A CLIMATOLOGY OF GLOBAL AEROSOL MIXTURES TO SUPPORT SENTINEL-5P AND EARTHCARE MISSION APPLICATIONS

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ABSTRACT

Since constraining aerosol type with satellite remote sensing continues to be a challenge, we present a newly derived global climatology of aerosol mixtures to support atmospheric composition studies that are planned for Sentinel-5P and EarthCARE.

The global climatology is obtained via application of iterative cluster analysis to gridded global decadal and seasonal mean values of the aerosol optical depth (AOD) of sulfate, biomass burning, mineral dust and marine aerosol as a proportion of the total AOD at 500nm output from the Goddard Chemistry Aerosol Radiation and Transport (GOCART). For both the decadal and seasonal means, the number of aerosol mixtures (clusters) identified is ≈10. Analysis of the percentage contribution of the component aerosol types to each mixture allowed development of a straightforward naming convention and taxonomy, and assignment of primary colours for the generation of true colour-mixing and easy-to-interpret maps of the spatial distribution of clusters across the global grid. To further help characterize the mixtures, aerosol robotic network (AERONET) Level 2.0 Version 2 inversion products were extracted from each cluster's spatial domain and used to estimate climatological values of key optical and microphysical parameters.

The aerosol type climatology represents current knowledge that would be enhanced, possibly corrected, and refined by high temporal and spectral resolution, cloud-free observations produced by Sentinel-5P and EarthCARE instruments. The global decadal mean and seasonal gridded partitions comprise a preliminary reference framework and global climatology that can help inform the choice of components and mixtures in aerosol retrieval algorithms used by instruments such as TROPOMI and ATLID, and to test retrieval results.

1. INTRODUCTION

Retrieval algorithms used by instruments such as TROPOMI and ATLID rely on knowledge of source regions to characterize aerosol types. Results from global circulation models [1] help paint a picture of how aerosols are globally distributed [2, 3] and studies of decadal trends in aerosol load are helping to constrain their temporal behaviour [4].

A proper characterization of aerosol requires knowledge of aerosol size, shape, and composition provided by optical and microphysical parameters [5]. However, there is still a lack of consensus on the optimal combination of such parameters that should be used as proxies to characterize aerosol types [6]. An additional and important challenge is to improve the way that satellite algorithms address aerosol *mixtures*. This is of increasing prominence due to cross-boundary and/or inter-continental transport of aerosol [7].

This paper presents a new methodology that provides a decadal and seasonal global partition for identifying, naming and visualizing mixtures of aerosols directly from the output of global circulation or chemical transport models. The motivation for this is the work of [8] which analyzed the sensitivity of multi-angle imaging to AOD and natural mixtures of aerosols, particularly over the ocean. Here we present global decadal and seasonal partitions.

2. METHODOLOGY

The partitioning is driven by the percentage contribution of distinct tropospheric aerosol types to the total AOD in each pixel of the global grid (2.5 x 2 degree longitude x latitude) provided by the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model [4, 9, 10, 11]. The GOCART model provides global, continuous, gridded 3-hourly values of total AOD at 500nm 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). We also calculated a "biomass burning" (BB) component from the sum of the BC and OC components.

The spatial partition and global climatology was obtained by applying cluster analysis (with a k-means algorithm and smart seeding with 10 random starting points) to the global mean percentage contribution of BB, SU, DU and SS components. Clustering was performed separately on: i) the global

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decadal mean and ii) the global seasonal means for the winter (DJF), spring (MAM), summer (JJA) and autumn (SON) seasons calculated from 7 years of GOCART simulated outputs for the period 2000-2006 (inclusive). **Fig. 1** shows the global seasonal and 7-year aggregate total AOD.

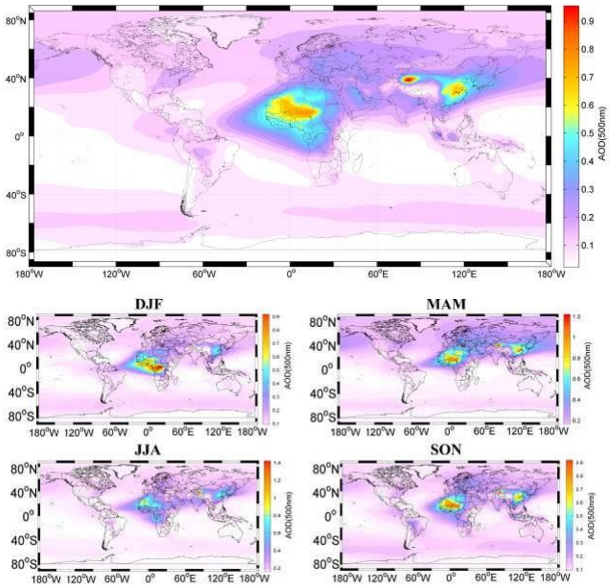

Figure 1. (Upper Panel) the global decadal mean total AOD at 500nm output from the GOCART model (Lower Panels) the global seasonal mean total AOD.

