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Global aerosol mixtures and their multiyear and seasonal characteristics

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HIGHLIGHTS

- Cluster analysis of 7 years of global GOCART aerosol optical depth data.
- Global composition of ≥10 multiyear and seasonal aerosol mixture clusters.
- A taxonomy and visualization scheme for mapping aerosol mixtures.
- AERONET-derived optical and microphysical properties of aerosol mixtures.
- Spatiochemical assessment in the context of other classification studies.

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ABSTRACT

The optical and microphysical characteristics of distinct aerosol types in the atmosphere are not yet specified at the level of detail required for climate forcing studies. What is even less well known are the characteristics of mixtures of aerosol and, in particular, their precise global spatial distribution. Here, cluster analysis is applied to seven years of 3-hourly, gridded 2.5° × 2° aerosol optical depth data from the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model, one of the most-studied global simulations of aerosol type currently available, to construct a spatial partition of the globe into a finite number of aerosol mixtures. The optimal number of aerosol mixtures is obtained with a k-means algorithm with smart seeding in conjunction with a stopping condition based on applying the 'law of diminishing returns' to the norm of the Euclidean distance to provide upper and lower bounds on the number of clusters. Each cluster has a distinct composition in terms of the proportion of biomass burning, sulfate, dust and marine (sea salt) aerosol and this leads rather naturally to a taxonomy for labeling aerosol mixtures. In addition, the assignment of primary colors to constituent aerosol types enables true color-mixing and the production of easy-to-interpret maps of their distribution. The mean multiyear global partition as well as partitions deduced on the seasonal timescale are used to extract aerosol robotic network (AERONET) Level 2.0 Version 2 inversion products in each cluster for estimating the values of key optical and microphysical parameters to help characterize aerosol mixtures. On the multiyear timescale, the globe can be spatially partitioned into 10 distinct aerosol mixtures, with only marginally more variability on the seasonal timescale. In the context of the observational constraints and uncertainties associated with AERONET retrievals, bivariate analysis suggests that mixtures dominated by dust and marine aerosol can be detected with reference to their single scattering albedo and Angstrom exponent at visible wavelengths in conjunction with their fine mode fraction and sphericity. Existing multivariate approaches at classification appear to be more ambiguous. The approach presented here provides gridded (1° × 1°) mean compositions of aeorosol mixtures as well as tentative estimates of mean aerosol optical and microphysical parameters in planetary regions where AERONET sites do not yet exist. Spreadsheets of gridded cluster indices for multiyear and seasonal partitions are provided to

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facililate further study of the global distribution of aerosol mixtures and possibly for the selection of new AERONET site locations.

1. Introduction

It is now known that aerosols contribute strongly to the change in radiative climate forcing and prediction uncertainty (Mishchenko et al., 2007a,b,c; Kahn, 2012; IPCC, 2013). However, much is still not known about the optical and physical properties of the aerosol components. For example, there is no suitable optical model for radiance retrievals of coarse-mode dust, even from the Sahara (e.g., Kalashnikova et al., 2005), and the time-varying, spectral single-scattering albedo of biomass burning particles is much too loosely constrained for many radiative forcing applications. Reduction of the aerosol uncertainty has been hindered by several things. Firstly, there is a gap in our detailed understanding of the global spatial and temporal variation of aerosol types (Hansen 2005), and secondly there is an absence of consensus on the optimal combination of optical and microphysical parameters that should be used for characterizing aerosol types (IPCC, 2013). Thirdly, satellite remote sensing data, while able to provide global spatiotemporal detail on aerosol amount and to some extent type, by itself it is insufficient for providing detailed aerosol microphysics.

