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Measurement of Particulate Matter Under Transient Conditions with Self-Calibrating Laser-Induced Incandescence

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Laser-induced incandescence (LII) is a powerful optical diagnostic for the study of PM which can be applied in any environment encountered as the exhaust gas travels from the combustion chamber to the atmosphere. In other words, neither cooling nor dilution are required, and measurements can be made either *in situ* or by continuous sampling. LII measures the volume concentration, active surface area, and primary particle (spherule) size for elementary carbon particles, and is insensitive to the presence of other species.

Artium Technologies, NRC, and Sandia are collaborating to develop, evaluate, and commercialize LII as a portable, safe, rugged, reliable instrument with automated, push-button simplicity for online exhaust PM monitoring.

Principle

Soot (elemental carbon based particles emitted from combustion sources) absorbs and emits light predominantly on the scale of the primary particles. Conventionally, a high-energy pulsed laser beam is used to rapidly heat the soot particles from the local ambient temperature to the carbon vaporization temperature (~ 4000 to 4500 K). The laser heating is independent of particle size, and the emitted light is nominally volumetric. Thermal emission, i.e., incandescence, from the particles is then recorded, using collection optics and photodetectors, as the particles slowly cool to the ambient temperature. Using appropriate calibration and analysis of the incandescence signal, the particle volume fraction and active surface area/primary particle size are estimated, where the former is obtained from the amplitude of the signal and the latter from the signals' rate of decay.

LII has a well-defined but complex response to volatile particulate matter. It is totally insensitive to liquid particles, because they absorb a negligible amount of laser energy compared to carbon. For carbon particles coated with volatile material, the latter will vaporize very early in the LII laser-heating period. In general, it is reasonable to state that LII measures the volume fraction of carbonaceous material in the exhaust. Other constituents may include trace metals and ash. Although trace metals may contribute to the incandescence if they survive the peak temperatures, there concentration is typically so low that the contribution will be negligible. Ash has a low absorptivity and emissivity relative to carbon, and is unlikely to contribute significantly to the incandescence, provided it has not evaporated.

Fundamental Research

A state-of-the-art numerical model of nanoscale (time and space) heat transfer to and from the particles has been developed to support understanding of the physical processes occurring during the LII event. This model has been enhanced to the point where it will well predict the time-resolved behavior of the LII signals for a range of laser fluences.

The common LII technique is inherently simple when the following assumptions apply: (1) soot primary particles are small compared to the laser wavelength (Rayleigh regime); (2) laser heating increases the temperature of all particles at the same rate, regardless of size; (3) when the particles reach the vaporization temperature, additional absorbed energy goes into vaporization rather than sensible energy, so that the particles remain at the same temperature for the duration of the laser heating period; (4) vaporization causes negligible particle-size reduction, so that the incandescent radiation from the particles is independent of laser fluence once above the vaporization threshold.

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, and thus extending the capabilities of LII for making practical quantitative measurements of soot. The use of the absolute intensity approach provides for continuous *in situ* self-calibration of the LII technique, and allows use of lower laser fluences and lower maximum soot temperatures. This low fluence approach simplifies interpretation of the data and lessens the burden of the assumptions noted above. Thus, issues associated with evaporating a significant portion of the soot are avoided. The absolute intensity method is a time-resolved approach that 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. This is shown to provide a stable measure of the soot concentration at every point during the signal decay (a period of ~ 1 μs). The primary particle size is determined from the time constant of temperature decay in conduction phase of the particle cooling.

Research on aggregate sizing with a combined LII/elastic scattering technique is under development and may provide size distributions over the range 1 nm – 1 μm. A potentially powerful use of scattering that has been demonstrated in flames is the Rayleigh-Debye-Gans polydisperse fractal aggregate (RDG/PFA) interpretation of scattered light. This analysis procedure provides quantitative measurements of particle aggregates, including fractal dimension, surface area, and aggregate size distribution. In applying this theory, the necessary particle volume fraction can be determined with LII, which also provides a redundant measure of primary particle size. Knowledge of the primary particle size and number provides insight about the aggregate morphology.

Measurement Capabilities

Quantitative LII is known to provide a sensitive, precise, and repeatable measure of the elemental carbon concentration over a wide measurement range. In the exhaust of a heavy-duty diesel engine, LII and gravimetric measurements have been demonstrated to correlate well over a wide range of operating conditions and particulate levels, while only LII was capable of providing real-time results due to the high speed data acquisition and analysis inherent to the technique.

The sensitivity of the LII technique is limited only by the size of the measurement volume. As an example, with a measurement volume of ~2 mm^3, reliable measurements of ~500 ppt (1 mg/m^3) were recorded in a diluted diesel exhaust. By increasing the dimensions of the probe volume, concentrations of <5 ppt (<0.010 mg/m^3) were measured in the undiluted exhaust of a gasoline automobile. In flames, concentrations as high as 10 ppm (20 g/m^3) have been measured.

Primary particle sizes have been measured over the range of 10 – 100 nm. The measurement of size and concentration are not limited by aggregate size, as only the primary particle size must be in the Rayleigh regime.

Applications

LII has become a widely used diagnostic for the investigation of soot in combustion systems ranging from fundamental burners to practical devices such as diesel and spark-ignition engines, and to study carbon black. Further applications may include elemental carbon atmospheric particulates and stack emissions from boilers, furnaces, and incinerators.

The measurement frequency is only limited by the repetition rate of a high-power pulsed laser (typically 10-30 Hz, which corresponds to one measurement per engine cycle at 1200-3600 rpm). While it thus is not possible to obtain crank-angle resolution in real-time, ensemble-averaging for many engine cycles can be used to reconstruct cycle-resolved transient behavior. However, this measurement frequency is more than adequate to observe engine and vehicle transients, such as those that occur in driving cycles.

Particulate measurements in the exhaust of a gasoline-powered vehicle demonstrated that maximum emission rates occur during acceleration transients. Steady–state driving and deceleration produced much lower levels of particulates. In comparison to concurrent ELPI measurements, 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.

Results are also shown for measurements acquired in the exhaust of a light duty diesel engine operated over a variety of engine transient conditions, in which LII is shown to provide greater temporal response than concurrent single size bin SMPS measurements.

The LII technique is capable of real-time soot measurements over all vehicle transient operations, making it a valuable tool in tuning gasoline and diesel engine soot emissions performance.

Summary

LII is a potential method for online monitoring of particulate emissions, with very high precision (repeatability), a large measurement range and high sensitivity, even at low particulate concentrations, and rapid data acquisition and analysis. Measurements can be made with or without dilution the latter avoiding sampling issues. A practical and effective calibration method for measuring soot concentration and active surface area has been successfully developed.

LII is applicable to a wide range of applications. Comparisons to gravimetric methods demonstrate the accuracy of the LII method. LII has superior time response and capability in measuring transient particle emissions in comparison to SMPS and ELPI

LII combined with other methods may best fulfill future monitoring requirements.

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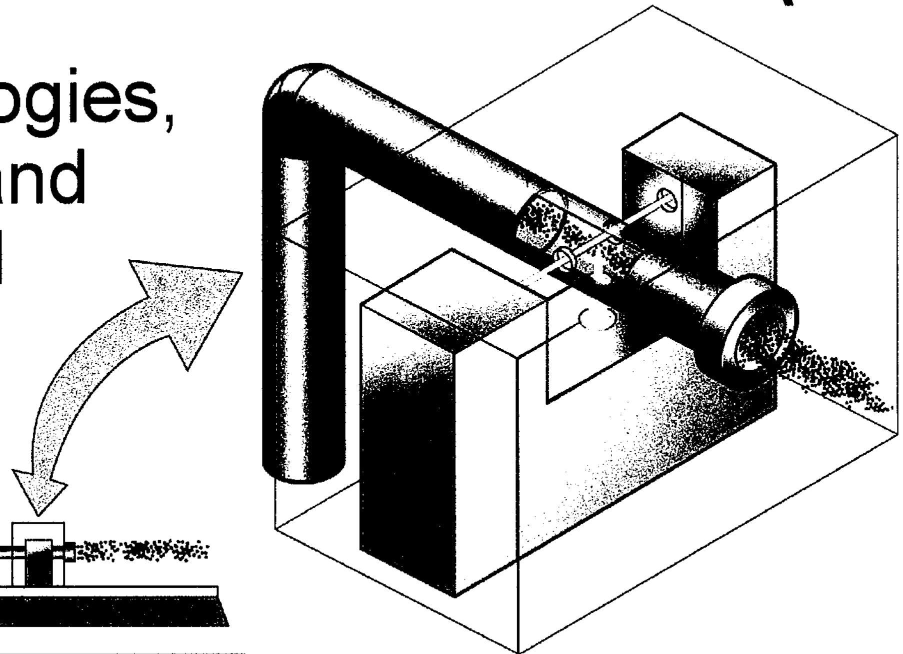
Outline

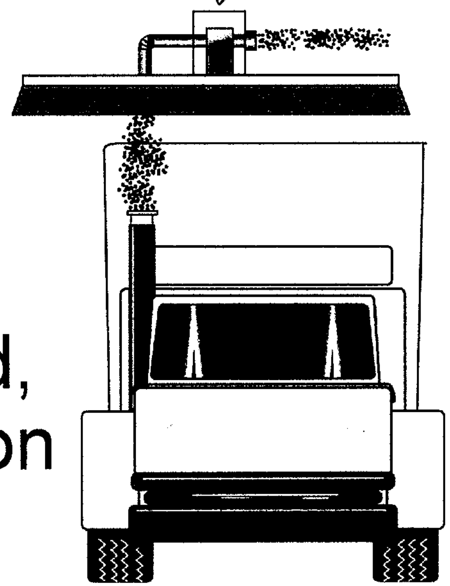
- Introduction to LII
- LII Method
  - *in situ* continuous self-calibration
  - concentration and sizing
- Applications
  - heavy duty diesel
  - light duty diesel
  - gasoline direct injection
- Summary

