Unsupervised Regionalization of Particle-resolved Aerosol Mixing State Indices on the Global Scale

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Abstract

The aerosol mixing state significantly affects the climate and health impacts of atmospheric aerosol particles. Simplified aerosol mixing state assumptions, common in Earth System models, can introduce errors in the prediction of these aerosol impacts. The aerosol mixing state index, a metric to quantify aerosol mixing state, is a convenient measure for quantifying these errors. Global estimates of aerosol mixing state indices have recently become available via supervised learning models, but require regionalization to ease spatiotemporal analysis. Here we developed a simple but effective unsupervised learning approach to regionalize predictions of global aerosol mixing state indices. We used the monthly average of aerosol mixing state indices global distribution as the input data. Grid cells were then clustered into regions by the k-means algorithm without explicit spatial information as input. This approach resulted in eleven regions over the globe with specific spatial aggregation patterns. Each region exhibited a unique distribution of mixing state indices and aerosol compositions, showing the effectiveness of the unsupervised regionalization approach. This study defines “aerosol mixing state zones” that could be useful for atmospheric science research.

1 Introduction and Motivation

The concept of aerosol mixing state describes how different aerosol chemical species are distributed among the particles in a population [Riemer et al., 2019]. A completely “externally mixed” population contains only one species per particle, while a completely “internally mixed” population contains particles with the same composition as the bulk population. Many different intermediate mixing states are possible between those two extremes, and these commonly occur in the ambient atmosphere. Aerosol mixing state influences the particles’ properties such as hygroscopicity [Fierce et al., 2017, Holmgren et al., 2014], optical properties [Fierce et al., 2016, Jacobson, 2001], cloud condensation nuclei activity [Ching et al., 2012, Wang et al., 2010], ice nucleation potential [Knopf et al., 2018], the aerosols’ lifetime [Koch et al., 2009] in the atmosphere, and their deposition in the human respiratory system [Ching and Kajino, 2018]. However, Earth System models (ESMs) or regional climate models usually hold simplified mixing state assumptions. These can influence how accurately physicochemical properties of aerosols are predicted [Ching and Kajino, 2018, Fierce et al., 2017], and thereby limit the accuracy of estimating aerosol impacts on climate and health.

Riemer and West [2013] proposed an entropy-based diversity metric to quantify aerosol mixing state. This metric, the mixing state index (χ), has a range from 0% (a fully externally mixed population) to 100% (a fully internally mixed population). The index χ has been applied to both modeling [Zhu et al., 2016] and experimental [Healy et al., 2014, O’Brien et al., 2015, Fraund et al., 2017, Bondy et al., 2018, Ye et al., 2018] work. For example, χ has been used for quantifying errors in CCN concentration [Ching et al., 2017] prediction, as well as the prediction of soot particles depositing in the human respiratory system [Ching and Kajino, 2018].

Knowing the global distribution of aerosol mixing state index is desirable, as it can be used for error quantification at the large scale. But deriving this distribution directly by using a benchmarking particle-resolved model [Riemer et al., 2009] at the global scale would be computationally prohibitive over the time scales of interest. To overcome this limitation, supervised-learning emulators were developed for predicting the spatial distribution aerosol mixing state across the globe [Hughes et al., 2018, Zheng et al., 2020]. However, the spatial delimitation based on aerosol mixing state indices remains unclear, which hinders further spatiotemporal analysis.

The research questions for this paper are: (1) Is it possible to regionalize the global mixing state indices, i.e., define aerosol mixing state zones, similar to distinct climate zones [Fovell and Fovell, 1993, Mitchell, 1976]? (2) What are spatiotemporal patterns of the regionalized mixing state indices? This paper describes an effort to regionalize the global mixing state indices using a simple but effective unsupervised learning approach.
2 Methods

2.1 Mixing state indices definition

The mixing state index $\chi$ [Riemer and West, 2013] is given by the affine ratio of the average particle species diversity ($D_a$) and bulk population species diversity ($D_h$) as $\chi = \frac{D_a - 1}{D_h - 1}$. The diversities $D_a$ and $D_h$ are calculated based on per-particle species mass fractions and mass fraction of particles as described in detail in [Riemer and West, 2013].

