

Aerosol distribution in the northern hemisphere during ACE-Asia: Results from global model, satellite observations, and sunphotometer measurements

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Abstract

We analyze the aerosol distribution and composition in the northern hemisphere during the ACE-Asia field experiment in spring 2001. We use the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model in this study, in conjunction with satellite retrieval from the Moderate Resolution Imaging Spectroradiometer (MODIS) on EOS-Terra satellite and sunphotometer measurements from the world-wide Aerosol Robotic Network (AERONET). Statistical analysis methods including histograms, mean bias, root mean square error, correlation coefficients, and skill scores are applied to quantify the differences between the MODIS $1^\circ \times 1^\circ$ gridded data, the day-time average AERONET data, and the daily mean $2^\circ \times 2.5^\circ$ resolution model results. Both MODIS and the model show relatively high aerosol optical thickness (τ) near the source regions of Asia, Europe, and northern Africa, and they agree on major features of the long-range transport of aerosols from their source regions to the neighboring oceans. The τ values from MODIS and from the model have similar probability distributions in the extra-tropical oceans and in Europe, but the MODIS is approximately 2 – 3 times as high as the model in North/central America and nearly twice as high in Asia and over the tropical/subtropical oceans. Comparisons with the AERONET measurements in the northern hemisphere demonstrate that in general the model and the AERONET data have comparable values and similar probability distributions of τ , whereas MODIS tends to report higher values of τ over land, particularly North/central America. The MODIS high bias is primarily attributed to the difficulties in land algorithm dealing with surface reflectance over inhomogeneous and bright land surfaces, including mountaintops, arid areas, and areas of snow/ice melting and with land/water mixed pixels. The model estimates that on average, sulfate, carbonaceous, dust, and sea-salt comprise 30%, 25%, 32%, and 13% respectively of the 550-nm τ in April 2001 in the northern hemisphere, with about 46% of the total τ from anthropogenic activities and 66% from fine mode aerosols.

1. Introduction

The Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia), which studied the characteristics of aerosols from Asia and their radiative effects, took place in spring 2001 in the western Pacific region near the east coast of Asia. In the spring, dust emission in northern Asia is strong, biomass burning in Southeast Asia is at its peak, photochemical production of pollution aerosols is active, and the continental outflow from Asia to the western Pacific is at its strongest. In other words, the timing of ACE-Asia was optimal for studying the impact of maximum Asian aerosol concentrations on downwind regions. The ACE-Asia field experiment involved three aircraft and two ships in coordination with surface and lidar networks and satellite overpasses [Huebert et al., 2003]. Coincident to the ACE-Asia observations, which covered only a limited geographical location, measurements from satellites and a world wide, ground-based, sunphotometer network provided global-scale aerosol information. These larger scale measurements placed the ACE-Asia observations into a broader perspective. A global model can synthesize such a wide array of observations in order to assess the global impact of Asian aerosols and to quantify the processes that control the aerosol composition and distributions.

In this study, we use the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model to address the characteristics of aerosol distribution and composition in the northern hemisphere in spring 2001. During the ACE-Asia intensive operation period (March 30 – May 4, 2001), the GOCART model was used in the forecast mode, providing daily aerosol forecasts to support the flight planning [Chin et al., 2003; Huebert et al., 2003]. These results have been verified by the aircraft measurements [Chin et al., 2003]. The focus of this paper is to evaluate the model calculated aerosol optical thickness (τ) and aerosol size information by comparing to the satellite retrieval from the Moderate Resolution Imaging Spectroradiometer (MODIS) and

sunphotometer measurements from the Aerosol Robotic Network (AERONET). The comparisons cover over both source regions and downwind areas in the northern hemisphere. Several statistical parameters, including histograms, mean bias, root mean square error, correlation coefficients, and skill scores, will be introduced to quantitatively evaluate the model. Based on the model results, we estimate the aerosol composition and the anthropogenic contributions in the northern hemisphere during spring 2001. The present work leads to a companion paper (Chin et al., Intercontinental transport of aerosols in the context of ACE-Asia, manuscript in preparation) which specifically addresses the impact of long-range transport of pollution and dust aerosols originating from major source regions on regional and hemispheric scales.

Section 2 provides a short description of the GOCART model and the MODIS and AERONET measurements. In section 3, we first compare the distributions of τ and the fraction of fine mode aerosols (f_r) calculated from the model with those retrieved from the MODIS instrument. Then we use the measured τ from 57 AERONET sunphotometer sites in the northern hemisphere to evaluate the corresponding quantities from MODIS and the model. In section 4, we estimate the aerosol composition and fractions of anthropogenic versus fine mode aerosols in total τ . We discuss the results and possible causes of discrepancies between the model and observations in section 5 before we conclude in section 6.

2. Description of the model and the data

2.1. The GOCART model for tropospheric aerosols

Detailed description of the GOCART model has been presented and results have been extensively evaluated in our previous publications [Chin et al., 2000a, 2000b, 2002, 2003;

Ginoux et al. 2001, 2004]. Here we provide a brief summary of the GOCART model and its recent modifications. The model simulates major tropospheric aerosol types of sulfate, dust, organic carbon (OC), black carbon (BC), and sea-salt. It uses assimilated meteorological fields from the Goddard Earth Observing System Data Assimilation System (GEOS DAS) that include winds, temperature, pressure, specific and relative humidity, cloud mass flux, cloud fraction, precipitation, boundary layer depth, surface winds, and surface wetness. The spatial resolution of the model is currently at 2° latitude by 2.5° longitude, with total 30 vertical layers. Physical processes in the model include emission, advection, convection, boundary layer mixing, wet deposition (rainout and washout), dry deposition (a function of surface resistance and atmospheric stability), and gravitational settling. Chemical processes include gas and aqueous phase reactions that convert sulfate precursors (dimethylsulfide, or DMS, and SO₂) to sulfate.

Global emissions of aerosols and their precursors have been updated and modified from the earlier version of the GOCART simulations [Chin et al., 2002, 2003] to reflect the most recent knowledge about these sources. Anthropogenic emissions of SO₂, OC, and BC for Asia are from the most recent and detailed emission inventory for the year 2000 [Streets et al., 2003]. For the rest of the world, we use the IPCC emission scenario of SO₂ for 2000 [Nakićenović et al., 2000]. Biomass burning emissions of SO₂, OC, and BC for March and April 2001 are obtained based on the CO emissions estimated from the AVHRR satellite fire counts [Heald et al., 2003] and the burned biomass inventory constructed using other satellite data [Duncan et al. 2003; Chin et al., 2002].

The dust sources are assumed to be in topographic depression areas with bare soil surfaces and the dust uplifting probability is defined according to the degree of depression [Ginoux et al. 2001]. Over Asia, the dust source is modified that include the detailed surface information and

the recent desertification regions over China (Q. Gao, unpublished data) which seem to be mainly responsible for the boundary layer dust over the western Pacific observed during ACE-Asia [Chin et al., 2003]. Dust particles ranging from 0.1 to 10 μm in radius are simulated by the model.

Volcanic emissions take account of sources from both continuously and sporadically erupting volcanoes. The major source of volcanic emissions in the ACE-Asia region is the Miyakejima volcano in Japan (34.08°N, 139.53°E), which started erupting in September 2000. The Miyakejima volcanic plume was estimated to have reached 500 – 2000 m above the rim, emitting 20 – 50 kton SO_2/day (e.g. <http://hakone.eri.u-tokyo.ac.jp/vrc/erup/miyake.html>). Here we assume an emission rate of 28 kton SO_2/day from January to mid May 2001 with an emission height at 1500 m above the volcanic rim (813 m above sea level).

Other emissions in the model include biogenic emissions of OC and oceanic emissions of DMS and sea-salt (0.1 – 10 μm), which have been described previously [Chin et al., 2002, 2003]. Figure 1 shows the emissions in April 2001 of sulfur (SO_2 , DMS, and sulfate), carbonaceous (OC + BC), dust, and sea-salt emissions for the northern hemisphere in this study.

The aerosol optical thickness τ is determined from the dry mass concentrations and the mass extinction coefficients. The mass extinction coefficients are calculated from the Mie-theory based on size distributions, refractive indices, and hygroscopic properties of individual aerosol types. We assume single-mode lognormal size distributions for sulfate, OC, and BC aerosols as well as for each dust and sea-salt size bins (details in Chin et al., 2002). Although a recent study has shown that the difference of extinction coefficient between externally and internally mixed aerosols is between 0 to over 50% (internal mixture being lower) depending on the relative humidity and aerosol composition [Lesins et al., 2002], we assume here that all aerosol particles

are externally mixed due to the difficulties in realistically determining the degree of the mixing state.

2.2. Aerosol data from MODIS retrievals and AERONET measurements

The MODIS instrument aboard the EOS-Terra satellite, which has been taking aerosol measurements since 2000, provides daily nearly global coverage with local equatorial overpass time about 10:30 am [King et al., 1999]. The MODIS aerosol retrieval uses separate algorithms over land and ocean to obtain aerosol optical properties in cloud free areas, including total aerosol optical thickness τ and fine mode (submicron particle size) aerosol fraction f_{τ} [Tanré et al., 1997; Kaufman et al., 1997a, 2002; Remer et al., 2002, 2004; Chu et al., 1998, 2002, 2003; Levy et al., 2003, 2004]. We use in this study the version 4 level-3 quality-assured MODIS daily τ at 550 nm, which is a globally gridded dataset at $1^{\circ} \times 1^{\circ}$ horizontal resolution. There is no aerosol retrieval over bright land surfaces (such as desert and snow covered surfaces) and ocean sun-glint areas.

Over ocean, the τ in seven wavelength bands (0.47 – 2.13 μm) and f_{τ} are retrieved by a “least residual method” that minimizes the difference between measured and calculated reflectance at the top of atmosphere. The calculated reflectance is obtained from optimized combinations of four fine and five coarse aerosol property lognormal modes [Tanré et al., 1997]. Over land, the aerosol properties are derived in two visible wavelengths (0.47 and 0.66 μm) using the “dark target” method that assumes a globally fixed empirical ratio between the surface reflectance in the two visible wavelengths to the measured reflectance at 2.13 μm ($R_{0.47}/R_{2.13} = 0.25$, $R_{0.66}/R_{2.13} = 0.50$) [Kaufman et al., 1997b]. The path radiance is determined to be a function of the difference between the estimated surface reflectance and the satellite measured reflectance in the two visible channels. The fine mode fraction f_{τ} over land in the MODIS standard product is

obtained based on the ratio of the path radiance in the two visible wavelengths, a relatively narrow spectral range that may cause high bias of f_{τ} [Chu et al., 2003]. In this study, we use an alternative formulation of f_{τ} that reduces the high bias of f_{τ} over land. Here, the land f_{τ} is determined from the Ångström exponent (α) over land and the knowledge of the relationship of f_{τ} and α over ocean [Chu et al., 2004], thus taking advantage of more accurate ocean retrievals obtained using a wider spectral range (0.47 – 2.13 μm). Even with improved formulation of f_{τ} , retrievals of τ still have large uncertainties, because the assumption of the empirical surface reflectance ratios seems too simple to account for the complexities of land surface reflectance [Levy et al., 2004].

