AeroMix v1.0.1: a Python package for modeling aerosol optical properties and mixing states

Sam P. Raj [1], Puna Ram Sinha [1], Rohit Srivastava [2], Srinivas Bikkina [3], D. Bala Subrahamanyam [4]

[1]Department of Earth and Space Sciences, Indian Institute of Space Science and Technology Thiruvananthapuram, Kerala,
5   695547, India

[2] National Centre for Polar and Ocean Research, Ministry of Earth Sciences, Govt. of India, Vasco-da-Gama, Goa, 403804,
India

[3] Birbal Sahni Institute of Palaeosciences, 53, University Road, Lucknow, 226007, India

[4] Space Physics Laboratory, Vikram Sarabhai Space Centre, Thiruvananthapuram, Kerala, 695022, India

10   *Correspondence to*: P. R. Sinha, (prs@iist.ac.in)

Abstract. Assessing aerosol mixing states, which mainly depend on aerosol chemical compositions is indispensable to estimate aerosol direct and indirect effects. While the limitations in the measurements of aerosol chemical composition and mixing states persist globally, 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. However, the

15   representation of real atmospheric aerosol mixing scenarios in OPAC has perennially been challenged by the exclusive assumption of external mixing. A Python successor to the aerosol module of the OPAC model is developed, named 'AeroMix,' with novel capabilities to 1) model externally and core-shell mixed aerosols, 2) simulate optical properties of aerosol mixtures constituted by any number of aerosol components, 3) and define aerosol composition and relative humidity in up to 6 vertical layers. Designed as a versatile open-source aerosol optical model framework, AeroMix is tailored for sophisticated inversion

20   algorithms aimed at modeling aerosol mixing states and also their physical and chemical properties. AeroMix's performance is demonstrated by modeling the probable aerosol mixing states over Kanpur (urban), India, and the Bay of Bengal (marine). The modeled mixing states are consistent with independent measurements using single-particle soot photometer (SP2) and transmission electron microscopy (TEM), substantiating the potential capability of AeroMix to model complex aerosol mixing scenarios involving multiple internally mixed components in diverse environments. This work contributes a valuable tool for

25   modeling aerosol mixing states to assess their impact on cloud nucleating properties and radiation budget.

1. Introduction

Various sources of aerosol particles and their multi-scale dynamic nature in the atmosphere form a complex mixture of externally and internally mixed aerosol components, which are highly variable spatially and temporally (Ching et al., 2019;

30   Riemer et al., 2019). Knowledge of the size-resolved aerosol chemical composition, size distribution, and mixing state is required to predict cloud condensation nuclei (CCN) close to sources (Ervens et al., 2010). Thus, the impact of aerosol mixing

state and chemical composition on the activation of CCN needs to be determined and understood to the extent that this is importantly represented in global climate models (Ghan and Schwartz, 2007). However, in situ measurements of aerosol chemical compositions and mixing states are sparse due to the complexities associated with the measurement techniques

35   (Riemer et al., 2019). The standard aerosol model outlined in Optical Properties of Aerosols and Clouds (OPAC) (Hess et al., 1998; Koepke et al., 2015) has been used extensively to estimate the probable aerosol mixing state from the measured aerosol optical properties through Mie inversion. However, it is not the best approach to assess the aerosol composition and mixing state for a given location, but this is the viable and practical option when no direct measurements of aerosol chemical composition are available.

40   The OPAC model has significantly contributed to aerosol research by providing reliable simulations of the optical properties of different aerosol mixtures necessary to estimate their radiative effects. It can model user-defined aerosol mixtures by mixing up to seven aerosol components. Despite the experimental evidences that the aerosol mixing state lies between a purely externally mixed state and a purely internally mixed state (Li et al., 2016; 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. Most

45   current climate models also assume entirely externally or internally mixed aerosols, resulting in an error in modeled optical, hygroscopic, CCN, and cloud properties (Stevens and Dastoor, 2019). Additionally, restricting the number of components constituting an aerosol mixture further limits the modeling of complex aerosol mixtures using OPAC.

Several attempts have been made to incorporate internal mixing in OPAC by modifying the predefined components with the optical properties of internally mixed aerosols (Chandra et al., 2004; Dey et al., 2008; Ramachandran and Srivastava, 2016;

50   Srivastava et al., 2016, Srivastava et al., 2018). In these studies, the mixing state of aerosols using the OPAC model was modeled by iteratively comparing modeled optical parameters such as aerosol optical depth (AOD), single scattering albedo (SSA), and asymmetry parameter (g) with measured ones for different mixing scenarios until they converge within the observational error. While transmission electron microscopy (TEM) observations have shown the presence of multiple combinations of internally mixed aerosol components in a mixture (Li et al., 2016), the limitation in the number of aerosol

55   components permissible to constitute an aerosol mixture in OPAC constrained the number of internally mixed components considered for each case in the above-referred studies, thus limiting the number of possible combinations of aerosol components to determine the probable mixing states. Although the MATLAB tool AEROgui provides the option to define internally mixed aerosols through a user interface, its batch mode has limited functionalities (Pedrós et al., 2014).

In an effort to address this challenge, a Python-based package named 'AeroMix' for modeling aerosol optical properties and

60   mixing states is developed. AeroMix enables the modeling of optical properties of complex aerosol mixtures consisting of any number of components in externally mixed and/or core-shell mixed states. Furthermore, other internal mixing states can also be represented using appropriate mixing rules (Bohren and Huffman, 1998; Stevens and Dastoor, 2019). The scalability of the number of components in a mixture, the ability to model core-shell mixed aerosols, and the scope of integration with other programs make AeroMix a versatile open-source tool for inversion algorithms aimed at modeling the properties of aerosol

65   mixtures, including aerosol chemical composition, and mixing states. For this purpose, AeroMix performance is demonstrated

by determining the chemical composition and probable mixing states of aerosols from the measured aerosol optical properties using the Mie inversion technique over Kanpur, a representative urban location in India, in the Indo-Gangetic Plain (IGP) and a marine environment of the Bay of Bengal (BoB). Furthermore, the probable mixing states modeled with AeroMix are compared with independent measurements made by single-particle soot photometer (SP2) and TEM.

