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Measuring light absorption by freshly emitted organic aerosols: optical artifacts in traditional solvent-extraction-based methods

Nishit J. Shetty et al.

Correspondence to: Rajan K. Chakrabarty (chakrabarty@wustl.edu)

S1. Experimental methods

The combusted fuels along with the organic carbon (OC) to total carbon (TC) mass ratios, single scattering albedo (SSA) values, and absorption Ångström exponents (AÅE), are listed in Table S1. Collected filters were shipped to Sunset Labs (Tigard, OR) in covered aluminum lined containers with ice packs to prevent losses of condensed organics.

Table S1: List of the fuels burnt in this study along with the number of filter samples collected for each fuel type. The optical properties along with the range of observed OC/TC ratios are also given

Fuel	Number of burns	OC/TC ratio	SSA		
			375	405	1047
Cattle dung	6	1	0.85 – 0.87	0.94 – 0.95	0.98
Sage	9	0.55 – 1	0.55 – 0.87	0.7 – 0.95	0.85 – 0.98
Grass	4	0.77 – 0.99	0.72 – 0.76	0.84 – 0.89	0.95 – 0.97
Ponderosa pine	3	0.6 – 0.95	0.61 – 0.83	0.73 – 0.9	0.59 – 0.91
Lodgepole pine	4	0.9 – 1	0.83 – 0.87	0.91 – 0.95	0.95 – 0.98
Douglas fir	2	0.76 – 0.99	0.81 – 0.83	0.89 – 0.94	0.9 – 0.97

Table S2: List containing descriptions of filter use in each burn along with the measured OC/TC mass ratio.

Fuel	Burn	Filters collected	Filter for EC/OC	Filter for extraction	OC/TC ratio
Dung	BrC_1	1	1	0	1
	BrC_2	1	1	0	1
	BrC_3	1	0	1	N/A (assumed 1)*
	BrC_4	2	1	1	1
	BrC_5	2	1	1	1
	BrC_6	2	1	1	1
Sage	BrC_7	2	1	1	1
	BrC_8	2	1	1	1
	BrC_9	2	1	1	0.97
	BrC_10	2	2	0	0.78
	BrC_11	2	1	1	0.79
	BrC_12	2	1	1	0.55
	BrC_13	2	1	1	0.79
	BrC_14	2	2	0	0.79-0.8
	BrC_15	3	2	1	0.7-0.72
Lodgepole Pine	BrC_16	2	1	1	0.98
	BrC_17	2	1	1	0.98
	BrC_18	1	1	0	1
	BrC_19	2	1	1	0.9
Ponderosa Pine	BrC_20	2	1	1	0.95
	BrC_21	2	1	1	0.7

	BrC_22	2	1	1	0.6
Grass	BrC_23	2	1	1	0.99
	BrC_24	3	2	1	0.77-0.8
	BrC_25	2	1	1	0.77
	BrC_26	2	1	1	0.78
Douglas fir	BrC_27	2	2	0	0.76-0.88
	BrC_28	3	3	0	0.9-0.99
Total filters		55	34	21	

*All dung burns had no visible flaming combustion phase and smoldered throughout the combustion process

Table S3: Particle phase AÅE for entire aerosol obtained from IPN data at 375, 405 and 1047 nm.

