One Year Survey of Brown and Black Carbon Contributions in the U.S.

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Objectives

- Examine measurement reproducibility and filter loading effects for ~30,000 IMPROVE and CSN 2016 samples.
- Demonstrate the separation of brown carbon (BrC) from black carbon (BC) using multiwavelength analysis.
- Characterize transmittance attenuation for source and ambient samples.
- Present seasonal and spatial patterns of BrC and BC.

Methods to Calculate Aerosol Radiation Absorption

$$ATN_{\lambda} = ln\left(\frac{FT_{\lambda,f}}{FT_{\lambda,i}}\right) \quad (1)$$

$$b_{ATN}(_{\lambda}) = ATN_{\lambda} \times \left(\frac{A}{V}\right) \quad (2)$$

$$\frac{b_{ATN}(\lambda_1)}{b_{ATN}(\lambda_2)} = \left(\frac{\lambda_1}{\lambda_2}\right)^{-AAE} \quad (3)$$

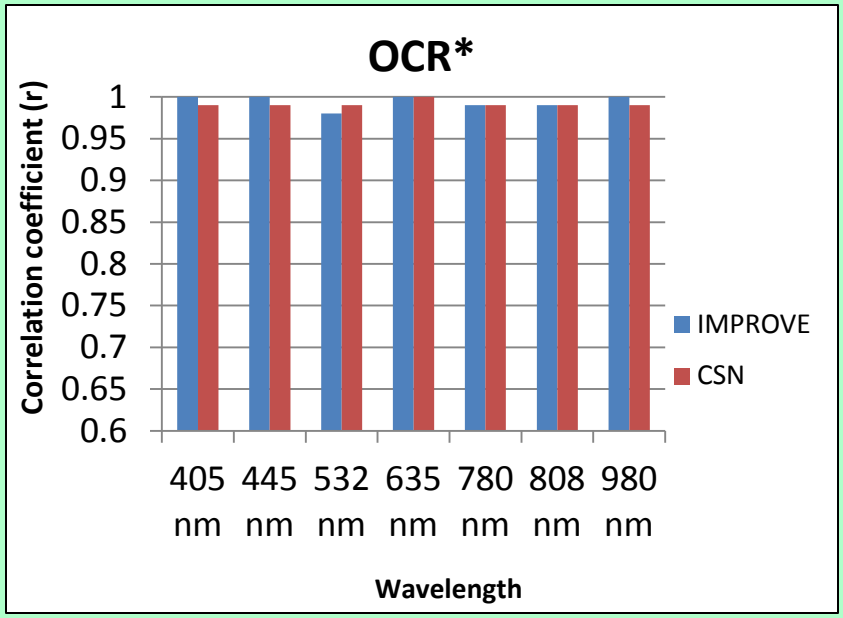
$$ATN_{\lambda} = q_{BC} \times \lambda^{-AAE_{BC}} + q_{BrC} \times \lambda^{-AAE_{BrC}} \quad (4)$$

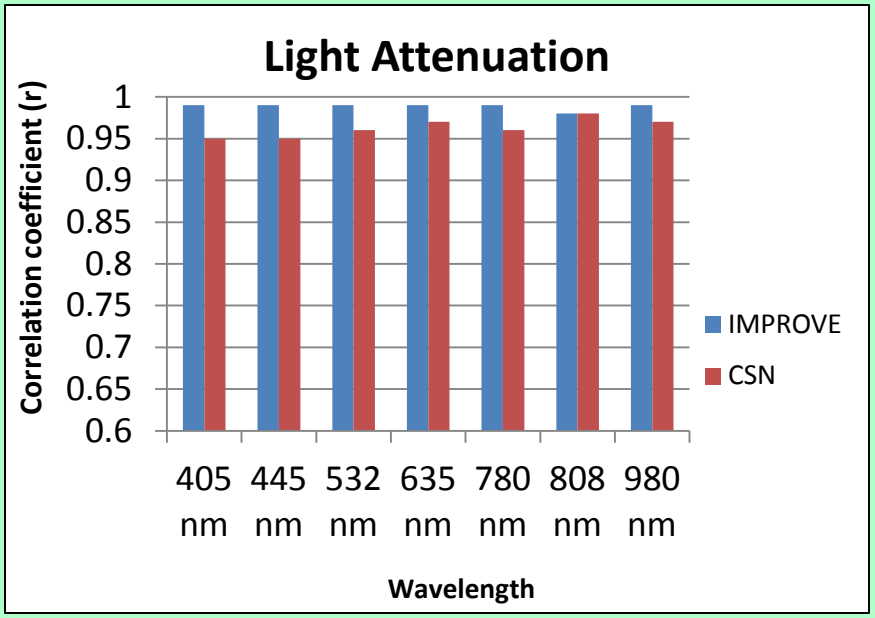
Where:

- ATN is the radiation attenuation
- $FT_{\lambda,i}$ and $FT_{\lambda,f}$ represent the filter transmittance before and after thermal carbon analysis, respectively
- b_{ATN} is the attenuation coefficient
- A is the filter deposit area (3.53 cm^2)
- V is the 25 mm IMPROVE sample volume (33.12 m^2)
- AAE is the absorption Angström exponent
- q_{BC} and q_{BrC} represent the fitting coefficients

High correlations across wavelengths are found for carbon and light attenuation replicates

(1867 IMPROVE and 1612 CSN)

