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Supplement of

Measurement and modeling of the multiwavelength optical properties of uncoated flame-generated soot

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Table S1. Summary of terminology

Terminology	*Definition*
Nascent soot	Soot particles that were not subject to additional thermal or chemical processing
Nascent-denuded soot	Soot particles that were heated to 270 °C for ~5 seconds in a thermal denuder
Coated-denuded soot	Soot particles that were first coated with either DOS, H_2SO_4 or SOA and subsequently heated to 270 °C for ~5 seconds in a thermal denuder
Forward-coating experiment	Experiments in which the particles were size selected, then the monodisperse particles were coated, and finally thermally denuded
Reverse-coating experiment	Experiments in which the polydisperse particle distribution was first coated, then the particles were size selected, and finally thermally denuded

Table S2. Summary of instrumentation used in BC2, BC3, BC3+, and BC4.

Instrument	Manufacturer	Measurement	Study
Centrifugal particle mass analyzer (CPMA)	Cambustion	Per particle mass in fg $particle^{-1}$	All Studies
Scanning mobility particle sizer (SMPS)	TSI	Particle mobility size distributions	All Studies
Condensation particle counter (CPC)	TSI	Number concentration of particles in particles cm^{-3}	All Studies
3 wavelength photoacoustic soot spectrometer (PASS-3) equipped with scattering sensor	Droplet Measurement Technologies	Aerosol absorption and extinction coefficients (b_{abs} and b_{ext}) at λ = 405 nm, 532 nm, and 781 nm in Mm^{-1}	BC2
NOAA photoacoustic spectrometer (PAS)	Custom-built	b_{ext} at λ = 532 nm in Mm^{-1}	BC2
NOAA cavity ringdown spectrometer (CRD)	Custom-built	b_{abs} at λ = 532 nm in Mm^{-1}	BC2
UC Davis CRD-PAS	Custom-built	b_{abs} and b_{ext} in Mm^{-1} at λ = 405 nm and 532 nm	BC3, BC3+, and BC4
Cavity attenuated phase shift single scatter albedo spectrometer (CAPS PM_{SSA})	Aerodyne Research	Scattering and absorption coefficients (b_{sca} and b_{ext}) in Mm^{-1} at λ = 630 nm	BC3, BC3+, and BC4
Soot particle aerosol mass spectrometer (SP-AMS)	Aerodyne Research	Chemical and physical properties of black carbon	BC2, BC3, and BC4
Compact time-of-flight aerosol mass spectrometer (c-ToF-AMS)	Aerodyne Research	Chemical composition of non-refractory aerosol	BC3

Table S3. Observed mean *MAC*s for the methane flame, averaged across BC3, BC3+ and BC4. Uncertainties are reported as 1σ standard deviations over the measurements

λ (nm)	*MAC* (m^2 g^{-1}) (all points)	*AAE*	*MAC* (m^2 g^{-1}) ($x > 0.9$)	*AAE*	*MAC* (m^2 g^{-1}) ($x < 0.9$)	*AAE*
405	11.8 ± 1.7	1.16*	12.1 ± 1.4	1.04	11.1 ± 2.0	1.20
532	8.6 ± 1.3	1.42#	9.1 ± 1.1	1.21	8.0 ± 1.4	1.35
630	6.3 ± 1.2	1.84+	7.1 ± 1.1	1.47	6.1 ± 1.2	1.60

* 405-532 nm; # 405-630 nm; + 532-630 nm

Table S4. Minimum and maximum binned *MAC*s for all flame types.

		Smallest *x* bin			**Largest *x* bin**		
λ (nm)	**Instrument**	**Size parameter**	**# Data Points**	***MAC*** (m^2 g^{-1})	**Size parameter**	**# Data Points**	***MAC*** (m^2 g^{-1})
		Methane Diffusion Flame (BC3, BC3+ & BC4)					
405	UCD CRD-PAS	0.54	13	9.7 ± 3.2	1.62	10	11.2 ± 1.0
532	UCD CRD-PAS	0.36	5	5.4 ± 0.8	1.26	11	8.7± 1.0
630	CAPS PM_{SSA}	0.36	14	4.4 ± 0.8	1.08	8	7±0.67
		Ethylene Premixed Flame Denuded (BC2)					
405	PASS-3	0.72	7	10.8± 1.5	1.08	5	11.5±1.3
532	PASS-3	0.36	5	4.5±2.1	0.9	6	9.0±0.7
532	PASS-3	0.54	8	6.9±1.0	0.9	6	7.6±0.9
781	PASS-3	0.36	9	3.7±0.6	0.54	20	3.7±0.6
		Ethylene Premixed Flame Nascent (BC2)					
405	PASS-3	0.36	6	5.6±1.9	1.08	7	10.54±2.2
532	NOAA CRD-PAS	0.36	5	4.1±0.9	0.9	6	6.9±1.5
532	PASS-3	0.54	6	5.5±1.0	0.9	6	6.4 ± 1.2
781	PASS-3	0.36	9	2.3±0.5	0.54	14	2.8± 0.5

Table S5. Observed mean *MAC*s from the BC3+ ethylene flame.

