Response notes

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

Reviewer: # 3

Overall the paper is written and presented very well, though I agree with the other referees about rearranging section 3 to make a smoother read.

Verdict: Accept with minor revisions (detailed below)

Reply: We appreciate and thank the reviewer's insightful 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 to be submitted.

We have also restructured sections 2 and 3 following the first and second reviewer's comments.

General Comments

1. In the introduction (e.g. line 53), the limit on number of aerosol components that are available in OPAC is presented as a key limitation on its ability to infer mixing state. However, on line 103, the authors explain how AeroMix relies on the OPAC aerosol database for its measurements. My understanding is that AeroMix is therefore limited to the number of components available in the OPAC aerosol database. Therefore, whilst both OPAC and AeroMix rely on the OPAC aerosol database, are they not identically limited to the number of aerosol components? (By the way, I note there is an argument that users could append aerosol components to the OPAC aerosol database, and therefore extend the number of aerosol components present in AeroMix, but is this not equally true for the OPAC model? If not, then please explain.)

Reply: While both OPAC and AeroMix use the OPAC aerosol database, the limitation of OPAC is not due to the number of components available in the database (which are nine). It is due to the maximum number of components permissible in a mixture is seven for calculating the optical properties.
This is specified in the OPAC input file as depicted below.

 # if you did select 0 (define new mixture), in the next 5 lines the
*# component number and the number density in [particles/cm**3] has to be*
 # given, divided by commas. The component numbers are the following:
#
 # There are not more than 7 components allowed to compose one aerosol
 # or cloud type.
#
 # (1) insoluble (6) mineral (nuclei mode, nonspherical)
 # (2) water-soluble (7) mineral (accumulation mode, nonspherical)
 # (3) soot (8) mineral (coarse mode, nonspherical)
 # (4) sea-salt (accumulation mode)
 # (5) sea-salt (coarse mode) (10) sulfate

In contrast, AeroMix offers more flexibility, which can account for any number of aerosol components to constitute a mixture. It does utilize all the nine predefined components available in the OPAC database. In addition, it also allows user-defined aerosols in the mixture as explained in lines #113-139. In this study, along with the predefined components, their core-shell mixed combinations at various mass mixing ratios are also considered, totaling 305 components as explained in line #194.

Adding new aerosol components to OPAC would require compromising the predefined components due to the program's limitation to account for more than seven components in a mixture. With the capability of AeroMix to model any number of aerosol components in a mixture, this 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, which is why we believe it offers a more comprehensive approach to assessing the aerosol mixing state as explained in line #309.

2. It is clear from Fig. 1 that AeroMix, as a stand-alone model predicts the optical properties of aerosol populations, and I can see from line 73 and line 98 that (as in OPAC) Mie theory is used to do this, and this is represented by the 'AeroMix' item in Figure 1. However, I am confused by the Mie inversion part of AeroMix: in line 163 several references are given for the 'Mie inversion' technique to find the aerosol mixing state, however none of these references use the phrase 'Mie inversion' for their techniques. Is this phrase newly coined in this paper? If so, it should be properly justified, including an explanation of why 'inversion' is the correct word, rather than iteration. I say this because it appears from Fig 1 and the description in line 241-260 that an iterative approach is used to find the mixing state that gives best fit to observations. Therefore, I question whether this is an inversion approach, or an iterative approach. Would 'Mie iteration' be a better phrase?

Reply: We acknowledge that the term 'Mie inversion' is not used in any of the cited references. However, related phrases such as 'inverting Mie model' or 'inverse Mie problem' are frequently used when estimating aerosol properties like refractive indices from optical properties. For example, Sumlin et al. (2018) discuss solving the 'inverse Mie problem' for a complex refractive index given inputs of scattering, absorption, and size distribution parameters. Similarly, Pedrós et al. (2014) refer to 'inverting the Mie model' to infer aerosol properties from measured optical properties.

The forward Mie calculation process involves determining scattering and absorption parameters at specific wavelengths by inputting the radius and complex refractive index of the particle. Consequently, estimating particle properties, in this case, the aerosol component characterized by its spectral refractive indices and size distribution parameters, from measured optical properties, presents an inverse Mie problem. The solution to this problem is obtained by iteratively finding a combination of aerosol components (an aerosol mixture) that yields aerosol optical depth and single scattering albedo spectra that best match the measured ones, with a minimum root mean square error (RMSE) between the modeled and measured spectra (as explained in line #241).

