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**Evaluation of aerosol
distributions in the
GISS-TOMAS model**

Y. H. Lee and P. J. Adams

Evaluation of aerosol distributions in the GISS-TOMAS global aerosol microphysics model with remote sensing observations

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Abstract

The Aerosol Optical Depth (AOD) and Angstrom Coefficient (AC) predictions in the GISS-TOMAS model of global aerosol microphysics are evaluated against remote sensing data from MODIS, MISR, and AERONET. The model AOD agrees well (within a factor of two) over polluted continental (or high sulfate), dusty, and moderate sea-salt regions but less well over the equatorial, high sea-salt, and biomass burning regions. Underprediction of sea-salt in the equatorial region is likely due to GCM meteorology (low wind speeds and high precipitation). For the Southern Ocean, overprediction of AOD is very likely due to high sea-salt emissions and perhaps aerosol water uptake in the model. However, uncertainties in cloud screening in high latitude make it difficult to evaluate the model AOD at high latitudes with the satellite-based AOD. AOD in biomass burning regions is underpredicted, a problem also seen in other global aerosol models but more severely in this work. Using measurements from the LBA-SMOCC 2002 campaign, the surface-level OC and EC concentrations in the model are found to be underpredicted severely during the dry season, suggesting the low AOD in the model is due to underpredictions in OM and EC mass. These, in turn, result from unrealistically short wet deposition lifetimes during the dry season in the GCM.

1 Introduction

Aerosols perturb the energy balance of the Earth-atmosphere system by scattering and absorbing solar and terrestrial radiation and by modifying cloud properties (e.g. Hansen et al., 2005; Forster et al., 2007). These changes caused by anthropogenic aerosols are termed the direct radiative forcing and indirect radiative forcing. Estimates of aerosol forcing mostly rely on computational modeling (e.g. Kinne et al., 2003; Hansen et al., 2005; Schulz et al., 2006; Forster et al., 2007) and contain significant uncertainties due to the challenges of representing aerosol microphysics, optical properties, and the spatial and temporal variability of aerosols. To gain confidence in their

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predictions, to characterize systematic weaknesses or biases, and to correct model deficiencies causing these weaknesses, the evaluation of aerosol models with observations is required. Measurements from ground and space help reduce these uncertainties (e.g. Kinne et al., 2003; Hansen et al., 2005; Schulz et al., 2006).

5 Increasing availability of satellite measurements makes it possible to evaluate the spatial and temporal variability of aerosols in global models more effectively (e.g. Kinne et al., 2003, 2006; Schulz et al., 2006). Remote sensing observations by satellites provide relatively well characterized spatial and temporal distributions and capture the large-scale transport of aerosols that are not available from in-situ observations, making them especially useful for the evaluation of global aerosol models. A radiometer used in remote sensing measures radiances that are influenced by aerosols, air molecules, surface reflectance, and cloud reflectance; a central challenge in satellite remote sensing of aerosols is distinguishing aerosols from these other contributors to radiance. Satellite and ground-based remote sensing platforms complement each other. Satellite platforms provide global distribution while ground-based radiometers, such as AEROSOL ROBOTIC NETWORK (AERONET), avoid the problem of distinguishing between aerosol and surface reflectance. The MODerate Resolution Imaging Spectroradiometer (MODIS) and Multiangle Imaging SpectroRadiometer (MISR) provide AOD observations that have frequently been used to evaluate global models (e.g. Stier et al., 2005; Kinne et al., 2003). Besides AOD, other aerosol optical and microphysical properties are also retrieved from radiometers but used more rarely due to lower confidence in data quality.

25 The AOD is a measure of light extinction in an atmospheric column. AOD is a spectrally dependent quantity, and a typical wavelength chosen for AOD is 500 nm or 550 nm because aerosols extinguish visible light effectively in the visible spectrum. The Angstrom coefficient (AC) describes the spectral dependence of AOD on the wavelength of light and provides important information about aerosol size.

Here we use remote sensing observations to evaluate the “GISS-TOMAS” model, a combination of the TwO-Moment Aerosol Sectional (TOMAS) aerosol microphysics

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model with the Goddard Institute for Space Studies General Circulation Model II-prime (GISS GCM II-prime). The goal of this study is to evaluate the GISS-TOMAS model against AOD data from MODIS, MISR and AERONET. Specifically, we seek to evaluate spatial and temporal distributions of aerosol AOD as well as aerosol size via the AC.

5 The descriptions of the GISS-TOMAS model and observational datasets used for the model evaluation are given in Sect. 2. Evaluation of the model against satellite and ground-based data is presented in Sect. 3. Section 4 provides an overall discussion of the comparisons, and Sect. 5 gives summary and conclusions.

2 Model description

10 2.1 Overview

The GISS GCM II-prime has horizontal grid dimensions of 4° latitude and 5° longitude, with nine vertical sigma layers including the stratosphere to the 10 hPa level (Hansen et al., 1983). A detailed description of the GISS GCM is found in Hansen et al. (1983). The parameterizations of convective and stratiform clouds are updated by Del Genio and Yao (1993) and Del Genio et al. (1996), respectively. The surface and boundary layer parameterization is improved by Hartke and Rind (1997), and the land-surface parameterization, e.g. surface air temperature and hydrological cycle over land, is improved by Rosenzweig and Abramopoulos (1997). Chemical tracers are advected every hour by the model winds using a quadratic upstream scheme (Prather, 1986); heat and moisture are advected with a similar scheme. Monthly average climatological sea surface temperatures data from the Atmospheric Model Intercomparison Project (AMIP) (e.g. Gates et al., 1999) are prescribed in the model, and the daily values are determined by linear interpolation.

25 The TOMAS aerosol microphysics model uses a sectional approach that represents the aerosol size distribution by predicting the amount of aerosol in several size categories or “bins”. TOMAS tracks two moments of the aerosol size distribution in each

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size bin: total aerosol number and mass. Total mass is decomposed into several aerosol species, allowing prediction of the size-resolved aerosol composition. The model has 30 size sections with the lower boundary of the smallest size bin being 10^{-21} kg dry mass, and each successive boundary has twice the mass of the previous boundary. This provides a size distribution that ranges approximately from 10 nm to $10\ \mu\text{m}$ in dry diameter, depending on aerosol density. TOMAS uses a moving sectional approach to treat water uptake; changes in water mass do not move particles between sections. Adams and Seinfeld (2002) provide a detailed description of the TOMAS model. The model tracks nine quantities for each size bin: sulfate mass, sea-salt mass, mass of pure (hydrophobic) elemental carbon (EC), mass of mixed (aged) EC, mass of hydrophobic organic matter (OM), mass of hydrophilic OM, mass of mineral dust, mass of water and the number of aerosol particles in that bin. In addition, the model tracks two bulk aerosol-phase species, methanesulfonic acid (MSA), and ammonium (NH_4^+), and five bulk gas-phase species: H_2O_2 , SO_2 , dimethylsulfide (DMS), H_2SO_4 , and ammonia (NH_3). For purposes of calculating condensation and coagulation rates, all aerosols are treated as internally mixed.