λ (nm)	*MAC* (m^2 g^{-1}) $d_{p,VED}$ 50-70 nm	*MAC* (m^2 g^{-1}) $d_{p,VED}$ 70-130 nm	*MAC* (m^2 g^{-1}) $d_{p,VED}$ > 130 nm
405	8.2 ± 2.6	9.5 ± 1.9	9.9 ± 1.7
532	4.8 ± 0.80	6.2 ± 1.0	5.7 ± 0.90
630	2.9 ± 0.90	3.7 ± 0.5	4.0 ± 0.7

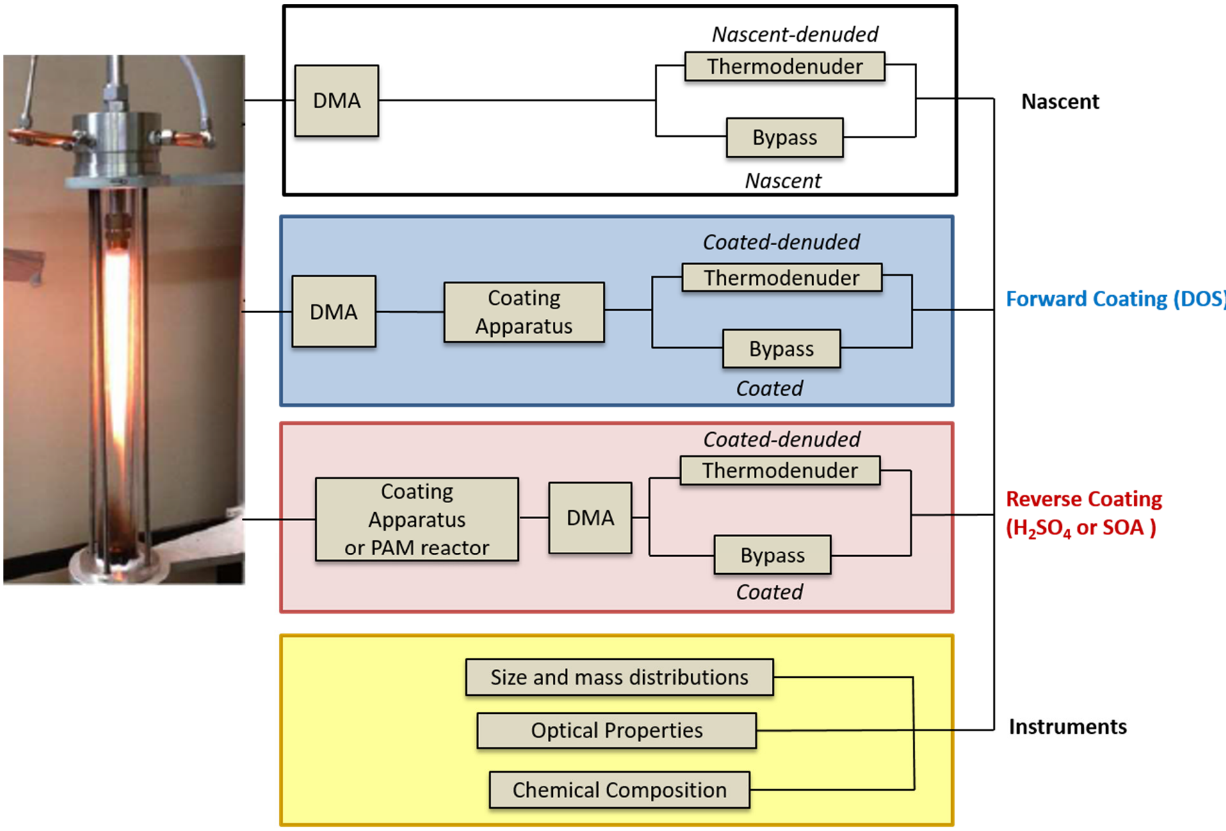

Figure S1. General experimental set-up for BC2, BC3, BC3+, and BC4, for the methane diffusion flame. Note that during BC4, particles were mass-selected with a CPMA, in addition to being size-selected.