Given the context above, we believe the term 'Mie inversion' is more appropriate than 'Mie iteration', as it accurately reflects the process of solving the inverse problem, even though it involves iterative steps.

3. Line 241 indicates that the iteration in AeroMix finds the probable existence of components, but Fig 1 and other part of the main text (e.g. line 253) suggest the iteration finds the probable mixing state. What does the iteration solve? Is it both the components present and their mixing state? If so, please make clearer.

Reply: The iteration specifically solves for the number concentration of aerosol components, with non-zero values indicating the probable existence of these components which refers to the aerosol mixing state. Line #241 is revised as follows for clarity:

"The probable existence of the aerosol components in the atmosphere, which refers to the aerosol mixing state is assessed by iteratively varying the number concentrations in the mixture in AeroMix until the RMSE between the measured and modeled AOD and SSA spectra are minimized."

4. The authors acknowledge that the iterative technique used to estimate probable mixing states is not unique (line 254). I would like to see more discussion, or reference (where this issue has been detailed before), around the range of mixing states that can be inferred from the iteration technique for a given set of inputs, i.e., a discussion around the probability that a given inferred mixing state is accurate. If I understand that AeroMix returns just one probable mixing state for a given set of inputs, then this seems to me to be a fundamental weakness, and future work should prioritise quantifying the uncertainty around the returned mixing state, and/or returning multiple mixing states so that users can quantify the range of probable solutions. I see that this issue is dealt with in lines 404-407, which explains that other algorithms could imbed AeroMix and therefore quantify the probable accuracy of returned mixing states. I also see that in sections 4.4 and 5, that the authors express the limitations of AeroMix, stating that only results useful for qualitative interpretation are returned. This is great, because it's key that the authors describe the model's limitations. However, I think this limitation should be mentioned in the model overview, so that readers are quickly aware of it. Furthermore, it would be useful to have some more description around what the 'inherent constraints' mentioned in line 401 are – this would greatly help authors of future minimization and machine learning algorithms to correctly utilise AeroMix.

Reply: The above reviewer comment has been segmented into four distinct sections for a more structured response:

a) The authors acknowledge that the iterative technique used to estimate probable mixing states is not unique (line 254). I would like to see more discussion, or reference (where this issue has been detailed before), around the range of mixing states that can be inferred from the iteration technique for a given set of inputs, i.e., a discussion around the probability that a given inferred mixing state is accurate.

   We mentioned in line #254 that the aerosol mixing state modeled by the Mie inversion technique is not unique, but a probable one. When solving a system of linear equations where the number of unknowns (number concentration of aerosol components) is greater than the number of constraints (spectral AODs and SSAs), multiple solutions are possible. This is because there are insufficient constraints to uniquely determine all the unknowns. While many possible solutions exist mathematically, not all are physically feasible for aerosols. To select a physically feasible solution for the location under consideration, we further constrained the modeled mixing states with concurrent and collocated measurements of component-wise aerosol mass concentration (line #254). The solutions in which the total aerosol mass concentration of each component (externally and core-shell mixed mass combined as explained in lines #262-270) within the bounds of ±1σ of the measured component-wise aerosol mass concentration is considered acceptable.

We have revised line #253 to clarify this point as follows.

"It is important to note that the aerosol mixing state modeled by the Mie inversion technique is not unique but a probable scenario. Solving a system of equations with number of unknowns greater than the constraints poses an undetermined system having multiple solutions (Sumlin et al., 2018). Hence, a physically feasible solution is selected from the AeroMix-modeled probable aerosol mixing states by further constraining with the measured component-wise M_a with the modeled M_a within ±1σ."

b) If I understand that AeroMix returns just one probable mixing state for a given set of inputs, then this seems to me to be a fundamental weakness, and future work should prioritise quantifying the uncertainty around the returned mixing state, and/or returning multiple mixing states so that users can quantify the range of probable solutions. I see that this issue is dealt with in lines 404-407, which explains that other algorithms could imbed AeroMix and therefore quantify the probable accuracy of returned mixing states.