The AERONET, an international federated sunphotometer network [Holben et al., 1998], currently has about 180 ground-based remote sensing monitoring stations. These sites represent virtually all aerosol regimes in a wide range of geographic locations, through seasonal and annual cycles. AERONET measures the total column aerosol spectral optical thickness at several visible and near infrared wavelengths, and derives a number of column-representative aerosol properties, including total and absorbing τ , size distribution, single scattering albedo, and complex index of refraction [Holben et al., 1998, 2001; Eck et al., 1999; Dubovik et al., 2002]. The accuracy of τ in the AERONET field instruments is ~ 0.01 - 0.02 [Eck et al., 1999]. Since they are from direct measurements, AERONET data are considered to be the “ground truth” for satellite and model validations [e.g., Remer et al., 2002; Chu et al., 2002; Chin et al., 2002]. Like satellite data, however, sunphotometer acquires aerosol data only during daylight in cloud free conditions. We use the quality-assured and cloud-screened level-2 AERONET data [Smirnov et al., 2000].

3. Aerosol distribution during ACE-Asia: Comparisons between the model and observations

There are spatial and temporal sampling differences among MODIS and AERONET data, both of which are subsets of the model. The model results are daily (24-hour) averages at 2° latitude \times 2.5° longitude spatial resolution, while the AERONET data are day time average at specific site locations and the MODIS retrievals are “instantaneous” at a local overpass time of 10:30 am with $1^\circ \times 1^\circ$ spatial resolution. In addition, the model results are for both cloudy and clear sky conditions, whereas the AERONET and MODIS data represent only clear sky conditions. This means that some differences among these results should be expected even in the most ideal situation (e.g., no error from measurements, retrievals or model). We compare the τ values at a commonly referred wavelength of 550 nm. Since the 550 nm τ is not directly measured by AERONET, it is interpolated from the 440 nm and 670 nm AERONET data assuming linear relationships between log wavelength and log τ .

We use a number of statistical parameters to evaluate the quality of the model output, which we refer to as “*HERBS*”: The histogram (H), which shows similarity between the peak, spread, and skewness of the observed and calculated aerosol distributions; the root mean square error (E), which reveals the magnitude of absolute difference between the model and observations; the correlation coefficient (R), which measures the linear correspondence; and the mean bias (B), which represents the ratio of the model results to the data. The skill score (S), which considers both correlation and standard deviation between two datasets, is defined as

$$S = \frac{4(1+R)}{(\sigma_f + 1/\sigma_f)^2(1+R_0)}$$

where σ_f is the ratio of the standard deviations of two datasets, and R_0 is the maximum attainable correlation coefficient [Taylor, 2001]. Here, we assume R_0 to be 1, although in reality it should always be less than 1 due to some intrinsic differences (e.g. spatial and time resolution) between the model and observations. The S value ranges from 0 to 1.

3.1. Aerosol distribution in the northern hemisphere in spring 2001

Monthly averaged northern hemispheric distributions of τ and fine mode fraction f_τ from MODIS and the model in April 2001 are shown in Figure 2a and 2b, respectively. The f_τ from the model is defined as the ratio of the sum of the τ from sulfate, OC, BC, and submicron dust and sea-salt to the total τ . As we mentioned in section 2, MODIS is unable to retrieve aerosol information over bright surfaces, in the presence of clouds, and over ocean sun-glint areas, therefore with fewer observational days over ocean than over land because of more frequent cloud cover and sun-glint interference. Also, there is no data over large desert areas in Sahara, Arabia, and Asia and most of the areas north of 60°N (Figure 2c). The monthly averaged results from the model in Figure 2a and 2b are constructed from the daily values that match the days and locations of the available MODIS data (Figure 2c) for a more meaningful comparison.

Both MODIS and the model show relatively high τ near the source regions such as eastern Asia, Europe, and northern Africa. They also reveal some major features of the long-range transport, such as from Asia to North Pacific, from North America to North Atlantic, and from Africa to the equatorial North Atlantic (Figure 2a). The most noticeable differences occur in North America and in the subtropical/tropical oceans, where the τ from MODIS is a factor of two to three higher than those from the model. We will discuss these discrepancies later.

When the monthly averaged f_τ for April 2001 from MODIS and the model are compared (Figure 2b), both show that f_τ is greater than 0.5 over most areas in the northern hemisphere with

the highest values (> 0.8) in and near the anthropogenic and biomass burning source regions. However, the f_{τ} from MODIS is 0.1 to 0.5 higher than the model in the eastern U.S., Europe, and equatorial Africa, and is 0.3 to 0.5 lower than the model at high latitudes (about 60°N) over land (North America and Eurasia) and over the equatorial Pacific and Indian oceans. In addition, the MODIS f_{τ} data also show discontinuities at some land-ocean borders, presumably from the use of different retrieval algorithms for land and ocean.

To further analyze the similarities and differences in spatial distributions between the MODIS and the model, we plot in Figure 3 the probability distributions (or normalized histograms) of τ and f_{τ} at different land and oceanic regions in the northern hemisphere. Here, the western North Pacific, eastern North Pacific, and North Atlantic oceans are divided into northern (latitude $\geq 30^{\circ}\text{N}$) and southern (latitude $< 30^{\circ}\text{N}$) parts, with a total of six oceanic sub-regions. The land is also divided into six sub-regions: Northern and southern Asia, northern and southern America, Europe, and Africa/Middle East, all separated by the 37°N latitudinal line.

The τ values from MODIS and the model have similar distributions (close to lognormal) over the northern part of North Pacific and North Atlantic oceans where the most probable, or modal, τ (τ_m) is about 0.2 – 0.3, with the model being 25 to 33% lower (Figure 3a, left column). In the southern parts, however, they differ by almost a factor of two; the τ_m in the model is 0.08 – 0.11 whereas MODIS reports 0.15 – 0.2 (Figure 3a, right column). These differences are consistent with Figure 2. Over land (Figure 3b), the model displays similar τ_m distributions to MODIS in Europe (τ_m at 0.36), but are significantly lower than MODIS for other regions. The largest discrepancy is clearly over North America where the τ_m from the model (0.11) is less than a third of MODIS (0.36).

Figure 3c and 3d show the probability distributions of f_{τ} over the same ocean and land sub-regions that are displayed Figure 3a and 3b. Both MODIS and the model show similar normal distributions over the oceanic regions, except in the southern NW and NE Pacific where the model indicates a second peak at f_{τ} of 0.9 where MODIS has only a single peak at 0.5 (Figure 3c). Over land, MODIS exhibits a relatively constant distribution of f_{τ} , between 0.2 and 1, in northern Asia and northern North America, whereas the model displays a well defined symmetrical distribution with a modal f_{τ} of 0.68. In other land regions, the MODIS data indicate that aerosols are predominantly composed of small particles with a modal $f_{\tau} > 0.9$. The model agrees with MODIS in southern Asia but shows considerably lower f_{τ} (differences between 0.12 – 0.47) in other areas (Figure 3d).

The regions where MODIS and the model have the largest discrepancies include North/Central America, northern Asia, and tropical/subtropical oceans, where the model values are less than one-half of those from MODIS. In other areas, the model agrees with MODIS reasonably well, although the τ from the model is generally lower than that from MODIS. These differences are further analyzed in the next section, by including AERONET data.

3.2. Daily variations of aerosols

The daily variations of τ from the model calculations and from the MODIS retrievals are compared with that measured by the AERONET sunphotometers. There are 57 AERONET sites in the northern hemisphere that have at least 3 days of measurements during April, 2001. These sites include in anthropogenic and dust source regions of Asia, North America, Europe, Africa, and Middle East, oceanic regions immediately downwind of aerosol sources, , and cleaner locations in the tropical oceans. Locations of the 57 sites are listed in Table 1 and are also shown in Figure 4 superimposed with the modeled monthly average τ for April 2001. (We will use the

site numbers in Table 1 to facilitate our discussions throughout the text.) No MODIS data are available at sites #1 – 2 in the Asian dust area and #48 – 49 in the Arabian Desert, due to their very bright surfaces.

Figure 5 shows the time series of daily τ in April 2001 from AERONET, MODIS, and the model at a subset of the AERONET sites. There are three sites each in Asia (1st row), North America (2nd row), Europe/Africa (3rd row), and oceans (last row), demonstrating daily variability seen in these geographic regions.

In Asia, Beijing (site #4) has high local pollution sources all year long but is heavily under the influence of dust in spring time. Large aerosol episodes observed by AERONET on April 4, 8, 10, 18, and 28 are mostly associated with dust events. Here the model calculated τ is about half of that measured by AERONET, even though the temporal variations from the model are similar to those of AERONET. At Je-Ju Island of South Korea (site #6) and Shirahama of Japan (site #8), the model indicates that pollution aerosols (mostly sulfate) contribute almost 60 – 70% to the total τ . These two sites also receive large amount of dust in spring, where 40% and 30% of τ respectively at Je-Ju and Shirahama are from dust that has been transported from the Asian continent. The observed large dust peaks on April 13 at Je-Ju and on April 14 at Shirahama are successfully captured by the model. The model predicts a sulfate insurgence on April 18 at Shirahama from the nearby Miyakajima volcano, but there are no available AERONET or MODIS data to verify the model results.

In North America (2nd row, Figure 5), Missoula (site #15) in the western U.S. is proximate to dust transported from Asia during the spring season, KONZA_EDC (site #23) in the central U.S. (Kansas) is influenced by both local sources and transport, whereas GSFC (site #26) in the eastern U.S. is dominated by pollution and sulfate aerosols. As shown in Figure 5, sulfate

amount increases whereas dust decreases eastward across North America. Yet, the Asian dust can arrive to east coast of North America, suggested by the model. About 15% of the τ at GSFC during April is dust, most which is transported from Asia. At Missoula, model estimates of τ are similar to AERONET, about half of the values retrieved by MODIS. The high values of MODIS may be attributed to the complexity of Missoula's land surface; which is arid and surrounded by mountains and snow. At KONZA_EDC, both AERONET and MODIS show a large episode on April 13 that is not reproduced by the model.

Avignon (site #36) is located near the coast in southern France where both the model and AERONET usually report τ as below 0.2. The τ from MODIS, on the other hand, is nearly double that from both AERONET and the model. The high values from MODIS are related to the coastal nature of the site with mixed land/water pixels, leading to a high bias [Chu et al., 2002]. The large dust event on April 21 over Bucarest (site #42 in Romania) is indicated by both the model and the AERONET data (no MODIS data on that day), although the τ from the model is 50% higher than that from AERONET. It appears that this dust was transported from Africa, where a similar maximum occurred 1 – 2 days earlier at THALA (site #45 in Tunisia) detected by both measurements and the model. During the last 5 days in April, the model simulates a large dust episode at THALA, which is consistent with MODIS observations. The τ values, however, are more than double those of AERONET.