Laser Induced Incandescence (LII)

- Artium Technologies, NRC Canada, and Sandia National Laboratories



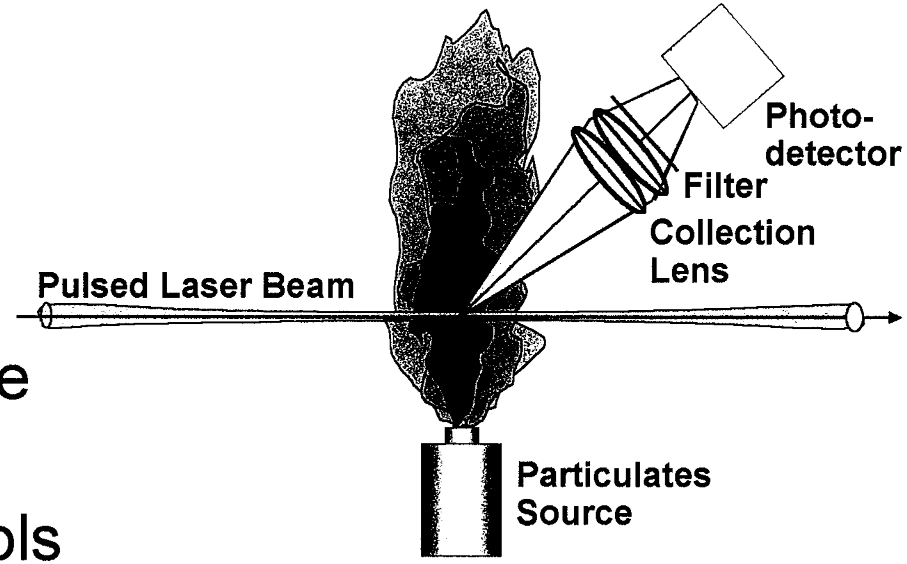
- portable
- safe, rugged, reliable
- automated, push-button simplicity



- collaborating to develop, evaluate, and commercialize LII for online exhaust PM monitoring

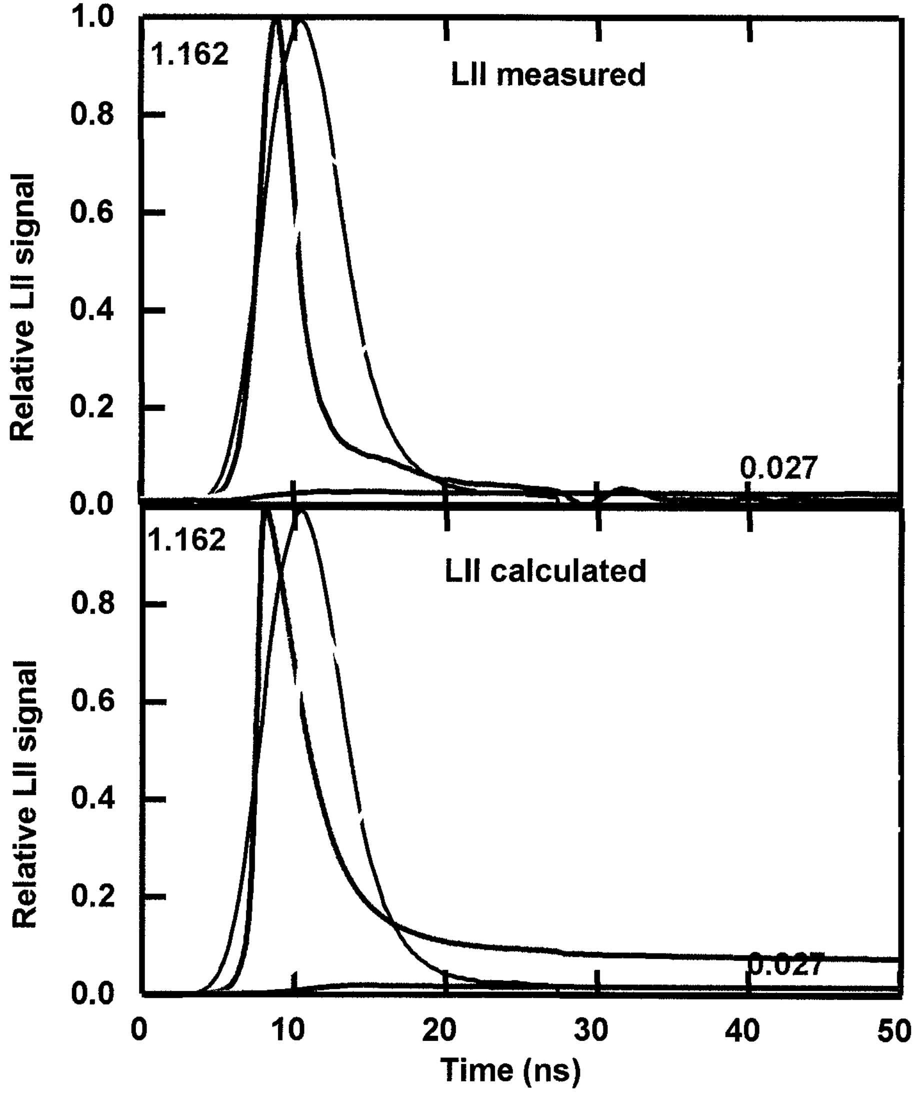
LII Concepts

- LII experiment:
  - pulsed laser beam
  - rapid heating of soot to evaporation temperature
  - soot radiates incandescence as it cools to ambient temperature
  - incandescence signal is collected to determine soot concentration and primary particle (spherule) size



- LII theory
  - a state-of-the-art numerical model of nanoscale (time and space) heat transfer to and from the particles

LII Theory vs. Experiment



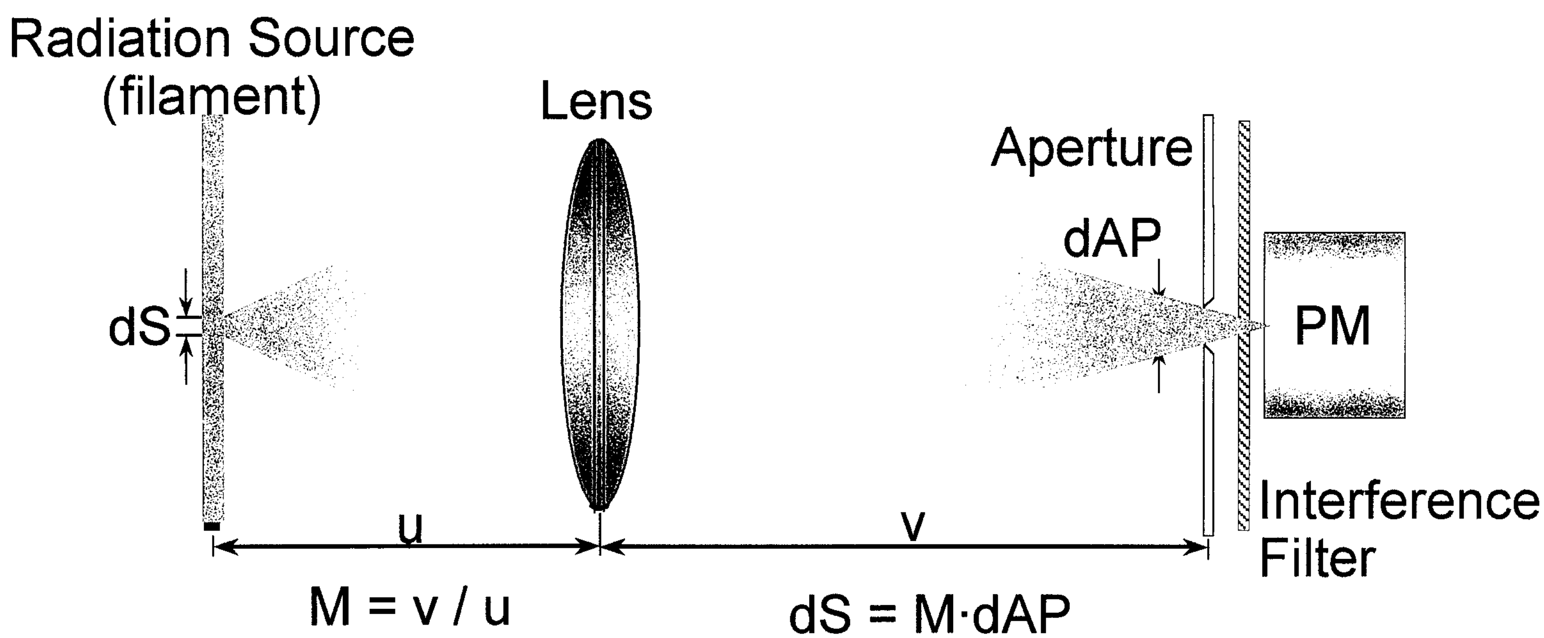
LII Features

- *in situ* and nonintrusive
- signal is proportional to soot volume fraction
- spatially resolved
- time resolved
- large measurement range
  - not limited by aggregate size
- high precision and repeatability
- high speed data acquisition and analysis

