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from MISR and
chemistry models

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Climatology of the aerosol optical depth by components from the Multiangle Imaging SpectroRadiometer (MISR) and a high-resolution chemistry transport model

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and clouds. For example, Li et al. (2008) showed that the recent positive trend in wintertime precipitation over the North Pacific is related to increased aerosol emissions. Zhang et al. (2007) and Wang et al. (2014) suggested that increased anthropogenic aerosol emissions in Asia can strengthen cyclones along the Pacific storm track. Both direct and indirect radiative forcing of aerosols are expected to be more important under a changing climate. For example, Ganor et al. (2010) and Lu et al. (2010) reported increasing dust aerosols in Africa and sulfate aerosols in China, respectively. However, global climate models (GCMs) have a hard time producing consistent radiative forcing responses to varying concentrations of aerosols (IPCC, 2013). In fact, indirect radiative forcing due to aerosols is one of the dominant sources of uncertainty in the energy budget of many GCMs (Regayre et al., 2014).

Despite substantial improvements in the representation of physical and chemical processes related to aerosols in global-scale chemistry models relative to most GCMs, recent work has shown that chemistry models still exhibit considerable biases and uncertainties in aerosol concentrations and related radiative forcings (Lee et al., 2013; Shindell et al., 2013). Therefore, validating simulated aerosols in chemistry models is critical in order to better understand the root causes of these biases and uncertainties.

Aerosol–radiation interactions are determined by the size distribution of aerosols, as well as their shape and light-absorption properties (Boucher et al., 2013). Understanding the optical and microphysical characteristics of natural and anthropogenic aerosols is critical for advancing the ability of CCMs to correctly simulate the climate impact of aerosols. Nevertheless, many previous studies evaluating simulated aerosol optical depth (AOD) in models against satellite observations, such as those available from the Moderate Resolution Imaging Spectroradiometer (MODIS) and the Multi-angle Imaging SpectroRadiometer (MISR), have used only the total column AOD without taking into account aerosol type information (e.g., Tilmes et al., 2015). Shindell et al. (2013) compared AODs for each aerosol type simulated in nine chemistry climate models (CCMs). Not surprisingly, the difference in component AODs among models is much greater than the difference in total AODs (see Fig. 3 in Shindell et al., 2013). To un-

derstand the diverse partitioning of AOD among dust, sea salt, sulfate, nitrate, black carbon, and organic carbon in CCMs, it is important to compare the simulated component AODs against global climatological maps of observed AOD by components, if possible.

Unfortunately, the retrieval of AOD by type from satellite observations and using the retrieved AOD for chemistry model evaluation have been, and remain, a significant challenge. Aerosol polarization measurement by the POLDER3 (POLARization and Directionality of the Earth Reflectance) instrument onboard the PARASOL (Polarization and Anisotropy of Reflectances for Atmospheric Sciences coupled with Observations from a Lidar) enables classifying observed aerosols into various types (Russell et al., 2014). CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation) also provides aerosol classification according to surface type (Omar et al., 2009). Higurashi and Nakajima (2002) suggested detecting a dominant aerosol type using radiances from four channels on the Sea-viewing Wide Field-of-view Sensor (SeaWiFS). Kim et al. (2007) used both MODIS observations and data from the Ozone Monitoring Instrument (OMI) to classify retrieved aerosol types, but the algorithm was similarly limited to providing a single, dominant aerosol type. In the AOD climatology by type reconstructed in Nabat et al. (2013), total AOD is from a satellite instrument, but the AOD by type was derived solely from the fractions of the five aerosol types (sulfate, black carbon, organic carbon, dust, and sea salt) simulated in two CCMs. Holzer-Popp et al. (2008) provided an overview of currently available aerosol type datasets from satellites, and used the the Advanced Along Track Scanning Radiometer (AATSR) and the Scanning Imaging Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY) onboard the European Environmental Satellite (ENVISAT) to produce total column AOD and speciation by aerosol mixtures. In their study, total column AOD and surface reflectivity were derived from AATSR observations and these variables were used to simulate spectra for pre-defined aerosol mixtures, which were selected by comparison with the observed SCIAMACHY spectra.