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.
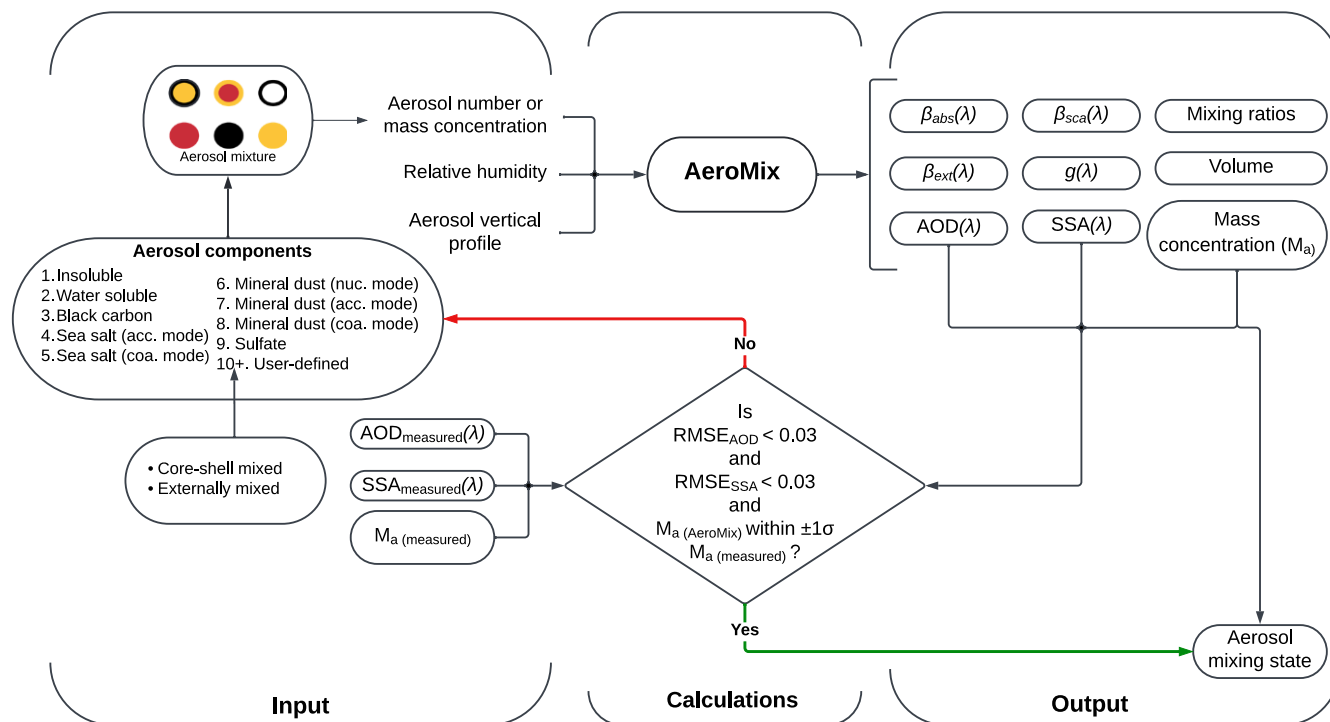
Figure 1: Overview of the AeroMix workflow for modeling aerosol mixing states using the Mie inversion technique.

80   Aerosol mixtures in AeroMix can be defined in terms of the number or mass concentration of the constituent aerosol components. Nine predefined aerosol components 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). The IS aerosols represent soil dust, fly ash, and non-hygroscopic organic matter from biomass burning, biogenic and anthropogenic sources. WS aerosols consist of hygroscopic particles

85   formed through gas-to-particle conversion, including SO_4^{2-}, NO_3^-, NH_4^+, the hygroscopic fraction of organic matter, and other hygroscopic aerosols except SS and SU. BC, produced by fossil fuel combustion or biomass burning, is non-hygroscopic in its pure form and is distinguished by its strong absorption of solar radiation. Though the aged BC aerosols form fractal clusters, individual monomers in the chain are treated independently for Mie calculations. The bursting of air bubbles caused by wind-wave interaction leads to the formation of hygroscopic SS aerosols. MD aerosols, originating from arid regions by wind erosion

90   of desert soils, consist of non-hygroscopic quartz and clay minerals. The SU describes stratospheric sulfate aerosols, while anthropogenic sulfates are accounted for in the WS component (Koepke et al., 1997; Hess et al., 1998). 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 (β_{ext} (λ), β_{sca} (λ), β_{abs} (λ), SSA (λ) and g (λ)) at sixty-one wavelengths from 0.25 to 40 µm and eight

95   RH values, adopted from the Global Aerosol Data Set and OPAC database (Koepke et al., 1997; Hess et al., 1998; Koepke et al., 2015). 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 taken as 1.8 g cm^{-3} (Bond et al., 2013; Kondo, 2015). The optical properties of each component except for the MD components, are calculated using the Mie theory (Mie, 1908). This calculation assumes that the particles are spherical in shape and follow a

100   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. MD 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).

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

105   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

110   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.

In addition to the nine predefined aerosol components, AeroMix offers the flexibility to model any number of user-defined components. An externally mixed aerosol component can be defined by its size distribution parameters, specific density, and

115   spectral refractive indices described above. A core-shell mixed aerosol component can be defined by specifying the core and shell components and their core-to-shell radius ratio (CSR) or mass fractions in the core-shell mixed state. 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.

