



1 **Evaluating Model Parameterizations of Submicron Aerosol**
2 **Scattering and Absorption with In Situ Data from ARCTAS**
3 **2008**

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1 **Abstract**

2 Accurate modeling of the scattering and absorption of ultraviolet and visible radiation by
3 aerosols is essential for accurate simulations of atmospheric chemistry and climate. Closure
4 studies using in situ measurements of aerosol scattering and absorption can be used to
5 evaluate and improve models of aerosol optical properties without interference from model
6 errors in aerosol emissions, transport, chemistry, or deposition rates. Here we evaluate the
7 ability of four externally mixed, fixed size distribution parameterizations used in global
8 models to simulate submicron aerosol scattering and absorption at three wavelengths using in
9 situ data gathered during the 2008 Arctic Research of the Composition of the Troposphere
10 from Aircraft and Satellites (ARCTAS) campaign. The four models are the NASA Global
11 Modeling Initiative (GMI) Combo model, GEOS-Chem v9-02, the baseline configuration of a
12 version of GEOS-Chem with online radiative transfer calculations (called GC-RT), and the
13 Optical Properties of Aerosol and Clouds (OPAC v3.1) package. We also use the ARCTAS
14 data to perform the first evaluation of the ability of the Aerosol Simulation Program (ASP
15 v2.1) to simulate submicron aerosol scattering and absorption when in situ data on the aerosol
16 size distribution is used, and examine the impact of different mixing rules for black carbon
17 (BC) on the results. We find that the GMI model tends to overestimate submicron scattering
18 and absorption at shorter wavelengths by 10-23%, and that GMI has smaller absolute mean
19 biases for submicron absorption than OPAC v3.1, GEOS-Chem v9-02, or GC-RT. However,
20 the changes to the density and refractive index of BC in GC-RT improve the simulation of
21 submicron aerosol absorption at all wavelengths relative to GEOS-Chem v9-02. Adding in
22 situ size distribution information, as in ASP v2.1, improves model performance for scattering
23 but not for absorption, likely due to the assumption in ASP v2.1 that BC is present at a
24 constant mass fraction throughout the aerosol size distribution. Using a core-shell mixing
25 state in ASP overestimates aerosol absorption, especially for the fresh biomass burning
26 aerosol measured in ARCTAS-B, suggesting the need for time-varying mixing states in future
27 versions of ASP.

28



1 **1 Introduction**

2 Atmospheric aerosols can both scatter and absorb ultraviolet and visible (UV-VIS) light,
3 thereby altering the actinic flux and the rates of photolytic reactions in the atmosphere (e.g.,
4 Michelangeli et al., 1992; He and Carmichael, 1999). The absorption of UV-VIS light by
5 atmospheric aerosols is dominated by light absorbing carbon (LAC) and mineral dust particles
6 (Bian et al., 2003; Martin et al., 2003). Produced by the incomplete combustion of fossil fuels
7 and biomass, LAC has two major forms: “black carbon” or BC, which is primarily composed
8 of soot; and organic aerosols (OA) that strongly absorb UV-VIS light, called “brown carbon”
9 or BrC (Andreae and Gelencsér, 2006). Both forms of LAC can be internally mixed with or
10 coated by less absorbing, more reflective inorganic and organic species, altering their optical
11 properties (e.g., Liao et al., 1999; Yang and Levy, 2004; Lack and Cappa, 2010).

12 In situ and regional studies of the impact of LAC aerosols on photolysis rates have found that
13 absorbing aerosols can reduce local photolysis rates and OH concentrations by as much as
14 40% (Tang et al., 2003; Lefter et al., 2003; Alvarado et al., 2009), substantially reducing the
15 net production rate of O₃ in urban airsheds (Jacobson, 1998; Li et al., 2005) and biomass
16 burning plumes (Tang et al., 2003; Alvarado et al., 2009), with the magnitude of the impact
17 dependent on the concentrations of NO_x and VOCs (He and Charmichael, 1999; Yang and
18 Levy, 2004). Global modeling studies have found similar impacts of LAC on photolysis rates,
19 OH concentrations, and net O₃ production (e.g., Liao et al., 2003). For example, Bian et al.
20 (2003) found that the scattering and absorption of UV-VIS light by aerosols increased global
21 tropospheric mean O₃ by ~1 ppbv and decreased OH by 8%. Martin et al. (2003) found that
22 the light absorption by externally mixed black carbon aerosols decreased the modeled
23 photolysis rate of O₃ to form O(¹D) by a factor of 2 in biomass burning regions and the
24 Ganges Valley, thus decreasing OH concentrations by as much as 40%. Tie et al. (2005)
25 found 10 to 40% reductions in the formation of O(¹D) by photolysis in Europe, eastern Asia,
26 and the Amazon due to externally mixed anthropogenic and biomass burning aerosols. This
27 caused 5 to 40% reductions in HO_x concentrations along with modest changes to O₃ (-4 to
28 +5%).

29 Furthermore, the scattering and absorption of UV-VIS light by LAC aerosols can lead to a
30 significant climate forcing (the direct effect), but the magnitude of this forcing is uncertain.
31 For example, the review of Bond et al. (2013) estimated the direct radiative forcing (DRF) of
32 atmospheric BC is +0.71 W m⁻² with 90% uncertainty bounds of (+0.08, +1.27) W m⁻².



1 Chung et al. (2012) used data from the ground based Aerosol Robotic Network (AERONET,
2 Dubovik and King, 2000) to estimate a similar global DRF of $0.65 \pm 0.15 \text{ W m}^{-2}$ from all
3 LAC aerosols, but in this study brown carbon was estimated to account for ~20% of the total
4 forcing. However, Wang et al. (2014) used the GC-RT model (Heald et al., 2014) combined
5 with AERONET data to get a lower DRF estimate of 0.32 W m^{-2} from all LAC aerosols
6 (uncertainty range 0.04 to 0.50 W m^{-2}), with 34% of the forcing coming from BrC.

7 Accurately accounting for the scattering and absorption of UV-VIS light by LAC aerosols is
8 thus critical for models of atmospheric composition, air quality, and climate change.
9 However, in order to reduce computational intensity, most global chemical transport models
10 (CTMs), such as the Global Modeling Initiative (GMI) Combo model (Duncan et al., 2007) of
11 the US National Aeronautics and Space Administration (NASA) and GEOS-Chem (Bey et al.,
12 2001), account for this absorption assuming that all aerosol species are externally mixed (i.e.,
13 sulfate, sea salt, dust, OA, and black carbon aerosols are not present in the same particle), and
14 that each of these aerosol types have fixed, prescribed size distributions. These simplifications
15 can lead to substantial errors in simulating the impact of LAC aerosols on photochemistry, as
16 these impacts can vary substantially with aerosol size and mixing state. For example, the
17 studies of Liao et al. (1999) and Yang and Levy (2004) showed that internal mixtures of
18 sulfate and BC aerosols can cause larger reductions of photolysis rates than external mixtures.
19 Other theoretical (e.g., Jacobson, 2001) and observational (e.g., Schwarz et al., 2008;
20 Shiraiwa et al., 2010; Lack et al., 2012) studies suggest that coatings on BC aerosol can
21 enhance absorption by 30% or more. For example, Kim et al. (2008) showed that accounting
22 for internally-mixed aerosols and changing aerosol size distributions with time gave a much
23 smaller total negative TOA forcing (-0.12 W m^{-2}) of all carbonaceous and sulfate aerosol
24 compounds compared to the cases using one-moment scheme either excluding or including
25 internal mixtures (-0.42 and -0.71 W m^{-2} , respectively). However, core-in-shell Mie
26 calculations carried out by Lack and Cappa (2010) suggested that a black carbon particle
27 coated with brown carbon can actually absorb less light than a black carbon particle coated in
28 non-absorbing material, with reductions in absorption of up to 50% relative to clear coatings.
29 In addition, Cappa et al. (2012) found little (~6%) enhancement of BC absorption by coatings
30 in California during the US Department of Energy Carbonaceous Aerosols and Radiative
31 Effects Study (CARES) in June of 2010.

32 Thus, while the simplifications used in the global CTMs greatly reduce the computational



1 expense of global studies of the impact of LAC aerosols on photochemistry, it is important to
2 quantify the errors in the simulation of aerosol scattering and absorption that results from the
3 assumption of an external mixture and the chosen size distributions for each aerosol type. In
4 situ closure studies, like the one in this work, allow the accuracy of the aerosol scattering and
5 absorption calculations in these models to be assessed independently of the potential errors in
6 other model processes such as the treatment of aerosol emission, secondary organic aerosol
7 (SOA) formation, and aerosol wet and dry deposition. In these closure studies, ambient
8 measurements of aerosol mass and composition are used as inputs to the aerosol optical
9 property routines of the global models, with the model-calculated aerosol optical properties
10 evaluated using simultaneous in situ measurements of aerosol scattering and absorption. In
11 addition, more detailed aerosol models that allow for time varying size distributions and more
12 complicated internal mixtures of aerosol, such as AER's Aerosol Simulation Program (ASP;
13 Alvarado, 2008; Alvarado and Prinn, 2009; Alvarado et al., 2015) can also be evaluated in
14 these closure studies to help determine if the errors in the global model routines are primarily
15 due to their fixed size distributions, assumptions about external mixtures, or their assumptions
16 about the refractive indices of LAC.

17 In this study, we evaluate four aerosol optical property parameterizations used in global
18 models with in situ data on submicron aerosol scattering and absorption at three wavelengths
19 (450, 550, and 700 nm for scattering, 470, 532, and 660 nm for absorption) gathered during
20 the 2008 Arctic Research of the Composition of the Troposphere from Aircraft and Satellites
21 (ARCTAS) campaign. The four parameterizations evaluated are from the Optical Properties
22 of Aerosol and Clouds (OPAC v3.1; Hess et al., 1998) software package, the GMI Combo
23 model, GEOS-Chem v9-02 (Bey et al., 2001), and the baseline configuration of a version of
24 GEOS-Chem with online radiative transfer calculations (called GC-RT; Heald et al., 2014;
25 Wang et al., 2014). We also use the ARCTAS data to perform the first evaluation of the
26 aerosol optical property calculations in ASP v2.1, and investigate how the use of in situ size
27 distribution information and the use of different mixing rules for BC affects the match with
28 observations.

29 Section 2 describes the five aerosol optical property models examined in this study, including
30 the ASP v2.1 model, while Section 3 describes the ARCTAS data used. Section 4 summarizes
31 the methodology for the closure studies for both the global models (Section 4.1) and ASP
32 v2.1 (Section 4.2). The results of the global model and ASP closure studies are discussed in



1 Sections 5 and 6, respectively. The conclusions of the study and recommendations for future
2 model development are summarized in Section 7.

3 **2 Aerosol Optical Property Models**

4 **2.1 OPAC v3.1**

5 The OPAC package was first described by Hess et al. (1998), and version 3.1 is available
6 online at <http://opac.userweb.mwn.de/radaer/opac-des.html#ftp>. OPAC v3.1 includes
7 microphysical and optical properties of six water clouds, three ice clouds, and 10 aerosol
8 components, with size distributions and complex refractive indices chosen to represent typical
9 cases. The optical properties calculated include normalized extinction, scattering, and
10 absorption coefficients, single scattering albedo, asymmetry parameter, and the phase
11 function at 61 wavelengths between 250 nm and 40 μm for up to 8 values of relative
12 humidity. The aerosol components included are water-insoluble aerosols, water-soluble
13 aerosols, soot, two size modes of sea salt, four size modes of mineral dust, and sulfate
14 droplets. A given aerosol is then modeled as an external mixture of these ten aerosol
15 components.