The TOMAS model used in this work includes previously developed modules for sulfate (Adams and Seinfeld, 2002), sea-salt (Pierce and Adams, 2006), and carbonaceous aerosols (Pierce et al., 2007), and mineral dust (Lee et al., 2009). Compared to the TOMAS model used in Lee et al. (2009), there are several modifications in the TOMAS model used in this work. These modifications are fully described in Sect. 2.2 of Pierce and Adams (2009a). Briefly, the pseudo-steady state assumption for sulfuric acid is assumed to solve the nucleation and condensation rates simultaneously (Pierce and Adams, 2009b), and the growth of nucleated particles up to the first size bin, a diameter of 10 nm, is taken into account using the parameterization of Kerminen et al. (2004). The primary sulfate emission is assumed to be 1% of aerosol sulfur (instead of 3%), and the size distribution of biofuel and biomass burning carbonaceous emission is changed such that the number mean diameter is now 100 nm instead of 30 nm. Finally, the sub-grid coagulation of freshly emitted primary sulfate and

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carbonaceous particles is accounted for (Pierce et al., 2009). The descriptions of wet deposition and dry deposition are available in Adams and Seinfeld (2002) and Lee et al. (2009). Briefly wet deposition is occurred in large-scale (stratiform) and convective clouds. For in-cloud scavenging, the modified Köhler theory is applied for the large-scale and convective clouds that are assumed to have a supersaturation of 0.2% and 1.0%, respectively. Dry deposition is used the series resistance approach that treats a size-dependent gravitational settling of particles and a size-dependent resistance in the quasi-laminar sublayer. Table 1 presents a summary of annual global aerosol budgets and burdens for each aerosol component in this model simulation. Water uptake by sulfate, sea-salt, and hydrophilic OM is accounted for in the model. For sulfate and sea-salt, it uses a polynomial fit based on ISORROPIA, a thermodynamic equilibrium model for inorganic aerosols (Nenes et al., 1998). For organic carbon, it is based on the observations of Dick et al. (2000). Table 2 shows the hygroscopic growth factor of those aerosols at relative humidities of 60%, 80%, and 95%. (Note that 99% is the maximum relative humidity allowed in the model for aerosol water uptake).

2.2 Aerosol optical depth module

A module for AOD (aerosol optical depth) calculation has been developed for the GISS-TOMAS model as an offline process. The AOD module uses a look-up table of aerosol optical properties that are calculated based on Mie theory. The module requires input data including refractive index, aerosol density for each species, aerosol particle size, and aerosol concentration data. The AOD is calculated using the GISS-TOMAS model monthly average output, which provides aerosol size distributions and aerosol concentrations for every grid cell, with assumed refractive index values and aerosol densities for each species as listed in Table 2. In the AOD prediction, all aerosol species are assumed to be internally mixed except pure EC. The volume averaging method is applied to get the complex refractive index for the internally mixed aerosol particle (including water). The model AOD prediction is an offline process and does not distinguish the cloud-free sky with the cloudy sky, while the remote sensing observations retrieve

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AOD only under the cloud-free (clear) sky. It is important to mention that the satellite retrieved data has limitations, including potential biases due to measurements in only clear-sky conditions and the possibility of cloud contamination. Recent studies have sought to improve cloud screening algorithms and to try to retrieve aerosol properties near clouds (e.g. Brennan et al., 2005; Koren et al., 2007; Redemann et al., 2009; Twohy et al., 2009). The module calculates AOD at four wavelengths: 440 nm, 500 nm, 550 nm, and 675 nm. The 500 nm and 550 nm wavelengths are used to evaluate the model AOD with the AERONET and satellite measurements, respectively. The wavelengths of 440 nm and 675 nm are used to calculate an AC (Angstrom coefficient) that is evaluated with AERONET.

2.3 Remote sensing observations

AERONET is a worldwide federation of ground-based and automated Sun photometers that measure aerosol optical properties and precipitable water (Holben et al., 1998). Sun- and sky-scanning CIMEL Sun photometers utilized in the AERONET program measure AOD every 15 min at 340, 380, 440, 500, 675, 870, 935, and 1020 nm wavelengths bands (e.g. Holben et al., 1998). AERONET spectral AOD data, and by definition the AC as well, have been widely used as a standard for validating other remote sensing aerosol retrievals due to well characterized uncertainties in the AERONET measurements (e.g. Abdou et al., 2005; Kahn et al., 2005; Liu et al., 2004; Torres et al., 2002). However column-integrated aerosol microphysical properties from AERONET have not been systematically evaluated against in-situ data (Kahn et al., 2004). In this paper, only AOD and AC are used. The data used in this work are monthly average AERONET Level 2.0 AOD. We selected 28 AERONET sites that represent the following characteristic regions: polluted continental, marine, biomass burning, and mineral dust-dominated. The AERONET site information is presented in Table 3. The following sites do not provide AOD at 500 nm: Banizoumbou, Barbados, Bidi Bahn, Capo Verde, and Yulin. For these sites, an AOD at 500 nm is estimated using AC obtained from AOD at 440 and 670 nm. Where possible, we select sites providing multi-year measurement

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data, but some sites (Los Fieros and Yulin) do not provide this.

Terra, the first Earth Observing System satellite launched in December 1999, carries MODIS and MISR. MODIS has 36 spectral channels, covering the wavelength range from 0.405 to 14.385 μm with a swath width of 2330 km. It covers the globe in 1–2 days. In this paper, monthly average AOD level 3 (Collection 5) products, MOD08_M3.005, are used. A description of the Collection 5 algorithm is available online (http://modis.gsfc.nasa.gov/data/atbd/atbd_od02.pdf). One important change in the Collection 5 retrieval algorithm is a new surface reflectance parameterization for the over-land algorithm that improves a bias over bright land surfaces (Li et al., 2005). MOD08_M3.005 is 1° by 1° resolution data measured for the time period 2000 to 2005 and are obtained from the Giovanni MODIS On-line Visualization and Analysis System website (http://acdisc.sci.gsfc.nasa.gov/Giovanni/modis/Terra.MOD08_M3.shtml). In some locations, e.g. bright land surfaces such as Saharan desert regions and snow/ice cover at the high latitudes and cloudy regions, Terra MODIS AOD data are not available due to failure of the retrieval process due to cloud or surface reflectance contamination.

MISR has four visible/near infra-red spectral bands (446, 557, 671, and 886 nm) and nine cameras at discrete viewing angles and has a swath width of 360 km (Diner et al., 1998). Global coverage requires approximately 9 days but depends strongly on latitude: over 9 days at the equator and 2 days near the poles. In this work, we use monthly averaged MISR Level 3 retrieved AOD data, MIL3MAE, that is considered to be “Stage 2 Validated” in the MISR data quality description. According to the MISR Level 3 quality statements, the global Level 3 MISR aerosol products over Greenland and Antarctica (snow/ice fields) are currently being excluded due to low spatial contrast and also due to inadequate cloud screening. These data are obtained from the Giovanni MISR On-line Visualization and Analysis System website (<http://g0dup05u.ecs.nasa.gov/Giovanni/misr.MIL3MAE.2.shtml>). The MISR AOD data provided are 1° by 1° resolution data for the time period 2000 to 2005. Different angular information (e.g. radiation) measured in MISR is utilized to reduce surface reflection

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contamination and cloud screening and to identify the surface type (e.g. Di Girolamo and Wilson, 2003; Jin et al., 2002). However, similar to MODIS, surface reflection contamination is still a challenge in MISR as Kahn et al. (2005) shows the lowest correlation coefficient of MISR AOD and AERONET AOD over bright desert sites. Also, like MODIS, MISR AOD data over high latitude is either not available or more uncertain. Despite challenges over bright surfaces and cloudy regions, MODIS and MISR provide long-term global aerosol distributions that are very useful for global aerosol model evaluation.