120   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,

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

125   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,

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

130   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,

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

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

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

Equation (4) contains only the terms of the specific density (ρ) of core and shell components and CSR, which remain same across the size distribution. Complex aerosol mixing states are represented in AeroMix by treating the mixture of particles composed of single chemical species (externally mixed) and particles composed of multiple chemical species (internally mixed) as an external mixture (see Fig. 1). Modeling the optical properties of diverse aerosol mixtures requires inputting the

140   number or mass concentrations of the constituent aerosol components in external and/or core-shell mixed states, along with their vertical distribution and the vertical profile of RH. For total column AOD calculation, up to 6 vertically arranged layers can be defined to specify the vertical distribution of aerosols. Unlike OPAC, in AeroMix, aerosol concentrations at the layer base 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,

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

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). It can also be modeled as a cubic function given by,

$$N(h) = ah^3 + bh^2 + ch + d, \tag{6}$$

where a, b, c, and d are the coefficients (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,

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

$$\tag{7}$$

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).

3. Modeling of 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 (Chandra et al., 2004; Dey et al., 2008; Kaskaoutis et al., 2011; Ramachandran and Srivastava, 2016; Srivastava et al., 2016, Srivastava et al., 2018).

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).

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. A summary of various aerosol and meteorological datasets used in the present study is provided in Table 1. A detailed description of the datasets used can be found in the reference therein and presented in supplementary information (SI). In brief, we utilized collocated spectral AODs, spectral SSAs and asymmetry

175   parameters, β_{ext} profiles, aerosol chemical composition, mixed layer height (MLH), and RH measured from January 2007 to December 2009 at Kanpur and collocated spectral AODs, β_{ext} profiles, aerosol chemical composition, MLH, and RH measured over BoB during the Winter-Integrated Campaign for Aerosols, gases and Radiation Budget (W-ICARB) conducted from December 2008 to January 2009. The MLH and β_{ext} profiles at 532 nm are obtained from NASA Micro-Pulse Lidar Network (MPLNET) collected over Kanpur. B_{ext} profiles at 532 nm over the western-BoB (W-BoB) and northern-BoB (N-BoB) are

180   obtained from Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP). The locations of Kanpur, W-BoB, N-BoB, and the W-ICARB cruise track are presented in Fig. 2.

Table 1: Aerosol and meteorological data used in this study.

Kanpur

Parameter	Period	Resolution			Reference
		Temporal	**Spatial**	**Vertical**	
Aerosol optical depth (AOD) at 0.34, 0.38, 0.44, 0.5, 0.675, and 0.87 μm	Jan 2007 – Dec 2009	Monthly	-	-	AERONET Holben et al., 2001
Single scattering albedo (SSA) at 0.44, 0.675, 0.87, and 1.02 μm					
Asymmetry parameter (g) at 0.44, 0.675, 0.87, and 1.02 μm					
Aerosol extinction coefficient (β_{ext}) at 532 nm	May 2009 – Nov 2015	1 minute	-	75 m	MPLNET Welton et al., 2001
Aerosol chemical composition	Jan 2007 – Mar 2008	1-2 samples/ week	-	-	Ram et al. 2010
Mixed layer height (MLH)	May 2009 – Nov 2015	1 minute	-	-	MPLNET Lewis et al., 2013
Relative humidity (RH)	Nov 2007 – Dec 2009	Hourly	-	-	MOSDAC

Bay of Bengal

Parameters	Period	Resolution			Reference
		Temporal	**Spatial**	**Vertical**	
Aerosol optical depth (AOD) at 0.38, 0.44, 0.5, 0.675, and 0.87 μm	27 Dec 2008 – 9 Jan 2009	10 minutes	-	-	Kaskaoutis et al. 2011
Aerosol extinction coefficient (β_{ext}) at 532 nm	Dec 2008 – Jan 2009	Monthly	2°×5° (lat×lon)	60 m	CALIOP Tackett et al., 2018
Aerosol chemical composition	27 Dec 2008 – 9 Jan 2009	Daily	-	-	Srinivas et al. 2011

| Mixed layer height (MLH) | 27 Dec 2008 – 9 Jan 2009 | 2 launches/ day | - | - | Subrahamanyam et al. 2012 |
| Relative humidity (RH) | 27 Dec 2008 – 9 Jan 2009 | Daily | - | - | Sinha et al., 2011b |

