RC1: 'Comment on egusphere-2025-454', Anonymous Referee #1

The authors use K-means clustering on AERONET data to identify different regimes of aerosol optical properties over the Iberian Peninsula, and subsequently train random forests to predict these regimes from aerosol column densities provided by MERRA-2. While the paper is interesting and fits the journal, I would still recommend major revisions, please refer to comments below:

We would like to thank the Anonymous Referee #1 for the willingness to review the manuscript and for the insightful questions and suggestions, which have undoubtedly contributed to improving the clarity of our paper. While recognizing the importance of certain questions and concepts, we have chosen to further discuss them here for clarification, but omitted or summarized some of them in the manuscript since we understand that they are aspects familiar to the atmospheric aerosol modelling community. We aim to keep the article as concise as possible, given its already long extension. We hope the reviewer understands this need.

The main objective of the manuscript is to present an alternative approach for atmospheric aerosol/climate modeling communities to prescribe aerosol spectral optical properties(optical models) in climate models. Our prescription strategy incorporates a decision criterion based on the combination of mass loading of aerosol species (Dust, Sea salt, Black carbon etc.) present in each grid point air mass. This approach seeks to produce a more realistic representation of the spatial and temporal dynamics of intensive optical properties in climate models, which are needed for calculating the Aerosol Optical Depth(AOD), the key variable for simulating the Direct Radiative Effect of aerosol particles. This represents an advancement over conventional methods that rely on static areas of influence and predefined seasonality based on aerosol source emission activity (ex. Hoelzemann et al., 2009, Rosario et al., 2013, Chen et al., 2024), and also a reference to confront the approach based on prescription of individual aerosol types' optical properties and assumptions of mixing states (Collow et al., 2024).

Major comments:

Ln 44. "limitations in the current address information?" What do you mean with "address information"?

R: We refer to the limitation in the current global aerosol monitoring system (ground-based and satellite-based) dedicated to characterizing the spatial and temporal distribution of the spectral complex refractive index and size distribution of aerosol particles. We recognized that the expression "address information" is not clear enough to express what we meant, so we adjusted the text as follows:

"For instance, when it comes to aerosol direct interaction with radiation, the current global aerosol monitoring system does not provide a comprehensive spatial-temporal characterization of spectral complex refractive index and size distribution of the aerosol particles, two critical information to characterize the particle absorption and scattering (Samset et al. 2018, Li et al., 2022). This lack of observational data contributes significantly to uncertainty in aerosol modeling and, therefore, on the aerosol radiative forcing.."

Ln 48. "geographical representativity" Can you be more elaborate on what you mean here? Adebiyi et al. (2023) mention that libraries typical assume identical mineral compositions of certain aerosol species regardless of location. Is that what you mean here?

R: Yes, mineralogy is a good example, dust physical properties do vary from region to region depending on the soil and mineralogy composition. Additionally, other types of aerosol mixtures (urban, smoke) also differ from region to region, depending on the energy matrix and the type of biomass burned. Historically, look-up tables of optical and microphysical properties have been important to characterize and represent aerosol direct radiative effects in climate models, but their geographical limitations have also been acknowledged. So, alternative ways to capture more realistic geographical representativity of aerosol optical properties are still a matter of ongoing research. We adjusted the text as follows:

"The difficulty of the traditional libraries of aerosol optical and microphysical properties (Shettle and Fenn, 1979; Koepke et al., 1997; Hess et al., 1998) to describe aerosols properties geographical variation, for instance soil dust mineralogy (Adebiyi et al., 2023), has been central in the aerosol optical properties uncertainty debate."

Ln 63-66. I do not fully understand the contrast between "absorption" and "relative contribution of .." implied here, are absorption problems not one of the consequences of misrepresenting the fine and coarse modes?

R: To a certain degree, yes, but not necessarily; some aspects drive absorption and those are not regulated by size. Soil dust, which is a dominant component of coarse mode, can be strongly absorbing, and black carbon (BC), a fine mode aerosol type, is also strongly absorbing, and the state of mixture (internal or external) of BC with other aerosol types is a critical aspect driving uncertainties in aerosol absorption in climate models. Also, one can prescribe (or predict) dust size distribution accurately and miss its absorption due to a lack of accuracy in its mineralogy composition. Regarding the misrepresentation of fine/coarse mode relative contributions, it has important implications for the scattering process.