Targeted suborbital measurements are expected to help to better contrain the uncertainty associated with direct aerosol radiative forcing (Kahn, 2012). Results from global circulation models (e.g. Mann et al., 2014) are helping to paint a picture of how aerosol is globally distributed (Stier et al., 2013; Kinne et al., 2013) and new studies of global decadal and/or multi-decadal trends in aerosol load are helping constrain its behavior in time (Zhang and Reid, 2010; Chin et al., 2014). It should be borne in mind that the convergence of results between IPCC/AR4 and AR5 in model intercomparison exercises suggests a reduction in model diversity but not necessarily in model uncertainty, due to the limitations of available observational constraints (Samset et al., 2014; Schwartz et al., 2014). Of note is the fact that there is a rapid growth in understanding 3D aerosol climatologies thanks to vertical profiles provided by LIDAR (e.g. Winker et al., 2013; Yu et al., 2010). These are likely to play a key role in comparative studies involving targeted suborbital measurements. With regard to the characterization of aerosol types, numerous studies have attempted to classify aerosol types from ground-based observations (e.g. Dubovik et al., 2002; Eck et al., 1999; Kalapureddy et al., 2009; Lee et al., 2010; Mielonen et al., 2009; Omar et al., 2005, 2009), and many satellite algorithms now adopt procedures for classifying aerosol types to improve the accuracy of their AOD retrieval (Higurashi and Nakajima, 2002; Jeong and Li, 2005; Kahn et al., 2010; Kaufman et al., 2005; Kim et al., 2007, 2013; Lee et al., 2009; Remer et al., 2005, 2008; Russell et al., 2014). Despite such studies at the local or regional level using bivariate or multivariate combinations of optical and/or microphysical parameters as proxies, there is no unambiguous protocol for the identifying aerosols of different types or mixtures. The latter is of particular importance and of increasing prominence due to cross-boundary and/or intercontinental transport of aerosol (e.g. Yu et al., 2013).

A proper characterization of aerosol requires knowledge of aerosol size, shape, and composition provided by optical and microphysical parameters (Dubovik and King, 2000). Inversion of radiation measurements made by ground-based CIMEL sunphotometers (typically several per day) operated at sites contributing to AERONET (Holben et al., 1998) provide size, index-of-refraction, and shape information, but sites in this point-sampling network are unevently distributed – particularly over ocean and uninhabited regions (e.g. deserts, ice caps oceans, and mountains). It should also be noted here that AERONET particle property retrievals are not as well-constrained as direct-sun spectral AOD measurements, and also that some of the assumptions in the sky scan retrieval are not physical (e.g. assuming that the indices of refraction are the same for both fine and coarse modes). The highest level of chemical detail is provided by surface or low altitude *in situ* measurements but laboratories and supersites tend to be located at urban centers and sample the atmosphere at altitudes well below the cloud layer (IPCC, 2013). Furthermore, there is a need for more studies comparing mass size distributions with columnar volume size distributions (e.g. Gerasopoulos et al., 2007) to test such ascertions, especially considering that AERONET aerosol type has only been validated against *in situ* data in a few cases. From space, radiation measurements from spectroradiometer instruments onboard satellites (typically $\approx$ 1 per day) provide full-Earth viewing capacity but discerning aerosol types is still not unambiguous. Nevertheless, progress is being made in this direction. For example, inversion algorithms that retrieve aerosol properties from spectral multi-angle polarimetric satellite observations suggest that polarization information is expected to help with regard to determination of particle size and the real part of the refractive index with reference to volume size distributions (Dubovik et al., 2011), and some constraints on size, shape and also the SSA have been shown to be derivable from multi-angle, multispectral observations with MISR (Kalashnikova and Kahn, 2006; Chen et al., 2008; Kahn et al., 2010).

Given the current limitations in coverage and detail associated with using AERONET and satellite retrievals of aerosol type data, this paper presents a method for identifying, naming and visualizing global mixtures of aerosols from the output of global circulation or chemical transport models. The motivation for this is the work of Kahn et al. (2001) which analyzed the sensitivity of multi-angle imaging to AOD and aerosol type, based on global-scale model simulations of these quantities drawn from several sources. They produced a global climatology of likely aerosol mixtures by month from the model results, and showed the degree to which multiangle imaging is sensitive to natural mixtures of aerosols, particularly over the ocean. Here, we develop a methodology for spatially partitioning the globe into zones of aerosol mixtures of distinct characteristics. The partitioning is driven by the contribution of distinct tropospheric aerosol types to the AOD in each pixel of the global grid ($2.5^\circ \times 2^\circ$ longitude $\times$ latitude) provided by the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model (Chin et al., 2000, 2002; 2014; Ginoux et al., 2001). The GOCART model provides global, continuous, gridded 3-hourly values of total AOD at 500 nm as well as the contribution to the total AOD of sulfate (SU), black carbon (BC), organic carbon (OC), desert (mineral) dust (DU) and sea salt (SS). With reference to the values of optical and microphysical parameters provided by AERONET sites in each cluster, we then assess the characteristics of the global partitioning obtained for a multiyear mean and for seasonal means of the GOCART chemical simulations in the context of