RC2: 'Comment on egusphere-2025-454', Anonymous Referee #2, 15 Jul 2025 reply

The study characterizes the typical aerosol intensive optical properties affecting the Iberian Peninsula (IP), comprising Spain and Portugal, using the atmospheric column inversion products from the AERONET sites. The authors employed K-means clustering to analyze historical aerosol intensive properties across all AERONET that operated for at least 2 years and has the highest quality dataset level 2.0 available. Five distinct aerosol optical regimes affecting the IP were identified based on the clustering technique, followed by the utilization of aerosol-type columnar mass density data (dust, organic carbon, black carbon, sea-salt, and sulphates) from MERRA-2 reanalysis to predict the aerosol optical regime using the Random Forest supervised learning methodology.

The performance of the trained model was tested under various aerosol scenarios, and the predictions ranged from 60% to 75% with accuracy exceeding 90% when predicting solely dust or non-dust optical regimes. Overall, the study is very interesting and its to the journal scope. The manuscript is well-written but requires some improvement in clarity on certain aspects before re-consideration. Recent literature needs to be cited.

We truly appreciate the time and effort Anonymous Referee #2 put into reviewing our manuscript. Their thoughtful questions and suggestions helped clarify our work and offered meaningful ideas for how we might build on it in the future. We are grateful for the constructive feedback, which will undoubtedly be very helpful in shaping the next steps of our research.

Comments:

Line 37: Statement starting with 'Via'?

Reply: The text was adjusted to attend reviewer 01 requirement and also to avoid to start “Via” (Ln 39, new version)

Line 70: compositions -> composition,

Reply: replaced

Line 70: It should be 'microphysical properties'
Reply: corrected

Line 70: computations -> computation
Reply: corrected

Line 76: What parameters are being referred to in 'aerosol simulation'?
Reply: The text was adjusted to better described Zhong et al. (2002) results. (Ln 89, new version)

Former text 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).*

Line 197: What do you mean by observation-constrained approaches? Are you referring to the threshold based aerosol type classification methods? Please clarify.
Reply: The term "observation-based approaches" better describes what we meant, so we adjusted the text accordingly.
Traditionally, climate models assume, constrained by computational capacity, a limited set of aerosol species, including organic carbon, black carbon, dust, sulfate, and marine aerosols. These models explicitly resolve the transport and removal processes of these species, treating them as an external mixture. Therefore, the optical properties involved in radiative interactions are calculated as a mass-weighted average of individual species at each grid point. The assumption of external mixing may not always be accurate, leading to significant uncertainties, such as excessive absorption by smoke aerosols and inaccuracies in dust size fractions.
Observation-based approaches, such as those provided by AERONET retrieval climatology, attribute intensive optical properties to an effective aerosol based on actual observations. This method aims to reduce the uncertainties arising from the explicit simulation of these properties in climate models. The Iberian Peninsula, influenced by a variety of aerosol types—including dust, smoke, urban-industrial emissions, and marine aerosols—presents an interesting region to test this hypothesis.

We've adjusted the text to make the idea of "restricted observational approaches" clearer (Ln 216-218).

Lines 211-215: What is the rationale for choosing these aerosol intensive properties? How is Lidar Ratio (LR) and Linear Depolarization Ratio (LDR) derived with AERONET sky radiance measurements? How reliable are the LR and LDR derived from AERONET?

Reply: This set of aerosol intensive properties is expected to capture most of the important aspects that differentiate the distinct aerosols optical regime that affect the study region. For instance, the imaginary part of the complex refractive index and single scattering albedo (SSA) are properties indicated to separate highly absorbing aerosol regimes from moderate and low absorbing regimes. Angstrom Exponent (AE) and Asymmetry Parameter (ASY) are properties that help separate aerosol regimes characterized by distinct size distribution. LR is highly sensitive to size and composition-related information, for instance, the real part of the complex refractive index. Meanwhile, LDR has high sensitivity to particle morphology, and it is widely used to separate dust particles from other aerosol types. The combination of LR x LDR has proved to be very helpful to categorize aerosol regimes(Groß et al., 2012*).