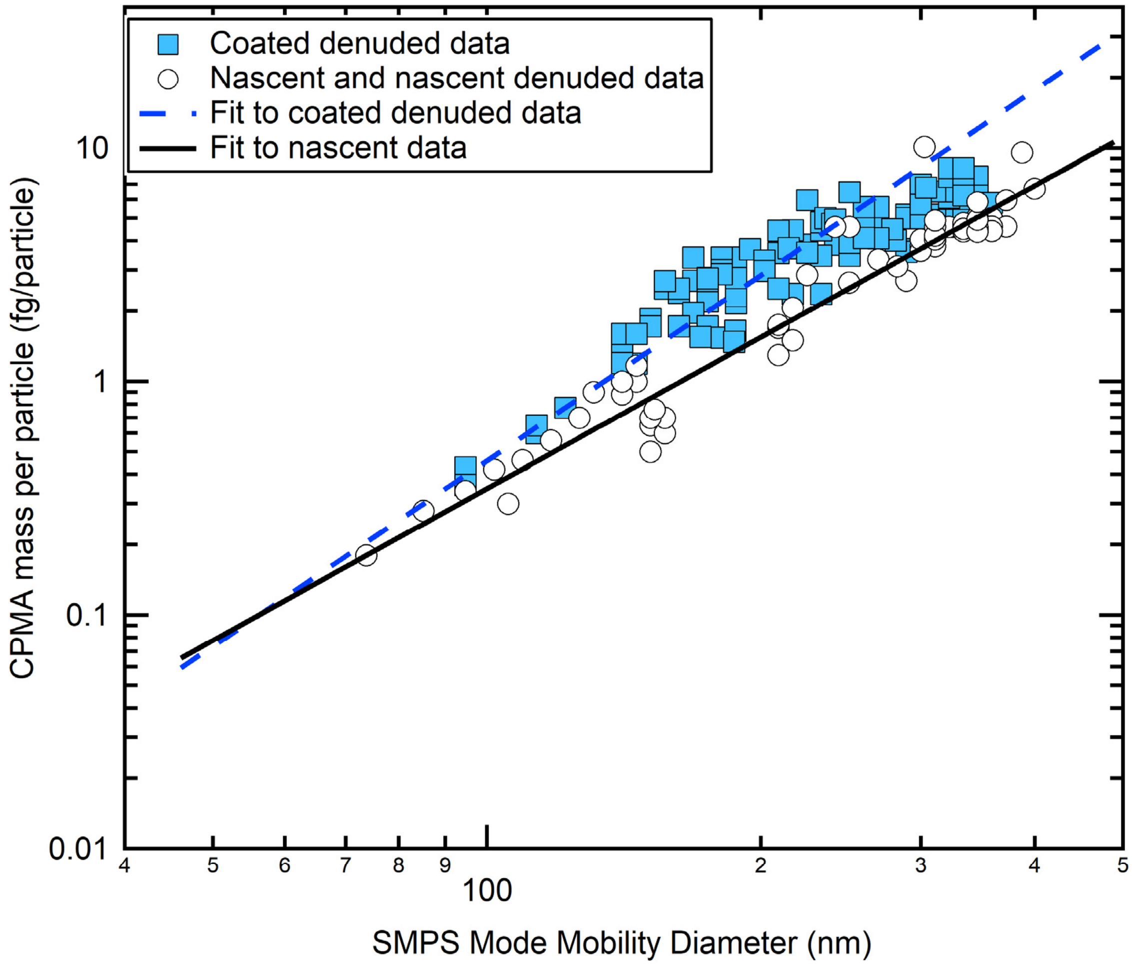

Figure S2. Observations and fits of CPMA mass per particle as a function of the SMPS mode number-weighted mobility diameter for methane diffusion flame soot particles. The blue squares show coated denuded observations and the dashed blue line is the power law fit to the coated-denuded data. The white circles are nascent observations and the black line is the power law fit to the nascent observations. Note that the fits to the coated-denuded data are for particles with mobility diameter < 260 nm since the large particles generally have thinner coatings and are therefore more fractal-like. The data were fit to a power law function (see eqn. 8 in the main text). For nascent soot, fit parameters C and $D_{f,m}$ were 1.70 x 10^{-5} and 2.12, respectively, and for coated-denuded soot, fit parameters C and $D_{f,m}$ were 2.44 x 10^{-6} and 2.64, respectively.