LII Benefits

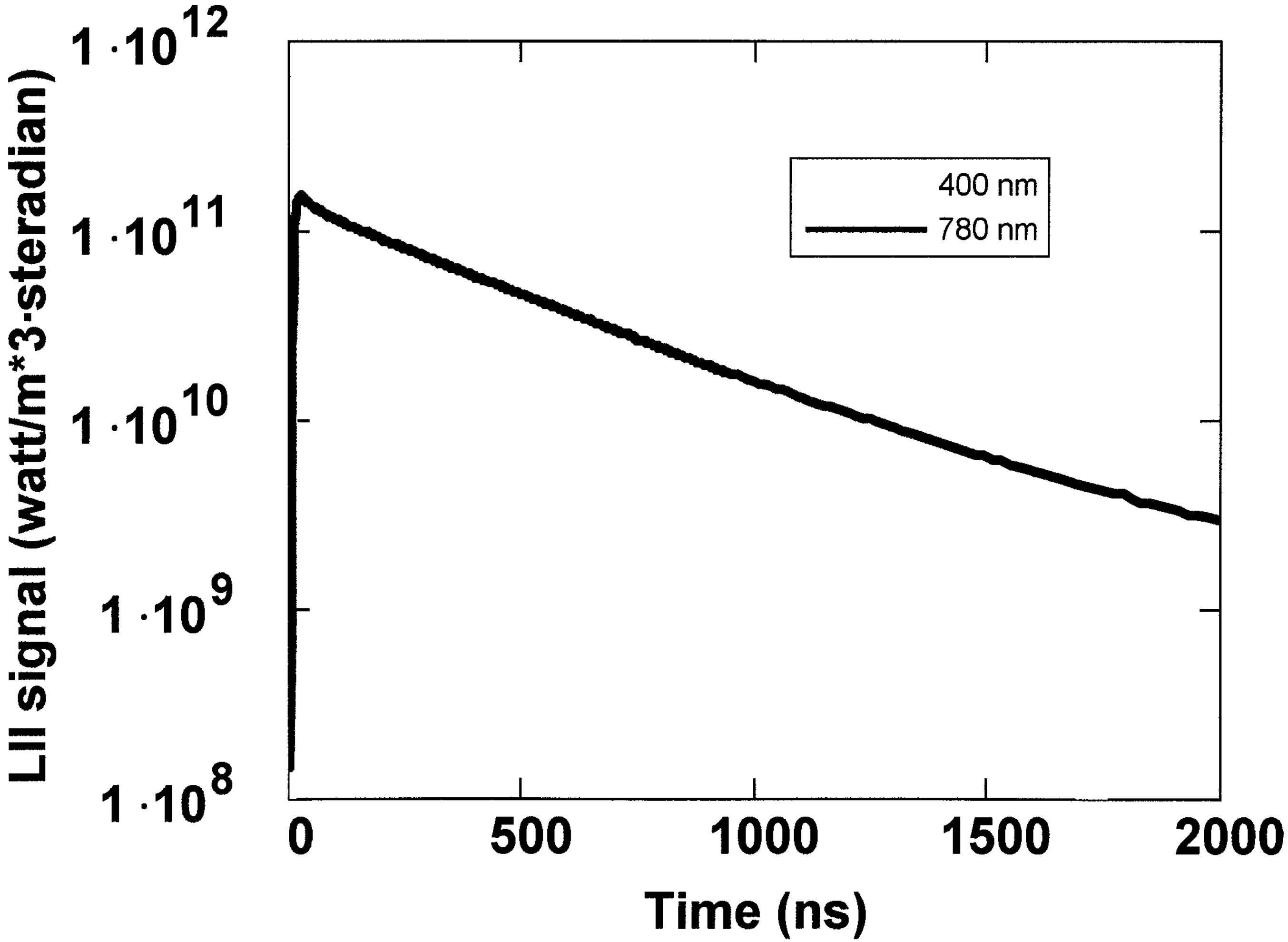
- dilution of sample not required
- stable measurement of elemental carbon
- insensitive to presence of other species
- can operate at very low concentrations
- real-time results
- cycle-resolved measurements possible
- can provide particulate morphology (size, size distribution, number density) when combined with scattering

In-Situ Absolute Intensity Continuous Self-Calibration Concept

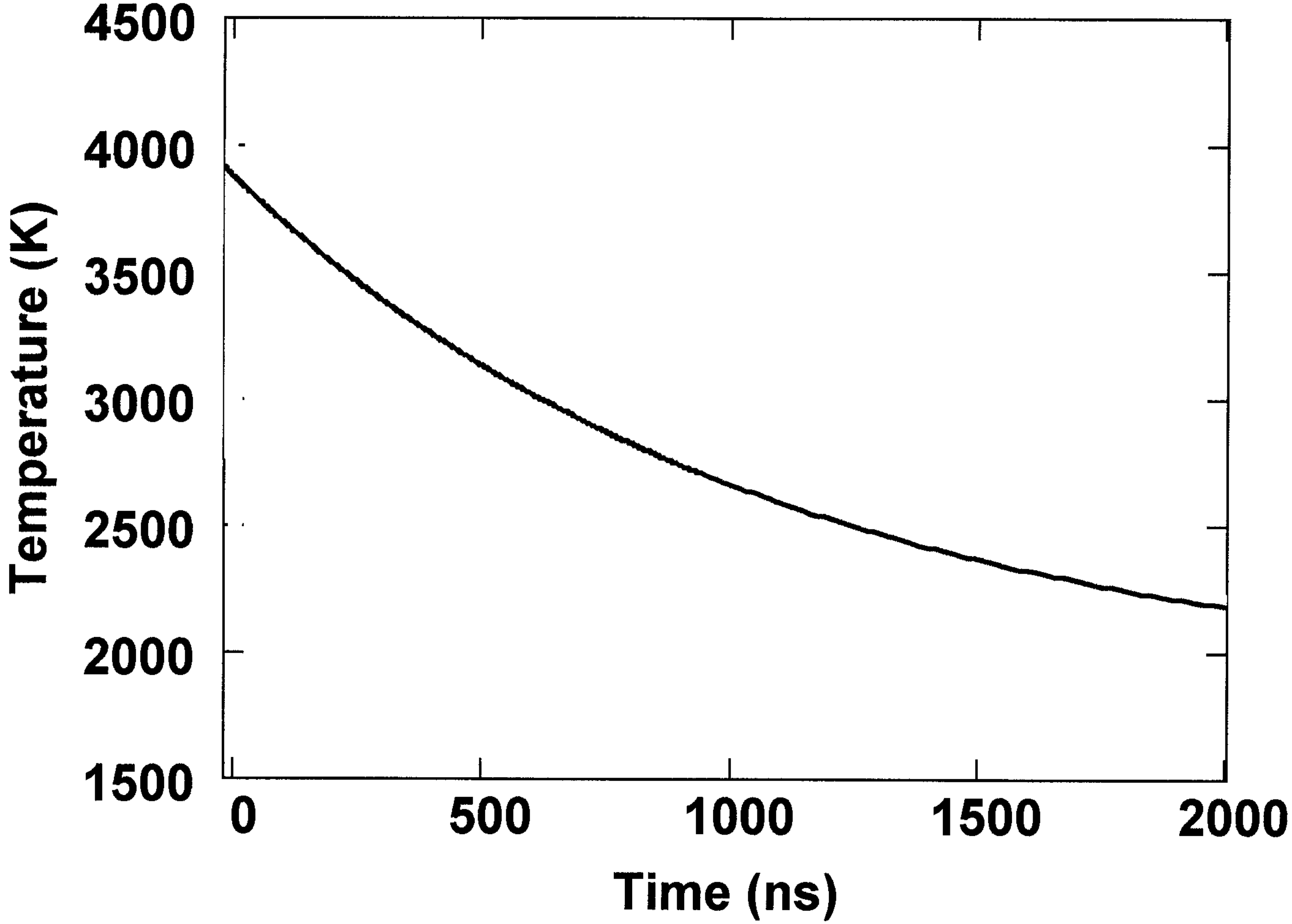


- two-color pyrometry
- use the temperature to determine primary particle radiation

Laminar Diffusion Flame

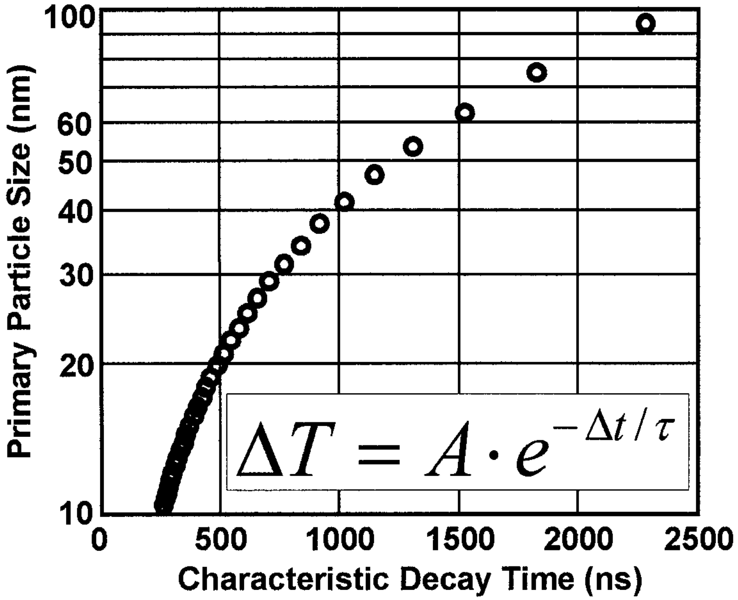


Laminar Diffusion Flame – Low Fluence



Particulate Concentration and Size

- concentration is determined from knowledge of absolute intensity of radiation and particle temperature
- concentration determined at every point during signal decay