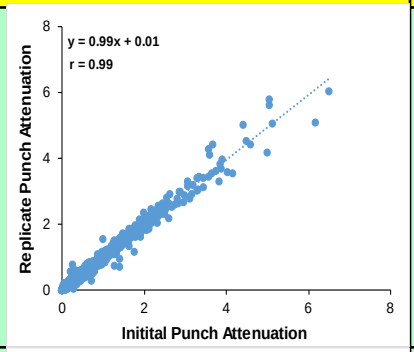
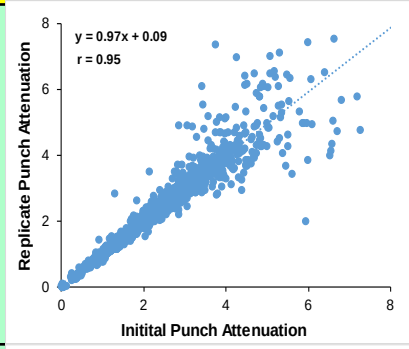
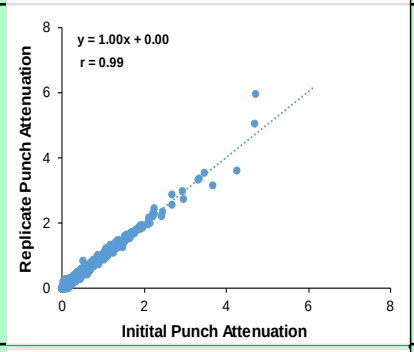
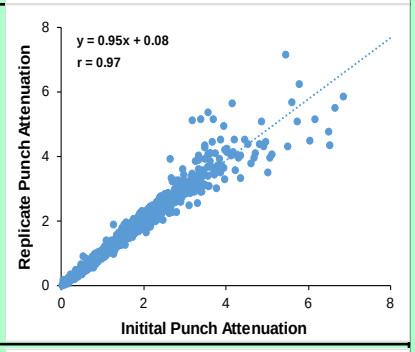
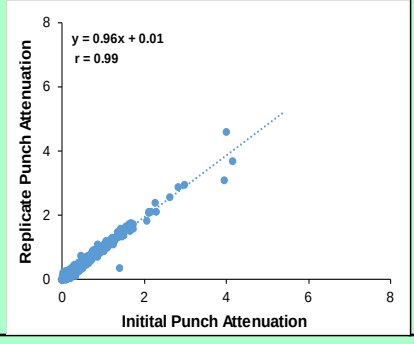
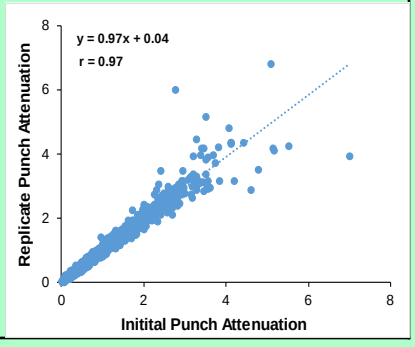




*Organic carbon by reflectance

Replicate ATN is less comparable for ATN>4

(filter loading and multiple scattering effects not compensated)

	IMPROVE	CSN
ATN_{405}		
ATN_{635}		
ATN_{980}		

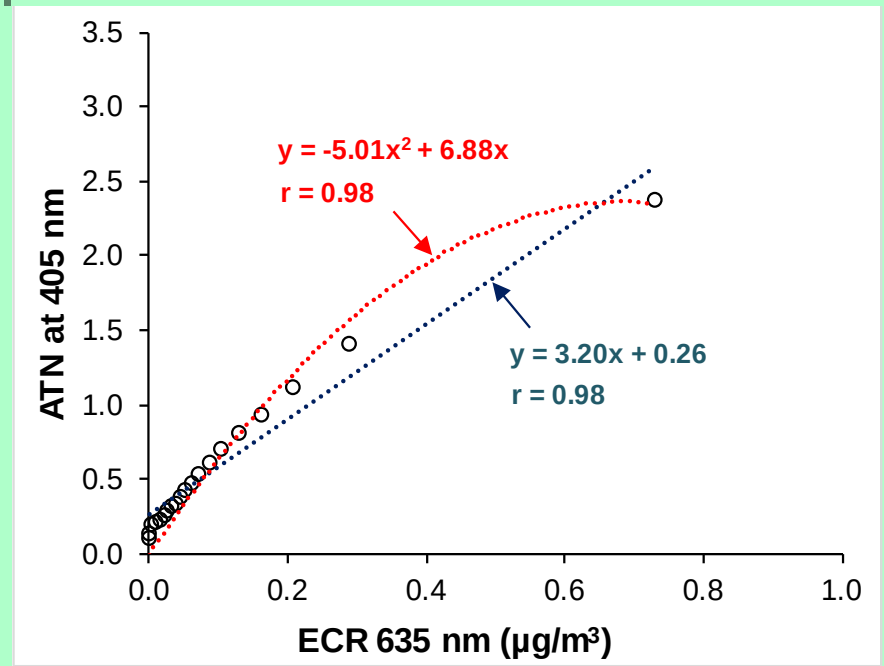
$$ATN_{\lambda} = ln\left(\frac{FT_{\lambda,f}}{FT_{\lambda,i}}\right)$$

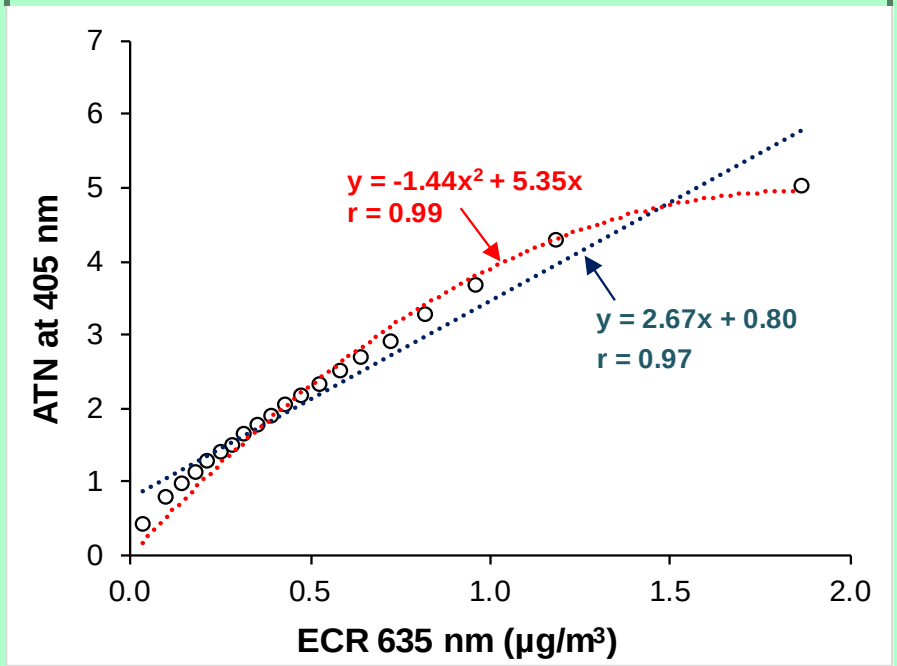
- *ATN* is the radiation attenuation
- $FT_{\lambda,i}$ and $FT_{\lambda,f}$ represent the filter transmittance before and after thermal carbon analysis, respectively

ATN_{405} becomes less related to EC at higher concentrations*

(due to both loading and BrC)