AERONET retrievals of LR and LDR are based on Dubovik et al (2006). Below is presented a summary based on Shin et al. (2018), which evaluated AERONET version 3 products evaluation focusing on LR and LDR.

For each AERONET observation, the elements F11(r,n) and F22(r,n) of the Müller scattering matrix (Bohren and Huffman, 1983) are computed from the particle size distribution and the refractive index that have been inferred from the AERONET inversion product. The element F11(r,n) is proportional to the flux of scattered light in the case of unpolarised incident light, while F22(r,n) strongly depends on the angular and spectral distribution of the radiative intensity (Bohren and Huffman, 1983) as measured with AERONET's instruments (Dubovik et al., 2006). From the element F11(r,n) at the scattering angle of 180◦ and the concurrently inferred single-scattering albedo (SSA), the lidar ratio can be computed.

$$S_\lambda^{\mathrm{p}} = \frac{4\pi}{\varpi_\lambda F_{11,\lambda}(r,n,180°)}.$$

The calculation of the particle linear depolarisation ratio requires knowledge of the elements F11(r,n) and F22(r,n) at a scattering angle of 180◦:

$$\delta_\lambda^{\mathrm{p}} = \frac{1 - F_{22,\lambda}(r,n,180°)/F_{11,\lambda}(r,n,180°)}{1 + F_{22,\lambda}(r,n,180°)/F_{11,\lambda}(r,n,180°)}.$$

Several other assumptions affect AERONET quality, for instance, external mixture. However, according to Shin et al. (2018) study, the quality of the retrievals of LR and LDR from AERONET have increased for the network products version 3(used in this study), LR and LDR show an overall improvement by moving to more realistic values when compared with previous evaluation(Müller et al., 2012).

Line 235: Which climate models are being referred here?
Reply: Climate models in general, but more specifically the CMIP6 models (Zhao et al., 2022). This was included in the text in order to be more specific.(Ln: 258, new version)

Table 1: Are these VMR-F, VMR-C, STD-F, STD-C, Reff-F, Reff-C provided by the AERONET inversion products or these are derived by the authors? Please clarify. Since these intensive properties are inversion products of AERONET, how did you account for their uncertainty impacting the aerosol optical regimes identified through K-means clustering (Section 2.4)? There is no much discussion on the influence of the observational/inversion uncertainty of aerosol intensive properties on the identified clusters and interpretation of your results.
Reply: The mentioned variables are the AERONET inversion products. We adjusted the title of Table 1 to make it more straightforward.
The reviewer is correct; all AERONET inversion products are subject to uncertainty. There are the traditional variables that have been exhaustively applied and evaluated in the literature, like SSA, and others included in recent years, like LDR, that are still subject to further evaluation to quantify their uncertainty and reliability better. Nevertheless, AERONET data undoubtedly has the best spatial and temporal coverage worldwide. Therefore, balancing data availability and uncertainties, we chose to proceed with our work.
In this study, we did not consider how the different degree of uncertainty that characterizes each inversion product influences the clustering performance. It would be indeed an interesting exercise for the next phase of our work. We need to consider how to approach this task, given the diversity of variables used, and it requires a much deeper discussion. Nevertheless, the clustering process results consistently capture in aerosol regimes that are captured by previous works in the study region (Cachorro et al., 2016; Gómez-Amo et al., 2017). So, we believe that our results are solid for this initial exploration of the method. We will include a discussion in the text addressing the impact of AERONET inversion products uncertainty on clustering methods performance, identifying this as a possible limitation that warrants further evaluation in future research.

Line 286: Use Sulphate or sulfate consistently throughout the manuscript.
Reply: Thanks, sulfate was adopted throughout the manuscript.

