GCM Aerosol Radiative Effects Using Geographically Varying Aerosol Sizes Deduced from AERONET Measurements

GLEN LESINS AND ULRIKE LOHMANN

Department of Physics and Atmospheric Science, Dalhousie University, Halifax, Nova Scotia, Canada

(Manuscript received 15 August 2002, in final form 15 April 2003)

ABSTRACT

Aerosol optical properties, and hence the direct radiative effects, are largely determined by the assumed aerosol size distribution. In order to relax the fixed aerosol size constraint commonly used in general circulation models (GCMs), measurements from the Aerosol Robotic Network (AERONET) and a new method to deduce a geographically and monthly varying aerosol size are used. Within the limitations of the GCMs’ ability to predict aerosol mass concentrations, which are shown to be reasonable, the globally averaged modal radius of an internally mixed fine mode that best matches the AERONET-deduced Ångström exponent curve is found to be 0.04 μm. This corresponds to a direct top-of-the-atmosphere total aerosol solar forcing in clear skies computed using monthly averages of -2.2 W m⁻², which is about 0.5 W m⁻² greater than assuming a globally constant modal radius of 0.04 μm.

1. Introduction

Aerosols in cloud-free air scatter and absorb solar radiation altering the shortwave transmission through the atmospheric column. The aerosol radiative direct effect (Chylek and Coakley 1974; Charlson et al. 1992) has contributions from the enhanced backscattering to space, which cools the atmosphere and the earth’s surface, and the enhanced absorption of solar radiation which heats the aerosol layer but cools below the layer. Model estimates of the direct radiative effect depend on a number of assumptions that must be made concerning the aerosol type and size. The aerosol optical properties are very sensitive to the size distribution and hence reliable model results are dependent on an accurate aerosol modal radius. In this paper we propose a method to help narrow the uncertainty in the general circulation model (GCM) aerosol size assumption by using the Aerosol Robotic Network (AERONET) surface observations of the aerosol Ångström exponent to create a plausible geographically and monthly varying fine aerosol modal radius to improve the predicted direct radiative effect.

Numerous global climate model studies have examined the optical properties of aerosols and their radiative effects. They typically impose a dry aerosol size distribution since size prognostic models are computationally expensive for GCMs. These sizes can vary for models with multiple externally mixed modes, but are typically fixed in space and time. Lohmann et al. (1999b) uses nine lognormal aerosol types derived from the dataset of Hess et al. (1998). Grant et al. (1999) used a fixed sulfate mode and three fixed carbonaceous modes in examining sulfate and biomass burning aerosols. Penner et al. (2002) used fixed size assumptions from Chuang et al. (1997) for continental aerosols and Quinn and Coffman (1998) for maritime aerosols. Chin et al. (2002) used the fixed sizes for each external mode from the Global Aerosol Data Set (GADS; Kopke et al. 1997). There are some global models that do have size-resolved aerosols: Gong et al. (1997) has eight size bins for sea salt aerosols, Jacobson (2001) has comprehensive size-resolved aerosols, and Ginoux et al. (2001) has seven size bins for dust aerosols. Some models, such as Tegen et al. (2000), parameterize the aerosol optical depth (AOD) directly from the burden and completely eliminate making an explicit assumption of size; however, an implicit size assumption is made in going from the burden to the AOD. Other models such as Wilson et al. (2001) are increasing the number of prognostic variables that describe the various aerosol modes but are still not explicitly solving for a spatial and temporal varying size. Most model studies deal with aerosol components as being externally mixed and hence the fixed size assumption is slaved to the particular aerosol type. However, most aerosols are in fact internally mixed (e.g., Murphy and Thomson 1997; Middlebrook et al. 1998) making the assignment of fixed model radii to a pure chemical composition somewhat dubious.
In section 2 the GCM model and aerosol characterization is described. In section 3 the GCM aerosol mass concentrations are validated against field observations. In section 4 the seasonal aerosol optical depths predicted by the GCM are compared against AERONET observations. In section 5 the new method of comparing the wavelength-dependent Ångström exponent between the GCM model and AERONET measurements is described and the results are discussed. The final section summarizes the findings.