Since the GCM meteorology is generic and does not correspond to any specific year, multiyear datasets are used for comparison to minimize biases that may be magnified by a single year observation. The AOD data from both remote sensing measurements have a finer resolution than the GISS-TOMAS model. Therefore, the satellite retrieved AOD data in 1° by 1° resolution are averaged to obtain AOD on a 4° by 5° grid.

3 Results

3.1 Global aerosol budgets

Figure 1 shows the spatial distribution of column mass concentration [mg m^{-2}] of each aerosol species including water and column aerosol number concentration [m^{-2}]. To put these results in the context of other published global aerosol models, we compare our predicted column burdens to those from the AEROCOM global model intercomparison activity (Kinne et al., 2006). In Kinne et al. (2006), 20 global aerosol models reported speciated column masses. A subset of 15 global models is selected by eliminating 5 outliers including one model without AOD calculation and a multi-model and global average column mass is calculated for each species from these 15 models. Our column masses and these AEROCOM average values are compared in Table 1. All species in our model are within $\sim 20\%$ of the AEROCOM averages, except OM and sea-salt, which differ by a factor of approximately two. Our model OM column mass

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(1.55 mg m^{-2}) is half of the OM column mass (3.3 mg m^{-2}) in Kinne et al. (2006) and our model sea-salt column mass (27.5 mg m^{-2}) is almost double the column mass (12.6 mg m^{-2}) in Kinne et al. (2006). Note that Kinne et al. (2006) presents the results of the “EXPERIMENT A” simulations, which requests outputs from simulations with the model in its standard configuration.

3.2 Evaluation against MODIS and MISR retrieved AOD

Figure 2 shows the GISS-TOMAS model, MISR, and MODIS AOD global distributions for each season. Three-month average in satellite retrieved data is obtained in each grid with available monthly average data. For example, when there is only one monthly average data available among the three months, the available data is used as the three-month average. However, when a grid cell does not have any available data due to the failure to retrieve AOD data from the satellite measurements, a grey color indicating no data available is shown in Fig. 2. Figure 2 illustrates several important features of the model AOD prediction as compared to satellite observations. First, the model AOD plume from North Africa is quite well predicted throughout the year except during JJA. Second, compared to the two satellite retrievals, our model AOD is underestimated in biomass burning regions in South Africa and South America. MODIS and MISR observed a significant AOD (typically over 0.2) throughout the year, while the model shows very low AOD (less than 0.1). Third, an underestimation of AOD is observed along the equator especially in the Pacific Ocean and the Indian Ocean. Finally, the model predicts a high AOD in the Southern Ocean due to the presence of sea-salt.

To illustrate spatial trends in the comparison of model predictions to remote sensing measurements, the ratios of annual average model AOD to MODIS and MISR retrieved AODs are presented in Fig. 3. To get an annual-average MODIS/MISR retrieved AOD, we use the three-month average AOD data shown in Fig. 2. To avoid biases in the annual-average observed AOD, the averaging is only performed when a retrieved AOD is available in all four seasons. Otherwise, the annual-average is shown as “no data”

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with the grey color in Fig. 3. There is a strong latitudinal trend in model performance. Near the equator, the model tends to underestimate AOD compared to satellites, related to sea-salt from oceanic regions and carbonaceous aerosols from biomass burning regions. Conversely, the model tends to overpredict AOD compared to satellite data at high latitudes.

Figure 4 presents four scatter plots of annual-average model AOD versus MODIS and MISR retrieved AOD. These scatter plots are categorized by areas with high concentrations of a given species to diagnose the model AOD prediction biases. Column mass concentration is used to identify where an aerosol species is significant. High sulfate columns (called “polluted continental” regions) are those greater than 15 mg m^{-2} ; high OM columns (called “biomass burning” regions), greater than 5 mg m^{-2} ; high mineral dust columns, greater than 50 mg m^{-2} ; high sea-salt columns, greater than 50 mg m^{-2} . Additionally, we define “moderate sea-salt” regions as those having sea-salt column mass concentrations between 20 and 50 mg m^{-2} . High EC areas are mostly the same as high OM areas. In this scheme, it is possible for a model column to fall into more than one category. Also, we see from Fig. 1 that portions of India and China fall into the “biomass burning” category using this scheme. Table 4 presents the log-mean normalized bias (LMNB) and log-mean normalized error (LMNE) of each plot in Fig. 4. From Fig. 4 and Table 4, we see that model AOD agrees with MODIS and MISR within a factor of two for dusty (high dust), polluted continental (high sulfate), and moderate sea-salt regions. However, consistent with finding above, model AOD in high OM regions is significantly underestimated, while that in high sea-salt region is significantly overestimated. For the high sea-salt region, the LMNB of 0.47 (MODIS) and 0.41 (MISR) indicates an average overestimation by a factor of 3.0 and 2.6, respectively. In the high OM region, the model is systematically underpredicts AOD by a factor of 3 compared to MODIS and MISR.

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3.3 Evaluation against AERONET AOD and AC

The geographical locations of the 28 AERONET sites are presented in Fig. 5. Figure 6 shows a scatter plot of annual-average AOD prediction and AERONET AOD at 28 measurement sites divided into four categories: biomass burning (green), dusty (pink), polluted continental (orange), and oceanic sites (blue). Table 4 shows the corresponding LMNB and LMNE for each regional category. Figure 7 shows temporal distributions of model AOD and AERONET AOD from January to December.

The model predictions are mostly underestimates in the biomass burning sites (1 to 6) and are more severe in South American sites (1, 2, and 3) and the South African site (4). Ilorin (5) and Banizoumbou (6) are also influenced by mineral dust aerosols, which may explain why they are better predicted than sites 1 to 4, which are dominated by biomass burning. Model AOD is underestimated by a factor of 4.2 (LMNB: -0.62) on average for these sites. Excluding the two sites in North Africa where mineral dust is also transported (sites 5 and 6), the model AOD is underestimated by a factor of 7.2 (LMNB: -0.86). Possible causes of the underprediction are discussed in Sect. 4.

Most dusty sites (7 to 14, except 9) agree with AERONET AOD within a factor of two. The AOD in dusty regions is underpredicted by only 10% on average (LMNB: -0.05), and the model predictions are typically within a factor of 1.6 of observed values (LMNE: 0.21). In Fig. 7, dusty sites (7 to 14) agree well with AERONET except summer and autumn periods in regions influenced by Africa dust (sites 5 though 9).

All polluted continental sites (15 to 22) show good agreement except Mexico City (19), which is underpredicted by an order of magnitude. The AOD prediction in polluted continental sites is underestimated by a factor of 1.4 on average (LMNB: -0.15), and the model predictions are within a factor of 1.7 of observed values (LMNE: 0.23). Excluding Mexico City, the AOD predictions are better, with a LMNB of 0.95 and LMNE of 0.11. The oceanic sites (23 to 28) are underpredicted, more severely near the equator (28). The AOD prediction in oceanic sites is underestimated by a factor of 2.7 on average (LMNB: -0.43), and the model predictions are within a factor of 2.8

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of observed values (LMNE: 0.44). Bermuda (23) and Rottneast Island (27) are also influenced by other aerosol species, e.g. long-range transported mineral dust. Note that Nauru (28) is not shown in Fig. 6 because its model AOD (0.005) is very low. Without Nauru (28), which suffers from low wind speeds predicted by the model (see Sect. 4), the model prediction is within a factor of 2, consistent with good model prediction in moderate sea-salt region for the evaluation with MODIS and MISR.