Lines 285-290: It was mentioned that the MERRA-2 Aerosol Diagnostic Product (ADP) for aerosol types is considered in this study. Dust, Black Carbon, Organic Carbon, Sea-Salt and Sulphate aerosol mass concentration at specific levels are integrated in the entire atmospheric column to obtain columnar aerosol optical properties such extinction, scattering and absorption optical depth. It is not clear on how the mass concentrations of individual species are converted to optical depths. At least proper citation of references to the method adopted might have been included. At which wavelength these are obtained? Did you validate extinction optical depth derived from MERRA-2 with the aerosol optical depth from AERONET? Similarly, how does the SSA from MERRA-2 validate with the corresponding SSA from AERONET?

Reply: In this part of the manuscript, we aimed to describe the aerosol variables available in Merra-2 Aerosol Diagnostic Product and provide an example of their potential to derive additional information about aerosol. We did not perform the calculations inquired by the reviewer (extinction, scattering and absorption optical depth); these are by-products of running the MERRA-2 reanalysis system and made available to the community via MERRA-2 ADP. We agree that the inclusion of the citation reference for MERRA-2 ADP (Buchard et al., 2017*) is in order, so we added that in the revised manuscript.

* Buchard, V., and Coauthors, 2017: The MERRA-2 Aerosol Reanalysis, 1980 Onward. Part II: Evaluation and Case Studies. J. Climate, 30, 6851–6872, https://doi.org/10.1175/JCLI-D-16-0613.1.

The Merra-2 system provides optical properties at multiple wavelengths, but 550 nm is used as a reference to provide aerosol optical properties information in Merra-2 ADP.

In this study, we did not aim to validate Merra-2 AOD as such analysis falls outside the primary goal of this paper and would require a more comprehensive approach. Moreover, several studies have already compared between Merra-2 data and AERONET retrievals worldwide, highlighting both the strengths and challenges aspects. In this study, we primarily focus on what we refer to as intensive aerosol optical properties (ex. SSA). These properties are independent of the aerosol loading and depend solely on the characteristics of the aerosol itself. Both extinction and scattering optical depth depend on the amount of aerosol.
Regarding SSA, we did compare and discuss our results with Merra-2 (Topic 3., Figure 13, 14, 15, 3)

Line 309: There exist several methods and indices to decide on the appropriate number of clusters such as Elbow, Silhouette, Davies Bouldin, and Calinski-Harabasz indices. I have noticed that in the following study: https://doi.org/10.1016/j.atmosres.2022.106518, the authors have stated that the correct number of clusters derived from different approaches may not lead to a single solution. What is the rationale for adopting the Elbow method, except the fact that it is a widely used method for determining the optimal number of clusters?
Reply: The referee is correct; there are several methods to decide the number of clusters, and we can also add the Silhouette Method to the list. Depending on the method selected, one can end up with a different number of clusters. Indeed, among the cited methods, there are those more rigorous than the Elbow Method, which is regarded by its simplicity. To reduce subjectivity, we combined Elbow and Stability methods to evaluate the optimal cluster number that best captures the diversity of aerosol regime affecting the study region. As illustrated on Figure 2, the results from the stability and elbow methods indicated potential for defining a distinct number of clusters, though we found that the number is likely to be around five in our case. Based on our clustering results and our experience with optical properties regime studies, we believe that the decision based on the combined methods effectively identified five optical properties regimes that characterizes coherently the variability of aerosol regimes affecting the Iberian Peninsula. Nevertheless, apart from its positive aspects, we agree it is valuable to include in the manuscript a brief description of the limitations that characterize Elbow Methods(described below).

