



Dark brown carbon and aerosol mixing impacts on absorbing aerosol optical depth in the southeastern United States

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Abstract. Wildfires are becoming more prevalent in the southeastern United States. Wildfires emit light absorbing organic aerosols such as black carbon and dark brown carbon (tar balls) that influence regional air quality and radiative forcing. Recent experimental work has identified dark brown carbon as a potentially significant source of warming in the atmosphere.

However, dark brown carbon is not typically included in air quality and climate models. We added dark brown carbon from wildfires as a species in the Weather Research and Forecasting modeling system coupled with chemistry (WRF-Chem) version 4.2 with the Quick-Fire Emissions Database (QFED). The model simulations were run over the southeastern United States from June 21st to June 30th 2013 in conjunction with in-situ measurements of aerosol extinction from the Southeast Nexus (SENEX) aircraft campaign and aerosol optical depth (AOD) measurements from the ground-based remote sensing Aerosol Robotic Network (AERONET). The impacts of imaginary refractive index, aerosol mixing, and Mie method were investigated with an offline radiation model for computationally efficient evaluation. Dark brown carbon was found to increase absorbing aerosol optical depth (AAOD) by up to 5.2% in the simulation time and region. We derived a linear equation to estimate the correlation between modeled AAOD and columnar dark brown carbon ($AAOD = 0.0023[D-BrC] + 0.0007$) with core-shell considerations. Core-shell aerosol mixing and explicit Mie computation enhanced increases in AAOD from dark brown carbon at AERONET sites (up to 6.42%). These results indicate that models omitting dark brown carbon may underestimate aerosol absorption in wildfire-influenced regions, and that including it, along with a realistic mixing state and Mie treatment, is important for estimating radiative forcing as wildfire activity increases.

1 Introduction

Carbonaceous aerosols emitted by wildfires play an important role in global and regional climate systems, directly through absorption (warming) or scattering (cooling) of solar radiation, and indirectly by acting as surfaces for clouds to form (Chylek



and Wong, 1995; Dennison et al., 2014; Intergovernmental Panel on Climate Change (IPCC), 2023a; Jacobson, 2014). Many organic aerosols scatter light and cool the Earth's surface below; however, black and brown carbon are absorbing carbonaceous aerosols contributing to global positive radiative forcing (Ackerman and Toon, 1981; Chapman et al., 2009; Chou and Suarez, 1994; Chylek and Wong, 1995; Feng et al., 2013; Chou and Suarez, 1999).

Wildfires emit a diverse mixture of carbonaceous aerosols including black carbon (BC) and brown carbon (BrC) (Abatzoglou and Williams, 2016; Dennison et al., 2014; Ichoku et al., 2008; McClure et al., 2020; Wooster et al., 2005). Black carbon absorbs solar radiation across a wide spectral range (ultraviolet to infrared) and has a well-constrained, wavelength independent imaginary refractive index (k), which represents the absorbance of a particle (Bond and Bergstrom, 2006). BrC is classified as the non-BC absorbing fraction of organic aerosols. The water-soluble organic carbon (WSOC) fraction of BrC absorbs light from the visible to near-ultraviolet (300–400 nm) spectral range (Laskin et al., 2015). WSOC BrC can be measured by collecting particles on Teflon filters followed by water and methanol extraction, with insoluble compounds filtered out with in-line syringe filters (Zeng et al., 2020). Dark brown carbon (d-BrC), also known as tar balls or chromophores, is a subtype of brown carbon that is made up of water-insoluble organic carbon (WIOC), absorbs over a wide spectral range (visible and near-infrared), and is resistant to daytime photobleaching with atmospheric lifetimes up to 41 days (Alang and Aggarwal, 2024; R. K. Chakrabarty et al., 2010; Rajan K. Chakrabarty et al., 2023; Chelluboyina et al., 2024; Fleming et al., 2020; Laskin et al., 2015; Shrivastava, 2025; Xuan Wang et al., 2025). Dark brown carbon has been measured with aerosol collection through particle inlets followed by scanning electron microscopy (SEM) (R. K. Chakrabarty et al., 2010).

Dark brown carbon can contribute a direct radiative effect of $+0.208 \text{ Watts m}^{-2}$, which is comparable to BC direct radiative effect (DRE) (Xuan Wang et al., 2025). Global modeling with BrC found that direct radiative forcing (DRF) of BC may be overestimated due to previous lack of BrC DRF attributions (X. Wang et al., 2014). Adler et al., 2019 measured intermediate absorbers that could not be classified as black or brown carbon, but could potentially be dark brown carbon tar balls (Adler et al., 2019). The global annual direct radiative effect from BrC absorption was estimated to be $+0.03$ and $-0.08 \text{ Watts m}^{-2}$ at the surface (Hammer et al., 2016). A study in the Mediterranean basin considered bleaching effect and estimated a DRE of 0.04 W m^{-2} (Methymaki et al., 2023). Brown carbon in Chiang Mai, Thailand was estimated to contribute between 33% to 40% of carbonaceous aerosol absorption at 440 nm wavelengths (Ying Zhang et al., 2021). Aerosol-snow radiative transfer studies found that d-BrC increased annual mean snow radiative forcing by 0.6 to 17.9 W m^{-2} (Chelluboyina et al., 2024). Washenfelder et al., 2015 found that organic brown carbon aerosols contributed to approximately $9 \pm 9\%$ of organic aerosol absorption at 365 nm (Washenfelder et al., 2015). Experimental measurements of smoke particles from smoldering biomass combustion analyzed with SEM found that over 95% of particles were dark brown carbon tar balls (R. K. Chakrabarty et al., 2010).