The global total AOD shows that the regions of peak aerosol load for both the decadal and seasonal means are broadly located over the Sahara and Nigeria in Northern Africa, in the Gobi desert to the north of the Himalayas, and over a large region centered on Beijing with relatively minor spatial variation. In the case of global seasonal means, the mean total AOD exhibits only a mild variation in the location of peak aerosol load because large inter-annual variations in dust and biomass burning are averaged-out on the decadal timescale. **Fig. 2** shows the global decadal mean percentage contribution of BC, OC, BB, SU, DU and SS.

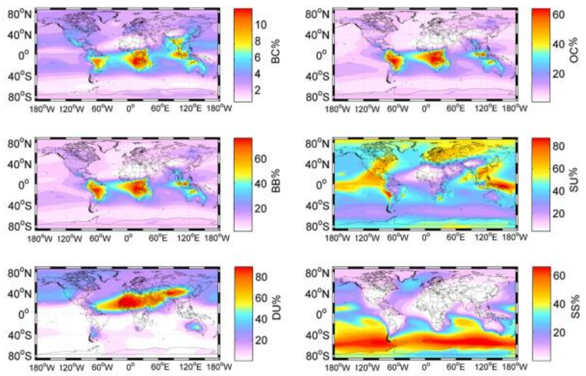

Figure 2. The global 7-year mean % contribution of BC, OC, BB, SU, DU & SS to the total AOD.

With the value of BC being < 10% over much of the globe, we proceeded to work with 4-species data: BB, SU, DU and SS. In terms of anthropogenic pollution particles, a part of their contribution to the total AOD is included in BB and the remaining portion is included in SU.

When performing cluster analysis, a stopping condition (that cluster centres do not change by more than 10%) was used to ensure convergence and repeatability, and led to identification of 10 clusters for the case of the global 7-year mean and 10-11 clusters for the global seasonal means. **Fig. 3** presents the percentage composition of the aerosol mixtures obtained by clustering the global 7-year and seasonal mean using this condition.

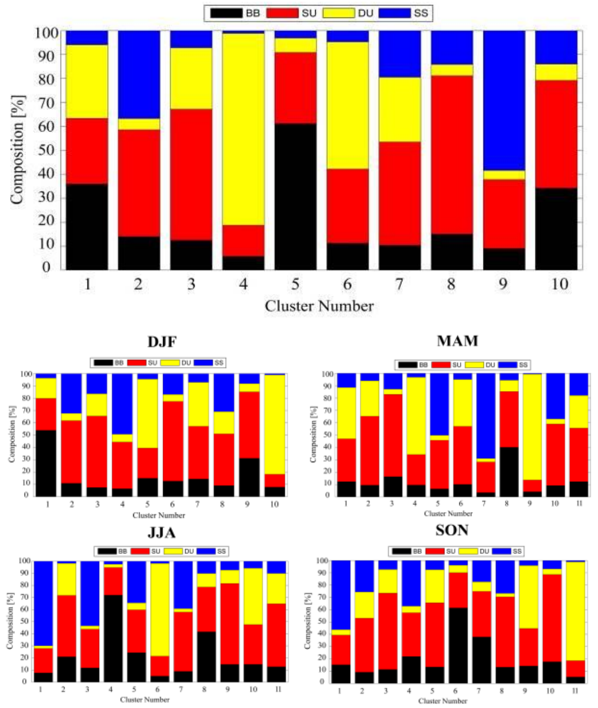

Figure 3. (Top Panel) the composition of each of the 10 mixtures that result from clustering the 4-species GOCART data (Lower Panels) the composition of the 10-11 mixtures resulting from seasonal clustering of the 4-species data.

Although the GOCART model has uncertainties associated with it, the 10% stopping condition used to cluster the data should therefore be considered a lower uncertainty bound. Having performed the cluster analysis, we then developed a naming convention and taxonomy for clusters using an ‘ordered adjective-object’ format whereby aerosol types are listed in increasing order of their percentage contribution. **Table 1** presents the taxonomy for the clusters in **Fig. 3** obtained for the global 7-year mean of the 4-species GOCART data with the caveat that “marine” refers to SS and “smoke” refers to BB aerosol.

Table 1. The taxonomy of the 10 clusters obtained from the global decadal mean.