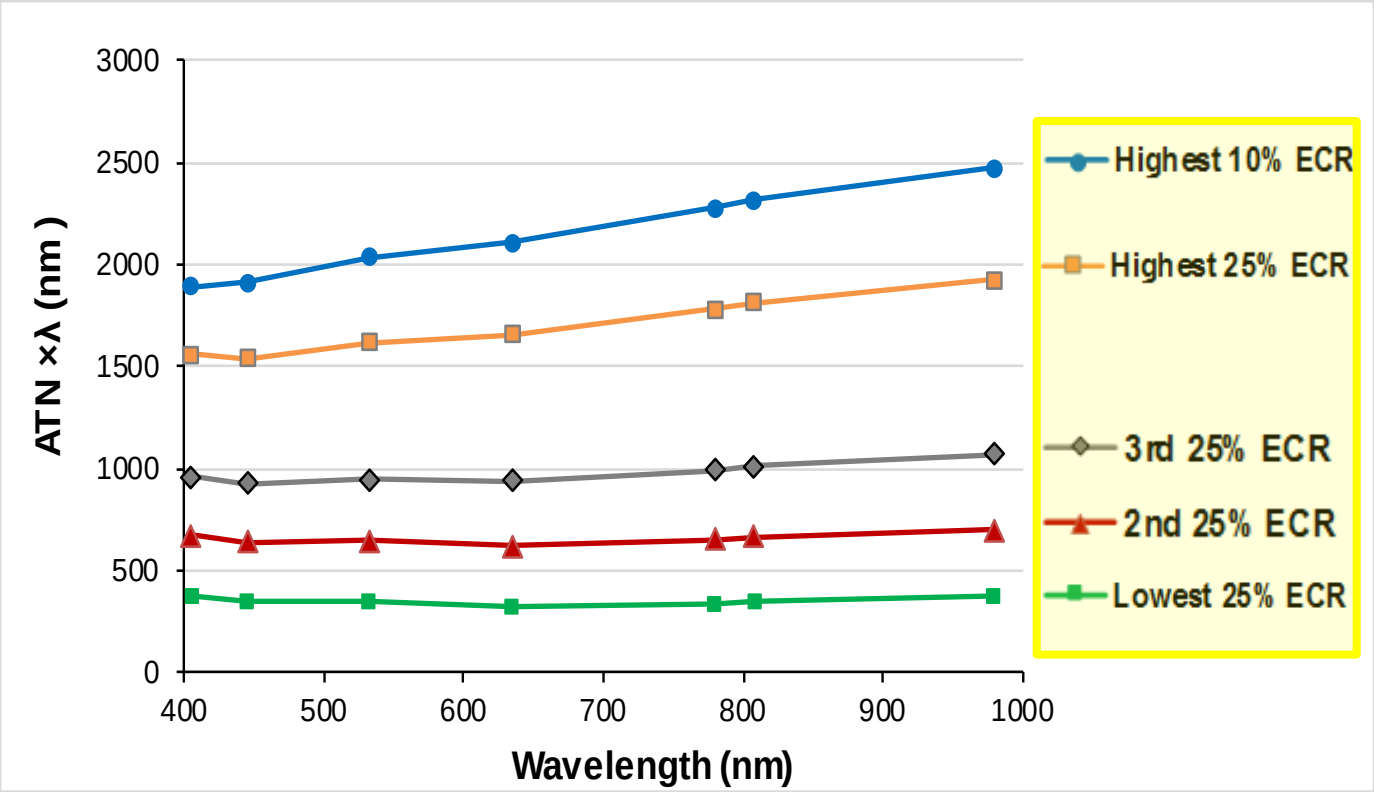
IMPROVE	CSN



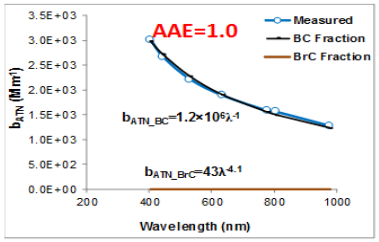
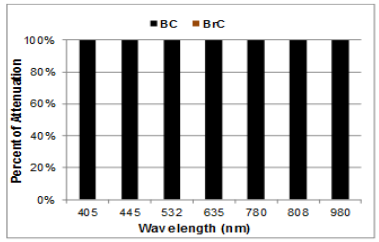
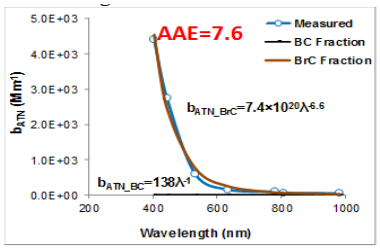
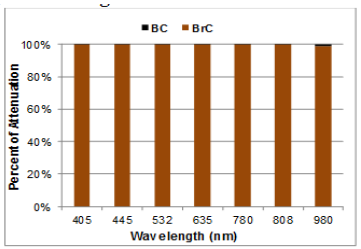
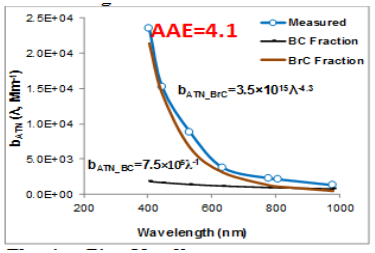
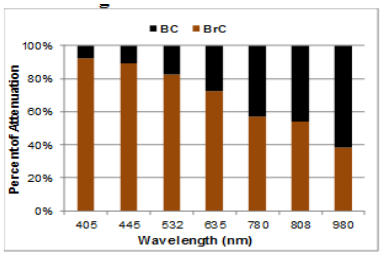
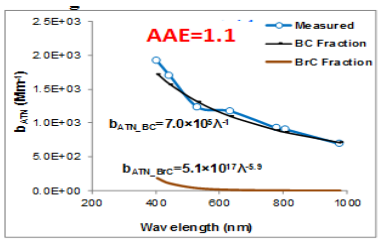
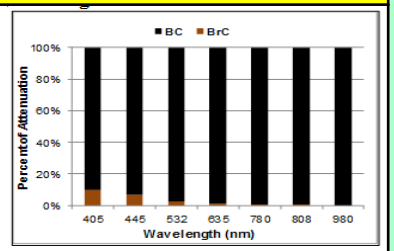


*Averaged over each 5th percentile range of ECR by reflectance at 635 nm

Wavelength-dependent decrease in attenuation occurs for highest 25% of EC concentrations.



ATN multiplied by wavelength (λ) for the highest 10th percentile and each 25th percentile of EC by reflectance (ECR) concentrations for the 2016 CSN samples.