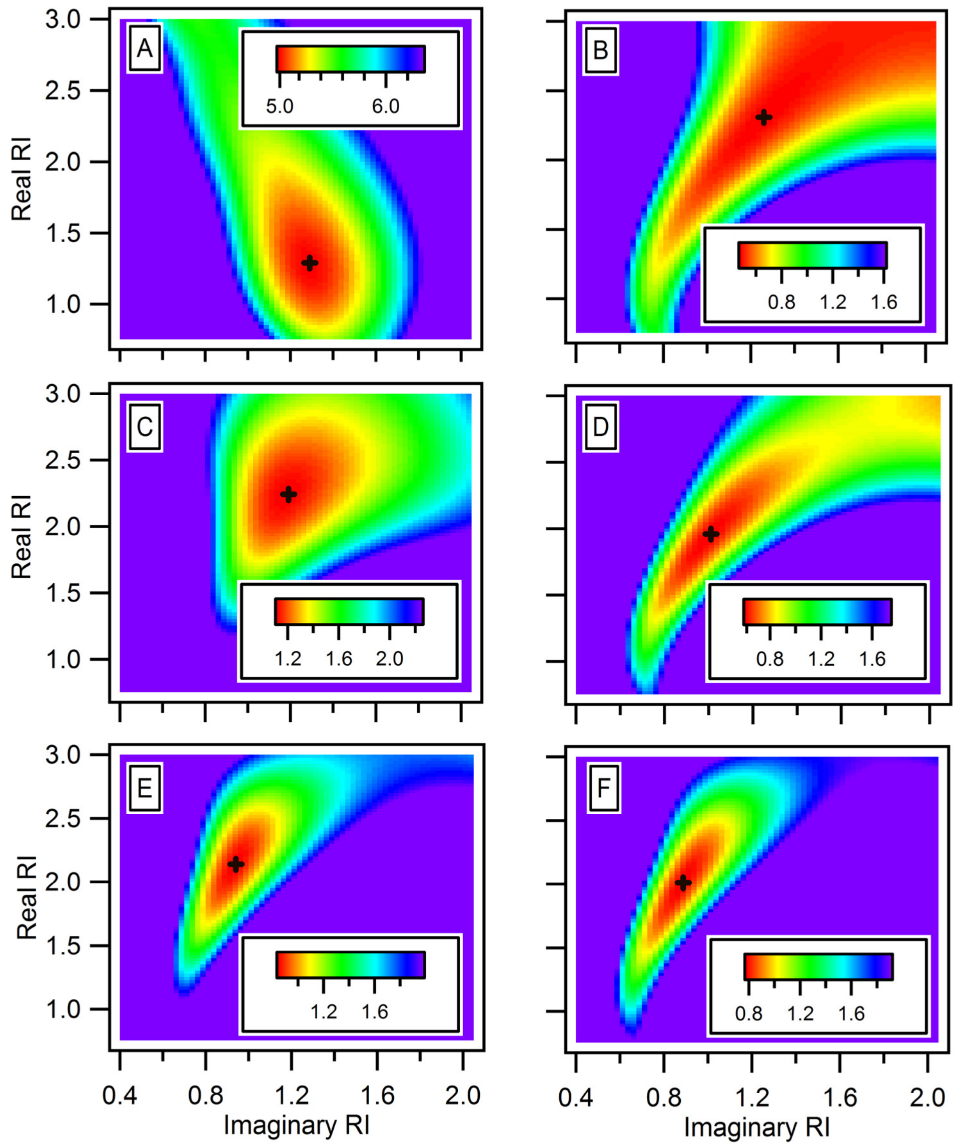

Figure S3. Image plots of the reduced χ^2 values of Mie Theory fits to absorption for all methane flame data. The colors represent the reduced χ^2 value at each effective refractive index. Each panel differentiates between different wavelengths. The column on the left is the fit over all size parameters and the column on the right corresponds to size parameter (x)<0.9. The minimum reduced χ^2 are shown as black crosses. Panels A and B correspond to λ = 405 nm data, panels C and D correspond to λ = 532 nm data, and panels E and F correspond to λ = 630 nm data. Purple on the image plots indicates the region beyond 1 standard deviation of the reduced χ^2 value (χ^2+1.11).

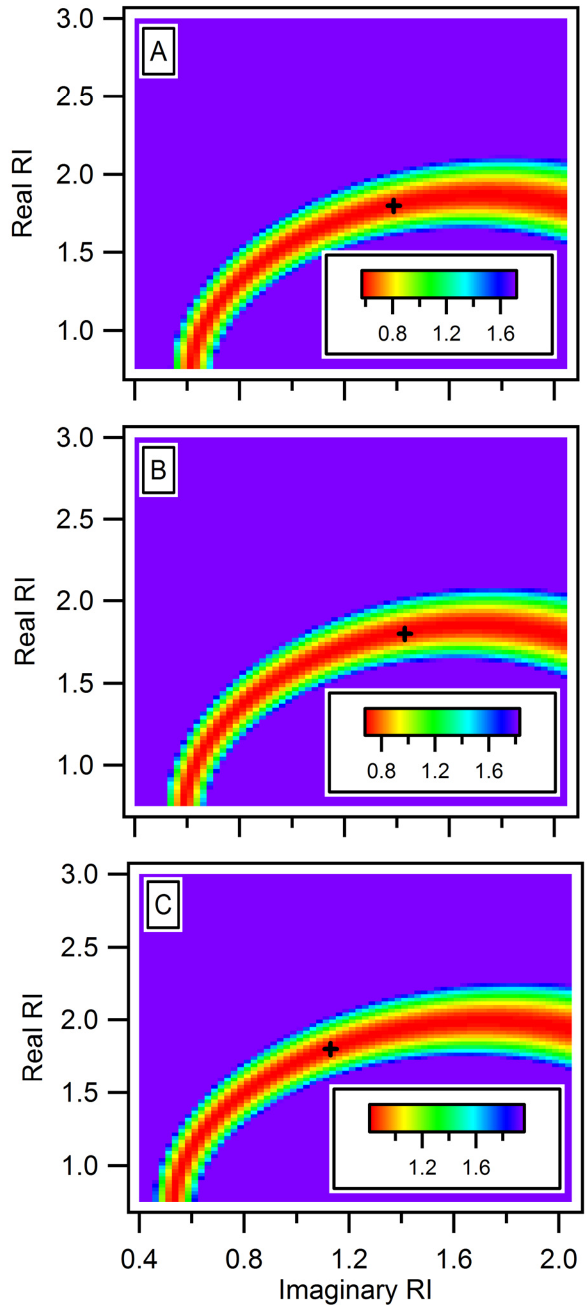

Figure S4. Same as Figure S3, except for RDG fits instead of Mie fits over all size parameters. Panel A, B, and C correspond to λ = 405 nm, 532 nm, and 630 nm data, respectively. Note that the crosses shown are not global minima, but selected to have m = 1.80. This is because there is no unique solution for the RDG fits.