The effects of land surface properties on the MODIS retrieval quality are reflected in the results at Bucarest (site #42) and Missoula (site #15, in the U.S.). Although they are located at similar latitudes, Bucarest, elevation at 44 m, was snow free in early April 2001, where the MODIS retrieval agrees with the AERONET data within 25%; by contrast, Missoula, at 1028 m

altitude and surrounded by mountains, had snow on the surface in April, where MODIS overestimates the τ by more than a factor of 2.

At three oceanic sites, Midway (site #50) in the open Pacific Ocean, Bermuda (site #54) and Azores (site #55) in the western and eastern North Atlantic, the model shows a sea-salt contribution of 13 – 19% to the total τ in April 2001. The model indicates that dust is the most important aerosol component at both Midway and Azores (35%), originating from either Asia or from northern Africa, whereas the pollution sulfate from North America contributes to about 50% of total τ at Bermuda.

3.3. Statistical analysis of aerosol optical thickness at AERONET sites

We grouped the 57 AERONET sites into four geographic regions in order to perform statistical analysis. Of those sites, 12 are in Asia, 23 in North America plus Surinam (we use “America” for this region), 14 are in Europe/Africa/Middle East, and 8 are in the islands over the oceans (Table 1). The statistical parameters of mean bias (B), root mean square error (E), correlation coefficient (R), and skill score (S), between the model and observations for the 57 sites are summarized in Figure 6. Only days in April 2001 when either AERONET or MODIS data are available are analyzed (days shown in the last panel of Figure 6). While the B (1st panel) measures the magnitudes of the model results relative to the data, the E (2nd panel) points out the absolute differences that are generally proportional to the magnitude of the τ . The model shows relatively large differences (ranging between one-half and double) as compared to AERONET data at the Asian dust sites Dunhuang (#1) and Dalanzadgad (#3) and also the immediate downwind sites Beijing and Xianghe (sites #4 and #5). This finding illustrates the challenge in modeling the large spatial and temporal variability of dust near its source by a relatively coarse resolution model. Similar to the results shown in Figures 2 and 3, the model reports τ values

much lower than by MODIS, ranging between a factor of two to four. As compared to AERONET, however, model has relatively small errors and biases (mostly within 50%). At several sites in Africa and the Middle East (#43 – 46), the model agrees better with MODIS than with AERONET. Over ocean, the model agrees with AERONET to within 50% at all sites, except at Male (#57), but consistently reports much lower values (50 – 120%) than MODIS. At Male, located near the tropics, has the lowest τ among 8 oceanic sites shown in Figure 4. Here the modeled April averaged τ (0.03) is much lower than both AERONET and MODIS data, with a mean bias $B = 0.3$.

The correlation coefficients R (3rd panel in Figure 6) reflects the model's performance at simulating the observed daily variation of τ . The R varies from -0.23 to 0.96 between the model and AERONET and -0.20 to 0.92 between the model and the MODIS. These two sets of R usually track each other with a few exceptions, such as Rodgers Dry Lake in Southwest U.S. (#17) and two African sites of Ouagadougou (#43) and Ilorin (#44). At Rodgers Dry Lake, the R is 0.7 between the model and AERONET but -0.1 between the model and MODIS; note that the τ from the model is 4 times lower than MODIS but about the same as AERONET. In contrast, the R values between the model and MODIS at Ouagadougou and Ilorin are 0.92 and 0.70 , respectively, but the model is negatively correlated to AERONET at these two sites ($R = -0.05$). This may be attributed to the fact that MODIS has far fewer days of coverage than AERONET (last panel in Figure 6) missing some extreme cases, particularly at Ilorin. The skill score (S) offers a more comprehensive assessment of the model performance than other parameters of B , E , and R , because it considers both correlation and variance between the model and the data. As shown in the 4th panel of Figure 6, whereas the model indicates comparable skills in simulating the AERONET and MODIS data in Asia, Europe, and Middle East, it is much more skillful in

reproducing the AERONET data than MODIS at almost every site in North America and over oceans.

The probability distributions of the daily τ at sites in each geographic region are shown in Figure 7. Days included in the histograms are those when both MODIS and AERONET measurements are available, so that some of the dust sites are not included here for lack of MODIS retrievals. The model results for the matching dates are also plotted. The τ typically exhibits a probability distribution that is approximately lognormal, as shown previously from AERONET observations [O'Neill et al., 2000]. Over Asia (Figure 7a), the model and AERONET have similar distributions with the modal value, τ_m , at 0.35. The shape of the MODIS distribution is somewhat skewed toward high τ , with a τ_m at 0.64. In America (Figure 7b), the model and AERONET have a similar τ_m at 0.13, with AERONET showing a wider range. The τ values from MODIS in this region are, as seen before, considerably higher than both AERONET and the model, with a τ_m at 0.27. In Europe/Africa/ Middle East region (Figure 7c), we find the τ_m of the model (0.23) in between MODIS (0.32) and AERONET (0.16). Over the 8 oceanic sites (Figure 7d), the AERONET, MODIS, and model all illustrate a bimodal distributions, but the model and AERONET have a dominant peak with τ_m of 0.13 whereas the MODIS shows equal magnitude between the two modes.

Figure 8 summarizes the monthly averaged τ for April 2001 from AERONET, MODIS, and the model at the 57 AERONET sites, obtained by averaging only the days with available observations from both AERONET and MODIS. The exceptions are the dust sites #1 – 2 and #48 – 49 where only AERONET and the model are available. The statistical parameters B , E , R , and S for each region are listed in Figure 8. The AERONET data can be considered as “calibration reference” because they are from direct measurements of high accuracy, even

though their data may not always be representative of the 100 – 200 km grid area in the MODIS or model results. Figure 8 clearly shows the τ values from MODIS at most sites in America that are significantly higher than both AERONET and the model, especially from sites #17 and 19 – 21 located in the Southwest U.S. and sites #29 – 31 at high latitudes in northeast of North America. MODIS differences for these sites may be attributed to the complexity of the land surfaces, including: snow/ice melting during spring in high latitude regions, mixed land/water surface sub-pixels at coastal or swamp sites, and high surface albedo at mountaintop and arid areas. The model agrees with AERONET within a factor of 2 at all sites except for Dunhuang (#1), Taiwan (#10), and Male (#57) where it differs from AERONET by a factor of 3. Assuming the AERONET data to be a reference standard, the model has generally lower bias and higher correlation (except in Asia) than does MODIS. For example, the regional averaged B values of the model are 0.88, 0.89, 1.19, and 0.97 for Asia, America, Europe/Africa/Middle East, and ocean regions, respectively, as compared to 1.30, 2.13, 1.55, and 1.41 of MODIS for the same regions (Figure 8). Overall, the model shows no systematic bias against the AERONET data ($B = 0.97$) while MODIS has demonstrated a high bias ($B = 1.64$) in the northern hemisphere during April 2001.

4. Aerosol composition and anthropogenic component in the northern hemisphere during spring 2001

As we have seen in Figure 2 and Figure 5, aerosol distribution is highly inhomogeneous and its composition varies significantly from one place to another. Crucial for assessing the anthropogenic aerosol climate forcing is the knowledge of the chemical composition of the aerosol that cannot be directly measured by AERONET or MODIS, even though the fine mode

fraction or size distribution plus spectral single scattering albedo can provide useful information on anthropogenic aerosol components (e.g., Kaufman et al., 2002; Dubovik et al., 2002; Christopher et al., 2004). The model has an obvious advantage when quantifying the chemical composition and anthropogenic contributions because they are directly simulated in the model.

The monthly averaged τ for April 2001 in the model, shown in Figure 4, has the highest values located near the Asian dust/pollution and African dust source regions. Figure 9a – 9d depicts the model calculated percentage of the τ from sulfate, carbonaceous (OC+ BC), dust, and sea-salt aerosols in April 2001. As expected, the highest percentage of each aerosol component is concentrated in its source regions (ref. Figure 1), especially dust which dominates in its regions (80 – 100% of total τ). Over land, dust and sulfate comprise the major portion (total 70 – 90%) of total τ over extra-tropical (latitudes $> 30^\circ\text{N}$) regions, whereas in subtropical and tropical areas, carbonaceous or dust dominates. Carbonaceous aerosols dominate (60 – 90%) in southern Asia and Central America, mainly from biomass burning, whereas dust dominates (60 – 100%) in Africa and the Middle East. Over ocean, dust dominates the tropical/subtropical North Atlantic (40 – 80%), sea-salt controls the equatorial North Pacific (60 – 80%), and carbonaceous aerosol is the major constituent in the subtropical North Pacific and Indian Ocean (40 – 80%). Dust influence is also shown over a substantial area in the extra-tropical North Pacific and eastern North Atlantic (20 – 40%), with sulfate and sea-salt sharing the remaining τ . We note that over the ACE-Asia experiment area in the Yellow Sea and the Sea of Japan, sulfate and dust contributes about equally to 40 – 50% of the total τ (Figure 9). On average, sulfate, carbonaceous, dust, and sea-salt comprise 30%, 25%, 32%, and 13% of the τ at 550 nm in April 2001 in the northern hemisphere. It should be pointed out that BC is optically thin, contributing to only 10 – 20% of τ in the tropical/subtropical regions and in central Europe, and less than 10%

everywhere else. However, the low single scattering albedo of BC makes it a very important absorbing aerosol in the atmosphere, with a significant impact on climate change [e.g. Hansen et al., 2000; Jacobson, 2001].

Figure 9e shows the anthropogenic fraction of the 550 nm τ . We define anthropogenic aerosol as the sum of sulfate, OC, and BC that are either directly emitted or chemically produced from precursors which are emitted from fossil fuel, biofuel, and biomass burning sources; the natural aerosols are composed with dust, sea-salt, and sulfate and OC that are formed from their gaseous precursors emitted from volcanoes, ocean, and vegetations. This assumption is somewhat oversimplified since, for example, not all biomass burning is human-caused and some dust is “anthropogenic” emitted from desertification area caused by land-use changes [e.g., Tegen et al., 2004]. The model shows that anthropogenic aerosols account for more than 50% of the τ over most land areas except dust dominated regions of northern Africa, Middle East, and Asia, and western part of North America (west of 125°W). Over oceans, 20 – 40% of τ is of anthropogenic origin except the eastern North Atlantic and the tropical/subtropical western North Pacific and Indian oceans where anthropogenic aerosol contributes to 40 – 90%. The model estimates that roughly 46% of the aerosol in the northern hemisphere in April 2001 is anthropogenic.

It is interesting to compare the anthropogenic fraction of τ with the fine mode fraction, because there have been several attempts to use the fine mode aerosol from satellite retrievals as a proxy of anthropogenic aerosol [e.g., Kaufman et al., 2002; Christopher et al., 2004]. We plot in Figure 9f the fine mode fraction in the northern hemisphere in April 2001 from the model. As we mentioned in section 3, the τ of fine mode aerosol from the model is the sum of values of sulfate, OC, BC, and submicron dust and sea salt. Figure 9f shows that the northern hemispheric

average of fine mode aerosol τ is 66%, i.e. 20% higher than the anthropogenic τ . This is because the contributions from natural sulfate and OC aerosols (formed from oxidation of DMS, volcanic SO_2 , and terpene, see section 2.1) and submicron dust and sea salt aerosols. Together, they contribute to 30% of fine mode aerosol or 20% of total aerosol.