Chemical transport models, such as the Weather Research and Forecasting modeling system coupled with Chemistry (WRF-Chem) and the Community Multiscale Air Quality modeling system (CMAQ), typically assume aerosols are externally or homogeneously mixed (Binkowski and Roselle, 2003; Chapman et al., 2009). Aerosol optical properties are calculated based on volume-averaging at each wavelength band. In reality, aerosols can be internally mixed and exist in a variety of morphologies. Core-shell morphologies can have a strong influence on aerosol chemistry (Anttila et al., 2006; Chen et al.,



2024; Farrell et al., 2026; Ng et al., 2025; J. Zhang et al., 2023; Yue Zhang et al., 2018) and aerosol optical properties (Ackerman and Toon, 1981; Bond and Bergstrom, 2006; Cheng et al., 2024; Toon and Ackerman, 1981).

The morphology of black and dark brown carbon can vary (externally mixed spherical particles, core shell spherical particles, or aggregates of spherules). Spherical particle morphology was found to more accurately estimate direct radiative effect in comparison to aggregate morphology (Saleh et al., 2016). Coatings on black and dark brown carbon can enhance absorption by up to 1.7 times, known as lensing enhancement (Lack et al., 2012). Recent experiments found evidence of absorption enhancement from organic and water coatings on dark brown carbon tar balls (Cheng et al., 2024). Field measurement-based modeling of brown carbon cores with coatings found that one-third of absorption in Shanghai was attributed to brown carbon (Mao et al., 2026).

Dark brown carbon, however, is not typically accounted for in regional-scale models, such as WRF-Chem. Measurements found that dark brown carbon contributed up to 75% of shortwave visible light absorption and half of longwave visible light absorption in western United States smoke plumes (Rajan K. Chakrabarty et al., 2023). The fate and transport of dark brown carbon in the southeast U.S. as well as its effects on radiative forcing are uncertain (Devi et al., 2016; Intergovernmental Panel on Climate Change (IPCC), 2023b). A laboratory experiment tested combustion of fuels representative of the southeastern United States and identified about half of BrC to be potential chromophores (Glenn et al., 2024). Although aerosol optical extinction in the southeastern United States in summer 2013 was dominated by scattering, biomass burning was found to be the dominant source of absorbance in the region (Washenfelder et al., 2015). Additionally, a significant amount of ground-based, aircraft, and remote sensing data available provides opportunity to conduct robust analysis of tar ball radiative effects in a previously uninvestigated region.

We added dark brown carbon into WRF-Chem to assess the regional radiative effects in the southeast United States. We simulated the southeast U.S region from June 21st to June 30th, 2013 and evaluated WRF-Chem simulated chemical species and radiative forcing with in-situ aircraft measurements of aerosol extinction and remote sensing measurements of aerosol optical depth (AOD). Offline radiation sensitivity simulations were conducted on the dark brown imaginary refractive index, aerosol mixing method, and explicit Mie computation impacts on absorbing aerosol optical depth (AAOD).

2 Methods

2.1 WRF-Chem model configuration

We ran WRF-Chem version 4.2 from June 21st to June 30th 2013 with an hourly output of chemical species concentrations, transport, and feedbacks with weather. Physical processes such as wet and dry removal were applied to all species. Weather was reinitialized every three simulation days. This model was run with a 12-kilometer grid spacing, 45 vertical layers, and a Lambert conformal map projection. Boundary conditions were obtained from the European Centre for Medium-Range Weather Forecasts (ECMWF) Reanalysis version 5 (ERA5) with analysis nudging for weather and the Community Atmosphere Model with Chemistry (CAM-Chem) (Hersbach et al., 2020). The Model for Simulating Aerosol Interactions and Chemistry



(MOSAIC) with 4 aerosol size bins was used for the aerosol treatment (Zaveri et al., 2008). MOSAIC is an aerosol model that couples chemical and microphysical processes and accounts for thermodynamic partitioning. The Statewide Air Pollution Research Center 1999 (SAPRC-99) chemical mechanism was used for gas-phase species (Chen et al., 2015). We used the Rapid Radiative Transfer Model for General Circulation Model Applications (RRTMG) for the longwave and shortwave radiation schemes (Iacono et al., 2008; Mlawer et al., 1997). The Morrison double-moment scheme was used for microphysics (Morrison et al., 2009). The RRTMG shortwave scheme calculates extinction based on water vapor, Rayleigh scattering, gases (carbon dioxide, ozone, oxygen, methane and nitrogen), aerosols, and clouds. RRTMG shortwave radiation flux is quantified for 14 spectral bands at 0.2 to 12.2 μm . The Yonsei University (YSU) scheme was used for the planetary boundary layer scheme (Hong et al., 2009).

We used the 2014 National Emissions Inventory for anthropogenic emissions and the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.1 for biogenic emissions (A. Guenther et al., 2006; A. B. Guenther et al., 2012). The MEGAN model uses plant functional types to model biogenic emissions. High resolution spatial and temporal data for fire emissions of biomass burning species including organic carbon (OC), black carbon (BC), sulfur dioxide (SO_2), fine particulate matter ($\text{PM}_{2.5}$), ammonia (NH_3), nitrogen oxide (NO), carbon dioxide (CO_2), carbon monoxide (CO), ethane, propylene, methyl ethyl ketone, propane, n-butane, i-butane, acetone, methane, formaldehyde, and acetaldehyde were generated with the Quick Fire Emissions Dataset (QFED) (Darmenov and Silva, 2015). QFED emissions are quantified with a top-down fire radiative power method. Utilizing fire radiative power improves estimates of biomass burning emissions in comparison to using emission factors (Wooster et al., 2005).

