<div align="center">**Response notes**</div>

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

<div align="center">**Reviewer: # 1**</div>

General comments

The study fits into the scope of GMD. Minor organization of the methods section is needed as described below.

Reply: We appreciate and thank the reviewer's valuable and detailed comments and recommendation. All comments have been taken into account, as described below. We have incorporated the necessary corrections in the revised manuscript being submitted.

Introduction

1. When discussing motivation, the mixing state of aged aerosol (far from source) is important for CCN (Farmer, Cappa, and Kreidenweis 2015). McFiggans et al. 2006 is another source discussing CCN from close to source and aged populations.

    Reply: As per your suggestion the text is revised as;

    "Knowledge of the size-resolved aerosol chemical composition, size distribution, and mixing state is required to predict cloud condensation nuclei (CCN) concentrations for freshly emitted and aged aerosols (McFiggans et al., 2006; Ervens et al., 2010; Farmer et al., 2015)."

Model Overview

2. For the readers' convenience, add a table for chemical species modeled (IS, WS, BC, SS, MD). Table columns should include the name, shorthand name (e.g. IS), species considered/represented (IS includes soil dust, fly ash, and non-hygroscopic organic matter from biomass burning), and modes present (nucleation (nm), accumulation (am), coarse (cm)).

    Reply: A new table presenting the compositions of predefined components, their shorthand name, constituting species, along with their dry state (0% RH) size distribution parameters and refractive index (at 0.5 µm) is added. The discussion about constituting species in the lines from #83-91 is removed to avoid redundancy.

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)	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	0.0212	2.24	1.8	$1.53 + 5 \times 10^{-3}$i

	hygroscopic fraction of organic matter.				
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	$1.431 + 1 \times 10^{-8}i$

3. It is unclear if the optical properties for coated MD particles are calculated using Mie theory or TMM. Clarify if TMM only applies to particles comprised of 100% MD.

Reply: Optical properties of particles composed solely of MD are calculated using the TMM code. These values are adopted from Koepke et al. (2015). To avoid further any confusion, the component names is mentioned explicitly. These texts are revised as

"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)."

Optical properties of all the core-shell mixed particles are modeled using the coated-sphere Python Mie calculation program PyMieScatt; which is mentioned in the revised manuscript (lines # 116-119).

4. The exponential function for the vertical profile of aerosol concentration is mentioned in Eq. 5; however, only the cubic function is utilized in the modeling section considering it provides the better fit based on R2 and RMSE values. The authors do not necessarily need to include the exponential function since it is not used to model vertical profiles in this paper.

Reply: We prefer to retain the exponential function to model aerosol vertical profile. It is primarily due to the considerations aimed at offering flexibility in the AeroMix model to adopt under different scenarios based on the following considerations:

a. The predecessor model, OPAC, uses the exponential function to model the aerosol vertical profile. Retaining this function allows for comparisons between AeroMix-modeled AOD values with OPAC-modeled ones, facilitating consistency in analyses.

b. The cubic function is employed for modeling due to its superior fit based on R^2 and RMSE values described in this paper. However, it may not always be applicable in locations lacking aerosol vertical profile measurements. Therefore, selecting suitable generalized exponential functions for the aerosol mixture (e.g., marine, urban, desert, continental) becomes necessary.

c. Offering users the option to choose from the given three types of aerosol vertical profiles (exponential, cubic, and homogeneous) enhances flexibility, enabling selection based on the specific study location and layer characteristics.

The section is revised as following to provide more clarity;

"The vertical profile of aerosol concentration in each layer can be modeled as homogenous or as an exponential function given by,

$$N(h) = N(0)exp\left(-h/z\right), \qquad (5)$$

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,

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

where a, b, c, and d are the coefficients for $N(0) = 1$ (Russo et al., 2006)."

We hope you will agree that maintaining the exponential function alongside other options to provide users a comprehensive package, ensuring adaptability to diverse research contexts.

5. This section was jumpy and difficult to follow. I suggest further dividing into subsections:

Reply: In accordance with your suggestion, we have carefully divided it into subsections as follows:

"

3. Modeling aerosol mixing state with AeroMix

The primary objective of AeroMix is to provide a versatile open-source aerosol optical model framework tailored to support inversion algorithms for modeling both aerosol mixing states and their physical and chemical characteristics. The probable aerosol mixing states are modeled with AeroMix using the widely used Mie inversion technique by iteratively

comparing the multispectral measurements of aerosol optical properties with modeled ones for different mixing scenarios until they converge within the observational error (Fig. 1) (Chandra et al., 2004; Dey et al., 2008; Kaskaoutis et al., 2011; Ramachandran and Srivastava, 2016; Srivastava et al., 2016, Srivastava et al., 2018).

