Response notes

Manuscript Title: AeroMix v1.0.1: a Python package for modeling aerosol optical properties and mixing states [egusphere-2024-62]

Reviewer: # 2

This manuscript is generally well-composed.

Reply: We appreciate and thank the reviewer for his/her constructive comment and recommendation. We have incorporated the necessary corrections in the revised manuscript being submitted.

Dividing the content between lines 80 to 120 into more succinct paragraphs could significantly improve readability.

Reply: Following your suggestion, we have divided section 2 into two subsections and added a table to explain pre-defined components to improve readability. The section 2 is restructured and revised as follows.

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2. Model overview

AeroMix is an open-source Python package which is developed to model the optical properties of aerosol mixtures, including AOD, SSA, asymmetry parameter, extinction coefficient (β_{ext}), scattering coefficient (β_{sca}), and absorption coefficient (β_{abs}) at sixty-one wavelengths ranging from 0.25 to 40 µm and eight relative humidity (RH) values, following Hess et al. (1998). The workflow of AeroMix for modeling the aerosol properties and assessing the mixing state is illustrated in Fig. 1. A methodology for determining the aerosol mixing state using AeroMix is detailed in the subsequent section.

2.1. Mixing of aerosols

Aerosol mixtures in AeroMix can be defined in terms of the number or mass concentration of the constituent aerosol components in external and/or internal (core-shell) mixed states, along with their vertical distribution and the vertical profile of RH. Optical properties of the complex aerosol mixing states are modeled by accounting for any number of both externally mixed particles (composed of single chemical species) and internally mixed particles (composed of multiple chemical species), with no presumed chemical or physical interaction among the particles within the mixture (see Fig. 1).

2.1.1. Externally mixed aerosol components

Nine predefined externally mixed aerosol components in AeroMix include water-insoluble (IS), water-soluble (WS), black carbon (BC), accumulation and coarse modes of sea-salt (SSam and SScm), nucleation, accumulation and coarse modes of mineral dust (MDnm, MDam, and MDcm) and stratospheric sulfate (SU). These components are represented in terms of their lognormal size distribution parameters (geometric standard deviation (σ), mode radius (r_m) and the upper and lower limits of radius, r_{min} and r_{max}), specific density (ρ), spectral refractive indices (m) and optical properties averaged for one particle ($\beta_{ext}(\lambda)$, $\beta_{sca}(\lambda)$, $\beta_{abs}(\lambda)$, SSA (λ) and $g(\lambda)$) at sixty-one wavelengths from 0.25 to 40 µm and eight RH values, adopted from the Global Aerosol Data Set and OPAC database (Koepke et al., 1997; Hess et al., 1998;

Koepke et al., 2015). A brief overview of the parameters used to describe the predefined aerosol components at dry state (0% RH) is given in Table 1. Detailed descriptions of the aerosol components, along with their size distribution parameters and spectral refractive indices, can be found elsewhere (Koepke et al., 1997; Hess et al., 1998; Koepke et al., 2015). The density of BC is set at 1.8 g cm^{-3} supported by observations, diverging from the 1.0 g cm^{-3} given in OPAC (Bond et al., 2013; Kondo, 2015). The optical properties of each component, except for MDnm, MDam, and MDcm components, are calculated using the Mie theory (Mie, 1908). This calculation assumes that the particles are spherical in shape and follow a lognormal size distribution. The calculated optical properties are normalized to one particle cm^{-3} and stored, which can then be scaled to any given number concentration. MDnm, MDam, and MDcm components are modeled using the T-Matrix Method (TMM) (Waterman, 1971; Mishchenko et al., 1999) to account for their non-sphericity (Koepke et al., 2015). In addition to the nine predefined aerosol components, AeroMix offers the flexibility to model any number of user-defined components. A new externally mixed aerosol component can be defined in AeroMix by its size distribution parameters, specific density, and spectral refractive indices described above.

Table 1: Size distribution parameters, specific density and refractive index of predefined aerosol components at dry state (0% RH).

Aerosol component	Constituting species	Mode radius (µm)	Geometric standard deviation	specific density (g cm^{-3})	Refractive index at 0.5 µm
Insoluble (IS)	Soil dust, fly ash, and non-hygroscopic organic matter.	0.471	2.51	2.0	$1.53 + 8 \times 10^{-3}i$
Water-soluble (WS)	SO_4^{2-}, NO_3^-, NH_4^+ and hygroscopic fraction of organic matter.	0.0212	2.24	1.8	$1.53 + 5 \times 10^{-3}i$
Black carbon (BC)	Black carbon aerosols	0.0118	2	1.8	$1.75 + 4.5 \times 10^{-1}i$
Sea-salt accumulation mode (SSam)	Sea salt aerosols	0.209	2.03	2.2	$1.5 + 1.55 \times 10^{-8}i$
Sea-salt coarse mode (SSam)		1.75	2.03		
Mineral dust nucleation mode (MDnm)	Desert dust	0.007	1.95	2.6	$1.53 + 7.8 \times 10^{-3}i$
Mineral dust accumulation mode (MDam)		0.39	2		
Mineral dust coarse mode (MDcm)		1.9	2.15		
Stratospheric sulfate (SU)	Sulfate aerosols from volcanic eruption.	0.0695	2.03	1.7	$1.431 + 1 \times 10^{-8}i$