5. Discussions

The use of the MODIS and AERONET data to evaluate the model results and characterize aerosol distributions paves the way for a quantitative assessment of the regional and global consequences of pollutant and dust emissions from different areas. The global model and satellite data together can extend the limited regional ACE-Asia measurements to a much larger spatial scale.

The MODIS instrument has provided aerosol products since 2000 with accuracies much higher than the products from previous satellite sensors not originally designed for measuring aerosols [Remer et al., 2004]. MODIS retrieves aerosol optical depth over both land and ocean and separates it into contributions from fine and coarse modes, providing multiple parameters for evaluating global aerosol models. Yet, uncertainties in the MODIS products over land are still relatively large, due to fewer usable wavelengths and much more complex surface properties than over ocean. The globally fixed ratio of surface reflectance between the visible and infrared wavelengths (see section 2.2) used in the land retrieval may be too simple to account for the variability of the ratio for different land cover types. It is an attempt to represent the global mean of the surface reflectance correctly, but the ratio at individual locations and specific surface types will deviate from the mean. While the empirical ratio works relatively well over dark, vegetated land surfaces, it is especially difficult to deal with the land surfaces with somewhat high reflectance, such as arid regions, elevated terrain (e.g. over the southwest U.S.), and snow/ice

melting areas (e.g. northeast North America in the spring), as well as in the coastal or swamp areas with mixed land/water pixels [e.g., Levy et al., 2004]. It is in these regions that MODIS has the largest disagreement with both AERONET and the model.

One unique product derived by MODIS is the fine mode fraction of aerosols (f_{τ}), which can be directly related to the fraction of anthropogenic aerosol contributions to total aerosol loading, which in turn, could lead to estimates of anthropogenic climate forcing, although the model estimates that the f_{τ} is about 20% higher than the anthropogenic fraction in the spring time of northern hemisphere. The f_{τ} over ocean from MODIS is well defined and statistically similar to the modeled values. Over land, however, the f_{τ} from MODIS is rather qualitative, indicating only clear dominance of either fine or coarse aerosol (e.g., southern Asia and southern North America in Figure 3d) or a comparable mixture of both (e.g., northern Asia and northern part of North America in Figure 3d where the MODIS shows a flat distribution). Thus, quantitative use of the current MODIS f_{τ} is, at present, only appropriate for areas over oceans. Again, the low spectral contrast from the two usable wavelengths over land, coupled with complex surface properties, leads to large uncertainties in land retrieval of f_{τ} . Our comparisons have shown that improvements of the MODIS land retrievals are needed. As future MODIS land aerosol retrievals will include improved screening for melting snow and other complicated land surfaces, we expect an improvement in MODIS data quality. Eventually the MODIS land algorithm will incorporate surface reflectance ratios dependent on land cover type, which should decrease error even further.

Over ocean, the MODIS and the model have shown similar distributions of τ in the extra-tropical oceans, but the model is usually a factor of 2 lower than the MODIS in the tropical/subtropical oceans. From the comparisons shown in Figure 6 and 8, the model seems to

have relatively small mean biases against the AERONET data at 5 subtropical Pacific sites (sites #50 – 53 and #56, $B = 0.76 – 1.3$) where MODIS is 40 – 110% higher than AERONET; on the other hand, the model is a factor of 3 lower than AERONET at the tropical Indian Ocean site Male (#57), where MODIS agrees with AERONET to within 10%. Recent studies comparing several global model results with multiple satellite products have shown that the τ values at the tropical oceans from all models are considerably lower than those from all satellite products [Penner et al., 2001; Kinne et al., 2003]. These studies (including this one) suffer, however, from a lack of direct measurements from AERONET and other instruments in the tropical oceans. Evidently, more direct measurements over the clean tropical/subtropical oceans are needed to help resolve the model-satellite discrepancies.

The world-wide AERONET aerosol measurement data are invaluable for model evaluations. Because AERONET represents most major aerosol regimes around the globe, the data are very helpful for statistical evaluations. Although the AERONET data are “point” measurements which are not necessarily representative of the 2° grid model results, the daily averaged data should be relatively unbiased. The spatial inhomogeneity in the 2° model grid should be much reduced by the time averaging, except near emission sources that have very large spatial and temporal variations. Even though we have shown that the model successfully reproduces aircraft measured dust concentrations downwind of the Yellow Sea and the Sea of Japan [Chin et al., 2003], the dust τ from the model is different by a factor of two as compared to AERONET valued in the Asian dust source region (one-half Dunhuang but double in Dalanzadgad). This discrepancy reflects not only the difficulties in simulating inhomogeneous dust at the source by a relatively coarse resolution model, but also the need to improve the dust emission parameters

within the model. For regions where aerosol emission is relatively constant and well known, such as North America, the model simulations are quite accurate and can be used quantitatively.

The AERONET data are very useful in evaluating errors and biases in both the satellite and the model products. These direct measurement data can help improve the satellite retrieval algorithms and the model's physical processes, and resolve discrepancies between their corresponding products. Although the comparison between the $1^\circ \times 1^\circ$ gridded MODIS retrieval and the daytime averaged AERONET data is not optimal for satellite data validation (the MODIS validation has been performed using the 10-km resolution data, e.g. Remer et al., 2004), the persistent high bias of MODIS retrieval over some land regions shown in this study is beyond the issue of resolution comparability. Instead, it reflects a general difficulty in MODIS land retrieval. This is highly relevant to some recent "aerosol assimilation" efforts that integrate satellite products with global model simulations to better describe the global aerosol distributions [e.g., Collins et al., 2001; Yu et al., 2003]. We argue that the AERONET data should be an important part in generating such integrated products, in order to reduce large errors or biases in the satellite retrievals or model results in the integrated system. This is especially appropriate over surfaces that have complicated physical and optical properties.

We have emphasized the use of statistical parameters to quantitatively address the degree of agreement between the model and data. The quantitative evaluation is particularly important when a model is used to estimate unavailable or immeasurable quantities (e.g., anthropogenic contributions and intercontinental transport fluxes) and to project future atmospheric changes. "Eyeball" verifications that look the model and data side by side are valid for demonstrations and qualitative judgment, but they are subject to individual interpretations and are not quantitative, while as shown in this study a few statistical parameters (e.g. *HERBS*) generate much more

effective and insightful evaluations. While similarities and differences are compounded in the skill scores, they are revealed individually in the bias, error, and correlation analysis. Given the differences in the spatial and temporal resolutions among AERONET, MODIS, and the GOCART model that make point-by-point comparisons uncertain, histogram is especially appropriate in assessing the behavior of each dataset on a common ground. Our more extensive comparisons and analyses of the AERONET, MODIS, and the model including other seasons and the southern hemisphere are currently underway for a more comprehensive assessment.

6. Conclusions

We have compared the GOCART model simulated aerosol optical thickness τ at 550 nm with the AERONET sunphotometer data and the MODIS satellite retrievals for the ACE-Asia period of April 2001. These comparisons are made not only for the dust and pollution source regions in Asia and its immediate downwind regions, which comprised the ACE-Asia measurement area, but also for other regions of North/South America, Europe, Africa, Middle East, and oceans in the northern hemisphere that are connected by long-range transport. This exercise has produced a quantitative assessment of the model's performance and credibility in estimating the impact of aerosols originating from different source regions on the global atmosphere.

We have used a set of statistical parameters, including histograms H , root mean square error E , correlation coefficient R , mean bias B , and skill scores S (*HERBS*), to evaluate the model and quantify the similarities and differences between the datasets and the model results. We have shown that these statistical parameters can provide effective and insightful evaluations, which are especially helpful when the differences in the spatial and temporal resolutions among different datasets make point-by-point comparisons uncertain. We recommend that these

statistical methods be more widely used in model evaluation, data validation, and in the intercomparisons of models and data.

Both MODIS and the model have shown relatively high τ near the source regions such as eastern Asia, Europe, and northern Africa, and are consistent regarding the major features of the long-range transport of aerosols from their source regions to the neighboring oceans. The probability distributions (or normalized histograms) between the τ values from MODIS and from the model are very similar for the northern part (north of 30°N) of the oceans, but they are a factor of 2 apart for the tropical/subtropical oceans (model being lower). Over land, the distributions of τ from the MODIS and the model are very similar over Europe, but the model is considerably lower than the MODIS in other land regions, especially over North America where model is a factor of 2 – 3 lower than the MODIS. Comparisons with the AERONET measurements have demonstrated that in general the model and AERONET has comparable values and similar probability distributions of τ , but MODIS has a high bias especially in America (greater than a factor of 2), which is largely attributed to the difficulties in the MODIS retrieval dealing with the high surface reflectance at the mountaintop, arid areas, and snow/ice melting places during the spring. Further improvements of the MODIS land retrieval that applies a more rigorous snow/ice mask and deals more effectively with land-cover type dependent surface reflectance are needed to reduce the bias. The discrepancy in the tropical/subtropical regions remains to be resolved, as the comparisons with a few available AERONET sites in this region are still inconclusive, although it is possible that the model has underestimated the sources or overestimated the sinks in the tropics. Since such discrepancy is common between global models and satellite data, more direct measurements focusing on the tropical/subtropical regions

are needed. In general, the model has shown no systematic bias against the AERONET data whereas the MODIS is about 60% too high compared to AERONET in spring 2001.

We have applied the model results to estimate the composition of aerosols in spring 2001 and the anthropogenic contributions. On average, sulfate, carbonaceous, dust, and sea-salt comprise 30%, 25%, 32%, and 13% respectively of the 550-nm τ in April 2001 in the northern hemisphere. Over land, dust and sulfate are the major contributors to the total τ in the extra-tropical region, while carbonaceous aerosol is the most significant component over the subtropical and tropical area in southern Asia and Central America, mainly from biomass burning. Over oceans, dust dominates the tropical/subtropical North Atlantic, sea-salt controls the equatorial North Pacific, and carbonaceous aerosol affects the subtropical North Pacific and Indian Ocean. Dust also influences a substantial area in the extra-tropical North Pacific and North Atlantic. The model estimates that anthropogenic aerosols contribute, on average, nearly 46% to the τ at 550 nm in the northern hemisphere in April 2001. Anthropogenic activities account for more than 50% of the τ over substantial land areas in Asia, North/central America, Europe and Eurasia, and subtropical western North Pacific and Indian oceans, and 20 – 40% over rest of the oceans. Although the fine mode aerosol fractions retrieved from MODIS can potentially be used to derive information on anthropogenic contributions, the model shows that on average the τ at 550 nm from fine mode aerosol is about 20% higher than that from anthropogenic aerosols in the northern hemisphere during April 2001, because of the natural sources of sulfate and OC and the submicron dust and sea-salt that contribute to 30% of total fine mode aerosols.