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Due to its unique multiangle viewing approach, the MISR instrument on NASA's Terra satellite is capable of distinguishing mixtures of aerosol types without relying on data from other instruments (Diner et al., 2005a). MISR measures radiation in four spectral bands (blue – 446 nm, green – 558 nm, red – 672 nm, and near infrared – 866 nm) from nine different viewing directions (± 70.5 , ± 60.0 , ± 45.6 , ± 26.1 , and 0.0° along the direction of satellite motion), allowing retrievals of aerosol particle size and shape (Kahn et al., 2001; Diner et al., 2005b). The operational (Version 22) aerosol retrieval algorithm is based on matching observed top of atmosphere (TOA) radiances to radiances modeled for AODs ranging from 0.0 to 3.0 from 74 “mixtures” based on eight “pure” particle types, each one representing a single size distribution with specific optical properties (Kahn et al., 2010). Within the algorithm, a mixture is acceptable, or “passing,” if the difference between the observed and modeled radiances are less than a pre-determined value. In the Level 2 (swath) product, the mean of the passing mixtures is reported in the field “RegBestEstimateSpectralOptDepth” at a spatial resolution of $17.6 \text{ km} \times 17.6 \text{ km}$. The best-fitting mixture out of the 74 candidates is reported as the “RegLowestResidMixture,” but the AOD and information about which mixtures were considered “passing” are retained in the fields “OptDepthPerMixture” and “AerRetrSuccFlagPerMixture,” respectively, for further evaluation. Validation of the Level 2 Version 22 product against ground-based observations from the Aerosol Robotic Network (AERONET) (Holben et al., 1998), including assessments of particle type retrievals, can be found in Kahn et al. (2010) and Kahn and Gaitley (2015). The mixture type information in the MISR Version 22 aerosol product has been exploited by Li et al. (2013, 2015), for example, who used CCM information to select from the complete set of “passing” mixtures to improve agreement with AERONET in the continental United States.

The main objective of the current study is to compare multi-year MISR AOD by component climatologies with simulated AODs, broken down by aerosol type. The results of this work highlight the added value of using AOD by components from MISR in evaluating chemistry transport models (CTMs) and CCMs. Due to the relatively short

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lifetime of aerosols compared to trace gases, it is also important to consider the spatial inhomogeneity of aerosol distributions and resulting regional effects. Here we focus, in particular, on characterizing AOD distributions in the regions near major aerosol emission sources: East Asia, the Sahara Desert, and West Africa with comparisons between MISR climatological observations and high-resolution simulations from the the SPectral RadlatioN-TrAnSport (SPRINTARS) model for Aerosol Species (Takemura et al., 2002, 2005) interactively coupled to the Nonhydrostatic Icosahedral Atmospheric Model (NICAM) (Sato et al., 2008, 2014; Suzuki et al., 2008). We also demonstrate why it is important to consider spatio-temporal distributions of AOD when comparing satellite observations and models rather than simply using spatially and temporally averaged AODs for model evaluation.

The remainder of the paper is structured as follows. The data and model used in this study are described in Sect. 2. Comparisons between MISR retrievals and model simulations are presented in Sect. 3, followed by a summary of key findings in Sect. 4.