Spectral Light Attenuation	BC and BrC Contributions to Attenuation
Diesel Exhaust	
	
Smoldering Pine Cones	
	
Smoldering Peat	
	
Flaming Pine Needles	
	

Spectral attenuation varies by source

←(up to 100% BrC with high AAE for smoldering pine cones)

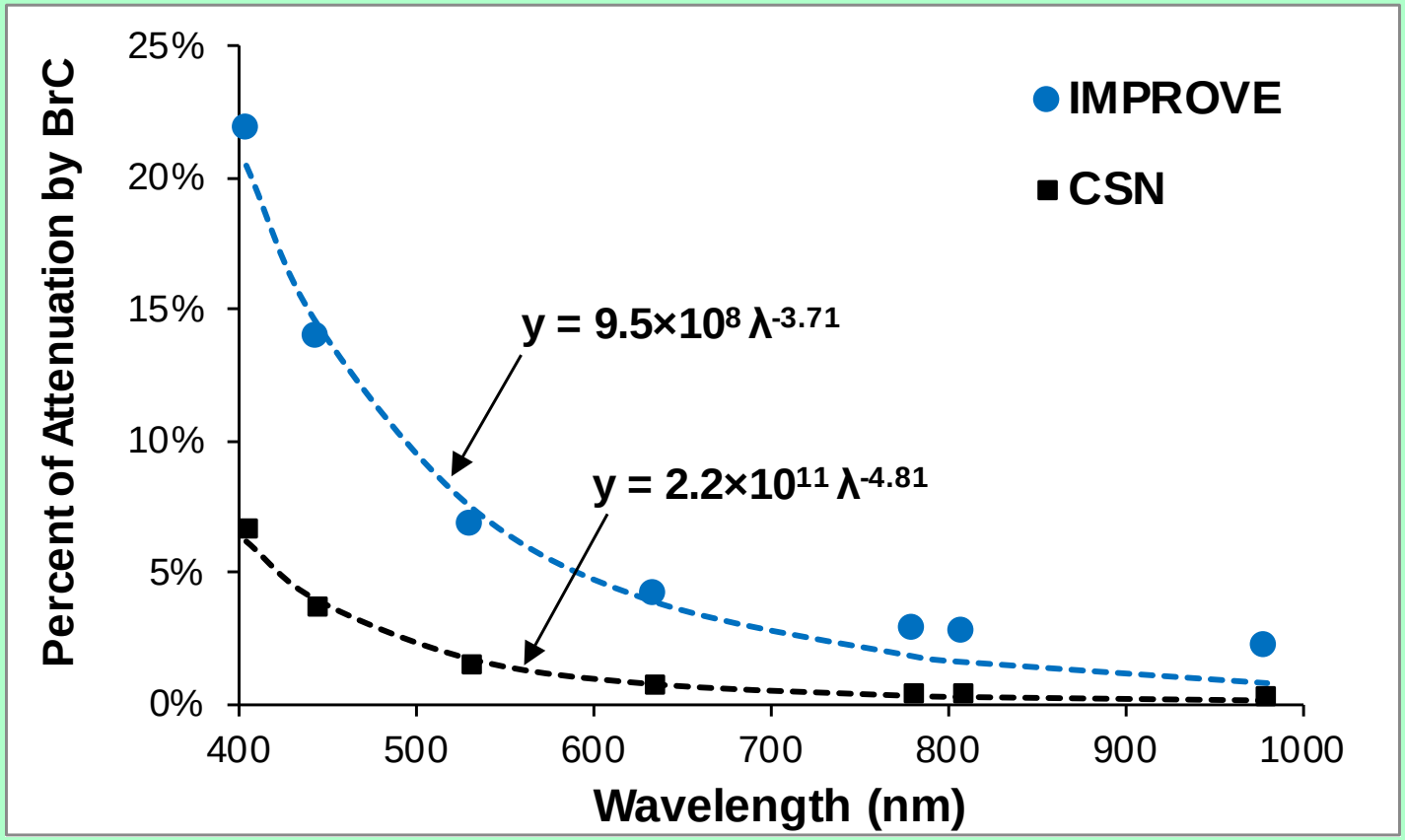
AAE varies with different calculation methods

Source	AAE (7λ fit)	AAE (405_980 nm ratio)	AAE (405_635 nm ratio)
Diesel	0.94	0.96	1.03
Diesel	0.87	0.87	0.89
Diesel	0.90	0.92	0.97
Diesel	0.85	0.86	0.87
Diesel	0.98	1.08	1.36
Diesel	0.88	0.93	1.01
Tunnel	0.81	0.80	0.88
Smoldering Pine Cone	5.73	5.39	7.63
Flaming Pine Needle	1.09	1.16	1.09
Peat	9.05	8.02	7.66
Peat	3.34	3.30	4.08
Prescribed Burning	1.81	1.77	2.77
Prescribed Burning	2.17	2.16	2.94
Prescribed Burning	3.06	3.04	4.13
Prescribed Burning	2.68	2.73	3.64
Yosemite Rim Fire	1.34	1.35	1.84

The wavelength ratios at 405 and 635 nm are used to derive the power-law fit of b_{ATN}

Enhanced light attenuation by BrC is found at shorter wavelengths

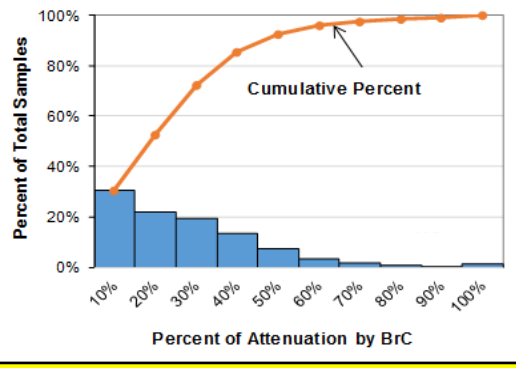
(average attenuation due to BrC_{405} is 6.7% for CSN and 21.9% for IMPROVE)



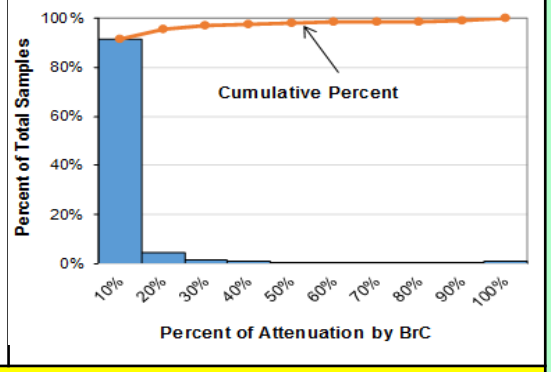
The wavelength ratios at 405 and 635 nm are used to derive the power-law fit of b_{ATN}