"The Elbow method has been widely used because of its straightforward approach to estimating the most appropriate number of clusters. However, it still carries a certain degree of subjectivity, as it relies on visual interpretation. To reduce this subjectivity, we combined the Elbow and Stability methods to evaluate the optimal number of clusters that best represent the major aerosol regimes affecting the study region. Although more rigorous methods are available in the literature, defining the number of clusters remains a challenge,

and different approaches often lead to distinct solutions (Krishnaveni et al., 2023). Despite the Elbow Method limitations, the number of clusters identified in our study seems to provide a coherent characterization of optical regimes affecting the Iberian Peninsula"

Lines 325-328: What do you mean by 'clusters average'?
Reply: After defining the number of clusters (number of optical properties regimes) and the clustering process is performed, we end up with five (5) individual clusters. Each one is characterized by sets of AERONET instantaneous retrievals of optical and microphysical properties that are expected to express its optical regime. Each AERONET instantaneous retrieval is tagged with the cluster number that it belongs to. By averaging the properties of each cluster,we aim to produce a typical optical and microphysical properties set of values that represent the mentioned aerosol optical regimes.

Lines 334-336: It was not mentioned anywhere how the times were synchronized between the AERONET inversion parameters and MERRA-2 data of aerosol species column mass density. Each of the AERONET inversion parameters and MERRA-2 aerosol species column mass densities might have different ranges of variability and units. How is this accounted for in the ML model while identifying the clusters? I mean to ask if the ML model does any scaling and normalization of different parameters. If not, won't the range of variability and units have any impact on the aerosol classification?

Reply: AERONET retrievals (each one tagged with its cluster reference number) and Merra-2 column mass density are synchronized considering their proximity in time. Merra-2 column mass density of aerosol types is available with a frequency of 1 hour, while AERONET instantaneous retrievals are provided with irregular time, due to its dependence on cloud cover and AOD criteria (AOD440 nm > 0.4). So, for each AERONET retrieval, our script searches for the MERRA-2 closest hour to synchronize the two dataset.

Line 461: Large radius spread for C3 ... What does this infer?
Reply: A larger fine-mode radius (C3 case) generally means the fine particles have grown (water uptake, aging, coagulation) or that the aerosol regime mixture includes sources that naturally produce slightly larger fine particles (e.g., smoke vs. fresh urban soot). It usually indicates more aged, more hygroscopic, or more humidified aerosol compared to freshly emitted, dry fine particles.

Lines 505-506: There is no mention about categorization of seasons till now. How are months categorized into seasons?
Reply: We chose to analyze the cluster's temporal occurrence as a function of month (Figure 7) instead of seasons (ex. June-July-Aug; December-January-February) which is why we did not categorize the seasons. Where is reading "seasons" in the sentence (Ln 5050-506) should be "months". We corrected in the revised manuscript.

Table 3: What does the values in brackets correspond to? Standard deviation or error? This may be mentioned in the table caption.

Reply: Yes, the values in the brackets correspond to standard deviation, this description was included in the table caption. Thanks.

Line 555: How can you say that this would not introduce a substantial error in the radiative effect calculations? In terms of what metrics radiative effect is calculated? Radiative forcing or heating rates? Better to quantify this error. I suggest you to check this study: https://doi.org/10.1016/j.jqsrt.2024.109179, and see if this might provide some insights on the errors associated with direct radiative effects.

Reply: The idea behind this sentence is that if the classifier's confusion is between the two dust-regime models (clusters) the induced error in radiative transfer calculations would be lower than that if the confusion was between a dust and a non-dust regime, especially like C2, which is substantially different from any of the dust regime. This conclusion seems to be reasonable, even without metrics on the radiative effect. However, the reviewer pointed out an important aspect, the need for further and conclusive quantification of the radiative effect accuracy of each cluster, along with an examination of the implications of eventual misclassification. This would require an analysis not only of the columnar influence of aerosol optical properties but also of the heterogeneity of aerosol types across atmospheric layers, as stated in the suggested article. Thank you, this feedback will be very helpful for exploring the focus of a subsequent manuscript to complement the current one.