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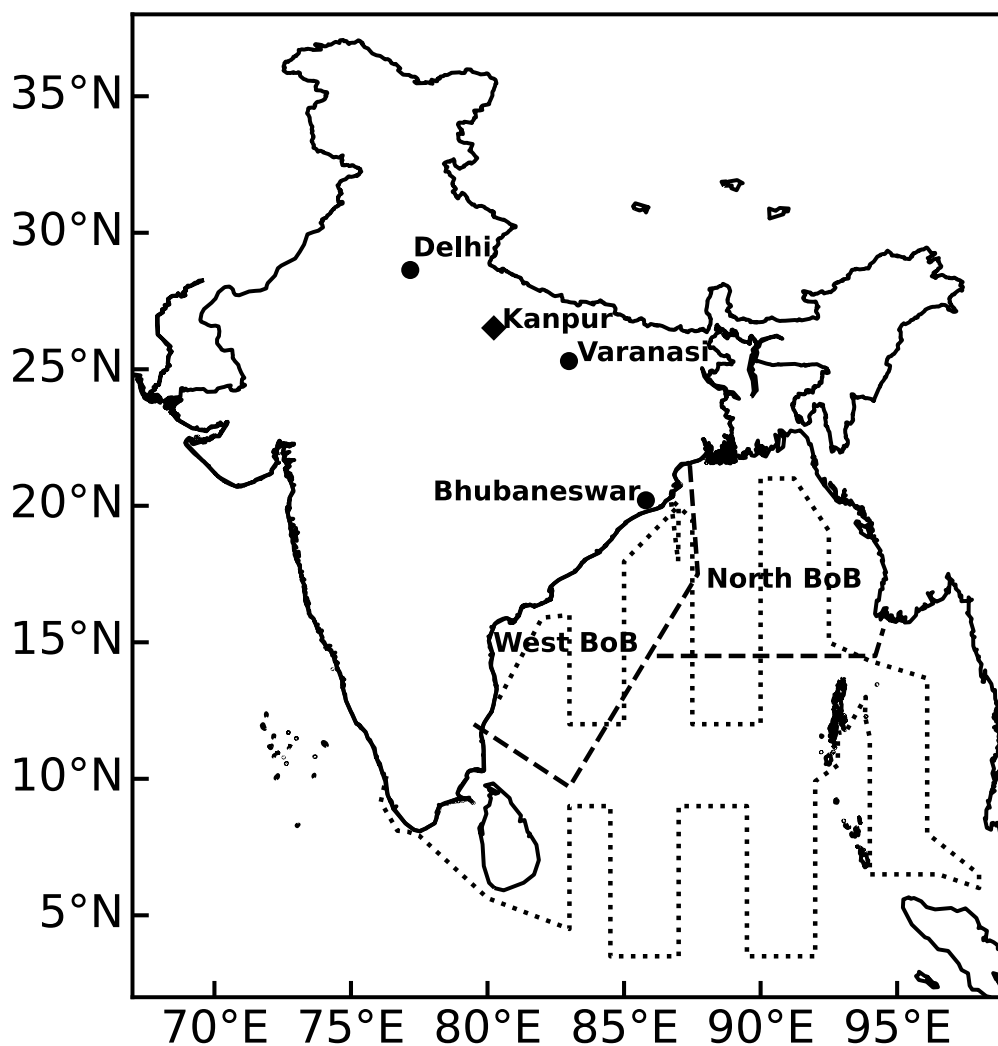
Figure 2: Location of Kanpur (diamond) and the W-ICARB cruise track (dotted lines). Measurements of AOD, chemical composition, and relative humidity over the West-BoB and North-BoB (demarcated with dashed lines) were utilized in this study. Additionally, the locations of Delhi, Varanasi, and Bhubaneswar (circles) are indicated, where mixing state measurements were obtained for comparison with the modeled mixing states.

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
190   parameters, encompassing AOD, SSA, vertical aerosol 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, 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.

The analysis encompasses eight aerosol components (IS, WS, BC, SSam, SScm, MDnm, MDam, and MDcm) in externally
195   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
200   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
205   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, which offers a more flexible approach than relying on measured M_a to assess the
210   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.

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
215   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.

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
220   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

225   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

230   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

235   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

240   AOD lacks meaningful interpretation.

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). Since AeroMix models optical properties at specific wavelengths, modeled AOD values are interpolated to the measurement wavelengths for

245   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.

$$ln\,AOD(\lambda) = a_2(ln\,\lambda)^2 + a_1\,ln\,\lambda + a_0 \qquad (8),$$

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.

250   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).

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

255 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

260 measured asymmetry parameter values is provided in the next section.

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

265 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

270 component in the mixture.

4. AeroMix modeled aerosol mixing state over Kanpur and the Bay of Bengal

4.1. Kanpur

Aerosol mixing state over Kanpur is deduced for winter (December-February), premonsoon (March-June), and postmonsoon (October and November) seasons by constraining AeroMix modeled spectral AODs, SSAs, and M_a with the measured ones

275 obtained at Kanpur during 2007-2009. The monsoon (July-September) season is excluded from the study due to the lack of aerosol chemical composition measurements. The selection of this study period is based on the availability of collocated aerosol chemical composition data over Kanpur and the concurrent W-ICARB campaign data over the BoB during winter, of which a detailed description is provided in the following section. Seasonal averages of MLH and β_{ext} at 532 nm derived with MPLNET for the entire operational period from May 2009 to November 2015 are utilized to model the aerosol vertical

280 distribution due to a lack of concurrent MPLNET observations for representing seasons under consideration.

Figures 3a-3c present the AeroMix modeled spectral AODs, SSAs, component-wise M_a in external and core-shell mixed states compared with the measured ones for winter, premonsoon and postmonsoon seasons of 2007-2009 over Kanpur. The modeled percentage mass fraction of aerosols to the total M_a is depicted in Fig. 3d. A summary of the probable core-shell mixed aerosol components in each season, their CSR value, and the mass fraction of components in each core-shell mixed component is

285 presented in Table 2. The AeroMix modeled spectral AODs and SSAs of the probable aerosol mixture agreed with the measured values for all the seasons within RMSE values of <0.03 (Figs. 3a-3b). The corresponding modeled and measured M_a

of all components agreed to within ±1σ (Fig. 3c). AeroMix estimated M_{MD} could not be compared due to the unavailability of mineral dust measurements during the study period.