16 **2.2 NASA GMI Combo Model**

17 The NASA GMI Combo model is a modular chemical transport model (Duncan et al., 2007;
18 Strahan et al., 2007; Bian et al., 2009) that includes treatment of both stratospheric and
19 tropospheric processes. Major atmospheric aerosol components included in the model are
20 sulfate, black carbon, OA, dust, and sea-salt using either GOCART (Chin et al., 2002, 2009;
21 Ginoux et al., 2001, 2004) or the University of Michigan/Lawrence Livermore National
22 Laboratory IMPACT model (Liu et al., 2007).

23 Within the NASA GMI Coupled model, the impact of aerosols on photolysis rates is
24 calculated using the FAST-JX model (v6.5). FAST-JX contains lookup tables of the
25 wavelength dependent extinction efficiencies, single scattering albedos, and phase function
26 coefficients for 14 aerosol types at 4 wavelengths (300, 400, 600, and 1000 nm) and at 7
27 values for relative humidity (0%, 50%, 70%, 80%, 90%, 95%, and 99%; see Supplement).
28 The 14 aerosol types include OA, black carbon, tropospheric, volcanic, and stratospheric
29 sulfate, two modes of sea salt, and seven modes of mineral dust. The optical properties in
30 these tables are based on the Mie theory calculations (Mishchenko et al., 2002; Martin et al.,



1 2003), which were initially performed for GEOS-Chem. The relative humidity dependent
2 complex indices of refraction and lognormal size distributions are taken from the Global
3 Aerosol Data Set (GADS) of Köpke et al. (1997), which is in turn based on OPAC v3.1
4 except that (a) all standard deviations of the modes are set to 2.0 and (b) the dry mode radius
5 of sulfate used in OPAC v3.1 (0.0695 μm) is reduced to 0.05 μm . FAST-JX then interpolates
6 the aerosol parameters contained in the look-up tables and calculates the average parameters
7 for external mixtures of the aerosol types.

8 **2.3 GEOS-Chem v9-02**

9 The aerosol optical property parameterizations within GEOS-Chem v9-02 (Bey et al., 2001)
10 follow a similar approach to the NASA GMI model, but the refractive indices and size
11 distributions of several aerosol components have been updated based on the work of Wang et
12 al. (2003a,b), Drury et al. (2010), and Jaegle et al. (2011). Table 1 shows the differences in
13 the lognormal size distribution parameters, densities, and refractive indices for the aerosol
14 types examined in this study from OPAC v3.1, GMI, GEOS-Chem v9-02, and the baseline
15 configuration of GC-RT (discussed below). In general, the geometric standard deviation of
16 the size distribution σ used in GMI was reduced from the values of 2.0 to 1.6 for tropospheric
17 sulfate, OA, and BC, and to 1.5 for the accumulation mode of sea salt. Furthermore, following
18 Drury et al. (2010) the assumed geometric mean radius (r_g) of OA was increased by a factor
19 of 3, the mean radius of BC was doubled, and the mean radius of sulfate was increased from
20 0.05 μm to 0.07 μm . Following Jaegle et al. (2011), the mean radius of the accumulation
21 mode sea salt was reduced from 0.21 μm to 0.09 μm . The refractive index of tropospheric
22 sulfate was also updated to reflect that of ammonium sulfate, rather than sulfuric acid
23 aerosols.

24 **2.4 Baseline GC-RT**

25 GC-RT (Heald et al., 2014; Wang et al., 2014) is a configuration of GEOS-Chem that is
26 coupled with the radiative transfer model RRTMG (Iacono et al., 2008) with modified aerosol
27 optical properties relative to the standard GEOS-Chem code. Here we test the aerosol optical
28 properties calculated by the “baseline” configuration of GC-RT described by Wang et al.
29 (2014). The aerosol optical property calculation in the baseline configuration of GC-RT
30 differs from GEOS-Chem v9-02 in that the BC density and refractive index are adjusted to the
31 values recommended by Bond and Bergstrom (2006), which have been found to agree better



1 with observations (Park et al, 2003; Stier et al, 2007; Kondo et al., 2011). These changes are
2 shown in Table 1.

3 **2.5 ASP v2.1**

4 ASP (Alvarado and Prinn, 2009) simulates the gas-phase, aerosol-phase, and heterogeneous
5 chemistry of young biomass burning smoke plumes, including the formation of O₃ and
6 secondary inorganic and organic aerosol. ASP is a flexible, sectional size-resolved aerosol
7 model that includes modules to calculate aerosol thermodynamics, gas-to-aerosol mass
8 transfer (condensation/evaporation), coagulation of aerosols, and aerosol optical properties.
9 ASP is generally run as a single box model, but it can be implemented as the chemistry
10 subroutine of larger Eulerian and Lagrangian chemical transport models (e.g., Alvarado et al.,
11 2009). ASP has been extensively used to study the chemical and physical transformations of
12 aerosols within biomass burning smoke plumes and the optical properties of aerosols
13 (Alvarado and Prinn, 2009; Alvarado et al., 2015) including the first simultaneous simulations
14 of the fluid dynamics, radiative transfer, gas-phase chemistry, and aerosol-phase chemistry in
15 a young biomass burning smoke plume (Alvarado et al., 2009). However, the aerosol optical
16 property routines of ASP have not been previously evaluated with in situ data.

17 In this study we are using ASP v2.1 (Alvarado et al., 2015). The modules of ASP v2.1 most
18 relevant to the current study are the modules for aerosol size distribution, thermodynamics,
19 and optical properties. These modules are described in detail below.

20 **2.5.1 ASP Aerosol Size Distribution and Thermodynamics**

21 Aerosols are represented in ASP v2.1 as a single, internally-mixed moving-center sectional
22 size distribution (Jacobson 1997, 2002, 2005). In this representation, size bin boundaries
23 remain fixed while the mean particle size within the bin is allowed to change with time, and
24 each particle in a size bin is assumed to have the same composition. In this study, the aerosol
25 size distributions were modeled at a high resolution by using 40 size bins, 38 logarithmically
26 distributed between diameters of 10 nm and 20 μm and two bins for particles smaller than 10
27 nm or larger than 20 μm. Our tests found that increasing the number of bins used in ASP v2.1
28 to 100 changed the calculated optical properties by only ~1%. In ASP v2.1, the mass fractions
29 of different aerosol components are assumed to be independent of aerosol size. This
30 assumption is likely to be violated for aerosols that contain significant amounts of BC (see
31 Section 6.2), and is planned to be relaxed in future model development.



1 The inorganic thermodynamics module in ASP v2.1 includes H₂O, NH₃, the acids HNO₃,
2 HCl, and H₂SO₄, the ions H⁺, NH₄⁺, Na⁺, K⁺, Ca²⁺, Mg²⁺, SO₄²⁻, HSO₄⁻, NO₃⁻, Cl⁻, OH⁻, and
3 their various salts (Alvarado and Prinn, 2009; Alvarado, 2008). Binary and mixed activity
4 coefficients for the various ion pairs are calculated using the Kusik-Meissner approach (Kusik
5 and Meissner, 1978) when binary coefficient data are available; otherwise they are
6 constructed using an appropriate combination of the available binary activity coefficients, as
7 in Kim et al. (1993a) and Steele (2004). Equilibrium constants for electrolyte and gas-particle
8 equilibrium reactions, as well as the deliquescence relative humidities (DRHs) of the
9 electrolytes have been updated to match ISORROPIA II (Fountoukis and Nenes, 2007). The
10 water associated with the inorganic aerosol is calculated via an iterative solution based on the
11 Gibbs-Duhem equation (Steele, 2004; Alvarado, 2008). Equilibrium concentrations of the gas
12 and aerosol species are then calculated using the Mass Flux Iteration (MFI) approach of
13 Jacobson (2005). This approach to inorganic aerosol thermodynamics compares well with
14 other inorganic aerosol thermodynamics models such as ISORROPIA (Nenes et al., 1998;
15 Fountoukis and Nenes, 2007), as shown by Steele (2004) and Alvarado (2008).

16 The ASP organic aerosol thermodynamics routine is based on the assumption that the organic
17 aerosol species can partition to both the aqueous phase containing H₂O and the inorganic ions,
18 and to a hydrophobic organic phase consisting solely of organic compounds (Alvarado,
19 2008); this approach is based on the one used in the Model to Predict the Multiphase
20 Partitioning of Organics (MPMPO) of Pun et al. (2002). Partitioning of organics between the
21 gas and hydrophobic phase is governed by Raoult's law, while the partitioning of organics
22 into the aqueous phase is governed by Henry's law. Following Pun et al., we assume that (1)
23 there is no interaction between the aqueous phase inorganic ions and the aqueous phase
24 organics and (2) the activity coefficient for the organic ions (formed by the dissociation of
25 organic acids) are equivalent to those of the corresponding molecular solute. Equilibrium
26 parameters and activity coefficient estimates were taken from available data or estimated
27 using structure-activity relationships such as the Myrdal and Yalkowsky (1997)
28 parameterization for organic vapor pressures and the UNIFAC group contribution method
29 (Fredenslund et al., 1977). These parameters can be uncertain to a factor of 10 or more,
30 however the surrogate compounds used in ASP in this study to represent water-soluble and -
31 insoluble organic aerosol (CBIO and POA1, see Section 4.2 below) both have very low vapor
32 pressures and thus partition almost completely to the aerosol phase, so these uncertainties
33 have little impact on this study.



1 For aqueous organic solutes, we have updated the calculation of the associated water content
2 to use the “kappa” (κ) parameterization of organic hygroscopicity of Petters and Kreidenweis
3 (2007). As in Alvarado and Prinn (2009), the water content for the aqueous organic solutes is
4 then added to the inorganic aerosol water content calculated as described above. In this study,
5 we assume that $\kappa = 0.04$ for the organic aerosol, corresponding to an O:C ratio of 0.25
6 (Jimenez et al., 2009). While this value may be highly uncertain, the fact that the aerosol
7 optical property measurements in this study were generally made at very low relative
8 humidity (below 20%, see Section 4.1) minimizes the impact of this assumption.

9 **2.5.2 ASP Aerosol Optical Properties**

10 As part of this work, we have extensively updated the calculations of aerosol optical
11 properties within ASP v2.1 beyond those described by Alvarado (2008) and Alvarado and
12 Prinn (2009). We have implemented spectrally-varying complex refractive indices for
13 wavelengths between 250-700 nm for five aerosol components (H_2O , soot, sulfate, sea salt,
14 and OA) based on those from OPAC v3.1 (Hess et al., 1998; see also Section 2.1 above).
15 Similar to the procedure used in the NASA GMI Combo model and GEOS-Chem, we assume
16 (1) that organics follow the OPAC v3.1 refractive indices of so-called “water-soluble
17 particulate matter”, (2) that all sulfate and nitrate salts follow the OPAC sulfate indices, (3)
18 that all chloride salts follow the OPAC sea salt indices, and (4) that all BC follows the OPAC
19 soot indices. The real refractive index of the inorganic aqueous solution (if present) is
20 calculated using the molar refraction approach of Tang (1997) and Tang et al. (1997).

21 As in Alvarado and Prinn (2009), we assume here that all particles are spherical for the
22 purposes of calculating their optical properties. ASP v2.1 has also been updated to include
23 four mixing rules for the refractive indices of black carbon and the other aerosol components:
24 (1) a volume-average (VA) dielectric constant mixing rule for all aerosol components; (2) a
25 core-shell (CS) mixing rule, where a spherical core of BC is surrounded by a spherical shell
26 of all other aerosol components (with the refractive index of the shell calculated using the
27 volume-average dielectric constant mixing rule); (3) the Maxwell Garnett (MG) mixing rule,
28 which assumes that BC is present in randomly distributed inclusions within the (Maxwell
29 Garnett, 1904); and (4) an external mixture (EXT) of BC and the other aerosols components,
30 with both sets of particles having the same size distributions but with the relative number of
31 particles determined by the relative volume concentrations. Mie calculations of aerosol optical



1 properties for each bin of the size distribution are performed within ASP using the publicly
2 available program DMiLay, which is based on the work of Toon and Ackerman (1981).

