Fundamentals and applications of laser-induced incandescence

Smallwood, Gregory J.

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Fundamentals and Applications of Laser-Induced Incandescence

Gregory J. Smallwood
Institute for Chemical Process and Environmental Technology
National Research Council Canada
Ottawa, ON, Canada K1A 0R6

1st Asian Workshop on Laser-Induced Incandescence
30 October 2009 Hefei, China

National Research Council Canada | Conseil national de recherches Canada

Canada

Outline

- Background
- Laser-Induced Incandescence (LII)
  - Basics
  - Autocompensating LII
- Applications
- Summary

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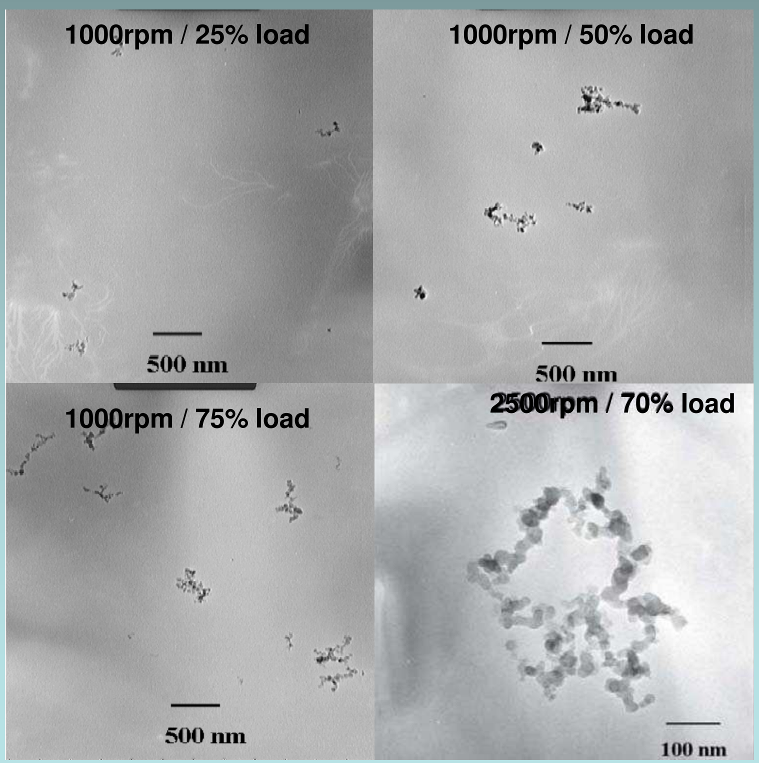
Why LII?

What is soot?

- dry solid particles produced through incomplete combustion of hydrocarbon fuels
- terminology varies by scientific field
  - elemental carbon, black carbon, refractory carbon, carbon black
- LII is effective at measuring all of these

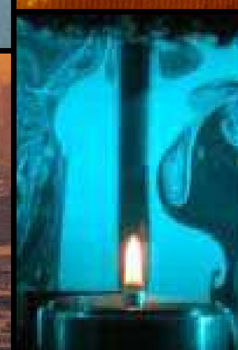


[Lee al., SAE Paper No. 2003-01-3169, 2003]

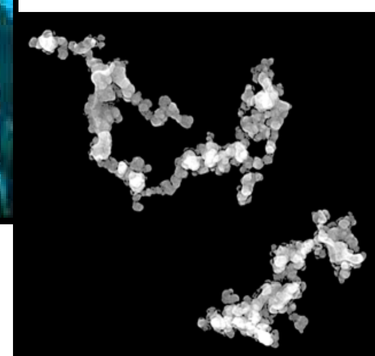
- there was a need for substantially improved instruments to quantify nanoparticle characteristics
- laser-induced incandescence technique for the quantitative measurement of soot nanoparticles
  - concentration, active surface area, and primary particle diameter
  - species selective technique
  - good sensitivity
- enhance the state of measurements for practical applications
  - nonvolatile particulate matter emissions

What are the issues with soot?

Improve efficiency of combustion technology



Engineered nanoparticle synthesis



Assess the ecological and health impacts of combustion

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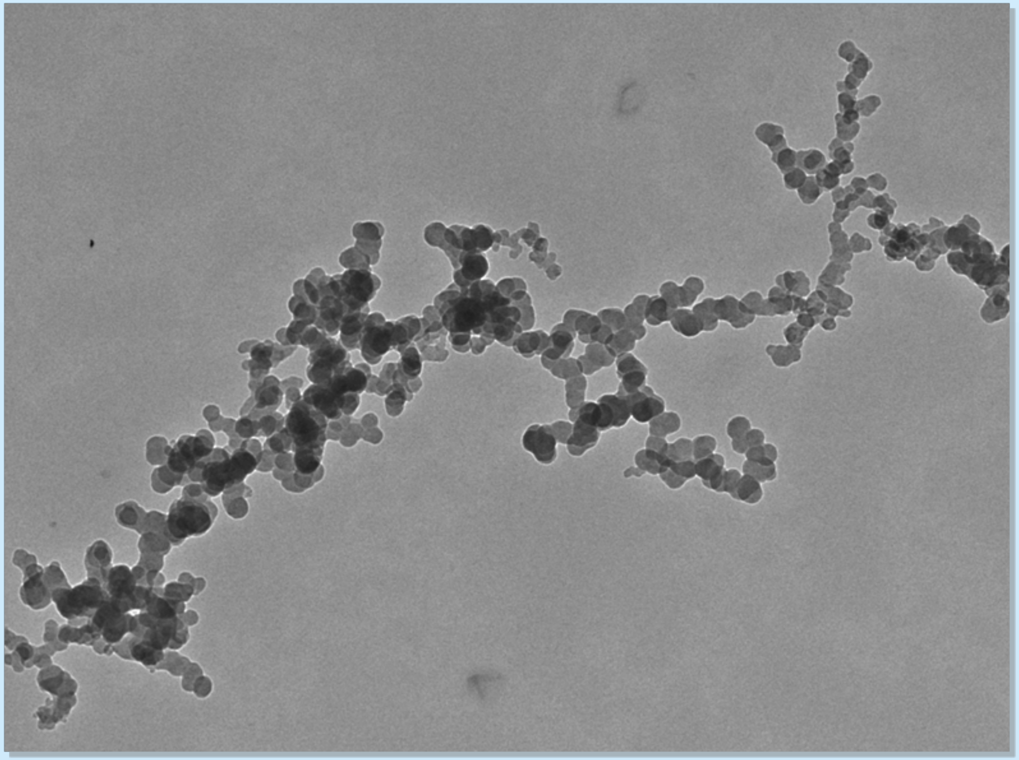
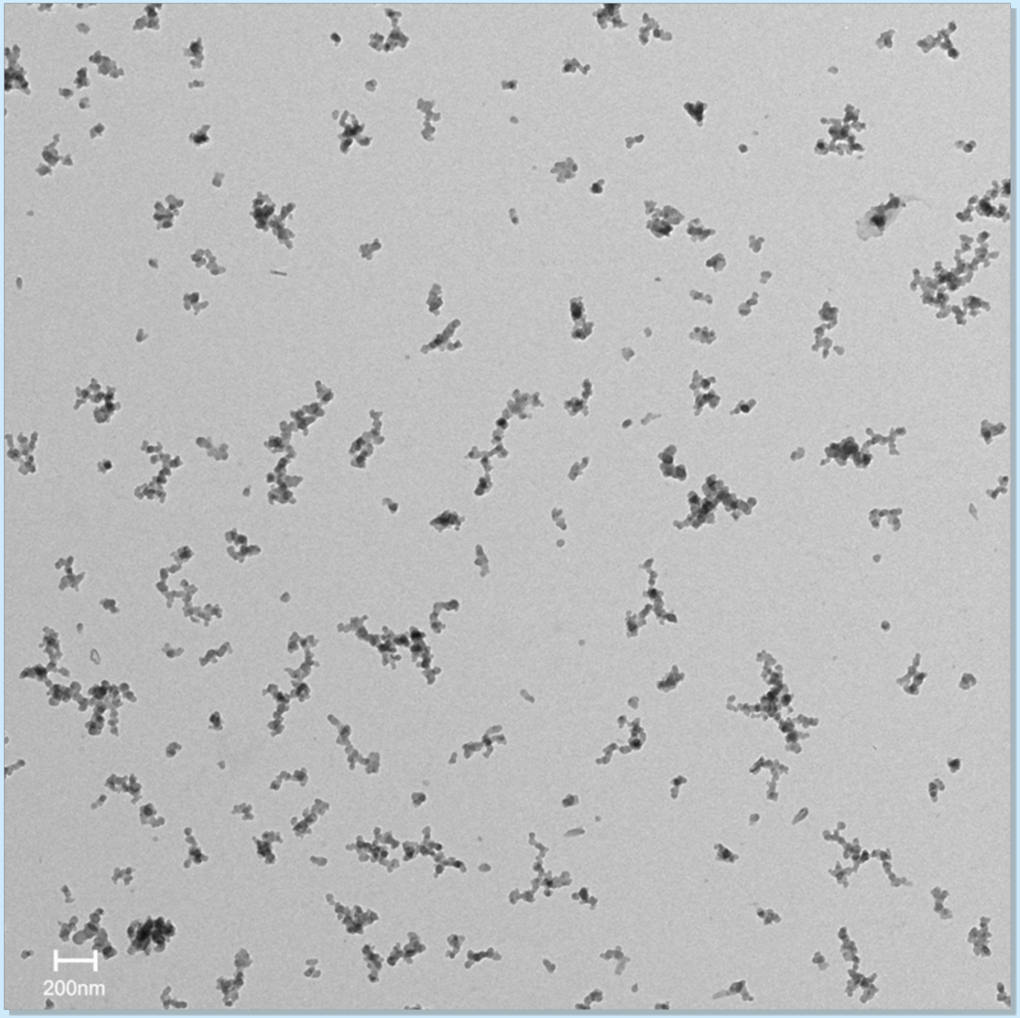
TEM Images of Nanoparticles Sampled From a Flame



TEM Images of Nanoparticles Sampled From a Flame



- particulate matter properties of interest:
  - concentration
  - active surface area
  - primary particle diameter distribution
  - aggregate size distribution
  - optical properties
  - volatile fraction
  - composition

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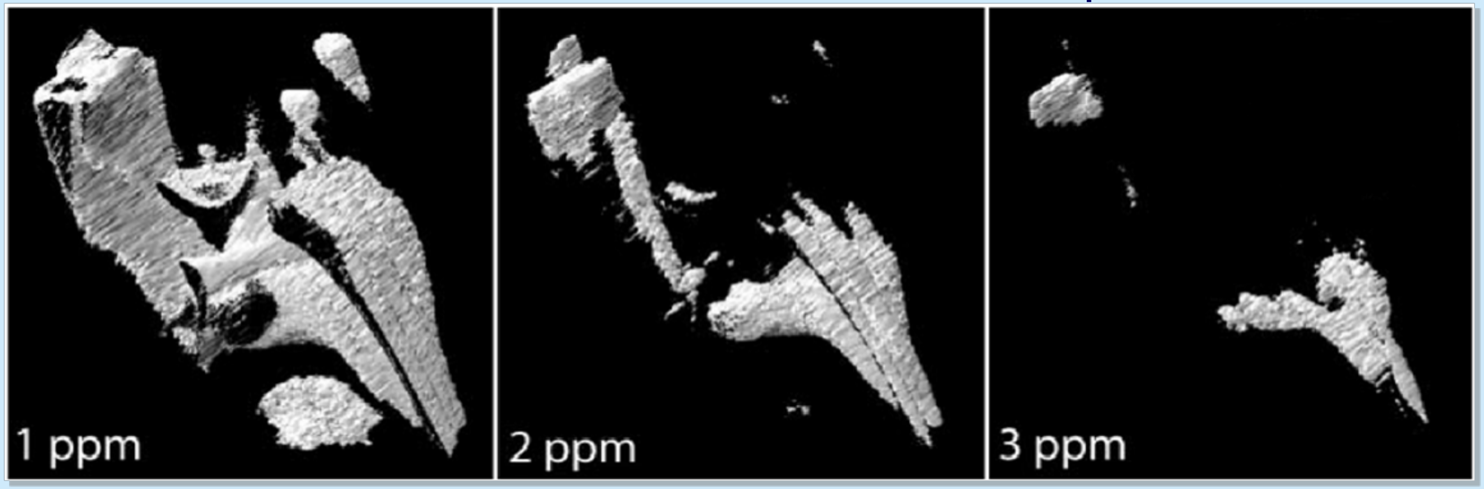
What is Laser-Induced Incandescence (LII)?

- laser-induced incandescence is a generic name for the physical process of rapidly heating refractory nanoparticles with a laser to the point that the radiative emission from the particles is discernable from the background
- many variants of LII have appeared
  - high fluence (most common)
    - particles are heated to their sublimation temperature
  - low fluence
    - particles are heated to lower than sublimation temperature
  - remote *in situ* nonintrusive measurements (some instruments)
    - fundamental studies on open flames
  - extractive sampling (most instruments)
    - engine and combustor development and emissions measurement

Further LII Variants

- pulsed laser or cw laser
- time-resolved (TiRe-LII) or gated
- single (narrow or broadband), two, or multiple wavelengths
- 0-D (point measurements); 1-D (line measurements); 2-D (area measurements); or 3-D (volume measurements)
  - iso-concentration surfaces in a turbulent non-premixed flame



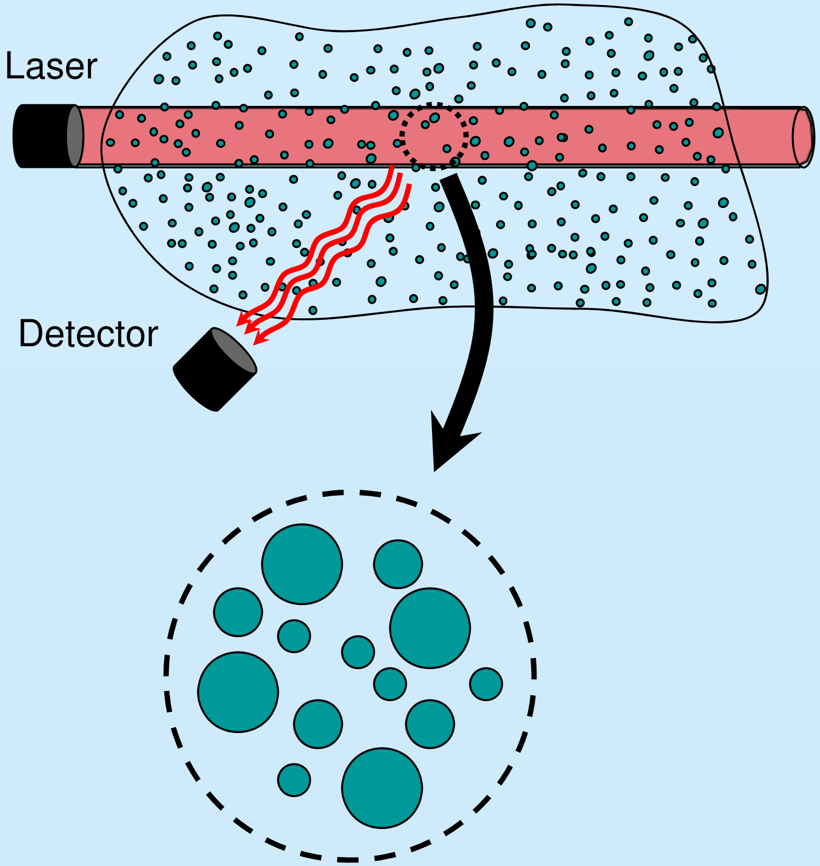
[Hult *et al.*, Experiments in Fluids **33**, 2002]

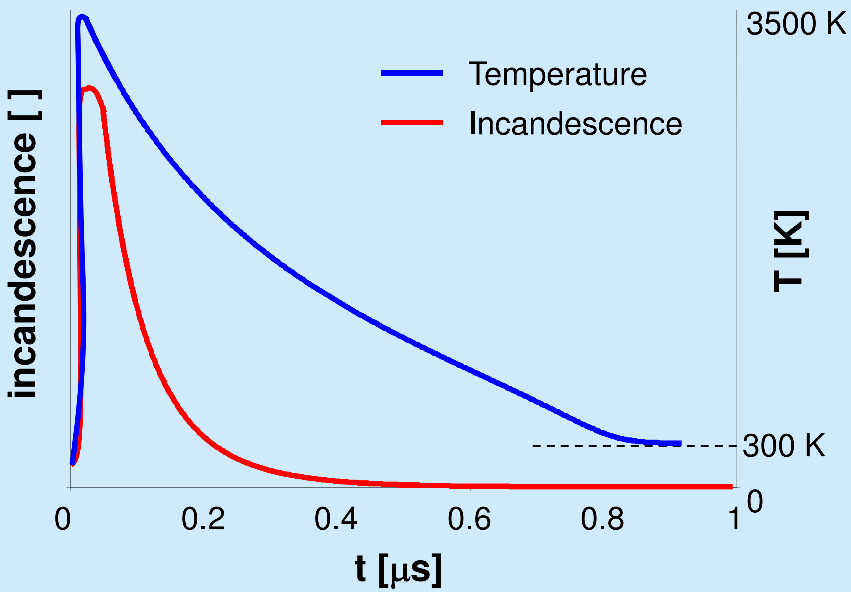
What Does LII Do?