Ln 76. Bias in what quantity?

R:We adjusted the text as follows, so it can be clearer

Former text that include "bias in aerosol simulations" was replaced for new one:

Former text: "Zhong et al. (2022) used relationships from an ensemble of aerosol models and satellite observations to identify the primary source of uncertainty in aerosol modelling results. Their study pointed out the incorrect lifetimes and the underestimation of mass extinction coefficients as the most critical drivers of bias in aerosol simulations."

New text: "Zhong et al. (2022) used relationships from an ensemble of aerosol models and satellite observations to identify the primary source of uncertainty in aerosol modelling results in biomass burning regions. Their study pointed out the incorrect simulations of lifetimes and the underestimation of mass extinction coefficients as the main reasons for their difficulty in matching observed aerosol optical depth (AOD).

Ln 107-110: This sentence is a bit ambiguous to me, do you mean that (1) dust regimes and (2) smoke regimes are the major source of differences? If so, could you rephrase?

R: The reviewer is correct; the sentence needs clarification. We adjusted the text to better express its goal.

"They found larger differences between the strong and moderately absorbing aerosol regimes, namely dust and smoke regimes, when comparing global and regional clustering results. This is a consequence of the differences between China's regional dust and smoke aerosol particles' physical and chemical characteristics and those of global dust and smoke mean features."

Could you discus/comment on the accuracy of AERONET retrievals, either in section 2.2 or later in the results? To what extent can we consider these the ground truth?

R: AERONET retrievals` accuracy is well documented (Dubovik et al., 2000a, 2000b, 2002, 2006, Sinyuk et al., 2020) and acknowledged when compared with alternative remote sensing platforms(specially satellite-based). With more than 25 years of operating a vast network of Cimel Electronique Sun—sky radiometers across the world the Aerosol Robotic Network (AERONET) has provided highly accurate, ground-truth measurements of aerosol optical optical depth and other properties (Giles et al.,2019). It has been widely used as the main reference to evaluate and validate satellites (Gupta et al., 2018) and model products (Gliß et al., 2021).

It would take a long text describing uncertainty for all AERONET inversion products, but two worth mentioning here are the uncertainties in SSA (single scattering albedo) and ASY(asymmetry parameter), two critical intensive optical properties that are retrieved by AERONET and that are related, respectively, to absorption and size of the aerosol. For AOD > 0.4 at 440 nm (or >0.2 at longer λ), SSA uncertainty $\approx \pm 0.03$, for lower AOD, uncertainty can be ± 0.05 –0.07 or larger. Regarding ASY, uncertainty is about ± 0.02 –0.05 when AOD is high (≥ 0.4 at 440 nm, ≥ 0.2 at longer wavelengths), but can be significantly larger at low AOD.

As suggested, a brief description on the reliability and accuracy of the AERONET product is introduced in section 2.2.

Section 2.1. To me, this section felt to elaborate and sometimes irrelevant for your study. I would suggest to limit it to a short overview of what aerosol types may be expected where (and when), perhaps aided by showing (long-term averages of) MERRA column densities or something similar, without the climate information

R:The authors respectfully disagree with the reviewer's perspective. We believe that characterizing the study region is indeed crucial for understanding the aerosol sources and transport patterns that we aim to model. This is especially important when dealing with natural aerosols, such as dust, where emissions are primarily driven by winds.

Figure 1. Related to the previous comment, I am not sure surface elevation is the most relevant quantity to show here.

R: Elevation was included on the basis that it is an important aspect to understand the context of AERONET sites. For instance, coastal proximity and the depth of inland location

are important factors in discussing the nature of the air mass affecting the site, which is also influenced by its elevation. That is why we combined the site's geographical location with its elevation to gain a complementary perspective.

Section 2.2: Could you provide more information on the time resolution of the AERONET data, and the time periods used for the study?

R: AERONET time resolution and the periods used for this study are informed in section 2.2 (in the new version of the manuscript).

"The interval between direct sun measurements is typically 15 minutes, but only cloud-free conditions are considered for aerosol retrievals. For sky radiance measurements up to nine times a day at the wavelengths 0.44, 0.67, 0.87, and 1.02 µm are performed(Sinyuk et al., 2020). Regarding the time period for the current study it extends from 2003 to 2023. However, due to calibration and other operational aspects, some AERONET sites present different time ranges within this period."