3 **3 ARCTAS Data**

4 The objective of the NASA ARCTAS campaign (Jacob et al., 2010) conducted in April and
5 June-July 2008 was to better understand the factors driving current changes in Arctic
6 atmospheric composition and climate. It used chemical and radiative measurements from
7 three research aircraft (DC-8, P-3, B-200) to interpret and augment the continuous
8 observations of Arctic atmospheric composition from satellites. The aircraft were based in
9 Alaska in April (ARCTAS-A) and in western Canada in June-July (ARCTAS-B). The focus
10 of ARCTAS-A was to examine the long-range transport of anthropogenic pollution to the
11 Arctic, while ARCTAS-B was more focused on the impacts of boreal forest fires on regional
12 and global atmospheric composition. The summer ARCTAS-B deployment was preceded by
13 one week of flights over California sponsored by the California Air Resources Board (CARB)
14 to address regional issues of air quality and climate forcing.

15 Here we use data from the DC-8 aircraft during all three phases of ARCTAS, as described in
16 detail below, to evaluate the aerosol optical property models. All analyses in this study used
17 the “merged” data set averaged to the 10s time resolution of the Ultra-High Sensitivity
18 Aerosol Spectrometer (UHSAS).

19 **3.1 Aerosol Mass Concentrations and Composition**

20 On the NASA DC8, submicron black carbon mass was measured with the University of
21 Tokyo Single Particle Soot Photometer (SP2; Moteki and Kondo, 2007, 2008). The SP2
22 measures BC volume for particles with volume equivalent diameters between 90 nm and 1.0
23 μm . The measured BC volume is then converted to BC mass using an assumed density of 1.8
24 g/cm^3 . The uncertainty in the BC mass measurements is $\pm 30\%$.

25 An Aerodyne high-resolution, time-of-flight aerosol mass spectrometer operated by the
26 University of Colorado, Boulder (HR-ToF-AMS, hereafter AMS; DeCarlo et al., 2008;
27 Cubison et al., 2011) was used to measure ammonium, chloride, nitrate, sulfate, and organic
28 aerosol mass concentrations. The AMS primarily samples submicron aerosols, with 0%
29 transmission for vacuum aerodynamic diameters below 35 nm and an approximate PM_{10} size
30 cut in vacuum aerodynamic diameter (DeCarlo et al., 2004; Canagaratna et al., 2007). The



1 uncertainty of the AMS mass concentrations measurements (2σ) is $\pm 34\%$ for the inorganics
2 and $\pm 38\%$ for the organics.

3 In addition, data on the concentration of water-soluble organic carbon (WSOC) in submicron
4 aerosol is provided by the Particle In Liquid Sampler (PILS-WSOC) of the Georgia Institute
5 of Technology (Sullivan et al., 2006), with an uncertainty of $\pm 45\%$. The measured WSOC
6 was converted to total organic mass using a factor of 1.6. This value is uncertain to at least
7 ± 0.4 , but as our total OA concentration is determined by the AMS and the relative humidities
8 of the optical property measurements were low, this assumption has little impact on our
9 results. The PILS-WSOC data is used in the ASP closure study to separate water-soluble and
10 water-insoluble organic aerosol (see Section 4.2 below).

11 The AMS data were also supplemented with measurements of additional inorganic cations
12 from the University of New Hampshire Soluble Acidic Gases and Aerosol (UNH SAGA,
13 Dibb et al., 2003) instrument. SAGA collects non-size selected (“bulk”), isokinetically-
14 sampled aerosols onto a teflon filter. The ions are then extracted off the filter with deionized
15 water and analyzed via ion chromatography. In addition to the ions measured by the AMS,
16 SAGA provides measurements of the refractory cations sodium ($\pm 0.1 \text{ ug/m}^3$ at 1013 hPa and
17 273.15 K), potassium ($\pm 0.017 \text{ ug/m}^3$), magnesium ($\pm 0.011 \text{ ug/m}^3$), and calcium (± 0.018
18 ug/m^3). In order to convert the SAGA bulk measurements of these ions into submicron
19 concentrations consistent with the AMS time-resolution, we use the bulk SAGA data to
20 determine a bulk ratio of these refractory cations to aerosol sulfate, and to combine these
21 ratios with the AMS measured sulfate concentrations to estimate the submicron mass
22 concentrations of the refractory cations.

23 To test this procedure, we compare AMS measured submicron nitrate, ammonium, and
24 chloride mass concentrations versus the concentrations estimated with the above SAGA-based
25 procedure. The match is very good for nitrate and ammonium (not shown), but submicron
26 chloride (not shown) is larger by the SAGA based procedure, as expected since SAGA is
27 sensitive to refractory chlorides such as NaCl and the AMS is not. However, as we expect the
28 aerosol in the ARCTAS campaign to be dominated by OA and BC aerosols, this should have
29 little impact on our closure study results.



1 **3.2 Aerosol Size Distribution**

2 In this study, we use the in situ measurements of dry aerosol size distribution provided by the
3 instruments of the NASA Langley Aerosol Research Group (LARGE; Anderson et al., 1998).
4 Specifically, we use the dry aerosol size distribution data from the TSI Scanning Mobility
5 Particle Sizer (SMPS), the Droplet Measurement Technologies (DMT) Ultra-High Sensitivity
6 Aerosol Spectrometer (UHSAS), and the TSI Aerodynamic Particle Sizer (APS) Model 3321.

7 The UHSAS is our primary source of size distribution information as the size range measured
8 by the UHSAS (optical particle diameters between 60 nm to 1000 nm) measures the particles
9 most likely to affect optical properties in the UV-VIS. The UHSAS has 99 bins geometrically
10 distributed in this size range, and gathers data every 10 s. The estimated precision of the
11 UHSAS is 5% in the particle size, and 20% in the particle number concentrations in each bin.

12 The TSI SMPS measures dry aerosol size distributions in 54 size bins with geometric
13 diameters between 8.8 nm and 399.7 nm. The time resolution is slower than the UHSAS
14 (105s for the SMPS versus 10 s for the UHSAS), and so care has to be taken in matching
15 SMPS size distributions with the UHSAS, as described in Section 4.2. The estimated
16 precision of the SMPS is 5% in the particle size, and 25% in the particle number
17 concentrations in each bin.

18 The TSI APS measures dry aerosol size distributions in 13 size bins with aerodynamic
19 diameters between 0.583 μm and 7.75 μm . The time resolution is the same as the UHSAS, but
20 the measured diameters are different (optical diameter for the UHSAS, aerodynamic diameter
21 for the APS). Converting continuum-regime aerodynamic diameter D_{aero} to geometric
22 diameter D_{geo} is done through the equation:

$$23 \quad D_{geo} = D_{aero} \sqrt{\frac{X}{\rho}}. \quad (3)$$

24 where ρ is the particle density and X is the “dynamic shape factor” that accounts for the non-
25 sphericity of the particles (for spheres, $X = 1.0$, otherwise $X > 1$). The estimated precision of
26 the APS is 10% in the particle size, and 20% in the particle number concentrations in each
27 bin.



1 **3.3 Aerosol Optical Properties**

2 In this study, we use the in situ measurements of dry aerosol scattering and absorption
3 provided by the LARGE suite of instruments. During ARCTAS, LARGE measured dry total
4 aerosol scattering and hemispherical backscattering coefficients at three wavelengths (450
5 nm, 550 nm, and 700 nm) using a TSI model 3563 nephelometer with an estimated precision
6 of 0.5 Mm^{-1} . These total scattering coefficients were then corrected for truncation errors using
7 the procedure described by Anderson and Ogren (1998). A Radiance Research (RR)
8 nephelometer with a $1 \mu\text{m}$ cut cyclone measured the scattering of submicron aerosols at 532
9 nm. This data allowed an estimate of the submicron scattering at 450nm, 550 nm, and 700 nm
10 by comparison of the two nephelometers when they were sampling mainly submicron
11 particles (i.e., a fine mode fraction > 0.6).

12 Dry total and submicron absorption was also measured at three wavelengths (467 nm, 532
13 nm, and 660 nm) using a RR Particle Soot Absorption Photometer (PSAP) with an estimated
14 precision of 0.2 Mm^{-1} . These filter-based absorption measurements were corrected to in situ
15 values using two methods: one from Virkkula (2010) and one from Lack et al. (2008). These
16 two corrections can differ by about 20-30%, with Virkkula (2010) giving lower aerosol
17 absorption. Most of our analysis is based on the correction of Lack et al. (2008), but we
18 discuss the sensitivity of our conclusions to the choice of correction as well.

19 **4 Closure Study Methodology**

20 **4.1 Fixed Size Distribution Parameterizations**

21 As OPAC, GMI, GEOS-Chem, and GC-RT all share a common heritage and features (i.e.,
22 external mixtures of fixed size distributions of the various aerosol components) our closure
23 study methodology for all four parameterizations is also similar. The general procedure is
24 shown in Figure 1a. The first step is to assign the measured aerosol mass concentrations to the
25 different aerosol types. In this study we focus on tropospheric, submicron aerosol, as detailed
26 composition data is available from the AMS and SP2 for this size range, and thus we exclude
27 the mineral dust, stratospheric sulfate, volcanic sulfate, and coarse mode sea salt aerosol
28 types. The SP2-measured submicron mass of BC is assigned to the BC (for GMI, GEOS-
29 Chem, and GC-RT) or SOOT (for OPAC) aerosol types. The AMS-measured submicron OA
30 mass is assigned to the OA (for GMI, GEOS-Chem, and GC-RT) or WASO (for OPAC)
31 aerosol types. For the inorganic species measured by the AMS and SAGA, we calculate



1 “equivalent electrolytes” consistent with the measured and estimated submicron ion
 2 concentrations (see Equation 17.72 of Jacobson, 2005). The sulfate- and nitrate-containing
 3 electrolytes are then assigned to the tropospheric sulfate (for GMI, GEOS-Chem, and GC-RT)
 4 and SUSO (for OPAC) aerosol types, while the chloride-containing electrolytes are assigned
 5 to the accumulation mode sea salt type.

6 The second step involves determining the submicron scattering coefficient, absorption
 7 coefficient, single scattering albedo (SSA), and asymmetry parameter for each aerosol type at
 8 the measured wavelengths and relative humidities (RHs). This is done through linear
 9 interpolation of the values present in the look-up tables for each aerosol parameterization. As
 10 the LARGE instruments measure dry optical properties, the RH used in the interpolation
 11 should not be the ambient RH, but instead is the RH in the inlet prior to the measurement.
 12 Here, we used the measured inlet RH in all comparisons. This “dry” RH is generally near 0%,
 13 but can get as high as 20%.

14 For GMI, GEOS-Chem, and GC-RT, the tabulated properties include the extinction efficiency
 15 (Q_{ext}), effective radius (r_{eff}), and SSA (ω) for each aerosol type j . After correction for
 16 wavelength and RH as described above, the extinction coefficient ($k_{ext,j}$) for each aerosol type
 17 is calculated from these properties by the equation:

$$18 \quad k_{ext,j} = \frac{3 Q_{ext,j} m_j}{4 r_{eff,j} \rho_j}. \quad (4)$$

19 where m_j is the mass concentration for each aerosol type ($\mu\text{g}/\text{m}^3$, corrected to ambient
 20 temperature and pressure) and ρ_j is the particle density. The scattering and absorption
 21 coefficients are then calculated as $k_{scat,j} = \omega_j k_{ext,j}$ and $k_{abs,j} = (1 - \omega_j) k_{ext,j}$. The k_{ext} , k_{scat} , k_{abs} ,
 22 and ω for each aerosol type are then combined together to give the model estimate of the
 23 optical properties for the submicron aerosol mixture. For k_{ext} , k_{scat} , and k_{abs} this is a simple
 24 sum, e.g. $k_{scat} = \sum_j k_{scat,j}$, and ω is the ratio of k_{scat} to k_{ext} .