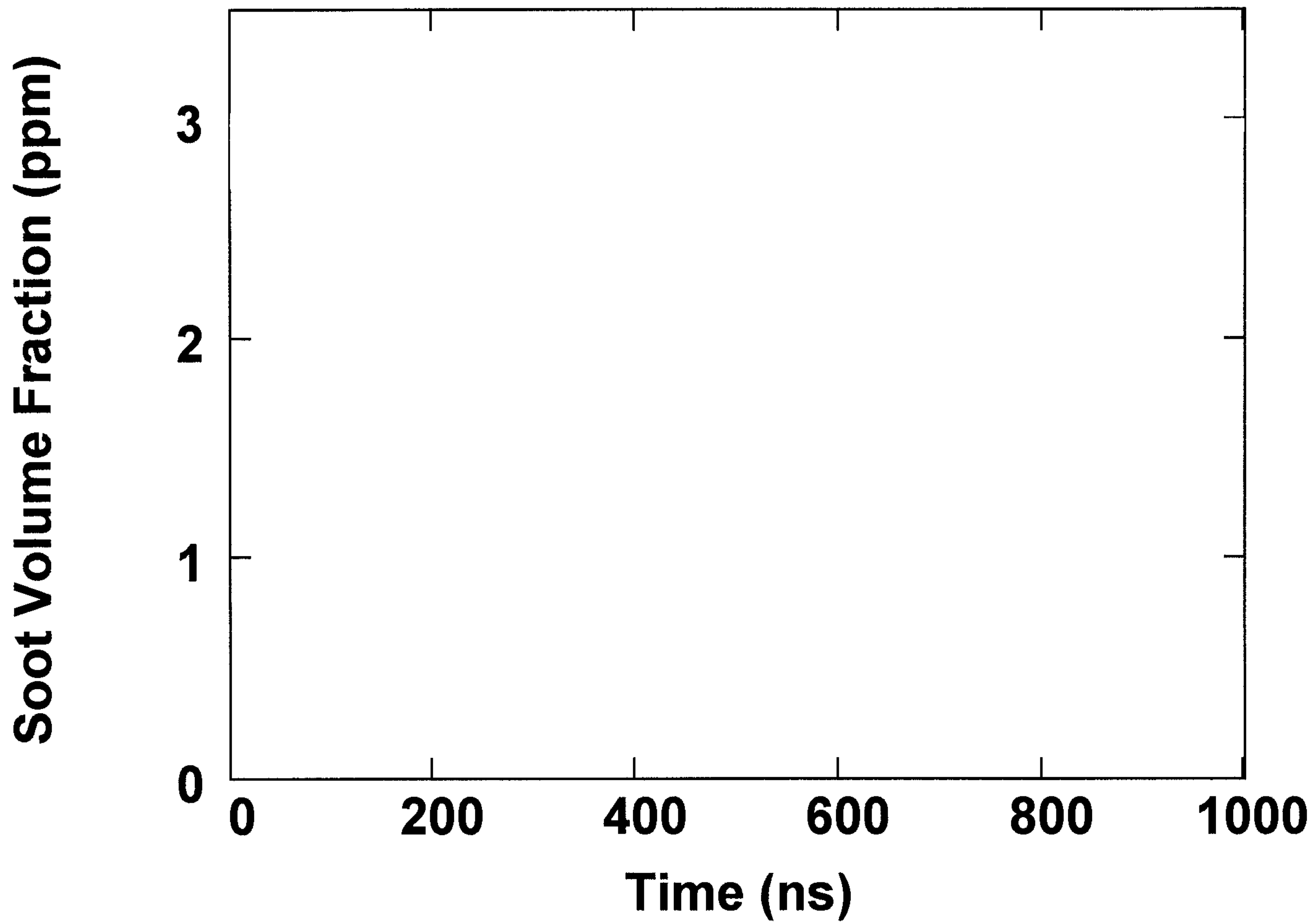


$$\Delta T = A \cdot e^{-\Delta t / \tau}$$

- primary particle size determined from time constant of temperature decay in conduction phase

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

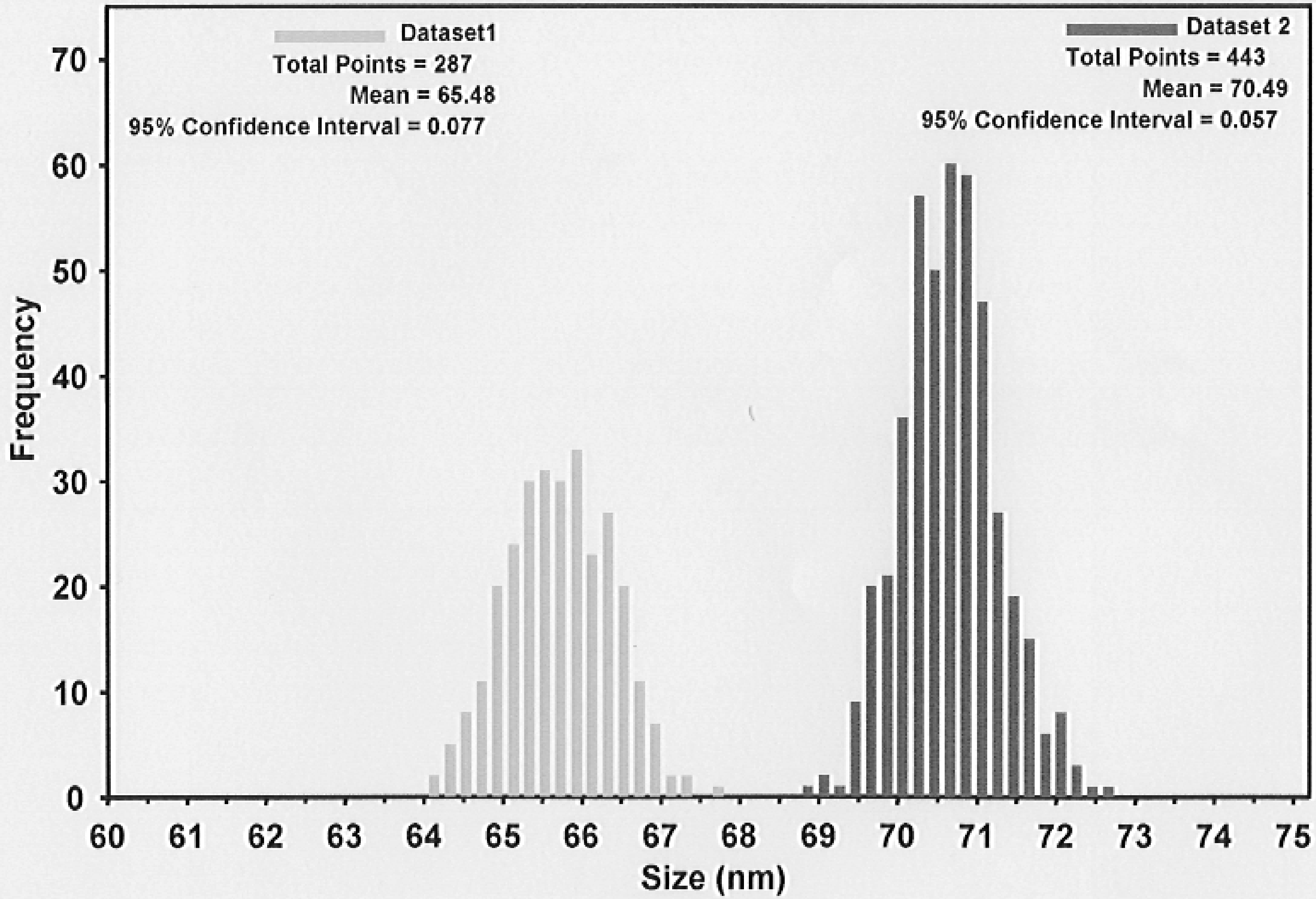
Laminar Diffusion Flame – Low Fluence



Primary Particle Size: Discussion

- the number of primary particles is also determined
- primary particle size can also be used to determine active surface area
- aggregate size is of greater interest from the health, environment, and regulation perspectives
- knowledge of primary particle size and number provides insight about aggregate morphology