Fuel	OC/TC ratio	$AÅE_{375\text{-}1047}$	$AÅE_{405\text{-}1047}$
Dung	1	2.96 ± 0.18	2.29 ± 0.16
	1	3.05 ± 0.04	2.3 ± 0.13
	1	2.8 ± 0.05	2.06 ± 0.05
	1	2.96 ± 0.17	2.24 ± 0.19
Sage	1	4.31 ± 0.6	3.83 ± 0.65
	1	3.71 ± 0.14	2.99 ± 0.19
	0.97	3.53 ± 0.21	3.03 ± 0.19
	0.79	2.52 ± 0.15	2.21 ± 0.06
	0.79	2.5 ± 0.2	2.24 ± 0.2
	0.71	1.92 ± 0.16	1.6 ± 0.18
	0.55	1.42 ± 0.07	1.26 ± 0.08
Pine	0.98	3.32 ± 0.18	2.96 ± 0.2
	0.98	3.14 ± 0.15	2.58 ± 0.16
	0.95	3.65 ± 0.24	2.91 ± 0.22
	0.9	2.75 ± 0.25	2.43 ± 0.29
	0.7	1.25 ± 0.18	1.07 ± 0.16
	0.6	1.28 ± 0.18	1.11 ± 0.17
Grass	0.99	3.14 ± 0.16	2.68 ± 0.07
	0.78	3.13 ± 0.19	2.7 ± 0.24
	0.78	2.7 ± 0.15	2.46 ± 0.16
	0.77	3 ± 0.16	2.66 ± 0.21

S2. Monte Carlo Simulation

The pseudocode for determining the errors using the Monte Carlo simulation is as below:

for i from 1 to 10000

$x_1(i) = PDF\ (\mu_1, \sigma_1)$

.

$x_n(i) = PDF\ (\mu_n, \sigma_n)$

% PDF (μ_n, σ_n) denotes probability distribution function with mean value of μ_n and a standard deviation of σ_n%

%x_n denotes the variables with uncertainties in these experiments which include: AÅE, Extraction volume, Absorbance, Absorption coefficients and pump flowrate%

$b_{abs,OA}(i) = b_{abs,IPN}(i) - b_{abs,BC}(i)*(1047/\lambda)^{(AÅE(i))}$

$b_{abs,sol}(i) = A(i)*Volume\ extract(i)/Volume\ sampled(i)*ln(10)$

end

$Bias = mean(b_{abs,OA}/b_{abs,sol})$

$Error = standard\ deviation(b_{abs,OA}/b_{abs,sol})$

Table S4: Typical values for mean and standard deviations used for different variables in the Monte Carlo simulation

	AÅE	Extract Volume (ml)	Absorbance	b_{abs} (Mm^{-1})	Pump flowrate (L/min)
μ	1	2-5	UV-Vis Measured	IPN Measured	5
σ	0.05	0.1*	0.002**	IPN calculated	0.06

* Based on volume measurements pre and post extraction

** Based on specifications of the UV-Vis instrument

S3. Number concentration and size distribution calculations

To perform Mie theory calculations, a proxy size distribution for the organics dissolved in water was obtained using size distribution data from the scanning mobility particle sizer (SMPS) and the total organic carbon (TOC) analyzer. The proxy size distribution was assumed to follow a lognormal distribution with the geometric mean size and standard deviation assumed to be the same as that of the distribution measured by the SMPS.

The lognormal size distribution of the measured aerosol can be written as:

$$n_d(d_p) = \frac{N_{tot}}{2\pi^{1/2}\, d_p \ln(\sigma_g)} . \exp\left[-\frac{(ln\, d_p - ln(d_{pg}))^2}{2\, ln^2(\sigma_g)}\right] \quad \text{(S1)}$$

Here, d_p is the mobility size of particles measured by the SMPS, d_{pg} is the geometric mean size of the particles in the distribution, σ_g is the geometric standard deviation, and N_{tot} is the total number concentration of particles in the aerosol stream . The frequency distribution function for the same would simply be:

$$f(d_p) = \frac{n(d_p)}{N_{tot}} \quad \text{(S2)}$$

Thus, the mass concentration for dissolved organics can be given as:

$$M = N_{tot,sol} * \sum_{d_{p,1}}^{d_{p,n}} \rho * \frac{\pi}{6} * d_{p,x}^3 * f(d_{p,x}) * (d_{p,x+1} - d_{p,x}) \quad \text{(S3)}$$

Here, M is the mass concentration of organics dissolved in water obtained from the TOC analyzer, ρ is the density of dissolved organics which is 1.6 g/cc, and $N_{tot,sol}$ is the number concentration of the proxy size distribution for the dissolved organics. Eq. S3 can be rearranged to find a value for $N_{tot,sol}$, and this value can then be used to generate the substitute size distribution for dissolved organics using Eq. S1. The surrogate size distribution was then used as an input for the Mie calculations.

