

Light scattering and extinction measurements combined with laser-induced incandescence for the real-time determination of soot mass absorption cross-section.

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Abstract

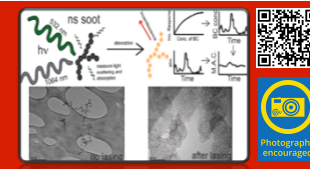
An aerosol albedometer was combined with laser-induced incandescence (LII) to achieve simultaneous measurements of aerosol scattering, extinction coefficient, and soot mass concentration. Frequency doubling of a Nd:YAG laser line resulted in a colinear beam of both $\lambda = 532$ and 1064 nm. The green beam was used to perform cavity ring-down spectroscopy (CRDS), with simultaneous measurements of scattering coefficient made through use of a reciprocal sphere nephelometer. The 1064 nm beam was selected and directed into a second integrating sphere and used for LII of light-absorbing kerosene lamp soot. Thermal denuder experiments showed the LII signals were not affected by the particle mixing state when laser peak power was 1.5–2.5 MW. The combined measurements of optical properties and soot mass concentration allowed determination of mass absorption cross section (M.A.C., m²/g) with 1 min time resolution when soot concentrations were in the low microgram per cubic meter range. Fresh kerosene nanosphere soot (ns-soot) exhibited a mean M.A.C and standard deviation of 9.3 ± 2.7 m²/g while limited measurements on dry ambient aerosol yielded an average of 8.2 ± 5.9 m²/g when soot was >0.25 $\mu\text{g}/\text{m}^3$. The method also detected increases in M.A.C. values associated with enhanced light absorption when polydisperse, laboratory-generated ns-soot particles were embedded within or coated with ammonium nitrate, ammonium sulfate, and glycerol. Glycerol coatings produced the largest fractional increase in M.A.C. (1.41-fold increase), while solid coatings of ammonium sulfate and ammonium nitrate produced increases of 1.10 and 1.06, respectively. Fresh, ns-soot did not exhibit increased M.A.C. at high relative humidity (RH); however, lab-generated soot coated with ammonium nitrate and held at 85% RH exhibited M.A.C. values nearly double the low-humidity case. The hybrid instrument for simultaneously tracking soot mass concentration and aerosol optical properties in real time is a valuable tool for probing enhanced absorption by soot at atmospherically relevant concentrations.



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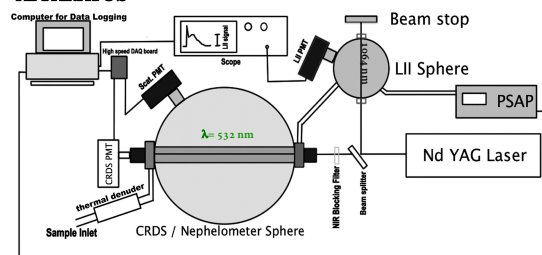
OVERVIEW: Laser - induced incandescence (LII) combined with an aerosol albedometer allowed simultaneous measurements of scattering & extinction coefficient, and BC mass concentration. The apparatus allowed determination of mass absorption cross section (M.A.C., m^2/g) with 60 s time resolution on dispersed aerosol.

Fresh kerosene soot exhibited a mean M.A.C of $9.3 \pm 2.7 \text{ m}^2/\text{g}$ while limited measurements on ambient aerosol at low R.H. yielded an average of $8.2 \pm 5.9 \text{ m}^2/\text{g}$ when indicated B.C. was $> 0.25 \mu\text{g}/\text{m}^3$.

Measurements of M.A.C. were made on laboratory-generated soot particles embedded within or coated with ammonium nitrate, ammonium sulfate, and glycerol. Glycerol coatings produced the largest fractional increase in M.A.C. (1.41 - fold increase), while solid coatings of ammonium sulfate and ammonium nitrate produced fractional increases of 1.10 and 1.06, respectively.

Fresh soot did not exhibit increased M.A.C. at high relative humidity (RH). However, soot coated with ammonium nitrate and held at 85% RH indicated M.A.C. values nearly double the low - humidity values.

APPARATUS



- The sphere nephelometer / cavity ring-down setup measures aerosol optical properties.
- The smaller sphere measures BC mass concentration with LII.
- The PSAP was used to calibrate the LII measurement using fresh kerosene soot.
- R-134a was used to calibrate the scattering measurement.
- This apparatus was developed to explore absorption enhancement by coatings on soot.
- We explored ambient measurements and internal mixing of several materials with soot

Figure 1. Experimental setup.

RESULTS

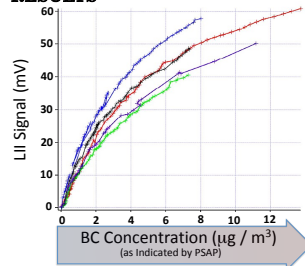


Figure 2. Variability of calibration responses of LII signal. A polynomial fit was used to relate LII signal to BC concentration.

The LII signal was related to BC mass concentration for fresh, uncoated, kerosene lamp soot. The lamp soot was directed to the LII chamber, followed by the particle soot absorption photometer (PSAP). The absorbance indicated on the green channel of the PSAP, and a mass absorption cross section of $13.1 \text{ m}^2/\text{g}$ was used for a reference measurement in calibration.