Primary Particle Sizing Precision

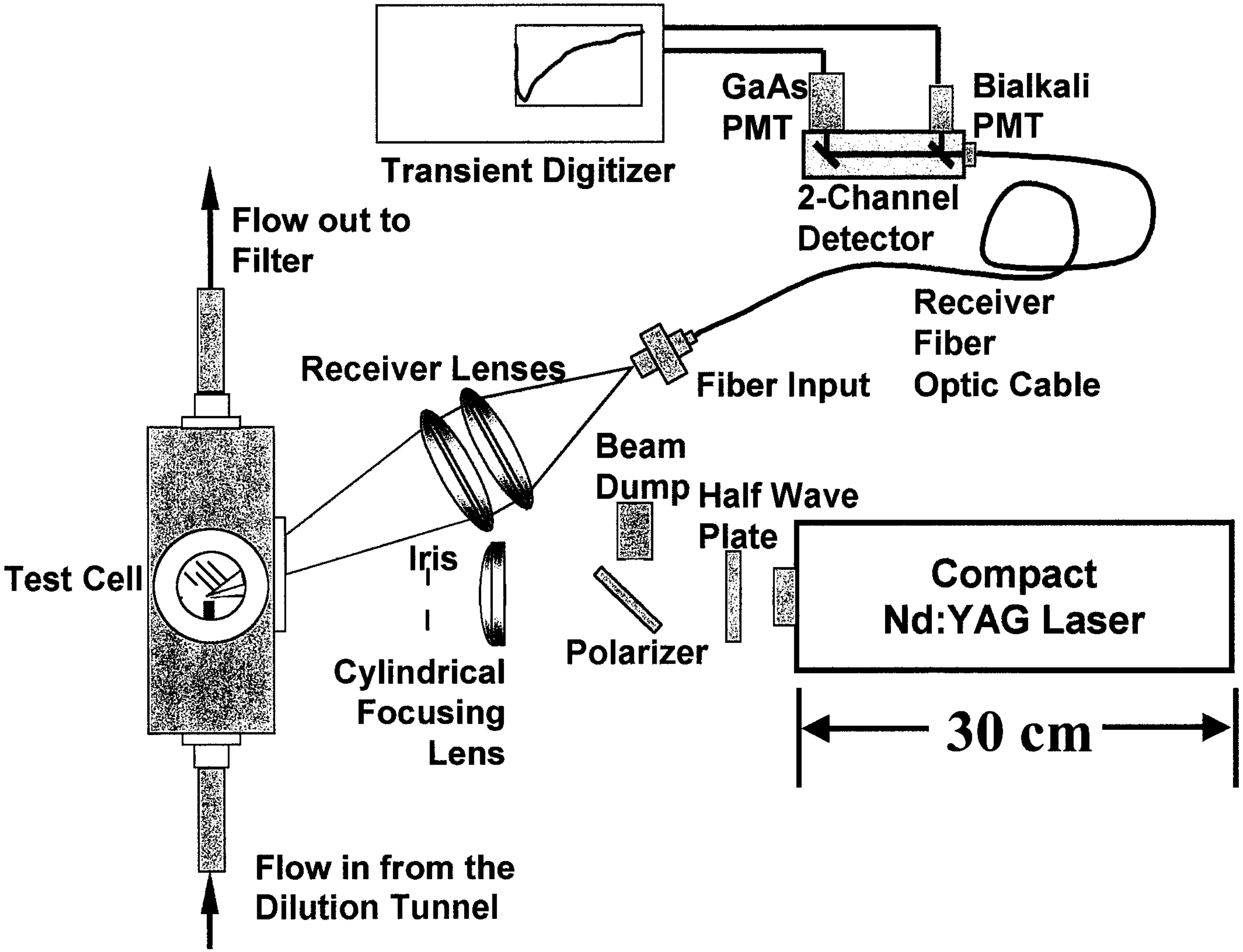


Sandia National Laboratories NRC · CNRC Artium Technologies Inc.

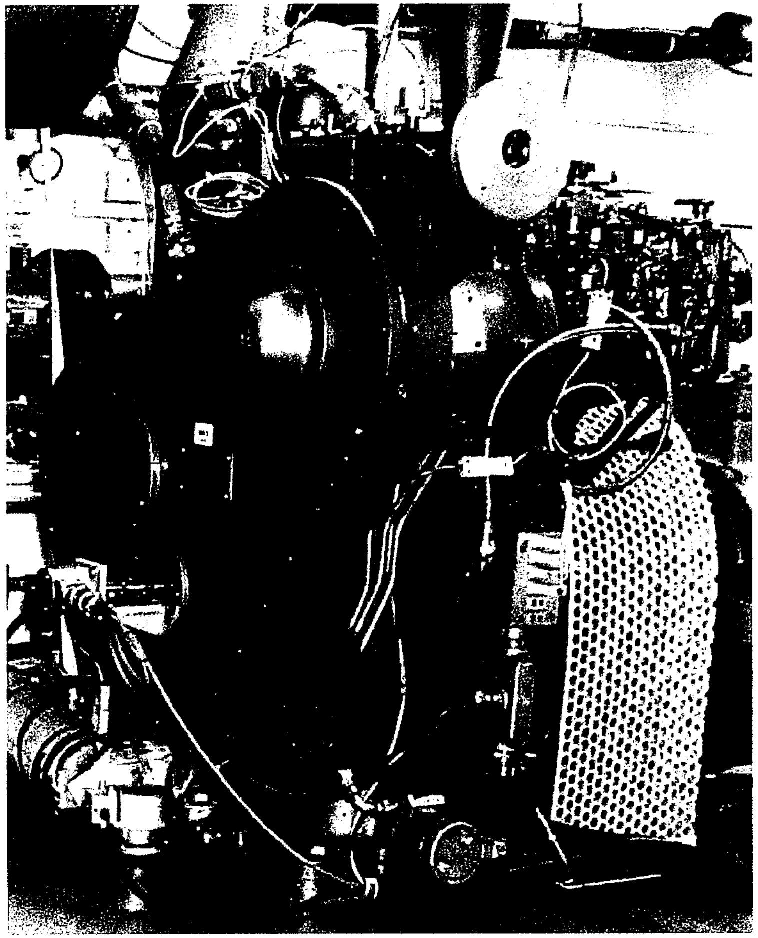
LII Applications

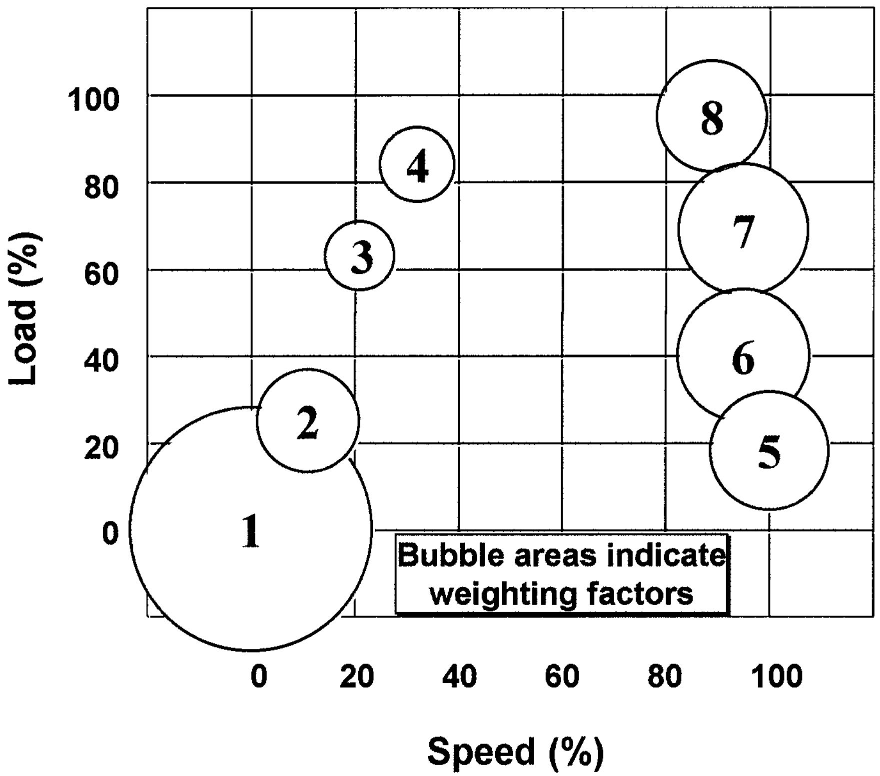
- Diesel and spark ignition engines
  - engine development
  - roadside particulates measurements
- gas turbine engines
- stack emissions from boilers, furnaces, and incinerators
- process control – carbon black
- elemental carbon atmospheric particulates

LII Apparatus Schematic (HD Diesel)