2 Data

2.1 MISR Level 3 Joint Aerosol product

As more observations become available for model intercomparison projects, such as AeroCom (Schulz et al., 2006; Myhre et al., 2013), the Coupled Model Intercomparison Project Phase 5 (CMIP5) (Taylor et al., 2012), and the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) (Lamarque et al., 2013), it is important that these data become more accessible (Teixeira et al., 2014). Getting the mixture information from the MISR Level 2 aerosol product is an indirect procedure, requiring access to Hierarchical Data Format (HDF) Vertex Data (VDATA) fields that map integer-valued mixture identifiers in the Level 2 files to the MISR “pure” particle types and corresponding mixing proportions. Each “pure” particle is assumed to have uniform composition and a specified size distribution. To make this information more

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of absorbing AOD plotted against non-absorbing AOD over East Asia for the month of April. One-dimensional sampling distributions are shown as the histogram on the top of the figure for non-absorbing particles and to the right of the figure for absorbing particles. The scatterplot itself shows how these two particle types covary with one another in this region for this time period. From the histograms, the MISR V22 product identifies non-absorbing particles as the dominant aerosol component, which tend to span the range from about 0.025 to 0.15, whereas absorbing aerosols are retrieved only for AOD values less than about 0.02, where aerosol-type retrieval sensitivity is very low (Kahn et al., 2010; Kahn and Gaitley, 2015). Because of this, there is very little covariance between the absorbing and non-absorbing AOD in this case.

From the information used to construct the marginal histograms it is simple to calculate the moments (mean, variance, and skewness) of the AOD distributions for different aerosol types. The k th central moment of the distribution (M_k) with a sample size, N , is conventionally defined as follows:

$$\text{first moment (mean)} = \bar{x} = \frac{1}{N} \sum_{j=1}^N x_j \quad (1)$$

$$k\text{th central moment} = M_k = \frac{1}{N} \sum_{j=1}^N (x_j - \bar{x})^k \quad (2)$$

where x_j corresponds to the j th AOD. Using the above definitions, the skewness of the distribution can be represented as:

$$\text{skew} = N \frac{M_3}{M_2^{1.5}}. \quad (3)$$

If the data follow a normal (Gaussian) distribution, the skewness of the data should be close to zero. If a distribution has positive skewness, the tail representing values larger than the median of the distribution is longer than the tail representing smaller values.

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Conversely, if the distribution has a negative skewness, the tail representing smaller values is larger. If the skewness of the distribution is not close to zero, the mean and standard deviation are not enough to appropriately represent the distribution. However, for a normal distribution the standard deviation of the sample skewness is approximately $\sqrt{15/N}$, where N is the sample size. Skewness values less than a few times as large as this (e.g., $|\text{skew}| < 3$ for a sample size of 15) should be viewed with suspicion. On the other hand, when the distribution of data is highly skewed, this indicates that it is necessary to analyze individual values or at least a summary histogram of the data in order to understand how the data are actually distributed. As we will show, the ability to easily determine the moments and distributions of the MISR AODs by component is an important feature of the JOINT_AS product.

2.2 The SPRINTARS model

The SPRINTARS model coupled to high-resolution NICAM model reports AOD every three hours for four different types of aerosols (carbonaceous, dust, sea salt, and sulfate) with a horizontal resolution of 7 km globally. As described by Suzuki et al. (2008), the SPRINTARS CTM reasonably reproduced global distributions of total AOD in comparison with MODIS near major aerosol emission sources. SPRINTARS is also one of the models included in the AeroCom intercomparison (Huneeus et al., 2011). Although due to computational limitations the SPRINTARS simulation period used in this study covers only eight days from 1 July through 8 July 2006, the high spatial and temporal resolution of the model output allow us to compare simulated AOD distributions with those from MISR under the assumption that the AOD distribution does not change significantly from one year to the next during the month of July (“the stationarity assumption”). We adopt this approach because we found that the JOINT_AS product for the single month of July 2006 contained a significant number of missing values even at $5^\circ \times 5^\circ$ spatial resolution. The missing data are likely due to cloud screening and locations being flagged as inappropriate for aerosol retrievals, as discussed in Kahn

et al. (2009). Comparison of long-term CTM simulations driven by reanalysis datasets with MISR component AODs may provide insight into the interannual variability of AOD type distributions. Specifically, the variability among the Julys from MISR might indicate how representative one July from SPRINTARS might be. However, this investigation is beyond the scope of the current work.