Figure 8 displays seasonal cycles of monthly averaged model AC and AERONET AC. Model AC in African regions (4, 5, and 6) generally agrees fairly well, while those in South America exceed the AERONET AC. Generally dusty sites tend to have lower model AC than AERONET AC. The dust model in the GISS-TOMAS model produces fewer sub-micron dust particles compared to the limited available observations (Lee et al., 2009), which may explain the lower AC. Polluted continental sites show very good agreement to AERONET, while oceanic sites except Bermuda tend to lie on low AERONET AC or lower.

4 Discussion

Generally, the GISS-TOMAS evaluation of AOD against MODIS, MISR, and AERONET shows good agreement (within a factor of 2) with the best agreement occurring in polluted continental regions (high sulfate regions), dusty regions, and moderate sea-salt regions. One of polluted continental locations, Mexico City, is significantly underpredicted as shown in Fig. 6. This underprediction is very likely because the model grid size is too large to capture the unique topographical features of the Mexico City air basin. This underprediction is also found in other models (Kinne et al., 2003). The underprediction of AOD in dusty regions during JJA is illustrated in Figs. 2 and 7. This is due to low surface wind speeds that results in underestimation of mineral dust during summer and autumn (Lee et al., 2009).

Significant underpredictions in AOD are found over the equatorial oceanic regions (e.g. Nauru 28), while significant overpredictions are found over high-latitude marine

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regions. To investigate this further, Fig. 9 shows a scatter plot of simulated and observed sea-salt surface mass concentrations from Savoie and Prospero (1977), divided into NH high latitude, SH high latitude, and near equatorial regions. In order to match the PM₁₀ inlet of typical aerosol samplers in an environment with 80% relative humidity, similar to the procedure used by Guelle et al. (2001), predicted sea-salt mass is associated with particles smaller than 4 μm dry diameter (Pierce and Adams, 2006). The model significantly underpredicts sea-salt mass concentrations at Nauru (0.5° S 167° E) and Enewetak Atoll (11.3° N 162.3° E), where the observed sea-salt mass concentrations are highest. In contrast, the model agrees well with observations at Fanning Island (3.9° N 159.3° W), where the observed concentrations are moderate. The model's tendency to underpredict sea-salt mass (and therefore AOD) in equatorial marine areas is mostly due to low biases in GISS GCM wind speeds that reduce sea-salt emissions (Pierce and Adams, 2006). In addition, a potential issue of heavy precipitation near the equator that causes low aerosol loading in GCM ModelE (Miller et al., 2006), with some of the same physics as the GISS GCM II', may partly explain the underprediction at the equator.

For high latitude, the model AOD is overpredicted noticeably. It is important to mention that the clear-sky AOD from satellite is expected to be much lower than the all-sky AOD from our model over the high latitude. Figure 2 in Schmidt et al. (2006) shows AOD from the clear-sky and all-sky in GISS ModelE, and AOD in SH/NH high latitude is much higher in all-sky due to the particle growth by water uptake at high relative humidity in cloudy-sky. In fact, the model high AOD is mostly contributed by aerosol-associated water mass. The average hygroscopic growth factor in SH high latitude (50° S to 65° S), defined the wet particle mass divided by the dry particle mass, is about 12, corresponding to the relative humidity of 95%. Even though the mass extinction efficiency decreases with particle size and is generally small in coarse mode, high sea-salt mass as well as water mass can increase AOD significantly.

For NH high latitude, the model sea-salt and mineral dust in Mace Head (53° N 10° W) and Heimaey (63° N 20° W) are underestimated, but the discrepancy is less than a

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factor of two, except the mineral dust in Heimaey (63° N 20° W), which is underpredicted more than an order of magnitude. However the model AOD is overpredicted by a factor of 2–3 compared to MISR data and a factor of 3–4 for the MODIS data: this difference between MISR and MODIS in high latitude could be due to cloud screening algorithms (Myhre et al., 2005). The overprediction of AOD prediction in NH high latitude may be mostly due to including cloudy-sky.

For SH high latitude, Fig. 9 show the overprediction of sea-salt mass by about a factor of 2.6 in Palmer Station (65° S 64° W). Worse agreements are found in Marsh (62° S 58° W) and Mawson (68° S 62° E). The overprediction of sea-salt mass concentration in SH high latitude region may be mainly due to the emission scheme used. Figure 5 in Pierce and Adams (2006) presents four different sea-salt emission schemes and the model simulation with Clarke et al. (2006) results in the overprediction of sea-salt mass concentration at Palmer Station. No AOD retrieved from satellite is available for either of these stations, but the nearest available satellite AOD show worse biases: the overprediction of about a factor of 4 against MISR and more than a factor of 5 against MODIS-retrieved data (Fig. 3). Therefore, the overprediction of AOD prediction in SH high latitude is probably partly by including cloudy-sky model AOD in these areas but also partly by sea-salt emission overprediction.

The model underpredicted AOD in biomass burning regions compared to both AERONET and satellite measurements. Our model simulations shown here use the Bond emissions inventory (Bond et al., 2004). To investigate any problems associated with that inventory in biomass burning regions, two additional biomass burning emission inventories were tested: the Global Fire Emission Database (GFED) inventory (Dentener et al., 2006) and an emission inventory used by the IPCC Third Assessment Report (IPCC, 2001), which is based on Penner et al. (1993) and Liousse et al. (1996) (see more detailed description in Pierce et al., 2007). Using the same OM:OC ratio assumption as our model, the GFED emission inventory gives similar total OM emission rate as BOND emission, but the seasonality of AOD prediction in South America in GFED emission is improved. The IPCC inventory has higher emissions than the BOND

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inventory (54 Tg yr^{-1} compared to 45 Tg yr^{-1} for primary OM and 5.7 Tg yr^{-1} compared to 3.3 Tg yr^{-1} for EC). Using the IPCC inventory, the model's AOD prediction in South American sites (1, 2, 3) is improved (not shown), but it is still underestimated by a factor of 5, as compared to a factor of 7.5 with the original Bond et al. (2004) emissions. In summary, our model underpredicts AOD in South America with any of the commonly accepted biomass burning emission inventories.

Global models have generally underestimated AOD over biomass burning regions albeit less than in our model. In Kinne et al. (2006), global models based on the same GFED emission have generally underpredicted AOD over the tropical biomass burning regions compared to AERONET AOD. According to Bond et al. (2004), Chin et al. (2002) used a biomass inventory based on Yevich and Logan (2003) that uses much higher emission factors for biomass burning than those assumed in Bond et al. (2004) and obtains a good agreement between their model and AERONET AOD. This may suggest that higher emission factors are needed. Emission-related factors could explain why global models have generally underestimated the AOD over biomass burning regions.