3.1 Comparison with OPAC

Initially, the AeroMix modeled aerosol properties were compared with OPAC for ten externally mixed cases given in OPAC, namely continental clean, continental average, continental polluted, urban, desert, maritime clean, maritime polluted, maritime tropical, Arctic, and Antarctic. The AeroMix computed aerosol mass concentrations (M_a) of all ten cases, as well as AOD, SSA, and asymmetry parameters, showed excellent agreement ($r = 0.99$, slope = ~ 1) with OPAC-derived ones (Figs. S1 and S2).

3.2 Model setup

3.2.1 Study region and data

AeroMix performance was further assessed by determining the probable aerosol mixing states over two contrasting environments: Kanpur, India (26.513° N, 80.232° E, 123 m AMSL; urban) and the Bay of Bengal (11.99°- 20.61°N, 80.52°-92.55° E; BoB; marine) representing diverse aerosol mixtures. Past endeavors to deduce intricate aerosol mixing states through Mie model inversion were hindered by the OPAC model's constraints on the number of plausible mixing state cases. The efficacy of this method relies on the accuracy of input parameters, encompassing AOD, SSA, aerosol vertical profile, RH, refractive indices, size distribution, and mixing state assumptions. Therefore, collocated and concurrent measurements of quality-controlled spectral AODs, spectral SSAs, spectral asymmetry parameter, vertical profile of aerosols, aerosol chemical composition, mixed layer height (MLH), and RH were used for the first time over Kanpur and the BoB; in South Asia to model the probable mixing states of aerosols using AeroMix. These measurements were taken from January 2007 to December 2009 at Kanpur and during the Winter-Integrated Campaign for Aerosols, gases and Radiation Budget (W-ICARB) conducted from December 2008 to January 2009 over the BoB (Moorthy et al., 2010). The seasonal mean MLH and β_{ext} profiles at 532 nm obtained from NASA Micro-Pulse Lidar Network (MPLNET) collected over Kanpur during the period May 2009 – Nov 2015 are utilized in this study. β_{ext} profiles at 532 nm over the western-BoB (W-BoB) and northern-BoB (N-BoB) are obtained from Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP). A summary of various aerosol and meteorological datasets used in the present study is provided in **Error! Reference source not found.**. A detailed description of the datasets used can be found in the reference therein and presented in supplementary information (SI). The locations of Kanpur, W-BoB, N-BoB, and the W-ICARB cruise track, along with seasonal average surface wind directions, are presented in Fig. 2.

3.2.2. Mixing state assumptions

The analysis encompasses eight aerosol components (IS, WS, BC, SSam, SScm, MDnm, MDam, and MDcm) in externally mixed state and their combinations forming core-shell mixed structure at various CSR values, ranging from 0.1 to 0.9 in increments of 0.1. This results in a total of 305 distinct and plausible aerosol components under consideration. This excludes combinations involving MD as the shell component and the homogenous internal mixing of these aerosol components, as they are unlikely to occur in the atmosphere (Jacobson, 2000; Dey et al., 2008). The SU component is not explicitly considered, as the WS component accounts for anthropogenic sulfate. Various CSR values

represent different mixing scenarios of core and shell components in varying mass fractions within a particle, with 0.1 representing thick coating and 0.9 representing thin coating.

Previous studies have assumed either the entire mass of the constituent aerosol components or a specific fraction of it to be involved in core-shell mixing (Chandra et al., 2004; Dey et al., 2008; Srivastava and Ramachandran, 2013). The latter assumption aligns more closely with observed mixing scenarios, wherein the entire mass of an aerosol component may not necessarily be in a core-shell mixed state with another; rather, a fraction of it may be, while the rest can exist in an externally mixed state or be internally mixed with other aerosol components (Arimoto et al., 2006; Shamjad et al., 2016; Thamban et al., 2017). However, both assumptions necessitate prior knowledge of the component-wise M_a for computing the CSR value of core-shell mixed aerosols to determine their optical properties. In this study, we propose allowing the variation of CSR values of core-shell mixed aerosols in the model. This offers a more flexible approach than relying on measured M_a to assess the probable aerosol mixing state. Subsequently, the mass fraction of the core and shell components participating in the mixing can be calculated from the CSR value of the probable core-shell mixed components.