Ln 213: What is the Lidar Ratio and what is it used for?

R: Lidar Ratio represents the ratio of the extinction coefficient (scattering and absorption) to the backscatter coefficient (scattering specifically at 180 degrees). It is a key parameter for identifying and categorizing different types of aerosols since light scattering depends on the size of the particle relative to the wavelength of light. Small particles, such as those from smoke plumes, usually have a high Lidar Ratio, while large particles, such as sea salt, have a small Lidar Ratio. We included a brief description of Lidar Ratio in the manuscript as follows:

"The Lidar Ratio is the ratio of the extinction coefficient to the backscatter coefficient and is crucial for identifying different aerosol types. It reflects how light scattering varies with particle size relative to the light wavelength. Small particles, like smoke, have a high Lidar Ratio, while large particles, like sea salt, have a low Lidar Ratio."

Ln 216: I assumed "mixed" in this context means multiple aerosols occurring in one column, irrespective of height. Does it matter for your optical properties and retrieval thereof whether at what height different aerosol species are occurring and (how) does that affect your work?

R: Mixed state in the context of aerosol particles refers to either external or internal mixing. In an external mixing state, you may have different aerosol particles, but still each particle corresponds to a specific composition or aerosol type (ex. Black-carbon, Dust, Sulphate). In an internal mixing state, a particle is a combination of different compositions or aerosol types, for instance, with a core of Black Carbon and a coated sulfate shell. This variability in mixing states introduces uncertainty into aerosol modeling. Traditional aerosol models typically describe the emission of specific aerosol types and assume a particular mixing state, most often external. They then calculate optical properties as a weighted mean of the individual aerosol components. Our approach, however, avoids any assumptions about the mixing state.

Ln 246-247: Consider introducing table 1 at the start of previous paragraph (ln. 221). Lines 221-229 feel out of place now.

R:Thanks, Table 1 was moved accordingly

Table 1. Am I right that you don't use aerosol optical depth? How would that be derived when your model is used? Also, would your approach also work if you only include the optical properties are used in radiative transfer calculations (SSA, ASY at different wave lengths)?

R: Aerosol optical depth (AOD) is an extensive variable, meaning it does not solely express the intrinsic features of aerosols but also the aerosol loading in the vertical column. Actually, the final goal of this method is to provide an aerosol optical properties model (including SSA and ASY) that will allow the models to improve radiative transfer calculations, via better simulations of aerosol optical depth, assuming the aerosol loading is adequately represented in the models. If climate models present a more accurate prescription of SSA, and ASY, they will better simulate aerosol optical depth and, consequently, better simulate radiative transfer.

Ln 296: What exactly is the Optical Model in this context?

R: Optical Model here refers to typical (climatological) spectral values of aerosol intrinsic optical properties used in atmospheric radiative transfer calculations. By averaging the components of each cluster obtained, we produce mean values of spectral optical properties or mean values of microphysical properties(eg. size distribution) that can be used to calculate mean spectral optical properties for the aerosol regimes. Usually, aerosol optical models are composed of spectral single-scattering albedo (SSA), asymmetry parameter(ASY), and extinction efficiency (Qext). For the sake of clarity, we replaced "optical model" with "aerosol spectral optical model". We also made a few adjustments to the sentence to improve its reading.(Ln 323 - 325, new version). We also believe that the introductory text at the beginning of this reply helps to better understand the importance and the context of optical models here discussed.

Ln 314-320: I am not sure I fully understand the equation and accompanying text, are you computing WCSS by minimising sum_1^k W(ck), or are you seeking to minimise WCSS? Also, could you explain more what you mean by Elbow?

R: Within-cluster sum of squares (WCSS) measures how compact or tight the clusters are. For each cluster, it calculates the distance of each point in the cluster to the cluster centroid, then squares it and sums them up. Then, the sums were added up for all clusters. If clusters are tight and well-separated, WCSS will be small, because points are close to their centroids. If clusters are loose or overlapping, WCSS will be large. The Elbow method is based on the computation of WCSS for different numbers of clusters. As the number of clusters increases, WCSS always decreases (more clusters = tighter groups). At some point, the decrease becomes less significant. The plot of WCSS against the number of clusters will display a feature similar to an "elbow". The number of clusters where the WCSS decrease becomes less significant is the one to be selected.

