrument Features

- Completely enclosed laser, optics, and sampling cell
- Built-in computer and display, touchscreen control
- Built-in pneumatics controller and sampling system
- Includes real-time pressure and temperature measurements to reduce data to STP
- Fail safe valve prevents sample from entering cell if purge air or power are off
- Compact rugged and portable instrument
- Easy to use, low maintenance system
- Very high sensitivity
- Low operating costs