2. Model description

The global and regional radiative effects of various aerosol size scenarios are studied using the Canadian Climate Centre for Modeling and Analysis (CCCma) GCM run at spectral truncation T48 corresponding to a $3.75^\circ \times 3.75^\circ$ Gaussian grid. Nine aerosol types are tracked as prognostic mass mixing ratios during the simulations. The aerosol dry size distributions are given by lognormal functions for two internally mixed modes for the fine and coarse size classes, which are modified for relative humidity effects by subdividing the distribution into 100 size bins and computing the water uptake exactly using the Kohler equation (Lesins et al. 2002). The aerosol types are sulfate, hydrophobic and hydrophilic black carbon, anthropogenic and natural organic carbon from monoterpene emissions (Guenther et al. 1995), desert dust using monthly mean emission rates (Ginoux et al. 2001), and sea salt where the fluxes are based on the instantaneous surface wind speed (Monahan et al. 1986). Details about the treatment of the various aerosol types and their sources can be found in Lohmann et al. (1999a) for sulfate, Lohmann et al. (1999b) for carbonaceous aerosols, and Lohmann (2002) for dust.

The dry deposition flux to the ground is assumed to be proportional to both the concentration in the lowest model layer (40 m above the ground) and to a prescribed dry deposition velocity that varies with surface type and aerosol type. Removal of SO$_2$ and aerosols by precipitation (in-cloud and below-cloud scavenging) is calculated explicitly in terms of the model’s precipitation formation rate. A bulk cloud microphysical scheme is used with prognostic equations for liquid water and ice water mixing ratio.

The GCM is run with an initial 6 months spin up and then 12 consecutive months are simulated and used to compute monthly means of the aerosol optical properties from which the annual means are determined. The extinction coefficient, single scattering albedo $\omega$ and asymmetry factor $g$ for the various mixing scenarios are computed offline using Mie theory for each grid point in the GCM. The optical depth $\tau$ is computed by vertically integrating the extinction coefficient from the ground to the top of the atmosphere, for various wavelengths.

3. Aerosol validation

The aerosol optical properties and hence the radiative effects are determined by the aerosol size distribution and the mass concentrations of the various components. Hence before attempting to adjust the aerosol dry sizes in order to better match modeled and measured optical properties, it would be prudent to investigate how well the model is able to predict the mass concentrations. The aerosol measurements are provided by Chin et al. (1996); Cooke et al. (1999); Quinn et al. (1996); D. L. Savoie and J. M. Prospero (2001, personal communication) and from the Canadian Air and Precipitation Monitoring Network (CAPMoN) and the Clean Air Status and Trends Network (CAST) datasets (Ro et al. 1997). This section compares the concentrations of various aerosol types.

a. Scatter diagrams for different aerosol types

A comparison between modeled and observed annual averaged surface aerosol concentrations is shown in Fig. 1 for five aerosol chemical species. Overall the model overestimates the sulfate concentrations by about 50%, however the bias is concentration dependent. Between about 500 and 5000 ng m$^{-3}$ there is almost no bias. At larger concentrations, corresponding to polluted continental sites, the model is slightly underestimating the sulfate concentration. This may be because we are comparing the average concentration between the GCM grid box area to a measurement from a single station, which can be located near an urban center where the air will be more polluted. At low concentrations below 500 ng m$^{-3}$ the model significantly overestimates the concentrations over oceanic areas where dimethylsulfide is the main source for the sulfate. This suggests that the DMS exchange rate between the surface ocean and the lower atmosphere may be too large in the model.