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References

- Chin, M., R. B. Rood, S.-J. Lin, J.-F. Müller, and A. M. Thomson, Atmospheric sulfur cycle in the global model GOCART: Model description and global properties, *J. Geophys. Res.*, *105*, 24,661-24,687, 2000a.
- Chin, M., D. Savoie, B. J. Huebert, A. R. Bandy, D. C. Thornton, T. S. Bates, P. K. Quinn, E. S. Saltzman, and W. J. De Bruyn, Atmospheric sulfur cycle in the global model GOCART: Comparison with field observations and regional budgets, *J. Geophys. Res.*, *105*, 24,689-24,712, 2000b.
- Chin, M., P. Ginoux, S. Kinne, O. Torres, B. N. Holben, B. N. Duncan, R. V. Martin, J. A. Logan, A. Higurashi, and T. Nakajima, Tropospheric aerosol optical thickness from the GOCART model and comparisons with satellite and sunphotometer measurements, *J. Atmos. Sci.*, *59*, 461-483, 2002.
- Chin, M., P. Ginoux, R. Lucchesi, B. Huebert, R. Weber, T. Anderson, S. Masonis, B. Blomquist, A. Bandy, and D. Thornton., A global model forecast for the ACE-Asia field experiment, *J. Geophys. Res.*, *108(D23)*, doi:10/1029, 2003JD003642, 2003.
- Christopher, S. A., J. Zhang, Y. J. Kaufman, and R. A. Remer, Measurement-based assessment of aerosol radiative forcing over the global oceans, submitted to *Science*, 2004.
- Chu, D. A., Y. J. Kaufman, L. A. Remer, D. Tanré, and B. N. Holben, Remote sensing of smoke

- from MODIS airborne simulator during the SCAR-B experiment, *J. Geophys. Res.*, *103*, 31,979-31,988, 1998.
- Chu, D. A., Y. J. Kaufman, C. Ichoku, L. A. Remer, D. Tanré, and B. N. Holben, Validation of MODIS aerosol optical depth retrieval over land, *Geophys. Lett.*, *29*(12), doi:10.1029/2001GL013205, 2002.
- Chu, D. A., Y. J. Kaufman, G. Zibordi, J.-D. Chern, J.-M. Mao, C. Li, and B. N. Holben, Global monitoring of air pollution over land from EOS-Terra MODIS, *J. Geophys. Res.*, *108*, 4661, doi:10.1029/2002JD003179, 2003.
- Chu, D. A., et al., Characterization of aerosol properties by MODIS during ACE-Asia experiment, submitted to *J. Geophys. Res.*, 2004.
- Collins, W. D., P. J. Rasch, B. E. Eaton, B. V. Khattatov, J.-F. Lamarque, and C. S. Zender, Simulating aerosols using a chemical transport model with assimilation of satellite aerosol retrievals: Methodology for INDOEX, *J. Geophys. Res.*, *106*, 7313-7336, 2001.
- Cooke, W. F., C. Liousse, H. Cachier, and J. Feichter, Construction of a 1x1 fossil fuel emission data set for carbonaceous aerosol and implementation and radiative impact in the ECHAM4 model, *J. Geophys. Res.*, *104*, 22,137-22,162, 1999.
- Dubovik, O., B. N. Holben, T. F. Eck, A. Smirnov, Y. J. Kaufman, M. D. King, D. Tanré, and I. Slutsker, Variability of absorption and optical properties of key aerosol types observed in worldwide locations, *J. Atm. Sci.*, *59*, 590-608, 2002.
- Duncan, B. N., R. V. Martin, A. C. Staudt, R. Yevich, J. A. Logan, Interannual and Seasonal Variability of Biomass Burning Emissions Constrained by Satellite Observations, *J. Geophys. Res.*, *108*(D2), 4040, doi:10.1029/2002JD002378, 2003.
- Eck, T. F., B. N. Holben, J. S. Reid, O. Dubovik, A. Smirnov, N. T. O'Neill, I. Slutsker, and S. Kinne, 1999: Wavelength dependence of the optical depth of biomass burning, urban and desert dust aerosols, *J. Geophys. Res.*, *104*, 31,333-31,350, 1999.

- Ginoux, P., M. Chin, I. Tegen, J. Prospero, B. Holben, O. Dubovik, and S.-J. Lin, Sources and global distributions of dust aerosols simulated with the GOCART model, *J. Geophys. Res.* *106*, 20,255-20,273, 2001.
- Ginoux, P., J. Prospero, O. Torres, and M. Chin, Long-term simulation of dust distribution with the GOCART model: Correlation with the North Atlantic Oscillation, *Environ. Modeling and Software*, *19*, 113-128, 2004.
- Hansen, J., M. Sato, R. Ruedy, A. Lacis, and V. Oinas, Global warming in the twenty-first century: an alternative scenario, *Proc. Natl. Acad. Sci.*, *97*, 9875-9800, 2000.
- Heald, C. L., D. J. Jacob, P. I. Palmer, M. J. Evans, G. W. Sachse, H. B. Singh, and D. J. Blake, Biomass burning emission inventory with daily resolution: Application to aircraft observations of Asian outflow, *J. Geophys. Res.*, *108(D21)*, 8811, doi:10.1029/2002JD003082, 2003.
- Holben B. N., et al., AERONET - A federated instrument network and data archive for aerosol characterization, *Rem. Sens. Environ.*, *66*, 1-16, 1998.
- Holben, B. N., et al., An emerging ground-based aerosol climatology: Aerosol Optical Depth from aerosol climatology: Aerosol Optical Depth from AERONET, *J. Geophys. Res.*, *106*, 12,067-12,097, 2001.
- Huebert, B., T. Bates, P. B. Russell, G. Shi, Y. J. Kim, K. Kawamura, G. Carmichael, and T. Nakajima, An overview of ACE-Asia: Strategies for quantifying the relationships between Asian aerosols and their climate impacts, *J. Geophys. Res.*, *108 (D23)*, 8633, doi:10.1029/2003JD003500, 2003.
- Jacobson, M. Z., Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols, *Nature*, *409*, 695-697, 2001.
- Kaufman, Y. J., D. Tanré, L. A. Remer, E. Vermote, A. Chu, and B. N. Holben, Operational remote sensing of tropospheric aerosol over the land from EOS moderate resolution imaging

- spectroradiometer, *J. Geophys. Res.*, *102*, 17,015-17,061, 1997a.
- Kaufman, Y. J., A. Wald, L. A. Remer, B.-C. Gao, R.-R. Li, and L. Flynn, The MODIS 2.1 μm channel – correlation with visible reflectance for use in remote sensing of aerosol, *IEEE Trans. Geosci. Remote Sens.*, *35*, 1286-1298, 1997b.
- Kaufman, Y. J., D. Tanré, and O. Boucher, A satellite view of aerosols in the climate system, *Nature*, *419*, 215-223, 2002.
- King, M. D., Y. J. Kaufman, D. Tanré, and T. Nakajima, Remote sensing of tropospheric aerosols from space: Past, present, and future, *B. Am. Meteor. Soc.*, *80*, 2229-2259, 1999.
- Kinne, S. et al., Monthly averages of aerosol properties: A global comparison among models, satellite data, and AERONET ground data, *J. Geophys. Res.*, *108(D20)*, 4634, doi:10.1029/2001JD001253, 2003.
- Lesins, G., P. Chylek, and U. Lohmann, A study of internal and external mixing scenarios and its effect on aerosol optical properties and direct radiative forcing, *J. Geophys. Res.*, *107*, 4094, doi:10.1029/2001JD000973, 2002.
- Levy, R. C., L. A. Remer, D. Tanré, Y. J. Kaufman, C. Ichoku, B. N. Holben, J. M. Livingston, P. B. Russell, and H. Maring, Evaluation of the Moderate-Resolution Imaging Spectroradiometer (MODIS) retrievals of dust aerosol over the ocean during PRIDE, *J. Geophys. Res.*, *108(D19)*, 8594, doi:10.1029/2002JD002460, 2003.
- Levy, R. C., L. A. Remer, J. V. Martins, Y. J. Kaufman, A. Plana-Fattori, J. Redemann, P. B. Russell, and B. Wenny, Evaluation of the MODIS aerosol retrievals over ocean and land during CLAMS, *J. Atmos. Sci.*, in press, 2004.
- Nakićenović, N., et al., Emissions scenario, *A Special Report of Working Group III of the Intergovernmental Panel on Climate Change*, Cambridge University Press., 599 pp., 2000.
- O'Neill, N. T., A. Ignatov, B. N. Holben, and T. F. Eck, The lognormal distribution as a reference for reporting aerosol optical depth statistics; Empirical tests using multi-year,

- multi-site AERONET sunphotometer data, *Geophys. Lett.*, 27, 3333-3336, 2000.
- Penner, J. S. et al., Aerosols and their direct and indirect effects, Chapter 5, *Intergovernmental Panel on Climate Change*, Cambridge University Press, 2001.
- Remer, L. A., et al., Validation of MODIS aerosol retrieval over ocean, *Geophys. Res. Lett.*, 29(12), doi:10.1029/2001GL013204, 2002.
- Remer, L. A., et al., The MODIS aerosol algorithm, products and validation, *J. Atmos. Sci.*, in press, 2004.
- Smirnov, A., B. N. Holben, T. F. Eck, O. Dubovik, and I. Slutsker, Cloud screening and quality control algorithms for the AERONET database, *Rem.Sens.Env.*, 73, 337-349, 2000.
- Streets, D., et al., an inventory of gaseous and primary aerosol emissions in Asia in the year 2000, *J. Geophys. Res.*, 108 (D21), 8809, doi:10.1029/2002JD003093, 2003.
- Taylor, K. E., Summarizing multiple aspects of model performance in a single diagram, *J. Geophys. Res.*, 106, 7183-7192, 2001.
- Tanré, D., Y. J. Kaufman, M. Herman, and S. Mattoo, Remote sensing of aerosol properties over oceans using the MODIS/EOS spectral radiances, *J. Geophys. Res.*, 102, 16,971-16,988, 1997.
- Tegen, I., M. Werner, S. P. Harrison, and K. E. Kohfeld, Relative importance of climate and land use in determining present and future global soil dust emission, *Geophys. Res. Lett.*, 31, L05105, doi:10.1029/2003GL019216, 2004.
- Yu, H., R.E. Dickinson, M. Chin, Y.J. Kaufman, B.N. Holben, I.V. Geogdzhayev, and M.I. Mishchenko, Annual cycle of global distribution of aerosol optical depth from integration of MODIS retrievals and GOCART model simulations, *J. Geophys. Res.*, 108(D3), 4128, doi:10.1029/2002JD02717, 2003.

Table 1. Site number, name, location, and principal investigator of the 57 AERONET sites.