In this study, the aerosol mixing state is described based on the distribution of the mass of different aerosol components within the aerosol mixture. If each particle in the mixture is composed of a single aerosol component, the mixture is considered 100% externally mixed. Conversely, if all the particles are composed of two aerosol components as a core-shell structure, it is denoted as 100% core-shell mixed. The total mass of the particles composed of a single aerosol component contributes to the externally mixed aerosol mass fraction, while the total mass of particles with a core-shell structure contributes to the core-shell mixed mass fraction. The mass fraction of an aerosol component in a core-shell mixed component is determined by calculating the ratio of its mass in that core-shell mixed component to the total mass of that component in the aerosol mixture. The masses of core and shell components in a core-shell mixed component are computed using Eq. (1). The combined mass of an aerosol component in various core-shell mixed particles and in an externally mixed state contributes to the total mass of that aerosol component in the mixture. The combined mass concentrations of SSam and SScm together are referred to as SS mass concentration (M_{SS}), while MDnm, MDam, and MDcm collectively constitute the MD mass concentration (M_{MD}). Here onwards, IS, WS, and BC mass concentrations are denoted as M_{IS}, M_{WS}, and M_{BC}, respectively. The scalability of AeroMix in terms of the number of components that can be defined in one mixture enabled this study to assess the possible existence of different types of core-shell mixed particles in a mixture. Since AeroMix models optical properties at specified RH levels, the nearest RH value to the seasonal average of the daytime mixed layer RH for Kanpur and the regional average for BoB is chosen, respectively.

3.2.3. Aerosol vertical profile

The vertical distribution of aerosols in the mixed layer is modeled by fitting a cubic polynomial (Eq. (6)) to the measured β_{ext} at 532 nm profiles. The decision to opt for an alternative was made because the exponential function (Eq. (5)) may not consistently capture the actual vertical variation of aerosols, especially in scenarios with co-existing elevated aerosol layers. This results in an inaccurate representation of the measured β_{ext} profiles. In the cases examined in this study, the exponential function yielded a suboptimal fit, as indicated by R^2 values ranging from -0.24 to 0.7 and RMSE varying between 0.03 and 0.14. Conversely, the cubic polynomial provided the best fit, with R^2 and RMSE values ranging from 0.91 to 0.99 and 0.004 to 0.036, respectively. The aerosol chemical composition and vertical profiles in the free troposphere and stratosphere are modeled following Hess et al. (1998). The size distributions and optical properties of all aerosol components are assumed to be uniform throughout the atmospheric column. Assessing the impact of assumptions

about the aerosol vertical profile is challenging due to the limitation in determining the extent of changes in aerosol properties with altitude. Even if such variations are known, like other inversion algorithms (Lewandowski et al., 2010; Sinha et al., 2013), there is a limitation associated with the Mie inversion technique on estimating the effect of the changes on the inversion results, although it has been widely used as described above. Alternatively, sensitivity estimates on AOD of the parameters describing vertical profiles are also complicated owing to the interdependence of multiple factors influencing AOD, such as vertical profile and layer thickness of aerosols. Suppose the covariation of the aerosol vertical profile with changes in mixed layer thickness is disregarded by assuming a constant aerosol vertical profile while varying the MLH. In that case, the changes in AOD ($\Delta AOD/\Delta MLH \approx \partial AOD/\partial h$) are minimal (<0.0005) for a unit meter shift in MLH across the different vertical profiles considered (Fig. S3). A detailed description of this sensitivity analysis is given in SI. However, the $\Delta AOD/\Delta MLH$ is not uniform throughout the vertical column. Instead, it follows the given vertical distribution of aerosols within the layer. This suggests that alterations in MLH have a more pronounced effect on AOD at altitudes characterized by higher aerosol concentrations than those with lower aerosol concentrations. Given the highly heterogeneous spatial and temporal nature of aerosol vertical distribution in the real atmosphere (Kumar et al., 2023), attempting a generalized quantification of the sensitivity of vertical profile assumptions on AOD lacks meaningful interpretation.