25 For OPAC, the tabulated properties include values of $k_{ext,j}$, $k_{scat,j}$, $k_{abs,j}$, and ω_j , with $k_{ext,j}$, $k_{scat,j}$,
 26 and $k_{abs,j}$ normalized to an assumed particle number concentration of 1 cm^{-3} . These normalized
 27 values are multiplied by the ratio of the measured mass concentration m_j to the assumed mass
 28 concentration for 1 particle cm^{-3} for each aerosol type. These properties are then corrected for



1 wavelength and RH and combined together as described above for the GMI and GEOS-Chem
2 parameterizations.

3 **4.2 ASP v2.1**

4 The ASP v2.1 model closure studies (see Figure 1b) differ from the other closure studies
5 mainly in the use of the data on the in situ dry aerosol size distribution from the LARGE
6 instrument suite. As noted in Section 3.2 above, this data comes from three different
7 instruments (the SMPS, UHSAS, and APS) with different time resolutions and measuring
8 techniques. Thus combining these observations into a consistent picture of the size
9 distribution is not a straightforward task. Our approach uses the UHSAS observations as the
10 core of our size distribution estimate, as the submicron aerosol optical properties of interest
11 here are likely most sensitive to aerosol within the size range of the UHSAS (60 nm – 1000
12 nm). We start by creating a “combined” size distribution with the same size resolution as the
13 UHSAS observations, but with an expanded range (i.e., 246 bins with optical diameters
14 between 8.8 nm and 10 μm). For size bins with diameters between 60 nm and 850 nm, the
15 UHSAS data is used directly. For size bins below 60 nm, SMPS data (interpolated to the
16 UHSAS size resolution) is used. As the SMPS has a lower time resolution than the UHSAS,
17 we scale the SMPS data to match the UHSAS data in the size range 60 nm to 100 nm – the
18 scale factor is the slope of the linear regression of the (interpolated) SMPS and UHSAS data
19 in this size range. For size bins larger than 850 nm, the size distribution is based on the APS
20 data, with the conversion factor between aerodynamic and geometric diameter assumed to be
21 0.8. This value is consistent with the density and shape factors of urban aerosols and solid
22 ammonium sulfate (Reid et al., 2006), and for a spherical particle is equivalent to a density of
23 1.56 g cm^{-3} . The corrected APS data is used to define a power law that describes how the size
24 distribution decays at optical diameters larger than 850 nm, and this power law is used to
25 extrapolate the UHSAS data for diameters larger than 850 nm.

26 As ASP v2.1 requires that the dry aerosol size distributions be input as a sum of lognormal
27 modes, the “combined” size distribution described above is fit to three lognormal modes (see
28 Equations 13.18 and 13.20 from Jacobson, 2005). The fitting boundaries for the three modes
29 are fixed at 8 – 80 nm, 80 – 400 nm, and 400 nm – 10 μm , as these boundaries coincide with
30 minima in the ARCTAS size distribution data.



1 The submicron aerosol mass concentrations of BC, OA, and equivalent electrolytes were
2 calculated as described in Section 4.1. OA was assumed to be fairly involatile and was
3 assigned to the species CBIO (if water-soluble) and POA1 (if not water soluble; Alvarado,
4 2008). These mass concentrations define the relative mass composition (i.e. mass fractions) in
5 the ASP modes. This aerosol composition was assumed to be the same for all three modes
6 input to ASP v2.1 – while the AMS can be used to get size-resolved composition, the
7 averaging times required for this data are large (about an hour), and thus are not useful for
8 comparison to the 10 s resolution optical property data.

9 We then used ASP v2.1 to calculate total and submicron k_{scat} , k_{abs} , and ω for wavelengths
10 between 250 nm and 700 nm (at 1 nm resolution) using each of the four mixing rules
11 described above: volume-averaged, core-shell, Maxwell-Garnett, and external mixture. These
12 were compared with the in situ measurements of optical properties from the LARGE
13 instruments.

14 **5 Fixed Size Distribution Parameterization Results**

15 **5.1 Scattering**

16 Figure 2 shows a scatterplot of the measured submicron scattering coefficient at 550 nm
17 versus the value calculated using the optical property tables of the GMI Combo model. The
18 slope and correlation coefficient (r^2) of a linear fit to the data from the entire ARCTAS
19 campaign are used in evaluating the models; these values are summarized in Table 2. We see
20 that all four parameterizations explain 70-74% of the variability (e.g., $r^2 = 0.70$ - 0.74) in the
21 observed submicron scattering at all three wavelengths, except for the GMI model at 700 nm,
22 where only 58% of the variability is explained. The slopes of the linear fits are between 0.89
23 and 1.08 for the 450 and 550 nm channels, but the 700 nm channel shows more variability,
24 with a slope of 1.19 for OPAC v3.1 and the slopes for the other models between 0.63-0.68.

25 However, Figure 2 shows that there can be substantial differences in the results for the
26 different phases of the ARCTAS campaign. The parameterizations generally work best for the
27 ARCTAS-B campaign, which sampled several fresh biomass-burning plumes and thus likely
28 had more externally mixed aerosol samples than the other two phases that sampled more aged
29 pollution. For ARCTAS-B the r^2 values were ~ 0.75 , with slopes between 0.99 and 1.15. In
30 contrast, the models generally overestimate the relatively smaller scattering coefficients of the
31 aged arctic pollution sampled during the ARCTAS-A campaign, with r^2 values of ~ 0.63 and



1 slopes between 1.5 (OPAC v3.1) and 2.0 (GEOS-Chem v9-02). The ARCTAS-CARB phase
2 shows a clear bifurcation, with some samples overestimated by a factor of 2 or more and
3 some underestimated by similar factors, suggesting two distinct types of aerosols were
4 sampled in this phase. This leads to poor r^2 values for this phase (0.25-0.39) and, as the
5 largest values are generally underestimated, slopes between 0.40 (OPAC v3.1) and 0.70
6 (GEOS-Chem v9-02).

7 We also examined the distribution of the errors (modeled value – measured value) of the
8 submicron scattering coefficient, as shown in Figure 3 for 550 nm and the GMI model. To
9 reduce the impact of the large dynamic range of the measured scattering coefficients on our
10 analysis, we examined the errors in the logarithm (base 10) of the scattering coefficients,
11 which is equivalent to the logarithm of the ratio of the modeled to measured value. The mean
12 (μ) and standard deviation (σ) of these error distributions are also summarized in Table 2. We
13 prefer these metrics over mean normalized bias (MNB), as with the logarithmic (geometric)
14 approach an overestimate of a factor of 2 and an underestimate of a factor of 2 average out to
15 no mean error, while the MNB of these two observations would be 25% due to the asymmetry
16 of overestimates and underestimates when expressed as percentages. However, the use of
17 MNB instead of μ does not substantially alter the conclusions of our study, and values for
18 MNB are also discussed below. The spread of the errors is very similar for all models and
19 wavelengths, with σ of ~ 0.25 , which is equivalent to a standard deviation of a factor of 1.8
20 about the mean. The histograms of the errors show little skew to either side of the mean value.
21 The models give a positive bias at 450 and 550 nm, with the GMI model having the lowest
22 mean bias in these channels ($\mu = 0.06$ and 0.09 , respectively, equivalent to a geometric mean
23 overestimate of 15% and 23%, and an MNB of 35% and 46%). As the 450 nm channel is
24 closest to the UV wavelengths important in photolysis, we would thus expect the GMI model
25 to perform best in modeling the impact of aerosols on photolysis rates. GEOS-Chem v9-02
26 has a slightly smaller negative bias (-0.04 , equivalent to a geometric mean underestimate of
27 9% and an MNB of 8.5%) than GMI ($\mu = -0.05$, geometric mean underestimate of 11%, MNB
28 of 16%) in the 700 nm channel. The results for GC-RT are similar to GEOS-Chem v9-02 at
29 450 and 550 nm, but the negative bias at 700 nm is about twice as large on average in GC-RT
30 as it is in GMI or GEOS-Chem v9-02.



1 **5.2 Absorption and SSA**

2 Figure 4 shows the shows a scatterplot of the measured submicron absorption coefficient at
3 532 nm versus the value calculated using the optical property tables of the GMI model. The
4 PSAP measurements have been corrected using the approaches of Lack et al. (2008) (Figure
5 4a) and Virkkula (2010) (Figure 4b). As stated in Section 3.3 above, the Virkkula (2010)
6 correction generally gives 20-30% lower aerosol absorption coefficients than the Lack et al.
7 (2008) correction. As the models tended to overestimate aerosol absorption using both
8 corrections, we discuss our results relative to the Lack et al. (2008) corrected values. Results
9 for all model-wavelength combinations using the Lack et al. (2008) correction are
10 summarized in Table 3. Unlike for scattering, the absorption coefficient slopes and
11 correlations are fairly consistent between the ARCTAS-B and ARCTAS-CARB phases of the
12 campaign, but the ARCTAS-A phase shows larger model overestimates of aerosol absorption
13 for the aged Arctic pollution sampled in that campaign. The global model parameterizations
14 can explain 65-72% of the observed variability, comparable to but a little worse than their
15 performance for scattering (see Section 5.1), with slopes between 0.75 (GMI, 660 nm) and
16 1.21 (GEOS-Chem v9-02, 532 nm).

17 Figure 5 shows the histogram of the errors in the logarithm of the submicron aerosol
18 absorption coefficient for GMI at 532 nm, while the mean and standard deviation for all
19 model-measurement combinations are summarized in Table 3. The spread of the errors
20 (measured by the standard deviation σ) is between 0.24-0.29 for all model-wavelength
21 combinations, giving a standard error of a factor of ~ 1.7 to 2.0 around the mean bias. We can
22 see that while all the models show a positive mean bias for aerosol absorption at all
23 wavelengths, the GMI model has the smallest mean bias at all wavelengths, with a maximum
24 bias at 532 nm ($\mu = 0.06$, or a 15% geometric mean overestimate, and an MNB of 39%).
25 Similar results hold when the Virkkula (2010) correction is used, but the geometric mean
26 overestimate for GMI at 532 nm increases to 55%. Thus while we can conclude the GMI
27 parameterization performs the best for submicron aerosol absorption of the global model
28 parameterizations evaluated in this study, we can only conclude that its geometric mean error
29 is within the range of 0-55%, depending on wavelength and PSAP correction. Table 3 also
30 shows that the mean overestimate in aerosol absorption in GEOS-Chem v9-02 has been
31 substantially reduced, but not eliminated, by the improved values for BC density and
32 refractive index in GC-RT (e.g., from $\mu = 0.27$ and MNB of 120% in GEOS-Chem v9-02 to μ



1 = 0.22 and MNB of 95% in GC-RT at 532 nm), and the correlation coefficients are slightly
2 improved as well.