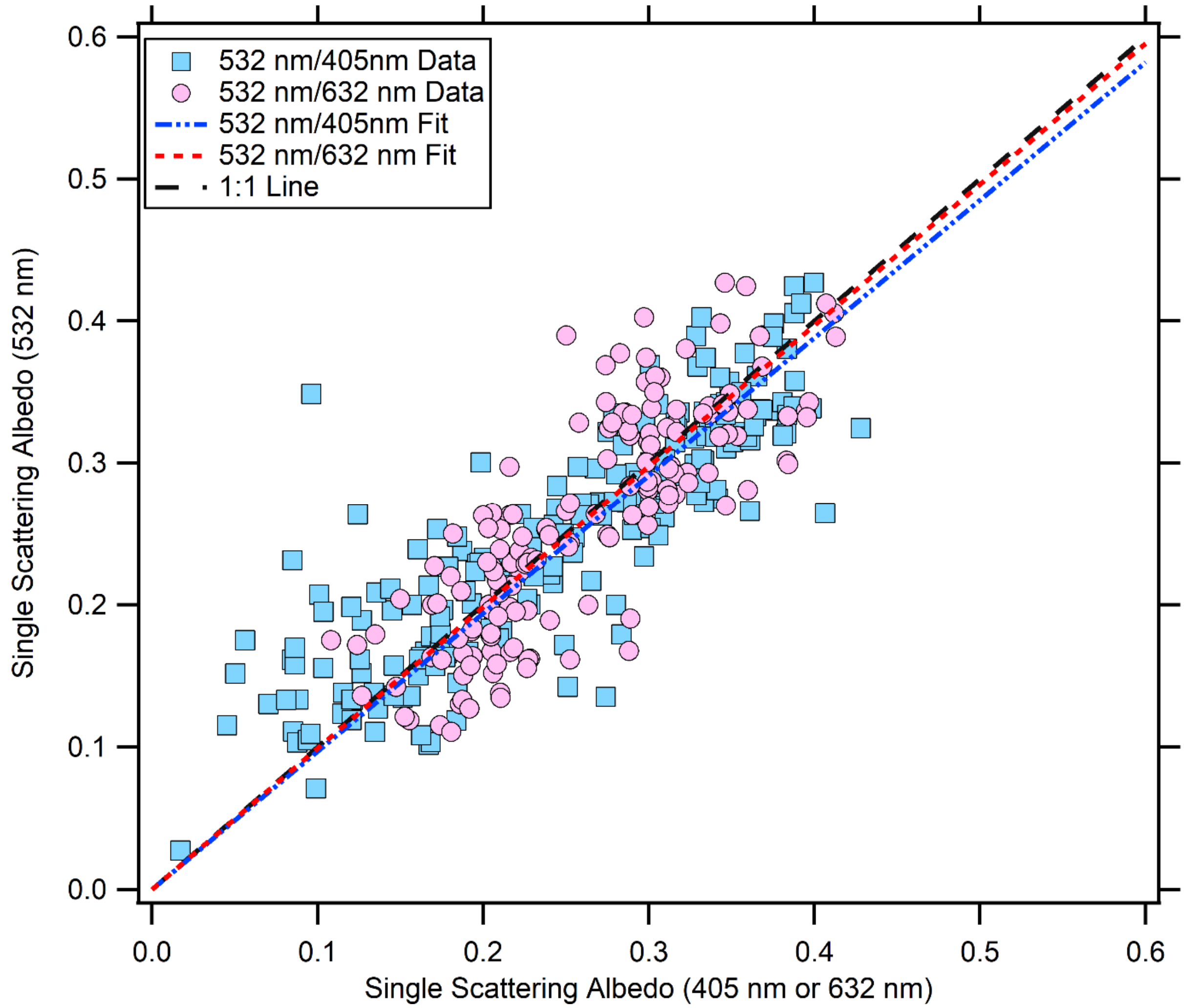

Figure S5. Comparison between observations of the single scattering albedo (*SSA*) at λ = 532 nm and at λ = 405 nm (blue squares) or at λ = 630 nm (pink circles) for the methane diffusion flame. The blue dashed line is a linear fit to the λ = 405 nm and 532 nm SSA pair, the red dashed line is the fit to the λ = 630 nm and 532 nm SSA pair. The black line is the 1:1 line. Note the generally good agreement between the *SSA*s between wavelengths. The best-fit slope was 0.97 for the λ = 405 nm/532 nm pair and 0.99 for the λ = 630 nm/532 nm pair.