For global surface dust the model predicts about 1.9 times the concentration as the observations based on monthly averages. Nevertheless it is a positive result that most of the differences are within a factor of 2 over a remarkably large range of concentrations from 100 to $3 \times 10^4$ ng m$^{-3}$. The sea salt results yield a smaller overall bias, with the model surface concentrations averaging about 34% greater than measurements. However, there are some outliers where the model is underpredicting the sea salt concentration by more than
FIG. 1. Comparison between surface-based aerosol measurements from various global sites and campaigns against the surface concentration predicted by the GCM model for five aerosol types: sea salt, dust, sulfate, black carbon, and organic carbon. Solid lines are shown for exact agreement and factors of 2 on either side. The overall average ratio of model to observed concentration is shown.

An order of magnitude. The limited black carbon measurements shows a large difference from model predictions with the model concentrations averaging about six times larger. Organic carbon surface concentrations from the GCM compare very favorably with measurements compiled by Liousse et al. (1996) and Cooke et al. (1999). The model averages about 1.5 times higher in concentration but good agreement is achieved over several orders of magnitude.

Considering the limitations in the spatial and temporal sampling, and the inherent difficulty in comparing relatively large grid box model predictions with point measurements, the comparison results given in these scatterplots can be considered to be very good and a measure of the model’s success in reproducing the average concentrations for the sulfate, organic carbon, dust and sea salt components. The biases in the CCCma are comparable to those models that participated in the International Panel on Climate Change (IPCC) intercomparison study (Penner et al. 2001).

b. Comparing with Quinn et al. (1996)

Quinn et al. (1996) measured sulfate and sea salt aerosol surface concentrations during Pacific cruises. This field data is an excellent source to compare the values from the GCM since it provides a latitudinal distribution of the aerosols from 60°S to 60°N over portions of the Pacific Ocean from 135°E to 140°W. It is also a valuable observation set since it separates the sulfate and sea salt aerosol components.

In Fig. 2 the comparison is shown for seven latitude bins. Excellent agreement was achieved for the sea salt concentration with typical differences of less than 20%. The largest difference is a model overestimation of about 40% in the 20°–40°N latitude bin. The GCM correctly captures the increase in sea salt concentrations that results from the strong westerlies in the midlatitudes. This favorable comparison is very encouraging since sea salt dominates the contribution towards the total optical depth over the oceans and because oceans...
Fig. 2. Comparison between Pacific sea level measurements of sea salt and sulfate from Quinn et al. (1996; solid line) against the GCM model (dashed line) as a function of latitude averaged from 135°E to 140°W. Also plotted are the 5th, 25th, 50th, 75th, and 95th percentiles from the model.
comprise the majority of the earth’s surface, the global optical properties should be reliable over the oceans. The model agrees very well with the sulfate measurements except from 40°S to 60°S where it overestimates by a factor of 2, probably due to an excess of modeled DMS flux to the atmosphere.

c. Seasonal cycle

The monthly average comparisons between the ground-based sulfate in the GCM and measurements are given in Fig. 3 for 30 stations around the globe. Overall there is very good agreement. The model generally slightly underpredicts the sulfate mixing ratios in the northern mid- and high latitudes and generally overpredicts the ratios in the Tropics and Southern Hemisphere. The model is unable to capture the magnitude of the summertime peak in sulfate concentrations for those northern continental stations that experience hot polluted air. The late winter maxima in sulfate at the Arctic stations Alert, Mould Bay, and Igloolik are not fully captured. This Arctic pollution depends on long transport from Eurasia (Barrie 1996) and strong trapping in the surface-based inversion, features that the GCM may not be completely able to reproduce. This is a common problem in models predicting sulfate (e.g., Feichter et al. 1996; Chin et al. 1996).

The surface sea salt concentrations show mixed results (Fig. 4). Of the 24 stations at least 15 show excellent agreement. The other stations are split between the model overestimating and underestimating the amount, and hence there is not any significant overall global bias. Nevertheless the results shown here are encouraging. The GCM captures the seasonality in the Northern Hemisphere but tends to exaggerate the seasonal variation in the Southern Hemisphere. The surface dust concentrations are reasonably predicted by the GCM for 22 stations (Fig. 5), but here again there are mixed results.