3 Figure 6 shows the results for Single Scattering Albedo (SSA) for the GMI model at 550 nm.
4 The Lack et al. (2008) measured absorptions at 532 nm and 660 nm were used to derive an
5 absorption Angstrom exponent that was then used to estimate the observed absorption at 550
6 nm. As expected, since both the GMI scattering and absorption comparisons showed small
7 positive biases at this wavelength (see Tables 2 and 3), the GMI calculation of SSA is
8 relatively unbiased, as shown in Table 4. However, the spread of the errors is large ($\sigma = 0.05$),
9 and the correlation between the modeled and measured values is poor ($r^2 = 0.06$). We
10 explored whether averaging the observations at 1 and 5 minute intervals would reduce the
11 spread in the SSA errors and improve the correlation. However, the spread of errors only
12 decreased to $\sigma = 0.03$ when the data is averaged to 5 minute intervals, and the correlation
13 coefficient only increased to $r^2 = 0.22$. Thus, while the GMI model gives reasonable mean
14 values for SSA, the calculated value tends to be significantly wrong for any given data point.
15 In addition, the GEOS-Chem v9-02 and GC-RT SSA predictions show similar biases and
16 spread of errors, with GC-RT performing slightly better than the other models at 450 nm.

17 **6 ASP v2.1 Results**

18 **6.1 Impact of Size Distribution Data on Aerosol Scattering**

19 As expected, when ASP v2.1 is given aerosol size distribution data from the LARGE
20 instrument suite, it does a substantially better job of modeling the observed aerosol scattering
21 than the global model parameterizations discussed in Section 5.1. Figure 7 shows the
22 scatterplot and histogram of the errors in submicron aerosol scattering for ASP v2.1 at 550
23 nm. The results for a core-shell (CS) BC mixing state are shown, but the results for all other
24 mixing states are similar, as shown in Table 5. Note that there are far fewer data points in
25 Figure 7a than in Figure 2 for the GMI model (1,771 versus 10,629). This is because the ASP
26 v2.1 closure requires all three LARGE size distribution instruments to be working at the same
27 time as the AMS, SP2, and other composition instruments, which reduces the amount of
28 available data. With the size distribution data, ASP v2.1 with the core-shell mixing state is
29 able to explain 88-89% of the observed variability in aerosol scattering, with linear regression
30 slopes of 0.99, 1.00, and 1.07 at 450, 550, and 700 nm, respectively. The maximum mean
31 (logarithmic) bias is $\mu = -0.03$ (equivalent to a mean underestimate of 7%), and the standard



1 deviation of the logarithmic errors (σ) is only 0.17, equivalent to a factor of 1.5. Given that
2 the size distribution data is itself uncertain to 20% before the instruments are stitched
3 together, this is remarkably good model performance. Together this implies that ASP v2.1 is
4 able to model more than 90% of the scattering data points to within a factor of 2.

5 **6.2 Impact of Size Distribution Data and Black Carbon Mixing State on** 6 **Aerosol Absorption**

7 In contrast to the results for scattering, ASP v2.1, with aerosol size distribution data from the
8 LARGE instrument suite, has difficulty reproducing the observations for aerosol absorption.
9 Figure 8a shows a scatterplot of the measured submicron absorption coefficient versus the
10 value calculated by ASP v2.1 using a CS mixing state. While overall slope (0.93 ± 0.12) is
11 reasonable, there are clear problems in the simulation of the absorption observations from
12 ARCTAS-B and CARB, leading to a poorer correlation coefficient ($r^2 = 0.44$) and a larger
13 spread in the errors ($\sigma = 0.32$, see Figure 8b) than was seen for the global parameterizations
14 using fixed size distributions and external mixtures.

15 Table 6 summarizes the submicron absorption results averaged over all ARCTAS phases for
16 the different BC mixing states that can be used in ASP v2.1. The relatively poor correlation
17 and wide spread of errors is consistent across mixing states. As expected, the assumption of
18 an external mixture (EXT) results in the lowest modeled absorption, significantly
19 underestimating absorption at 470 nm and 532 nm, but giving very little bias at 660 nm. In
20 contrast, the internal mixtures (CS, MG, and VA) all overestimate absorption, but show much
21 less dependence of this bias on wavelength. CS and VA both give regression slopes near 1,
22 but the VA mixing state shows a larger overestimate of absorption than the CS mixing state,
23 while the MG mixing state has a lower positive bias than CS.

24 However, the results vary significantly between the different phases of ARCTAS. For
25 example, Figure 9 shows the histograms of the ASP errors when the CS mixing state is used
26 separated for the three campaigns. We can see that both the mean bias and the spread of the
27 errors vary significantly between the campaigns. For example, ASP with CS appears
28 relatively unbiased for ARCTAS-CARB ($\mu = -0.02$) but the spread of the errors is large ($\sigma =$
29 0.36). The results for ARCTAS-A show a small positive bias ($\mu = 0.07$) similar to the overall
30 GMI results ($\mu = 0.06$, see Table 3), but with a small spread in the errors ($\sigma = 0.18$). In



1 contrast, ASP with CS substantially overestimates absorption during ARCTAS-B by an
2 average factor of 2 ($\mu = 0.32$), but again shows a relatively small spread of errors ($\sigma = 0.23$).

3 These differences between the campaigns make sense when we consider the types of pollution
4 sampled during each campaign. ARCTAS-A sampled primarily aged Arctic haze particles,
5 and so the BC in these particles would be expected to be internally mixed. In contrast,
6 ARCTAS-B sampled substantial amounts of fresh biomass burning smoke, where the BC
7 would be expected to be externally mixed and thus have lower absorption per mass of BC
8 than would be calculated by the core-shell assumption. Finally, ARCTAS-CARB sampled a
9 mixture of anthropogenic pollution and biomass burning smoke from a variety of sources.
10 These aerosols are likely in a variety of mixing states and have a variety of size distributions
11 of BC particles, and thus ASP CS would be expected to show the large spread of errors seen.
12 These results point to the need for further development of the ASP model to allow for time-
13 varying mixing states and to allow the BC size distribution to vary independently of the
14 overall size distribution.

15 In order to examine the benefit that including a time-varying mixing state for BC in ASP
16 could bring, we examined a third “variable” mixing state case where CS was used for
17 ARCTAS-A and ARCTAS-CARB while EXT was used for ARCTAS-B. The results are
18 shown in Figure 10 and Table 6. This variable mixing state generally shows lower mean
19 positive biases than any of the internally-mixed states (CS, VA, MG) while showing a slightly
20 smaller spread in the errors than any of the constant mixing state cases (σ of 0.28-0.29 versus
21 0.30-0.32), more consistent with the GMI results seen in Table 3. However, the correlation
22 coefficient is still very poor ($r^2 = 0.44$ -0.45), suggesting that the assumption in ASP v2.1 of a
23 constant mixing ratio of BC throughout the overall size distribution can lead to errors in
24 submicron aerosol absorption as large as those seen in the externally-mixed, fixed-size
25 distribution global models.

26 Table 7 summarizes the results for SSA for ASP v2.1 using different mixing states. When
27 assuming internal mixtures, ASP tends to underestimate the SSA by an average of 0.01 to
28 0.04, while assuming external mixtures gives fairly unbiased results (-0.01 to 0.01). We see
29 that the “variable” mixing state gives small negative biases similar to the results from the MG
30 mixing state, but has a higher correlation coefficient (r^2 of 0.30 at 532 nm, compared to 0.20
31 for MG). The SSA correlation coefficients for ASP v2.1 for all mixing states are generally
32 higher than those for GMI or GEOS-Chem v9-02.



1 7 Conclusions

2 We performed a closure study using in situ observations of submicron aerosol concentration,
3 composition, size distribution, scattering, and absorption from the NASA ARCTAS campaign
4 to evaluate the modeling of submicron aerosol scattering and absorption in four global
5 parameterizations (those used in the GMI Combo model, OPAC v3.1, GEOS-Chem v9-02,
6 and the baseline configuration of GC-RT) as well as the smoke plume chemistry model ASP
7 v2.1. Our closure study allowed for the evaluation of the predictions of aerosol scattering and
8 absorption by these models without the complications associated with different treatments of
9 aerosol emissions, transport, chemistry, and deposition. We find that the GMI model has
10 smaller mean biases in predicting submicron aerosol scattering and absorption than OPAC
11 v3.1, GEOS-Chem v9-02, or the baseline GC-RT. On average, GMI overestimates submicron
12 aerosol scattering during ARCTAS by 15% (1σ range -34% to 100%) at 450 nm and 23% (-
13 29% to 114%) at 550 nm, while it underestimates scattering at 700 nm by -11% (-53% to
14 66%). When the Lack et al. (2008) correction is applied to the ARCTAS PSAP observations,
15 GMI overestimates submicron aerosol absorption by 10% (1σ range -41% to 104%) at 470
16 nm, by 15% (-38% to 114%) at 532 nm, and by 0% (-42% to 74%) at 660 nm. On average
17 GMI slightly overestimates mean submicron SSA during ARCTAS (0.01 ± 0.05 at 470 and
18 532 nm, 0.02 ± 0.07 at 660 nm) while GEOS-Chem v9-02 slightly underestimates it ($-0.01 \pm$
19 0.05 at 470 and 532 nm, -0.02 ± 0.06 at 660 nm), but the correlation is very poor for all
20 models, suggesting that while the mean is reasonable the models tend to have little skill
21 predicting individual data points. We also find that the changes to the treatment of BC in the
22 baseline configuration of GC-RT reduce the positive bias in modeled absorption relative to
23 that in GEOS-Chem v9-02 (e.g., from a mean overestimate of 86% in GEOS-Chem v9-02 to a
24 mean overestimate of 66% at 532 nm).

25 The use of in situ size distribution information allows ASP v2.1 to accurately simulate
26 submicron aerosol scattering with a high correlation ($r^2 = 0.88-0.89$) and very little spread in
27 the error distribution compared to the GMI model. When a core-shell (CS) BC mixing state is
28 used, ASP v2.1 underestimates aerosol scattering during ARCTAS by 7% (1σ range -37% to
29 38%) at 450 and 550 nm and 2% (-34% to 45%) at 700 nm on average. However, the ASP
30 v2.1 results for submicron aerosol absorption show a substantially lower correlation ($r^2 =$
31 $0.44-0.50$) likely due to the assumption in ASP v2.1 of a constant mass fraction of BC
32 throughout the size distribution. When a CS mixing state is used, ASP v2.1 overestimates



1 submicron aerosol absorption by 29 to 35% with a weak dependence on wavelength, while
2 ASP v2.1 with an external (EXT) mixture in ASP tends to underestimate aerosol absorption,
3 with the average errors showing a strong dependence on wavelength (-21% at 470 nm, -11%
4 at 532 nm, and 0% at 660 nm). Examination of the distribution of errors for each phase of the
5 ARCTAS campaign suggests that an external mixture is best for the fresh smoke observations
6 in ARCTAS-B, while an internally mixed core-shell approach is better for the aged Arctic
7 haze in ARCTAS-A and the anthropogenic pollution in ARCTAS-CARB. Using this
8 “variable” mixing state in ASP v2.1 leads to an average overestimate of aerosol absorption of
9 10% (1σ range -42% to 109%) at 470 nm, 17% (-38% to 124%) at 532 nm, and 23% (-37% to
10 140%) at 660 nm.

11 These results suggest that the GMI model does a reasonable job modeling submicron aerosol
12 scattering and absorption, and that future refinements to the GMI approach should focus on
13 improvements that, on average, reduce scattering and absorption in the 550/532 nm and
14 450/470 nm bands. For GEOS-Chem, adopting the baseline GC-RT BC parameters from
15 Wang et al. (2014) for the standard GEOS-Chem model would substantially improve the
16 ability of the model to simulate aerosol absorption. However, further refinements to the
17 treatment of BC and OA absorption are needed to reduce the positive bias that remains in GC-
18 RT, such as the potential of BrC absorption to decrease with atmospheric age (e.g., Forrester
19 et al., 2015). For ASP v2.1, the results show that future model development should focus on
20 improvements to the simulation of submicron aerosol absorption by adding routines that allow
21 for a more complete description of aerosol mixing state (such as in the PartMC-MOSAIC
22 model, Tian et al., 2014) and adding the ability for the BC mass fraction to vary with aerosol
23 size. In addition, similar closure studies should be performed with data from other recent field
24 campaigns, such as NASA Studies of Emissions and Atmospheric Composition, Clouds and
25 Climate Coupling by Regional Surveys (SEAC⁴RS; Toon, 2013) and the US Department of
26 Energy Biomass Burning Observation Project (BBOP; Kleinman et al., 2014) campaign, to
27 investigate how the biases in the aerosol optical property models vary with location and
28 pollution source.