290 **Table 2: Probable core-shell mixed aerosol components modeled with AeroMix over Kanpur for winter, premonsoon, and postmonsoon seasons of 2007-2009 and over BoB during winter (Dec 2008-Jan 2009). The percentage mass fraction of aerosol components participating in core-shell mixing is also presented.**

Kanpur						
Season	Core-shell mixed components	CSR	Core		Shell	
			Component	%	Component	%
Winter	IS-WS	0.8	IS	86.5	WS	49.8
	BC-WS	0.4	BC	51.8	WS	42.0
	SSam-BC	0.9	SSam	50.2	BC	9.70
Premonsoon	IS-WS	0.9	IS	100	WS	62.6
	MDnm-BC	0.3	MDnm	0.04	BC	49.6
Postmonsoon	IS-WS	0.8	IS	97.1	WS	58.6
	BC-WS	0.4	BC	8.0	WS	7.80
Bay of Bengal						
Region	Core-shell mixed components	CSR	Core		Shell	
			Component	%	Component	%
W-BoB	IS-WS	0.8	IS	100	WS	34.7
	BC-WS	0.4	BC	23.6	WS	21.9
	MDnm-BC	0.4	MDnm	0.70	BC	76.4
N-BoB	IS-WS	0.9	IS	100	WS	13.4
	BC-WS	0.4	BC	100	WS	86.6
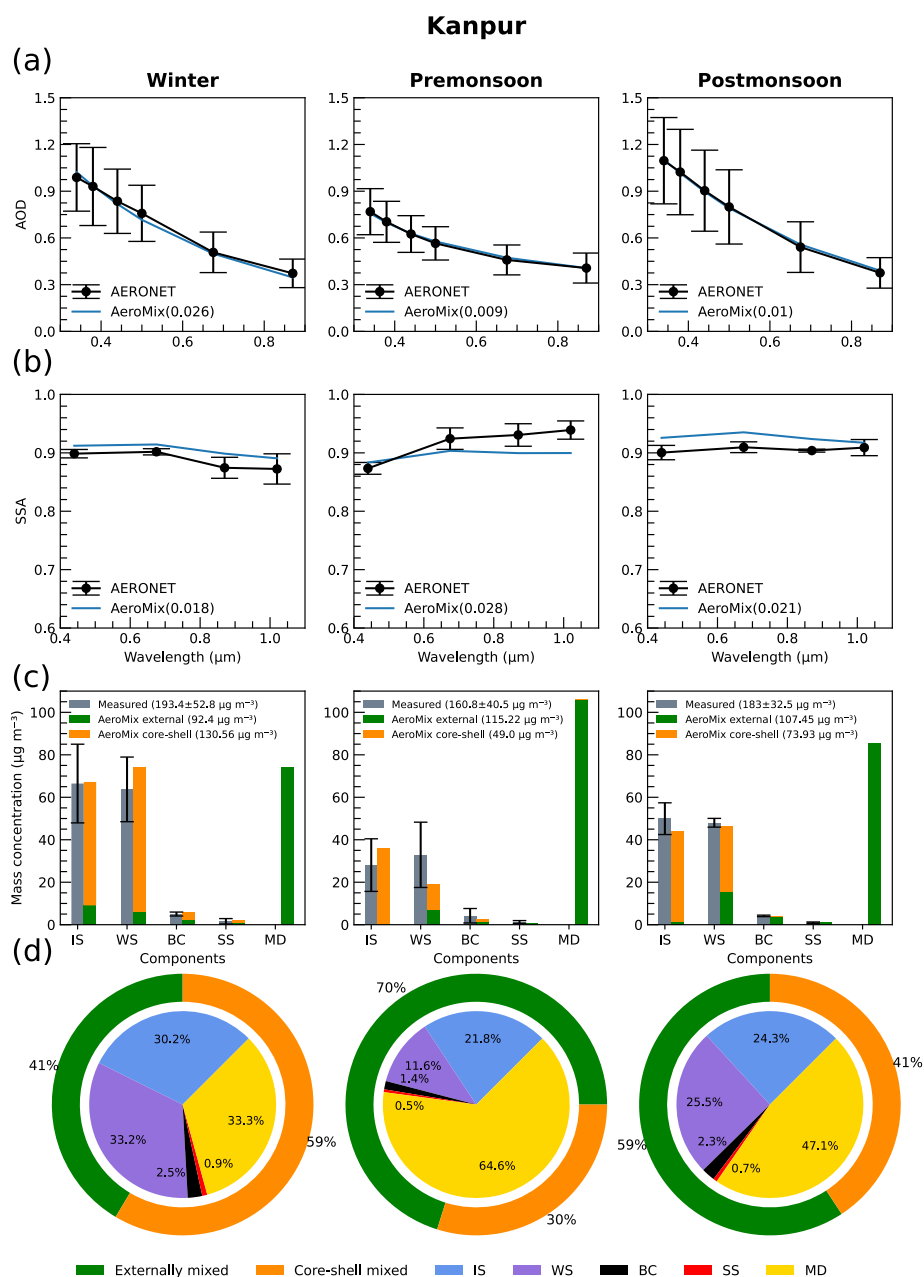
Figure 3: Measured and AeroMix modeled aerosol parameters over Kanpur during the winter, premonsoon, and postmonsoon seasons of 2007-2009. a) AeroMix modeled season-wise spectral AOD compared with AERONET AOD. Vertical bars represent the standard deviation of the mean values. Root mean square error of the fit is given in parenthesis, b) Same as (a) but for SSA, c) Component-wise aerosol mass concentration in externally mixed (green) and core-shell mixed (orange) state compared with measured aerosol mass concentration (grey). Vertical bars represent standard deviation of measurements, d) Modeled percentage mass fraction of aerosol components to the total aerosol mass (inner pie) and percentage mass fraction of externally mixed (green) and core-shell mixed (orange) aerosols to the total aerosol mass (outer pie).

295   Seasonal variability in the extent of core-shell mixing, the types of core-shell mixed particles, and the aerosol mass fraction participating in core-shell mixing are observed (Fig. 3; Table 2). The percentage mass fractions of the total M_a in core-shell mixed states varied from 30% to 59%, highest in winter and lowest in premonsoon (Fig. 3d). Insoluble aerosols thinly coated with water-soluble are probable in all three seasons. Water-soluble aerosols are also found to be thickly coated over BC aerosols during the winter and postmonsoon seasons. BC aerosols are thinly coated over half of the SSam mass in winter while

300   thickly coated over a tiny fraction of the MDnm aerosols in premonsoon season. Only SSam of sea-salt components and MDnm of mineral dust components are in the core-shell mixed state, and the majority (>99%) of mineral dust is in an externally mixed state in all seasons.