IMPROVE

405 nm

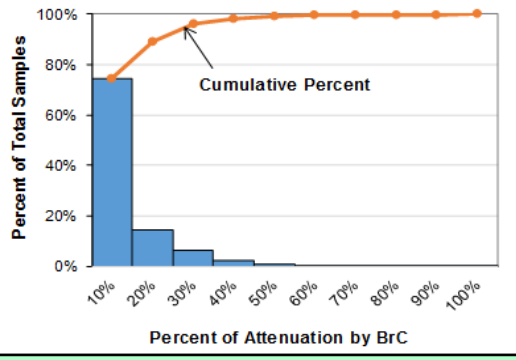


635 nm

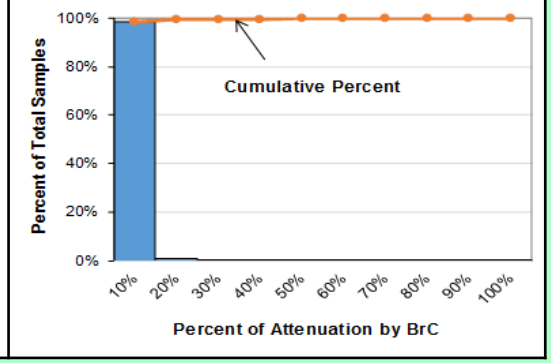


CSN

405 nm



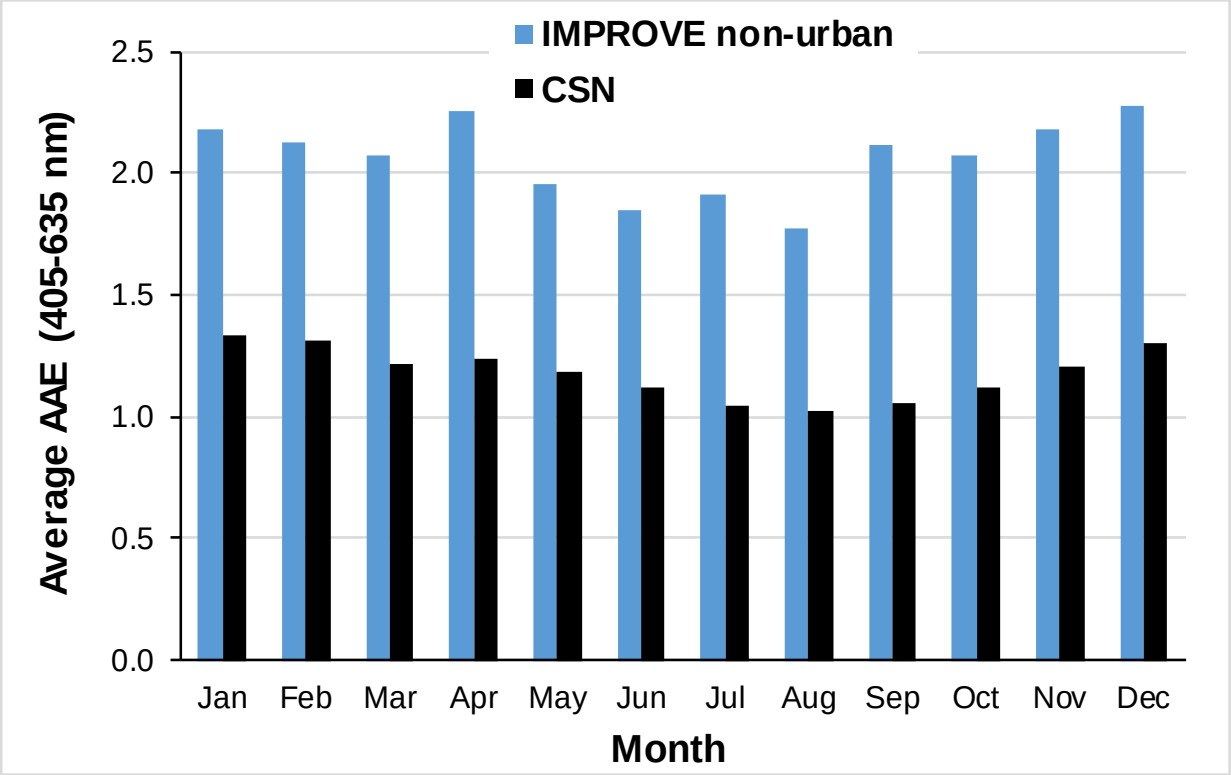
635 nm



Only 8.6% of IMPROVE and 1.4% of CSN samples have >10% attenuation by BrC at 635 nm

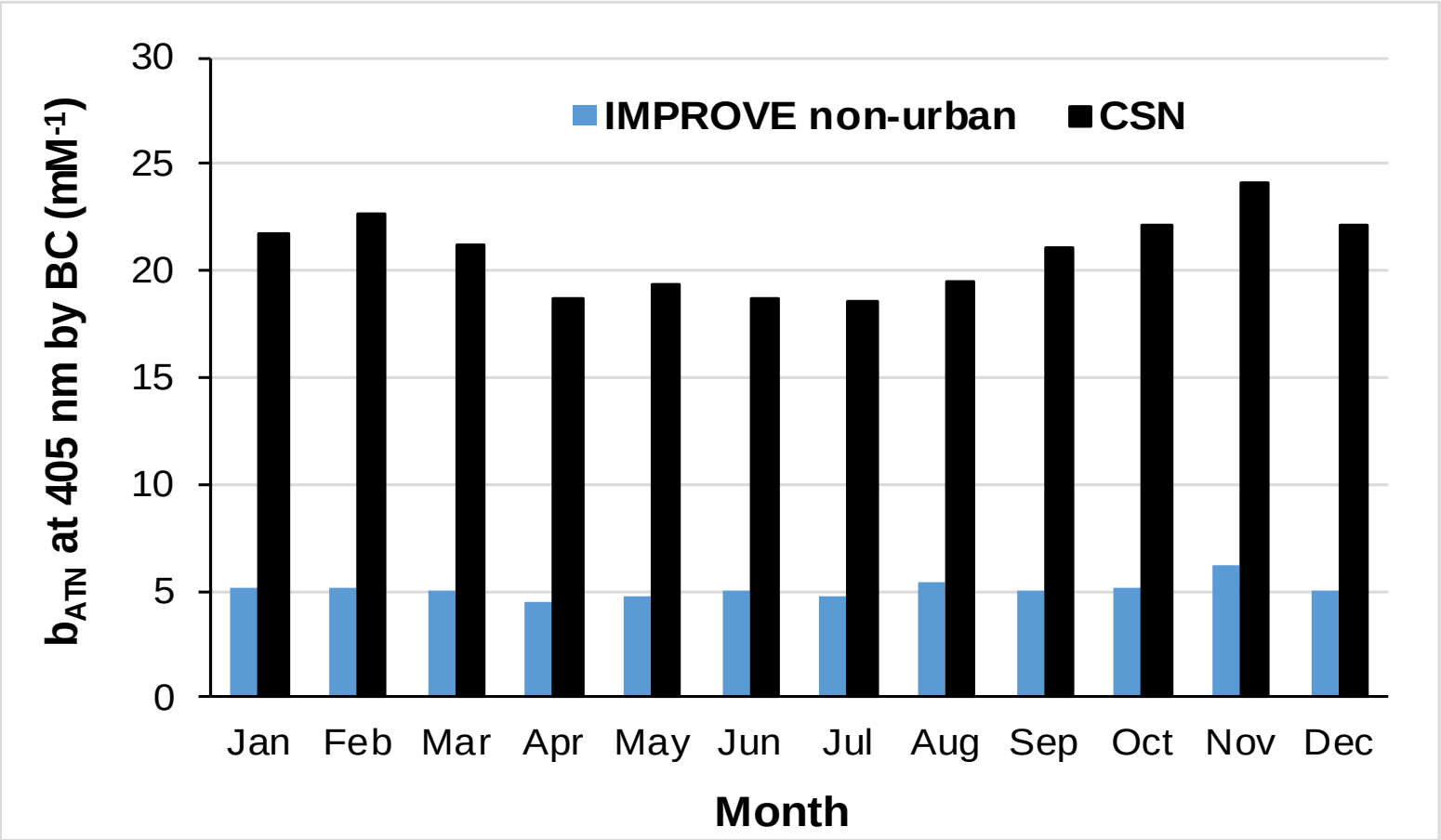
(showing that BC is a good indicator at 635 nm but misses BrC contributions)