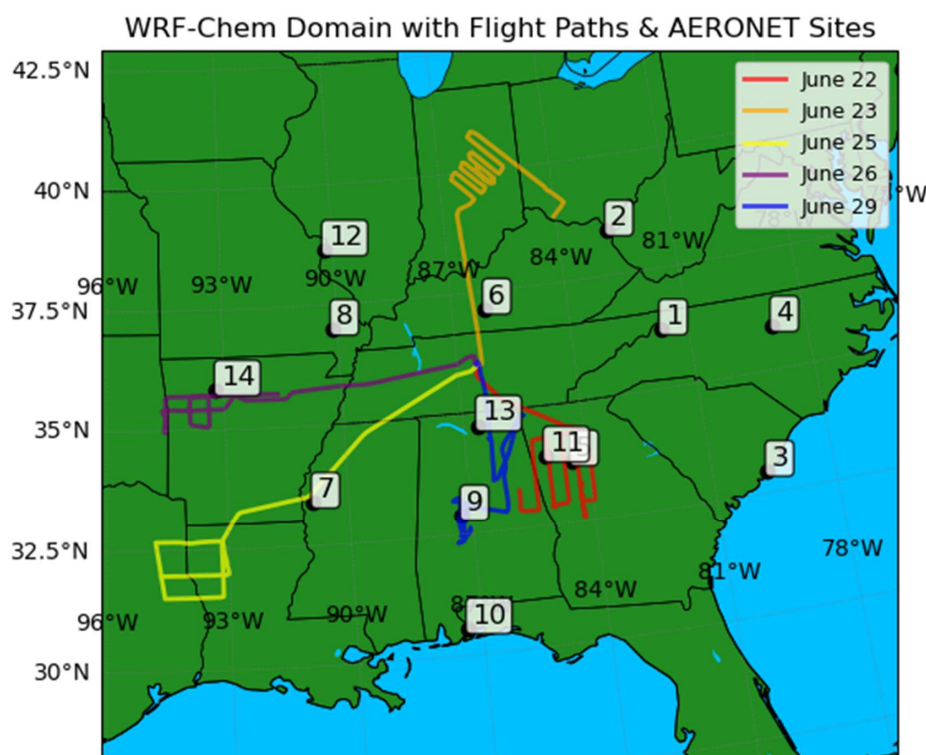


Figure 1. Locations of aircraft flights and AERONET remote-sensing measurement site. The corresponding AERONET site labels can be found in Table S1.

2.2 Aircraft measurements

120 The Southeast Nexus (SENEX) campaign conducted flights on June 22nd, 23rd, 25th, 26th, and 29th and collected aerosol, gas, and radiation measurement data aboard the National Oceanic and Atmospheric Administration (NOAA) WP-3D aircraft (Warneke et al., 2016). Dry, humidified, and thermodenuder aerosol extinction were quantified with an aerosol cavity ringdown spectrometer (Langridge et al., 2011). Aircraft CRDS measures in situ observations of aerosol extinction. Uncertainty was $\pm 2\%$ for dry measurements (Strawa et al., 2006). Dry aerosol extinction was measured at RH < 30%, 273.15 K and 101,300 Pa. Aerosol concentrations for organics, sulfate, nitrate, and ammonium were measured with an aerosol mass spectrometer (AMS) with uncertainty of $\pm 30\%$ (Warneke et al., 2016).

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2.3 Ground-based remote sensing data

Aerosol optical depth (AOD) is the amount of light scattered or absorbed by aerosols in a vertical column. AOD measurements were obtained from the Aerosol Robotic Network (AERONET) version 3, Level 2.0 which is cloud screened and quality-assured with 0.01 and 0.02 uncertainty at visible and ultraviolet wavelengths, respectively (O. Dubovik et al., 2000; Oleg Dubovik et al., 2001; Holben et al., 2006). AERONET is a global ground-based system of automatic sun and sky scanning radiometers that derive AOD from direct solar radiation at wavelengths between 340 and 1040 nm (Holben et al., 1998). AERONET AOD is often used as ground truth validation for remote sensing and reanalysis AOD products (Byrne et al., 2024; Z. Zhang et al., 2024). AOD at 14 AERONET locations were used to analyze model simulations (Figure 1, Table S1).

2.4 Modeling dark brown carbon

We added primary solid-state dark brown carbon as a new species in WRF-Chem. Zhang et al., 2020 estimated 23% of d-BrC to OC, Jo et al., 2016 estimated 20% d-BrC to OC, and Park et al., 2010 estimated between 16% to 30% of d-BrC to OC (Jo et al., 2016; B. Zhang et al., 2024). In this work, primary dark brown carbon was assumed to be 20% of organic carbon from biomass burning. In our online base model, d-BrC is considered non-reactive and has no chemical interactions with other species (Fleming et al., 2020). Complex refractive indices are composed of a real (n) and imaginary (k) refractive index. Dark brown carbon real refractive indices range from about 1.4 to 1.9 and imaginary refractive indices for dark brown carbon range from 0.01 to 0.4 (Alexander et al., 2008; R. K. Chakrabarty et al., 2010). We used a refractive index of $1.49 + 0.056i$ at 550 nm for dark brown carbon in our “online base” simulation (Cheng et al., 2024). For black carbon, we used a refractive index of $1.95 + 0.79i$ at 550 nm (Bond and Bergstrom, 2006).