The OPAC aerosol database is comprehensive, reliable, based on extensive observations, and widely employed in radiative transfer modeling, global climate modeling, and mixing state studies (Shettle et al., 1979; Deepak and Gerber, 1983; Koepke et al., 1997; Srivastava et al., 2016). Studies examining the sensitivity of refractive indices report negligible influences on modeled β_{ext} (Ramachandran and Jayaraman, 2002; Srivastava et al., 2016). Further, Srivastava et al. (2016) investigated the sensitivity of BC mode radius on SSA and β_{ext} for BC and sulfate in different mixing states, revealing differences of only up to 1.3%. Hence, the AeroMix modeled optical properties using the OPAC database are anticipated to be minimally affected by uncertainties in refractive indices and size distribution parameters. Along with the default database, AeroMix allows users to employ various datasets that characterize aerosols using the parameters described above. This flexibility not only enables users to choose datasets based on their preferences but also enhances AeroMix capability by incorporating more comprehensive datasets that consider the complex characteristics of aerosol particles, including morphology.

2.1.2 Internally mixed aerosol components

Core-shell mixed aerosol components can be defined in AeroMix by specifying the core and shell component names with their core-to-shell radius ratio (CSR) or mass fractions of the core and shell species in a core-shell mixed particle. Optical properties of the core-shell mixed aerosol components are modeled by using PyMieScatt (Sumlin et al., 2018), a coated-sphere Python Mie calculation program based on the BHCOAT program (Bohren and Huffman, 1998), which takes the spectral refractive indices of the core and shell components, and the radius of the core (r_c) and shell (r_s) as inputs.

The r_s for each particle is equivalent to the radius of the core-shell mixed particle and is assumed to follow the size distribution of the shell component (Srivastava et al., 2016), which is also supported by the observation (Arimoto et al., 2006). The r_c of each particle is calculated according to the CSR, expressed as the ratio of the r_c to the r_s, and is given by,

$$CSR = \frac{r_c}{r_s} = \left(1 + \frac{M_s \rho_c}{M_c \rho_s}\right)^{(-1/3)}, \qquad (1)$$

M_c and M_s are the mass contributions of core and shell components to the mixing, and ρ_c and ρ_s are the specific mass densities of core and shell components, respectively (Srivastava et al., 2016; Chandra et al., 2004). CSR can be specified in AeroMix either directly or in terms of the mass contribution of the core and shell components. The mass of the core-shell mixed components is calculated from their size distribution parameters and effective mass density (ρ_{eff}). The ρ_{eff} of the core-shell mixed particle can be defined as,

$$\rho_{eff} = \frac{M_c + M_s}{V}, \qquad (2)$$

where V is the volume of the core-shell mixed particle. Since M_c, M_s, and V vary along the particle size distribution as a function of r_c and r_s, ρ_{eff} needs to be defined in terms of parameters that remain fixed across the size distribution for modeling simplicity. For this, M_c and M_s can be written as,

$$M_c = \rho_c \frac{4}{3}\pi r_c^3 \ \text{ and } \ M_s = \rho_s \frac{4}{3}\pi (r_s^3 - r_c^3). \qquad (3)$$

Since, $CSR = \frac{r_c}{r_s}$,

$$\rho_{eff} = \rho_c CSR^3 + \rho_s (1 - CSR^3). \qquad (4)$$

Equation (4) contains only the terms of the specific density (ρ) of core and shell components and CSR, which remain the same across the size distribution.

2.2. Vertical profiles of aerosols and relative humidity

For total column AOD calculation, up to six vertically arranged layers can be defined to specify the vertical distribution of aerosols. Unlike OPAC, in AeroMix, aerosol concentrations at the layer base, aerosol profile type and layer mean RH can be defined separately for mixed layer, free troposphere, stratosphere, and elevated aerosol layers. In contrast, OPAC uses constant background extinction coefficient values and RH for the free troposphere and stratosphere. The vertical profile of aerosol concentration in each layer can be modeled as homogenous or as an exponential function given by,

$$N(h) = N(0)exp\left(-h/_z\right), \quad (5)$$

where *N(0)* is the number concentration of aerosol at the layer bottom, h is the height from the layer bottom in kilometers, and *z* is the scale height in kilometers representing the change in aerosol concentration with height (Hess et al., 1998). Standard exponential profiles for different aerosol types provided by Hess et al. (1998) can be utilized for locations lacking aerosol vertical profile measurements. Alternatively, when measured aerosol profiles are available, they can be modeled using a cubic function given by,

$$N(h) = N(0)(ah^3 + bh^2 + ch + d), \quad (6)$$

where *a*, *b*, *c*, and *d* are the coefficients when $N(0) = 1$ (Russo et al., 2006). The default values of aerosol concentration, RH, and profile type for the free troposphere, stratosphere, and elevated mineral dust layer are adopted from Hess et al. (1998). The total column AOD is calculated by,

$$Total\ AOD = \sum_{n=1}^{6} \beta_{ext_n} \int_{h_{min_n}}^{h_{max_n}} N_n(h)dh \quad (7)$$

where h_{min} and h_{max} are the layer bottom and layer top height for each layer n.

The AeroMix package and detailed documentation are available online at www.github.com/sampr7/AeroMix (P Raj and Sinha, 2024a).

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