This study focused on submicron aerosols because they are the dominant category of particles for radiation interactions and the provision of cloud condensation nuclei. We defined the mixing state indices in three different ways, namely based on chemical species abundance ($\chi_a$), based on the mixing of absorbing and non-absorbing species ($\chi_o$), and based on the mixing of hygroscopic and non-hygroscopic species ($\chi_h$). The index $\chi_a$ was defined based on all the model aerosol species. A lower value for $\chi_a$ indicates that the species tend to be present in separate particles. For $\chi_o$, we considered two surrogate species, black carbon (absorbing) and all other aerosol species grouped together (assumed to be non-absorbing). Thus, a lower value for $\chi_o$ indicates the absorbing species black carbon and the sum of all other (non-absorbing) species are more externally mixed. Similarly, $\chi_h$ was also calculated from two surrogate species. We combined black carbon, dust, and primary organic matter as one surrogate species, given their comparatively lower hygroscopicities, and salt, secondary organics, and sulfate as the other surrogate species. Correspondingly, a lower value for $\chi_h$ represents the case where hygroscopic and non-hygroscopic species tended to be present in separate particles. Note that mixing state indices defined for different purposes capture different aspects of the overall mixing state, and they are uncorrelated [Zheng et al., 2020].

2.2 Data

We used the supervised-learning surrogate models developed by [Zheng et al., 2020] to predict the mixing state indices from ESM (Earth System Model) output. The surrogate models were trained on an ensemble of particle-resolved model PartMC-MOSAIC [Riemer et al., 2009; Zaveri et al., 2008], with the training labels for $\chi$ calculated directly from the particle-resolved data. The strategy to generate the training and testing data was to vary 45 input parameters for the PartMC-MOSAIC model scenarios, including primary emissions of different aerosol types, primary emissions of gas phase species, and meteorological parameters. A Latin Hypercube sampling approach was employed to sample the parameter space efficiently. The surrogate models were trained by using XGBoost (eXtreme Gradient Boosting, Chen and Guestrin [2016]), and can be expressed as:

$$\chi_S = f_S(A, G, E).$$  \hspace{1cm} (1)

Where $\chi_S$ is the mixing state index ($\chi_a$, $\chi_o$, or $\chi_h$) and $f_S$ denotes the surrogate function for the corresponding mixing state index. The variants $A$ (aerosol), $G$ (gas), and $E$ (environment) represent the features (variables) from the PartMC simulations that are used for predicting the mixing state index. These variables are also available from the ESM. A detailed definition of the features for the surrogate models is given in [Zheng et al., 2020].

We used the simulations from the Community Earth System Model Version 2 [CESM2 version 2.1.0; Danabasoglu et al., 2020] with MAM4 [Liu et al., 2016] to provide variables at the global scale. Specifically, we used the component set “FHIST” for the global simulation configuration. This component set represents a typical historical simulation using an active atmosphere and land with prescribed sea surface temperatures and sea-ice extent, and a 1 degree finite volume dycore with the forcing data available from 1979 to 2015. We ran the model for the year 2011 with 6 years (2005-2010) spinup. The simulation was conducted at a resolution of 0.9$^\circ$ latitude by 1.25$^\circ$ longitude along with emission inventories from CMIP6 emissions [Emmons et al., 2020]. We stored the instantaneous outputs every three hours in the simulation, which yielded 2920 timestamps for each surface-layer grid cell for the entire year of simulation time.

With the surrogate models and the ESM simulation, we predicted the mixing state indices for each grid cell at each timestamp (i.e., every 3 hours). For each grid cell, the mixing state indices were averaged by month. Therefore, each grid cell has a vector of stacked mixing state indices with a length of 36 (12 months $\times$ 3 mixing state types), represented as $x = (\chi_{a,1}, \ldots, \chi_{a,12}, \chi_{o,1}, \ldots, \chi_{o,12}, \chi_{h,1}, \ldots, \chi_{h,12})$. 

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2.3 Regionalization strategy

Here we interchangeably use the terms “region” and “cluster” to refer to a group of grid cells. We used the \( k \)-means unsupervised clustering algorithm to partition \( n \) grid cells into \( k \) clusters, minimizing the within-cluster variances. The \( k \)-means method has been applied to environmental sciences for ecoregion delineation [Kumar et al. 2011], environmental risk zoning of the chemical industrial area [Shi and Zeng 2014], and clustering haze trajectory of peatland fires [Khairat et al. 2017], among other applications.