2.5 Offline radiation simulation

We conducted sensitivity simulations of dark brown carbon aerosol optical properties using an offline WRF Chem radiation model that can run computationally intense radiation schemes efficiently (Barnard et al., 2010; Shrivastava et al., 2013). The offline radiation model uses online WRF-Chem chemical concentrations and aerosol optical property outputs as inputs and calculates individual vertical layer AOD, single scattering albedo, and asymmetry with Mie theory (Ghan and Zaveri, 2007). We updated the offline model to include dark brown carbon and the 14 spectral bands from RRTMG. The sensitivity simulations for aerosol mixing state, Mie method, and imaginary refractive index are shown in Table 1. We used a real refractive index of 1.67 and imaginary refractive index of 0.27 at 550 nm wavelength, which is representative of the upper literature range for dark brown carbon (Alexander et al., 2008), for the high dark brown carbon absorption sensitivity.



Table 1. Offline radiation model sensitivity simulations for aerosol mixing, Mie method, and imaginary refractive index (k_{D-BrC}) at 550 nanometer wavelength.

Simulation Name	Aerosol Mixing	Mie Method	k_{D-BrC}
Base	Volume averaging	Simplified	0.00
D-BrC	Volume averaging	Simplified	0.27
Maxwell-Garnett	Maxwell-Garnett	Simplified	0.00
MG, D-BrC	Maxwell-Garnett	Simplified	0.27
Core-shell	Core-shell	Explicit	0.00
CS, D-BrC	Core-shell	Explicit	0.27
Explicit Mie	Volume averaging	Explicit	0.00
EM, D-BrC	Volume averaging	Explicit	0.27

160 2.6 Aerosol mixing

Aerosol mixing methodology impacts aerosol scattering, absorption, and single-scattering albedo. The online WRF-Chem default assumes volume-mixing for all aerosol chemical species in a given size bin for computationally efficient calculations of aerosol optical properties. With volume-mixing, refractive indices are weighted by volume for each aerosol size bin. However, volume-mixing is not physically accurate (Bond and Bergstrom, 2006). Core-shell are computationally expensive when run online. Therefore, the offline radiation model is advantageous for analyzing aerosol mixing state. The “Maxwell-Garnett” mixing simulation randomly distributes black carbon in a particle (Markel, 2016). We updated the offline model to include dark brown carbon as another species for random distribution in a particle. Lesins et al., 2002 and Schuster et al., 2005 found that volume mixing calculates refractive indices 13 to 30% greater than with the Maxwell Garnett approximation. The “core-shell” simulation uses the core-shell mixing method which assumes a core of black and a shell with all other chemical species (Ackerman and Toon, 1981; Bond and Bergstrom, 2006). We updated the model to include dark brown carbon in the core. The “core-shell” simulation uses the full Mie code (explicit Mie method) at each time step and accounts for core-shell mixing.

2.7 Mie optical property methodology

Mie theory is used for predicting absorption and scattering for spherical particles. Under Mie theory computation, absorption and scattering are dependent on complex refractive indices, particle size, and incident light wavelength. The base simulation replicates online WRF-Chem and uses Chebyshev economization with the full Mie code called a single time for computational efficiency (Fast et al., 2006). The explicit Mie method, which is not typically used for online WRF-Chem, calls Mie code for each time step and uses Mie code from Ackermann and Toon, 1981 which allows for core-shell morphology Mie calculations (Ackerman and Toon, 1981). Aerosol optical properties (single scattering albedo, extinction, and asymmetry) are calculated with a two-component model from Ackerman & Toon 1981.



2.8 Model evaluation and metrics

Aerosol extinction is the sum of both aerosol absorption and scattering. Single-scattering albedo (SSA) is aerosol scattering divided by extinction. SSA values close to 1 indicate that extinction is primarily from scattering while SSA less than about 0.85 indicate significant aerosol absorption (Haywood and Shine, 1995). Absorbing aerosol optical depth (AAOD) for the different offline sensitivity models were quantified with AOD and single scattering albedo (SSA) in the following equation:

$$AAOD = AOD \times (1 - SSA) \quad (1)$$

AOD and AAOD (τ) were calculated at 550 nm wavelength (λ) with the Angstrom exponent (α) equation:

$$\alpha = - \frac{\log \frac{\tau_{\lambda_1}}{\tau_{\lambda_2}}}{\log \frac{\lambda_1}{\lambda_2}} \quad (2)$$

$$\tau_{550} = \tau_{400} \left(\frac{550}{400} \right)^{-\alpha} \quad (3)$$

3 Results and discussion

3.1 Model-simulated dark brown carbon and black carbon

Dark brown carbon from wildfires was added to QFED and WRF-Chem. The model-simulated spatial distribution of average dark brown carbon and black carbon in a column over the simulation time period are shown in Figure 2.