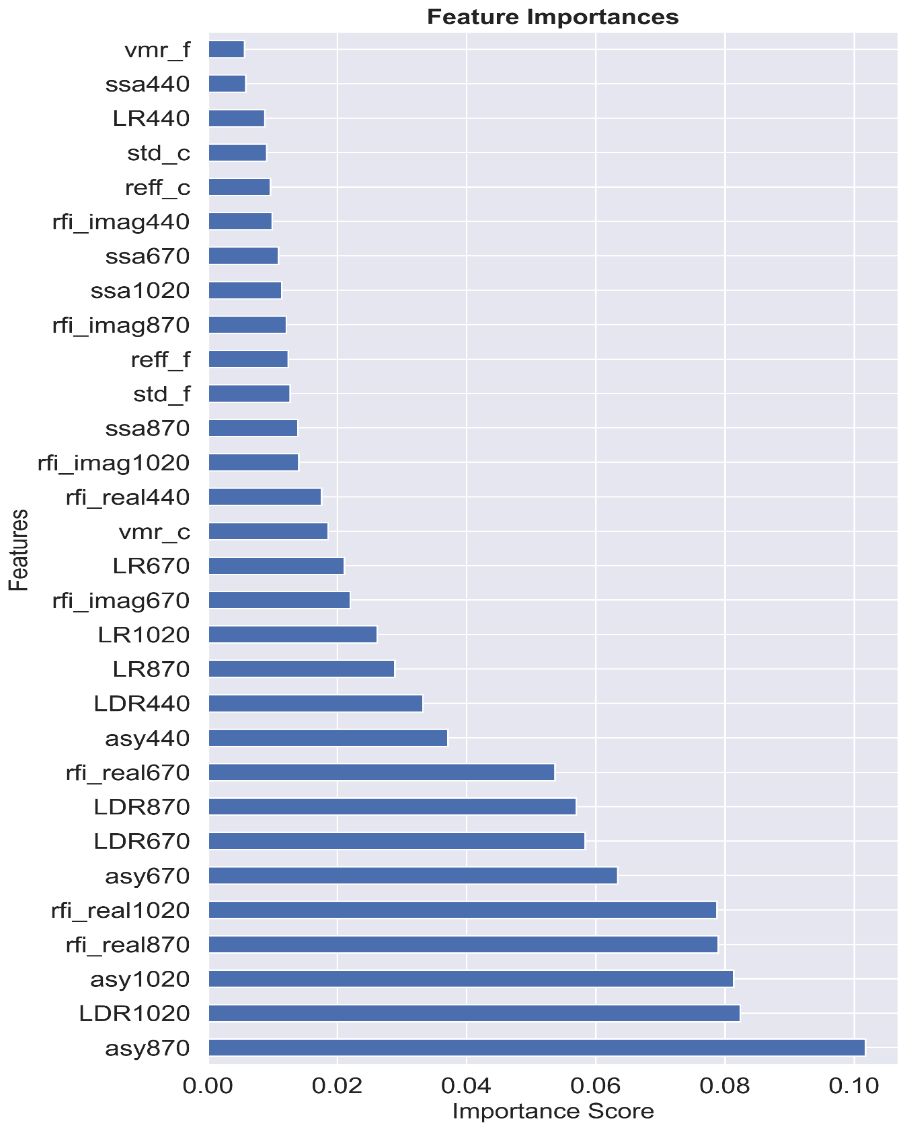
The text in the manuscript was adjusted to clarify and contextualize the expression *"introduce a substantial error in the radiative effect calculations*".

Lines 573-583: It appears that these details are repeated again. Please check and avoid repetitions.
Reply: We were unable to find the repetition mentioned. This is the only part of the text where we discuss the results in Table 4. Some of these figures are highlighted in the abstract and conclusion, but not repeated in other parts of the manuscript.

Line 598: reanalyzes -> reanalyses
Reply: Thanks, corrected.

