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Absorption coefficient measurements of particle-laden filters using laser heating: Validation with nigrosin

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ABSTRACT

A laser-heating technique, referred as the laser-driven thermal reactor, was used in conjunction with laser transmissivity measurements to determine the absorption coefficient of particle-laden substrates (e.g., quartz-fiber filters). The novelty of this approach is that it analyzes a wide variety of specific samples (not just filtered samples) and overcomes measurement issues (e.g., absorption enhancement) associated with other filter-based particle absorption techniques. The absorption coefficient was determined for nigrosin-laden, quartz-fiber filters and the effect of the filter on the absorption measurements was estimated when compared to the isolated nigrosin results. The isolated nigrosin absorption coefficient compared favorably with Lorenz-Mie calculations for an idealized polydispersion of spherical particles (based on a measured nigronsin/de-ionized water suspension size distribution) dispersed throughout a volume equivalent to that of the nigrosin-laden filter. To validate the approach. the absorption coefficient of a nigrosin/de-ionized water suspension was in good agreement with results obtained from an ultraviolet/visible spectrometer. In addition, the estimated imaginary part of the refractive index from the Lorenz–Mie calculations compared well with literature values and was used to estimate the absorption coefficient of optically opaque packed nigrosin.

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1. Introduction

1.1. Filter-based particle absorption enhancement

The class of filter-based particle absorption techniques (measuring the absorption characteristics of atmospheric aerosol particles collected on filters) include aethalometers, (e.g., Hansen et al. [1]), integrating plate/integrating sphere photometers (e.g., Fry et al. [2], Campbell et al. [3], Lawless et al. [4]), particle soot absorption photometers (e.g., Bond et al. [5]), and multi-angle absorption photometers (e.g., Petzold et al. [6]). Typically, the transmission of light passing through the filter is

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measured to determine the absorption coefficient in the Beer-Lambert law (or the extinction coefficient when scattering is considered significant). In turn, the absorption coefficient can be related to the imaginary part of the refractive index (depending on the state of the substance, i.e., if a bulk homogeneous substance or aerosolized powder). One important issue that has been reported extensively in the literature (e.g., Campbell et al. [3], Clarke [7], Bohren and Huffman [8], Petzold and Schönlinner [9], Cappa et al. [10]) is the effect of the filter substrate (and possibly the presence of nonabsorbing particles) on determining the correct value for the particle absorption coefficient, often referred to as absorption enhancement (e.g., Taha et al. [11]). This effect is attributed to absorption of backscattered laser light, which on first pass is not absorbed by the particles discretely distributed over and embedded within the fiber filter