We try to improve the explanation of WCSS and the Elbow Method in the text (Ln 337 – 341, new version)

Ln 251-253: what do you mean with class imbalance, and what issues regarding atmospheric measurements are you addressing exactly?

R: Class imbalance refers to the unequal representation of aerosol regimes in the dataset, where some clusters (such as C2, representing only 3.48% of observations) occur much less frequently than others (such as C1, representing 37.88%). This imbalance is typical in atmospheric measurements, where extreme but radiatively important aerosol events like intense smoke episodes are rare compared to more common background conditions. Despite their low frequency, accurate prediction of minority classes like C2 is crucial for radiative transfer calculations due to their high absorption characteristics and significant impact on aerosol direct radiative forcing.

Ln 394: What is cluster stability?

R: Stability is also used as a criterion to define the number of clusters in data analysis. It helps to evaluate whether the number of identified clusters is meaningful or merely an artifact of randomness or noise. The variation in the number of clusters k in k-means is used to help identify the value that yields the most stable clustering, or the value above which there is a strong decrease in stability. High stability suggests that the clusters represent genuine patterns in the data, rather than just random fluctuations. We used the stability criteria to corroborate the number of clusters identified by the Elbow Method as the ideal choice.

Part of the explanations described here were included in the text to clarify the importance of cluster stability. (Ln: 419 – 422, new version)

Ln 438: you mention marine particle scenarios here in relation to Clusters C2-C4, but marine aerosols are barely mentioned later on. Are they not important enough, or present in all regimes in similar quantities?

R: Although present in the regional atmosphere, especially in coastal areas, marine aerosols by themself do not generate high aerosol loading scenarios, and therefore high radiative forcing over the study region, as seen in dust, smoke, and urban-pollution scenarios. As the AERONET sky inversions are restricted for AOD at 440 nm higher than 0.4, it is expected that none of the clusters represent a pure or a dominant marine aerosol regime. That's why marine aerosols are not exhaustively discussed. However, as part of the background particles across the Iberian Peninsula, marine aerosol particles undoubtedly influence the clusters identified to some degree, particularly those dominant in coastal regions.

Ln 3.1: What are the mean aerosol column densities from MERRA-2 for each regime?

R: Unless we misunderstood the referee's request, plotting average aerosol column density for each aerosol species(6) and for each regime (5) will result in a significant number of maps. This would require a proper discussion, which would make the manuscript even longer. Also, generating these maps for the annual mean would not capture the regime dynamic we seek to represent, then we would have to do it monthly(12), which again would translate into an even higher number of plots. Moreover, we are not convinced that the aerosol column density for each aerosol species would add to the discussion, as it alone cannot be translated into radiative forcing. But we evaluate to include monthly climatology of Aerosol Optical Depth(critical to radiative forcing calculations) over the Iberian Peninsula (Figure below) in the supplements to provide a perspective on the spatial and monthly

dynamic of aerosol loading in the region. We believe that this also addresses the referee's comment regarding Figure 1. In the text where we discussed

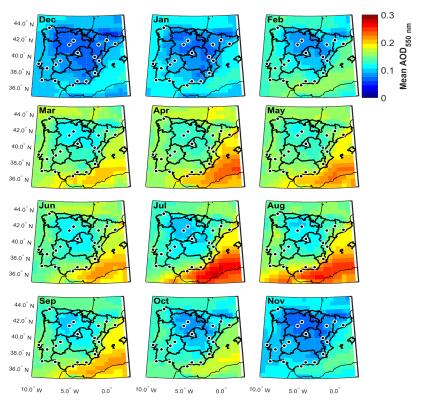


Figure R1: Monthly mean Aerosol Optical Depth at 550 nm over Iberian Peninsula and surroundings for the period from 2003 to 2023.

Figure 4: based on what data are these size distribution computed?

R: The size distributions mentioned are the average of the instantaneous size distributions retrieved by AERONET from each identified cluster. As shown in Figure 3, which includes ASY, SSA, and other properties, each cluster is composed of many instantaneous retrievals. Figure 4 displays the average size distributions for each cluster.

Ln 492-493, table 3: Where are the contest of this table discussed? Line 500 perhaps, I would refer to table 3 on that specific line.

R:Table 3 is introduced at line 453 (former version) and line 485 (new version), and is mentioned again at line 492 (former version) and line 524 (new version). To reinforce the contextualization of table 3, as suggested. We also referred to it at line 500 (former version) and 532 (new version).