It is important to note that the aerosol types in SPRINTARS are different from the “pure” particle distributions used in the operational MISR aerosol retrievals. In order to make the MISR AOD by components comparable with those of the model, MISR aerosol types were combined in the manner shown in Table 1. AODs from weakly and strongly absorbing aerosols in the MISR dataset, with single scattering albedo (SSA) in the mid-visible of 0.9 and 0.8, respectively, were combined to construct an analog to modeled carbonaceous aerosols. The combination of the more absorbing and less absorbing aerosol particles is intended to represent the range of such particles in nature (e.g., Liu et al., 2014). Given the limitation of MISR to represent the modeled dust, the AODs from the non-spherical grains and coarse spheroids from MISR (Kalashnikova et al., 2005) were combined. Finally, AODs from the three (small) non-absorbing particles were added to compare to the modeled sulfate and nitrate aerosols. The largest MISR particle type, with an effective radius of 2.8 μm , was excluded when calculating the non-absorbing AOD due to issues with retrieval sensitivity to this component (Kahn et al., 2010).

3 Results

3.1 East Asia

Rapid increases in emissions of aerosols and their precursors in East Asia have caused growing concern because of the broad impact of Asian aerosols on the North Pacific and mainland North America, especially the United States (Yu et al., 2008). Figure 2 compares climatological AODs from non-absorbing aerosols for July retrieved

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by MISR in East Asia with the sulfate AOD predicted by SPRINTARS for the beginning of July 2006. Panels a and b of Fig. 2 are the respective MISR non-absorbing and SPRINTARS sulfate AOD maps displayed using the same color scale. Peak AOD values correspond to source regions in the Shandong Province south of Beijing and are closely related to the emissions in this heavily industrial region (Streets et al., 2007). The spatial gradient in AOD is due to transport and deposition processes. In spite of the differences in time period, these figures show very good qualitative agreement in their representation of the spatial distribution of non-absorbing/sulfate AOD. It is worth noting that both MISR and SPRINTARS capture a secondary peak in AOD in southwestern China in Sichuan Province, which burns coal high in sulfur (Streets and Waldhoff, 2000), with peak loading seen in April, July, and October (Wang et al., 2010). The reason for this good agreement is likely that the industrial source regions are well considered in the emission database used for SPRINTARS.

Figure 2c and d shows the probability mass function (PMF) of the non-absorbing AOD from MISR and the sulfate AOD from SPRINTARS, respectively, corresponding to the shaded boxes on the maps in Fig. 2a and b. The PMF is simply the histogram scaled so that the total area of the bars corresponds to some fixed value. The boxes capture the region of highest AOD loading. It is important to recognize that Fig. 2c from MISR includes both temporal and spatial variability because it is averaged over a 15 year time period, whereas Fig. 2d from SPRINTARS represents primarily spatial variability. Even so, the histograms have similar shapes, with only a 13% disagreement in the mean non-absorbing/sulfate AOD (0.45 for MISR and 0.39 for SPRINTARS) and similar standard deviations (0.39 for MISR and 0.35 for SPRINTARS). What is noticeable about the shapes of the distributions is that they are non-Gaussian, with long positive tails. The ability to visualize the full AOD distribution is an important analysis technique enabled by the MISR JOINT_AS product. The skewness of the MISR distribution is 1.56 and for SPRINTARS the skewness is 2.47, showing that the model has more positive skewness corresponding to more high AOD values. This is consistent with Fig. 2b, which

shows values in the box tend to be purple ($AOD > 0.5$) for SPRINTARS with somewhat lower values for the boxed region in the MISR climatology in Fig. 2a.