- quantitative measurement for soot:
  - concentration (0.01 ppt – 10 ppm volume; 20ng/m^3 – 20g/m^3 mass)
  - active surface area (50 – 200 m^2/g)
  - primary particle diameter (typically 5-50 nm)
  - number density of primary particles
- properties are for an ensemble of particles
- measurement features:
  - very high precision and repeatability
  - transient concentration
  - nonintrusive (dilution unnecessary)
  - wide range of applicability
  - potential standardized method
  - measures soot
  - high selectivity

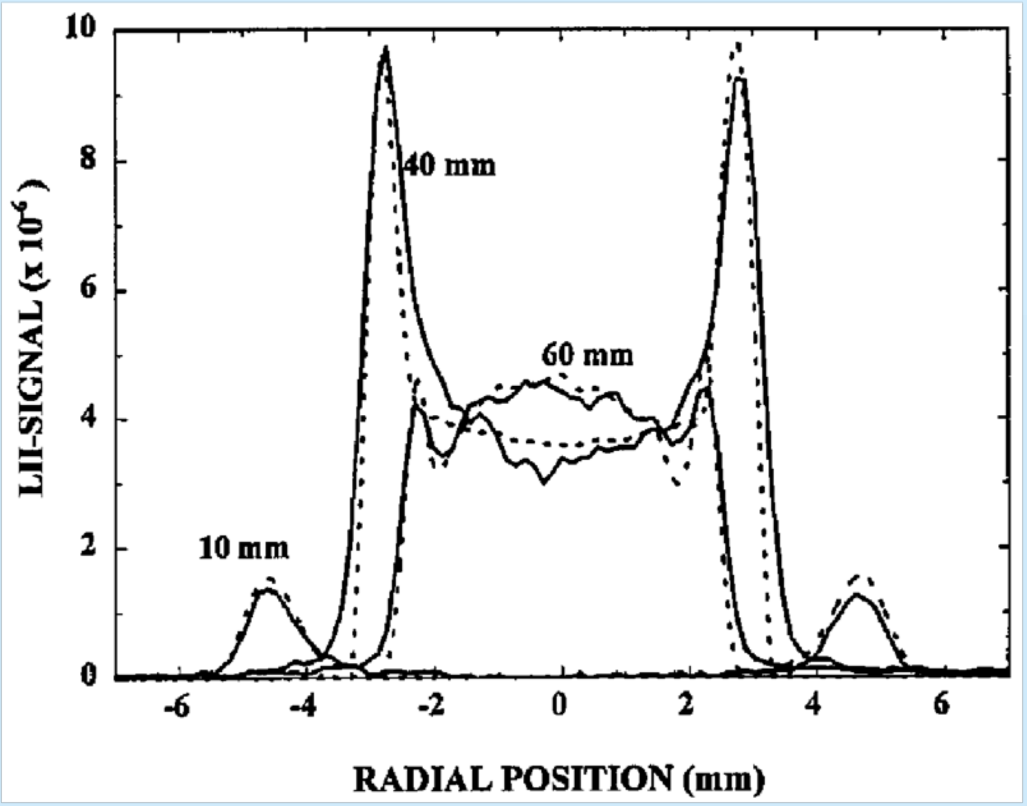
[Schulz *et al.*, Applied Physics B **83**, 2006]

Time-Resolved Laser Induced Incandescence





LII Calibration – Correlation to Other Flame Measurements



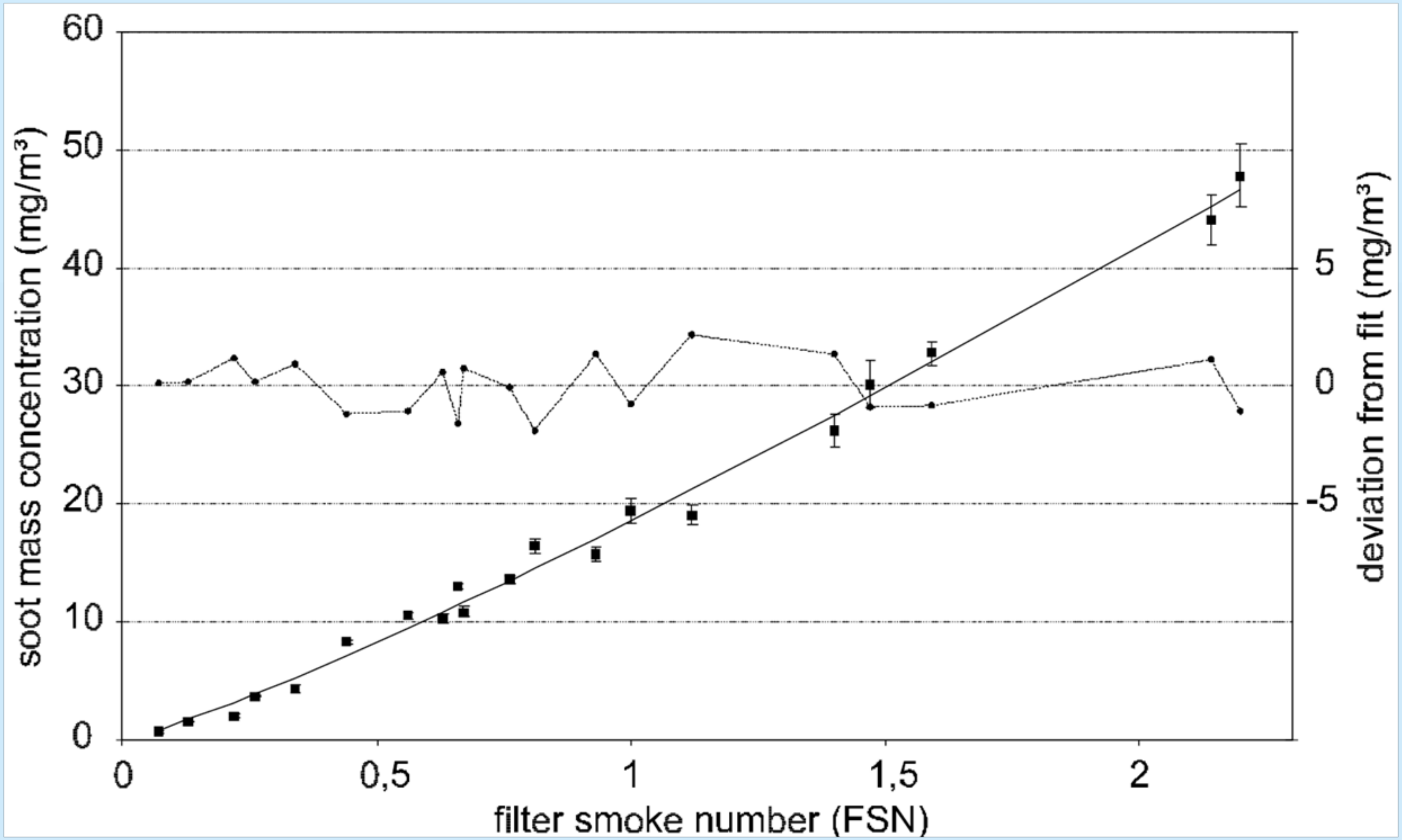
[Ni *et al*., Applied Optics **34**, 1995]

- calibration by comparison of the LII signal to the SVF profiles obtained by laser extinction

—— LII data

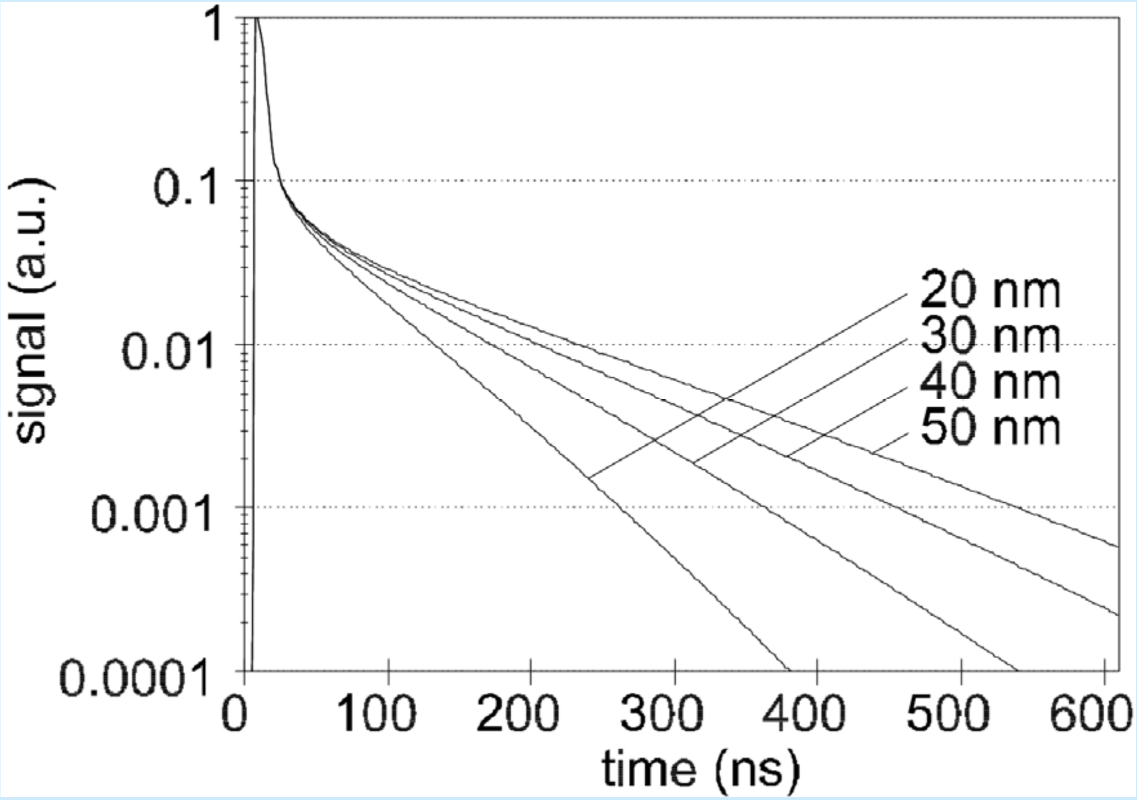
- - - - laser extinction/scattering data

LII Calibration – Comparison to Other Instruments



[Schraml *et al.*, SAE Paper No. 2000-01-2002, 2000]

Determination of Primary Particle Diameter



[Schraml *et al.*, SAE Paper No. 2000-01-2002, 2000]

- numerically modelled LII signal decay of soot at STP ambient conditions
- signal decay rate varies with primary particle diameter

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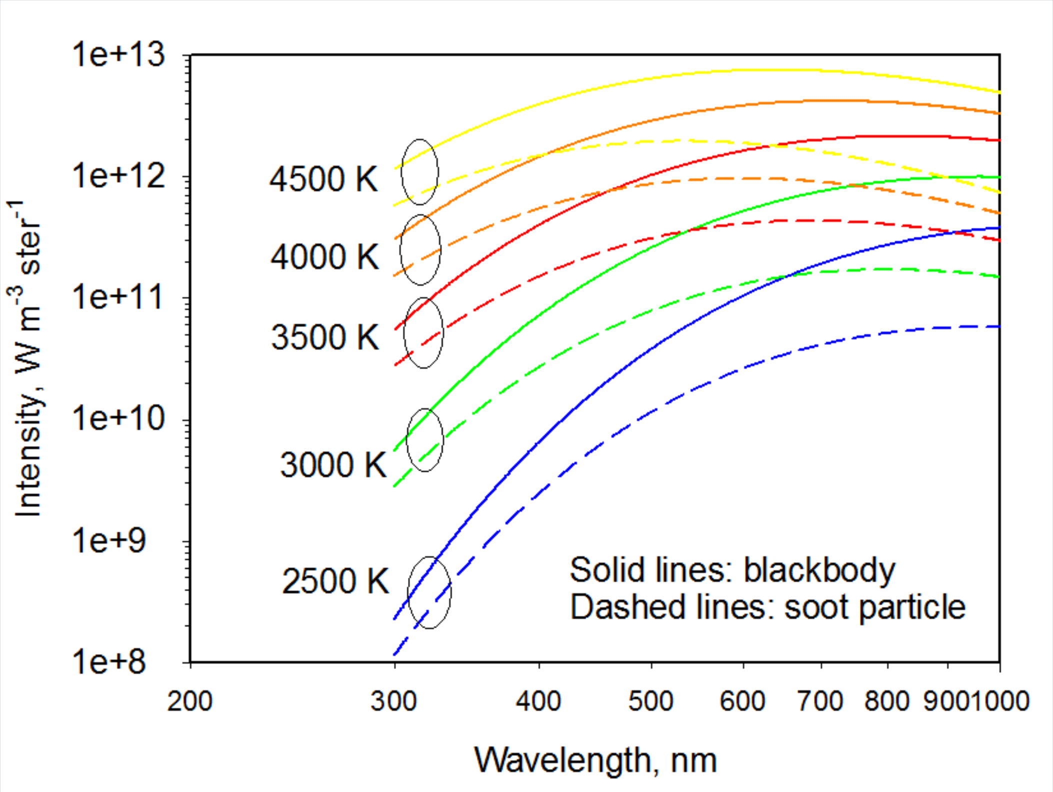
Auto-Compensating LII (AC-LII)

- two-color pyrometry coupled with LII to determine the time-resolved particle temperature
  - permits use of low-fluence
  - particles are kept below the sublimation temperature
- this new technique *automatically compensates* for any changes in the experimental conditions
  - fluctuations in local ambient temperature
  - variation in laser fluence
  - laser beam attenuation by the particulate matter
  - desorption of condensed volatile material

Particle Emission Intensities



[Smallwood, Ph. D. Thesis, Cranfield University, 2009]

- blackbody and soot particle emission intensity
  - over range of temperatures encountered in LII over the UV-VIS-NIR spectral range
- emissivity

$$\varepsilon_p = \frac{4\pi d_p E(m)}{\lambda}$$

- soot particles are calculated for d_p = 30 nm and $E(m)$ = 0.4

Soot Concentration from Two-Color Pyrometry

- temperature is determined from the spectral radiance signals at two wavelengths
  - varies with relative *E*(*m*) at the two wavelengths

$$T = \frac{hc}{k}\left(\frac{1}{\lambda_2} - \frac{1}{\lambda_1}\right)\left[\ln\left(\frac{V_{\exp_1}\lambda_1^{\,6}}{\eta_1 E(m_{\lambda_1})}\right) - \ln\left(\frac{V_{\exp_2}\lambda_2^{\,6}}{\eta_2 E(m_{\lambda_2})}\right)\right]^{-1}$$

- soot volume fraction is determined from the temperature and the spectral radiance signal at either one of the wavelengths
  - depends upon absolute value of *E*(*m*) at the selected wavelength

$$f_V = \frac{V_{EXP_\lambda}\,\rho}{\eta_\lambda w_b}\,\frac{\lambda^6\left(e^{\frac{hc}{k\lambda T}} - 1\right)}{12\pi c^2 h\, E(m_\lambda)} = V_{EXP_\lambda}\,\frac{K_1}{E(m_\lambda)}\left(e^{\frac{K_2}{T}} - 1\right)$$

[Smallwood, Ph. D. Thesis, Cranfield University, 2009]