A general comment on figures, I find the large variety in font sizes (for example, large bold text in Figure 6, small font size in Figures 11 and 12 in combination with the low dpi of some figures is distracting from their main message.

R: We managed to adjust and improve the font and resolution of the figures as suggested.

Ln 501: This hypothesis was already stated on line 470.

R: Thanks for pointing that out. To avoid repetition, as suggested, we omitted the sentence related to the comment.

Ln 508-509: "however, ..., but..." consider removing either however or but? **R**:"However" term was removed and the text adjusted. (Ln 540 new version).

Ln 535: do you have any metrics showing whether the random forests overfitting on its training data?

R:Thanks for this comment. Now we included metrics that allow us to evaluate overfitting. We generated classification reports for the training data to be compared with the previous report generated for the test data presented in Table 4. We created a new Table 4 that will replace the former one. These results indicate that the model generalizes well, without significant overfitting. Even for Cluster 2, which has a small number of occurrences, the model was able to maintain high precision and score. The general accuracy did not drop critically for the test data (0.70) when compared with the train dataset (0.88), another indicator of the model's ability to generalize.

The manuscript text has been adjusted to integrate the metrics and new results displayed here. Table 4 also was adjusted.

Table 4. Performance metrics values of the trained model applied to the test and the train(within parenthesis) dataset to predict aerosol optical regimes based on aerosol-type column mass density.

Clusters	Precision	Recall	F1-Score	Support (N)
0	0.62(0.89)	0.62(0.78)	0.62(0.83)	394(848)
1	0.68(0.86)	0.70(0.94)	0.69(0.90)	452(1140)
2	0.62(0.92)	0.60(0.76)	0.61(.83)	62(111)
3	0.76(0.93)	0.73(0.94)	0.74(0.94)	251(439)
4	0.68(0.91)	0.69(0.93)	0.69(0.92)	185(397)

Line 563: Could you elaborate on the "extra training", would you need extra variables, longer time series?

R: Sure, longer time series are an important aspect in the context of extra training, particularly when examining the C2 cluster, which has a lower occurrence compared with the other clusters. However, that is not the only aspect; training with additional predictors could also be beneficial. For example, brown carbon, an important aerosol component, is not yet available in the current MERRA-2 aerosol reanalysis products. This further discussion was included in the manuscript (Ln 595-598, new version).

Ln 573: Accuracy is not included in Table 4.

R: Well observed, thanks. Accuracy is referred to in the text and not displayed in Table 4. We adjusted the text to consider this observation. (Ln 607, new version)

Ln 580: Looking at Table 4, does C2 not have the lowest precision?

R: The reviewer is correct, thanks for pointing that out. The discussion was adjusted to describe and contextualize C2 precision properly. (Ln 614-620 new version)

Ln 590: Based on Figure 10, is organic carbon really one of the primary factors? It's relative important of organic carbon is very similar to sea salt, SO4, and SO2.

R: We rephrased the sentence to contextualize the role of organic carbon better. Although second in terms of importance, the organic carbon relative importance value is closer to sea salt and sulphate aerosol types when compared to dust. Dust occurrence, indeed, is by far the dominant predictor of the cluster. The sentence was adjusted to express this description. (Ln: 627 - 633, new version)

Ln 623/698: Could you discuss these AOD thresholds in more detail? Is your model only trained for cases with a higher AOD? What percentage of time is this threshold actually reached? I suppose that even for relatively low aerosol optical depths, you would still want optical properties to be well-represented.

R: This AOD threshold requirement is a widely recognized limitation of AERONET retrieval of aerosol properties, particularly regarding absorption (specifically SSA). The AERONET algorithm can only retrieve SSA when the AOD at 440 nm is higher than 0.4. Consequently, all studies based on the AERONET database to prescribe aerosol optical models must adhere to this constraint, i.e., optical models are built based on AOD conditions higher than 0.4. The frequency with which this threshold is met varies from region to region and site. In the Iberian Peninsula, the threshold was achieved only between 3% and 5 %. It is important to recognize that the accuracy of an optical model is influenced by aerosol loading. For conditions of low AOD, the error in radiative transfer calculation due to inaccuracies in optical models prescription is minimal. However, the significance of these errors increases as AOD rises. Ideally, optical properties should be accurately represented regardless of aerosol loading, but current climate models often struggle to predict their properties accurately. Therefore, enhancing the prescription of optical models for polluted scenarios is already a notable improvement.