Higher AAEs in IMPROVE than CSN suggest increased contributions from biomass burning and aged aerosol in non-urban environments



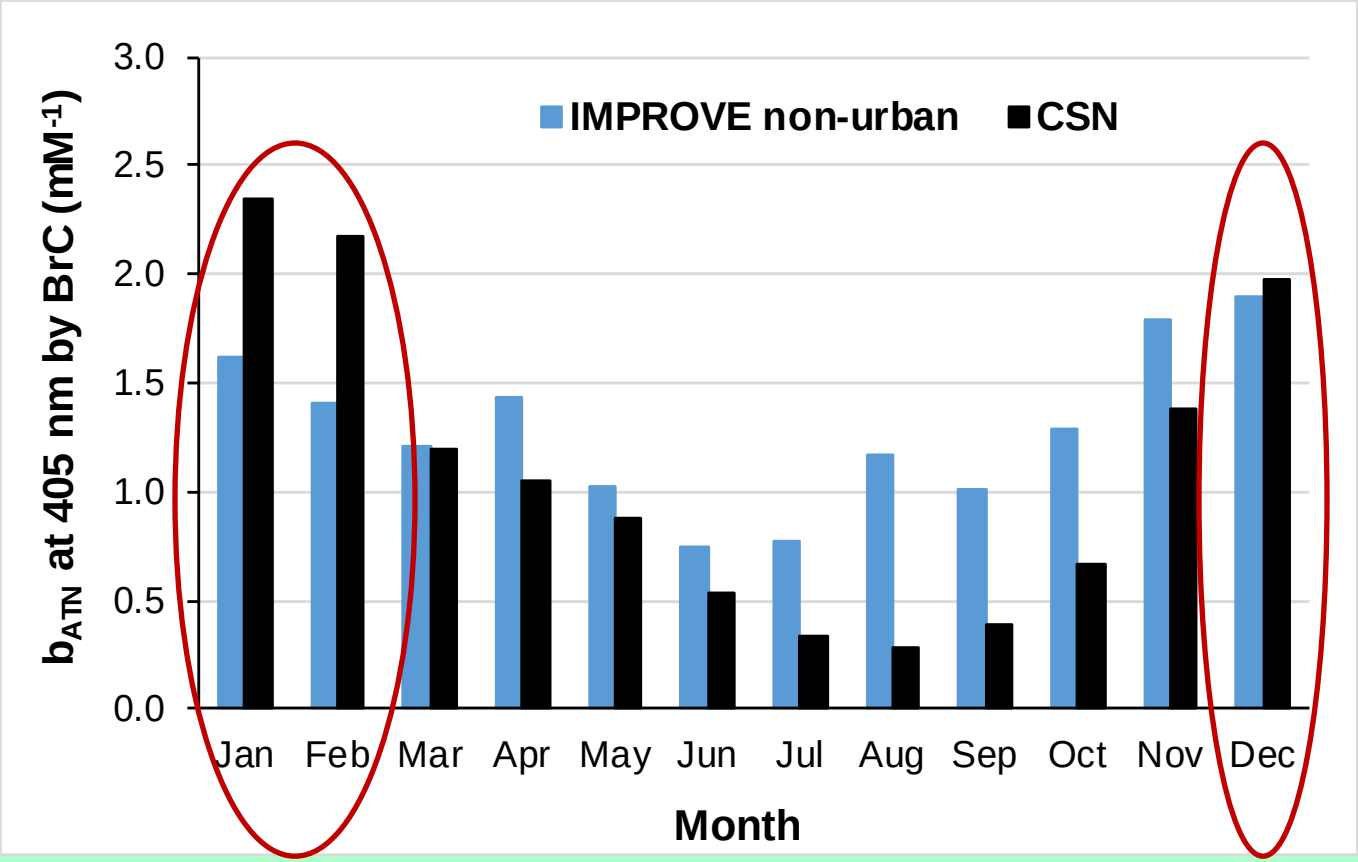
Monthly average AAE values derived from the wavelength ratios at 405 and 635 nm (Eq. 3) at the non-urban IMPROVE and urban CSN sites.

BC light attenuation (b_{ATN}) at 405 nm is ~4 times higher at CSN than IMPROVE sites

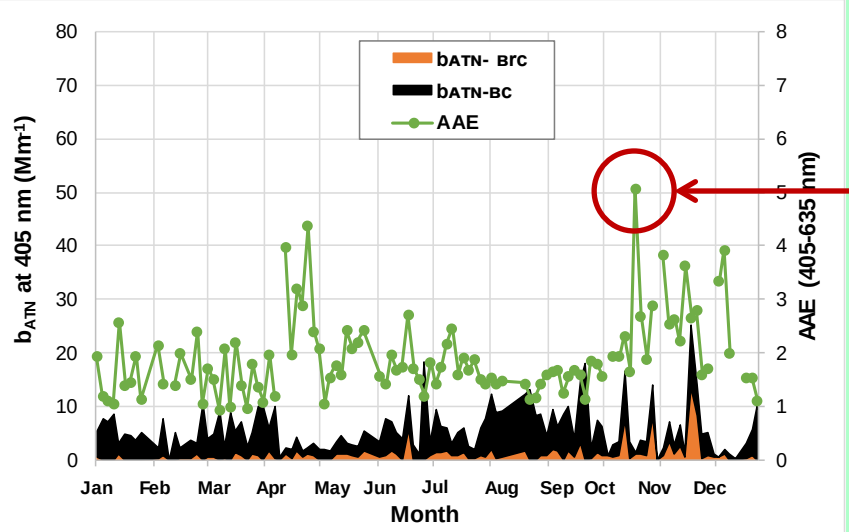


Where: $b_{ATN}(_{\lambda}) = ATN_{\lambda} \times \left(\frac{A}{V}\right)$ **and** $ATN_{\lambda} = ln\left(\frac{FT_{\lambda,f}}{FT_{\lambda,i}}\right)$