Black carbon surface concentration measurements (Fig. 6) are much more limited. The data collected at Mace Head was divided into marine and continental sectors depending on the wind direction as provided by Cooke et al. (1999). The GCM prediction lies in between these two sets of measurements except for the winter when it underestimates the concentrations. The strong peak in the spring is not evident in the measurements. There is excellent agreement at Barrow but an overestimate at Mauna Loa.

Overall there is good agreement between the model and measurements in the average surface mass concentrations as shown in Fig. 1. This suggests that, at least in a globally averaged sense, we cannot justify ascribing all the error in aerosol optical properties to errors in the mass concentration. Instead we should also consider aerosol size as a contributing factor to biases in the optical properties which would then affect the radiative transfer through the atmosphere. The effect of adjusting the dry aerosol sizes using surface-based sun photometer measurements will now be considered.

4. AERONET aerosol optical depth

The AERONET program consists of a ground-based remote sensing aerosol network that was established to assess aerosol optical properties and validate satellite retrievals of aerosol optical properties (Holben et al. 1998, 2001). The network imposes standardization of instruments, calibration, and processing. Data from this collaboration provides globally distributed observations of spectral aerosol optical depths, inversion products, and precipitable water in geographically diverse aerosol regimes.

The AERONET aerosol optical depths at 500 nm are compared with the GCM values for monthly means (Fig. 7). The AERONET averages are computed assuming both a normal and lognormal distribution. O’Neill et al. (2000) showed that the optical depths follow a lognormal distribution and hence the geometric mean is the more meaningful average to use. Using geometric means yields about 10% lower optical depths than arithmetic means. The nine aerosol types are distributed as internal mixtures into three lognormal modes with the following number modal dry radii: 1) 0.02 µm consisting of sulfate and all carbonaceous mass, 2) 0.2 µm consisting of accumulation mode sea salt and dust, and 3) 2.0 µm consisting of coarse mode sea salt and dust. All three modes have a geometric standard deviation width of 2.0. This simplified size distribution is motivated by the aerosol climatology from Hess et al. (1998) and is intended to provide a baseline for comparisons.

The model prediction agrees well with measurements at Goddard Space Flight Center (GSFC), Greenbelt, Maryland, with the exception of June, July, and August when hot and humid conditions create summer haze conditions with higher optical depths. At this time we see the impact of using the geometric mean for the observations since it significantly lowers the average optical depth bringing it closer to the model value. Excellent agreement was obtained at Sevilleta, New Mexico, and the H. J. Andrews Experimental Forest, Oregon, where the seasonality was captured. Semiarid conditions with limited vegetation are found near Sevilleta, while H. J. Andrews is located close to the forests of the Cascade Mountains. The model does not capture the very high optical depths measured during the dry season at Mongu, Zambia, on the grassy shores of the Zambezi River surrounded by tropical forests. The local biomass burning is a source of aerosols that may undergo a strong interannual variability or that is simply not captured in the emission inventories that may not include recent changes in emission strengths. The annual averaged AOD at Bermuda is in good agreement. This station is in the western North Atlantic where sea salt is a major component but with long range transport of sulfate and dust likely contributing. The model is unable to capture
the springtime peak. At Ilorin in southwest Nigeria, the model agrees well with observations except in the winter months when it is unable to capture the high AOD associated with biomass burning. Excellent agreement is found at Sede Boker in the Negev Desert and Bahrain in the Persian Gulf where dust is the main aerosol type. At Kaashidhoo in the Indian Ocean, the model underestimates the AOD during the winter monsoon period when northern winds transport aerosols from the Indian Subcontinent to the site. At Dalanzadgad, in the Gobi Desert of southern Mongolia, the model generally overpredicts the amount of predominantly dust aerosols. Very good agreement is found at many sites, such as Lanai, Hawaii; San Nicolas, California; Dry Tortugas, Florida; Cloud and Radiation Testbed (CART) site; and Bondville, Illinois. At Waskesiu, Saskatchewan, Cana-
da, the AODs during the summer are underpredicted since forest fires contribute to the aerosol loading then. The GCM does not include any boreal biomass burning sources. Sites such as Alta Floresta and Brasilia, in Brazil; and Los Fieros, in Bolivia, show disagreements during the burning seasons from August to October where much higher AODs are observed. This suggests that the biomass inventories may be underestimated, although local sources that the GCM cannot reproduce may also be responsible. Arica, Chile, is interesting in that the observed AODs are much higher than predicted for the entire year. Arica is on the northern coast of Chile in an extremely arid but narrow region where the local dust source may not be accounted for. In Mauna Loa, Hawaii, a high elevation site, the model overestimates the optical depth, which suggests that the vertical transport of aerosols may be too efficient in the model. This finding is supported by the results from the Comparison of Sulfur Cycle Models (COSAM) study (Lohmann et al. 2001). It was revealed that the CCCma boundary
layer scheme carries sulfate too rapidly away from the source.