Figure 10: Short forms (dst, oac, ssl, so4, so2, bcc) used as x-axis labels for features should be defined in the figure 10 caption. It is also not clear if this relative importance is obtained for the entire IP region or the grids consisting the AERONET sites. Can you bring out similar figures to ascertain the relative importance of aerosol intrinsic parameters (Table 1) for different clusters (or aerosol scenarios) identified in this study together with the predictor variables from MERRA-2.
Reply: The short forms were described in the caption of figure 10. The relative importance was obtained for the grids consisting of the AERONET sites. This is now clarified in the text. If we understood well, the referee is also asking for a similar analysis of relative importance but targeting the importance of the features presented in Table 1 in the clusters prediction. We managed to calculate the relative importance of predictors(Table 1) in cluster prediction (**Figure below**) . It is possible to observe consistency between this importance score scale and the importance score scale from MERRA-2, regarding aerosol types (Figure 10). In

Figure 10, the dust (dst) mass variability emerges as the most relevant factor for determining which cluster should be applied. In the figure below, which shows the importance of the optical parameters from Table 1 for clustering, the scores are well distributed (maximum close to 0.1). However, it became clear that higher wavelengths (near infrared 870 and 1020 nm) and optical parameters(ASYmmetry parameter and Linear Depolarization Ratio),that best differentiate dust from other aerosol types, presented the highest importances.



We understand that this new plot results corroborate the result presented in Figure 10 and the related discussion, which show that dust variability is central to the aerosol optical regime variability over Iberian Peninsula. However, to avoid increasing the manuscript extension since it is already long, we include the figure above in the supplement and in the manuscript we briefly refer to it and to this coherence between importance score obtained for the cluster predictors(figure above) and the relative importance of MERRA-2 aerosol types as aerosol optical regime predictor.

Line 625: All of a sudden MERRA-2 AOD field is taken as a reference. AERONET sites also provide the AOD and SSA values, which could have been checked during the period of various scenarios (Case#01, Case#02, Case#03, Case#04).
Reply: We aimed to test our prescription approach in these case studies for scenarios where AOD at 440 nm meets AERONET threshold used to perform retrievals of SSA (AOD@440 nm > 0.4). However, given that the prescription is done based on a MERRA-2 map of the combination of aerosol types column density, the only way to filter areas across IP where AOD@440 nm > 0.4 is by using the AOD field from MERRA-2. AERONET provides local AOD at specific sites. This is why we took MERRA-2 AOD as a reference, and this is the context in which the sentence at line 625 was written.
We adjusted the text to make it clearer.

Lines 673-674: 'lower computational cost' --> How is this quantified? Have you compared with any other methods of aerosol classification?
Reply: Thanks. Indeed, we should better contextualize the reference to lower computational cost here. The reduction of computation cost achieved by using prescribed aerosol optical models is well recognized and undisputed, particularly when compared to the modeling systems that simulate Mie calculations online or systems that perform 3-D simulation of optical properties based on aerosol types and their mixing state, which are computationally more expensive than columnar prescriptions.

Figure 11 Caption: MODIS Terra?
Reply: Corrected to MODIS Terra and Aqua since the images are selected from overpassing of both satellites. We adjusted the caption accordingly.

Lines 697-699: Earlier it was mentioned AERONET AOD > 0.4 but for MERRA-2 AOD > 0.3. Why?

Reply: This has to do with the difference in AOD as a function of wavelength. When describing AERONET threshold to provide sky retrievals such as SSA we were referring to AOD at 440 nm (0.4), here for MERRA-2 we are talking about AOD at 550 nm (0.3), that`s why the values are different. MERRA-2 ADP provides AOD at 550 nm. The MERRA-2 AOD at 550 nm threshold(0.3) goal is to provide and compare SSA only for region where aerosol loading is above the limited adopted for AERONET to perform SSA retrieval (AOD at 440 nm > 0.4)

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