In our model, this general tendency to underestimate biomass burning AOD values appears to be exacerbated by too much precipitation. Wet deposition is a major removal process for the carbonaceous aerosols, and its lifetime is expected to be long during the typical dry season, from August to September, when the maximum biomass burning emission is occurred. However, the model-predicted wet deposition lifetime over the AERONET sites in South America shows a short lifetime, less than 2 days, during the dry season but a long lifetime from March to June. The predicted organic and elemental carbon concentrations from the simulation with the GFED emissions are compared to the Large Scale Biosphere-Atmosphere Experiment in Amazonia – Smoke, Aerosols, Clouds, Rainfall, and Climate (LBA-SMOCC) experiment 2002, which was conducted in Rondônia ($10^\circ \text{ S } 62^\circ \text{ W}$), Brazil during September to mid-November 2002 (Decesari et al., 2006). The model fine OC concentration ($9.2 \mu\text{g m}^{-3}$) during the dry season is only about 30% of the observation ($31.6 \mu\text{g m}^{-3}$) while the transition and wet

season are within a factor of two or less. Unlike OC, the model fine EC concentration ($0.8 \mu\text{g m}^{-3}$) is over 70% of the observation ($1.1 \mu\text{g m}^{-3}$) during the dry season. Although this is a rough comparison since the observation is only for three months in 2002 and the model is a one-year simulation with the GCM climatology, it helps to link the underprediction of AOD with the underprediction of surface mass concentration during the dry season.

5 Summary and conclusions

A module for calculating AOD is implemented in the GISS-TOMAS global aerosol microphysics model, and model predictions are evaluated against remote sensing data from MODIS, MISR, and AERONET. Generally, evaluations against satellite-based and AERONET AOD lead to similar conclusions. However, the global coverage of satellite data greatly increases our confidence that these conclusions are true for large regions and are not significantly dependent on local factors specific to AERONET sites. The model AOD agrees with AOD retrieved from satellites within a factor of two over polluted continental, dusty, and moderate sea-salt regions. Similarly, the AOD evaluation against AERONET agrees generally within a factor of two over polluted continental locations (except Mexico City), dusty, and remote oceanic sites except equatorial oceanic sites.

In equatorial marine regions, the model AOD is significantly underpredicted compared remote sensing measurements. The low AOD results from an underprediction in sea-salt concentrations, which in turn result from wind speeds in the GCM. High precipitation rates may also play a role.

There are large errors (\sim a factor of 5) over high latitude marine regions in both the Northern and Southern Hemispheres. Overprediction of AOD in high latitude is likely due to inclusion of cloudy-sky in the model AOD calculation compared to satellites that sample clear sky only. However, in SH high latitudes, the model overprediction is also partly due to high sea-salt emission from the emission scheme used in the model. It is

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worthy to note that satellite retrievals over high latitude have limitations such as larger differences in AOD retrieved among satellites possibly due to cloud screening algorithms, difficulty in cloud screening (distinguish between cloud droplets from swelling aerosols by water uptake), and biases toward sampling clear sky conditions. In this regard, it is difficult to evaluate the model AOD in high latitude with the satellite-based AOD.

AOD in biomass burning regions is generally underpredicted in global models. However, our model AOD is more severely underpredicted during the dry season over South America. Compared to the surface-level OC and EC concentrations from LBA-SMOCC 2002 experiment data, the model shows significant underpredictions during the dry season. The underprediction is observed with all commonly used global emissions inventories. The underprediction is most likely due to very short wet deposition lifetime (<2 days) in the model that cause low OM and EC loading and thus low AOD during the dry season. Since global models have generally underestimated AOD over biomass burning regions, this might indicate that current biomass burning emissions inventories may be too low.

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Table 1. Globally and annually averaged aerosol budgets, burdens, and the comparison of column mass to AEROCOM column mass.

Aerosols	Total Sources [Tg yr ⁻¹]	Dry Deposition [Tg yr ⁻¹]	Wet Deposition [Tg yr ⁻¹]	Burden [Tg]	Lifetime [days]	Model Column mass [mg m ⁻²]	AEROCOM Column mass [mg m ⁻²]
Sulfate*	44	1.1	43	0.73	6.0	4.27	3.9
Sea-salt	7380	4740	2640	13.0	0.64	27.53	12.6
Mixed EC	1.6	0.25	7.61	0.14	7.0	–	–
Pure EC	6.4	0.13	0.01	0.03	1.5	–	–
Total EC	8.0	0.38	7.62	0.17	4.3	0.33	0.39
Hydrophilic OM	30.5	1.6	48.7	0.70	5.1	–	–
Hydrophobic OM	30.5	0.45	10.3	0.08	0.94	–	–
Total OM	61.0	2.0	59.0	0.78	3.5	1.55	3.3
Dust	2440	1900	560	17	2.6	37.1	39.1

* Note that, for sulfate, the units of sources and sinks are Tg S per year and Tg S for the burden.

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Table 2. Aerosol densities, refractive indices (RI), and hygroscopic diameter growth factors.

Aerosol Type	Density [kg m ⁻³]	Real RI	Imaginary RI	Hygroscopic growth factor		
				RH: 60%	RH: 80%	RH: 95%
Sulfate	1780	1.53	1.0e-7	1.20	1.39	2.02
Sea-salt	2165	1.50	1.0e-8	1.63	1.89	2.74
Elemental Carbon (EC)	2200	1.75	0.44		–	
Organic Matter (OM)	1400	1.53	0.006	1.15*	1.23*	1.31*
Dust	2650	1.53	0.0055		–	
Water	1000	1.33	1.96e-9		–	

* The hygroscopic growth factor for OM applies only to hydrophilic OM.

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Table 3. Locations of AERONET sites and corresponding measurement time periods.

	Sites	Longitude	Latitude	Years
1	Alta Floresta	56.0° W	9.9° S	1999–2005
2	Los Fieros	60.6° W	14.6° S	1996
3	Cuiaba-Miranda	56.0° W	15.7° S	2001–2005
4	Mongu	23.2° E	15.2° S	1995–2005
5	Ilorin	4.3° E	8.3° N	1998–2005
6	Banizombou	2.0° E	13.0° N	1995–2005
7	Capo Verde	22.9° W	16.7° N	1994–2004
8	Bidi Bahn	2.5° W	14.1° N	1996–1997
9	Barbados	59.5° W	13.2° N	1996–2000
10	Sede Boker	34.8° E	30.9° N	1998–2005
11	Bahrain	50.6° E	26.2° N	2004–2005
12	Solar Village	46.4° E	24.9° N	1999–2005
13	Dalanzadgad	104.4° E	43.6° N	1997–2005
14	Yulin	109.7° E	38.3° N	2001–2002
15	Sevilleta	106.9° W	34.4° N	1994–2005
16	Cart site	97.5° W	36.6° N	1996–2005
17	Bondville	88.4° W	40.1° N	1996–2005
18	GSFC	76.8° W	39.0° N	1995–2005
19	Mexico city	99.2° W	19.3° N	1999–2005
20	Ispra	8.6° E	45.8° N	2001–2005
21	Kanpur	80.3° E	26.5° N	2001–2005
22	Shirahama	135.4° E	33.7° N	2000–2005
23	Bermuda	64.7° W	32.4° N	1996–2005
24	Lanai	156.9° W	20.7° N	1996–2004
25	Dry Tortugas	82.9° W	24.6° N	1996–2003
26	Tahiti	149.6° W	17.6° S	1999–2005
27	Rottneest Island	115.5° E	32.0° N	2001–2004
28	Nauru	166.9° E	0.5° S	1999–2005

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Table 4. Log-mean normalized bias (LMNB) and log-mean normalized error (LMNE) of model-predicted AOD values compared with MODIS, MISR, and AERONET AOD observations.

