



## Supplement of

## Multi-wavelength optical measurement to enhance thermal/optical analysis for carbonaceous aerosol

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Abbreviations/Acronyms	Descriptions
А	Filter sampling area
ATN, ATN $_{\lambda}$	Transmittance attenuation (general, at wavelength $\lambda$ )
BC, BC <sub>d</sub>	Black carbon (general, diesel elemental carbon equivalent)
BrC	Brown carbon
CV-RMSR	Coefficient of variance of root mean square residual
EC, ECR, ECR $_{\lambda}$ , ECT,	Elemental carbon (general, reflectance adjusted, reflectance
$ECT_{\lambda}$ , [EC]	adjusted at wavelength $\lambda$ , transmittance adjusted, transmittance
	adjusted at wavelength $\lambda$ , areal concentration on filter)
Не	Helium
IMPROVE	Interagency Monitoring of Protected Visual Environments
MAE, MAE <sub><math>\lambda</math></sub> , MAE <sub><math>\lambda</math>-EC</sub>	Mass absorption efficiency (general, at wavelength $\lambda$ , of
	elemental carbon at wavelength $\lambda$ )
$M_{\lambda}$	Multiple scattering effect coefficient at wavelength $\lambda$
Ne	Neon
OC, OCR, OCT, OCR $_{\lambda}$ ,	Organic carbon (general, reflectance adjusted, reflectance
$OCT_{\lambda}$	adjusted at wavelength $\lambda$ , transmittance adjusted, transmittance
	adjusted at wavelength $\lambda$ )
PM, PM <sub>2.5</sub>	Particulate matter (general, less 2.5 µm aerodynamic diameter)
POC	Pyrolyzed organic carbon
R, LR, LR <sub><math>\lambda</math></sub> , FR, FR <sub><math>\lambda</math></sub>	Reflectance (general, laser relative, laser relative at wavelength $\lambda$ ,
	filter absolute, filter absolute at wavelength $\lambda$ )
RD	Relative difference
T, LT, $LT_{\lambda}$ , FT, $FT_{\lambda}$ ,	Transmittance (general, laser relative, laser relative at wavelength
$FT_{\lambda,i}, FT_{\lambda,f}$	$\lambda$ , filter absolute, filter absolute at wavelength $\lambda$ , filter absolute at
	wavelength $\lambda$ before thermal analysis, filter absolute at
	wavelength $\lambda$ after thermal analysis)
TC	Total carbon
ТОА	Thermal/optical analysis
TSA	Thermal/spectral analysis
V	Filter sampling volume
$\alpha, \alpha(\lambda), \alpha_{\rm BC}, \alpha_{\rm BrC}$	Absorption Angström exponent (general, at wavelength $\lambda$ , of
	black carbon, of brown carbon)
b	Regression intercept
b <sub>abs</sub>	Absorption coefficient
m	Regression slope
р	T-test p value
$\beta_{\lambda}$	Shadowing effect coefficient at wavelength $\lambda$
λ	Wavelength
$ au_{a},  au_{a,\lambda},  au_{a,\lambda,BC},  au_{a,\lambda,BrC}$	Absorption optical depth (general, at wavelength $\lambda$ , at wavelength
	$\lambda$ due to black carbon, at wavelength $\lambda$ due to brown carbon)

Table S1. List of abbreviations and acronyms used in the paper.



**Figure S1.** Configuration of (a) the multi-wavelength module and (b) interface to a thermal/optical carbon Analyzer. Example of typical thermal/spectral analysis (TSA) thermogram for: (c) Fresno ambient and (d) diesel exhaust samples. Laser reflectance (LR<sub> $\lambda$ </sub>, solid lines) and transmittance (LT<sub> $\lambda$ </sub>, dashed lines) at 7 wavelengths (405, 450, 532, 635, 780, 808, and 980 nm) are reported in millivolts (mV) as detected by the photodiodes. Absolute filter reflectance and transmittance (FR<sub> $\lambda$ </sub>/FT<sub> $\lambda$ </sub>) for (c) and (d) are shown in (e) and (f), respectively, after calibrating LR<sub> $\lambda$ </sub> and LT<sub> $\lambda$ </sub> against reflectance and transmittance measurements by the Lambda 35 integrating-sphere spectrometer. FR<sub> $\lambda$ </sub> is between 0.1 and 1 (i.e., 100%) while FT<sub> $\lambda$ </sub> is generally <0.1 (10%). The temperature steps follow the IMPROVE\_A protocol: OC1 (140°C), OC2 (280°C), OC3 (480°C), OC4 (580°C), EC1 (580°C), EC2 (740°C), and EC3 (840°C) with oxygen introduced at the beginning of EC1. The last carbon peak corresponds to the internal methane standard.



**Figure S2.** (a) Fresno Supersite  $PM_{2.5}$  samples acquired from year 2003 serving as transfer standards for calibrating the reflectance and transmittance measurement of a multi-wavelength carbon analyzer. Also shown are the spectral (b) reflectance and (c) transmittance of these samples as quantified by an integrating-sphere spectrometer (Lambda 35, Perkin Elmer, Massachusetts, USA). Dashed lines correspond to wavelengths of the thermal/spectral analysis by the multi-wavelength carbon analyzer.



**Figure S3.** Calibration of  $ATN_{\lambda}$  of the 7-wavelength carbon analyzer with EC loading ([EC]) measured during the EC2 step of IMPROVE\_A analysis of a diesel exhaust sample (CIFQ074, acquired from the Gasoline/Diesel Split Study). Regressions are based on Eq. (6).



**Figure S4.** Measured  $\tau_{a,\lambda}$  (450, 635, and 808 nm are shown) compared with  $\tau_{a,\lambda}$  fitted from Eq. (7) assuming a two-component model. All samples in this study are included.



**Figure S5.** Decomposition of measured absorption optical depth  $(\tau_{a,\lambda})$  from (a) Fresno ambient and (b) Reno wildfire samples into the BC and BrC contributions based on their distinct spectral dependence of light absorption. See text for details.