In Kanpur, the AeroMix modeled and measured spectral asymmetry parameter exhibited excellent closure for winter (RMSE = 0.024) and postmonsoon (RMSE = 0.02) but deviated slightly (RMSE = 0.065) for premonsoon (however, not shown), which

305   could be associated with the presence of BC-coated mineral dust aerosols (Table 2) of which the non-sphericity is not accounted in AeroMix.

The earlier modeling study assessing aerosol mixing states over Kanpur, also reported the probable existence of water-soluble coated BC aerosols during winter and postmonsoon, BC-coated mineral dust during premonsoon and water-soluble coated insoluble aerosols during postmonsoon seasons (Srivastava and Ramachandran, 2013). Expanding upon these findings, the

310   present study, leveraging AeroMix capability to model any number of aerosol components in a mixture, reveals the likely coexistence of multiple core-shell mixed aerosol components in the aerosol mixture over Kanpur. The mass fractions of core and shell components involved in core-shell mixing are anticipated to differ among the studies. The present study assessed the probable coexistence of aerosol components in externally mixed states and different core-shell mixed states at various mass-mixing scenarios, considering all possibilities simultaneously. In contrast, Srivastava and Ramachandran (2013) examined the

315   probable existence of each core-shell mixed component as a separate case, with core and shell components mixed at mass fractions varying from 20-100% of their total mass, while the rest remained externally mixed with other aerosol components. The findings in this study are further strengthened by constraining the probable mixing states with the concurrent and collocated measured aerosol chemical composition data (Ram et al., 2010), contributing to a more robust assessment of probable aerosol mixing states.

320   **4.2. Bay of Bengal**

Aerosol mixing states over the W-BoB and N-BoB are analyzed for the winter season from the spectral AODs, aerosol chemical composition, MLH, and RH data collected from the W-ICARB campaign during the period 27th December 2008 to 9th January 2009. MLH derived from radiosonde and β_{ext} at 532 nm from CALIOP are used to model the aerosol vertical distribution. In the absence of SSA measurement, only the measured spectral AODs and M_a are used as constraints to simulate

325   aerosol compositions and model the probable mixing state over the regions, which may influence the mixing state estimates. The modeled spectral AODs of the probable aerosol mixtures are in close agreement with the measured values (RMSE<0.03) (Fig. 4a), and modeled M_a are within ±1σ of measured values for both regions (Fig. 4b).
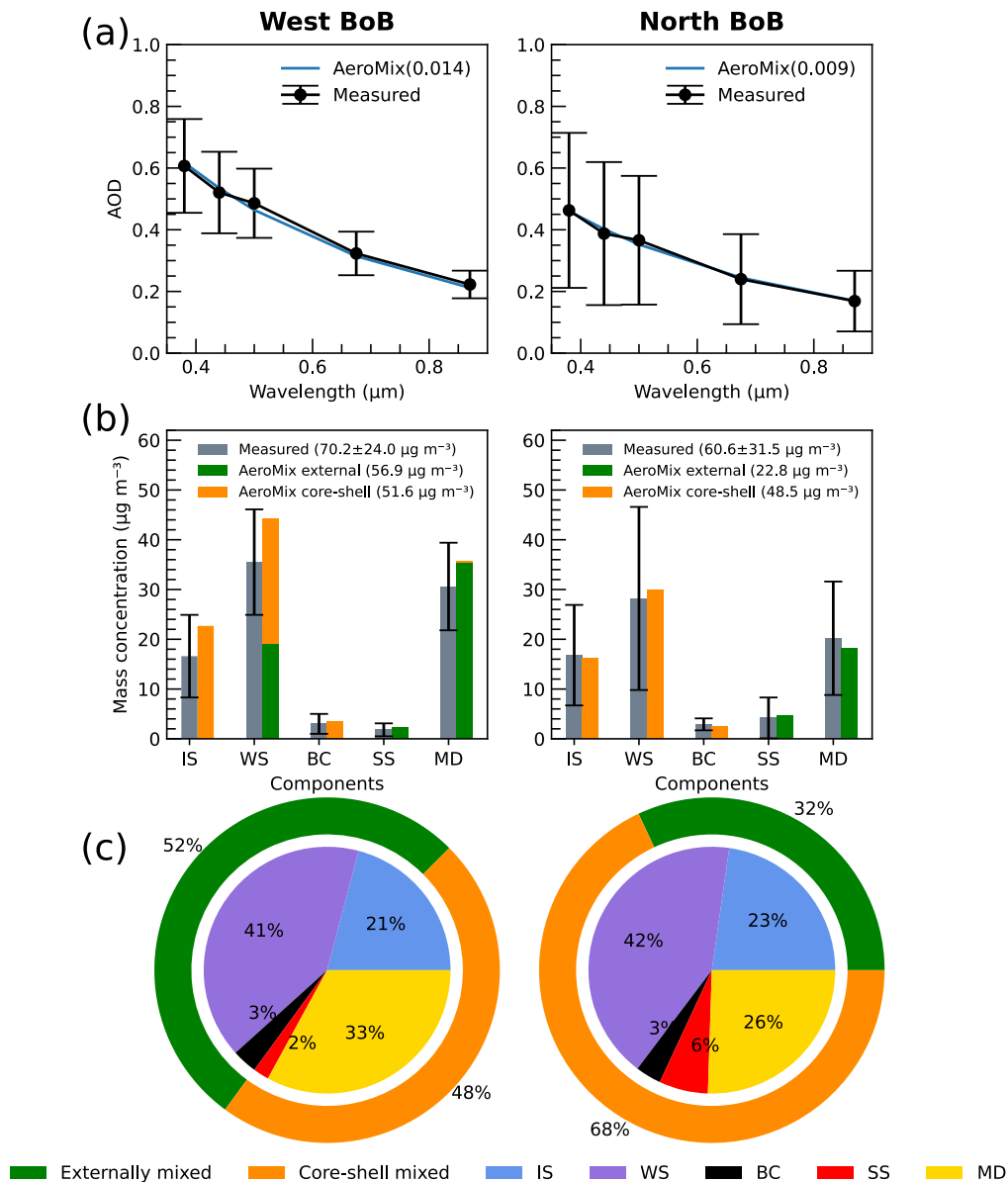
Figure 4: Measured and AeroMix modeled aerosol parameters over W-BoB and N-BoB during winter season (Dec 2008-Jan 2009). a) AeroMix modeled season-wise spectral AOD compared with measured AOD. Vertical bars represent the standard deviation of the mean values. Root mean square error of the fit is given in parenthesis, b) Component-wise aerosol mass concentration in externally mixed (green) and core-shell mixed (orange) state compared with measured aerosol mass concentration (grey. Vertical bars represent standard deviation of measurements, c) Modeled percentage mass fraction of aerosol components to the total aerosol mass (inner pie) and percentage mass fraction of externally mixed (green) and core-shell mixed (orange) aerosols to the total aerosol mass (outer pie).