In our case, given \( n = 55,296 \) (192 latitude \( \times \) 288 longitude) grid cells \((x_1, x_2, \ldots, x_{55296})\), where each grid cell contains a 36-dimensional vector (three mixing state indices over 12 months), \( k \)-means clustering aims to partition these grid cells into \( k \) \((k \leq n)\) sets \( S = \{S_1, S_2, \ldots, S_k\}\) so as to minimize the within-cluster variance. The objective is to find:

\[
\arg\min_{S} \sum_{i=1}^{k} \sum_{x \in S_i} \|x - \mu_i\|^2
\]

where \( \mu_i \) is the mean of the points in \( S_i \).

The \( k \)-means clustering method requires the a priori specification of the number of clusters \( (k) \). As the value of \( k \) increases, the variance \( V_k \) asymptotically approaches 0 until \( k \) equals \( n \). Here we used the relative difference in variance between \( k - 1 \) and \( k \) to determine the number of clusters. Starting from \( k = 2 \), we computed the criteria of relative difference \( V_{k-1} - V_k \leq 0.05V_{k-1} \) to determine the cluster number \( k \). In this study, since \( V_{10} - V_{11} \leq 0.05V_{10} \), we selected \( k = 11 \) clusters.

3 Results

The regional clusters of mixing state are shown in Figure 1. Unlike with supervised learning, the performance of unsupervised learning is challenging to evaluate since there is no ground truth. However, the clustering process of this study was merely based the information of mixing state indices, without the direct guidance such as aerosol species distribution or other explicit spatial information. Therefore a success indicator of the spatiotemporal clustering is whether spatially-contiguous regions emerge from this procedure. The clusters in Figure 1 exhibit notable regional patterns among the eleven clusters (mixing state zones), suggesting that this approach has indeed identified meaningful clusters.

Each cluster has a unique spatiotemporal distribution of three mixing state indices, and captures the spatiotemporal variations that could not be detected by the global overall averages. For example, ocean regions (e.g., clusters 8, 9, 10 in the Southern Hemisphere) tend to display a more pronounced seasonal cycle than land regions (e.g., cluster 7) and overall averages.

Clusters 1 (the Arctic) and 11 (the Antarctic) were characterized by high mixing state indices (with monthly means of at least 67%) throughout the year. The high values of all three mixing state indices means that the different aerosol species are rather internally mixed in these regions. Clusters 5 (tropical oceans), 8 (the South China Sea, Indian Ocean and the Pacific Ocean in the northern hemisphere near the equator, and the northern part of the Southern Ocean) and 9 (the southern part of the Southern Ocean) were ocean areas. Similar bulk aerosol composition was predicted in these regions, however the annual cycle of mixing state varies more for clusters 8 and 9 than for 5.

Clusters 2, 3, and 4 covered oceans at mid and high latitudes in the Northern Hemisphere, and lands including Northern and Southern America, Europe, Middle East, Southeast Asia, as well as Australia. Clusters 6 (the region across the Atlantic from Western and Northern Africa to the northeast coast of South Africa, as well as part of the Indian Ocean) and 7 (Middle, Eastern, and Southern Africa, as well as Asia) were characterized by their higher values in \( \chi_a \) compared to \( \chi_s \) and \( \chi_b \), meaning that black carbon and non-absorbing species are comparatively internally mixed.

4 Conclusions

In this paper, we developed a simple but effective unsupervised learning approach to regionalize global aerosol mixing state indices. We used the monthly averages of the spatially-varying aerosol mixing state indices as input data. Each grid cell was then assigned to a region using \( k \)-means clustering without explicit spatial information as input. This approach resulted in eleven distinct regions over the world with specific spatial aggregation patterns. Each region exhibited a unique
Figure 1: Aerosol mixing state zones based on unsupervised regionalization. Also shown are monthly average mixing state indices and annual averages of aerosol species mass fractions (bulk composition).

distribution of mixing state indices, suggesting that the unsupervised regionalization approach had identified meaningful regions. To the best of our knowledge, this is the first study to define aerosol mixing state zones, which we anticipate will be helpful for future studies of the global aerosol burden.

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