Levy et al. (2009) describe how the monthly mean AOD reported by satellite instruments is highly dependent on the averaging method selected. Reliance on the mean and standard deviation alone are particularly problematic when high outliers are common. A satellite instrument can miss extreme events due to its sampling characteristics (Colarco et al., 2014), but these events may be captured by a model with better temporal and complete spatial sampling. If this is the case, the mean AOD values could be very different, but, as shown here, the distributions are still similar.

It is worth noting that taking the base-10 logarithm of the non-absorbing/sulfate AODs in Fig. 2c and d results in distributions with skewnesses of -0.25 (MISR) and -0.26 (SPRINTARS). These skewness values are very close to zero, indicating that these modified distributions are very nearly log-normal, as is typical for atmospheric pollutant concentrations (Ott, 1990). The results in Fig. 2 as a whole indicate that combining MISR non-absorbing AOD in the manner described appears to be a good proxy for sulfate aerosol loading, at least in East Asia.

When comparing satellite-derived total column AOD with modeled AOD, it is common practice to assume that a single, dominant type of aerosol accounts for the majority of the modeled AOD in a region (e.g., Kim et al., 2007). However, this approach will not work if the dominant pollutant type varies over time (Wang et al., 2010). Another useful characteristic of the MISR operational aerosol retrieval is that the mixture climatology is applied globally to all locations and seasons, unlike MODIS over land, which relies on aerosol models that change as a function of location and season based on the AERONET climatology (Levy et al., 2013). A recent study by Eck et al. (2013), for example, showed that the seasonality in biomass-burning SSA in southern Africa is better captured by MISR than MODIS due to these different approaches. Figure 3 shows the monthly partitioning of aerosol type from MISR for April (top row) and July (bottom row) based on the 15 year climatology from the JOINT_AS product. This is possible because

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the case of large dust outbreaks (Kahn et al., 2010; Carboni et al., 2012; Banks et al., 2013). On the modeling side, in the first AEROCOM intercomparison, the SPRINTARS model also tends to underestimate the emissions in north Africa, with dust particles having too short a lifetime (1.6 days) (Huneeus et al., 2011). In this regard, it is important to recall that the MISR data represent a temporal average over 15 years, whereas the SPRINTARS model is likely representing a single, large dust event. In spite of these differences, there is generally good agreement between the maps in Fig. 4a and b. The latitudinal spread is less in the model than in the climatology, whereas the longitudinal extent of high dust AOD loading is greater in the MISR climatology. Between the end of March and mid-October, the location of the maximum dust emission shifted with time, consistent with the differences shown here in the latitudinal distributions (Prospero et al., 2002; Ben-Ami et al., 2012). The difference in longitudinal extent could be related to the lifetime of the dust particles in the SPRINTARS model (Huneeus et al., 2011).

Focusing on the shaded rectangles on the two maps, Fig. 4c and d shows the PMFs of the non-spherical AOD from MISR and dust AOD from SPRINTARS. The two distributions have similar means (0.37 for MISR and 0.35 for SPRINTARS) and standard deviations (0.21 for MISR and 0.18 for SPRINTARS). Both distributions also have significant positive skewness (0.97 for MISR and 1.56 for SPRINTARS), with the model showing much greater skewness than the satellite observations. Looking at the distributions themselves, it is apparent that the model distribution, which is dominated by spatial variability, is much better behaved than the MISR distribution, which contains contributions from both spatial and temporal variability. Although the peaks of the two distributions are nearly identical (0.350 for MISR and 0.325 for SPRINTARS), the SPRINTARS distribution is much more sharply peaked. The MISR distribution appears to have a secondary peak around 0.2, that is not represented in the SPRINTARS distribution, which instead has a peak around 0.075. Careful inspection of the two distributions shows that frequency of AOD values larger than the highest peak in the SPRINTARS model falls off more rapidly than they do in the MISR observations, an-