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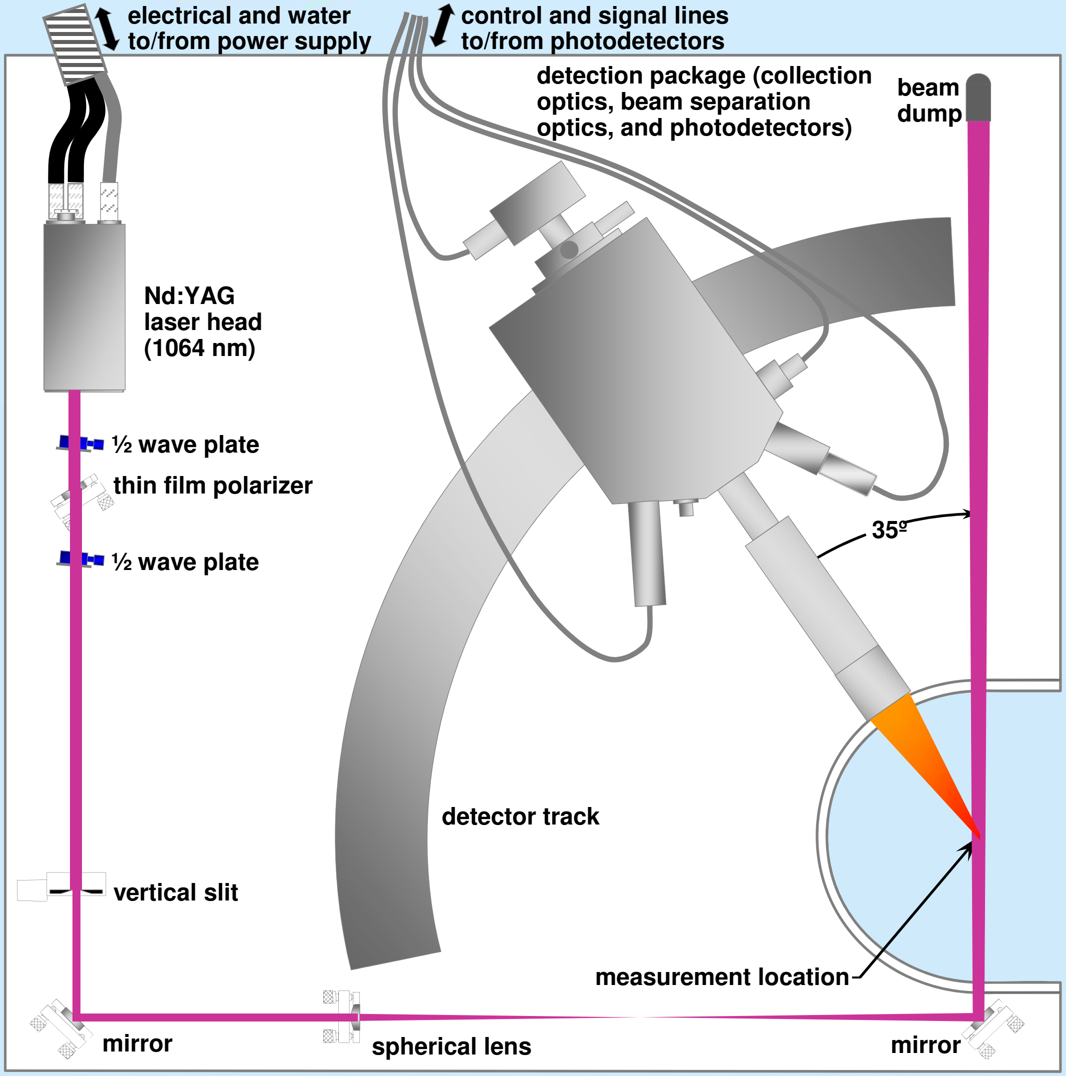
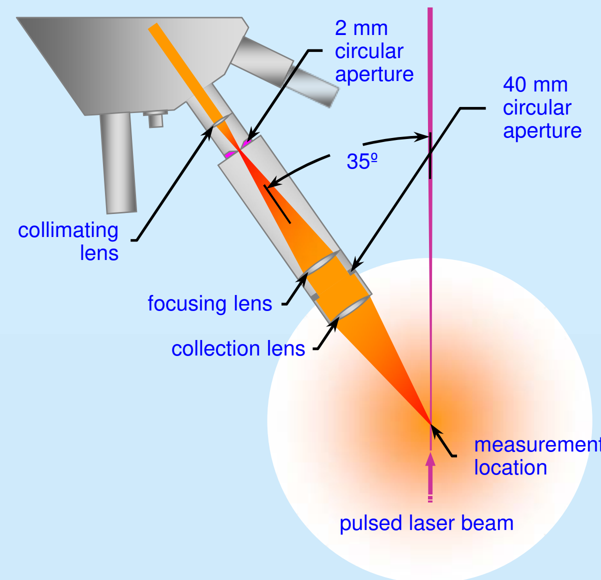
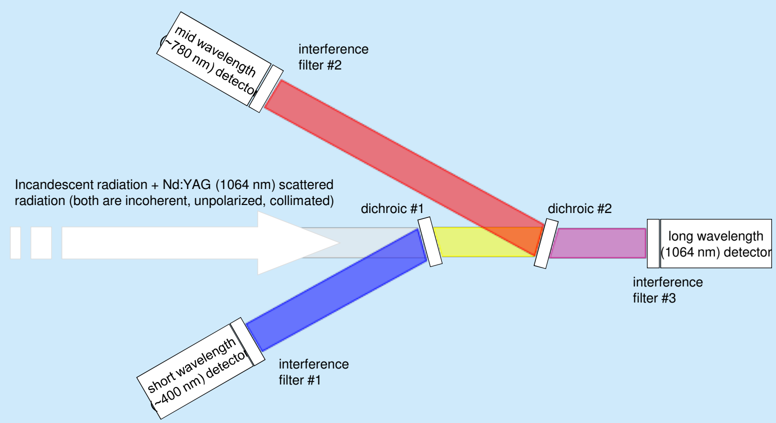
Experiment: LII Optical Apparatus

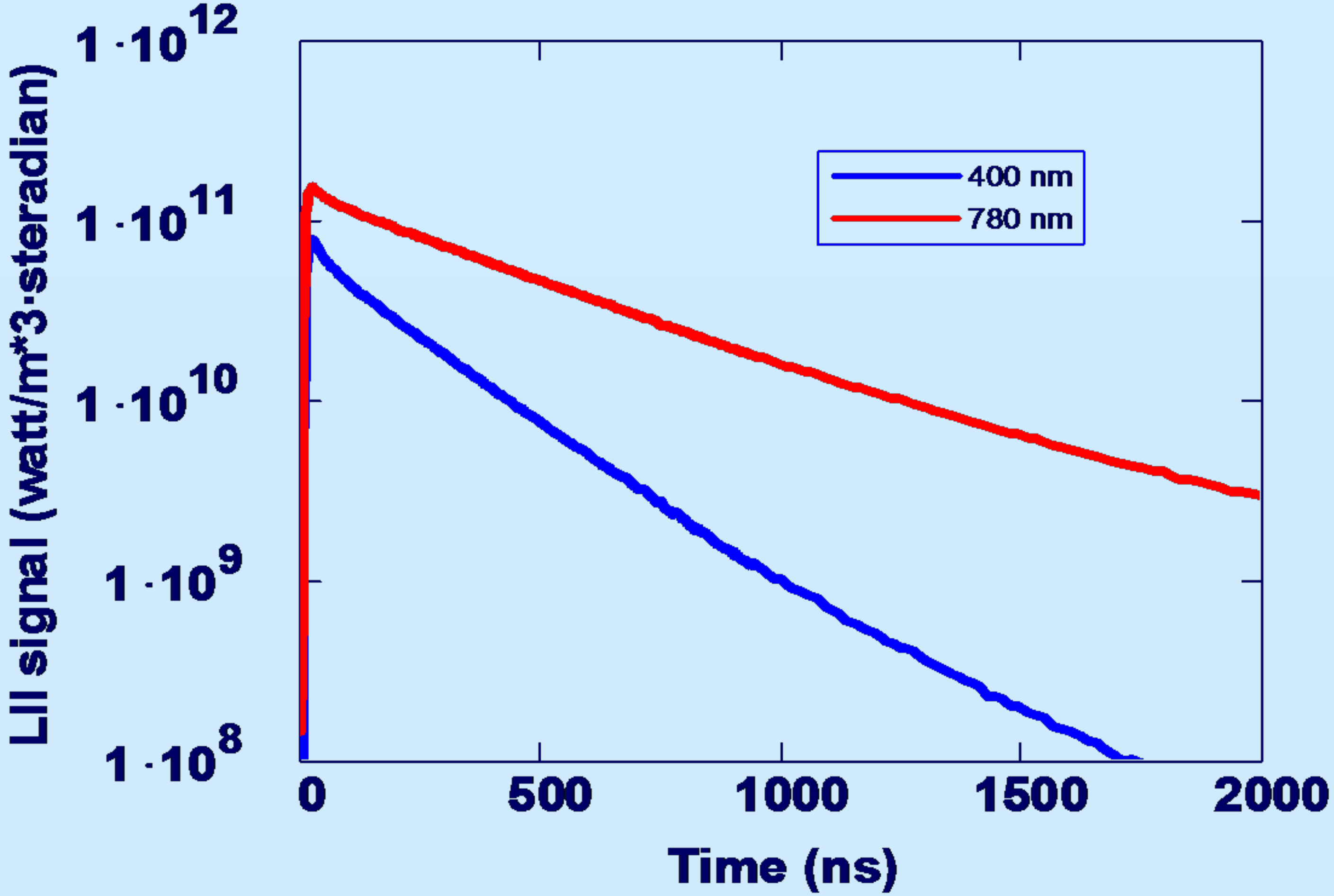


[Smallwood *et al.*, SAE Paper No. 2001-01-3581, 2001]



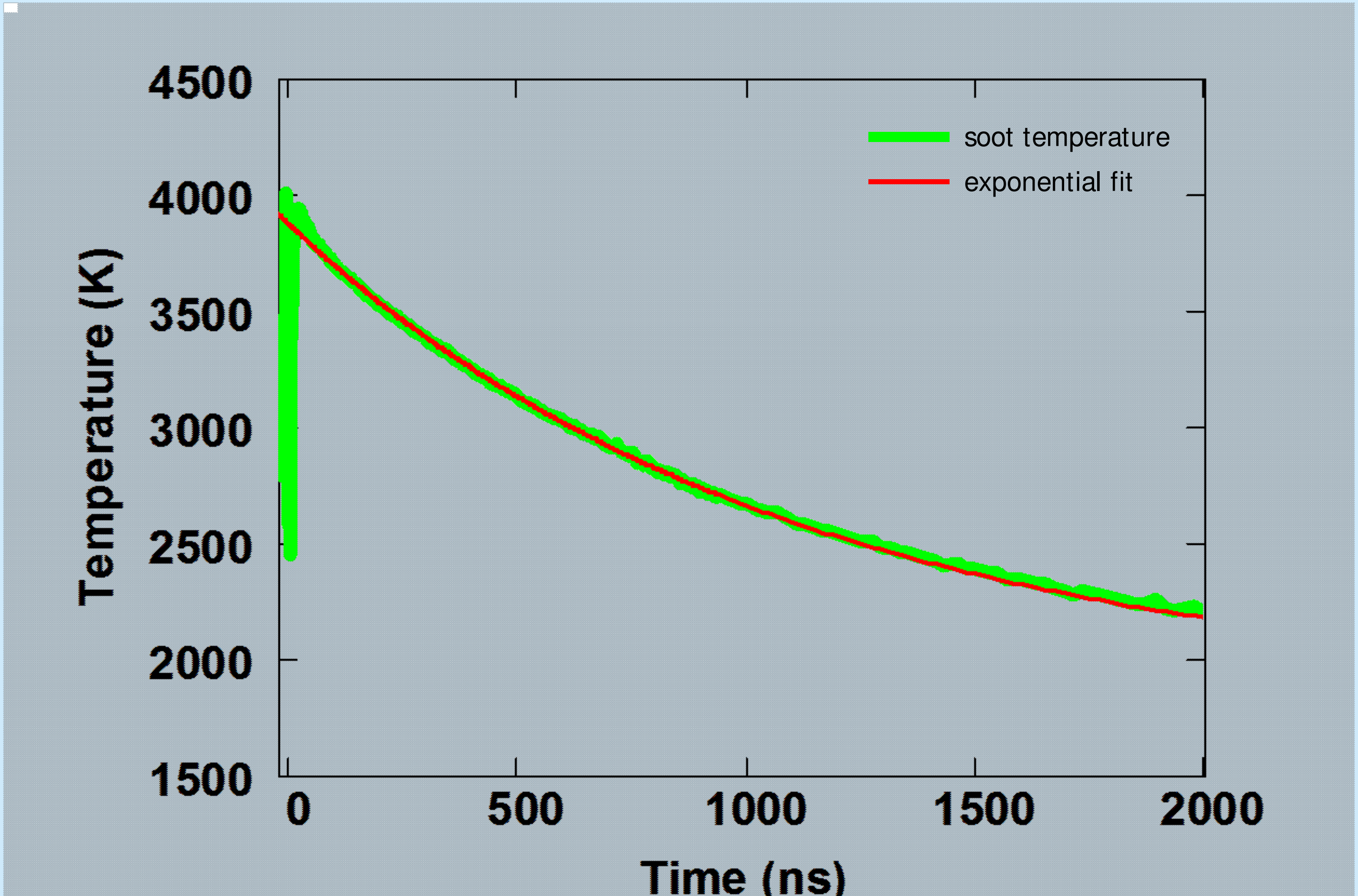


Absolute LII Signals



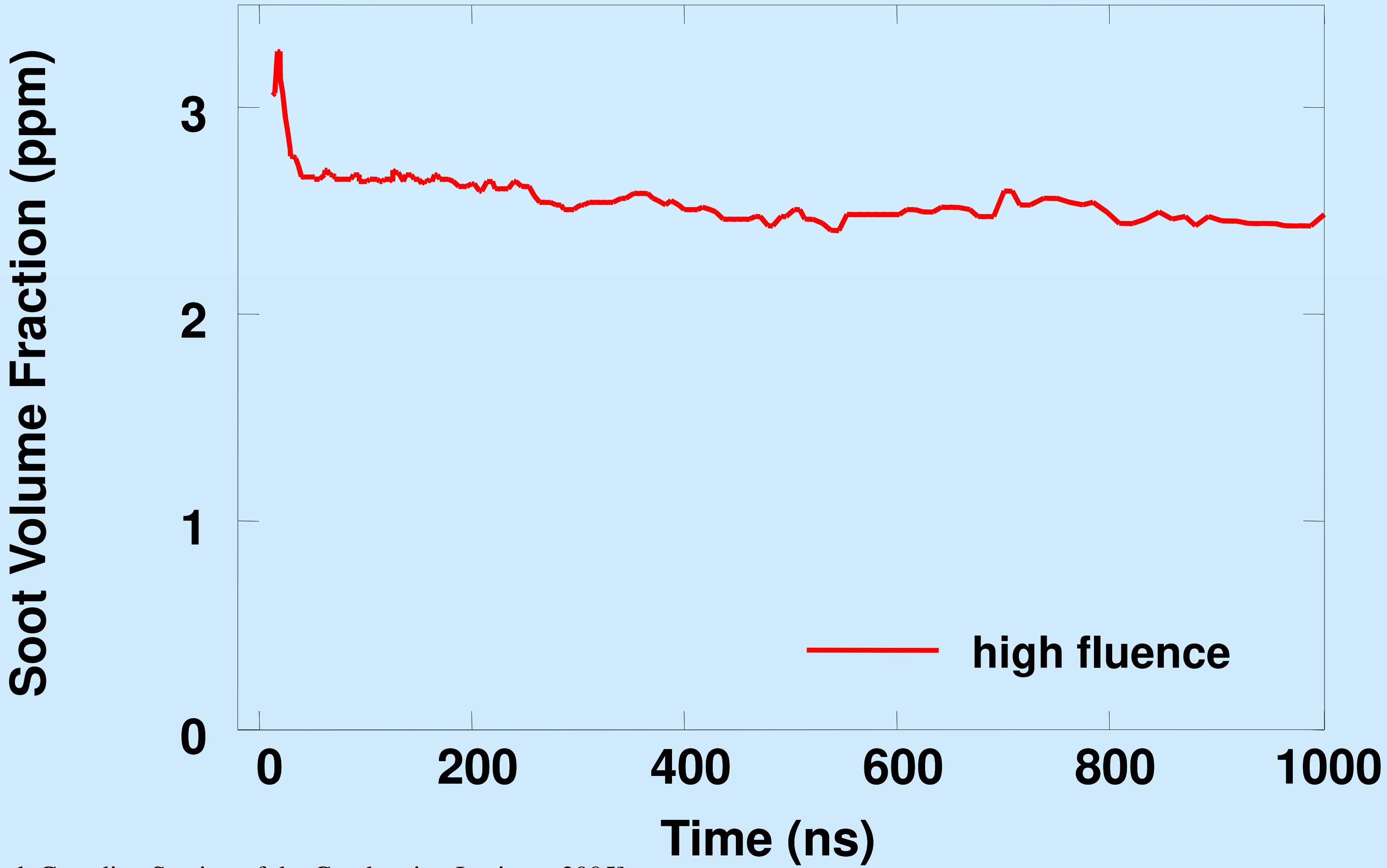
[Smallwood, Canadian Section of the Combustion Institute, 2005]