BrC light attenuation (b_{ATN}) at 405 nm increases at CSN sites during winter, indicative of residential wood combustion



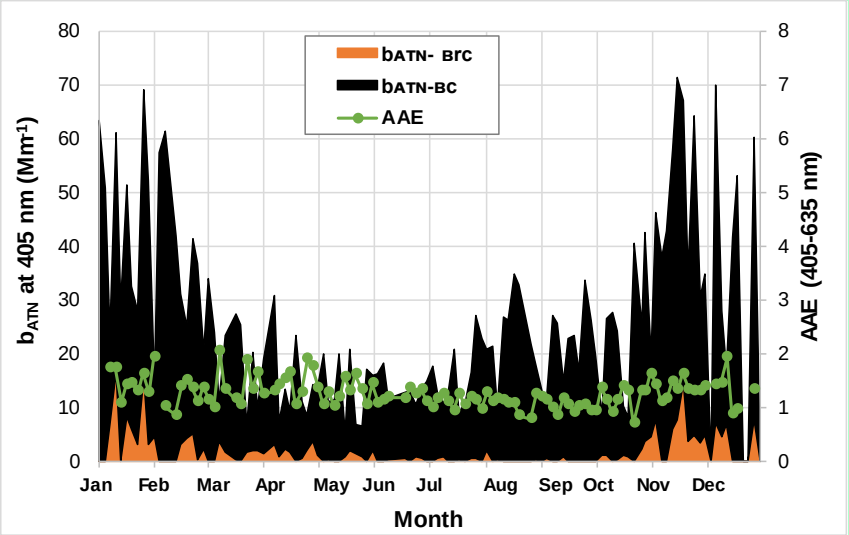
High BC and **BrC** in fall (non-urban Kaiser Wilderness*)



*73 km northeast of the Fresno site, located at 2698 m in the Sierra Nevada mountain range

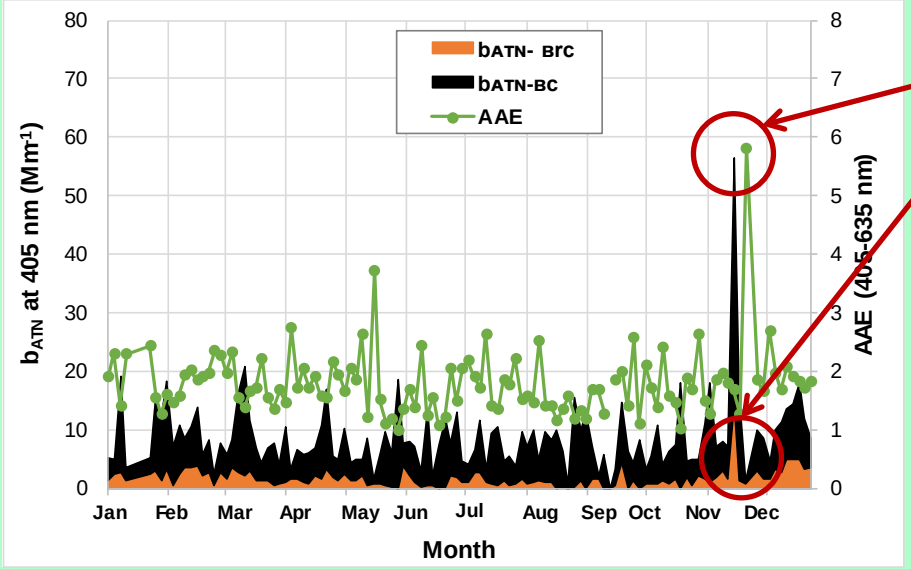
Elevated AAE attributed to 100-acre fire 36 km southeast

High BC and **BrC** in winter (urban Fresno, CA)



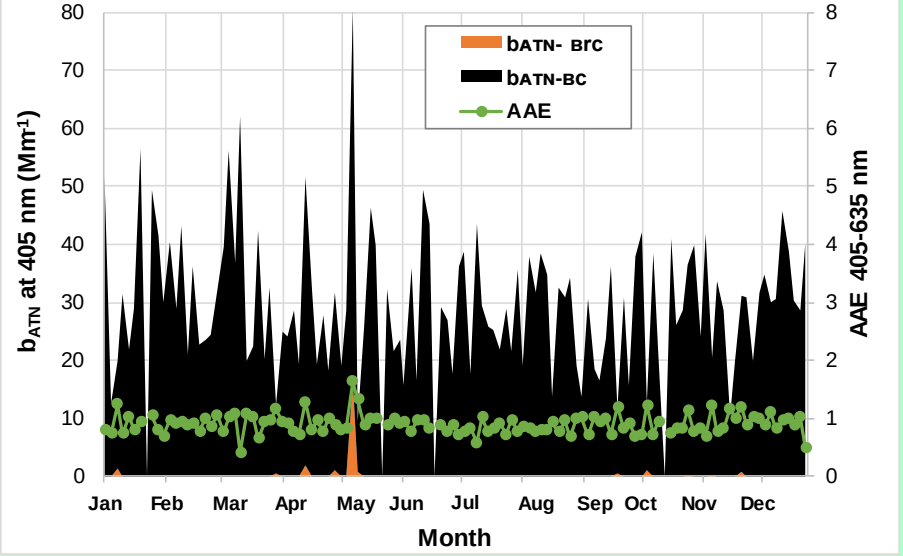
Temporal patterns of BC and BrC vary at non-urban versus urban sites

Non-urban Mohawk Mountain, CT*



*125 km northeast of Bronx at 505 m

Urban Bronx, NY



Possible influence from Flat Rock Fire (elevated hourly CO concentrations, hourly $PM_{2.5}$ [230µg/m^3], and 24-hour OC [20.7 µg/m^3] were found on 11/14/2016

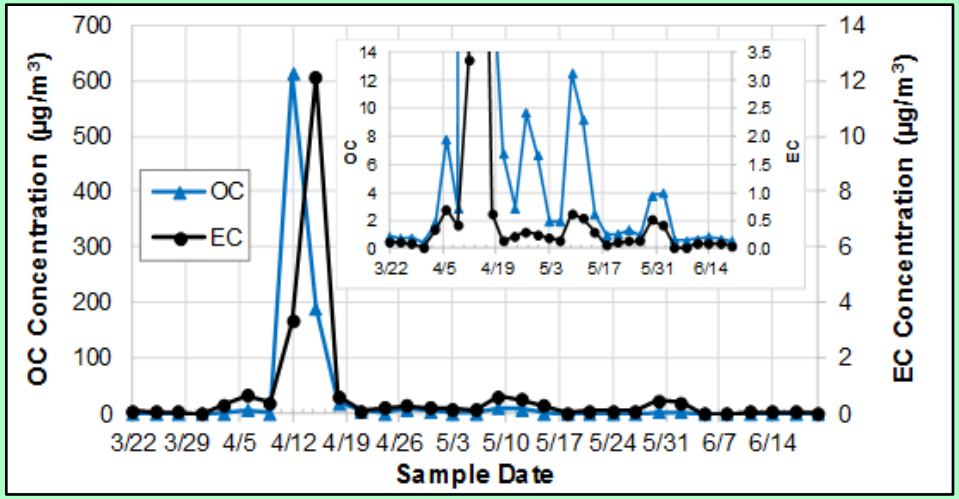
Mohawk Mountain site shows higher AAEs and higher BrC_{405} contributions than the Bronx site