Ln 703: what do you exactly mean with "randomly simulated following a Gaussian..."? Please elaborate.

R: We used the central value of each optical regime cluster (the mean) with a typical spread (the standard deviation), both calculated and presented in Table 3, to randomly generate normal distributions for SSA and AE.

Ln 705: "In addition ... size behaviors", does this refer to the Angstrom exponent (Figure 14)?

R: Yes, we adjusted the sentence to make this clearer.

Ln 714: "why does MERRA underestimate SSA in C3 and C4 despite the coarser particles?

R: There are several hypotheses for the underestimation of SSA (absorption overestimation) in MERRA-2. This issue was also highlighted in a previous study by Bakatsoula et al. (2023). One factor influencing the absorption in MERRA-2 is the lack of brown carbon in its aerosol type classification scheme. Additionally, MERRA-2 tends to underestimate SSA for other aerosol types, particularly for black carbon, as noted by Bakatsoula et al. (2023). The same study also pointed out that MERRA-2 shows excessive absorption for dust aerosol types. Therefore, the underestimation of SSA in MERRA-2 can be related to several factors. It is important to mention that despite its widespread use by the community, including this study, which indicates its importance and relevance for atmospheric studies, MERRA-2 aerosol products have inherent uncertainties that prevent them from being regarded as the absolute truth, especially regarding aerosol dynamic studies.

Ln 727:" What exactly are the expected and observed values referring to?

R:We are comparing the SSA and AE distribution as predicted by our model with MERRA-2.

We adjusted the text to clarify as follows:

Ln 723-727: The model preserves the distribution of optical properties of less frequent aerosol regimes while capturing MERRA-2 features without the need for explicit class imbalance treatment, with C2's highly absorbing and dominant fine mode conditions reflected in both SSA and AE predictions, with the distributions of values across clusters showing coherence with MERRA-2 values.

Figure 12: I am not sure if a diverging colormap is the most appropriate choice here. **R**:Thanks, we tried an alternative colormap in order to better depict the SSA variability.

Figures 13-15: Do the dashed lines show the means? If so, please state in the caption. **R**:Yes, thanks, stated in the caption.

Ln 784-786: could you elaborate on the generalizability of your approach and how your approach would benefit atmospheric models, e.g. climate simulations? Would you need to many random forests, each of a small part of the world, and use those to infer spectral optical properties given column amounts of the various aerosol species?

R: As described, we present an alternative approach for atmospheric aerosol/climate modeling communities to prescribe aerosol spectral optical properties(optical models) in climate models. With a decision criteria based on the predicted combination of mass loading of aerosol species(Dust, Sea salt, Black carbon etc.) in each grid point column, our prescription strategy search to produce a more realistic spatial and temporal dynamic in intensive optical properties in climate models required to calculate Aerosol Optical Depth(AOD), the primary variable to the simulation of aerosol Direct Radiative Effect. This is an important advance compared to the conventional prescription of optical models based on static areas of influence and on seasonality of aerosol sources emission (ex. Hoelzemann et al., 2009, Rosario et al., 2013, Chen et al., 2024). This approach can also contribute to understanding the results of climate models that, instead of prescribing an observed-based

intensive optical model, try to explicitly predict optical properties based on assumptions such as the state of mixture assumption (externally and internally).

Regarding the application of random forest: once implemented in the climate model, the model trained using random forest to prescribe the optical models will require to be fed with the combination of aerosol types column mass loading predicted by the climate model. Once the decision of the likely cluster is made, the corresponding spectral optical model will be taken from a look-up table and applied in radiative transfer calculations. The approach can rely either on look-up tables of global or regional optical models. Therefore, the model training to prescribe optical models can be done on a global or regional basis.

Ln 785: "aerosol radiative forcing" would it be possible to also illustrate the possible benefit your approach in terms of aerosol radiative effects? For example, using clear sky radiative transfer computations using either your aerosol optical properties or those from MERRA-2 aerosol optical properties.