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found by Liousse et al. (2010) when comparing POLarization and Directionality of the Earth's Radiation (POLDER) total column AOD measurements from the Polarization and Anisotropy of Reflectances for Atmospheric Sciences coupled with Observations from a Lidar (PARASOL) satellite with modeled AOD for July 2006. These authors attribute the displacement to errors in the location of the biomass burning emissions, errors in transport, or errors in the satellite products. In fact, the location of the maximum SPRINTARS carbonaceous AOD corresponds well with the location of the maximum BC emission in the Global Emissions Inventory Activities (GEIA) emissions (Liousse et al., 2010), which are used in the SPRINTARS model (Takemura et al., 2005). The GEIA emissions distribution in this region is consistent with version 3 of the Global Fire Emissions Database (GFED3) (van der Werf et al., 2010) as well as the Fire Energetics and Emissions Research version 1.0 (FEER.v1) (Ichoku and Ellison, 2014) database. The spatial distribution of absorbing aerosols from MISR shown in Fig. 6a is also consistent with the AOD map from POLDER shown in Liousse et al. (2010), even though the PARASOL satellite has a 13:30 LT equatorial crossing time compared to the 10:30 LT equatorial crossing time for the Terra satellite. These results implicate the transport and deposition processes in the SPRINTARS model, as opposed to the emissions inventory or the satellite products, themselves. Even so, recent work by Marlier et al. (2014) has shown that daily fire emissions lead to different model results compared to monthly fire emissions, and Veira et al. (2015) show that the injection height of aerosols can also play a role in the modeled distribution of AOD from biomass burning. The paper by Matichuk et al. (2007) includes a longer list of additional model sensitivities that complicate model predictions of carbonaceous aerosol loading, particularly in transport regions.

Figure 6c and d shows the distribution of absorbing/carbonaceous AOD from MISR and SPRINTARS for the shaded box on the maps in Fig. 6a and b, which corresponds to the region of highest AOD loading in both the observations and the model. In this case, the two distributions are quite similar. The mean MISR AOD is 0.37, and the mean SPRINTARS AOD is 0.33, with the difference likely due to the broader spatial distribu-

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Table 1. Combination of AOD by component for comparison between MISR and SPRINTARS. The names of aerosol components in MISR are from Table 1 in Kahn et al. (2015).

MISR	SPRINTARS
weakly+strongly absorbing aerosols : sph_absorb_0.12_ssa_green_09 + sph_absorb_0.12_ssa_green_08	carbonaceous aerosols
medium + coarse non-spherical aerosols : medium_grains + coarse_spheroids	dust
very small + small + medium non-absorbing aerosols : sph_nonabsorb_0.06 + sph_nonabsorb_0.12 + sph_nonabsorb_0.26	sulfate

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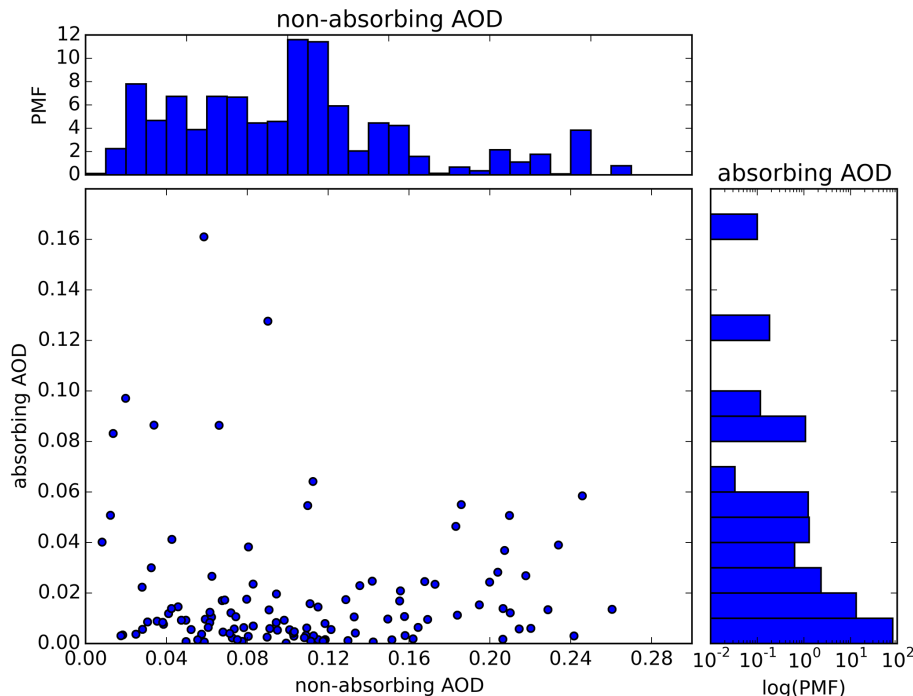