Real-time Temperature



[Smallwood, Canadian Section of the Combustion Institute, 2005]

Impact of Low Fluence LII



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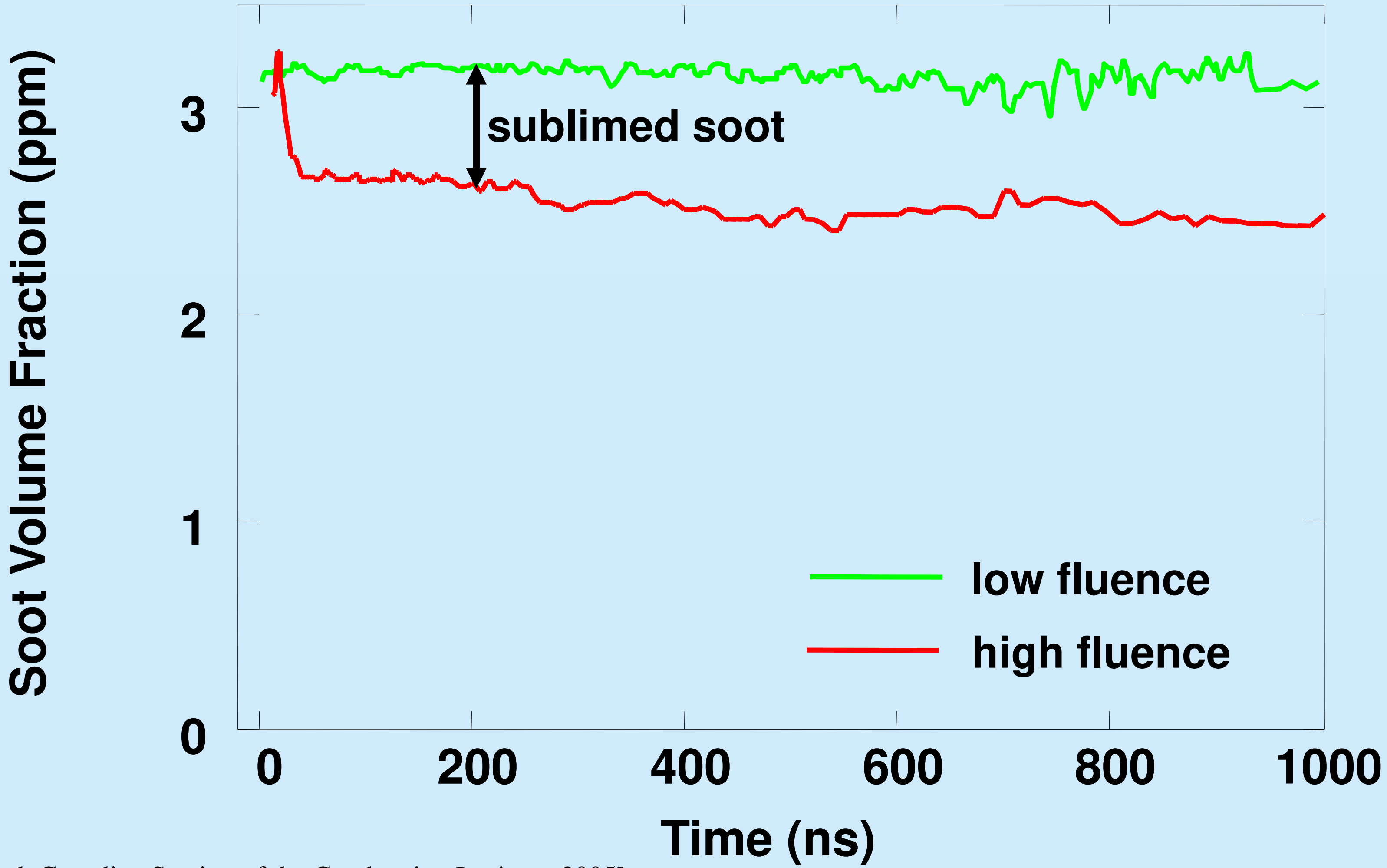
Impact of Low Fluence LII



[Smallwood, Canadian Section of the Combustion Institute, 2005]

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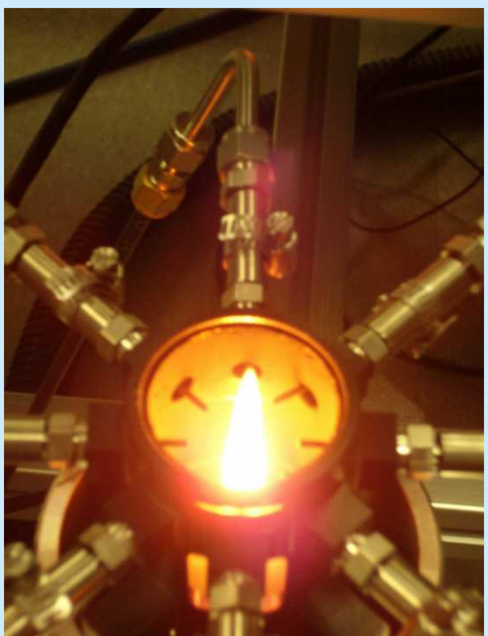
Demonstration of Fluence Effects in LII



.83 mJ/mm^2

1.25 mJ/mm^2

0.50 mJ/mm^2 Peak Fluence

2.50 mJ/mm^2

3.75 mJ/mm^2

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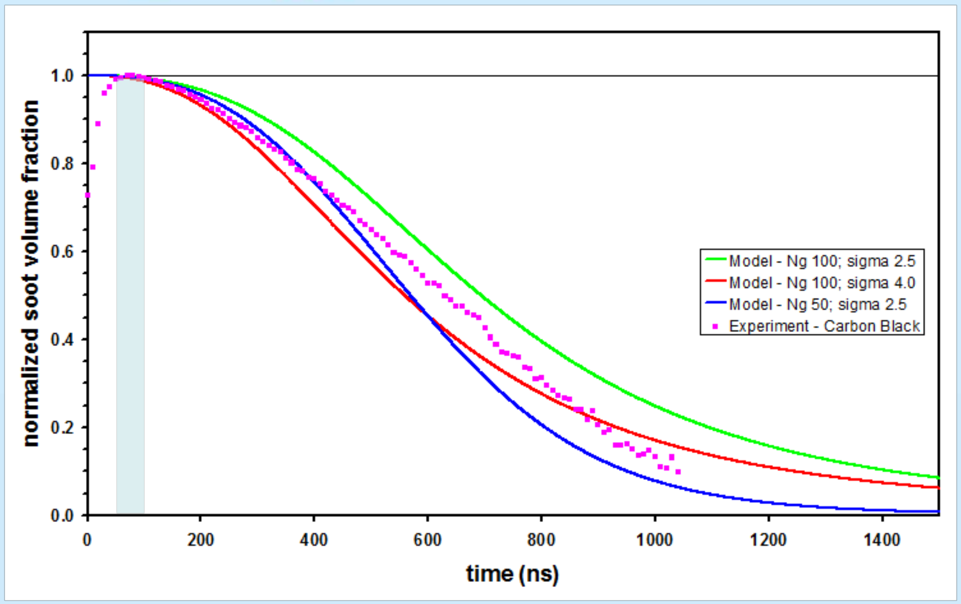
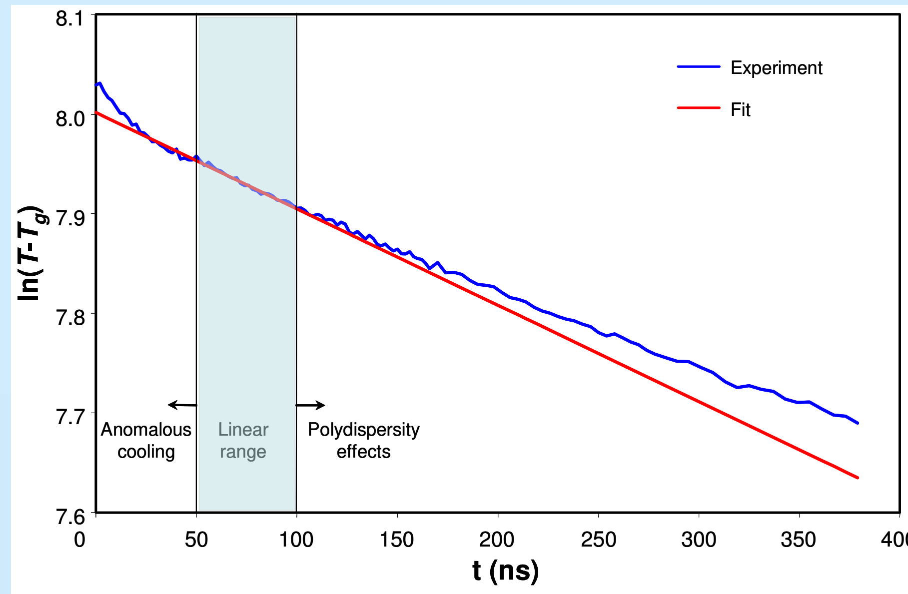
Experiment: Optimum Analysis Interval

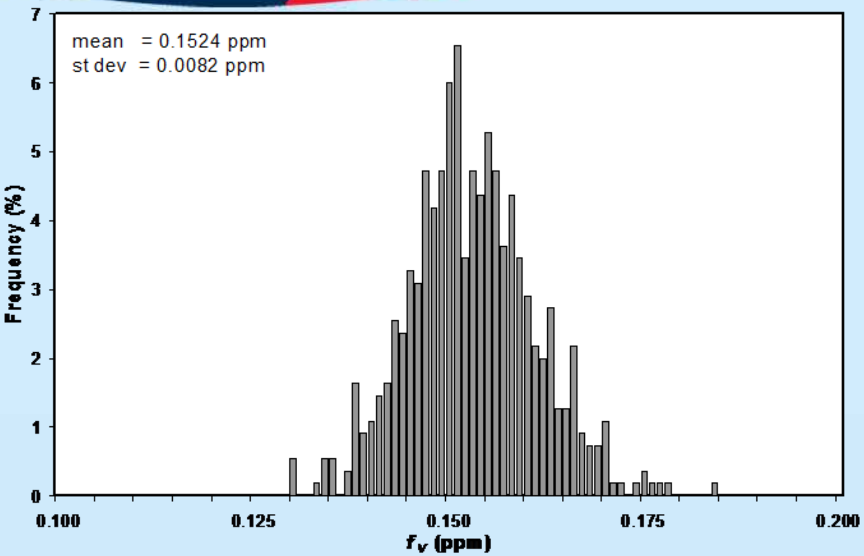


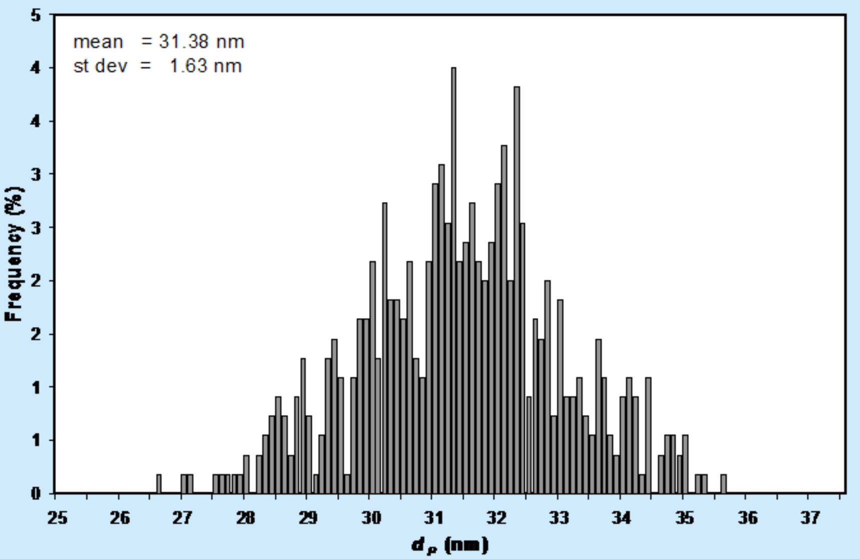


[Smallwood, Ph. D. Thesis, Cranfield University, 2009]

- for high quality AC-LII measurements, the optimum analysis interval was found to be approximately 50-100 ns after the peak of the laser pulse
  - maximum soot volume fraction and single exponential temperature decay
  - interval is dependent upon experimental conditions

LII Precision

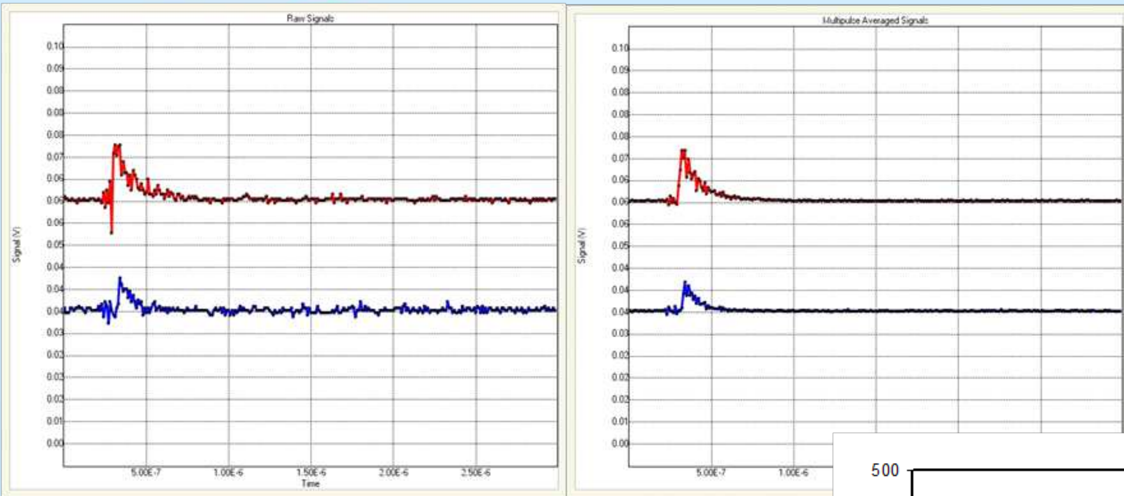




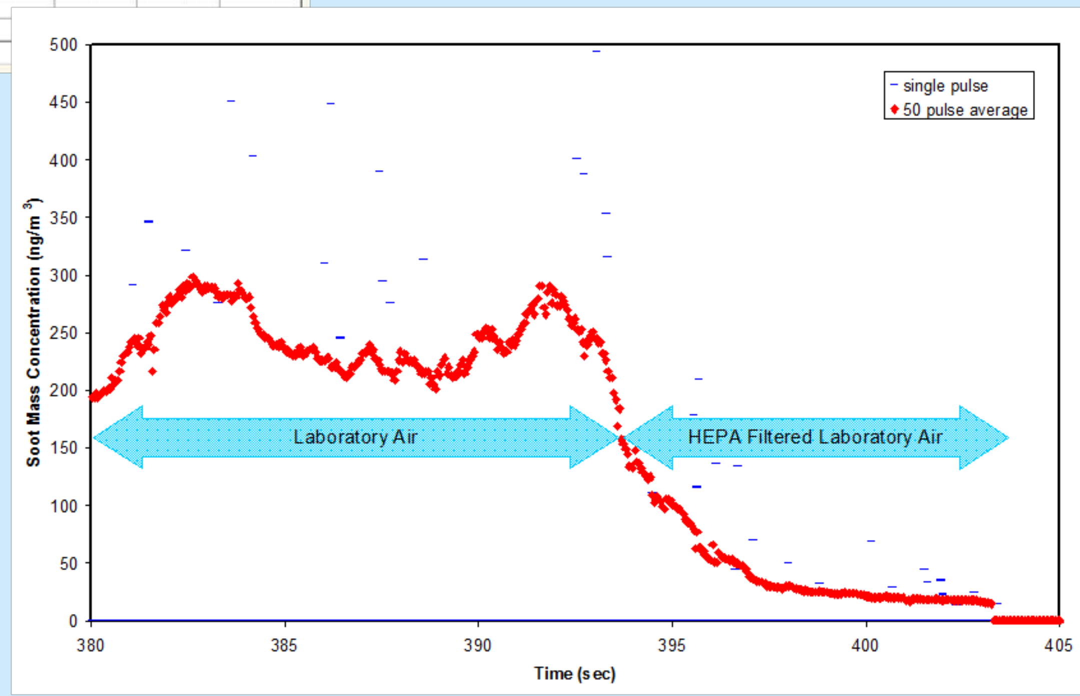
- single-shot precision of LII in measuring soot concentration and primary particle diameter is good
- standard deviation is about 5% for these examples acquired above a quenched laminar diffusion flame