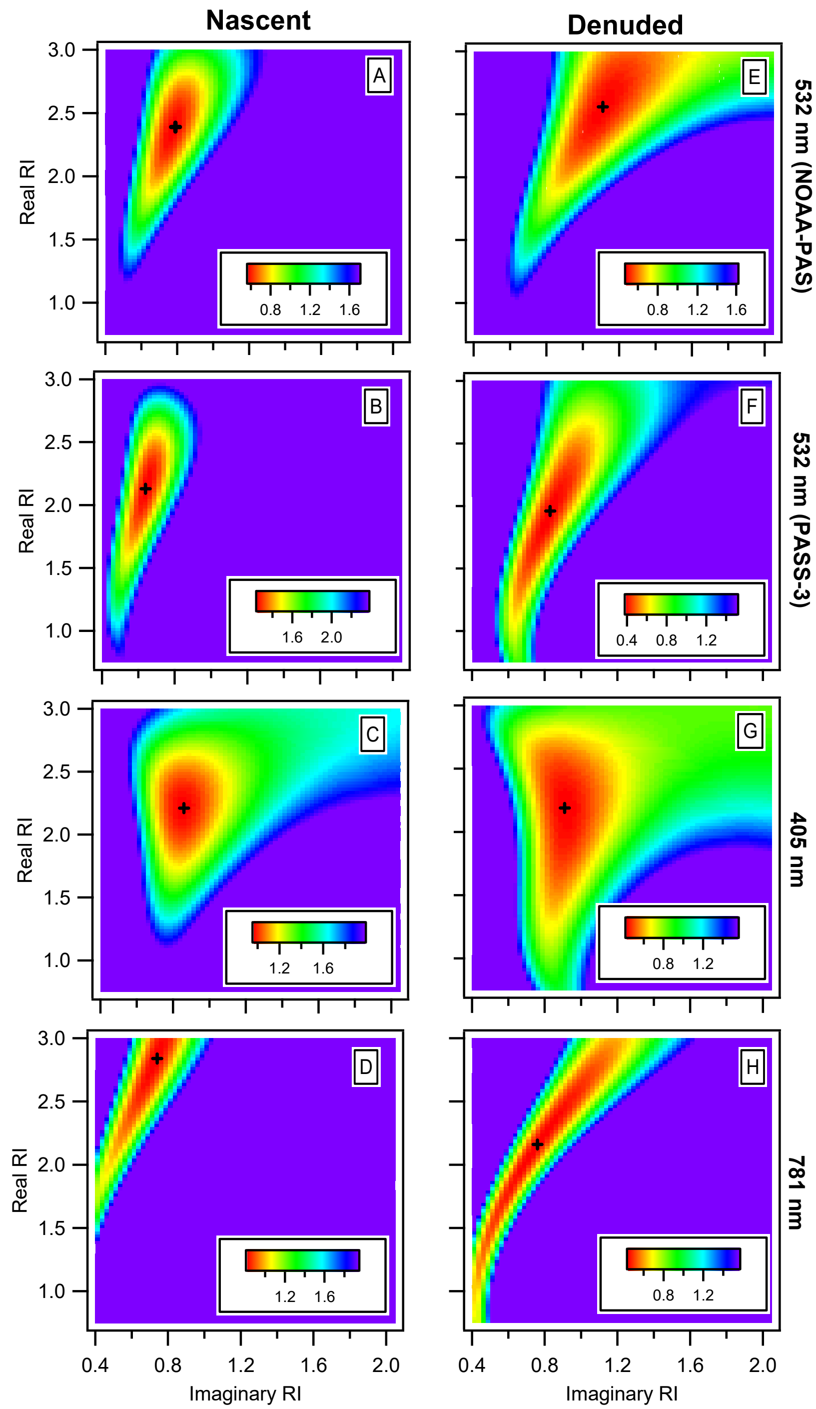

Figure S6. Image plots of the reduced χ^2 values of Mie Theory fits to absorption for all ethylene flame (20 cm sampling height) data from BC2. Colors indicate reduced χ^2 values, with the minima values shown as black crosses. Purple indicates the region beyond 1σ of the minimum reduced χ^2 value (χ^2+1.11). Each panel is for a particular wavelength and soot type (nascent vs. thermodenuded). The left column (A-D) is for nascent soot. The right column (E-H) is for denuded soot. Panels A and E are λ = 532 nm NOAA PAS data, panels B and F are λ = 532 nm PASS-3 data, panels C and G are λ = 405 nm PASS-3 data, and D and H are λ = 781 nm PASS-3 data.

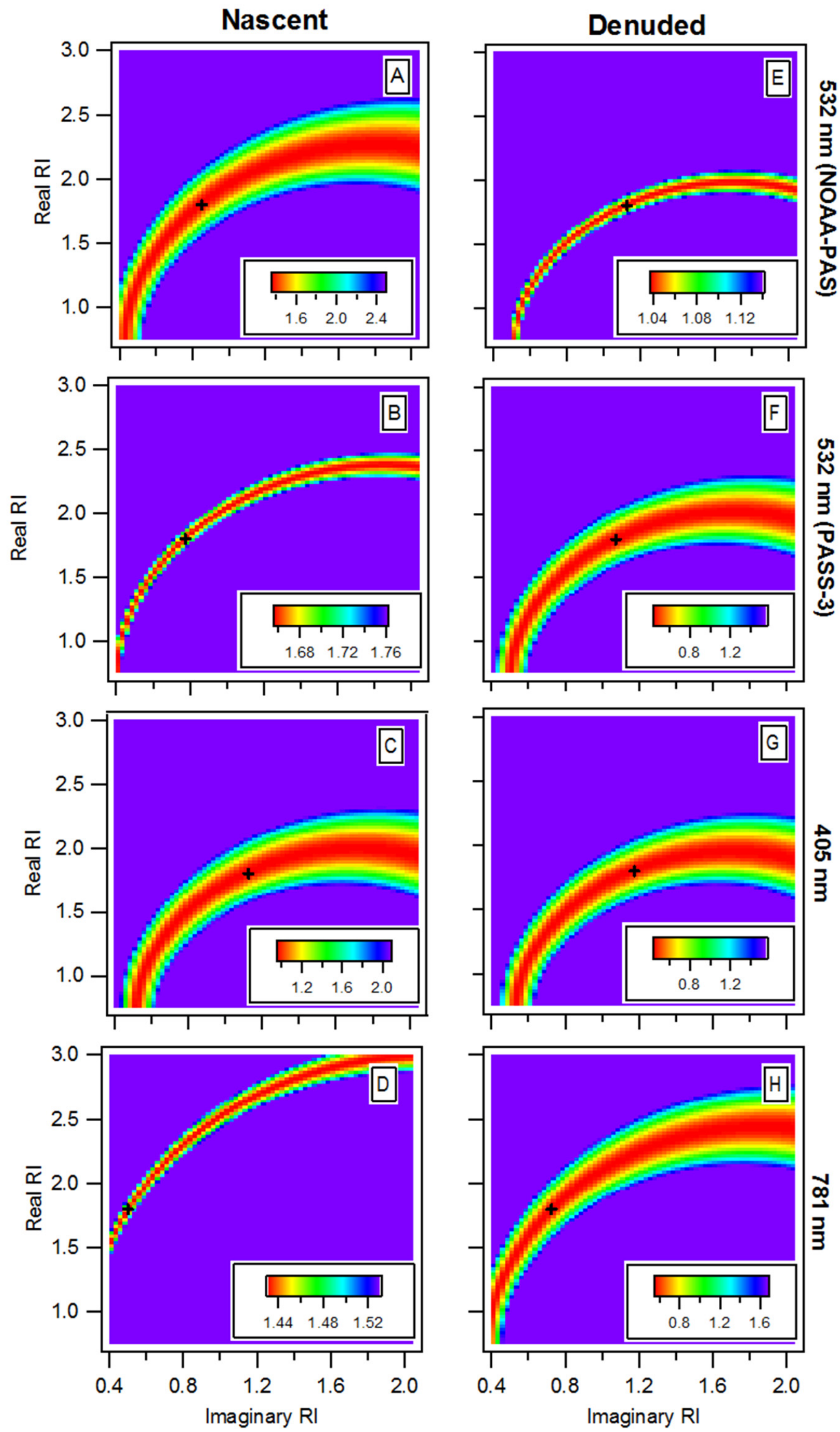

Figure S7. Same as Figure S6, except for RDG fits instead of Mie fits. Panel letters correspond to the same datasets as Figure S6. Note that the crosses shown are not global minima, but selected to have m = 1.80. This is because there is no unique solution for the RDG fits.