#	Site Name	Lat.	Long.	Principal Investigator
<i>Asia:</i>				
1	Dunhuang	40.04°N	94.79°E	Brent Holben
2	Inner Mongolia	42.68°N	115.95°E	Brent Holben
3	Dalanzadgad	43.58°N	104.42°E	Brent Holben
4	Beijing	39.98°N	116.38°E	Philippe Gouloub / Hongbin Chen
5	XiangHe	39.75°N	116.96°E	Brent Holben
6	Je-Ju	33.28°N	126.17°E	Brent Holben
7	Anmyon	36.52°N	126.32°E	Brent Holben / Chuck McClain
8	Shirahama	33.69°N	135.36°E	Brent Holben
9	Noto	37.33°N	137.14°E	Itaru Sano
10	Taiwan	24.90°N	121.10°E	Gin-Rong Liu
11	Okinawa	26.36°N	127.77°E	Brent Holben
12	Kanpur	26.45°N	80.35°E	Brent Holben / Ramesh Singh
<i>America:</i>				
13	Saturn Island	48.78°N	123.13°W	Norm O'Neill
14	Rimrock	46.49°N	116.99°W	Brent Holben
15	Missoula	46.92°N	114.08°W	Wei-Min Hao
16	San Nicolas	33.26°N	119.49°W	Robert Frouin
17	Rogers Dry Lake	34.93°N	117.89°W	Jeannette Vandenbosch
18	La Jolla	32.87°N	117.25°W	Robert Frouin
19	Maricopa	33.07°N	111.97°W	Brent Holben
20	Tucson	32.23°N	110.95°W	Kurt Thome
21	Sevilleta	34.35°N	106.89°W	Doug Moore
22	Cart Site	36.61°N	97.41°W	Mary Jane Bartholomew
23	KONZA_EDC	39.10°N	96.61°W	David Meyer
24	Bondville	40.05°N	88.37°W	Brent Holben
25	Walker Branch	35.96°N	84.29°W	Brent Holben
26	GSFC	39.03°N	76.88°W	Brent Holben
27	MD Science Center	39.28°N	76.62°W	Brent Holben
28	COVE	36.90°N	75.71°W	Brent Holben
29	Egbert	44.23°N	79.75°W	Norm O'Neill
30	CARTEL	45.38°N	71.93°W	Alain Royer / Norm O'Neill
31	Howland	45.20°N	68.73°W	Brent Holben
32	Stennis	30.37°N	89.62°W	Dean Noel
33	Dry Tortugas	24.60°N	82.80°W	Kenneth J. Voss
34	Mexico City	19.33°N	99.18°W	Brent Holben
35	Surinam	5.80°N	55.20°W	Brent Holben
<i>Europe:</i>				
36	Avignon	43.93°N	4.88°E	Michel Verbrugghe
37	IMC Oristano	39.91°N	8.50°E	Didier Tanré
38	Ispra	45.80°N	8.63°E	Giuseppe Zibordi
39	Venise	45.31°N	12.51°E	Giuseppe Zibordi
40	Rome Tor Vergata	41.84°N	12.65°E	Gian Paolo Gobbi
41	SMHI	58.58°N	16.15°E	Bertil Hakansson
42	Bucarest	44.45°N	26.52°E	Didier Tanré

<i>Africa:</i>				
43	Ouagadougou	12.20°N	1.40°W	Didier Tanré
44	Ilorin	8.32°N	4.34°E	Rachel T. Pinker
45	THALA	35.55°N	8.68°E	Brent Holben
<i>Middle East:</i>				
46	IMS-METU-ERDEMLI	36.56°N	34.26°E	Brent Holben
47	Nes Ziona	31.92°N	34.79°E	Brent Holben
48	Sede Boker	30.52°N	34.47°E	Brent Holben
49	Solar Village	24.91°N	46.41°E	Brent Holben
<i>Ocean:</i>				
50	Midway Island	28.21°N	177.38°W	Brent Holben
51	Coconut Island	21.43°N	157.79°W	Chuck McClain
52	Lanai	20.74°N	156.92°W	Chuck McClain
53	La Paguera	17.97°N	67.04°W	Brent Holben
54	Bermuda	32.37°N	64.70°W	Brent Holben
55	Azores	38.53°N	28.63°W	Brent Holben
56	Cape Verde	16.73°N	22.93°W	Didier Tanré
57	Male	4.19°N	73.53°E	Brent Holben

Figure captions

Figure 1. Emissions of sulfur (SO_2 , DMS, and sulfate), carbonaceous (OC and BC), dust, and sea-salt in the northern hemisphere in April 2001 used in the model.

Figure 2. Northern hemispheric distributions of (a) 550-nm aerosol optical thickness (τ) and (b) fine model fraction (f_τ) from the MODIS and the model for April 2001, averaged over the locations and days that MODIS measurements are available which are shown in (c).

Figure 3. Probability distribution of MODIS (dotted lines) and model (solid lines) products of aerosol optical thickness τ over (a) ocean and (b) land, and the fine mode fraction f_τ over (c) ocean and (d) land. The latitude and longitude borders for each sub-region are indicated on each panel. The most probable (or modal) values of MODIS (M) and GOCART (G) for each region are shown in the corresponding panel.

Figure 4. Locations of 57 AERONET sites in the northern hemisphere with > 3 days of measurements available during April 2001 (see Table 1 for details). Superimposed is the model calculated monthly average τ at 550 nm for April 2001.

Figure 5. Daily τ at 550 nm from AERONET, MODIS, and the model at 12 AERONET sites in April 2001. Vertical bars: AERONET data; grey circles: MODIS data; lines and shaded areas: GOCART model results with aerosol compositions. Statistical parameters of mean bias (B), root mean square error (E), correlation coefficient (R), and skill score (S) between the GOCART model and AERONET (G-A) or GOCART and MODIS (G-M) are listed on each panel.

Figure 6. Mean bias B (top panel), root mean square error E (2nd panel), correlation coefficients R (3rd panel), and skill score S (4th panel) between the model and AERONET (black vertical bars) or MODIS (grey vertical bars). The number of observation days in April 2001 for

AERONET (black) and MODIS (grey) is shown in the bottom panel. Dashed lines in the top panel are $B = 2$ and $B = 0.5$ lines, i.e, a factor of two bias.

Figure 7. Probability distribution of daily τ at 550 nm in April 2001 from AERONET (dashed lines), MODIS (dotted lines), and the model (solid lines) at the AERONET sites located in (a) Asia, (b) America (North America and Surinam), (c) Europe, Africa, and Middle East, and (d) islands in the oceans. The most probable (or modal) values of AERONET (A), MODIS (M), and GOCART (G) for each region are shown in the corresponding panel.

Figure 8. Monthly averaged τ at 550 nm for April 2001 at 57 AERONET sites. Data are averaged for the days when both AERONET and MODIS observations are available, except at sites #1, #2, #48, and #49 where no available MODIS data. Statistical parameters of mean bias B , root mean square error E , correlation coefficient R , and skill score S for each regions of Asia, America, Europe/Africa/Middle East, and oceans are also listed. “M-A” is for MODIS vs. AERONET, and “G-A” for GOCART vs. AERONET.

Figure 9. Model calculated percentage contributions of (a) sulfate, (b) carbonaceous, (c) dust, (d) sea-salt, (e) anthropogenic, and (f) fine mode aerosols to the monthly averaged τ at 550 nm for April 2001 in the northern hemisphere.

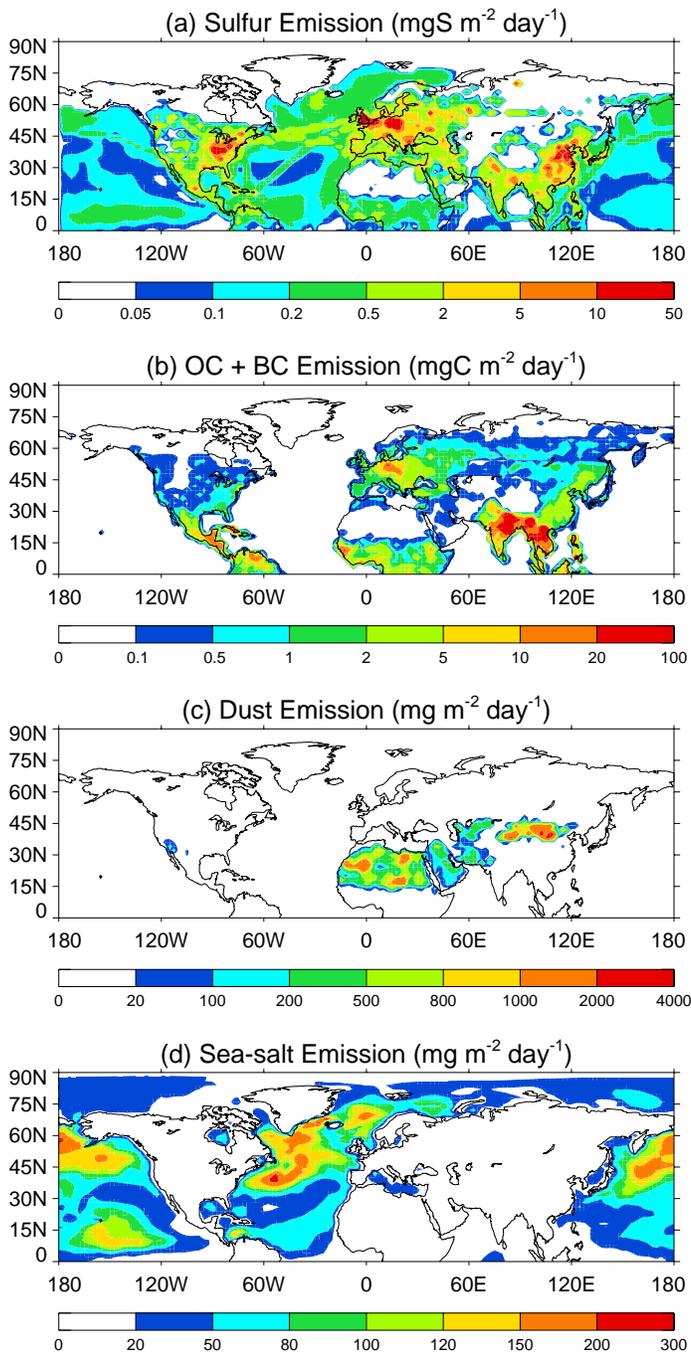
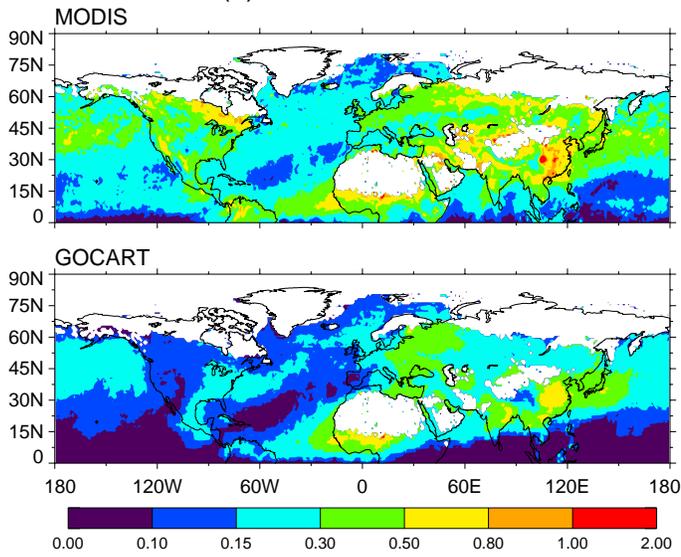
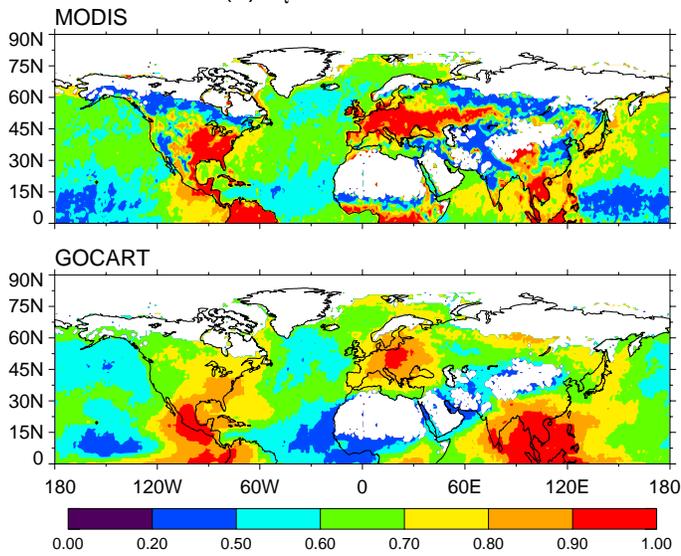