Cluster 1	Sulfurous Dusty **SMOKE**
Cluster 2	Marine **SULFATE**
Cluster 3	Dusty **SULFATE**
Cluster 4	**DUST**
Cluster 5	Sulfurous **SMOKE**
Cluster 6	Sulfurous **DUST**
Cluster 7	Marine Dusty **SULFATE**
Cluster 8	**SULFATE**
Cluster 9	Sulfurous **MARINE**
Cluster 10	Smokey **SULFATE**

We then assigned primary colours (black, red, yellow and blue) to the 4-aerosol types (BB, SU, DU and SS respectively) to allow for true colour-mixing in direct proportion to their percentage composition. The result was the generation of visually and conceptually easy-to-interpret maps. **Fig. 4** presents the global spatial partitions for the decadal mean and the seasonal means.

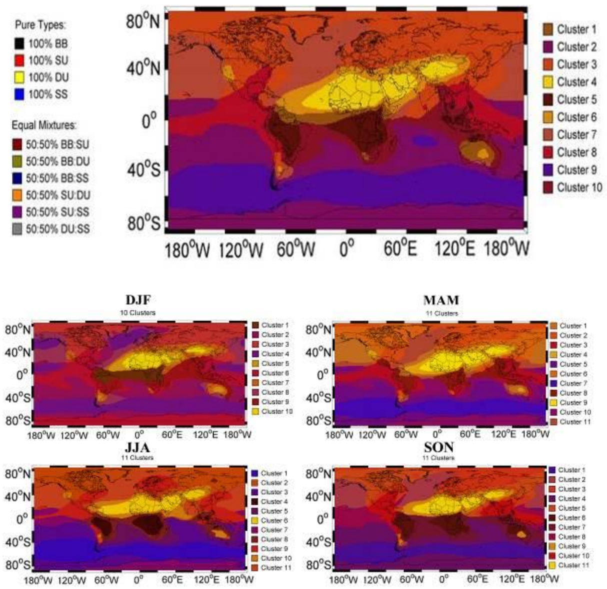

Figure 4. (Upper Panel) the spatial distribution of aerosol mixtures that result from clustering the global decadal mean (Lower Panels) the spatial distribution of mixtures that result from clustering the seasonal means.

3. CLIMATOLOGICAL MEANS

To further help characterize the mixtures, aerosol robotic network (AERONET) Level 2.0 Version 2 inversion products were extracted from sites located within each cluster and used to estimate mean climatological values of optical and microphysical parameters. **Fig. 5** shows the sites contributing full inversion records for the dust-dominated clusters 4 and 6.

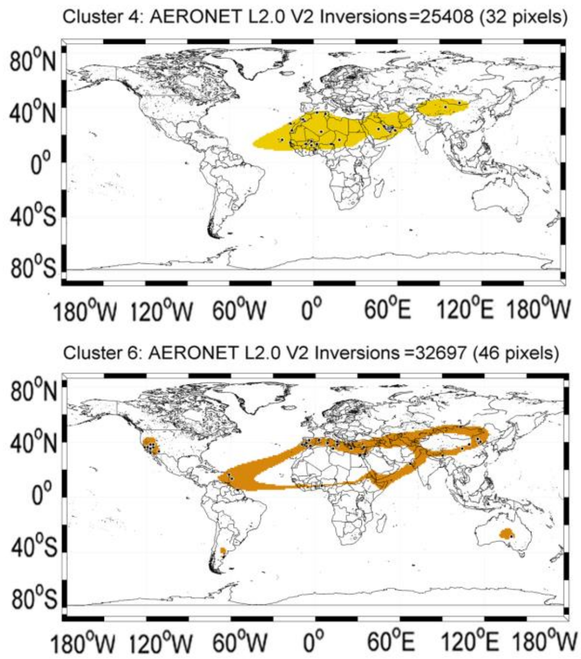

Figure 5. The spatial distribution of aerosol mixtures associated with the dust–dominated clusters 4 and 6.

The AERONET data extracted for each cluster includes key optical parameters: the spectral dependence at 440, 675, 870 and 1020nm of the asymmetry factor (ASYM), the absorption aerosol optical depth (AAOD), the single scattering albedo (SSA), the lidar ratio (LR) estimated from the SSA and the phase function at 180^o [12] and the real and imaginary parts of the complex refractive index (CRI-R and CRI-I respectively). In addition, microphysical parameters were also extracted from the clustered AERONET data record including: the fine fraction (η), the percentage of spherical particles in the retrieval ('% Sphericity'), and the volume size distribution (dV/dlnr) spanning the radial range 0.05 to 15μm shown in **Fig. 6**.