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- 8



1 Table 1. Lognormal mode parameters (r_g in μm , σ unitless), density (ρ , g cm^{-3}) and refractive
 2 indices (n , unitless) at 550 nm and 0% RH of selected aerosol types from OPAC v3.1, GMI,
 3 GEOS-Chem v9-02, and GC-RT.

Model	Parameter	BC/Soot	OC/WASO	Sea Salt (Acc. Mode)	Trop. Sulfate
OPAC v3.1	r_g	0.0118	0.0212	0.2090	0.0695
	σ	2.00	2.24	2.03	2.03
	ρ	1.0	1.8	2.2	1.7
	n	1.75-0.44 <i>i</i>	1.53-0.006 <i>i</i>	1.50-10 ⁻⁸ <i>i</i>	1.43-10 ⁻⁸ <i>i</i>
GMI	r_g	0.0118	0.0212	0.2090	0.05
	σ	2.0	2.0	2.0	2.0
	ρ	1.5	1.5	2.2	1.769
	n	1.75-0.44 <i>i</i>	1.53-0.006 <i>i</i>	1.50-10 ⁻⁸ <i>i</i>	1.43-10 ⁻⁸ <i>i</i>
GEOS-Chem v9-02	r_g	0.02	0.063	0.09	0.07
	σ	1.6	1.6	1.5	1.6
	ρ	1.0	1.8	2.2	1.7
	n	1.75-0.44 <i>i</i>	1.53-0.006 <i>i</i>	1.50-10 ⁻⁸ <i>i</i>	1.53-0.01 <i>i</i>
GC-RT ^a	r_g	0.02	0.063	0.09	0.07
	σ	1.6	1.6	1.5	1.6
	ρ	1.8	1.8	2.2	1.7
	n	1.95-0.79 <i>i</i>	1.53-0.006 <i>i</i>	1.50-10 ⁻⁸ <i>i</i>	1.53-0.01 <i>i</i>

4 ^aBaseline GC-RT configuration as described in Wang et al. (2014).

5



1 Table 2. Summary of results for modeling the submicron scattering coefficient throughout the
 2 entire ARCTAS campaign with OPAC v3.1, GMI, GEOS-Chem v9-02, and GC-RT. The
 3 mean (μ) and standard deviation (σ) of the \log_{10} error distributions are shown. The correlation
 4 coefficient (r^2) and slope of the linear fit between the modeled and measured values are
 5 shown as well.

Wavelength	Metric	OPAC v3.1	GMI	GEOS-Chem v9-02	GC-RT ^a
450 nm	$\mu \pm \sigma$	0.07 ± 0.24	0.06 ± 0.24	0.15 ± 0.25	0.15 ± 0.24
	r^2	0.74	0.74	0.72	0.73
	Slope	0.89 ± 0.02	0.91 ± 0.02	1.01 ± 0.02	1.05 ± 0.02
550 nm	$\mu \pm \sigma$	0.15 ± 0.24	0.09 ± 0.24	0.17 ± 0.25	0.16 ± 0.24
	r^2	0.72	0.72	0.72	0.73
	Slope	0.95 ± 0.02	0.94 ± 0.02	1.05 ± 0.02	1.08 ± 0.03
700 nm	$\mu \pm \sigma$	0.27 ± 0.25	-0.05 ± 0.27	-0.04 ± 0.24	-0.09 ± 0.25
	r^2	0.70	0.58	0.72	0.71
	Slope	1.19 ± 0.03	0.63 ± 0.01	0.68 ± 0.02	0.63 ± 0.02

6 ^aBaseline GC-RT configuration as described in Wang et al. (2014).
 7



1 Table 3. Summary of results for modeling the submicron absorption coefficient (using the
 2 correction of Lack et al., 2008) throughout the entire ARCTAS campaign with OPAC v3.1,
 3 GMI, GEOS-Chem v9-02, and GC-RT. The mean (μ) and standard deviation (σ) of the \log_{10}
 4 error distributions are shown. The correlation coefficient (r^2) and slope of the linear fit
 5 between the modeled and measured values are shown as well.

Wavelength	Metric	OPAC v3.1	GMI	GEOS-Chem v9-02	GC-RT ^a
470 nm	$\mu \pm \sigma$	0.12 ± 0.28	0.04 ± 0.27	0.26 ± 0.26	0.20 ± 0.26
	r^2	0.70	0.72	0.69	0.70
	Slope	0.81 ± 0.06	0.83 ± 0.04	1.09 ± 0.05	0.96 ± 0.04
532 nm	$\mu \pm \sigma$	0.28 ± 0.29	0.06 ± 0.27	0.27 ± 0.25	0.22 ± 0.25
	r^2	0.70	0.71	0.68	0.69
	Slope	0.94 ± 0.06	0.94 ± 0.05	1.21 ± 0.06	1.07 ± 0.06
660 nm	$\mu \pm \sigma$	0.14 ± 0.29	0.00 ± 0.24	0.15 ± 0.25	0.09 ± 0.24
	r^2	0.68	0.68	0.65	0.67
	Slope	0.99 ± 0.10	0.75 ± 0.04	0.95 ± 0.06	0.84 ± 0.05

6 ^aBaseline GC-RT configuration as described in Wang et al. (2014).
 7



1
2 Table 4. Summary of results for modeling the submicron single scattering albedo (SSA, using
3 the correction of Lack et al., 2008) throughout the entire ARCTAS campaign with OPAC
4 v3.1, GMI, GEOS-Chem v9-02, and GC-RT. The mean (μ) and standard deviation (σ) of the
5 absolute error distributions are shown. The correlation coefficient (r^2) and slope of the linear
6 fit between the modeled and measured values are shown as well.

Wavelength	Metric	OPAC v3.1	GMI	GEOS-Chem v9-02	GC-RT ^a
450 nm	$\mu \pm \sigma$	-0.01 ± 0.05	0.01 ± 0.05	-0.01 ± 0.05	0.00 ± 0.05
	r^2	0.17	0.07	0.09	0.10
	Slope	0.44 ± 0.03	0.65 ± 0.05	0.70 ± 0.04	0.61 ± 0.04
550 nm	$\mu \pm \sigma$	-0.01 ± 0.05	0.01 ± 0.05	-0.01 ± 0.05	-0.01 ± 0.05
	r^2	0.15	0.06	0.10	0.10
	Slope	0.43 ± 0.04	0.69 ± 0.06	0.73 ± 0.04	0.66 ± 0.04
700 nm	$\mu \pm \sigma$	0.01 ± 0.06	0.02 ± 0.07	-0.02 ± 0.06	-0.02 ± 0.06
	r^2	0.14	0.03	0.11	0.10
	Slope	0.32 ± 0.02	0.78 ± 0.14	0.79 ± 0.04	0.81 ± 0.04

7 ^aBaseline GC-RT configuration as described in Wang et al. (2014).
8



1 Table 5. Summary of results for modeling the submicron scattering coefficient throughout the
 2 entire ARCTAS campaign for ASP v2.1 using different mixing states. The mean (μ) and
 3 standard deviation (σ) of the \log_{10} error distributions are shown. The correlation coefficient
 4 (r^2) and slope of the linear fit between the modeled and measured values are shown as well.

Wavelength	Metric	ASP v2.1	ASP v2.1	ASP v2.1	ASP v2.1
		CS	EXT	VA	MG
450 nm	$\mu \pm \sigma$	-0.03 ± 0.17	-0.02 ± 0.17	-0.03 ± 0.17	-0.03 ± 0.17
	r^2	0.89	0.89	0.89	0.89
	Slope	0.99 ± 0.02	1.01 ± 0.02	0.99 ± 0.02	1.00 ± 0.02
550 nm	$\mu \pm \sigma$	-0.03 ± 0.17	-0.02 ± 0.17	-0.03 ± 0.17	-0.03 ± 0.17
	r^2	0.89	0.89	0.89	0.89
	Slope	1.00 ± 0.02	1.02 ± 0.02	1.00 ± 0.02	1.01 ± 0.02
700 nm	$\mu \pm \sigma$	-0.01 ± 0.17	0.00 ± 0.17	-0.01 ± 0.17	0.00 ± 0.17
	r^2	0.88	0.88	0.88	0.88
	Slope	1.07 ± 0.03	1.10 ± 0.03	1.08 ± 0.03	1.08 ± 0.03

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1 Table 6. Summary of results for modeling the submicron absorption coefficient (using the
 2 correction of Lack et al., 2008) throughout the entire ARCTAS campaign for ASP v2.1 using
 3 different mixing states. The mean (μ) and standard deviation (σ) of the \log_{10} error
 4 distributions are shown. The correlation coefficient (r^2) and slope of the linear fit between the
 5 modeled and measured values are shown as well.

Wavelength	Metric	ASP v2.1	ASP v2.1	ASP v2.1	ASP v2.1	ASP v2.1
		CS	EXT	VA	MG	Variable
470 nm	$\mu \pm \sigma$	0.11 ± 0.32	-0.10 ± 0.32	0.16 ± 0.30	0.07 ± 0.32	0.04 ± 0.28
	r^2	0.47	0.50	0.47	0.47	0.45
	Slope	0.85 ± 0.09	0.54 ± 0.06	0.90 ± 0.09	0.79 ± 0.08	0.60 ± 0.08
532 nm	$\mu \pm \sigma$	0.13 ± 0.32	-0.05 ± 0.31	0.17 ± 0.30	0.09 ± 0.32	0.07 ± 0.28
	r^2	0.46	0.48	0.47	0.47	0.44
	Slope	0.93 ± 0.12	0.62 ± 0.09	0.98 ± 0.13	0.85 ± 0.11	0.68 ± 0.11
660 nm	$\mu \pm \sigma$	0.13 ± 0.32	0.00 ± 0.32	0.17 ± 0.30	0.09 ± 0.32	0.09 ± 0.29
	r^2	0.46	0.47	0.46	0.46	0.44
	Slope	0.97 ± 0.16	0.72 ± 0.13	1.02 ± 0.16	0.89 ± 0.14	0.76 ± 0.14

6
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1 Table 7. Summary of results for modeling the SSA (using the correction of Lack et al., 2008)
 2 throughout the entire ARCTAS campaign for ASP v2.1 using different mixing states. The
 3 mean (μ) and standard deviation (σ) of the absolute error distributions are shown. The
 4 correlation coefficient (r^2) and slope of the linear fit between the modeled and measured
 5 values are shown as well.