Observation	Pollution type	LMNB	LMNE
MODIS	Dusty region	−0.15	0.23
MISR		−0.04	0.16
AERONET		−0.05	0.21
MODIS	Polluted continental region	0.11	0.19
MISR		0.16	0.19
AERONET		−0.15	0.23
MODIS	Biomass burning region	−0.56	0.56
MISR		−0.52	0.53
AERONET		−0.62	0.62
MODIS	High sea-salt region	0.47	0.47
MISR		0.41	0.41
MODIS	Moderate sea-salt region	−0.02	0.16
MISR		−0.06	0.19
AERONET	Oceanic region	−0.43	0.44

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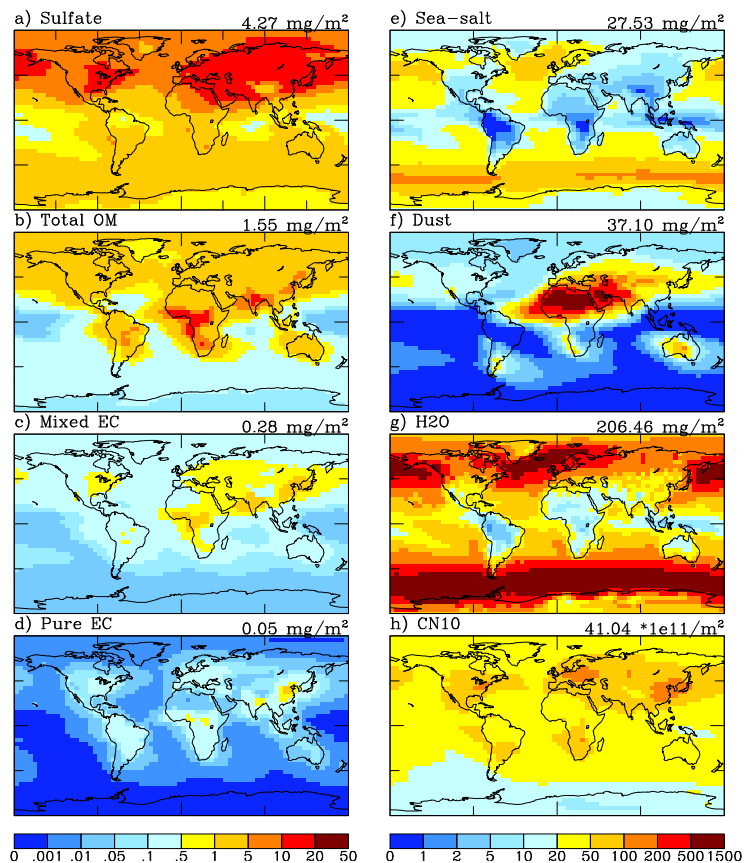


Fig. 1. Global distributions of annual-average aerosol column mass [mg m^{-2}] and column number [m^{-2}]. Values on the upper-right of each panel give global-average column mass and number concentrations.

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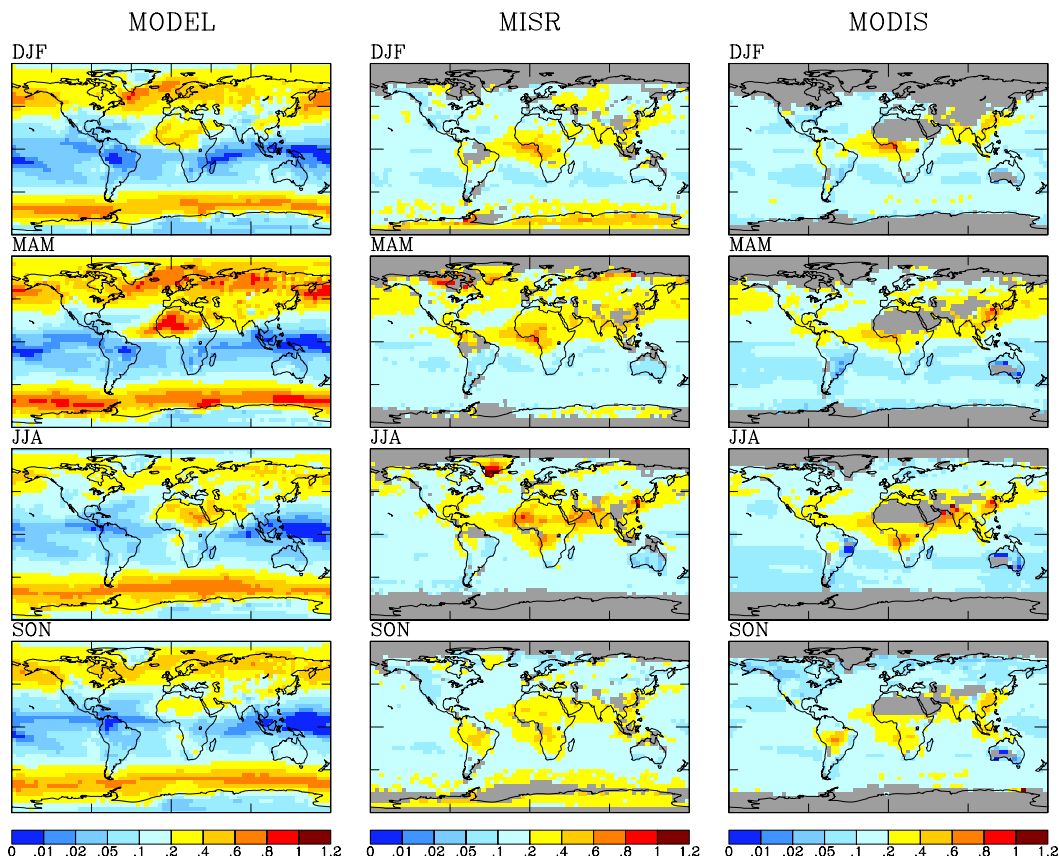


Fig. 2. Global distribution of AOD in the model, from MISR, and from MODIS in four seasons. Grey color in satellite observations indicates no data available. From top to bottom, the rows correspond to seasonal average AOD from December to February (DJF), March to May (MAM), June to August (JJA), and September to November (SON).

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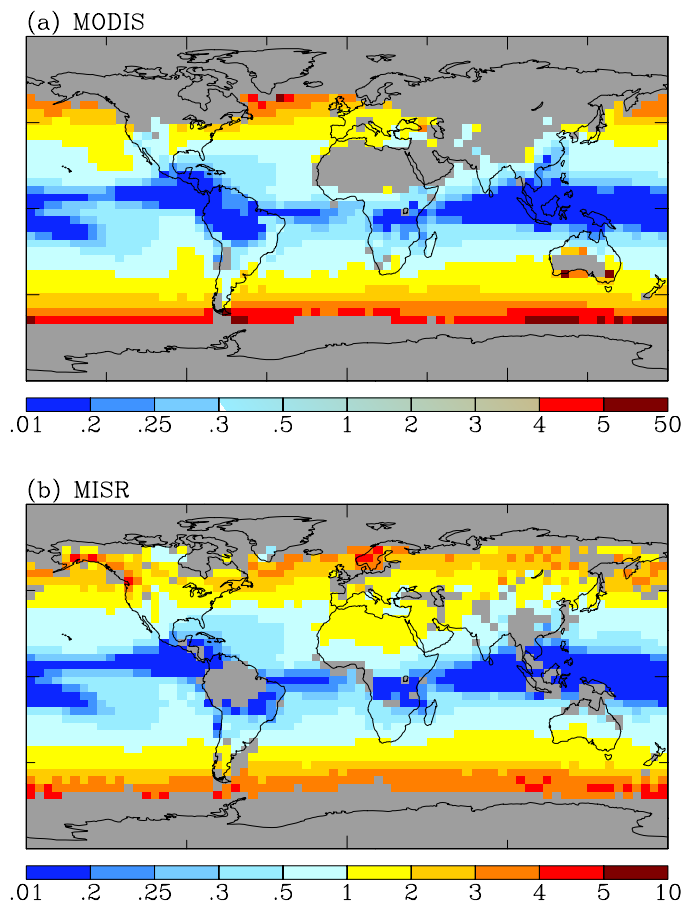