Nomenclature

u	fitting parameter for exponential decaying function (see Fig. 5)
Δ	sample geometric cross-sectional area $[m^2]$
Δ*	sample geometric cross-sectional area [iii]
л D;	$\frac{dDSOLDallCC}{dDSOLDallCC}$
DI	Blot Nulliber $(= nL/\kappa_{sam})$
С	mass specific absorption cross section (m^2g^{-1})
С	solute/solvent molar concentration (mol L^{-1})
$\overline{C}_{\text{ext}}, \overline{C}_{\text{sca}},$	\overline{C}_{abs} mean extinction, scattering, and absorp-
	tion cross sections, respectively (m^2)
$c_p(T)$	specific heat capacity $(Ig^{-1}K^{-1})$
\overline{C}_{y}	mean volume fraction
ď	characteristic path length through the sample
	(m)
(dT/dt)	sample temperature derivative (K s^{-1})
D	diameter (m)
D_{σ}	geometric mean diameter (m)
D32	volume-to-surface area mean diameter
52	(Sauter mean diameter) (m)
$F(T,T_{\alpha})$	heat transfer term (W)
h	convection heat transfer coefficient
	$(W m^{-2} K^{-1})$
L	incident radiation intensity (W m ^{-2})
I_	transmitted radiation intensity (W m ^{-2})
L _o	absorbed radiation intensity (W m ^{-2})
	reflected radiation intensity (W m ^{-2})
$\frac{1}{k}$	imaginary part of the complex
ĸ	refractive index
k.	coverage factor
kcom	sample thermal conductivity (W m ^{-1} K ^{-1})
I	sample characteristic length (m)
m(t)	sample mass with respect to time (g)
m _e	sample final mass (g)
m.,	complex refractive index $(=n_r+ik)$
M	particle mass loading $(g m^{-2})$
n	sample number
n	real part of the complex refractive index
N	
••	number density (particles m^{-3})
Pr	number density (particles m ⁻³) incident radiation power (see Table 3) (W)
P _I P-	number density (particles m ⁻³) incident radiation power (see Table 3) (W) transmitted radiation power (see Table 3) (W)
P_I P_{τ} P(D)	number density (particles m ⁻³) incident radiation power (see Table 3) (W) transmitted radiation power (see Table 3) (W) probability distribution function
P_I P_{τ} $P(D)$ $a(T)$	number density (particles m ⁻³) incident radiation power (see Table 3) (W) transmitted radiation power (see Table 3) (W) probability distribution function specific heat release rate due to chemical
$P_I P_{\tau} P(D) q(T)$	number density (particles m ⁻³) incident radiation power (see Table 3) (W) transmitted radiation power (see Table 3) (W) probability distribution function specific heat release rate due to chemical reactions (W σ^{-1})
P_{T} P_{τ} $P(D)$ $q(T)$	number density (particles m ⁻³) incident radiation power (see Table 3) (W) transmitted radiation power (see Table 3) (W) probability distribution function specific heat release rate due to chemical reactions (W g ⁻¹)
P_{I} P_{τ} $P(D)$ $q(T)$ $Q_{\text{ext}}, Q_{\text{sc}}$	number density (particles m ⁻³) incident radiation power (see Table 3) (W) transmitted radiation power (see Table 3) (W) probability distribution function specific heat release rate due to chemical reactions (W g ⁻¹) _a , Q_{abs} differential extinction, scattering, and absorption efficiencies respectively
P_{I} P_{τ} $P(D)$ $q(T)$ Q_{ext}, Q_{sc} $\overline{Q}_{ext}, \overline{Q}_{sc}$	number density (particles m ⁻³) incident radiation power (see Table 3) (W) transmitted radiation power (see Table 3) (W) probability distribution function specific heat release rate due to chemical reactions (W g ⁻¹) a, Q_{abs} differential extinction, scattering, and absorption efficiencies, respectively \overline{Q}_{abs} mean extinction scattering and absorp-
P_{I} P_{τ} $P(D)$ $q(T)$ Q_{ext}, Q_{sc} $\overline{Q}_{ext}, \overline{Q}_{sca}$	number density (particles m ⁻³) incident radiation power (see Table 3) (W) transmitted radiation power (see Table 3) (W) probability distribution function specific heat release rate due to chemical reactions (W g ⁻¹) a, Q_{abs} differential extinction, scattering, and absorption efficiencies, respectively \overline{Q}_{abs} mean extinction, scattering, and absorp- tion efficiencies respectively
P_{I} P_{τ} $P(D)$ $q(T)$ Q_{ext}, Q_{sca} $\overline{Q}_{ext}, \overline{Q}_{sca}$ $R_{r}(T)$	number density (particles m ⁻³) incident radiation power (see Table 3) (W) transmitted radiation power (see Table 3) (W) probability distribution function specific heat release rate due to chemical reactions (W g ⁻¹) a, Q_{abs} differential extinction, scattering, and absorption efficiencies, respectively \overline{Q}_{abs} mean extinction, scattering, and absorp- tion efficiencies, respectively rate at which heat is transferred from the
P_{I} P_{τ} $P(D)$ $q(T)$ Q_{ext}, Q_{sc} $\overline{Q}_{ext}, \overline{Q}_{sca}$ $R_{1}(T_{r})$	number density (particles m ⁻³) incident radiation power (see Table 3) (W) transmitted radiation power (see Table 3) (W) probability distribution function specific heat release rate due to chemical reactions (W g ⁻¹) a, Q_{abs} differential extinction, scattering, and absorption efficiencies, respectively , \overline{Q}_{abs} mean extinction, scattering, and absorp- tion efficiencies, respectively rate at which heat is transferred from the reactor at temperature T_{a} (W)
P_{I} P_{τ} $P(D)$ $q(T)$ Q_{ext}, Q_{sc} $\overline{Q}_{ext}, \overline{Q}_{sca}$ $R_{1}(T_{r})$ $R_{2}(T)$	number density (particles m ⁻³) incident radiation power (see Table 3) (W) transmitted radiation power (see Table 3) (W) probability distribution function specific heat release rate due to chemical reactions (W g ⁻¹) a, Q_{abs} differential extinction, scattering, and absorption efficiencies, respectively , \overline{Q}_{abs} mean extinction, scattering, and absorp- tion efficiencies, respectively rate at which heat is transferred from the reactor at temperature T_r (W) rate of heat loss from the sample (W)
P_{I} P_{τ} $P(D)$ $q(T)$ Q_{ext}, Q_{sc} $\overline{Q}_{ext}, \overline{Q}_{sca}$ $R_{1}(T_{r})$ $R_{2}(T)$	number density (particles m ⁻³) incident radiation power (see Table 3) (W) transmitted radiation power (see Table 3) (W) probability distribution function specific heat release rate due to chemical reactions (W g ⁻¹) a, Q_{abs} differential extinction, scattering, and absorption efficiencies, respectively \overline{Q}_{abs} mean extinction, scattering, and absorp- tion efficiencies, respectively rate at which heat is transferred from the reactor at temperature T_r (W) rate of heat loss from the sample (W) standard deviation of the mean
P_{I} P_{τ} $P(D)$ $q(T)$ Q_{ext}, Q_{sc} $\overline{Q}_{ext}, \overline{Q}_{sca}$ $R_{1}(T_{r})$ $R_{2}(T)$ s t	number density (particles m ⁻³) incident radiation power (see Table 3) (W) transmitted radiation power (see Table 3) (W) probability distribution function specific heat release rate due to chemical reactions (W g ⁻¹) a, Q_{abs} differential extinction, scattering, and absorption efficiencies, respectively \overline{Q}_{abs} mean extinction, scattering, and absorp- tion efficiencies, respectively rate at which heat is transferred from the reactor at temperature T_r (W) rate of heat loss from the sample (W) standard deviation of the mean time (s)
P_{I} P_{τ} $P(D)$ $q(T)$ Q_{ext}, Q_{sc} $\overline{Q}_{ext}, \overline{Q}_{sca}$ $R_{1}(T_{r})$ $R_{2}(T)$ s t t t t	number density (particles m ⁻³) incident radiation power (see Table 3) (W) transmitted radiation power (see Table 3) (W) probability distribution function specific heat release rate due to chemical reactions (W g ⁻¹) a, Q_{abs} differential extinction, scattering, and absorption efficiencies, respectively \overline{Q}_{abs} mean extinction, scattering, and absorp- tion efficiencies, respectively rate at which heat is transferred from the reactor at temperature T_r (W) rate of heat loss from the sample (W) standard deviation of the mean time (s) time initiating Regimes 2 and 3 respectively
P_{I} P_{τ} $P(D)$ $q(T)$ Q_{ext}, Q_{sc} $\overline{Q}_{ext}, \overline{Q}_{sca}$ $R_{1}(T_{r})$ $R_{2}(T)$ s t t_{a}, t_{b}	number density (particles m ⁻³) incident radiation power (see Table 3) (W) transmitted radiation power (see Table 3) (W) probability distribution function specific heat release rate due to chemical reactions (W g ⁻¹) a, Q_{abs} differential extinction, scattering, and absorption efficiencies, respectively \overline{Q}_{abs} mean extinction, scattering, and absorp- tion efficiencies, respectively rate at which heat is transferred from the reactor at temperature T_r (W) rate of heat loss from the sample (W) standard deviation of the mean time (s) time initiating Regimes 2 and 3, respectively (see Fig. 5) (s)
P_{I} P_{τ} $P(D)$ $q(T)$ Q_{ext}, Q_{sc} $\overline{Q}_{ext}, \overline{Q}_{sca}$ $R_{1}(T_{r})$ $R_{2}(T)$ s t t_{a}, t_{b} T	number density (particles m ⁻³) incident radiation power (see Table 3) (W) transmitted radiation power (see Table 3) (W) probability distribution function specific heat release rate due to chemical reactions (W g ⁻¹) a, Q_{abs} differential extinction, scattering, and absorption efficiencies, respectively \overline{Q}_{abs} mean extinction, scattering, and absorp- tion efficiencies, respectively rate at which heat is transferred from the reactor at temperature T_r (W) rate of heat loss from the sample (W) standard deviation of the mean time (s) time initiating Regimes 2 and 3, respectively (see Fig. 5) (s) sample temperature (K)