R: We appreciate the reviewer's suggestion; however, the inclusion of aerosol radiative forcing analysis and its intercomparison between our calculations and MERRA-2 would be a new work in itself, as it would require a comprehensive analysis and deeper discussion. In this paper, we focus on describing the developed approach and on evaluating its ability to produce optical properties prescription coherent with MERRA-2 predicted optical properties. With our approach, we not only achieved this but also identified differences between our and MERRA-2 optical properties prescriptions that are consistent with previous studies that evaluated MERRA-2 against AERONET (Bakatsoula et al., 2023).

Minor comments & technical corrections

Ln 16. "simulation aerosol" -> "simulation of aerosol"

R: Thanks, replaced

Ln 20. "a observational-based" -> "an observation-based" R: Thanks, corrected

Ln 37. "Aerosol particles' importance", maybe rephrase to "The importance of aerosols" R: Thanks, rephrased

Ln 38. What do you mean with "direct players"

R: The sentence (Ln 38-40) was adjusted to better express the meaning of direct players.

Ln 40. Could you be more specific with this sentence, it is not very clear to me **R**: We believe that with the adjustment made in the previous sentence and a minor adjustment (inclusion of the term "participation") the sentence is now clearer. (Ln 39-42)

Ln 60. "due to treatment of aerosol mixing state" can you formulate more clearly?

R: Aerosol particles can be found in the atmosphere externally mixed (each particle represents a specific chemical composition) and/or internally mixed (one particle is a combination of particles with different compositions). While all aerosols scatter light, only a few species are significant absorbers of incoming solar radiation. These absorbing aerosol species include Black-Carbon(BC), dust, and absorbing organic aerosol (called brown carbon (BrC)). Condensable gas-phase species create a secondary aerosol coating of predominantly scattering material that either fully or partially coats these absorbing aerosols, this is usually how the internal mixing state is produced. In the case of BC, secondary and primary coatings can enhance absorption of light. Climate models struggle to simulate properly an aerosol mixing state, several just assume external mixing, which is not often the case. While some try to simulate somehow the internal mixing. Brown et al. (2021) point out that idealized internal mixing assumptions used in climate models tend to overestimate BC absorption enhancement when compared to observations. This is just one of the aspects related

We reformulated the sentence(Ln 70 new version):

Ln 70. "microphysical" missing word?

R: Yes, thanks. We added properties, so now it reads "*microphysical properties*" (Ln 82 new version)

Ln 102-103. "aerosol scenarios variability", should "variability" be omitted here? **R**: Yes, it sounds better, thanks!. Variability term omitted. (Ln 115 new version)

Ln 241. When or where?

R: We omitted "where". (Ln 258 new version)

Ln 336-337: "data fitted to a training process,..." could you rephrase more clearly

Ln 339-341: "mass density, trying to" This sentence does not flow well.

R: Considering the reviewer comments on both lines references, we rewrote the entire sentence, from Ln 336 to 341(old version), as follows:

"With this, we built a time series of collocated MERRA-2 aerosol types of column mass density with the developed clusters occurrences over each AERONET site, which was used in a training process aiming to predict the suitable cluster given a specific combination of aerosol types of column mass density predicted."

We believe that the sentence is clearer. (Ln: 358-362 new version)

Ln 367: "takes .. account" is redundant.

R:Thanks, "into account" was omitted (In 338 new version)

Figure 3 misses (a) (b) (c) (d) labels

R: Thanks, labels included.

Figure 9: The colorbar misses a label **R**: Thanks, colorbar label included

Ln 582: What do you mean with "cost"?

R: We understand that replace cost would provide better understanding, so the sentence was adjusted to

"mislabeling this aerosol regime would translate in higher radiative error"

Ln 635: What do you mean with "corridor"?

R: Corridor meant the path (or way). We replace "corridor" with "path" so it can be clearer. (Ln: 652 new version).

Ln 658: "Regimes regarding" -> "regimes such as"?

R:Adjusted as suggested (Ln: 675)

Ln 682: "550 nm field" -> "550 nm"

R: "field" omitted. (Ln: 699 new version)

Ln 723: What do you mean with "physical distribution characteristics"?

R:We changed the words to clarify as follows:

Ln 723-727: The model preserves the distribution of optical properties of less frequent aerosol regimes while capturing MERRA-2 features without the need for explicit class imbalance treatment, with C2's highly absorbing and dominant fine mode conditions reflected in both SSA and AE predictions, with the distributions of values across clusters showing coherence with MERRA-2 values.

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