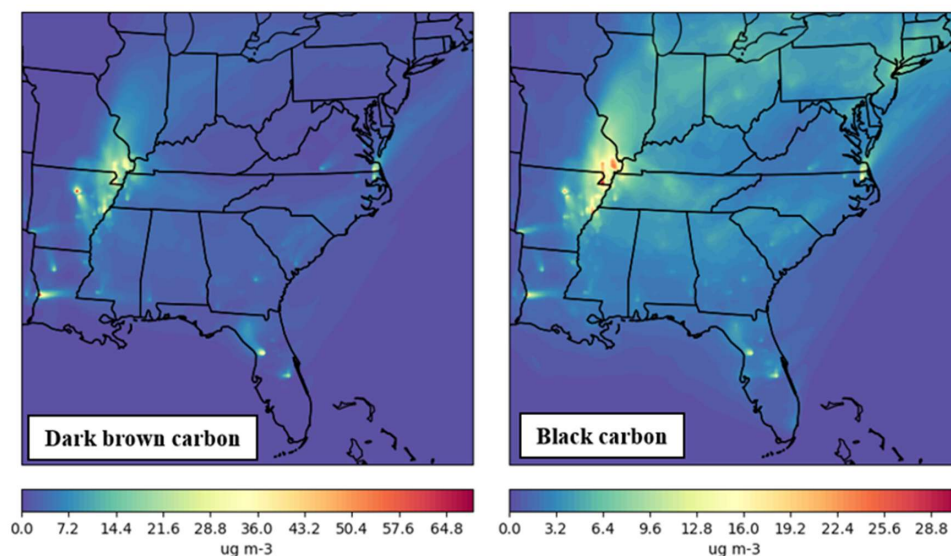


Figure 2. Predicted average total column dark brown carbon (left) and black carbon (right) concentrations simulated by WRF-Chem between June 21st and June 30th 2013.



From June 21st to June 25th, there was biomass burning in Arkansas, Tennessee, and Missouri and long-range transport of biomass burning emissions to the northeast. June 26th and 27th had biomass burning in Florida and Arkansas. On June 28th and 29th, there was biomass burning in Louisiana, Arkansas, Tennessee, and North Carolina. The average WRF-Chem modeled total columnar dark brown carbon during the time period was $1.64 \mu\text{g m}^{-3}$ (standard deviation = 2.41). While the maximum dark brown carbon in a vertical column ($68.7 \mu\text{g m}^{-3}$) was greater than maximum columnar black carbon ($30.7 \mu\text{g m}^{-3}$), the average black carbon ($2.09 \mu\text{g m}^{-3}$) in the simulation time period was greater.

3.2 Sensitivity analysis

The AAOD percent differences between the base simulation and the radiation sensitivity simulations for all grid cells and all hourly outputs during the simulation time period are shown in Table 2.

Table 2. Maximum percent difference for absorbing aerosol optical depth (AAOD) between base WRF-Chem and sensitivity simulations.

Simulation Name	D-BrC	Maxwell-Garnett	Maxwell-Garnett, D-BrC	Core-shell	Core-Shell, D-BrC	Explicit Mie	Explicit Mie, D-BrC
Maximum AAOD % increase	5.2%	0%	0%	29.7%	29.7%	30.2%	30.2%
Maximum AAOD % decrease	0%	-23.3%	-23.3%	-18.2%	-18.2%	-4.7%	-4.7%

The “D-BrC” simulation increased AAOD by up to 5.2%. The “Maxwell-Garnett” and “Maxwell-Garnett, D-BrC” simulations decreased AAOD by up to 23.3%. The “Core-Shell” and “Core Shell, D-BrC” simulations increased AAOD by up to 29.7% and decreased AAOD by up to 18.2%. The “Explicit Mie” and “Explicit Mie, D-BrC” simulations increased AAOD by up to 30.2% and decreased AAOD by up to 4.7%. The addition of D-BrC to the Maxwell-Garnett, core-shell, and explicit Mie simulations did not change the maximum AAOD percent increase or decrease. This is likely due to mixing method dominating at the locations where the maximum and minimums are located.

3.3 AERONET

Table 3 shows the mean percent bias between AERONET measurements of AOD and the base and sensitivity simulation AOD at 550 nm wavelength.



Table 3. Percent bias between AERONET and model simulation aerosol optical depth at 550 nm wavelength.

Simulation Name	Base	D-BrC	Maxwell-Garnett	Maxwell-Garnett, D-BrC	Core-shell	Core-Shell, D-BrC	Explicit Mie	Explicit Mie, D-BrC
All sites	-53.48%	-53.48%	-53.66%	-53.66%	-54.14%	-54.15%	-53.13%	-53.13%
App	-55.67%	-55.67%	-55.80%	-55.80%	-54.81%	-54.33%	-55.53%	-55.53%
Boyd	-48.45%	-48.45%	-48.63%	-48.63%	-42.51%	-42.23%	-48.27%	-48.27%
Cape	-68.72%	-68.72%	-68.82%	-68.82%	-68.86%	-71.28%	-68.63%	-68.63%
EPA	-53.54%	-53.54%	-53.69%	-53.69%	-53.15%	-53.15%	-53.33%	-53.33%
Geo	-46.19%	-46.19%	-46.43%	-46.43%	-47.89%	-47.80%	-45.86%	-45.86%
Imp	-65.24%	-65.24%	-65.38%	-65.38%	-62.79%	-62.62%	-65.04%	-65.04%
Lel	-41.05%	-41.05%	-41.24%	-41.24%	-37.08%	-41.79%	-41.37%	-41.37%
Mingo	-12.97%	-12.97%	-13.33%	-13.33%	-27.97%	-27.97%	-11.82%	-11.82%
SE-CTR	-52.64%	-52.64%	-52.82%	-52.82%	-50.78%	-50.66%	-52.33%	-52.33%
SE-OLF	-61.63%	-61.63%	-61.76%	-61.76%	-69.60%	-69.60%	-61.49%	-61.49%
SE-YOR	-30.64%	-30.64%	-30.97%	-30.97%	-32.78%	-39.87%	-30.01%	-30.01%
St Louis	-61.02%	-61.02%	-61.22%	-61.22%	-60.55%	-60.44%	-60.61%	-60.60%
UAH	-55.57%	-55.57%	-55.81%	-55.81%	-47.52%	-42.18%	-55.26%	-55.26%
UppBuf	-87.68%	-87.68%	-87.72%	-87.72%	-86.36%	-85.90%	-87.62%	-87.62%