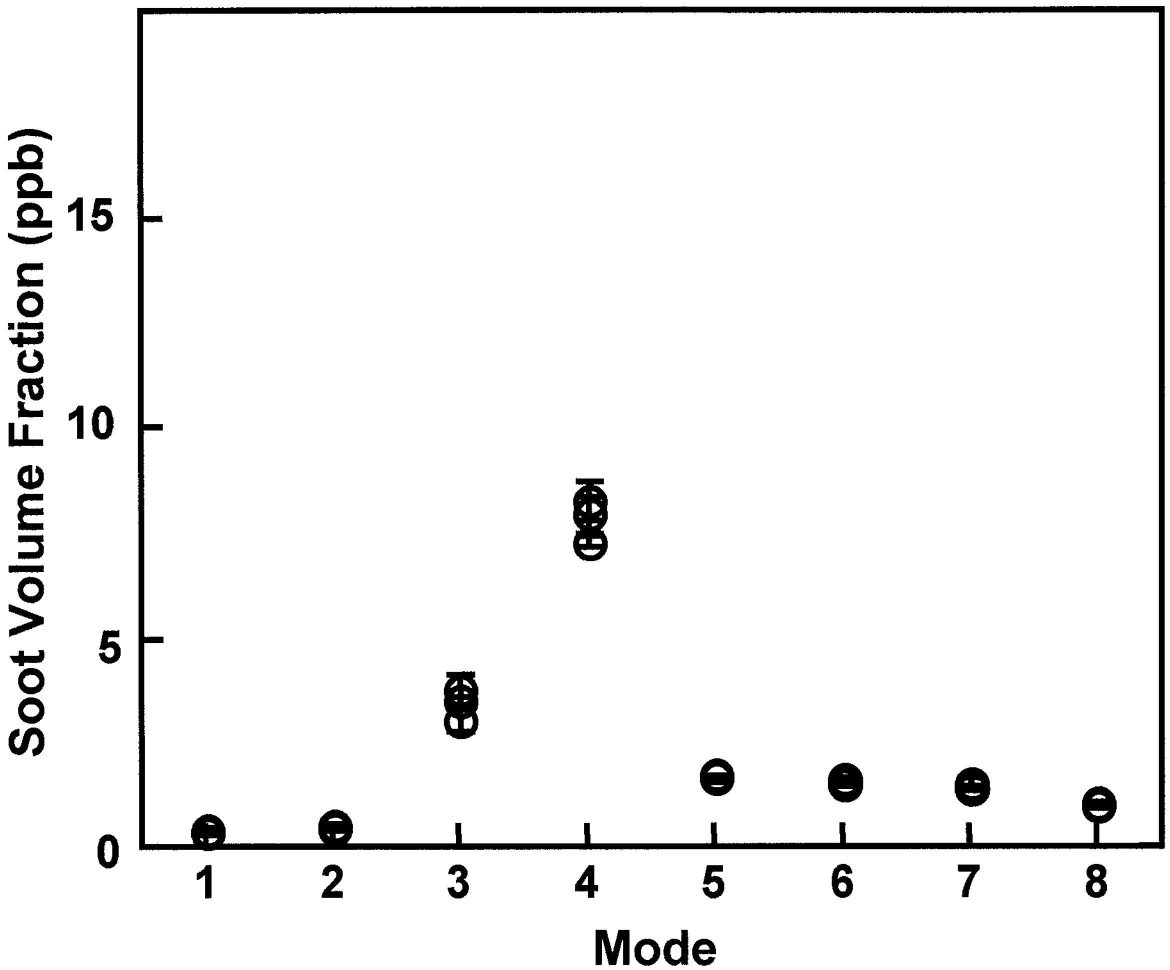
Heavy Duty Diesel Engine



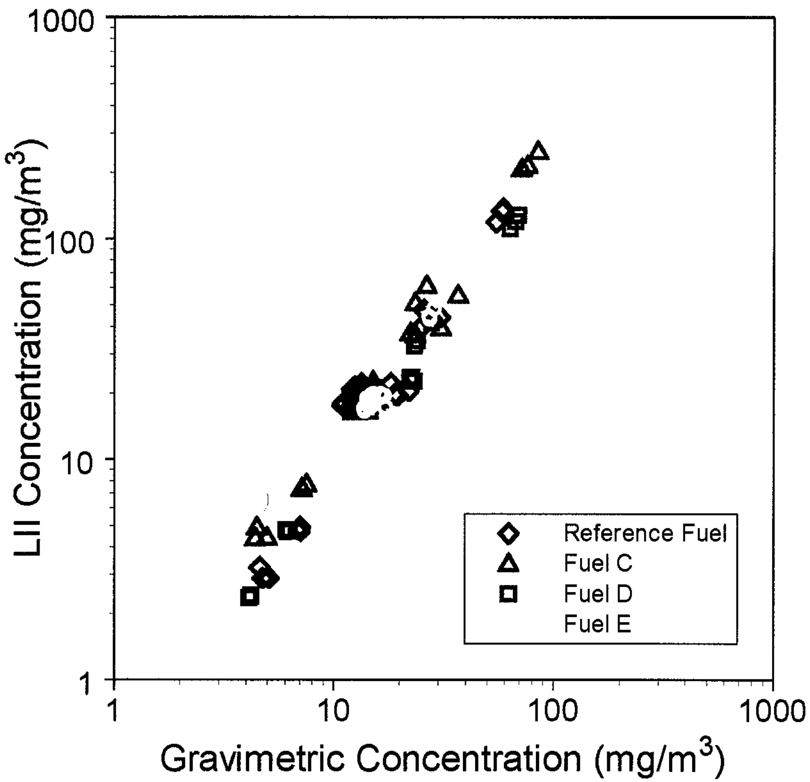


AVL 8-Mode Steady-State Simulation

HD Diesel Soot Concentration



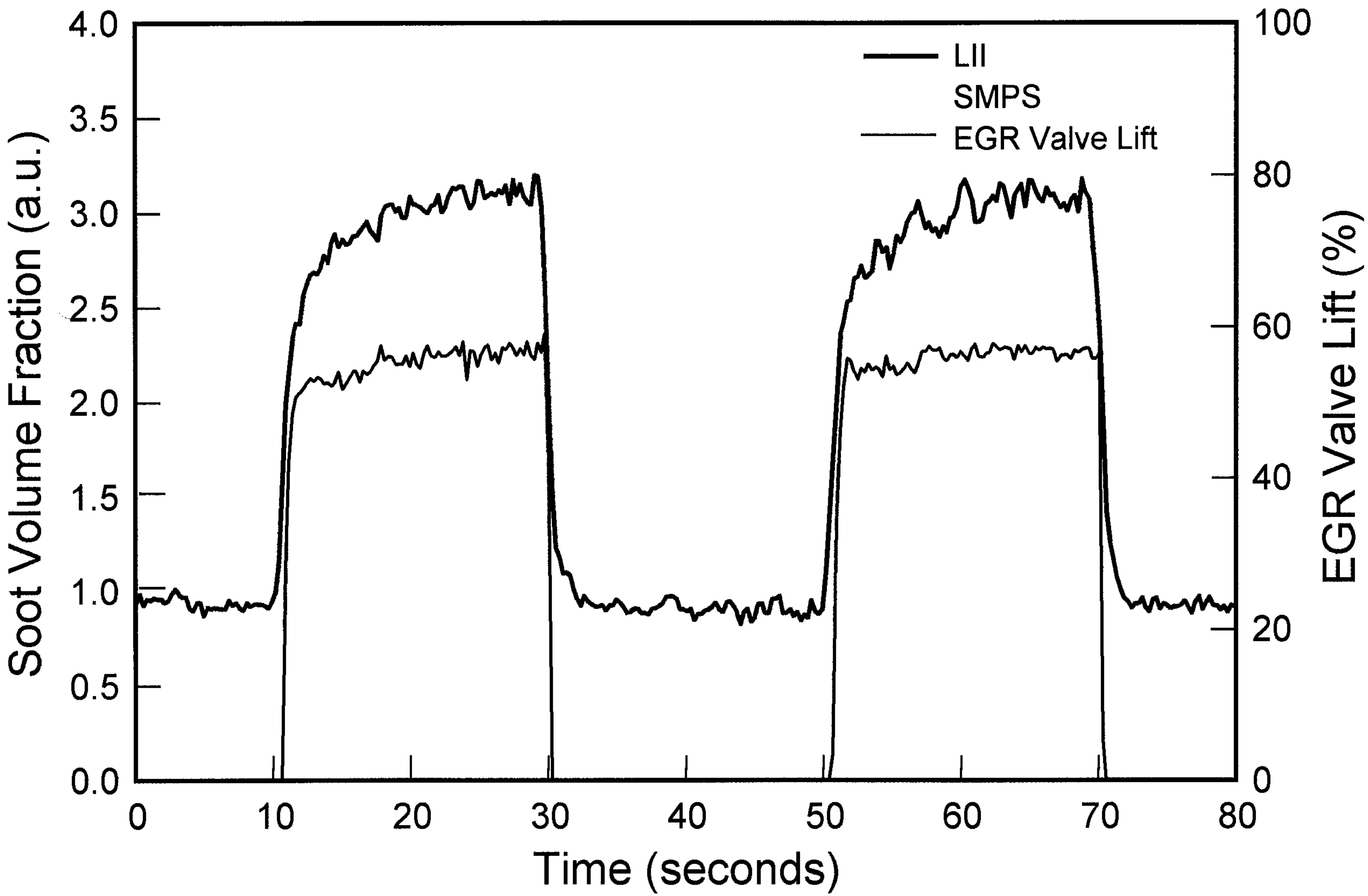
HD Diesel: LII vs. Gravimetric



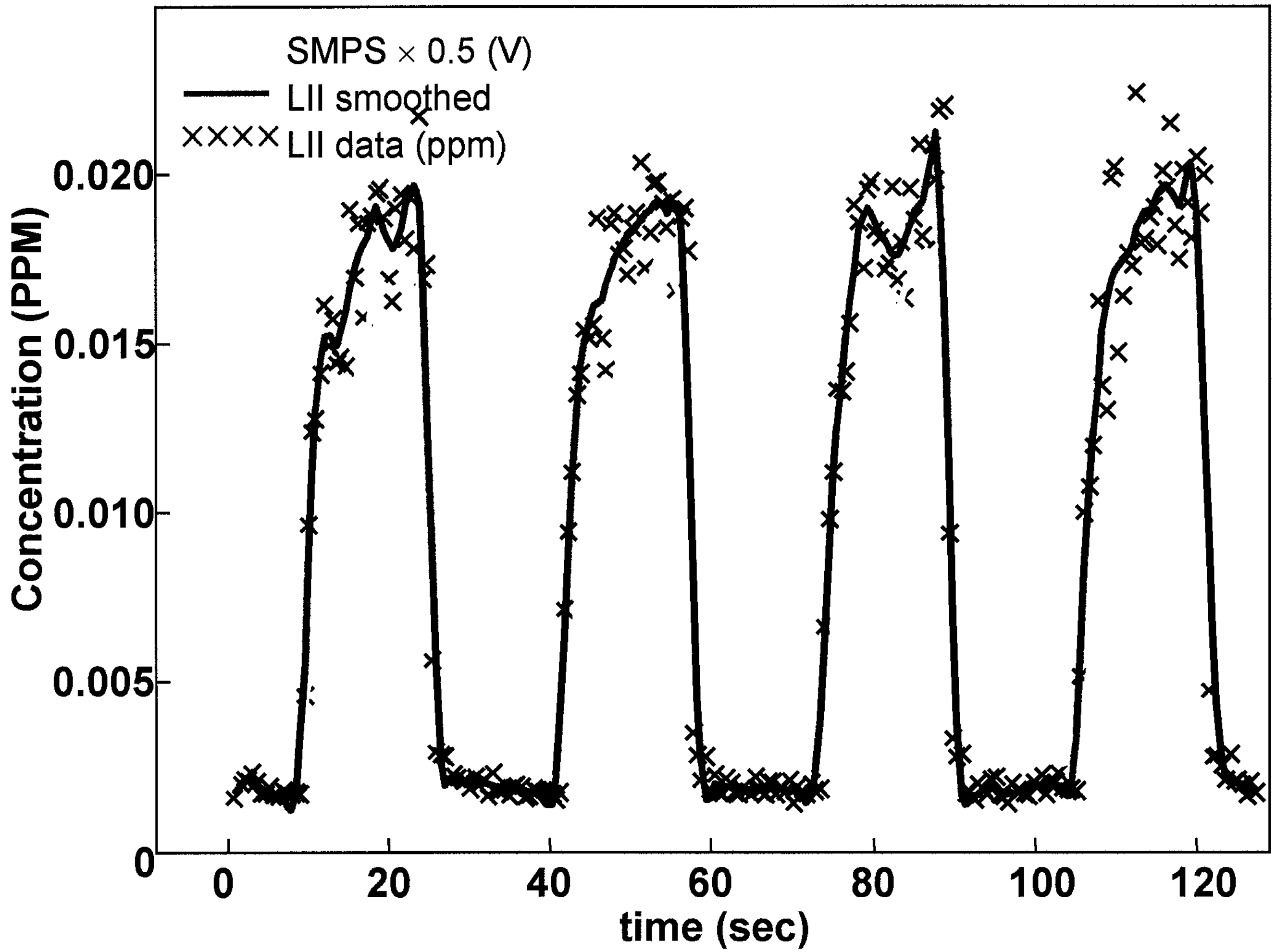
- LII measurements show good correlation with the filter mass measurements over a wide range of operating conditions and particulate levels
- the repeatability of LII measurements is far better than the repeatability of the standard gravimetric filter technique