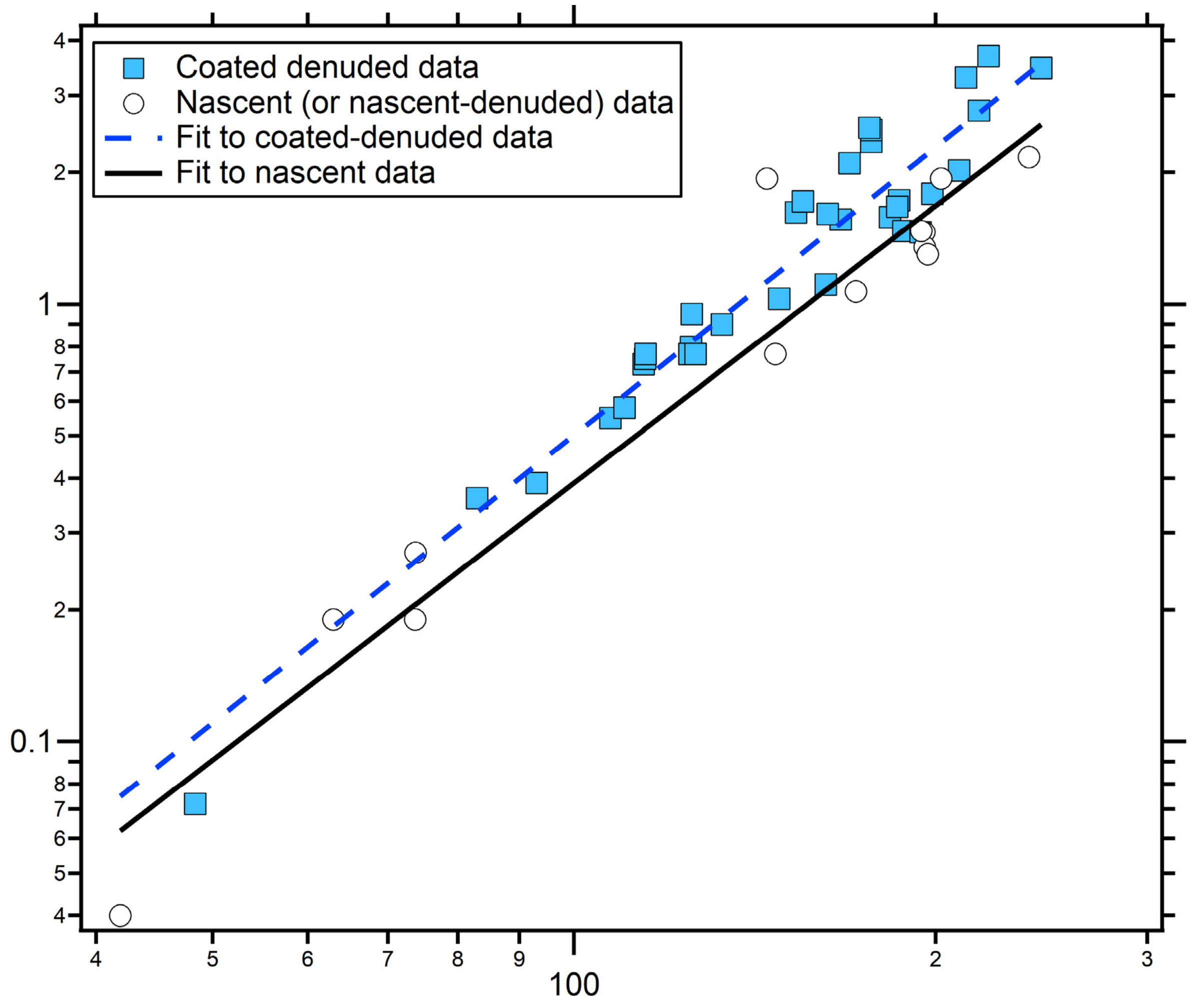

Figure S8. Fit of CPMA mass per particle as a function of the SMPS mode number-weighted mobility diameter for soot particles from the premixed ethylene flame from BC2, when the flame was sampled at 20 cm above the burner surface. The blue squares are coated denuded observations and the dashed blue line is the associated power-law fit. The white circles are nascent observations and the black line is the associated power-law fit. The data were fit to a power law function (see eqn. 8 in the main text). For nascent soot, fit parameters C and $D_{f,m}$ were 2.40 x 10^{-5} and 2.12, respectively, and for coated-denuded soot, fit parameters C and $D_{f,m}$ were 1.33 x 10^{-5} and 2.28, respectively.