$T_{\rm max}, T_o$	steady-state sample temperatures defined in
	Fig. 4 (K)

- T_r reactor temperature (K) u_c combined standard uncertainty
- *x* size parameter $(=\pi D/\lambda)$
- y absorption penetration depth (m)
- *z* transformation parameter

Greek symbols

α	sample absorption coefficient (m^{-1})	
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- $\beta(T, \lambda)$ spectral hemispherical absorptivity (fraction absorbed of incident radiation intensity)
- $egin{array}{ccc} eta_y & ext{differential} & ext{spectral} & ext{hemispherical} \\ & ext{absorptivity} & ext{} \end{array}$
- γ heat transfer parameter (W K⁻¹)
- $\Delta m(t)$ sample mass change after heating (g)
- ε sample extinction coefficient (m⁻¹)
- ε_m solute molar cross section (m² mol⁻¹)
- λ wavelength (m)
- ho spectral hemispherical reflectivity (fraction reflected of incident radiation intensity)
- ρ_p mass density (g m⁻³)
- σ sample scattering coefficient (m⁻¹)
- $\sigma_g \qquad \mbox{geometric mean standard deviation} \\ \tau \qquad \mbox{spectral hemispherical transmissivity (frac$ tion transmitted of incident radiation $intensity) }$
- τ^* temperature-dependent relaxation time (s)

Subscripts

abs	absorption
Reg#	regime
С	clean copper pan
ет	electromagnetic (see Table 2)
ext	extinction
f	clean filter
hb	laser heating beam
i	index $(=p, s)$
j	index (= <i>ext</i> , <i>sca</i> , <i>abs</i>)
п	isolated nigrosin
пс	nigrosin packed within the copper pan
nf	nigrosin-laden filter
р	isolated particles
ps	particle-laden substrate
pb	laser probe beam
S	clean substrate
	· · ·

sca scattering t thermal (see Table 2)

Superscripts

b fitting parameter for exponential decaying function (see Fig. 5)

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