LD Diesel LII System

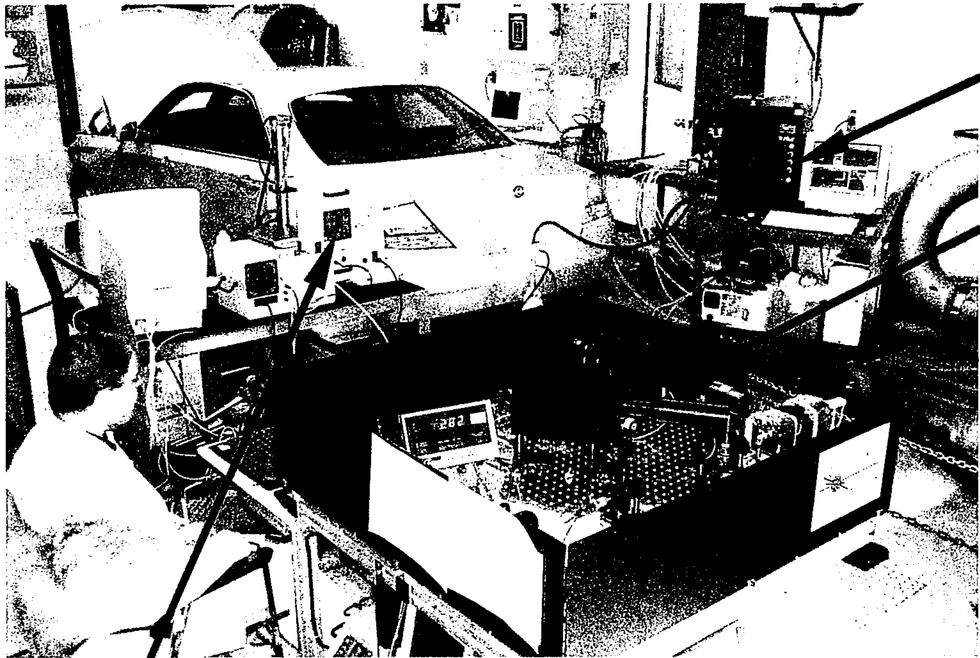
LD Diesel EGR transient

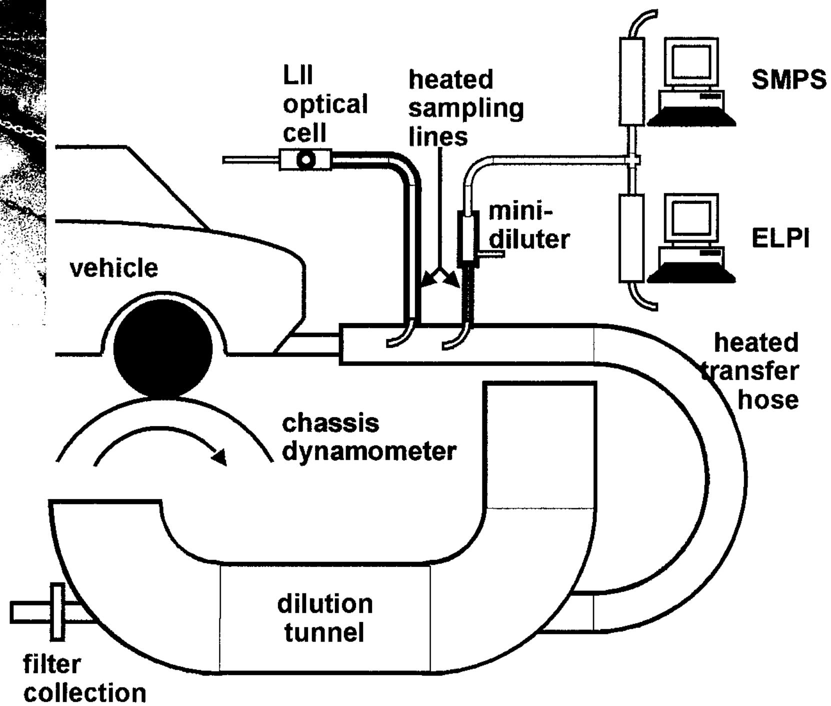


LD Diesel EGR Transient: LII vs. 120 nm SMPS

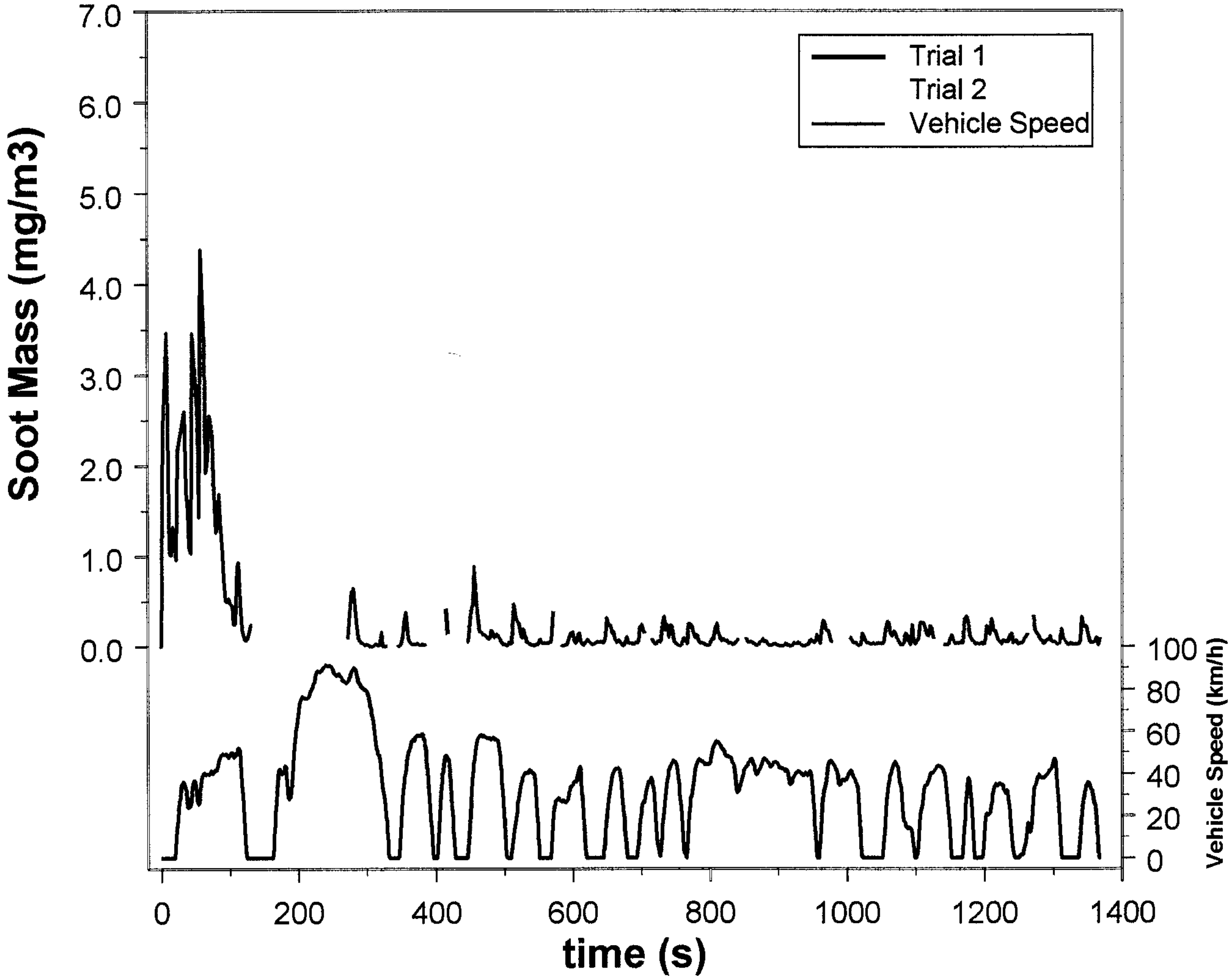


DISI Tailpipe Particulates Measurements

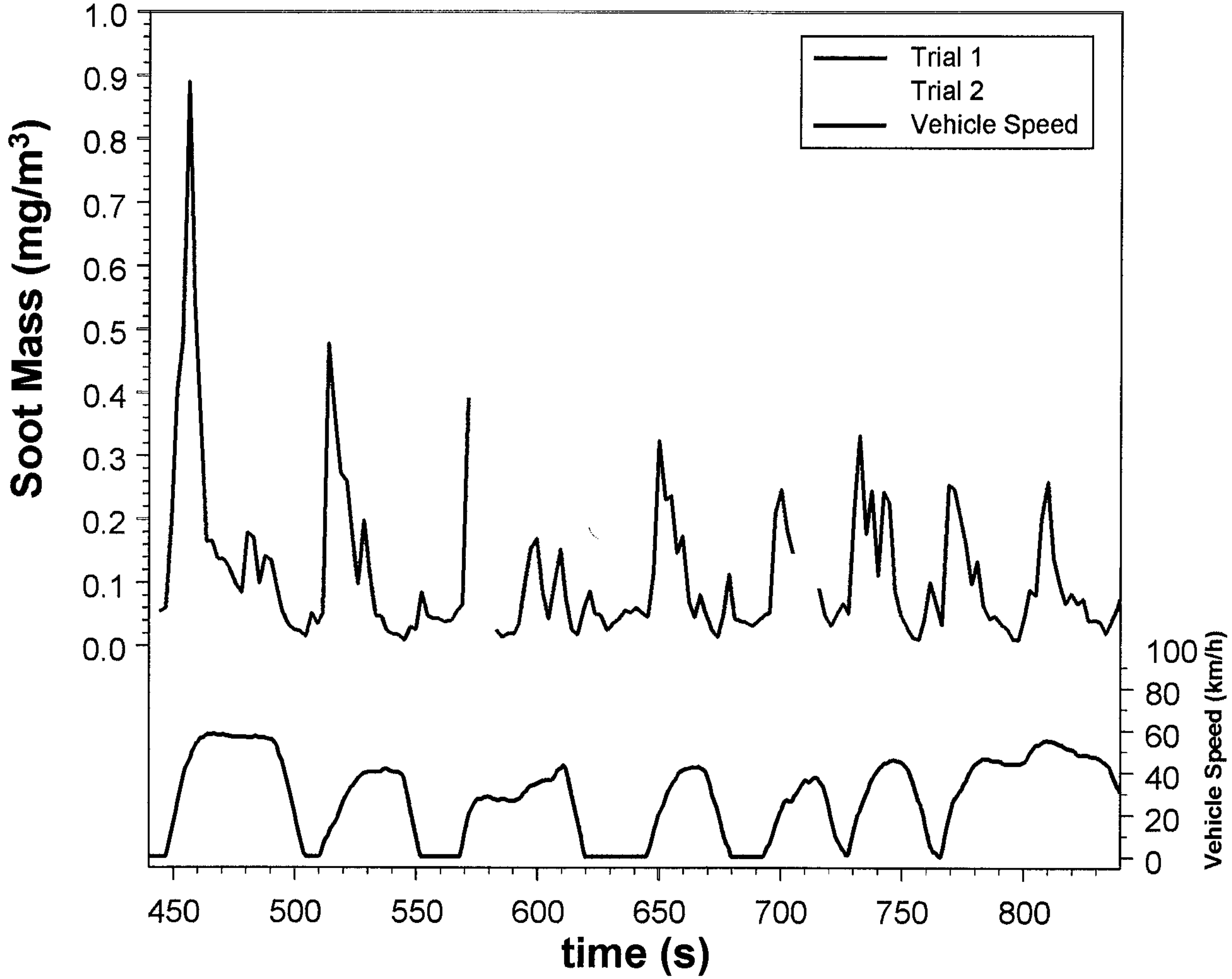




DISI LII LA4 Cold Start Cycle



DISI LII LA4 Cold Start Cycle - Detail



Summary

- LII is a potential method for online monitoring of particulate emissions
- method has very high precision (repeatability)
- shown to have a large measurement range and sensitivity even at low particulate concentrations

Application	Measured SVF Range
Laminar Diffusion Flame	100 ppb – 10 ppm
Carbon Black	10 ppb – 500 ppb
Diesel Exhaust	500 ppt – 100 ppb
DISI Vehicle	5 ppt – 500 ppt

- measurements can be made without dilution – avoiding sampling issues

Summary

- practical and effective calibration method for measuring soot concentration successfully developed
- effective for a wide range of applications
- comparisons to gravimetric methods show the accuracy of the LII method
- superior time response and capability in measuring transient particle emissions in comparison to SMPS and ELPI
- rapid data acquisition and analysis
- LII combined with other methods may best fulfill future monitoring requirements

For further information, please consult our publications and references therein:

1. Smallwood, G. J., Snelling, D. R., Gülder, Ö. L., Clavel, D., Gareau, D., Sawchuk, R. A., and Graham, L., "Transient Particulate Matter Measurements from the Exhaust of a Direct Injection Spark Ignition Automobile," SAE Paper No. 2001-01-3581, 2001.
2. Axelsson, B., and Witze, P. O., "Qualitative Laser-Induced Incandescence Measurements of Particulate Emissions During Transient Operation of a TDI Diesel Engine," SAE Paper No. 2001-01-3574, 2001.
3. Smallwood, G. J., Snelling, D. R., Liu, F. and Gülder, Ö. L., "Clouds over Soot Evaporation: Errors in Modeling Laser-Induced Incandescence of Soot," Journal of Heat Transfer, 123, pp. 814-818, 2001.
4. Witze, P. O., "Qualitative Laser-Induced Incandescence Measurements of Soot Emissions During Transient Operation of a Port Fuel-Injected Engine," The Fifth International Symposium on Diagnostics and Modeling of Combustion in Internal Combustion Engines (COMODIA), Nagoya, 2001.
5. Smallwood, G. J., Snelling, D. R., Neill, W. S., Liu, F., Bachalo, W. D., and Gülder, Ö. L., "Laser-Induced Incandescence Measurements of Particulate Matter Emissions in the Exhaust of a Diesel Engine," The Fifth International Symposium on Diagnostics and Modeling of Combustion in Internal Combustion Engines (COMODIA), Nagoya, 2001.