5. Narrowing uncertainties in aerosol size

Aerosol size assumptions that depend on climatologies do not include information on the actual state of the aerosols for a given location or time. It would be useful to have a geographical dependence of the aerosol sizes for each month that reflects the annual changes for different locations. Typically the dry size of each aerosol species is fixed for all locations and also for each month. Here we seek to loosen this restriction by allowing the dry aerosol sizes to vary both in space and time. The challenge is to generate such size parameters for the entire globe from observations taken at a limited number of stations.

One of the best continuous global datasets on aerosol properties is provided by the AERONET project. The aerosol optical depth is measured nominally at seven...
wavelengths from the UV to the near-IR (0.34, 0.38, 0.44, 0.50, 0.67, 0.87, and 1.02 μm). The dependence of optical depth \( \tau \) on wavelength \( \lambda \) is largely determined by the size distribution of the aerosols. The Ångström relation is commonly used to parameterize this relationship:

\[
\tau = \tau_1 \lambda^{-\alpha},
\]

where \( \alpha \) is the Ångström exponent and \( \tau_1 \) is the reference optical depth. The exponent will vary from near 0 for very large particles to 4 in the Rayleigh limit of small particles, but its exact value will depend on the convolution of the Mie extinction coefficient with the number concentration size dependence. Atmospheric aerosols typically have exponents in the range from 0 to 2 (e.g., Eck et al. 1999). AERONET radiance measurements of the solar aureole can also be inverted to provide size information but this is beyond the scope of this paper.

The Ångström exponent can be computed with as few as two optical depth measurements at different wavelengths. This is the approach used in satellite remote sensing of aerosol properties where dual channel measurements allow the exponent to be determined (Mishchenko et al. 1999). The AERONET data allows this approach to be extended since seven wavelengths are used. The Ångström exponent is computed as a function of wavelength by a least squares fit to the optical depth using three adjacent wavelengths. The AERONET instrumentation has an uncertainty of 0.01–0.02 in aerosol optical depth (Eck et al. 1999). This translates into a typical maximum error in the Ångström exponent of about 0.15, which could be higher in very low optical depth cases. For the seven wavelengths used in AERONET, five Ångström exponents can be determined as a function of wavelength. This extra information can potentially provide a better determination of the modal sizes of the aerosols (Eck et al. 1999; O’Neill et al. 2001a,b). These curves are computed for each of the 28 AERONET stations used in this study and for each month of the year. The monthly data are averaged over the total observation period which is about 3–5 yr for most of the stations.

Although we demonstrated reasonable agreement in the surface aerosol mass concentrations in Fig. 1, there do exist differences in the aerosol burdens between the model and observations. However, these differences do not influence the value of the Ångström exponent since aerosol optical depths vary linearly with aerosol burden, so that a constant multiplier, due to an error in the mass burden, applied to Eq. (1) will not change the computed exponent. Hence, the method of matching observed and modeled Ångström exponents is insensitive to errors in the burden.