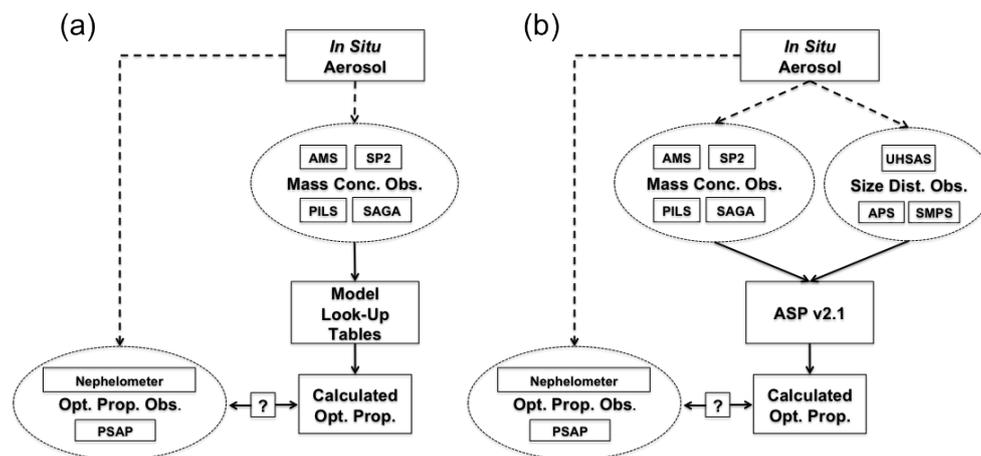
Wavelength	Metric	ASP v2.1				
		CS	EXT	VA	MG	Variable
450 nm	$\mu \pm \sigma$	-0.02 ± 0.04	0.01 ± 0.03	-0.03 ± 0.04	-0.01 ± 0.04	-0.01 ± 0.04
	r^2	0.20	0.18	0.24	0.20	0.30
	Slope	0.51 ± 0.06	0.30 ± 0.03	0.64 ± 0.07	0.46 ± 0.05	0.61 ± 0.06
550 nm	$\mu \pm \sigma$	-0.04 ± 0.04	-0.01 ± 0.03	-0.04 ± 0.04	-0.03 ± 0.04	-0.03 ± 0.04
	r^2	0.20	0.17	0.25	0.20	0.30
	Slope	0.54 ± 0.06	0.32 ± 0.03	0.67 ± 0.06	0.48 ± 0.05	0.64 ± 0.06
700 nm	$\mu \pm \sigma$	-0.02 ± 0.05	0.00 ± 0.05	-0.03 ± 0.05	-0.01 ± 0.05	-0.01 ± 0.05
	r^2	0.17	0.13	0.22	0.16	0.25
	Slope	0.44 ± 0.05	0.28 ± 0.03	0.55 ± 0.05	0.38 ± 0.04	0.52 ± 0.05

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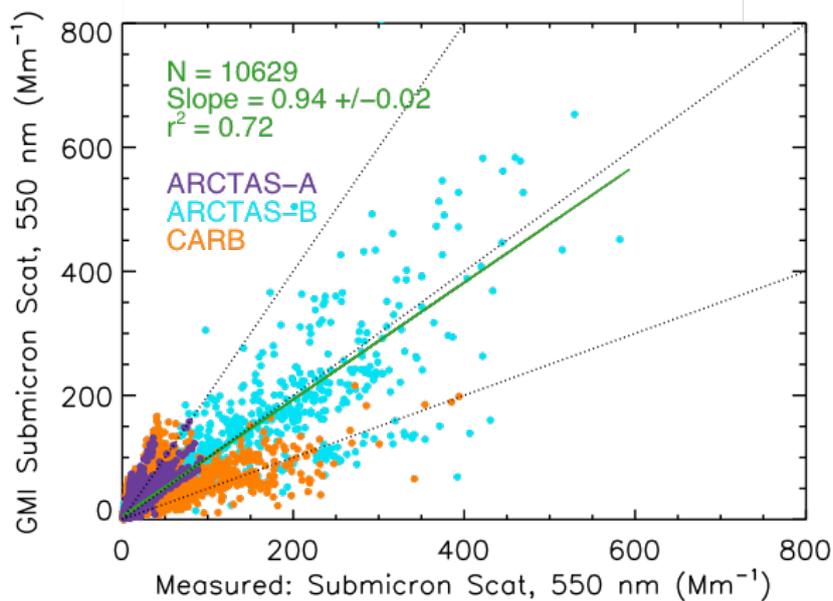
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4 Figure 1. (a) Schematic of aerosol optical property closure study methodology for the OPAC
5 v3.1, NASA GMI Combo model, GEOS-Chem v9-02, and baseline GC-RT aerosol
6 parameterizations. (b) Schematic of closure study for ASP v2.1.

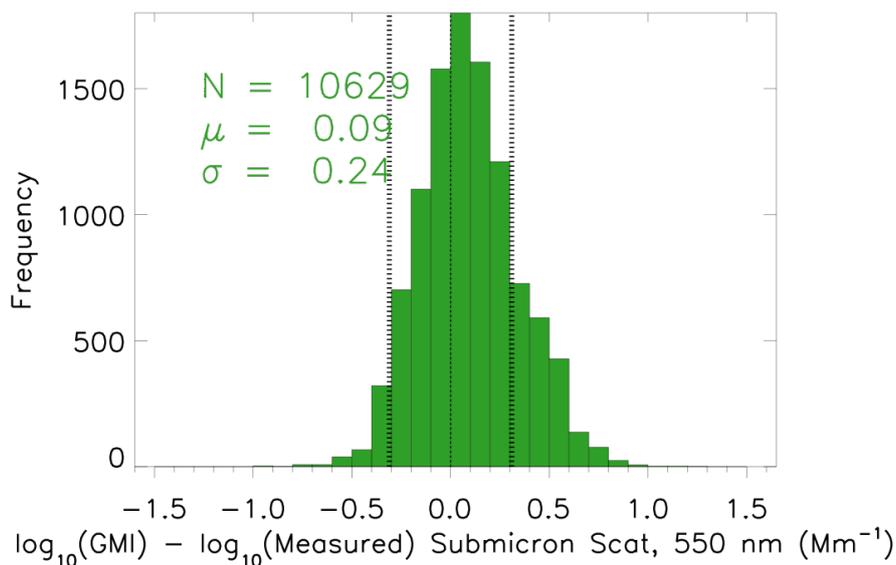


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3 Figure 2. Example scatterplot of the measured submicron scattering coefficient (Mm^{-1}) at 550
4 nm versus the calculated submicron scattering coefficient for the GMI model. The color of the
5 data points corresponds to the phase of the ARCTAS campaign (ARCTAS-A in purple,
6 ARCTAS-B in cyan, and ARCTAS-CARB in orange). The dotted black lines are the 1:1 line,
7 2:1 line, and 1:2 line. The green line is the linear fit to the data. The number of data points
8 (N), the slope of the linear fit, and the correlation coefficient (r^2) are shown as well.

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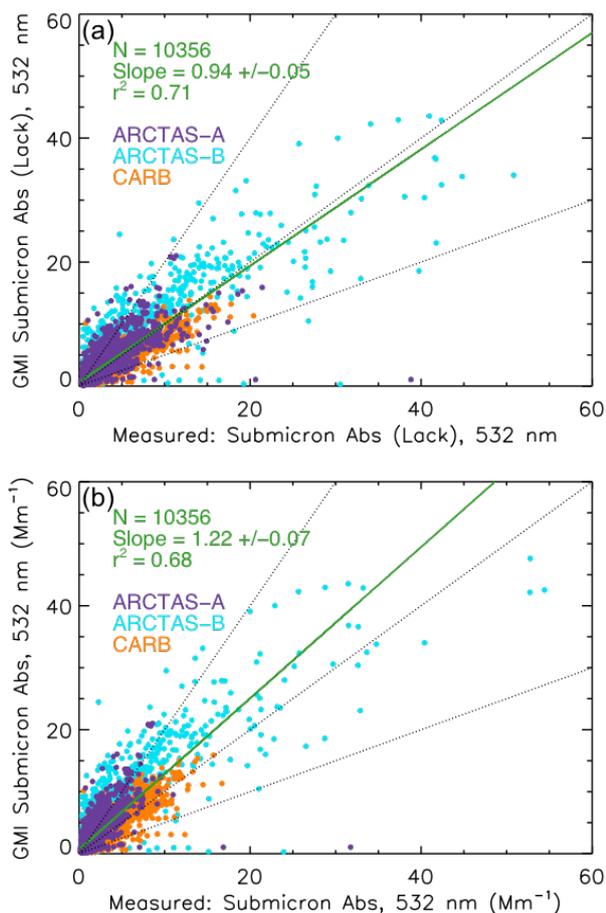
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4 Figure 3. Example histogram of the errors in the \log_{10} of the submicron scattering coefficient
5 (modeled value – measured value) at 550 nm for the GMI model. The vertical dashed lines
6 are at ± 0.31 , corresponding to an error of a factor of 2. The number of data points (N) and the
7 mean (μ) and standard deviation (σ) of the error distribution are also shown.

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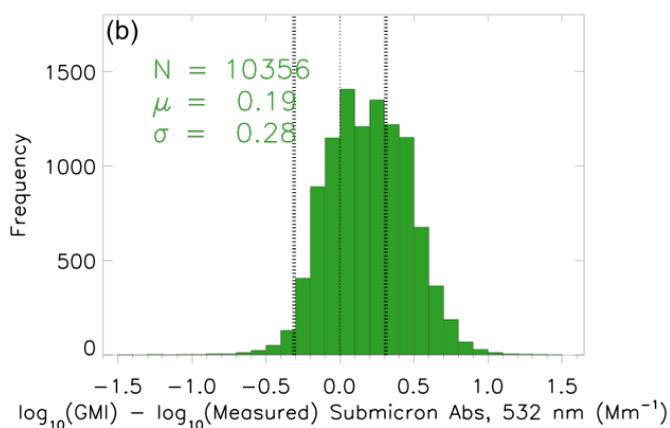
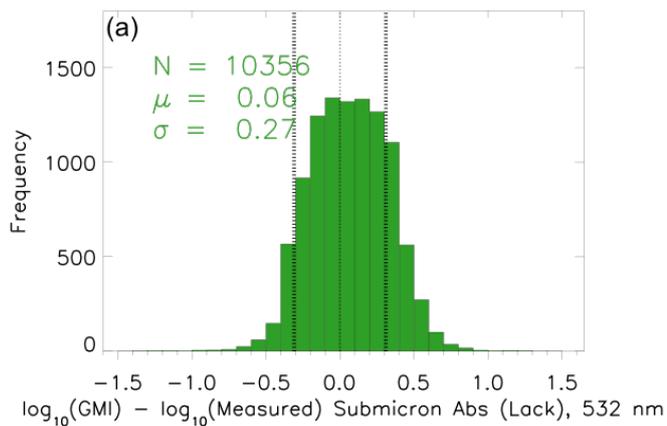
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3 Figure 4. As in Figure 2, but for submicron aerosol absorption coefficients (Mm^{-1}) at 532 nm.

4 The PSAP measured aerosol absorption has been corrected following (a) Lack et al. (2008)

5 and (b) Virkkula (2010).

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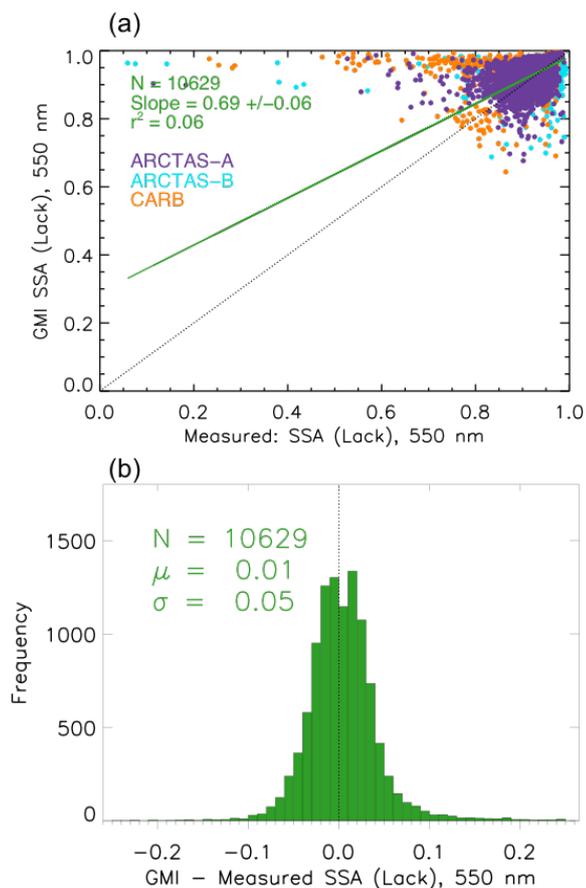
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3 Figure 5. As in Figure 3, but for submicron aerosol absorption coefficients (Mm^{-1}) at 532 nm.