The base simulation underpredicted AERONET AOD at all sites by 53.48% with an AOD mean bias of 0.128. The model performance varied by AERONET location. The base simulation had the lowest bias (-12.97%) at the Mingo AERONET site and the greatest bias at the Upper Buffalo AERONET site (-87.68%). The “D-BrC” sensitivity had the same average percent bias with AERONET AOD as the base simulation. The “Maxwell-Garnett” and “Maxwell-Garnett, D-BrC” sensitivity simulations slightly worsened AERONET AOD predictions by 0.18% on average for all sites in comparison to the base simulation, with the greatest change at Mingo (0.36%) and Yorkville (0.33%). The “Core-Shell” sensitivity on average for all sites worsened AERONET AOD predictions by 0.66% in comparison to the base simulation. However, the “Core-shell” simulation improved percent bias at App, Boyd, EPA, Imp, Lel, CTR, St. Louis, UAH, and Upper Buffalo by 0.86%, 5.94%, 0.39%, 2.62%, 1.86%, 0.47%, 9.05%, and 1.32%, respectively. The “Core Shell, D-BrC” sensitivity on average for all sites worsened percent bias by 0.67% in comparison to the base simulation. The “Core-shell, D-BrC” simulation improved percent bias at App, Boyd, EPA, Imp, Lel, CTR, St. Louis, UAH, and Upper Buffalo by 1.34%, 6.22%, 0.39%, 2.62%, 1.98%, 0.58%, 13.39%, and 1.78%, respectively. The “Explicit Mie” and “Explicit Mie, D-BrC” sensitivity simulations slightly improved AERONET AOD predictions by 0.35% for all sites in comparison to the base simulation, with the greatest change at Mingo (1.15%) and Yorkville (0.63%). The “Explicit Mie” and “Explicit Mie, D-BrC” sensitivity simulations slightly worsened AERONET AOD predictions by at Leland (0.32%).

The base model and all sensitivity simulations underpredicted AERONET AOD. Although QFED provides one of the most accurate predictions of biomass burning emissions, previous studies found that QFED still underestimates AOD and PM_{2.5} (Byrne et al., 2024; Wu et al., 2025) This could explain the underpredictions of AOD. Another reason for this underprediction could be the temporal resolution of AERONET. The AERONET AOD values used are daily averages while the model



simulated AOD is predicted for every hour. Therefore, differences are expected. Additionally, AOD has diurnal variations. Although the diurnal effects on AERONET AOD are minimal when averaged across all AERONET sites, individual sites can be strongly influenced by diurnal cycles (Arola et al., 2013). Table 3.4 summarizes the percent difference between model simulated AAOD with and without D-BrC at the AERONET sites.

Table 4. Percent difference between model simulations of absorbing aerosol optical depth at 550 nm wavelength with and without dark brown carbon at AERONET sites.

Sensitivity Simulation	Volume Mixing	Maxwell-Garnett	Core-shell	Explicit Mie
All sites	+0.02%	0.00%	+0.64%	+0.02%
App	+0.01%	0.00%	+0.46%	+0.01%
Boyd	+0.01%	0.00%	+1.32%	+0.01%
Cape	+0.02%	0.00%	-4.64%	+0.02%
EPA	+0.01%	0.00%	0.00%	+0.01%
Geo	+0.01%	0.00%	+0.91%	+0.01 %
Imp	+0.00%	0.00%	+0.69%	+0.01 %
Lel	+0.03%	0.00%	-1.55%	+0.04 %
Mingo	+0.01%	0.00%	-0.06%	+0.01 %
SE-CTR	+0.02%	0.00%	+3.46%	+0.02 %
SE-OLF	+0.04%	0.00%	-1.31%	+0.04 %
SE-YOR	+0.03%	0.00%	+4.35%	+0.03 %
St Louis	+0.02%	0.00%	+1.56%	+0.02 %
UAH	+0.01%	0.00%	+6.42%	+0.02 %
UppBuf	+0.02%	0.00%	+3.46%	+0.02 %

The addition of dark brown carbon increased AAOD by 0.02% for volume mixing, 0% for “Maxwell-Garnett”, 0.64% for core-shell, and 0.02% for explicit Mie on average for all sites. The base simulation (volume mixing) was minimally sensitive to the addition of D-BrC (maximum percent increase = 0.04%). The Maxwell-Garnett simulation was insensitive to the addition of D-BrC. The core shell mixing simulation was highly sensitive to the addition of D-BrC (maximum percent increase = 6.42%, maximum percent decrease = -4.64%), highlighting the importance of core-shell mixing in predictions of AAOD. The explicit Mie simulation was minimally sensitive to the addition of D-BrC (maximum percent increase = 0.04%).

3.4 Aircraft measured extinction comparison with WRF-Chem

The WRF-Chem simulated in-situ aerosol extinction was evaluated with aircraft CRDS measurements for June 22nd, 23rd, 25th, 26th, and 29th. Figure 3 shows the WRF-Chem “Core-shell, D-BrC” extinction comparison with aircraft measurements.