Fig. 3. Global maps of the ratio of annual-average AOD in the model prediction to **(a)** MODIS and **(b)** MISR AOD. Grey color indicates no data available from the satellite instrument.

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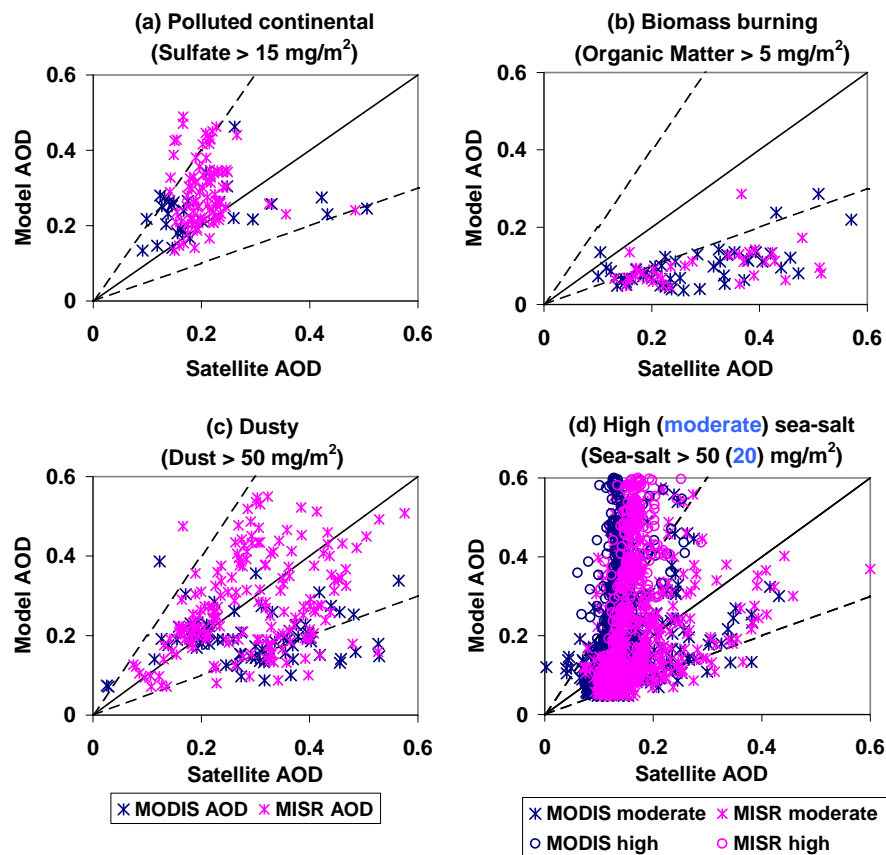


Fig. 4. Scatter plots of model-predicted AOD and satellite-retrieved AOD. The solid line and dashed lines are for 1:1 and 1:2 (or 2:1), respectively. In **(a)–(d)**, MODIS and MISR AOD are presented as blue and pink, respectively. In panel (d), for high sea-salt areas, MODIS and MISR AOD are presented as circles.

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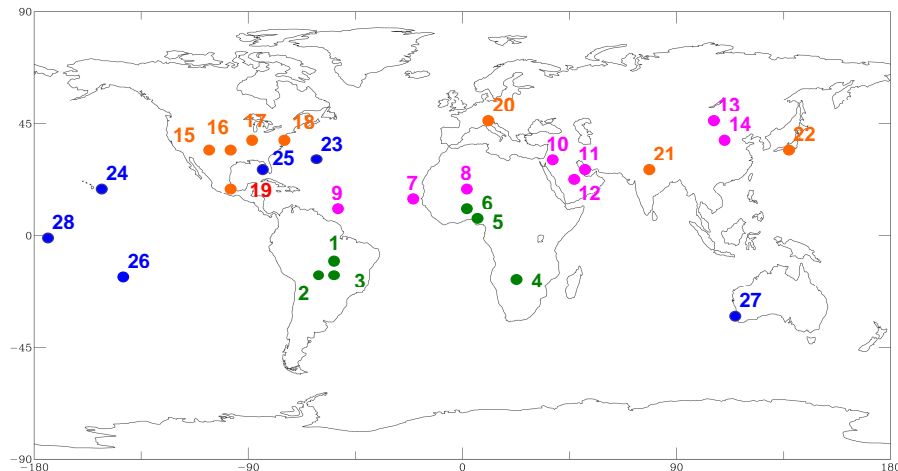


Fig. 5. Geographical location of 28 AERONET sites used in this work. Biomass burning sites are green, dust-dominated sites are pink, polluted continental sites are orange, and marine sites are blue. The site numbers correspond to those in Table 3.

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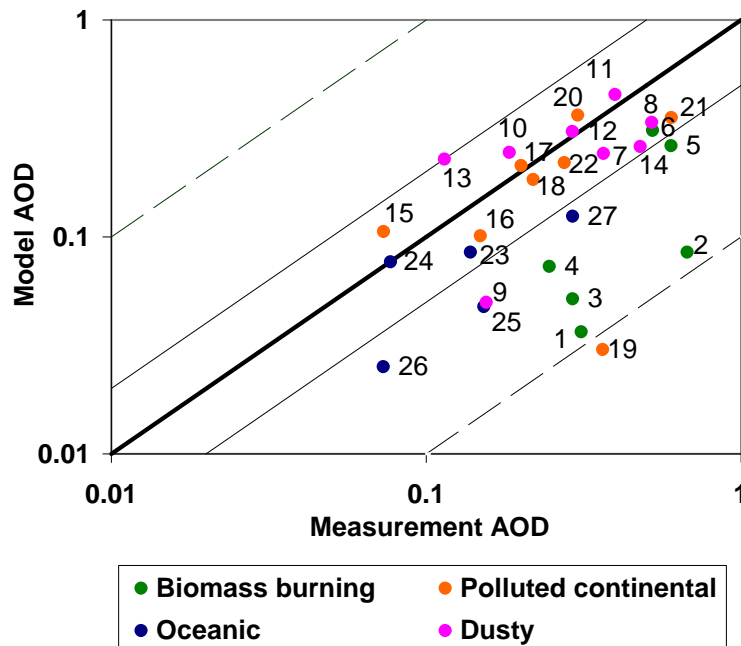


Fig. 6. Scatter plot of AOD from the model and AERONET. Biomass burning sites are green, dusty sites are pink, polluted continental sites are orange, and oceanic sites are blue. The thick and thin solid lines are for 1:1 and 2:1, respectively. The thin dashed line is for 10:1. The site numbers correspond to those in Table 3.