Figure 1

(a) τ at 550 nm 200104



(b) f_τ at 550nm 200104



(c) Available days 200104

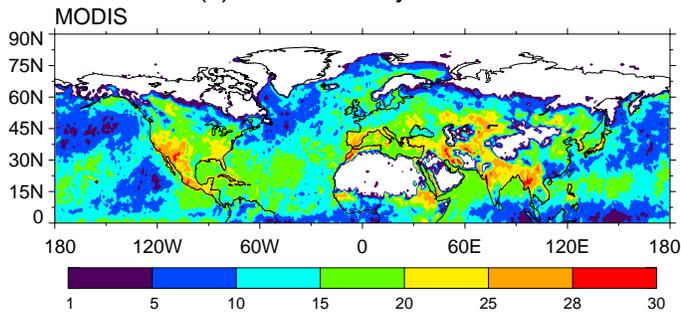


Figure 2

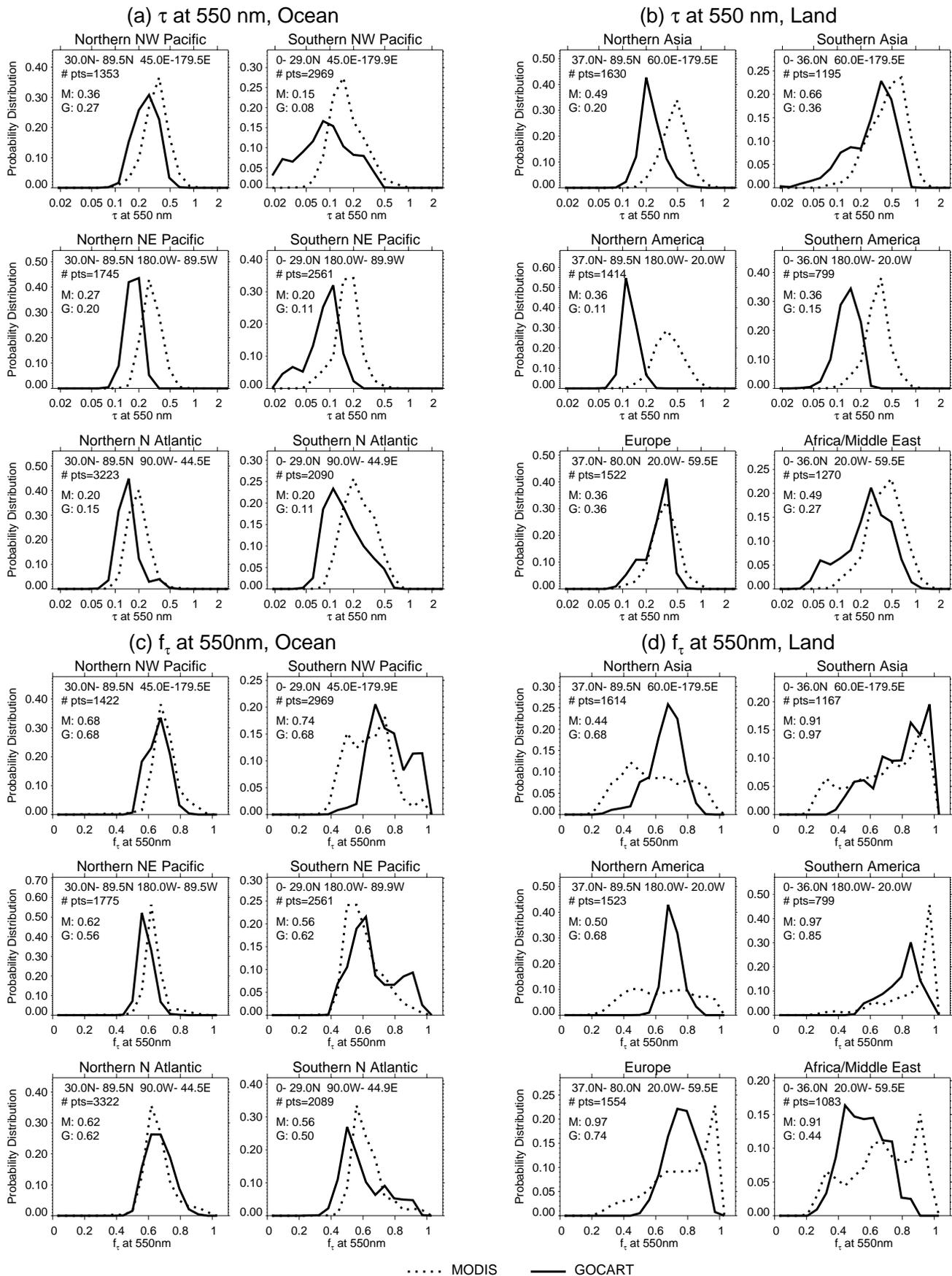


Figure 3

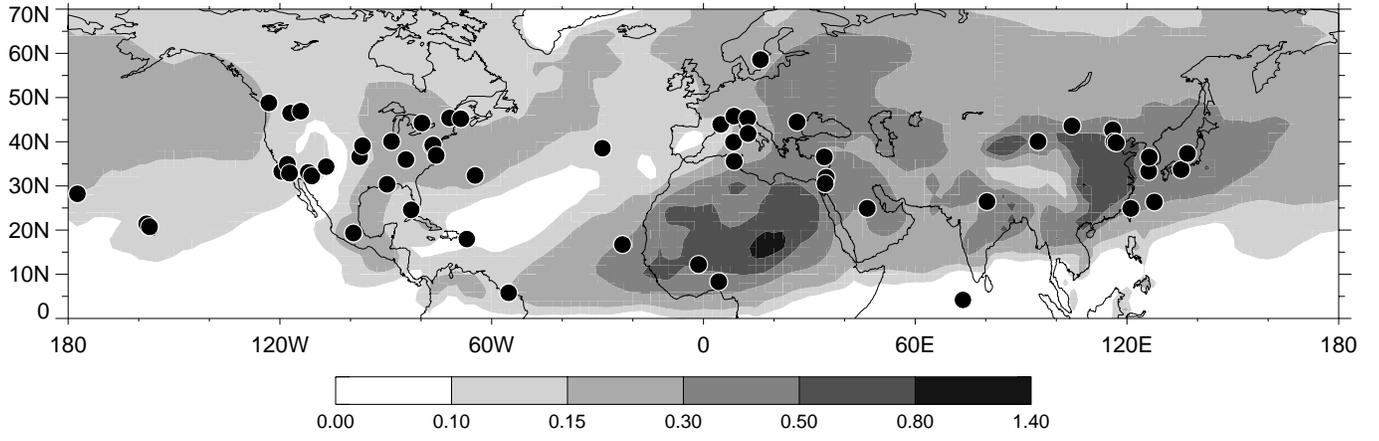


Figure 4