As explained above, AeroMix provides not just one solution for a given set of inputs, but different probable ones depending on the combinations examined. The key limitation of this technique is that it does not guarantee a unique solution but a probable one. To assess the probability of a particular solution being true or to quantify the range of uncertainty of the solution, all possible solutions need to be modeled. This is not practical considering the number of combinations to be examined and the required computational time. For example, if we check 1000 possible values for all 305 components, the total iterations required will be 1000^{305}. As the reviewer suggested for future work, the development of minimization and machine learning algorithms is already underway to ensure a quantitative and deterministic estimation of aerosol mixing states using AeroMix as discussed in lines #404-407.

c) I also see that in sections 4.4 and 5, that the authors express the limitations of AeroMix, stating that only results useful for qualitative interpretation are returned. This is great, because it's key that the authors describe the model's limitations. However, I think this limitation should be mentioned in the model overview, so that readers are quickly aware of it.

The limitation of the AeroMix in modeling aerosol mixing states using the Mie inversion technique is that the modeled mixing states are not unique, but probable scenarios. This inherently limits the analysis of mixing states using AeroMix to a qualitative examination. Lines #74-75 in the model overview section are revised as follows to indicate that the modeled mixing states are probable ones.

"The workflow of AeroMix for modeling the aerosol properties and assessing probable mixing states is illustrated in Fig. 1. A methodology for determining the probable aerosol mixing state using AeroMix is detailed in the subsequent section."

d) Furthermore, it would be useful to have some more description around what the 'inherent constraints' mentioned in line 401 are – this would greatly help authors of future minimization and machine learning algorithms to correctly utilise AeroMix.

The inherent limitations of the Mie inversion technique are detailed in response to the first part of this comment (a) as well as in section 3.3 in the revised manuscript. Section 5 is revised to reflect this.

"However, this study is limited to a qualitative examination of aerosol mixing states due to the inherent constraints of the inverted Mie model approach as discussed in Sect. 3.3."

5. Please detail what happens along the 'No' route of Fig. 1 – the authors explain in line 241 that iterative changes are made, but how is the amount and direction of change estimated?

Reply: The direction of change is decided towards where the root mean square error between the modeled and measured AOD and SSA spectra are converged as mentioned in lines #241-243. Determination of the amount of change of component number concentration is under development and will be addressed in future work.

Minor Comments

Line 35 – please provide some example references of studies that have used OPAC to estimate probable mixing state.

Reply: The references cited in lines #49 and #163 have used OPAC to estimate the probable mixing state.

Line 57 – more detail is needed about why AEROgui is inferior to AeroMix, so that readers can distinguish between the two. For example, what limits in functionality does AEROgui have that AeroMix overcomes?

Reply: We removed this discussion about AEROgui to avoid ambiguity following the first reviewer's comment.

Line 137 – I don't understand the sentence starting on this line. Specifically, how can an internally mixed aerosol be treated as an external mixture? This sounds like the components comprising the internally mixed aerosol are separated into separate particles so that an external mixture is formed.

Reply: To clarify, for the Mie inversion technique, all the probable aerosol components need to be modeled in AeroMix beforehand to examine their existence in the mixture. Core-shell mixed aerosols are modeled as user-defined aerosol components, as explained in line #113 before they are constituted into a mixture. AeroMix treats this core-shell mixed aerosol in the same manner as an externally mixed aerosol. In other words, for AeroMix, a core-shell mixed aerosol is another aerosol with characteristic spectral Mie coefficients, size distribution parameters, and specific density (ρ). However, these Mie coefficients are calculated using the coated-sphere Mie scattering theory and specific density as explained in line #124. AeroMix assumes no chemical or physical interaction among the particles within the mixture while calculating the optical properties.

Subsequently, different combinations of predefined externally mixed aerosols (composed of single chemical species) and internally (core-shell) mixed aerosols (composed of multiple chemical species) are used to constitute various aerosol mixtures in AeroMix to calculate their optical properties.

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 as core and shell), with no presumed chemical or physical interaction among the particles within the mixture (see Fig. 1)."

References

Pedrós, R., Gómez-Amo, J. L., Marcos, C. R., Utrillas, M. P., Gandía, S., Tena, F., and Lozano, J. A. M.: AEROgui: A graphical user interface for the optical properties of aerosols, Bull. Am. Meteorol. Soc., 95, 1863–1871, https://doi.org/10.1175/BAMS-D-13-00162.1, 2014.

Sumlin, B. J., Heinson, W. R., and Chakrabarty, R. K.: Retrieving the aerosol complex refractive index using PyMieScatt: A Mie computational package with visualization capabilities, J. Quant. Spectrosc. Radiat. Transf., 205, 127–134, https://doi.org/10.1016/J.JQSRT.2017.10.012, 2018.