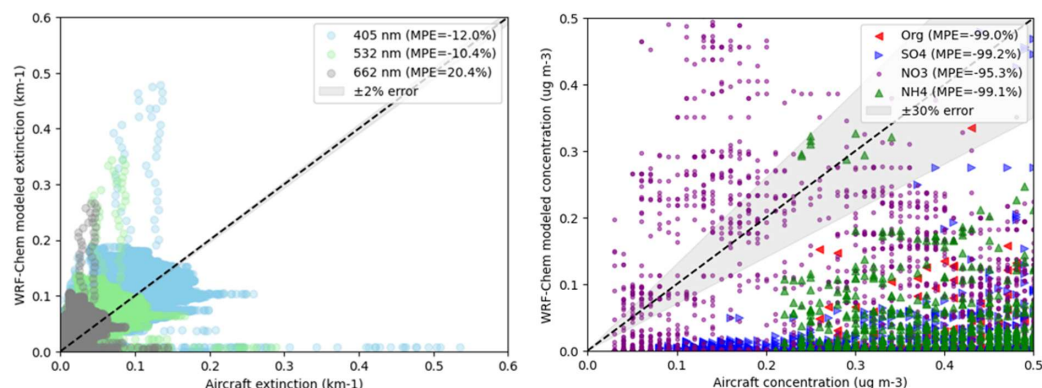


Figure 3. Comparison of aerosol extinction measured by aircraft and WRF-Chem “Core-Shell, D-BrC” simulated extinction at 405, 532, and 662 nm wavelengths (left panel). The mean percent error (MPE) is also shown. Aerosol concentrations $\leq 0.5 \mu\text{g m}^{-3}$ measured with an Aerosol Mass Spectrometer (AMS) on aircrafts were compared to WRF-Chem (right panel).

Averaged over all flights, WRF-Chem “Core-Shell, D-BrC” underpredicted aircraft extinction at 405 and 532 nm by 12.0% and 10.4% respectively, and overpredicted extinction at 662 nm by 20.4%. Major differences could be attributed to model underpredictions in aerosol loading. While sulfate, organics, and ammonium were mostly underpredicted by the WRF-Chem simulation, nitrate was overpredicted at many time points. Figure 4 shows the temporal plots of measured and modeled extinction from flights conducted on June 22nd, 23rd, 25th, 26th, and 29th.

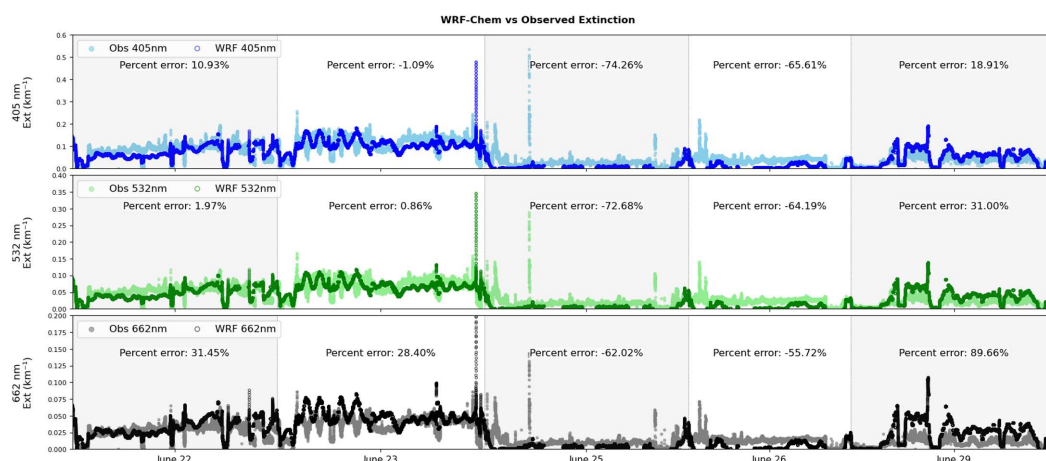


Figure 4. Aircraft measurement and WRF-Chem “Core-shell, D-BrC” modeled aerosol extinction at 405, 532, and 662 nm wavelengths over time. The timescale is not continuous between different aircraft flights.

Model performance varied with different flights. Overall, the model performed best at 405 nm and 532 nm. The model had excellent performance ($|\text{percent error}| < 5\%$) at 405 nm on June 23rd and at 532 nm on June 22nd and June 23rd. The model had poor performance ($|\text{percent error}| > 80\%$) at 662 nm on June 29th in which extinction was overpredicted by 89.66%.



3.5 Core-shell dark brown carbon and absorbing aerosol optical depth correlation

275 We investigated the relationship between core-shell dark brown carbon and absorbing aerosol optical depth with a linear regression. The correlation between dark brown carbon and absorbing aerosol optical depth is shown in Figure 5.