S4. Performance comparison of SSA and OC/TC parametrizations

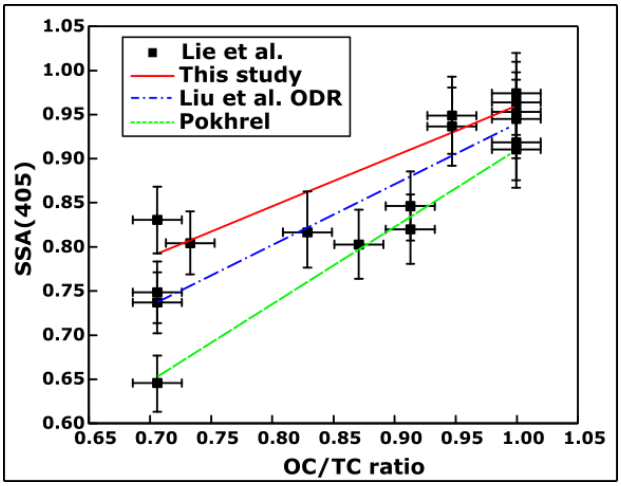

Fig. S1: Comparison of parametrizations obtained in this study with those provided by Pokhrel et al. (2016). The blue perforated line represents the ODR fit for data from Liu et al. (2014). The solid red line and dashed green line represent the parametrizations obtained during this study and Pokhrel at al. respectively.

The parametrizations determined during this study can be used in conjunction with those obtained by Pokhrel et al. (2016) to obtain reasonable estimates within which the SSA of BB aerosols might lie given the OC/TC ratio is known *a priori*.

S5. Comparison of particle and solvent phase AÅE

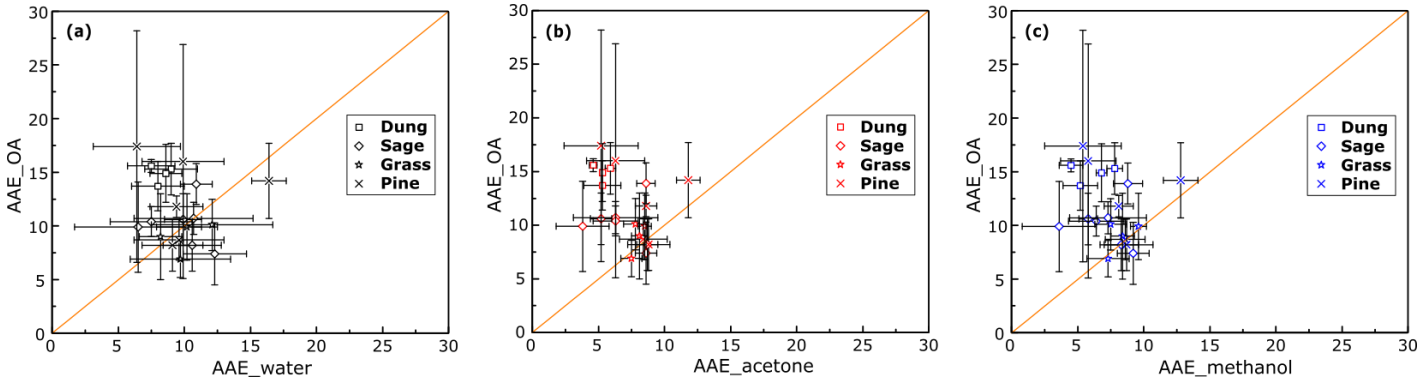

Fig. S2: Comparison of the particle phase AÅE with corresponding values for particles extracted in (a) water, (b) acetone, and (c) methanol along with a 1:1 line in orange

The AÅE for the particle phase appeared to be close to or higher than the corresponding values in the solvent phase and this was verified with a t-test as well.