The extent of core-shell mixing is greater over the N-BoB (68%) compared to the W-BoB (48%) during the winter season.
330 The mixtures consisting of BC-coated mineral dust; and water-soluble coated insoluble and BC particles are the probable core-shell mixing scenarios over W-BoB. Meanwhile, over the N-BoB, only water-soluble coated insoluble and BC aerosols are probable in the core-shell mixed state (Table 2). Interestingly, over the W-BoB, about 43% of the water-soluble aerosol mass remains externally mixed, whereas, in N-BoB, all water-soluble aerosols are coated over all of the insoluble and BC. A small fraction of the mineral dust is thickly coated with BC over W-BoB, but all of the mineral dust is externally mixed in N-BoB.
335 Size-segregated measurements of aerosol vertical profiles combined with airmass trajectory analysis over BoB during W-ICARB revealed the strong presence of coarse mode aerosols in the IGP outflow (Sinha et al., 2011a). There is no similar modeling study conducted over BoB to assess probable aerosol mixing states, making it impossible to compare the current findings directly. However, comparisons with the available aerosol mixing measurements from nearby regions at different time periods are provided in Sect. 4.4.

4.3. Comparison of modeled mixing states over Kanpur and Bay of Bengal during winter

The BoB, being located downwind of the IGP, is significantly affected by the continental outflow from the IGP during the winter (Sinha et al., 2011a). During this season, a significant fraction of aerosols are in the core-shell mixed state over Kanpur and BoB. The extent of core-shell mixing is higher over N-BoB (68%) and lower over W-BoB (48%) compared to Kanpur in winter (59%) (see Figs. 3d and 4c). The relative coating thickness of the core-shell mixed components remained similar over
345 Kanpur and BoB, though there is a difference in the mass fraction of aerosol components participating in the core-shell mixing. The similarity in the modeled mixing states, particularly water-soluble coated insoluble and BC particles in the BoB and Kanpur suggests the transport of core-shell mixed aerosol components from IGP to BoB during winter. Additionally, the thick coating of BC over a small fraction of mineral dust aerosols in the W-BoB region, which is not observed in Kanpur, indicates the effect of the long-range transport on the mixing state of the mineral dust and BC aerosols. While the M_{WS} over W-BoB is
350 merely half of that over Kanpur, 43.4% of the M_{WS} is in the externally mixed state over W-BoB compared to only 8% of M_{WS} over Kanpur. This suggests a possible contribution of water-soluble aerosols (anthropogenic) to W-BoB from proximal sources, along with continental outflow from IGP. The probable core-shell mixing scenarios presented in this study for Kanpur and the BoB are similar to those in other urban (Clarke et al., 2004; Arimoto et al., 2006) and marine regions (Guazzotti et al., 2001) and are discussed in detail in the next section.

4.4. Comparison of aerosol mixing states obtained with AeroMix, SP2, and Electron Microscopy

Measurements with an SP2 and an aerosol mass spectrometer (AMS) over Kanpur indicate 61.6±9.8% of the total number of BC aerosols are thickly coated in winter with low volatile oxygenated organic aerosols, ammonium, and nitrate (Shamjad et al., 2016; Thamban et al., 2017). These findings are qualitatively consistent with AeroMix, which models 51.8% of BC mass in a core-shell structure over Kanpur in winter. Due to the lack of concurrent aerosol mixing state inferred through TEM
360 analysis at Kanpur, we qualitatively compare AeroMix and TEM-derived core-shell structures available over Delhi in the

winter of 2014 (Mishra et al., 2018). Similar aerosol characteristics over Kanpur and Delhi in the Eastern IGP are anticipated, supported by recent findings indicating dominant anthropogenic aerosols over the entire IGP region in winter (Kumar et al., 2018; Ojha et al., 2020). The key anthropogenic aerosols, namely BC, sulfate, and organic aerosols, generally favor the core-shell structure. In winter over Delhi, observations revealed semi-aged carbon fractal aggregates are embedded into sulfate or
365 organic coats (Mishra et al., 2018). Other studies reported similar findings over Delhi and Varanasi in the IGP during winter (Tiwari et al., 2015; Murari et al., 2016).