An aerosol size distribution is required before the surface optical depths can be computed from the GCM aerosol mass concentrations for each aerosol type. To study the sensitivity of the size distribution assumption, six different size scenarios are considered. The total aerosol size spectrum is assumed to be comprised of two size modes, a coarse mode with a number mode radius of 2.0 μm that is an internal mixture of the coarse mode sea salt and coarse mode mineral dust components, and a fine mode whose size is varied for the different scenarios, which is an internal mixture of sulfate, carbonaceous and accumulation mode sea salt, and mineral dust. For this calculation only two modes are used in order to minimize the total number of free parameters while still allowing a meaningful size adjustment. Table 1 lists the different modal dry radii for each fine mode scenario, spanning the likely limits of the fine mode. The geometric standard deviation of the lognormal modes is set to 2.0 for both the coarse and fine modes. The actual modeled aerosol size will be larger due to water uptake due to the relative humidity. The Kohler equation is solved exactly for the two internally mixed modes. This is done by subdividing the dry lognormal distribution into 100 size bins and solving the Kohler equation for each size bin (Lesins et al. 2002). Hence, the wet distribution is not assumed to be lognormal, eliminating any errors that would arise from that assumption. The Ångström exponent that is com-

### Table 1. Fine modal dry radius used for each of the six size scenarios.

<table>
<thead>
<tr>
<th>Scenario</th>
<th>Fine modal radius (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.2</td>
</tr>
<tr>
<td>2</td>
<td>0.1</td>
</tr>
<tr>
<td>3</td>
<td>0.05</td>
</tr>
<tr>
<td>4</td>
<td>0.035</td>
</tr>
<tr>
<td>5</td>
<td>0.02</td>
</tr>
<tr>
<td>6</td>
<td>0.01</td>
</tr>
</tbody>
</table>

Fig. 6. Same as Fig. 3, except for black carbon.
The Ångström exponent is then computed for the five central wavelengths (0.38, 0.44, 0.50, 0.67, and 0.87 μm). This procedure is repeated for six different size scenarios for each of the 28 GCM grid points that contains an AERONET station, and for each of the 12 months. This results in 336 (28 × 12) wavelength-dependent Ångström exponents, which are shown in Fig. 8 for each of the size scenarios. The figure demonstrates the sensitivity that size has on the Ångström exponent. Also note that for the size scenarios considered, the variation due to size dominates over the variation due to time of year or location. Note that the slope of the Ångström exponent with wavelength is positive except for the smallest size scenarios.
Fig. 8. The model-predicted variation of the Ångström exponent with the wavelength for six size scenarios ranging from a fine modal radius of 0.2 to 0.01 μm. For each size scenario, a line is drawn for each of the AERONET sites and for each month of the year.
Fig. 9. Comparison between the wavelength-dependent Ångström exponent as computed from the GCM-modeled aerosols for each of the six size scenarios against the function derived from the AERONET measurements. (a) Jan results and (b) Jul results. Note that not all AERONET stations have observations in Jan.
The exponent curves (Ångström exponent as a function of wavelength) are computed using the AERONET measurements based on monthly averaged aerosol optical depths. The results for January and July are shown in Fig. 9a and 9b, respectively, along with the exponent curves for each of the six size scenarios from the GCM-derived optical depths at the same AERONET grids and for the same month. Although the modeled exponent
curves are unlabeled they follow the same pattern as in Fig. 8 where the average exponent typically increases with decreasing modal radius. The comparison between measurements and model size scenarios often shows different slope characteristics. The modeled exponent curves are generally more monotonic with smaller and usually positive slopes while the measured exponent curves often exhibit sharp variations in slope as a function of wavelength and frequently have a negative slope.

The fact that the modeled curves do not always fully capture the shape of the measured curves can have a number of explanations. First, the limited number of size scenarios considered here restricts the full range of possible curve shapes that could be modeled. This limitation was imposed to keep the analysis relatively simple at this juncture in order to introduce this new method of better determining the dry aerosol sizes as a function of time and location. The scenarios can be extended to include variations in the geometric width and adding other modes. Second, the AERONET measurement themselves could have errors. In spite of cloud screening algorithms that are applied, thin cirrus clouds could still be present (O’Neill et al. 2001a). Third, the errors in the UV measurements are typically larger, which will magnify when computing the Ångström exponent. Finally, errors could arise due to differences in the vertical distribution of the aerosols.