Figure 1. A scatter plot of absorbing AOD and non-absorbing AOD and their histograms over East Asia (117.5–127.5° E, 32.5–42.5° N) in April between 2000 and 2014. The AOD data are from MISR Version 1 Level 3 Joint Aerosol product (JOINT_AS) based on the Version 22 operational Level 2 aerosol retrievals. The two histograms for non-absorbing AOD and absorbing AOD are scaled to show a probability mass function (PMF) so that the total area of the bars in each histogram becomes one.

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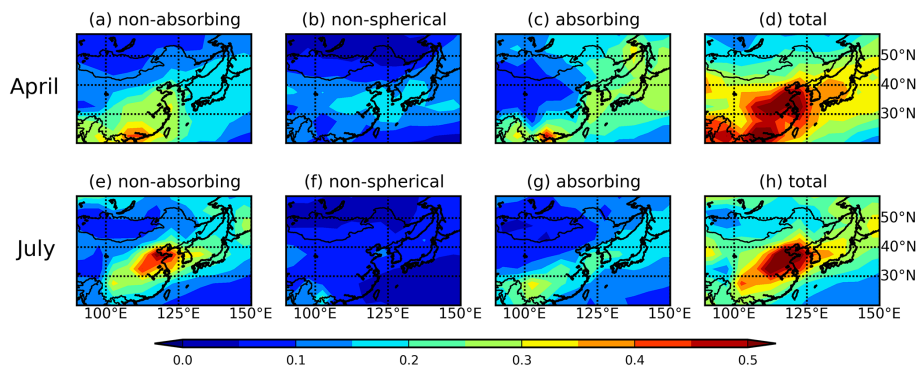


Figure 3. Map of average MISR optical depth of absorbing aerosols, non-spherical, non-absorbing aerosols and total aerosols in April (a–d) and July (e–h).