3.3. Assessment of probable aerosol mixing state

The probable existence of the components in the atmosphere is assessed by iteratively changing the number concentrations in the mixture in AeroMix until the root mean squared error (RMSE) between the measured and modeled AOD and SSA spectra are minimized. Only those spectra with RMSE minimum ≤ 0.03 are considered as the best fit (Fig. 1). The RMSE threshold of 0.03 is chosen to ensure that the RMSE remains within the lowest uncertainty (15%) in the Aerosol Robotic Network (AERONET) AOD and SSA retrievals (Srivastava and Ramachandran, 2013). Since AeroMix models optical properties at specific wavelengths, modeled AOD values are interpolated to the measurement wavelengths for calculating the RMSE using a second-order polynomial equation (Eq. (8)) (Eck et al., 1999), which provides greater precision compared to the Angstrom power law relationship.

$$ln\,AOD(\lambda) = a_2(ln\,\lambda)^2 + a_1\,ln\,\lambda + a_0 \qquad\qquad (1),$$

where λ is the wavelength at which AOD is calculated, and a_0, a_1, and a_2 are coefficients. However, for SSA, wavelengths closer to the measurement wavelengths are chosen in AeroMix since no equivalent relationship exists for SSA interpolation.

The aerosol mixture with the spectral AODs and SSAs matching well with the measured ones (RMSE≤ 0.03) is taken as a probable aerosol mixing state for that location during the particular season. It is important to note that the aerosol mixing state modeled by the Mie inversion technique is not unique but a probable scenario. Further, AeroMix-modeled probable aerosol mixing states are constrained using the measured component-wise M_a with the modeled M_a (within ±1σ) to minimize uncertainty in the modeled probable mixing scenarios. Measured M_a of organic carbon (OC), elemental carbon (EC), water-soluble organic carbon (WSOC), and water-soluble ionic species (WSIS) are grouped into the M_{IS}, M_{WS}, M_{BC}, $M_{SS,}$ and M_{MD} using appropriate fixed scaling factors for each component prescribed in the literature (see SI for detailed description). The asymmetry parameter is not considered to constrain the mixing states; however, a brief comparison between modeled and measured asymmetry parameter values is provided in the next section.

"

6. Little mention of future work/applications. Maybe mention potential model development projects and associated improvement to probable mixing state predictions.

Reply: The lines #404-408 is revised as;

"The prospective advancement linked to AeroMix involves the development of an optimization/machine learning algorithm utilizing AeroMix as the aerosol optical model. Such an algorithm will enable faster and more deterministic estimation of aerosol mixing states at fine temporal and spatial resolutions."

Minor comments

Line 43: Healy et al. 2014 and Ye et al. 2018 describe in situ measurements of mixing state between external and internal.

Reply: The single-particle mass spectrometry observations presented in these studies will help present the case in our manuscript. Suggested references are incorporated to strengthen the discussion on real atmospheric aerosol mixing states. The sentence is revised as;

"Despite the experimental evidences that the aerosol mixing state lies between a purely externally mixed state and a purely internally mixed state (Healy et al., 2014; Li et al., 2016; Ye et al., 2018; Riemer et al., 2019), the standard FORTRAN-based OPAC model considers the external mixing of aerosols alone and cannot treat complex aerosol internal mixing states."

Line 58: Unclear to reader why "batch mode" would have limited functionalities. Either clarify or reword.

Reply: We removed this discussion about AeroGUI to avoid ambiguity.

Line 97-98: Unclear why density of BC is mentioned here.

Reply: The specific densities for all components are sourced from the Global Aerosol Data Set and OPAC database. However, the observational studies on black carbon suggest using a specific density value of 1.8 g cm^{-3}, differing from the 1.0 g cm^{-3} value provided in OPAC. This clarification will be added to the manuscript as follows**:**

"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)."

Line 137 – 139: This sentence is confusing, specifically "and particles composed of multiple chemical species (internally mixed) as an external mixture."

Reply: The sentence is revised as follows for brevity.

"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)."

Equation (7): I believe index of summation should be layer, not n.

Reply: The equation is corrected as,

"

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

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

Line 163: Consider adding one sentence on the Mie inversion technique.

Reply: The sentence is revised as

"The probable aerosol mixing states are modeled with AeroMix using the widely used Mie inversion technique by iteratively comparing the multispectral measurements of aerosol optical properties with modeled ones for different mixing scenarios until they converge within the observational error (Fig. 1) (Chandra et al., 2004; Dey et al., 2008; Kaskaoutis et al., 2011; Ramachandran and Srivastava, 2016; Srivastava et al., 2016, Srivastava et al., 2018)"

Figure 2: Consider adding average wind directions for winter, pre-monsoon and post-monsoon. This would aid in the comparison of modeled mixing states discussion (Sections 4.3 and 4.4). Label IGP region.