[Smallwood, Ph. D. Thesis, Cranfield University, 2009]

Single-shot vs. Multi-pulse Averaging



[Smallwood, Ph. D. Thesis, Cranfield University, 2009]



ABOVE

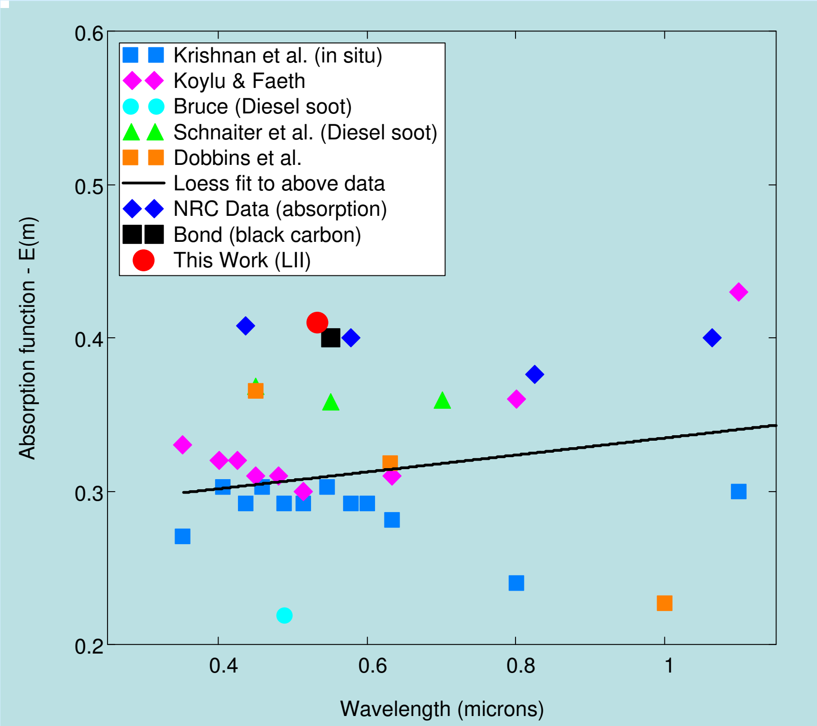
- single-shot (left) and 50-shot average (right)

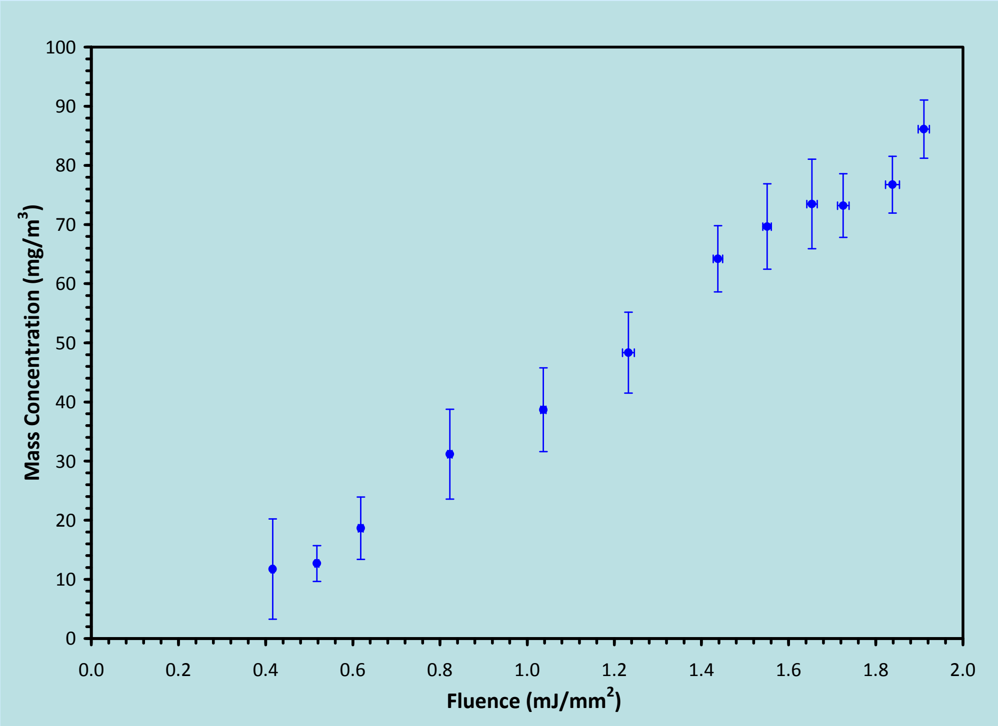
RIGHT

- effect of averaging on measurement validation rate

AC-LII Issues

- AC-LII does not always agree with gravimetric
  - need improved knowledge of *E*(*m*) as a function of temperature and wavelength
  - SVF determined by AC-LII varies with fluence





[Smallwood, Ph. D. Thesis, Cranfield University, 2009]

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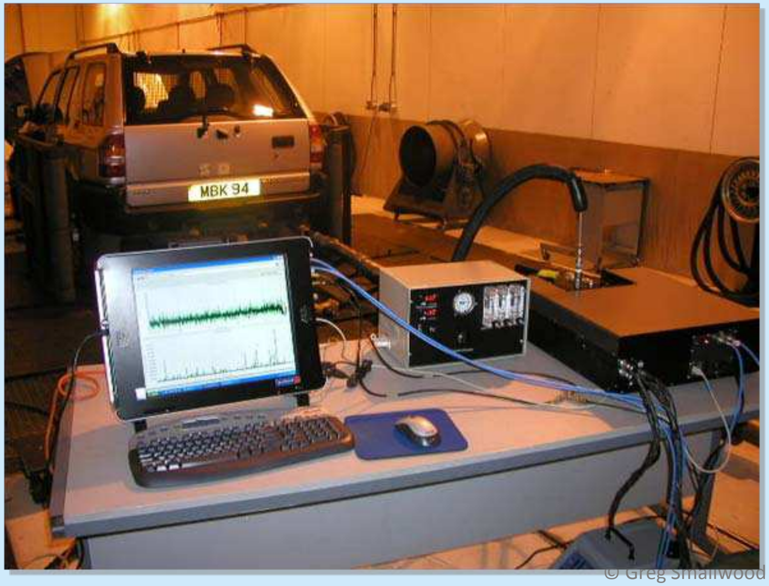
LII Applications: Present and Future

- process control of carbon black:
  - aggregate size distribution
  - higher sensitivity to changes in surface area
- air quality monitoring (urban and global):
  - greater concentration sensitivity
  - 0.05 parts-per-trillion (1 femtogram) detection limit
- engine emissions (manufacturers):
  - single-shot transient response
  - determination of volatile organic compound fraction
- vehicle emissions (regulators)
  - improved repeatability
  - on-road emissions measurements



LII Applications: Artium Technologies Instruments

- Artium Technologies takes an active role, with NRC's support, in working with customers who have purchased the LII 300 (top) or LII 200 (bottom) instruments
  - Easy to use
  - Low maintenance system
  - Low operating costs
  - Very high sensitivity
  - Compact rugged and portable instrument
  - Built-in computer and display, touchscreen control
  - Completely enclosed laser, optics, and sampling cell
  - 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
  - Technologies protected by US Patents 6,154,277 and 6,181,419 under license from National Research Council (NRC) Canada

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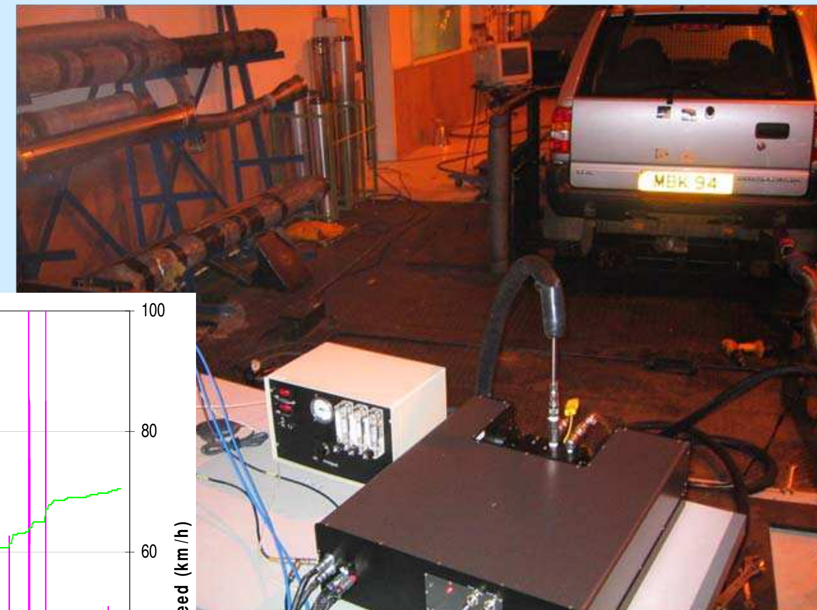
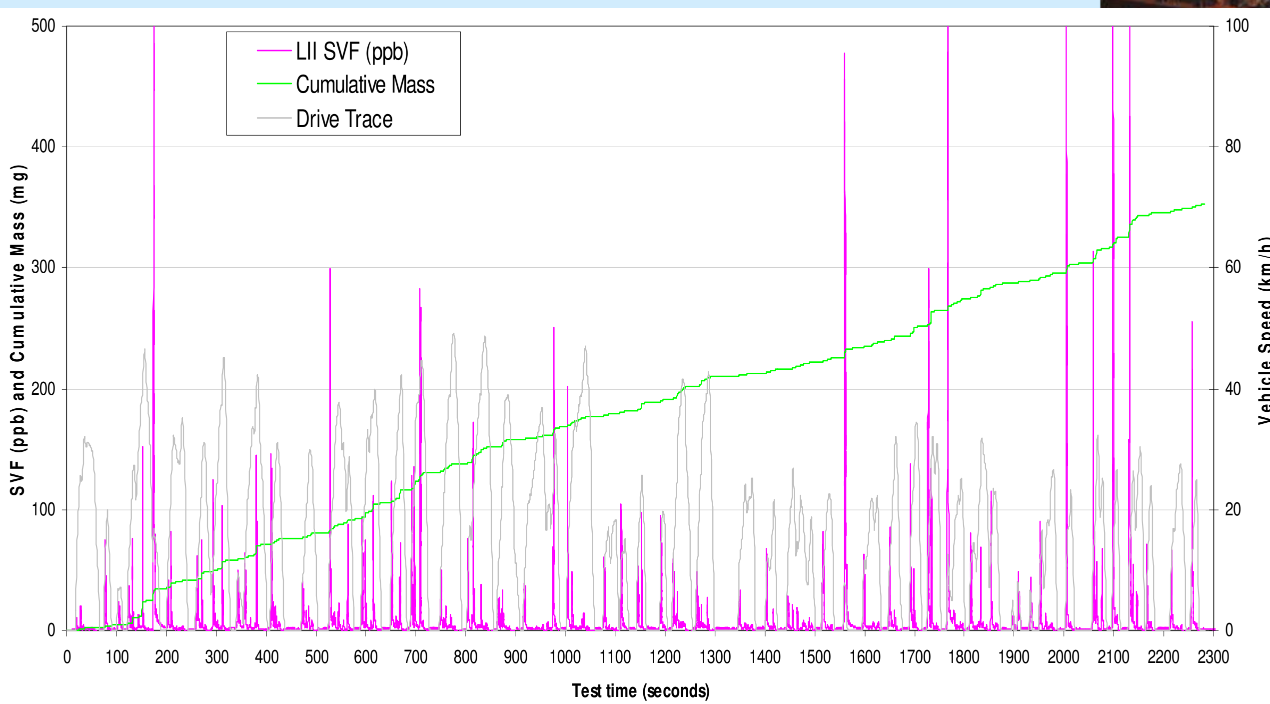
LII Measurement of Diesel Nonvolatile Particulate Emissions





[Smallwood *et al.*, 8th International ETH-Conference on Combustion Generated Particles, 2004]