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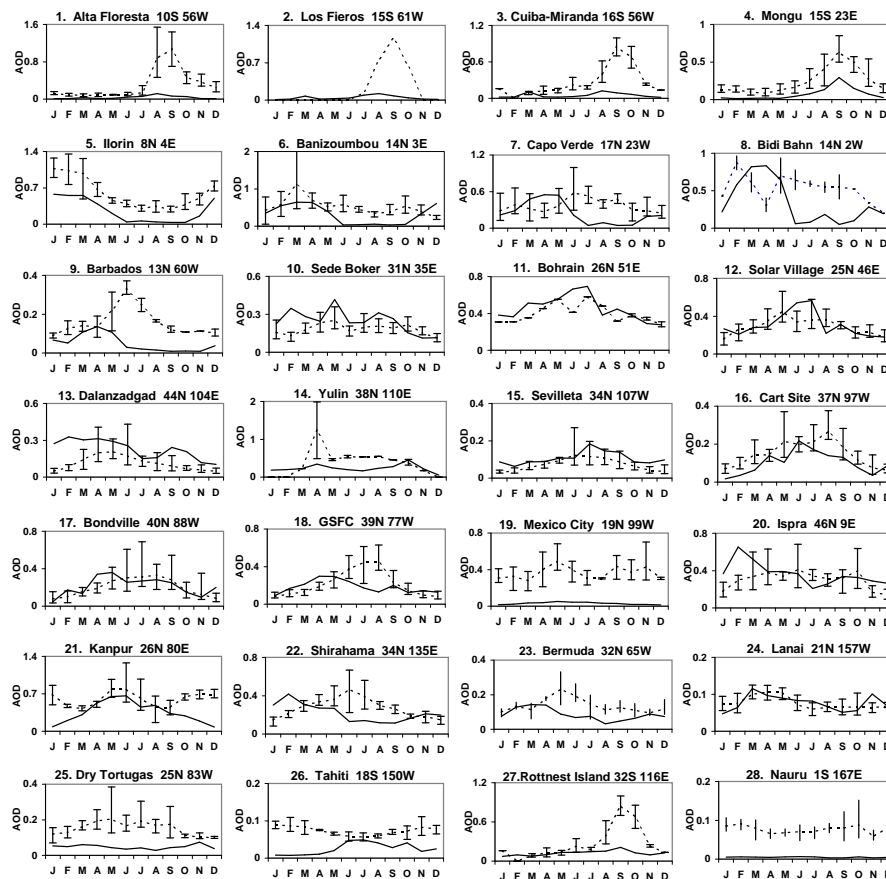


Fig. 7. A comparison of monthly averaged model AOD (solid lines) and AERONET AOD (dashed lines). The bars around the AERONET data denote minimum and maximum monthly average values in the multi-year measurements. The site numbers correspond to those in Table 3.

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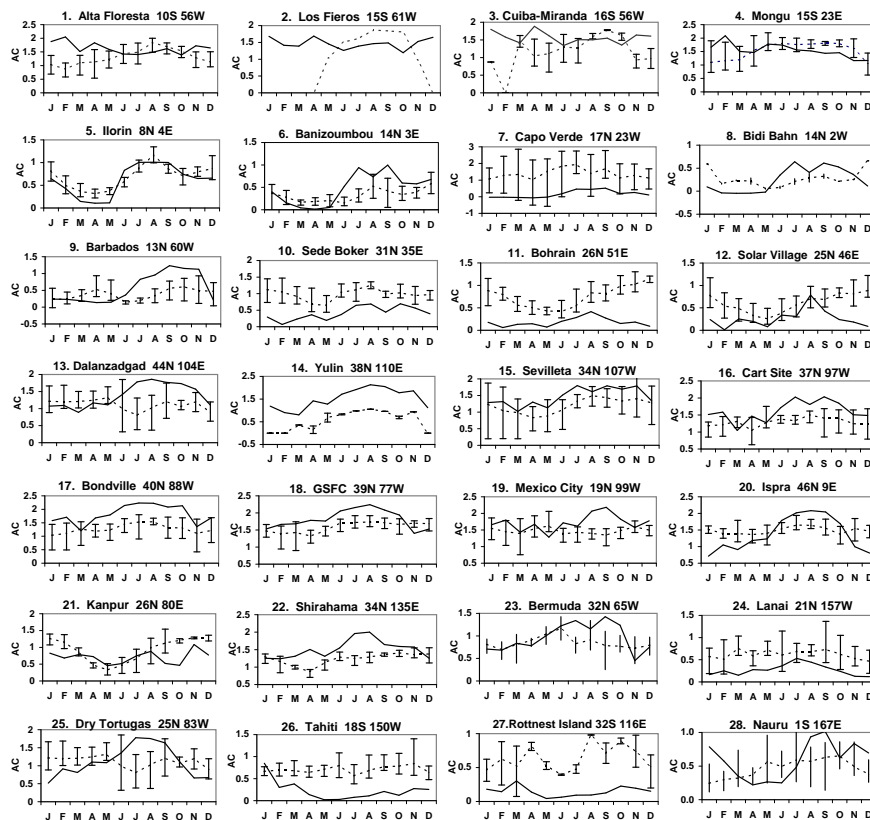


Fig. 8. Temporal distribution of AC (Angstrom coefficient) of the model and AERONET. The solid line is for the model prediction and the dashed line is for AERONET. The bars around the AERONET data denote minimum and maximum monthly average values in the multi-year measurements.

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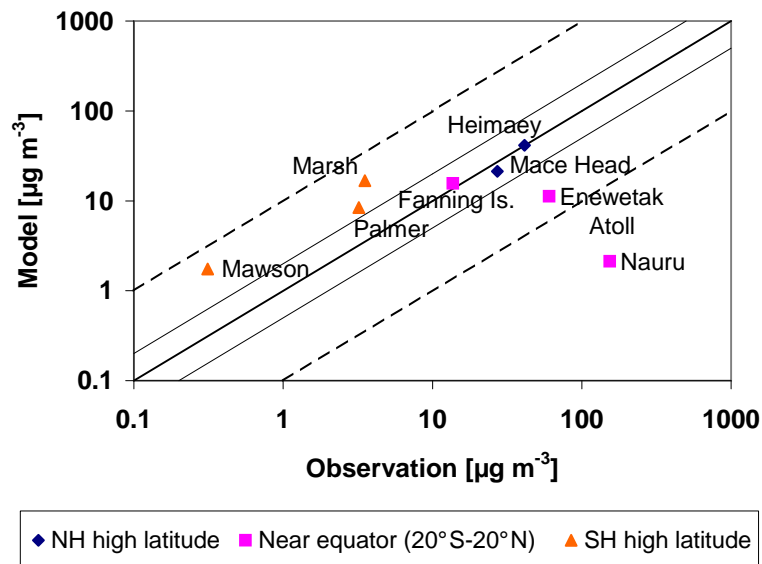


Fig. 9. Scatter plot of annually averaged surface mass concentrations of sea-salt predicted by the model and measurements in units of $\mu\text{g m}^{-3}$ at 273 K and 1013 hPa. Thick and thin solid lines are the 1:1 line and 2:1 lines, respectively. Dashed lines are 10:1 lines.

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