Reply: Average wind directions for different seasons are added to Figure 2, along with labeling the IGP region as shown below.
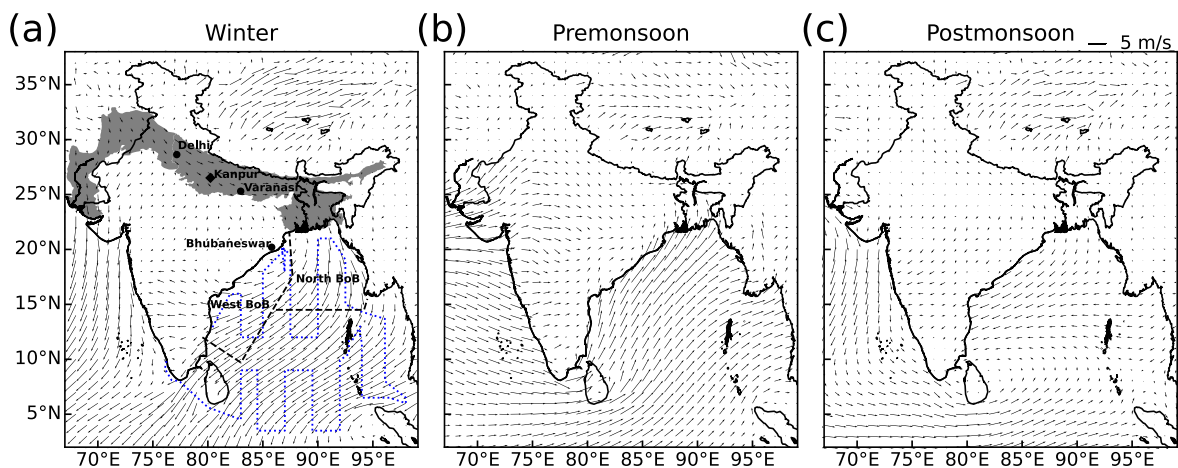
Figure 2. a) Location of Kanpur (diamond) and the W-ICARB cruise track (dotted lines). Measurements of AOD, chemical composition, and relative humidity over the West-BoB and North-BoB (demarcated with dashed lines) were utilized in this study. Additionally, the locations of Delhi, Varanasi, and Bhubaneswar (circles) are indicated, where mixing state measurements were obtained for comparison with the modeled mixing states. The shaded area marks the Indo-Gangetic Plain. Seasonal average surface winds over the region from ERA5 reanalysis are shown for a) winter, b) premonsoon and c) postmonsoon by vectors.

Line 243: Add reasoning for RMSE threshold after sentence: "Only those spectra with RMSE minimum 0.03 are considered as the best fit (Fig. 1). [Add here]." The last sentence of this paragraph "The RMSE threshold of 0.03 is chosen to ensure …" should be moved to this location.

Reply: Revised as suggested.

Figure 3 and 4: Unnecessary to include colors for externally mixed and core-shell mixed in caption.

Reply: Revised as suggested.

Technical corrections

Line 11: Add comma after "aerosol chemical compositions,"

Reply: Corrected as suggested.

Line 13: Remove "the" so the sentence reads "While limitations in the measurements of aerosol …"

Reply: The sentence is modified as per the following comment.

Line 13-15: Sentence "While the limitations in the measurements …" is a run on. Consider breaking into two sentences.

Reply: Following your suggestion, we corrected as the sentence as:

"*Limitations in the direct measurements of aerosol chemical composition and mixing states necessitate modeling approaches to infer the aerosol mixing states. The Optical Properties of Aerosols and Clouds (OPAC) model has been widely used to construct optically equivalent aerosol chemical compositions from measured aerosol optical properties using Mie inversion.*"

Line 18: Move position of "3)" – "components, and 3) define aerosol composition"

Reply: Corrected as suggested.

Line 98: Add comma "The optical properties of each component, except for the MD components, are calculated"

Reply: Corrected as suggested.

Line 209: Run-on sentence, rewrite as "…CSR values of core-shell mixed aerosols in the model. This offers a more flexible approach compared to relying on measured…"

Reply: Corrected as suggested.

Line 297: Change to "… mixed states varied from 30% to 59% in premonsoon and winter, **respectively."**

Reply: Changing to "mixed states varied from 30% to 59% in premonsoon and winter, respectively" may lead to confusion regarding whether these values represent intra-seasonal or inter-seasonal variation. To clarify, we propose revising it to:

"*The percentage mass fractions of the total M_a in core-shell mixed states increased from 30% in premonsoon to 59% in winter (Fig. 3d).*"