LII Measurement of Diesel Nonvolatile Particulate Emissions

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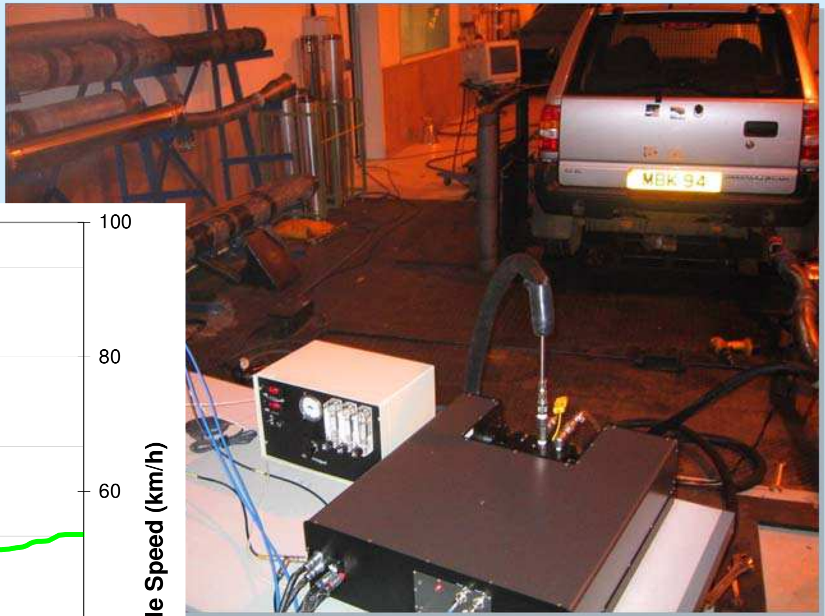
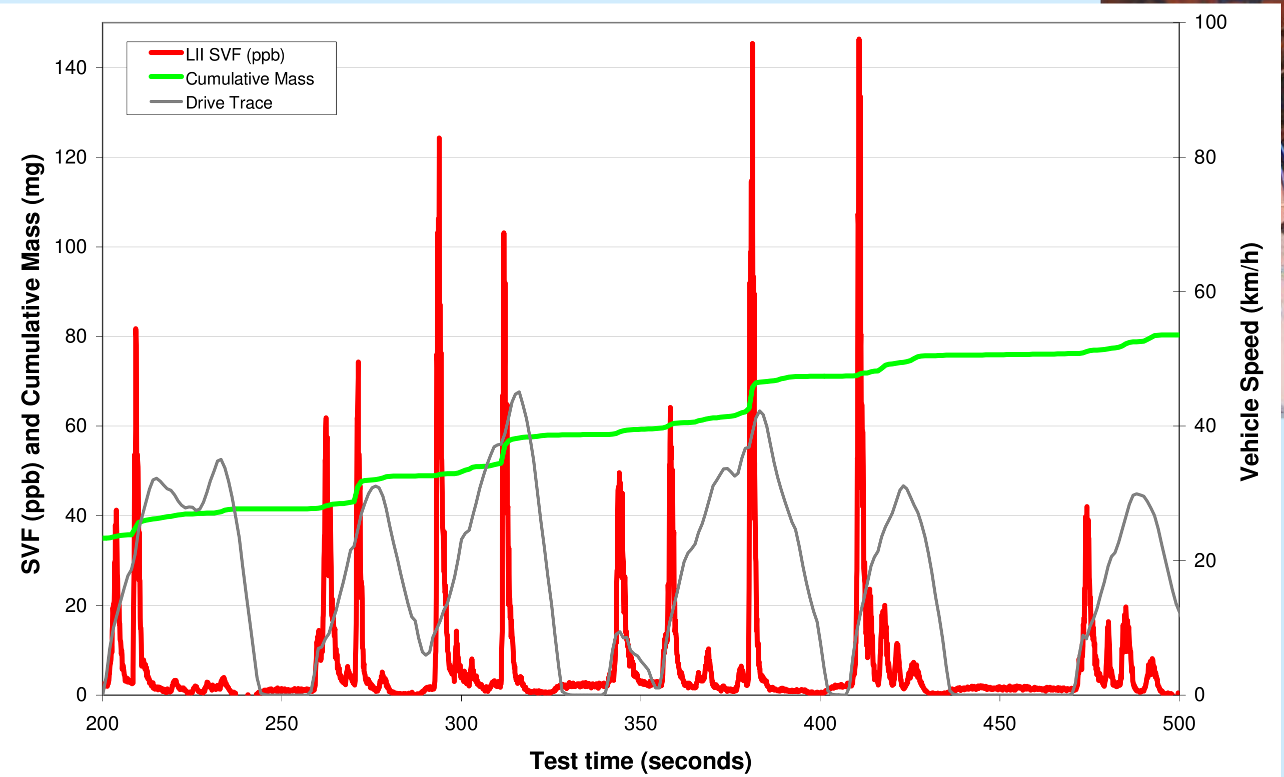
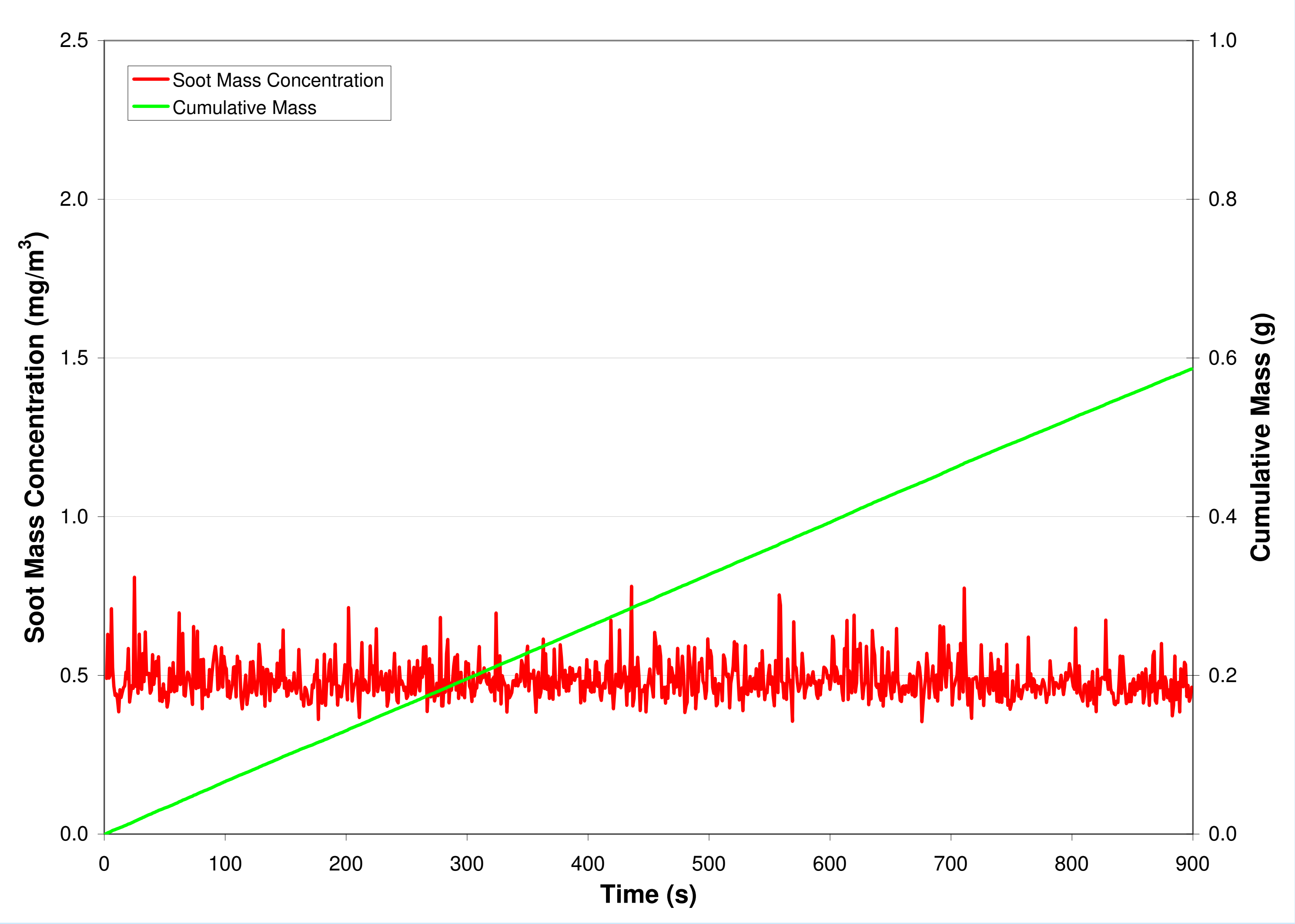




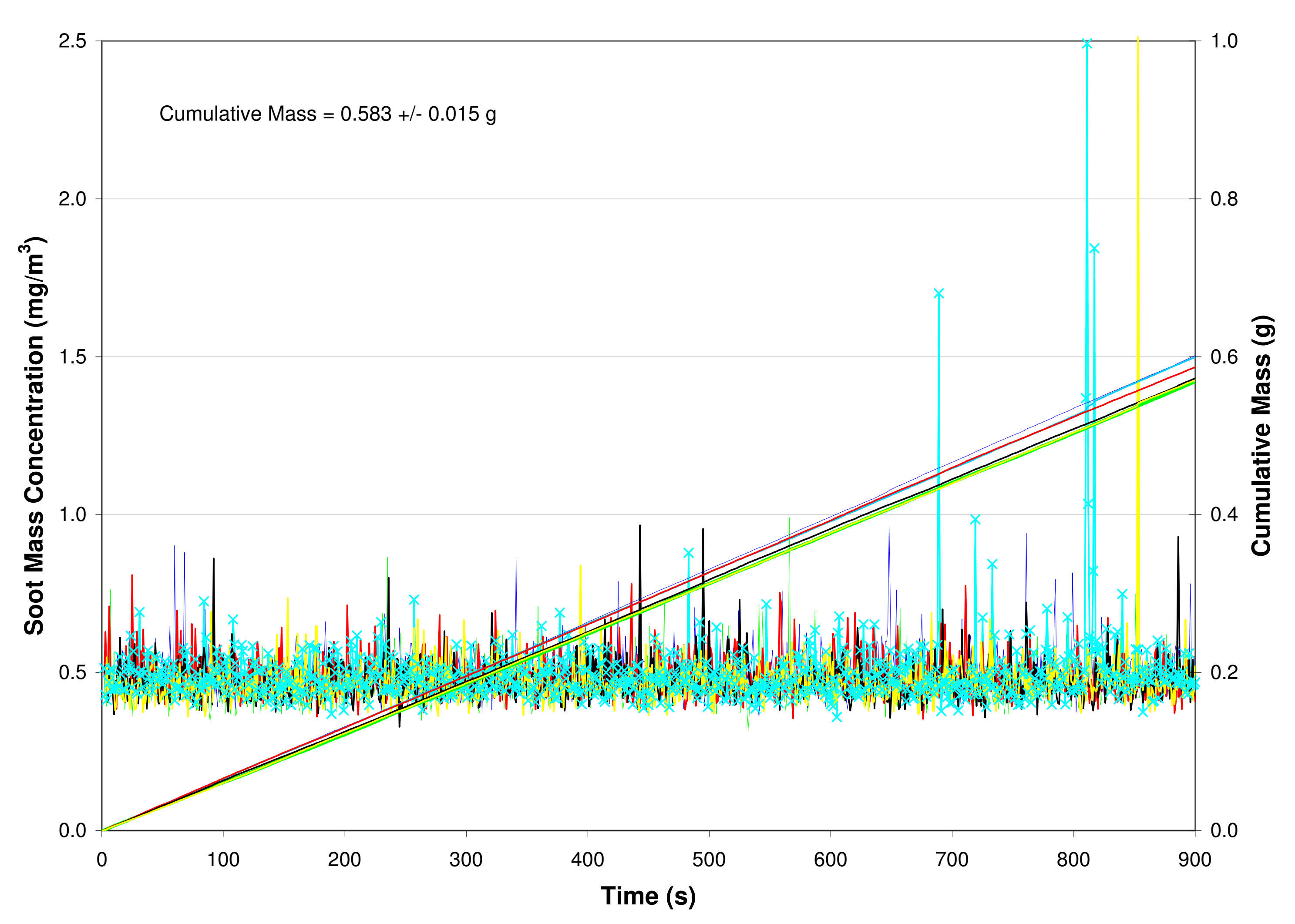
[Smallwood *et al.*, 8th International ETH-Conference on Combustion Generated Particles, 2004]

HD Diesel – Steady State



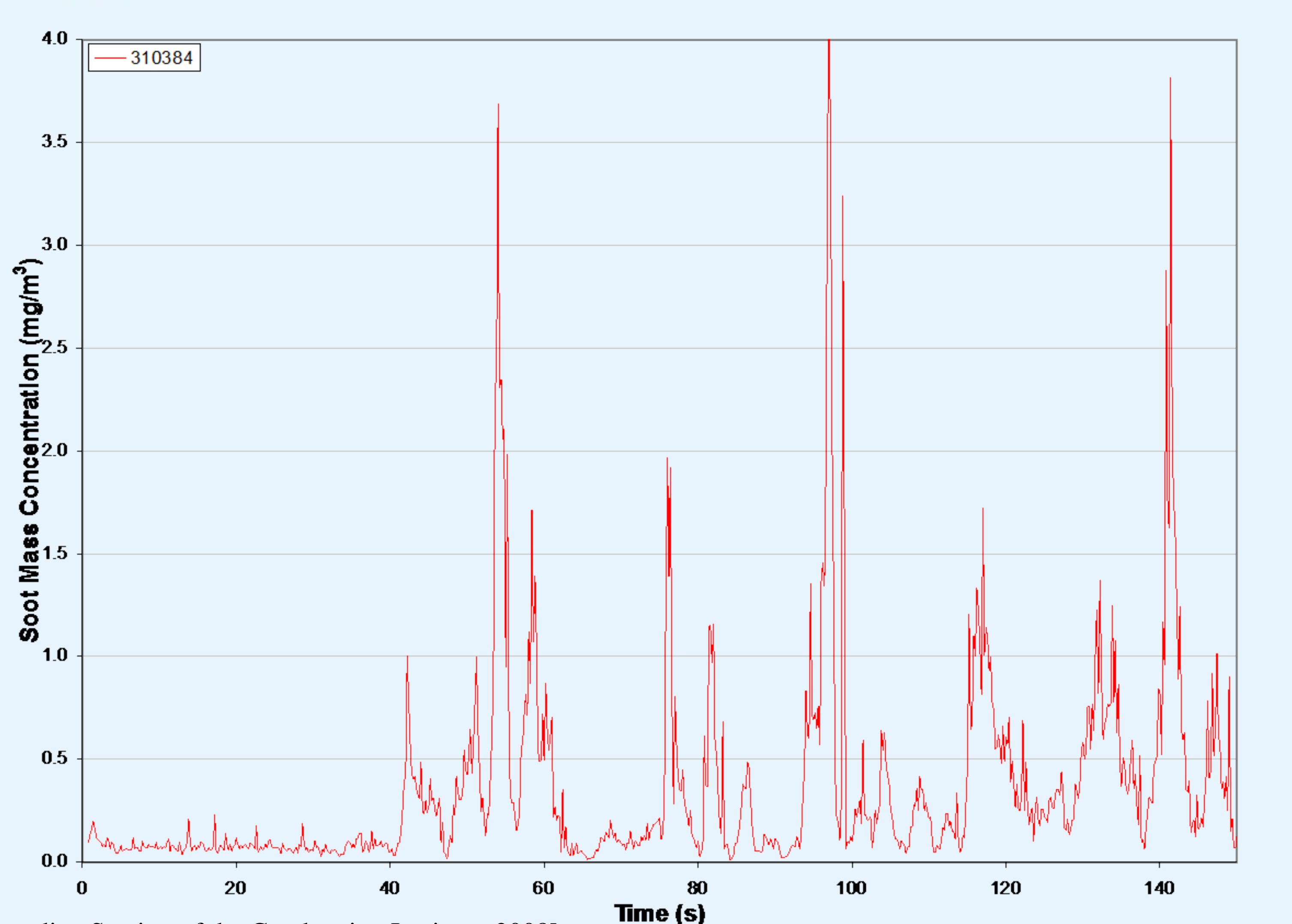
[Smallwood, Canadian Section of the Combustion Institute, 2008]

HD Diesel – Steady State – 6 Repeats



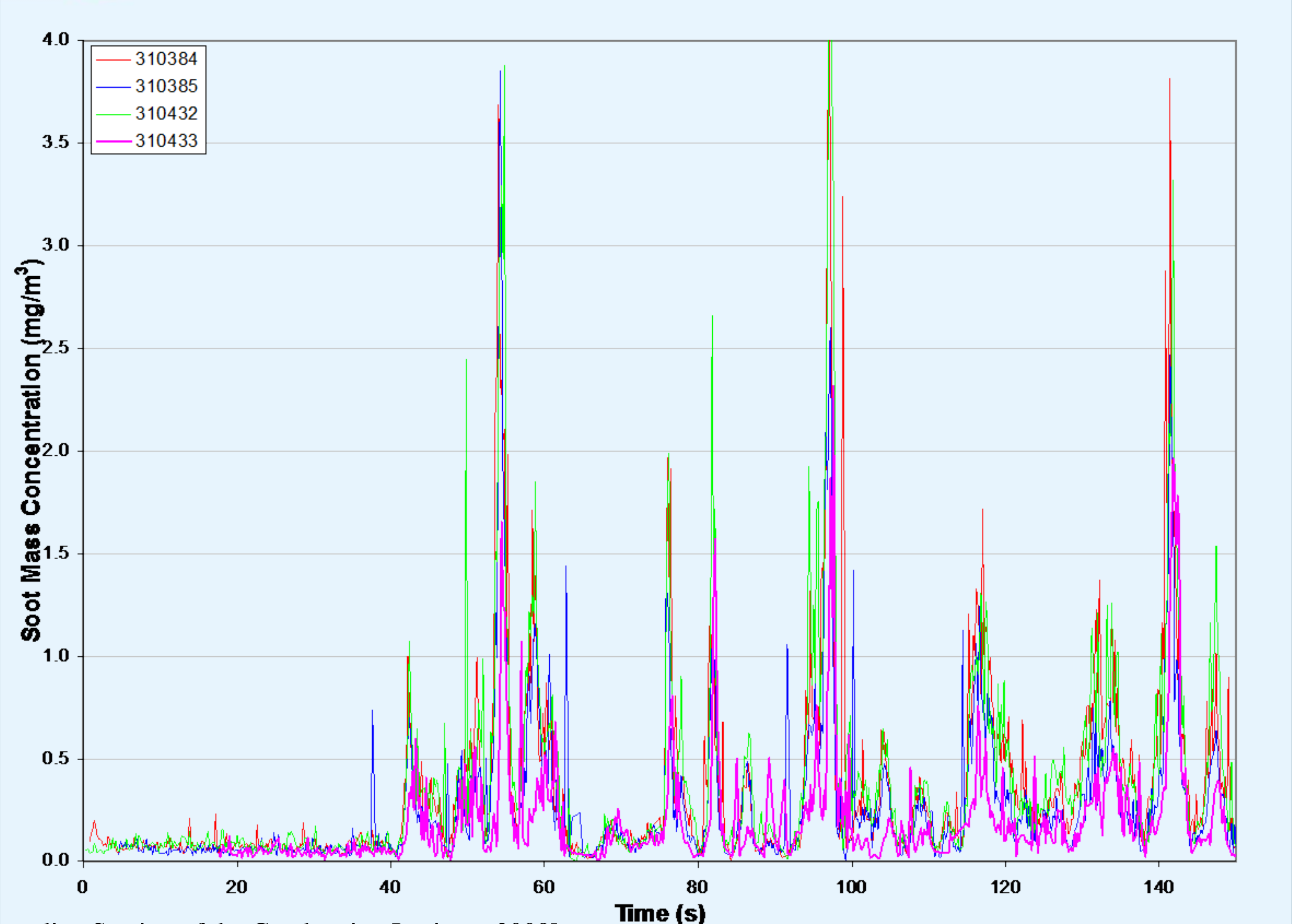
[Smallwood, Canadian Section of the Combustion Institute, 2008]

HD Diesel – Transients and Sensitivity



[Smallwood, Canadian Section of the Combustion Institute, 2008]

HD Diesel – Transients and Sensitivity – 4 rep.



