Design Optimization for High Sensitivity Two-Color LII

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The development of a high sensitivity LII (HS-LII) experimental apparatus is discussed. The target was to improve the sensitivity of an auto-compensating laser induced incandescence (AC-LII) system employing two-wavelength pyrometry and low-fluence excitation by two orders of magnitude. The design considerations and implementation through a number of iterations of improvement are discussed. All aspects of the system were considered for optimization. The principle of the *Lagrangian invariant* was found to be most effective in designing the optical receiver subsystem. Application of the HS-LII instrument to measurements of soot concentration in the ambient atmosphere is presented, demonstrating a 500-fold improvement.

Introduction

The IPCC National Greenhouse Gas Inventories Programme is now focusing on emission estimation of aerosols relevant to climate change, and there is a need to measure black carbon levels in the atmosphere at microgram per cubic metre or lower mass concentrations. At the same time, emission standards for Diesel particulate matter (PM) are being lowered dramatically, resulting in the adoption of Diesel particulate filters (DPFs) by manufacturers, and there is a need to measure solid carbon levels in the exhaust and in dilution tunnels at microgram per cubic metre or lower mass concentrations.

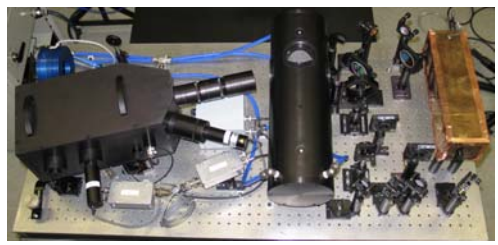
Development of high sensitivity LII to measure soot concentration at ambient levels for monitoring emissions from post-2007 Diesel engines, urban air quality, black carbon in atmosphere, and emissions from aircraft at altitude is required. The target for the high sensitivity LII system is a measurement limit of 0.05 ppt (~0.1 μg/m^3) or less.

Methodology

The current limit for measuring soot concentration with the NRC Mobile II AC-LII system is about 5 ppt (nearly 10 μg/m^3). The new design therefore requires a 100-fold improvement in sensitivity. Additionally, it is desirable to retain the low fluence and two-color pyrometry features of auto-compensating laser induced incandescence (AC-LII), and to make the system portable/mobile.

The approach was to optimize all aspects of the laser-induced incandescence method, including the laser, beam generation optics, sampling cell, receiver collection optics, receiver filters and dichroics, photodetectors, signal detection and digitization electronics, and the signal analysis software.

The *Lagrangian invariant* principle was applied to constrain the design of collection optics and receiver, as preservation of the Lagrangian invariant is essential for a lossless optical system. The concept is that the minimum product of the numeric aperture and aperture diameter cannot be improved upon, $L = NA1 \cdot R1 = NA2 \cdot R2$ = constant for optimum design. The probe volume diameter was based on the maximum practical Lagrangian invariant of receiver optical system, and the probe volume depth was set by desired laser fluence and maximum available laser energy. Optimization recommended that the crossing angle be minimized, but it was constrained by practicality.



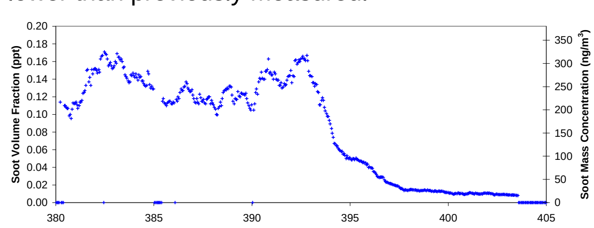
Optical layout of first generation high sensitivity system HS-LII-1

Results

A comparison of the calibration constants for a number of AC-LII systems shows that the target of a 100-fold improvement in sensitivity has been far exceeded.

System	λ_1 (nm)	RCS (W/m^3·sr·Volt)	Increase relative to Mobile II	λ_2 (nm)	RCS (W/m^3·sr·Volt)	Increase relative to Mobile II
Mobile II	397	$2.54 \cdot 10^{10}$	–	782	$4.22 \cdot 10^{10}$	–
HS-LII-1	445	$9.43 \cdot 10^{5}$	26,900x	746	$4.41 \cdot 10^{8}$	95.7x
HS-LII-2	445	$9.14 \cdot 10^{5}$	27,800x	753	$1.36 \cdot 10^{7}$	3100x
Artium LII-200	402	$4.39 \cdot 10^{8}$	57.9x	782	$4.62 \cdot 10^{9}$	9.13x
Artium ES-LII-200	447	$2.20 \cdot 10^{8}$	115x	829	$1.91 \cdot 10^{9}$	22.1x

Recent measurements of ambient air in Ottawa have demonstrated the ability to measure concentrations below 0.010 ppt ($<$20 ng/m^3), 500 times lower than previously measured.



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Proceedings of the 2nd International Discussion Meeting and Workshop: Laser-Induced Incandescence, Quantitative interpretation, modelling, application, Bad Herrenalb, Germany, 2-4 August, 2006