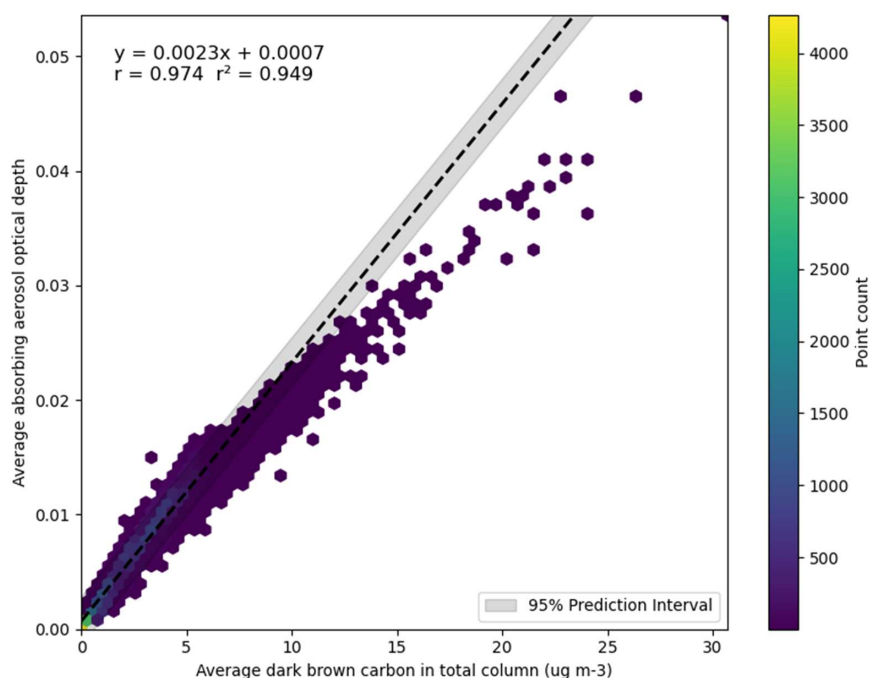


Figure 5. Correlation between modeled core-shell dark brown carbon concentration in column and absorbing aerosol optical depth in the full domain and simulation time period. The color bar represents the number of grid cells.

280 The modeled total column core-shell dark brown carbon and absorbing aerosol optical depth had a strong positive correlation with a Pearson correlation coefficient (R) of 0.745 and an R^2 of 0.554, as shown in Figure 5. This indicates a strong linear correlation. The linear regression line of best fit was $AAOD = 0.0023[D-BrC] + 0.0007$. This equation can be used to estimate increases in AAOD given dark brown carbon concentrations with core-shell considerations and using the parameters in this work. At D BrC columnar concentrations greater than $\sim 15 \mu\text{g m}^{-3}$, the correlation has reduced performance. This could
285 demonstrate a non-linear relationship between D-BrC and AAOD with core-shell considerations.

3.6 Uncertainties

In this study, we assumed 20% of organic aerosol from biomass burning. However, the emission rate of dark brown carbon from wildfires is still highly uncertain, as field measurements vary between about 15% to 80% of organic carbon classified as “dark” (R. K. Chakrabarty et al., 2010; Rajan K. Chakrabarty et al., 2023; Cheng et al., 2024). One method for identifying



290 dark brown carbon requires manual identification with a scanning electron microscope (SEM), which can result in uncertainty
(R. K. Chakrabarty et al., 2010; Rajan K. Chakrabarty et al., 2023; Cheng et al., 2024). Additionally, QFED has been found to
underestimate organic carbon loading from biomass burning (Barnard et al., 2010; Byrne et al., 2024; Wu et al., 2025). Both
black carbon and organic carbon could be underestimated in this study due to the usage of QFED. Future studies should
constrain the emissions of dark brown carbon and black carbon. While this study only considered spherical particles, particles
295 can exist as aggregates or fractals. Future work should investigate complex morphology impacts on AOD and AAOD. The
southeastern United States during summer 2013 was dominated by scattering (Washenfeller et al., 2015) so future work should
assess dark brown carbon for wildfire-specific time periods.

4 Conclusions

This study explored the radiative effects of dark brown carbon from wildfires in the eastern United States. Dark brown
300 carbon was found to increase AAOD by up to 5.2% in this simulation time period. A linear equation was derived from the
correlation between columnar core-shell dark brown carbon and absorbing aerosol optical depth that can be used to estimate
AAOD increases.

Aerosol optical depth measured at the AERONET sites was underpredicted by over 50% at all sites. Sensitivity analysis at
the AERONET sites demonstrated the importance of aerosol mixing on AAOD, as dark brown carbon increased AAOD by
305 0.02% for volume mixing, 0% for Maxwell-Garnett, 0.64% for core-shell, and 0.02% for explicit Mie on average for all sites.
At the AERONET sites, the core-shell simulation was the most sensitive to the addition of D-BrC out of all the mixing
sensitivities (maximum percent increase = 6.42%, maximum percent decrease = -4.64%). This demonstrates that the particle
mixing method for aerosol optical properties can enhance the impact of dark brown carbon on AAOD. These findings provide
insight on the imaginary refractive index, aerosol mixing, and Mie method impacts on AOD and AAOD.

310 This work emphasizes the importance of accurate particle mixing and explicit Mie calculations on AOD and AAOD. Even
so, these methods are highly computationally intense, even when run offline. Future work should aim to incorporate
computationally efficient methods for accurate particle mixing and explicit Mie into regional-scale methods, such as MieAI
(Kumar et al., 2024). This study highlights the importance of particle mixing and improves our understanding of carbonaceous
absorbing aerosols.

315

Code and data availability. Code is available upon request. Observational data from the Southeast Nexus (SENEX) 2013
field campaign was used with permission from Principal Investigators of associated instruments. Data analysis utilized WRF-
Python which can be found at <https://doi.org/10.5065/D6W094P1>.

320 **Author contributions.** All co-authors contributed to reviewing and editing the manuscript.



Competing interests. The authors declare that they have no competing interests.

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