[Smallwood, Canadian Section of the Combustion Institute, 2008]

Real-Time On-Road Particulate Measurements

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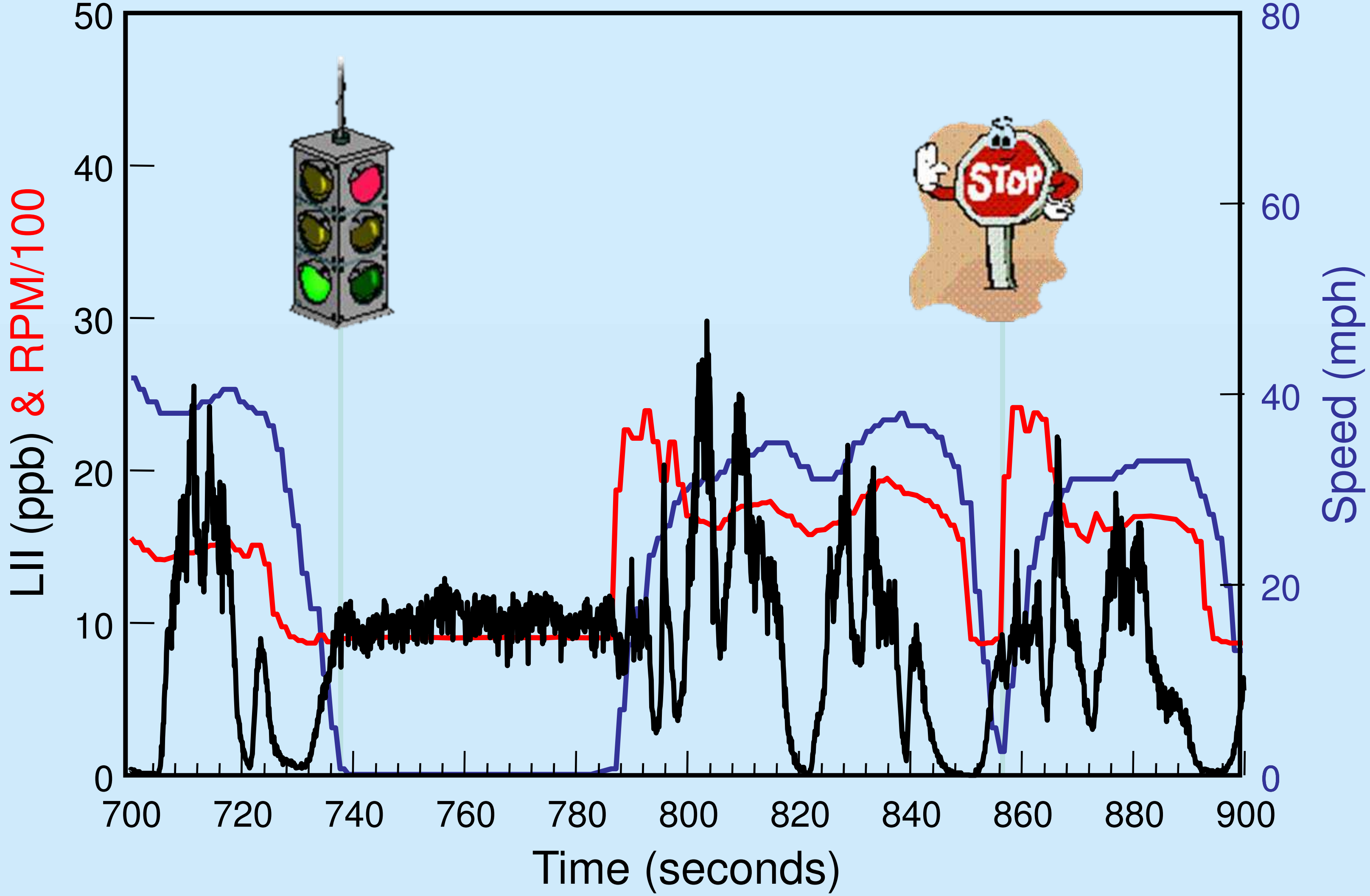
VW TDI: Stop-Start Urban Driving



[Witze *et al.*, 14th CRC On-Road Vehicle Emissions Workshop, 2004]

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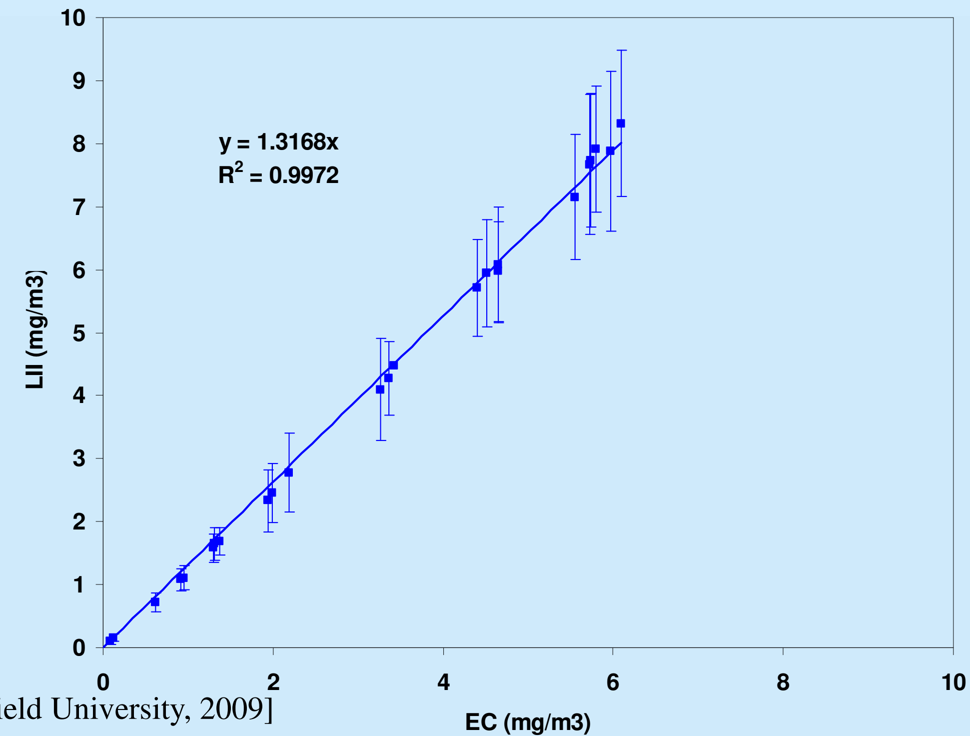
Comparison to Thermo-optical: EC

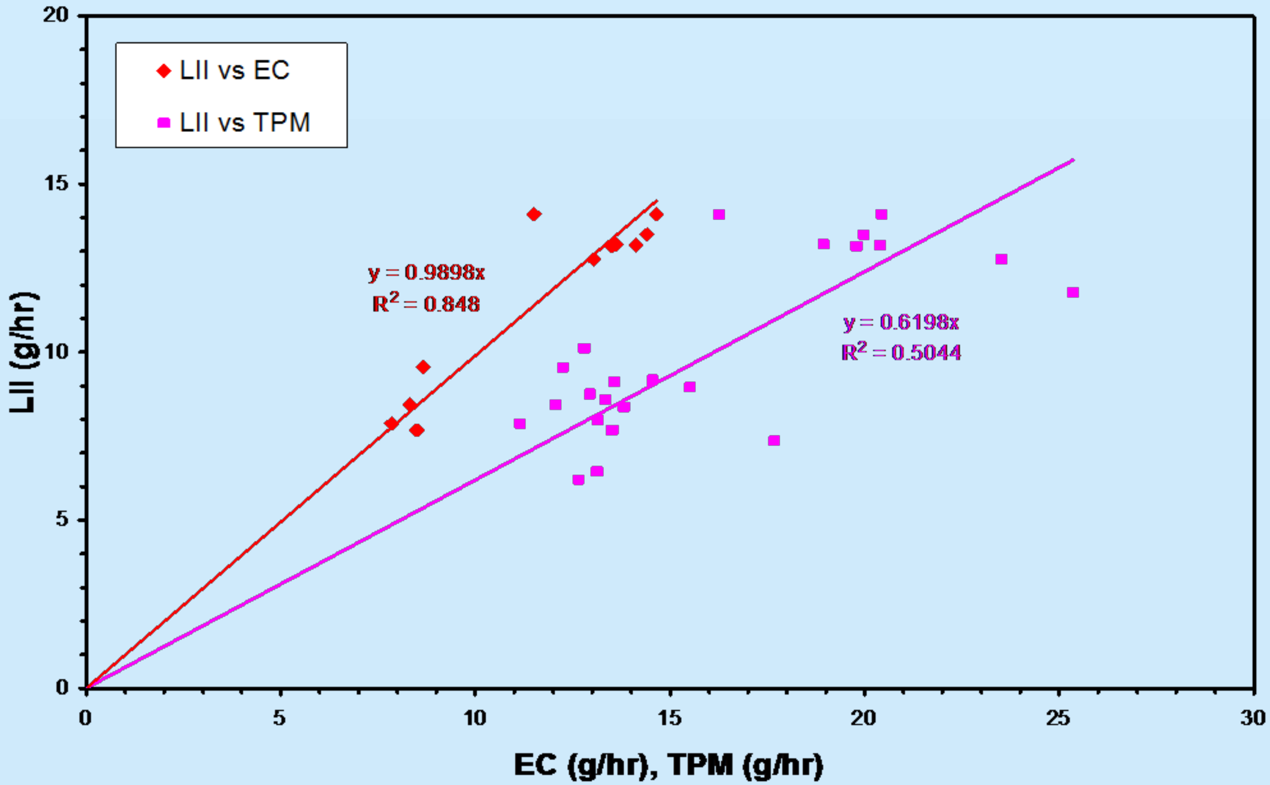
- AC-LII measurements of soot concentration compared to elemental carbon concentration determined by the NIOSH 5040 method
  - error bars represent single shot precision



[Smallwood, Ph. D. Thesis, Cranfield University, 2009]

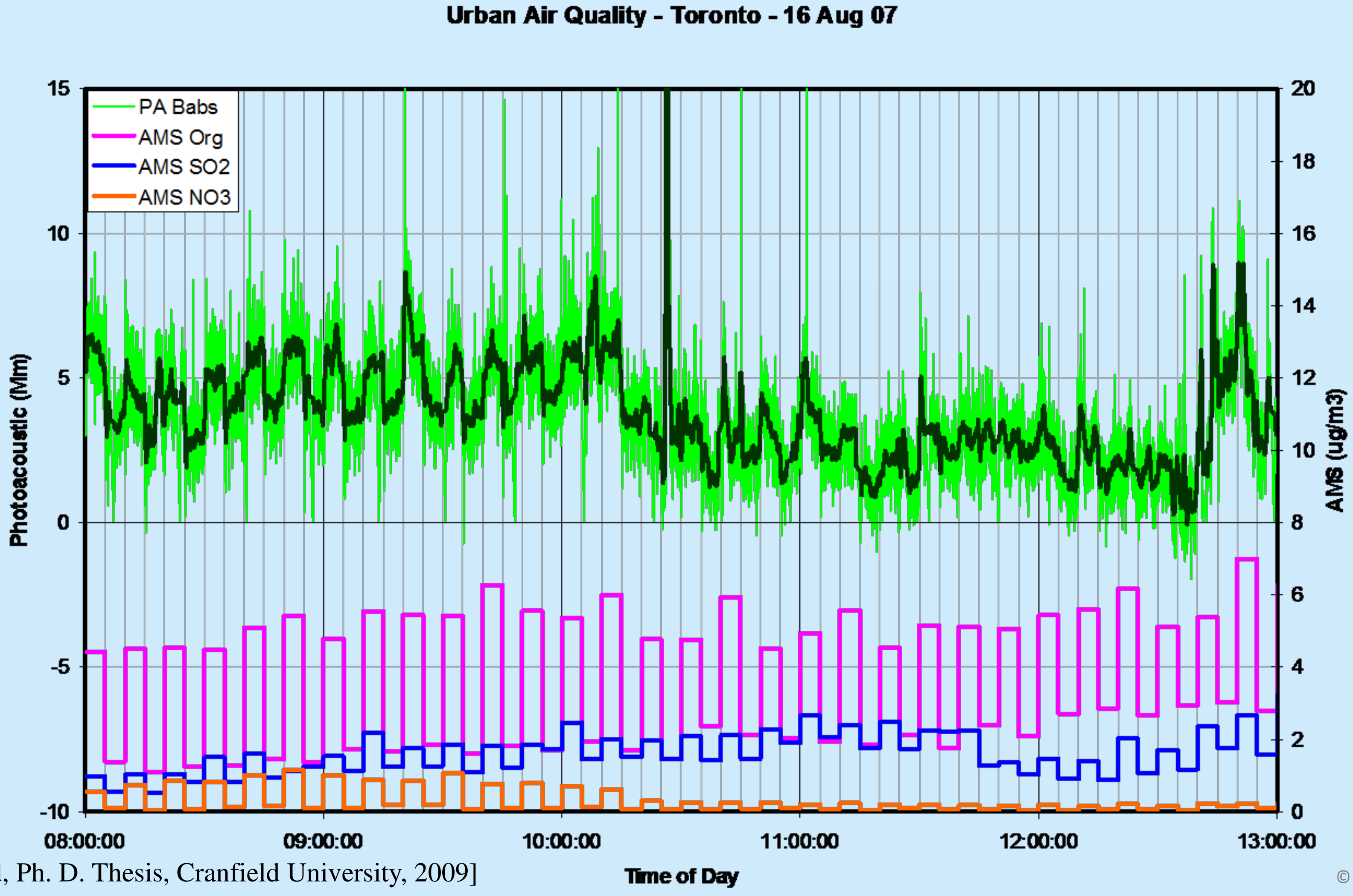
Comparison to Gravimetric: TPM & EC

- AC-LII measurements of soot emissions from a heavy-duty truck on a chassis dynamometer compared to total particulate matter (TPM) and nonvolatile particulate matter (EC) emissions