6. Snelling, D. R., Smallwood, G. J. Gülder, Ö. L., Liu, F., and Bachalo, W. D., "A Calibration-Independent Technique of Measuring Soot by Laser-Induced Incandescence Using Absolute Light Intensity," The Second Joint Meeting of the US Sections of the Combustion Institute, Oakland, CA, 2001.
7. Witze, P. O., Hochgreb, S., Kayes, D., Michelsen, H. A. and Shaddix, C. R., "Time-Resolved Laser-Induced Incandescence and Laser Elastic Scattering Measurements in a Propane Diffusion Flame," Applied Optics, 40, pp.2443-2452, 2001.
8. Snelling, D. R., Smallwood, G. J. and Gülder, Ö. L.,"Method and Apparatus for Applying Laser Induced Incandescence for the Determination of Particle Measurements," US Patent No. 6,181,419, 2001.
9. Snelling, D. R., Smallwood, G. J. and Gülder, Ö. L., "Absolute Light Intensity Measurements in Laser Induced Incandescence," US Patent No. 6,154,277, 2000.
10. Snelling, D. R., Liu, F., Smallwood, G. J. and Gülder, Ö. L., "Evaluation of the Nanoscale Heat and Mass Transfer Model of the Laser-Induced Incandescence: Prediction of the Excitation Intensity," NHTC2000-12132, Thirty Fourth National Heat Transfer Conference, Pittsburgh, PA, 2000.
11. Green, R. M. and Witze, P. O., "Laser-Induced Incandescence and Elastic-Scattering Measurements of Particulate-Matter Volume Fraction Changes During Passage Through a Dilution Tunnel," Proceedings of the 10th International Symposium on Applications of Laser Techniques to Fluid Mechanics, Lisbon, 2000.
12. Snelling, D. R., Smallwood, G. J., Gülder, Ö. L., Bachalo, W. D., and Sankar, S., "Soot Volume Fraction Characterization Using the Laser-Induced Incandescence Detection Method," Proceedings of the 10th International Symposium on Applications of Laser Techniques to Fluid Mechanics, Lisbon, 2000.
13. Snelling, D. R., Smallwood, G. J., Sawchuk, R. A., Neill, W. S., Gareau, D., Clavel, D., Chippior, W. L., Liu, F., Gülder, Ö. L. and Bachalo, W. D., "*In-Situ* Real-Time Characterization of Particulate Emissions from a Diesel Engine Exhaust by Laser-Induced Incandescence," SAE Paper No. 2000-01-1994, 2000.
14. Snelling, D. R., Smallwood, G. J., Sawchuk, R. A., Neill, W. S., Gareau, D., Chippior, W. L., Liu, F., Gülder, Ö. L., and Bachalo, W. D., "Particulate Matter Measurements in a Diesel Engine Exhaust by Laser-Induced Incandescence and the Standard Gravimetric Procedure," SAE Paper No. 1999-01-3653, 1999.
15. Snelling, D. R., Smallwood, G. J., Campbell, I. G., Medlock, J. E., and Gülder, Ö. L., "Development and Application of Laser Induced Incandescence (LII) as a Diagnostic for Soot Particulate Measurements," Proceedings of the NATO/AGARD Propulsion and Energetics Panel, 90th Symposium on Advanced Non-Intrusive Instrumentation for Propulsion Engines, Brussels, Belgium, 1997.