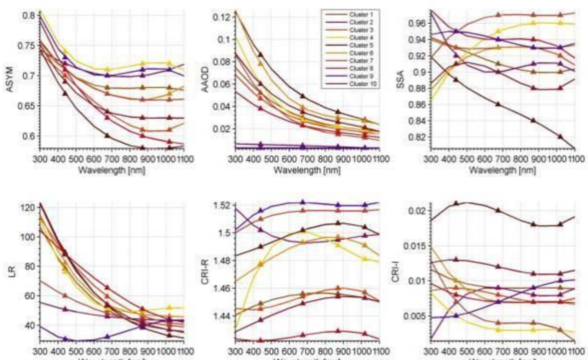

Figure 6. Spectral behaviour of the global mean values of key optical and microphysical parameters for each cluster

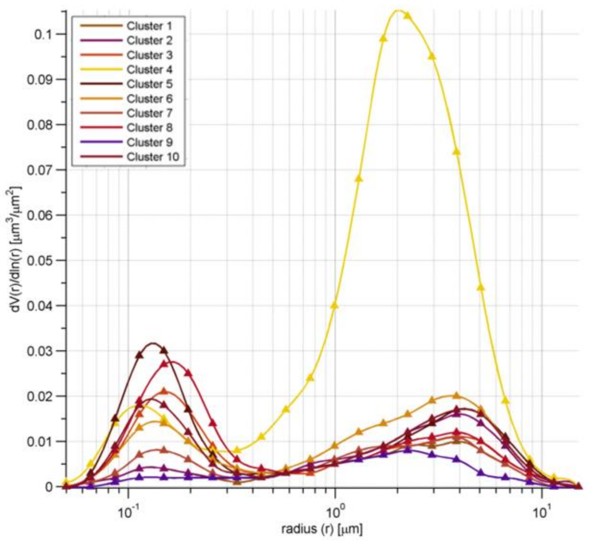

Figure 6. (continued) the global mean size distribution for each cluster.

In the context of the observational constraints and uncertainties associated with AERONET retrievals [13], bivariate analysis was also performed and suggested that mixtures can be detected with reference to their fine mode fraction (η) and % Sphericity as shown in **Fig. 7**.

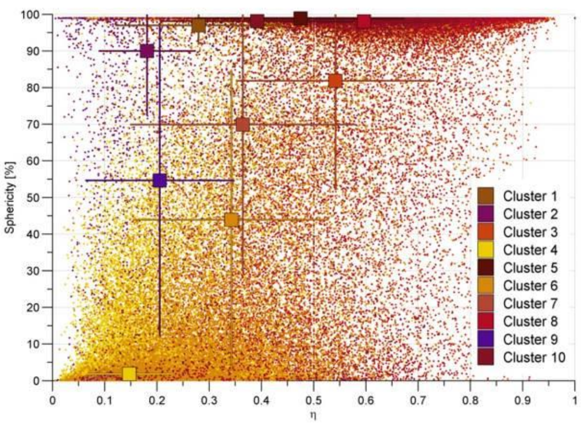

Figure 7. The location of cluster centres overlaid on AERONET inversion products for the parameter pair {% Sphericity, η}. Error bars extend out to 1 standard deviation on both parameters and all wavelengths are given in nm.

This choice of parameters was found to separate clusters visually in 2D and is line with a recent assessment of global aerosol type retrieved from MISR [14] which also found that the distinctions between spherical vs. non-spherical and fine vs. coarse mode are generally the most robust. Note also that the error bars are large and overlap in most cases, but should also be seen in the context of the density of points. The % Sphericity parameter should be considered an indication of dust only if the associated Angstrom exponent (AE) < 1.0, i.e. larger particles (Tom Eck, private communication). This may be a significant limitation because the transported dust can be strongly dominated by small particles having AE > 1.0.

4. CONCLUSION

The gridded global decadal mean (and also seasonal mean) partitions of AOD and compositional aerosol mixtures comprise a climatology that can be refined by high temporal and spectral resolution, cloud-free observations produced by Sentinel-5P and EarthCARE instruments. This preliminary reference framework can:

1. enable tests of the effect on look-up table derived retrievals of initializing retrieval algorithms used by OMI/TROPOMI or CALIOP/ATLID with aerosol type mixtures,
2. help fine-tune aerosol type selection methods used in existing algorithms by referring to mean and seasonal optical and microphysical properties of mixtures,
3. allow comparison of retrieved aerosol types with those expected from the climatology, and,
4. contribute to the assessment of region and season-specific aerosol type assumptions.

To help facilitate the uptake and further investigation and refinement of these results, tables of gridded global cluster indices for the decadal and seasonal means are available at: http://apcg.meteo.noa.gr/AEROMAP.

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