(22% vs. 0.8%)

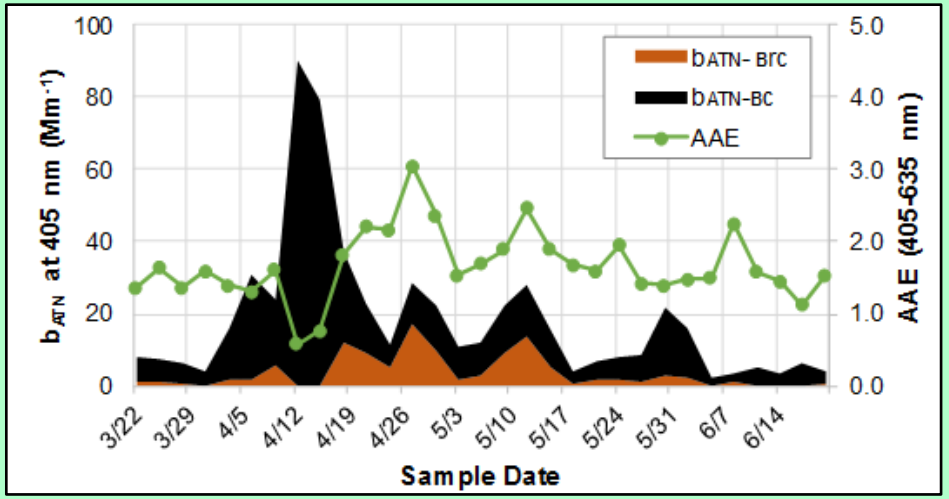
Elevated carbon and light attenuation by BrC is apparent during fires

(Long Pine Key Wildfire in the Everglades National Park; 4/10-17/16)

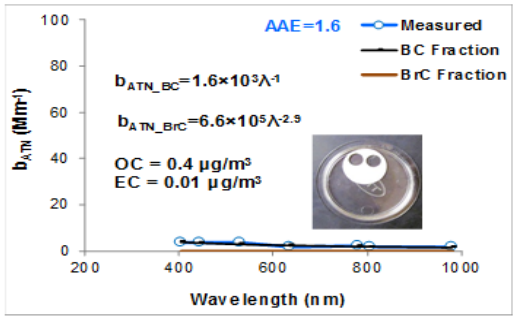
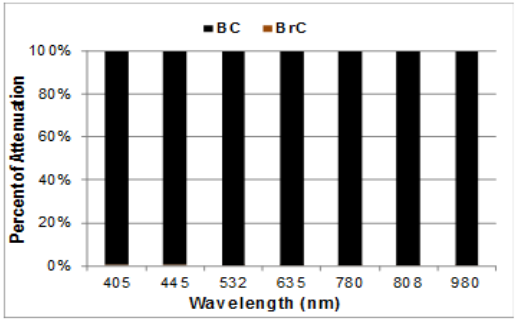
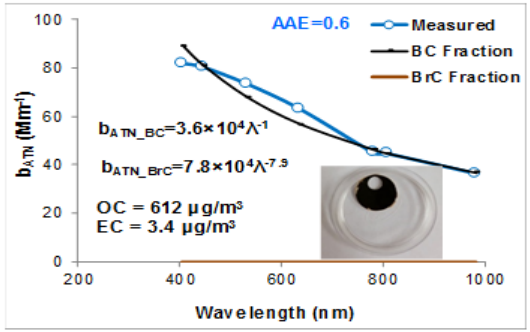
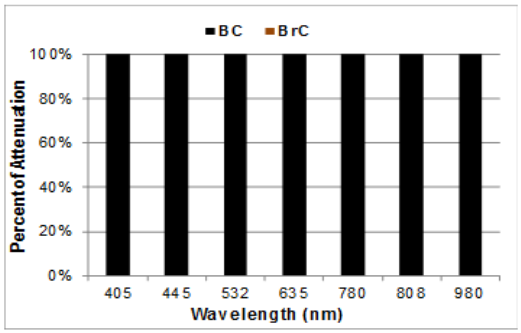
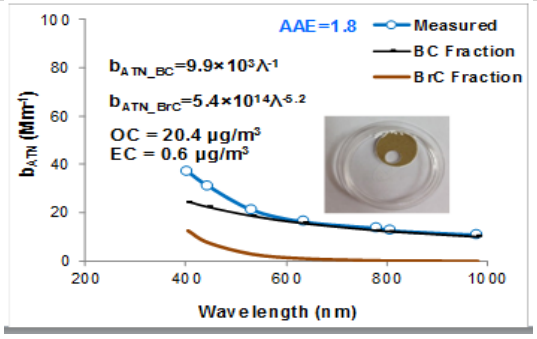
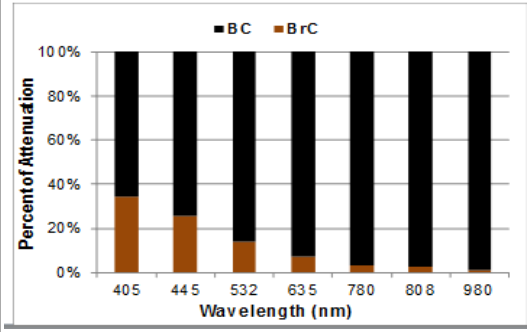
OC and EC Concentrations



Light Attenuation and AAE



Evolution of fire from flaming to smoldering is apparent

Spectral Light Attenuation	BC and BrC Contributions to Attenuation
Before fire (3/31/2016)	
	
During Fire (4/12/2016)	
	
After Fire (4/18/2016)	
	

Conclusions

- BrC and BC can be quantified based on multiwavelength light attenuation using Model 2015.
- Filter loading effect is apparent for ~25% of samples with the highest EC concentrations; loading corrections need to be implemented.
- Higher BrC absorption (at 405 nm) is found for IMPROVE than CSN samples, suggesting the influence of biomass burning and aged aerosol.
- Pairing non-urban and urban sites shows potential to distinguish local from regional exceptional events, which can be excluded from attainment designations.