Electron micrographs of the aerosol samples collected from the regions impacted by emissions from heavy industries suggest the presence of fine metal and fly ash particles coated with secondary aerosols (Li et al., 2016), supporting the probable existence of IS-WS aerosols, as modeled by AeroMix over the heavily industrialized Kanpur throughout the year and in the
370 outflow. Similar observations of the desert outflows at various locations have reported the presence of BC-coated MD aerosols (Arimoto et al., 2006; Clarke et al., 2004; Deboudt et al., 2010), which is probable during the premonsoon season in Kanpur and in the IGP outflow. BC aerosols coated with WS are commonly reported over urban locations (Clarke et al., 2004; Dong et al., 2019; Li et al., 2016; Kompalli et al., 2020), agreeing with results obtained over Kanpur using AeroMix.

A collocated measurement of BC aerosols with an SP2 and aerosol chemical composition with an Aerosol Chemical Speciation
375 Monitor (ACSM) in winter 2016-2017 over a coastal urban site of Bhubaneswar located in the IGP outflow to the W-BoB suggests coated BC aerosols (CSR ≈ 0.74), preferentially by sulfate particles (Kompalli et al., 2020). Similarly, for the continental outflow over the northern Indian Ocean from IGP via W-BoB during winter, collocated measurement with an SP2 and ACSM reported the presence of BC aerosols thickly coated (CSR ≈ 0.37-0.59) by sulfate and organic matter (Kompalli et al., 2021). These observations are consistent with the AeroMix modeled aerosol mixing states, indicating the probable
380 existence of the thickly coated BC with WS aerosols (CSR=0.4) over both W-BoB and N-BoB. This reasonable agreement between AeroMix and independent measurements, although some of these measurements are not spatially and temporally collocated, substantiates the potential capability of AeroMix to model intricate aerosol mixing states.

5. Summary and future scope

Atmospheric aerosols can be a complex mixture of different components due to their variety of sources, short residence time,
385 and dynamic nature. A Python package named 'AeroMix' was developed to model the optical properties and mixing states of complex aerosol mixtures using Mie inversion. The novel features in the AeroMix encompass the capability to model both externally and core-shell mixed aerosols, simulate optical properties of aerosol mixtures comprising any number of aerosol components, and define aerosol composition and relative humidity in up to 6 vertical layers. These features make AeroMix a valuable tool for modeling real atmospheric aerosol mixing scenarios by assessing the potential coexistence of aerosol
390 components in both externally mixed states and various core-shell mixed states by considering all possibilities simultaneously. AeroMix performance is demonstrated by modeling probable aerosol mixing states over Kanpur, India, and the Bay of Bengal (BoB), representing urban and marine environments. This study presents observationally constrained mixing states of complex

aerosol mixtures constituted by multiple aerosol components in externally mixed and/or core-shell mixed states with AeroMix using measured aerosol chemical and optical properties for the first time over Kanpur and BoB.

395   Utilizing AeroMix capability to model any number of aerosol components in a mixture, this study reveals the likely existence of multiple core-shell mixed aerosol components in the aerosol mixture over Kanpur and BoB. The AeroMix modeled aerosol mixing states are qualitatively consistent with the independent measurements using single-particle soot photometer (SP2) and transmission electron microscopy (TEM), although some of these measurements are not spatially or temporally collocated. The reasonable agreement between AeroMix and these measurements substantiates the potential capability of AeroMix to

400   model complex aerosol mixing states involving multiple core-shell mixed components. However, this study is limited to a qualitative examination of aerosol mixing states due to the inherent constraints of the inverted Mie model approach.

Although aerosols exhibit diverse morphology, AeroMix treats all aerosols as spherical particles except mineral dust. With the capability to incorporate various databases describing aerosol components, AeroMix ensures a better representation of aerosol properties as more refined aerosol data becomes available, accounting for morphological effects on optical properties. The

405   scope of AeroMix lies in its ability to provide a robust and versatile open-source framework for computationally intensive minimization and machine learning algorithms, enabling more quantitative and deterministic estimation of aerosol mixing states. Its potential applications extend beyond aerosol studies, including astrophysics and remote sensing, particularly for atmospheric correction.

Code availability

410   The AeroMix package is publicly available on Zenodo (https://doi.org/10.5281/zenodo.10552079) and GitHub (https://github.com/sampr7/AeroMix) under GNU General Public License-3.0 (P Raj and Sinha, 2024a). The key dependencies of AeroMix package include,

1. NumPy (www.numpy.org)
2. SciPy (ww.scipy.org)
415   3. PyMieScatt (www.github.com/bsumlin/PyMieScatt).

The model outputs and codes used to generate the results presented are available in a separate Zenodo repository: https://doi.org/10.5281/zenodo.10552114 (P Raj and Sinha, 2024b).

Data availability

The AERONET level 2 data (www.aeronet.gsfc.nasa.gov/), MPLNET data (www.mplnet.gsfc.nasa.gov/), CALIOP data
420   (www.subset.larc.nasa.gov/calipso/), and RH data over Kanpur (www.mosdac.gov.in/catalog/insitu.php) are openly available from respective websites. The aerosol chemical composition over Kanpur (Ram et al., 2010), AOD (Kaskaoutis et al., 2011),

aerosol chemical composition (Srinivas et al., 2011), and mixed layer height (Subrahamanyam et al., 2012) data collected during W-ICARB campaign are presented in respective sources.

Author Contribution

425   PRS conceived the ideas and designed the study. SPR developed the AeroMix package, performed analysis, and prepared the original draft with inputs from PRS. RS contributed to the scientific discussions on mixing state calculations. DBS and SB provided the radiosonde and chemical composition data, respectively, obtained during the W-ICARB cruise and contributed to the scientific discussions.

Competing interests

430   The authors declare that they have no conflict of interest.

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