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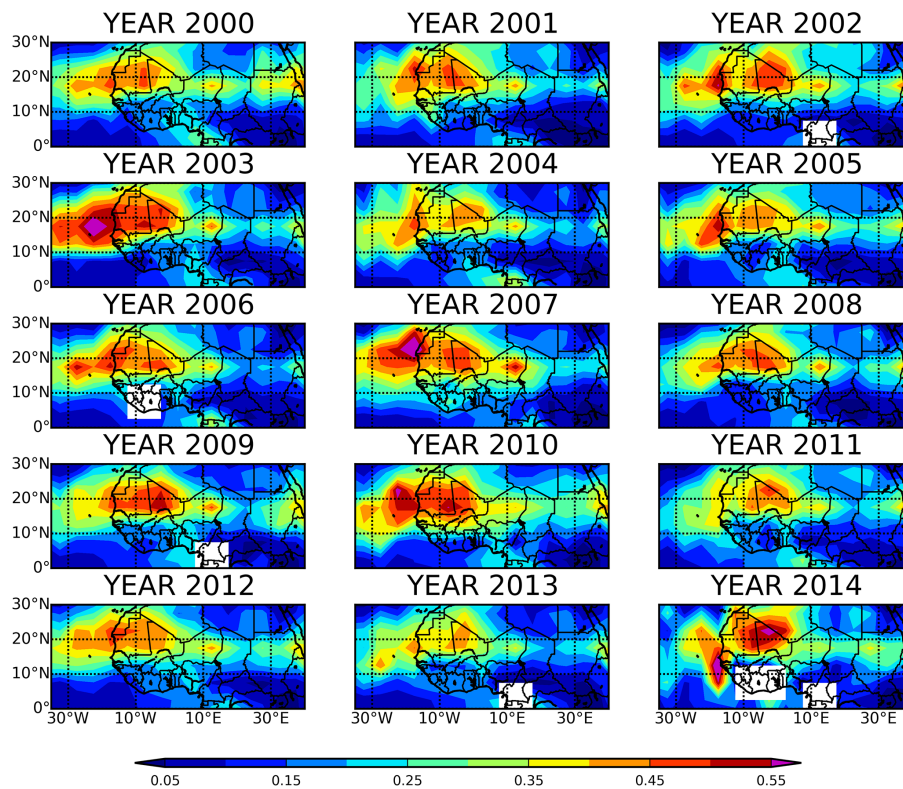


Figure 5. Monthly mean optical depth of the dust aerosols from MISR in July for 15 years between 2000 and 2014.

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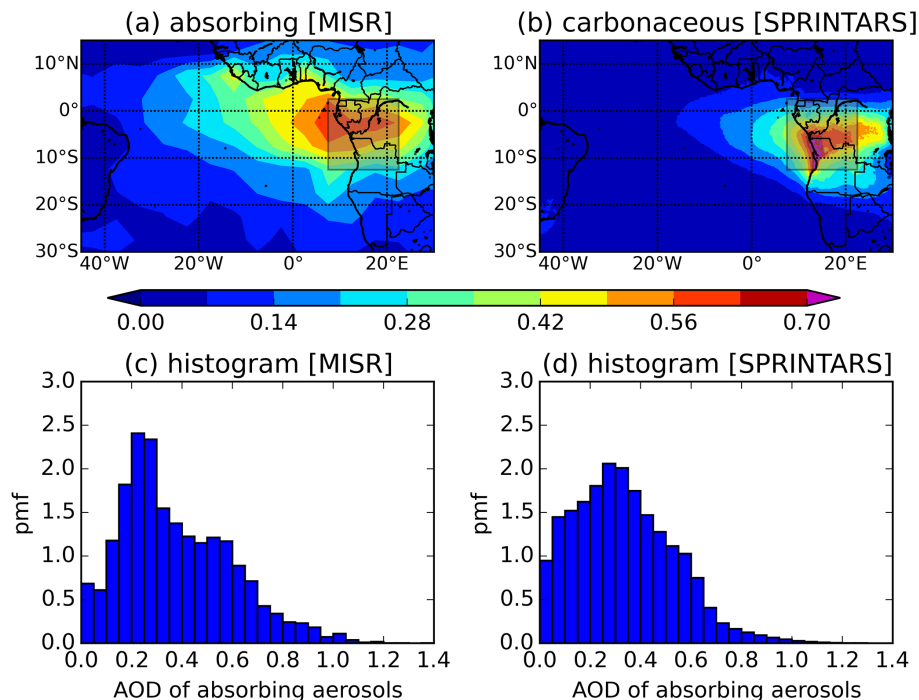


Figure 6. (a) Climatological mean optical depth of the absorbing aerosols in MISR and (b) carbonaceous aerosols optical depth in SPRINTARS over West Africa (7.5–22.5° E, 12.5° S–2.5° N). Spatio-temporal distributions of (c) the absorbing AOD in MISR for 15 Julys between 2000 and 2014 and (d) carbonaceous AOD in SPRINTARS for July 2006, covering the boxed emission source region are displayed.

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