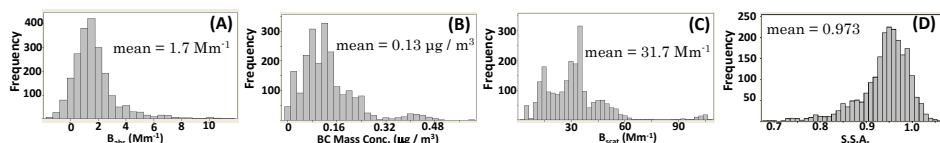


Figure 3. Summary of ambient monitoring data during sampling period at Lubbock, TX. (A) Histogram of observed aerosol absorption coefficient (b_{abs} , Mm^{-1}). (B) Histogram of observed BC mass concentrations ($\mu\text{g}/\text{m}^3$). (C) Histogram of observed aerosol scattering coefficient (b_{scat} , Mm^{-1}). (D) Histogram of observed SSA.

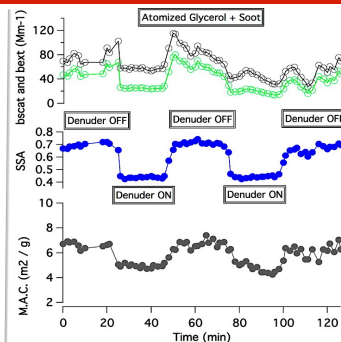
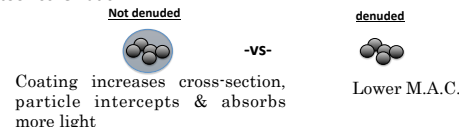


Figure 4. Plot of experimental data in time for lab-generated, atomized mixture of glycerol plus soot. At approximately 23 min, the thermal denuder (150-200 C) was turned on. At approximately 50 min the denuder was turned off. The cycle was repeated at approximately 75 and 100 min. As observed, the SSA dropped dramatically when the thermal denuder evaporated the glycerol coatings. The indicated M.A.C. also dropped when the coated soot was denuded indicating the glycerol coating caused an absorption enhancement. In the top trace the green data is scatter coefficient.

Cartoon schematic:



For laboratory experiments, lamp soot was collected and dispersed in water along with a coating material. The resulting mixture was atomized and particles dried. Three coating compositions were explored:

- Glycerol
- Ammonium nitrate
- Ammonium sulfate

expt	M.A.C. (coated/denuded) (m^2/g)	mean SSA (coated/denuded)	M.A.C. enhancement (coated/denuded)
atomized glycerol and soot	11.9/8.0	0.79/0.42	1.49
atomized glycerol and soot	14.3/10.3	0.78/0.39	1.39
atomized glycerol and soot	6.96/5.0	0.73/0.46	1.38
atomized glycerol and soot	6.6/4.8	0.70/0.44	1.36
mean enhancement observed ^a			1.41 \pm 0.06
atomized ammonium nitrate and soot	8.9/8.3	0.55/0.41	1.07
atomized ammonium nitrate and soot	8.9/9.1	0.91/0.39	0.98
atomized ammonium nitrate and soot	9.0/7.8	0.86/0.42	1.15
mean enhancement observed ^a			1.06 \pm 0.09
atomized ammonium sulfate and soot	9.12/7.93	0.65/0.45	1.15
atomized ammonium sulfate and soot	9.38/7.92	0.64/0.48	1.18
atomized ammonium sulfate and soot	13.9/13.3	0.63/0.48	1.04
atomized ammonium sulfate and soot	6.3/6.0	0.65/0.56	1.05
mean enhancement observed ^a			1.10 \pm 0.07
sample	mean M.A.C. \pm 1 σ (m^2/g)	mean SSA	M.A.C. enhancement
fresh kerosene soot	9.3 \pm 2.7	0.27	n.a.
dry ambient aerosol ^b	8.4 \pm 5.8	0.97	n.a.

^aCoating experiments were conducted on laboratory-generated, polydisperse, submicrometer aerosol. The large variability in M.A.C. observed between trials is believed to result from differences in soot samples used for analysis or for calibration. ^bAmbient measurements are for when BC mass concentration $> 0.25 \mu\text{g}/\text{m}^3$. This may bias data toward fresh soot plumes that have not been diluted/aged extensively. Also, relative humidity effects are not considered.

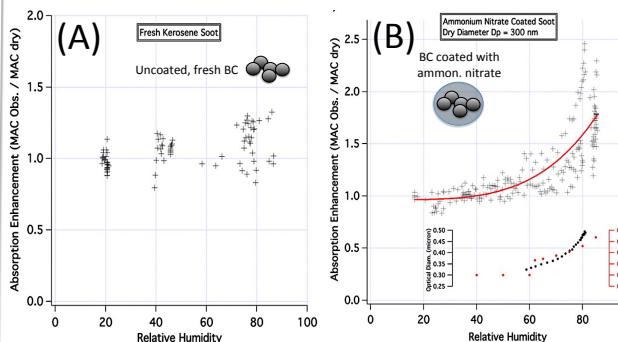


Figure 5. Effect of relative humidity on M.A.C. value for (A) fresh kerosene soot and (B) atomized BC internally mixed with ammonium nitrate. The ammonium nitrate makes the particle hygroscopic, and as the particles deliquesce the indicated optical absorption is enhanced at high RH. The inset in panel B plots the observed optical diameter and modeled particle diameter as a function of relative humidity. The particles in panel B were size-selected to 300 nm dry diameter using an electrostatic classifier prior to humidification.

CONCLUSIONS

- Coupling the aerosol albedometer with LII measurements provides a valuable tool for the research community for study of BC absorption and absorption enhancement due to coatings on BC.
- Significant absorption enhancement (1.41x) was observed for soot coated with glycerol, however, only marginal change in absorption ($< 10\%$) was observed for ammonium nitrate and ammonium sulfate coatings.
- Absorption enhancement of BC within internally mixed, hygroscopic particles at high R.H. remains an interesting, largely unexplored area of research that could prove to have important climate ramifications.