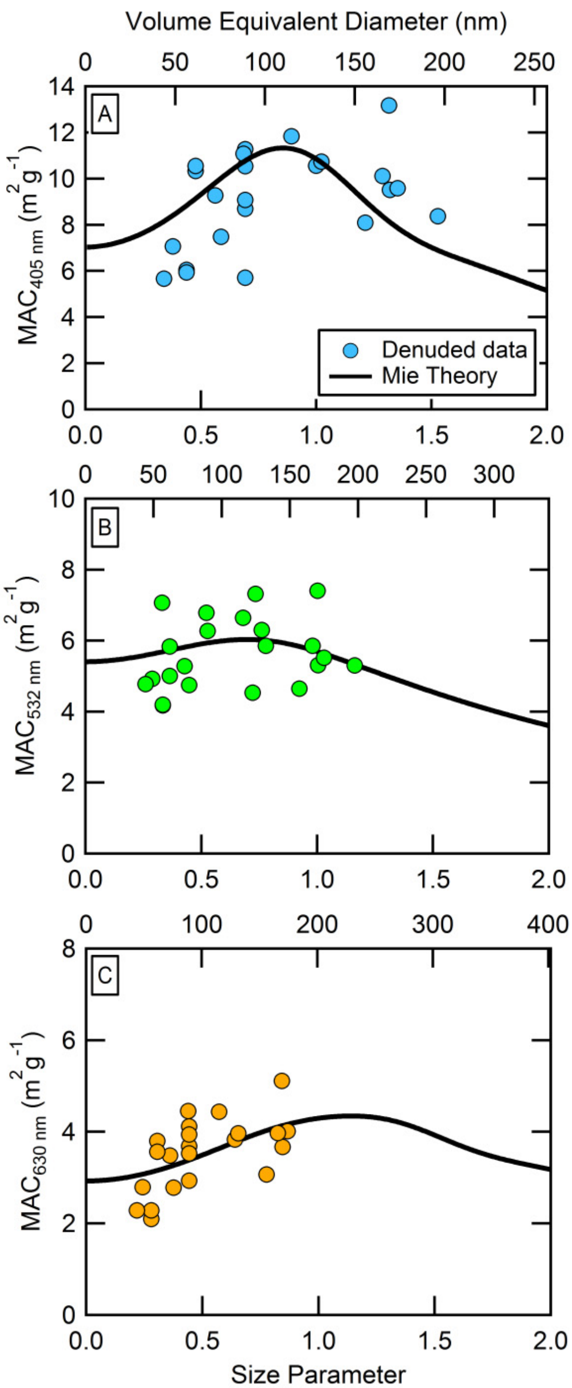

Figure S9. *MAC*s for ethylene soot sampled 5.1 cm above the burner surface as a function of size parameter (bottom) and volume equivalent diameter (top) for **(A)** λ =405 nm, **(B)** λ = 532 nm, and **(C)** λ = 630.

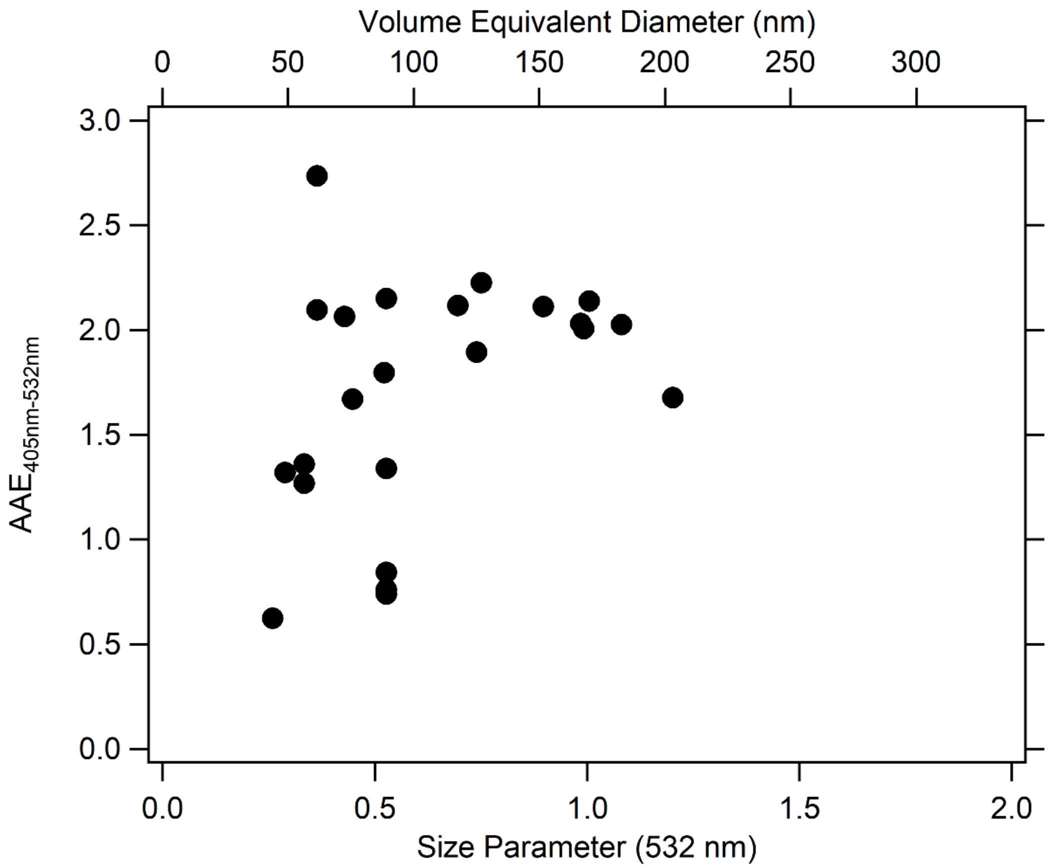

Figure S10. Absorption Ångström exponent (*AAE*) calculated using data measured at λ = 405 nm and λ = 532 nm as a function of size parameter (at λ = 532 nm) and volume equivalent diameter produced from the ethylene flame during BC3+.