4 The PSAP measured aerosol absorption has been corrected following (a) Lack et al. (2008)

5 and (b) Virkkula (2010).

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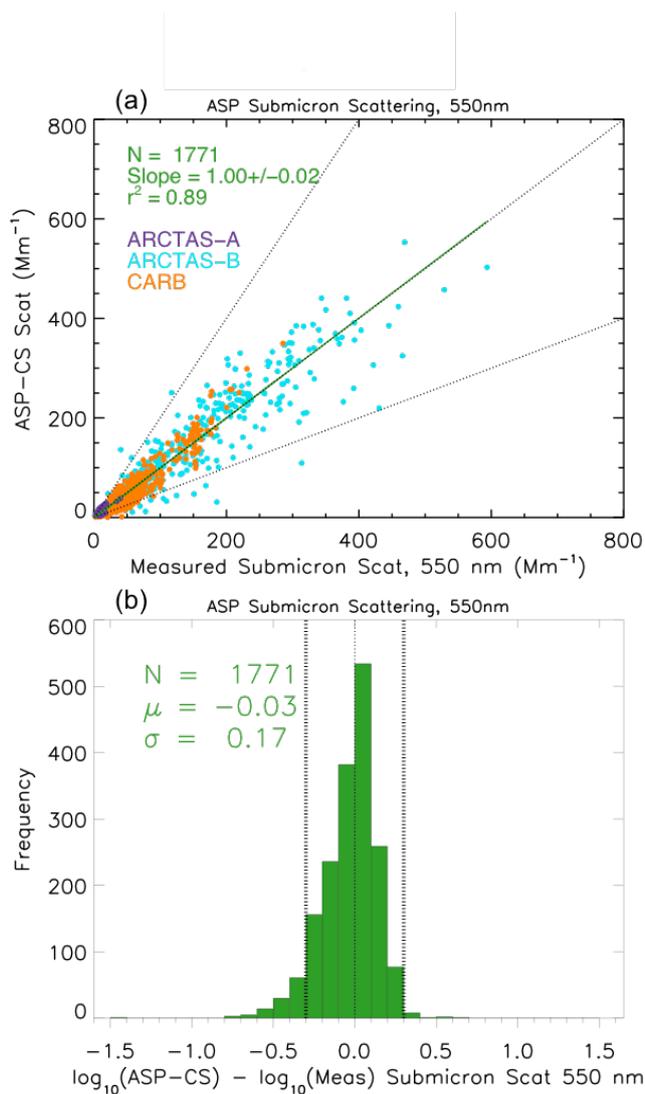


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3 Figure 6. (a) Scatterplot of the measured submicron single scattering albedo (SSA) at 550 nm
4 versus the calculated submicron SSA for the GMI model. The dotted black line is the 1:1 line.
5 The green line is the linear fit to the data. (b) Histogram of the errors in the SSA (modeled
6 value – measured value) at 550 nm for the GMI model.

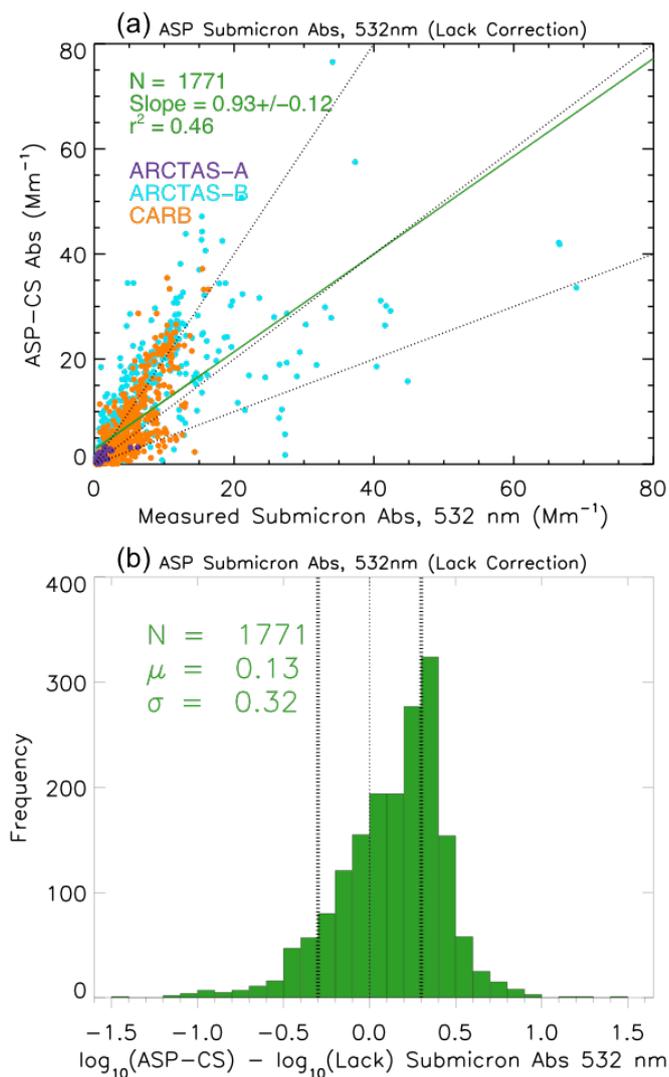
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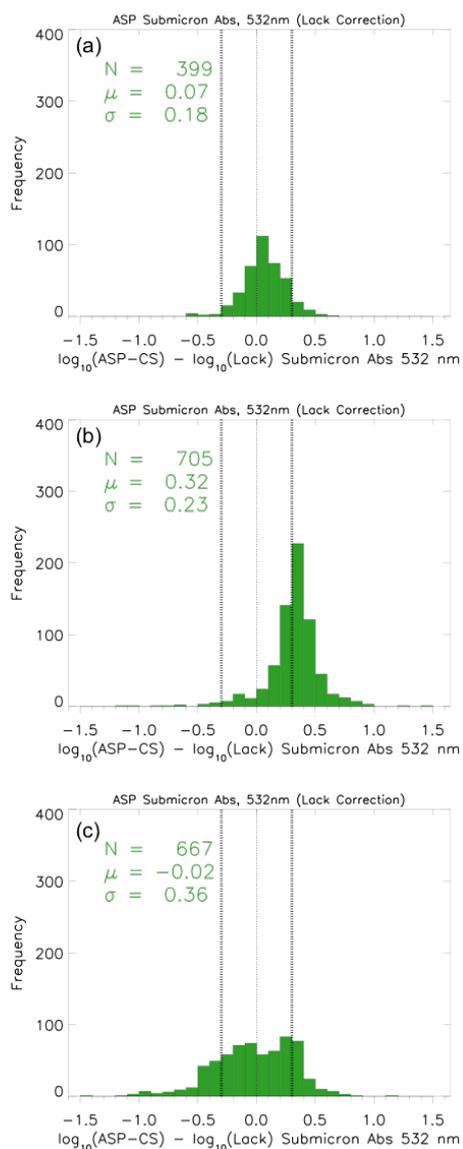
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3 Figure 7. (a) Scatterplot of the measured submicron scattering coefficient (Mm⁻¹) at 550 nm
4 versus the calculated submicron scattering coefficient for ASP v2.1. The dotted black lines
5 are the 1:1, 2:1, and 1:2 lines. The green line is the linear fit to the data. Results for core-shell
6 (CS) mixing state are shown, but the results for other mixing states are similar (see Table 5).
7 (b) Histogram of the errors in the log₁₀ of the submicron scattering coefficient (modeled value
8 – measured value) at 550 nm for ASP v2.1. The vertical dashed lines are at ± 0.31,
9 corresponding to an error of a factor of 2.



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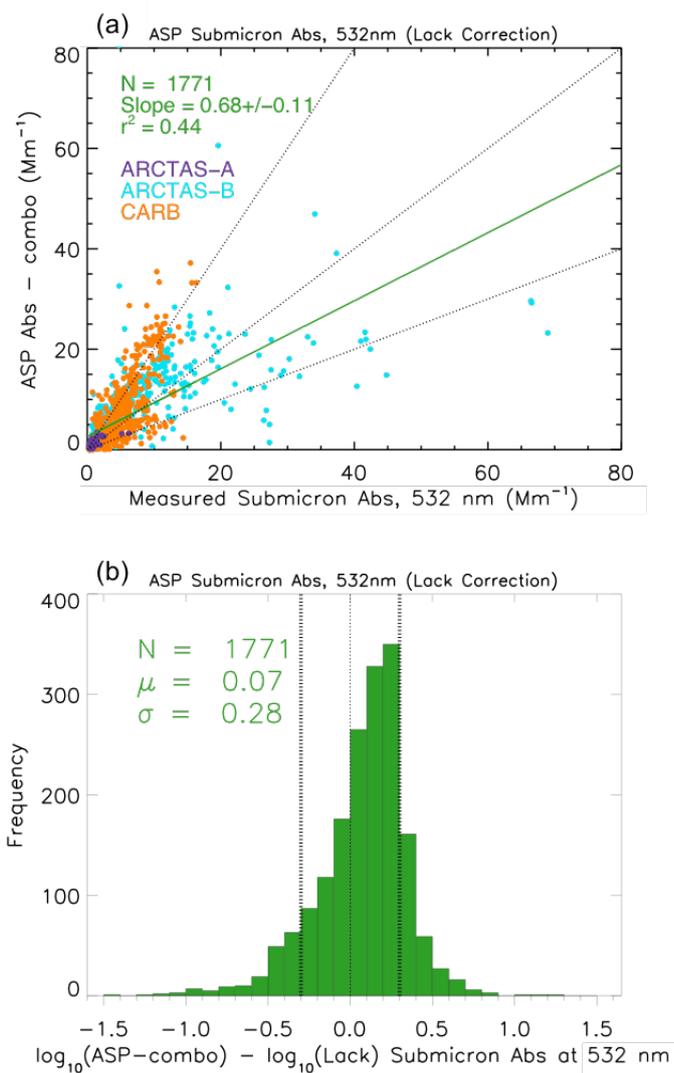
Figure 8. (a) Scatterplot of the measured submicron absorption coefficient (Mm^{-1}) at 532 nm versus the calculated submicron absorption coefficient for ASP v2.1. These results use the core-shell (CS) mixing state. (b) Histogram of the errors in the \log_{10} of the submicron absorption coefficient (modeled value – measured value) at 550 nm for ASP v2.1 with the CS mixing state.



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3 Figure 9. Histogram of the errors in the \log_{10} of the submicron absorption coefficient
4 (modeled value – measured value) at 550 nm for ASP v2.1 with the CS mixing state. The
5 PSAP measured aerosol absorption has been corrected following Lack et al. (2008). Results
6 are broken apart for (a) ARCTAS-A, (b) ARCTAS-B, and (c) ARCTAS-CARB.



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3 Figure 10. (a) As in Figure 8a, but using a variable mixing state - core-shell (CS) is used for
4 ARCTAS-A and ARCTAS-CARB, while an external mixture (EXT) is used for ARCTAS-B.

5 (b) As in Figure 8b but with this variable mixing state.

6