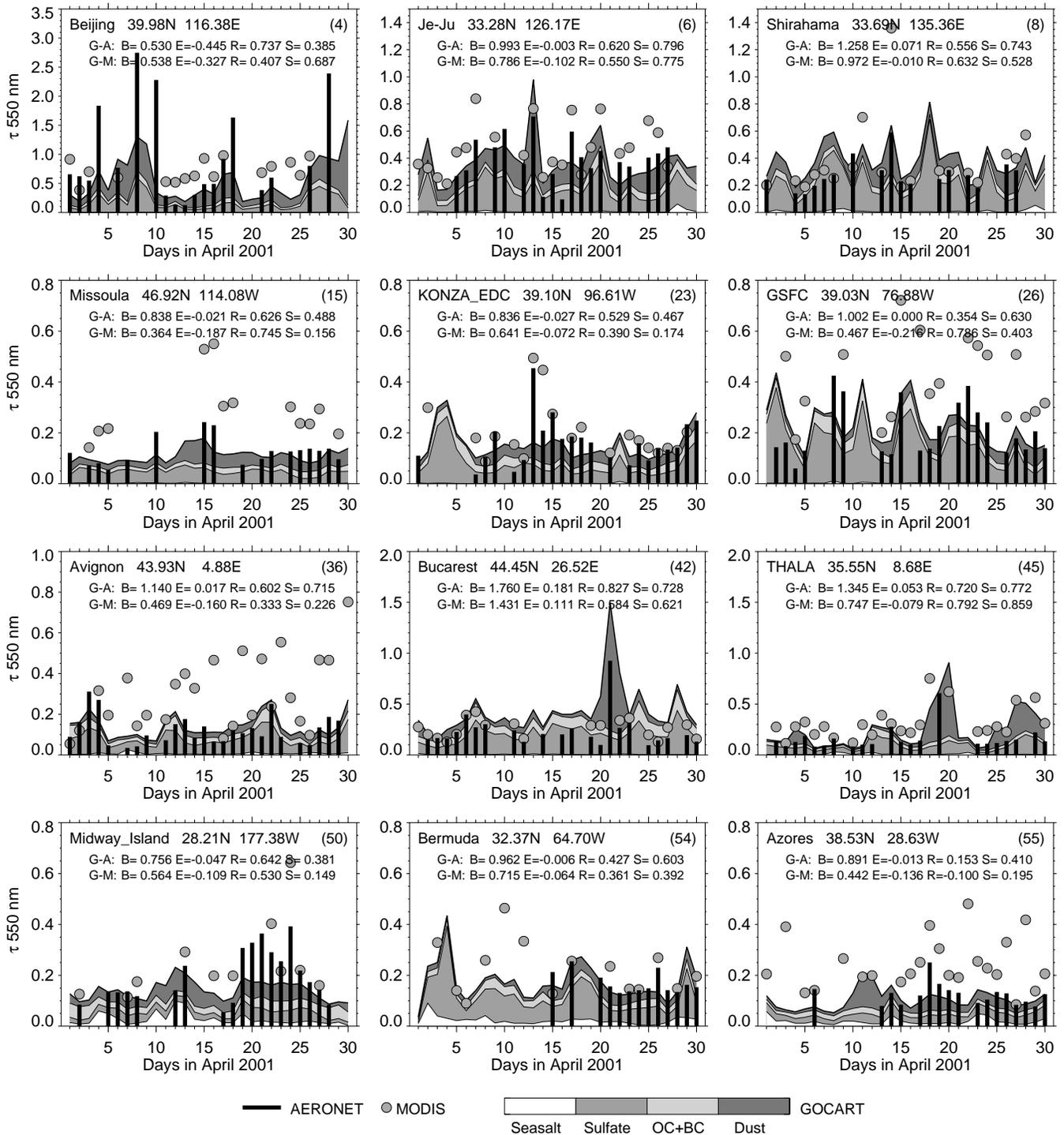


Figure 5

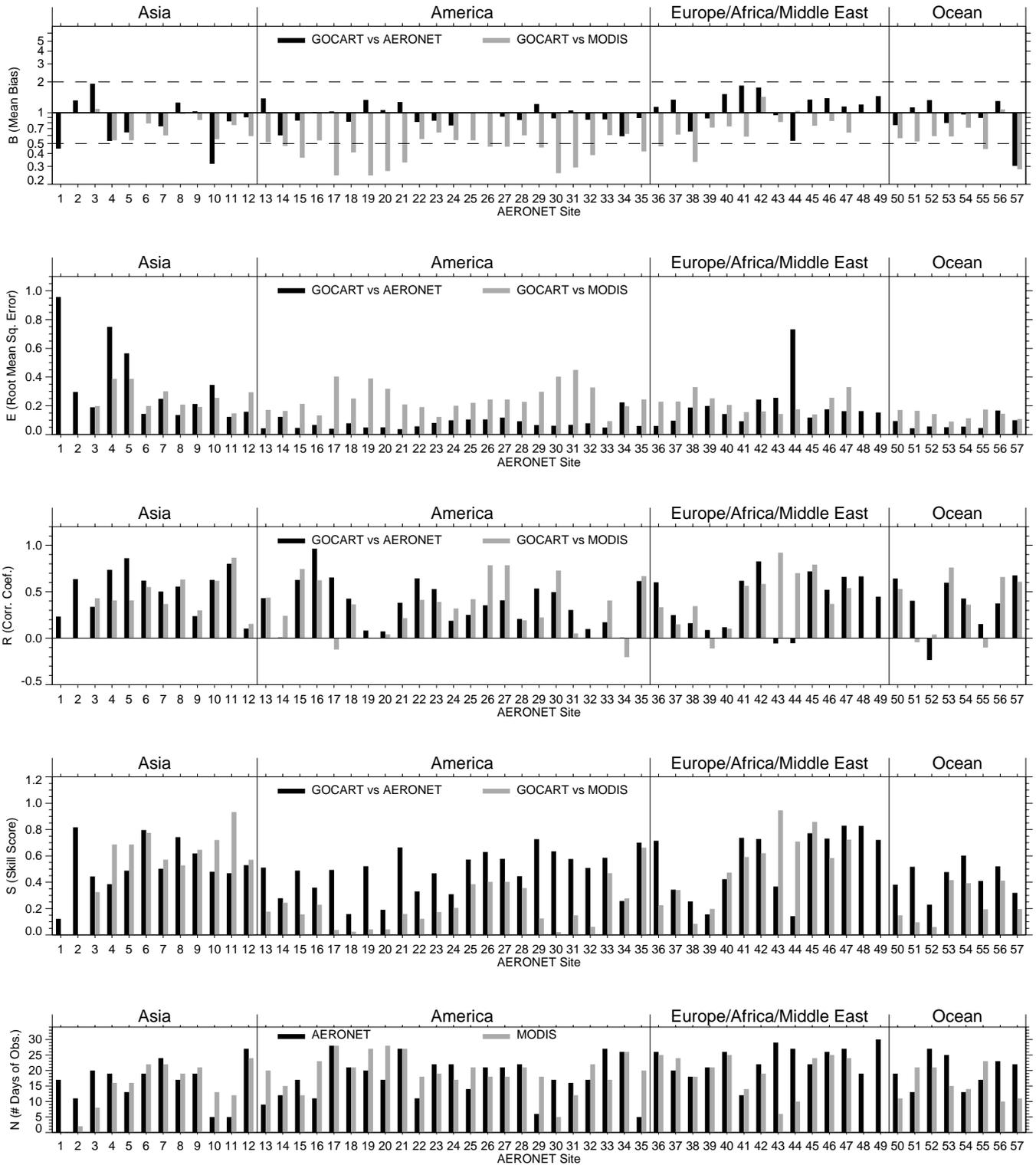


Figure 6

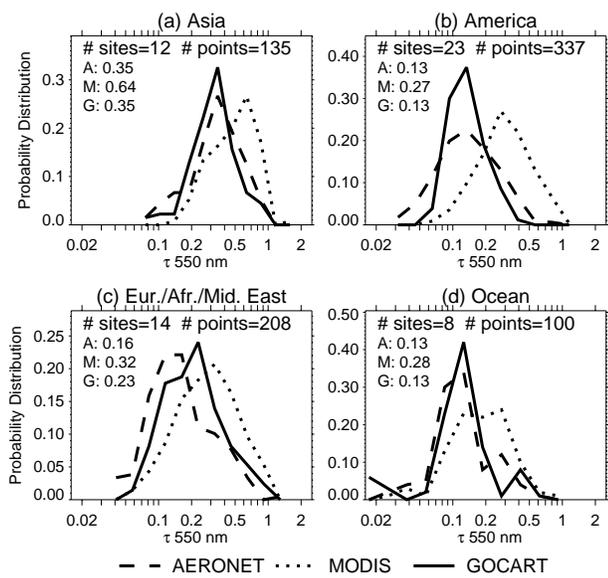


Figure 7

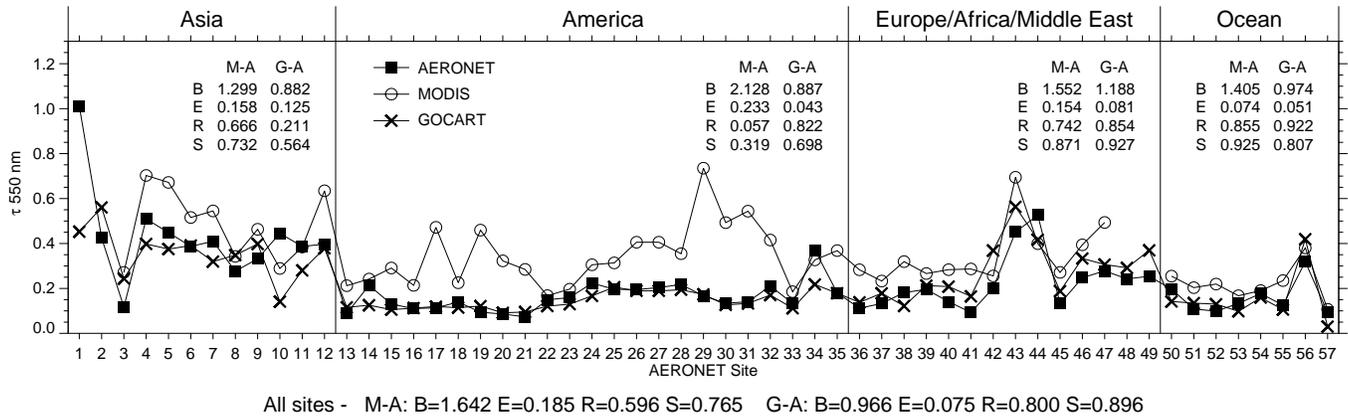


Figure 8

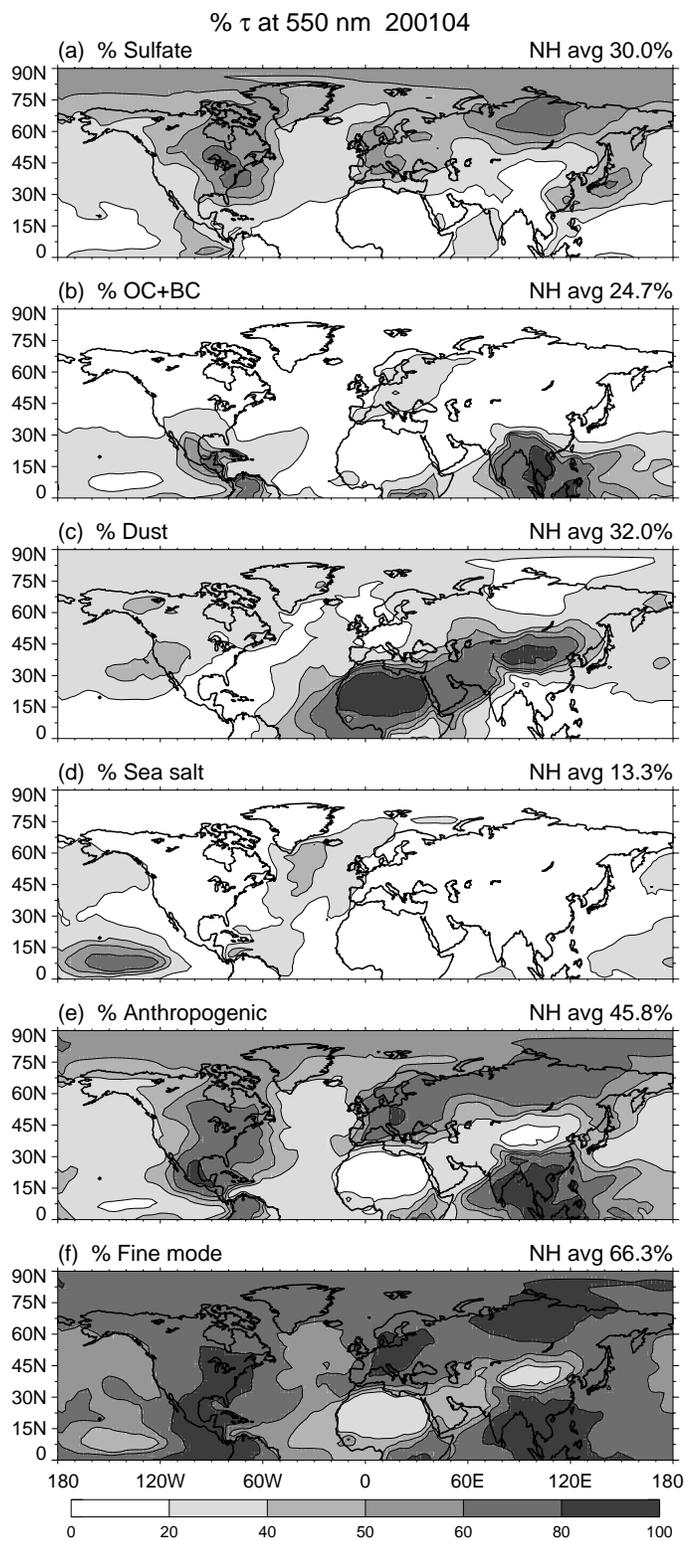


Figure 9