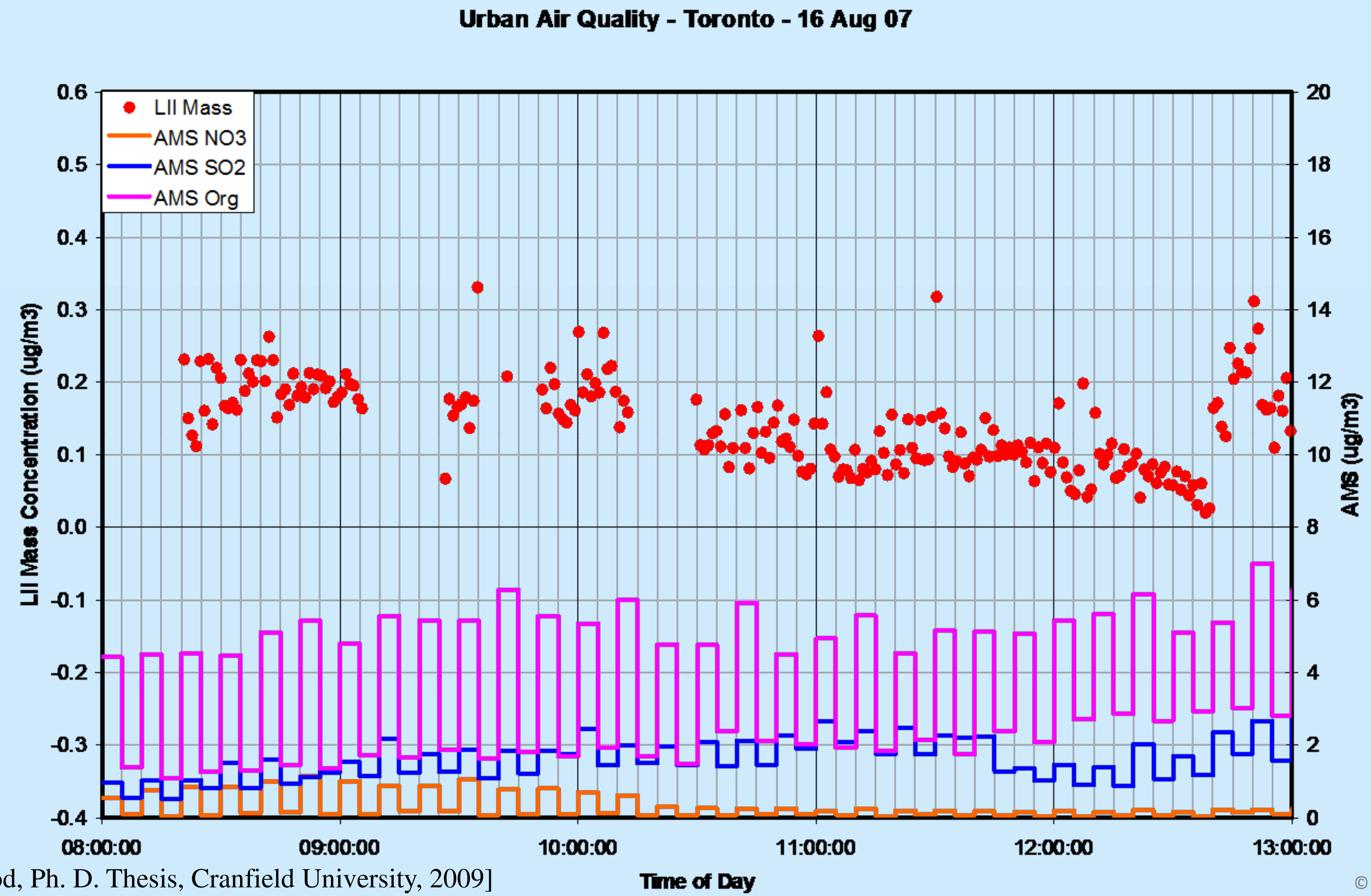
[Huai *et al.*, 16th CRC On-Road Vehicle Emissions Workshop, 2006]

Photoacoustic and AMS – Ambient and Denuded



[Smallwood, Ph. D. Thesis, Cranfield University, 2009]

LII and AMS – Ambient and Denuded



[Smallwood, Ph. D. Thesis, Cranfield University, 2009]

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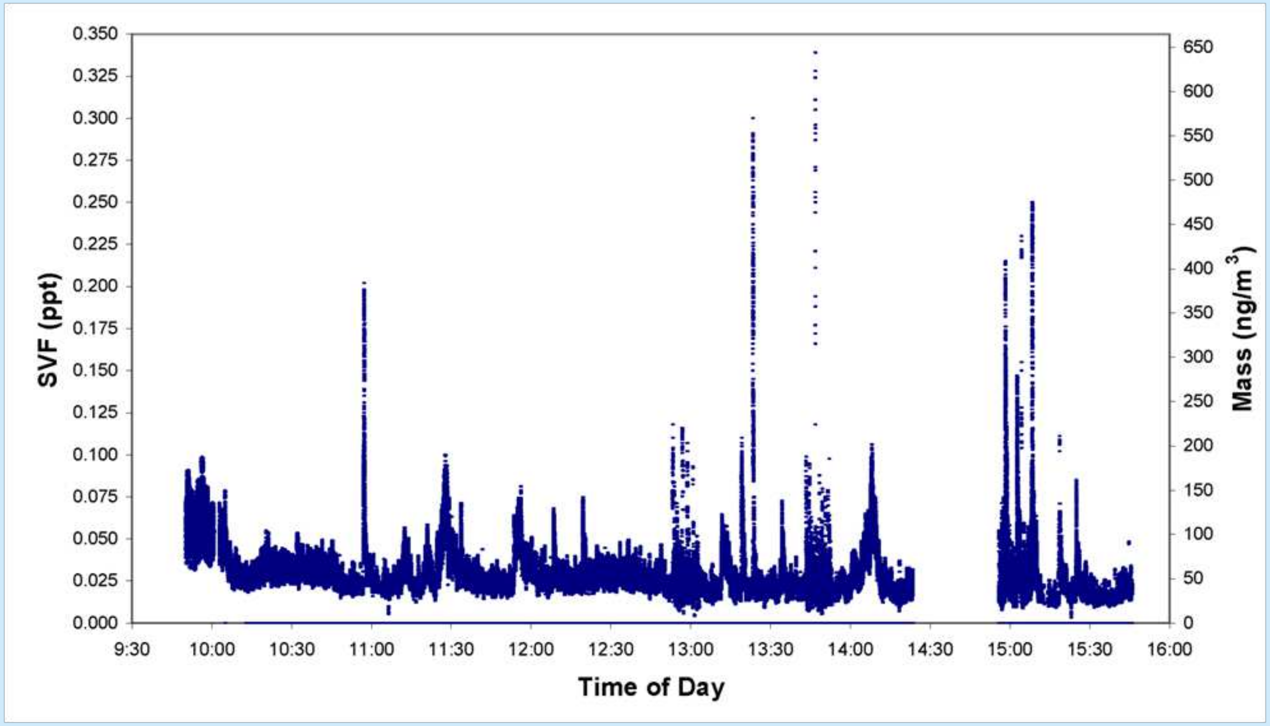
Experiment: High Sensitivity LII

- optimize all aspects of the laser-induced incandescence method
- use Lagrangian invariant principle to constrain design of collection optics and receiver
- resulting design was over 500 times more sensitive (ng/m^3 level)



[Smallwood, Ph. D. Thesis, Cranfield University, 2009]

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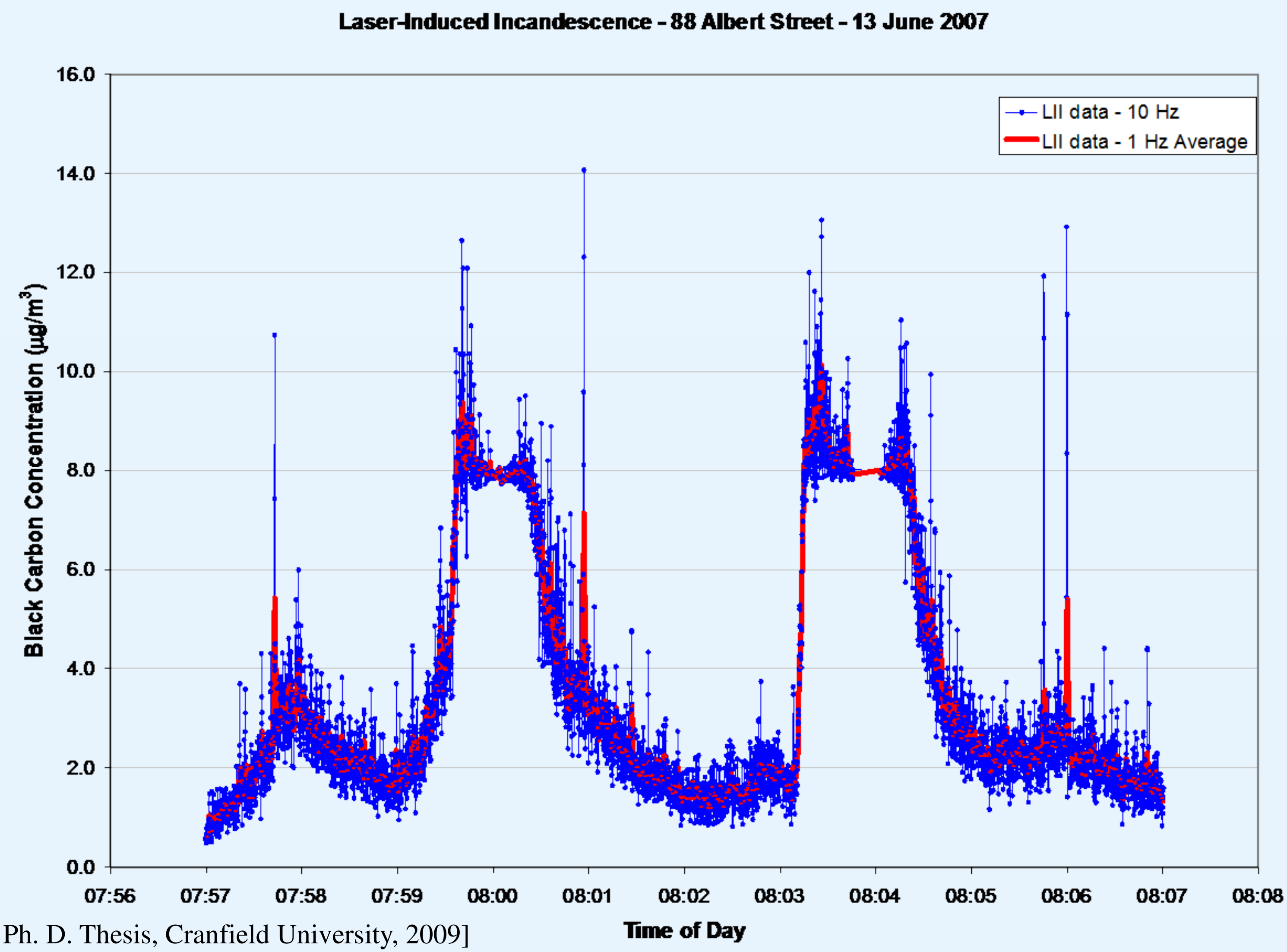
Urban Air Quality – High Sensitivity



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Summary

- a significant contribution has been made to improving the real-time measurement of nonvolatile particulate matter emissions
- autocompensating laser-induced incandescence (AC-LII) addresses some of the limitations of conventional LII, but also introduces new issues
- AC-LII was demonstrated to be highly repeatable, precise, selective, and linear with respect to some other particle measurement techniques
  - real-time measurements and high sensitivity also demonstrated
- LII however has shown uncertainty in the absolute concentration when compared to other methods

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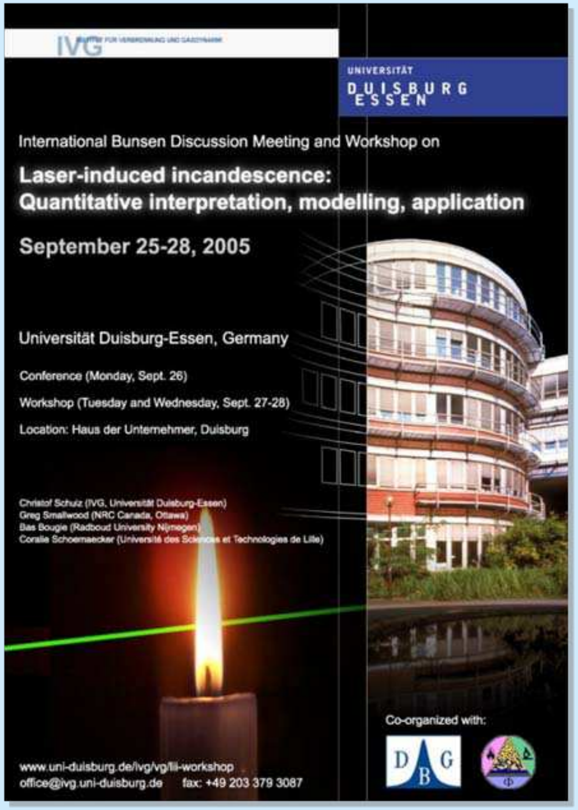
International Workshops on LII



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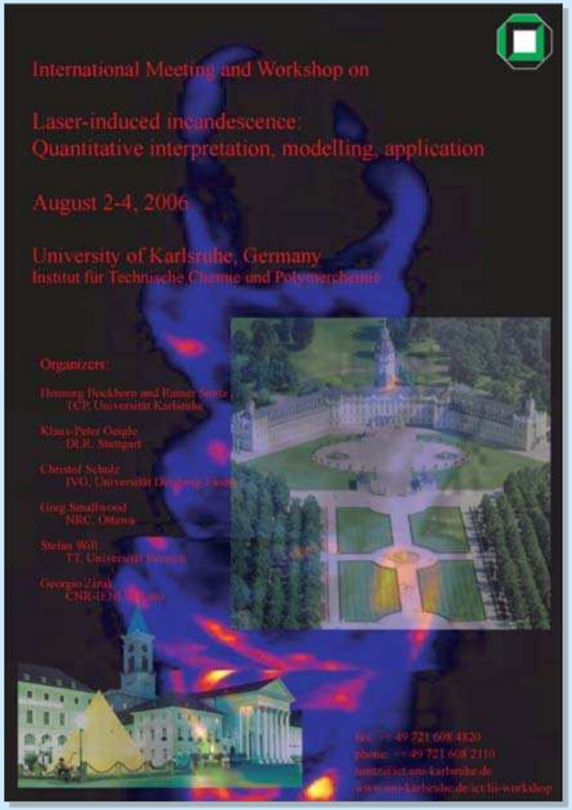

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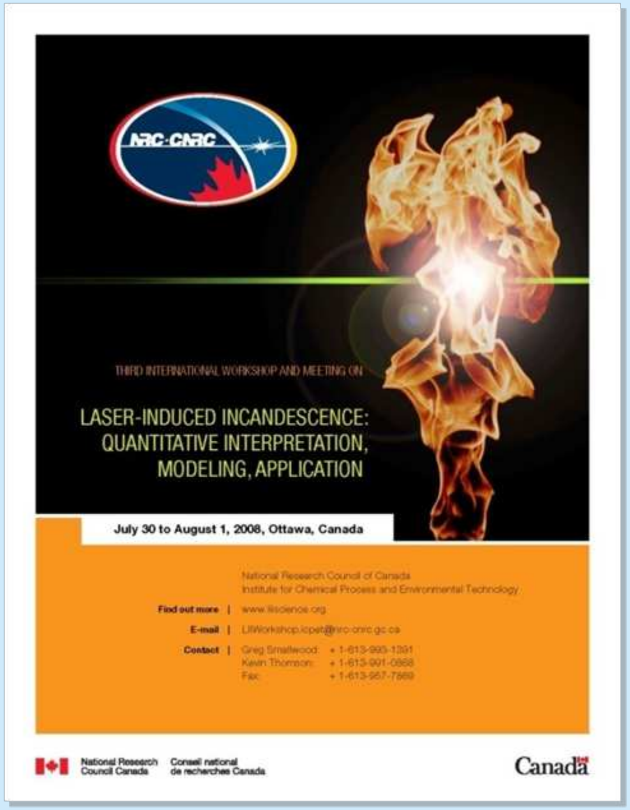

2008

4th International Workshop and Meeting on Laser-Induced Incandescence
19-20 April 2010, Lake Como, Italy

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International Workshops on LII

Fourth International Workshop on Laser-Induced Incandescence: Quantitative interpretation, modeling, and application

18 - 20 April 2010

Villa Monastero, Lake Como, Italy

Acknowledgements

- **NRC-ICPET**
  - Dave Snelling
  - Kevin Thomson
  - Fengshan Liu
  - Hongsheng Guo
  - Bob Sawchuk
  - Dan Clavel
  - Daniel Gareau
  - Reg Smith
  - Fazil Baksh
  - Ron Jerome
  - Dashan Wang
- **Carleton University**
  - Prof. Matt Johnson
  - Brian Crosland
  - James McEwen
- **Heriot-Watt University**
  - Prof. Doug Greenhalgh
  - Vivien Beyer
- **Universities of Waterloo**
  - Profs. Kyle Daun and James Sloan
- **British Columbia**
  - Profs. Ruth Signorell and Steve Rogak
- **Funding**
  - PERD AFTER Program
  - PERD P&E Program
  - PERD UPAIRI Progam
  - NRC/NSERC/BDC Nanotechnology Initiative Program
  - NRC/Helmholtz Program

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