For each GCM grid point which corresponds to a location of one of the 28 AERONET stations used in this study (Table 2), the size scenario that gives the smallest least squares error in the five Ångström exponents compared to the measured values is assumed to be the best size assumption. This provides a correlation between the exponent curve and aerosol size scenario. The resulting best scenarios are shown in Table 2 for the case of annual averaged exponent curves. Note that the best size scenario varies over the entire modal radius range from 0.01 to 0.2 μm, indicating the importance of including a geographically varying dry aerosol size for the assumed aerosol modes beyond a simple treatment where the dry size just differs between marine and continental regions. The monthly results also indicate that the best size scenario can vary for the given station depending on the season, but this variation is less than the geographic one (not shown).

The next step is to choose the most appropriate size scenario for each GCM grid point, not just the grid points containing an AERONET station. For each GCM grid point for a given month, we find the AERONET grid point for any of the 12 months of the year that has the best match in the modeled aerosol mass fractions and/or burdens. It is the GCM-predicted mass concentrations that are being used here and hence the technique’s success will be limited by the ability of the GCM to correctly predict the aerosol masses. In section 3 it was shown that fairly good agreement between model and measurements was achieved so there is an expectation that using the modeled mass fields is not unreasonable. The size scenario at the AERONET grid point with the best match in burden and/or mass fractions is then applied to the original GCM grid point. In this way one of the size scenarios is chosen for each GCM grid point.

The best match in modeled aerosol mass properties between a general grid point and the AERONET grid point is determined by one of three criteria: 1) total burden (BEST 1); 2) total burden and the five mass fractions for sea salt, dust, sulfate, organics, and black carbon (BEST 2); and 3) the five mass fractions only (BEST 3). The global total aerosol direct effect is then determined for each of these criteria.

The monthly averaged aerosol radiative solar effect at the top of the atmosphere (TOA) \( F_R \) is estimated using a simplified equation (Charlson et al. 1992) that is modified to include the effects of aerosol absorption of solar radiation (Chylek and Wong 1995):

\[
F_R = -S_{\text{tot}} T_{\text{atm}}^2 (1 - N_{\text{cloud}}) \\
\times 2 \tau (1 - \alpha)^2 \beta - 2 \alpha (1 - \omega),
\]

where \( S_{\text{tot}} \) is the top of atmosphere monthly mean solar irradiance, which is a function of latitude; \( T_{\text{atm}} \) is the transmittance of the atmosphere above the aerosol layer, which is assumed to be 0.79 (Penner et al. 1992); \( N_{\text{cloud}} \) is the fraction of sky covered by clouds obtained from the GCM; \( \alpha \) is the albedo of the underlying surface as given by the GCM; and \( \beta \) is the fraction of radiation scattered upward by the aerosol, which is approximated by \( \beta = (1/2)(1 - g) \) (Sagan and Pollock 1967). The overbars denote vertical means of the optical properties weighted by the extinction for the single scattering albedo \( \omega \) or the scattering coefficient for the asymmetry factor \( g \). In this formulation, the cloudy areas are assumed not to contribute to the solar direct effect.

The equation for \( F_R \) is applicable to the TOA total aerosol effect for optically thin layers in clear skies and hence is a measure of the direct effect only. It has been applied successfully to estimate the direct radiative effect by sulfate aerosols (Charlson et al. 1992) and by biomass burning aerosols (Chylek and Wong 1995). Its ease of use allows the radiative effect to be estimated without resorting to complicated radiative transfer codes and hence can be used offline from the GCM. The equation also clearly demonstrates the role of the optical depth, single scattering albedo, asymmetry factor, and the surface albedo in determining the radiative effect, making sensitivity studies easy to perform. In spite of this, the three optical parameters are obtained by Mie theory over the whole lognormal size distribution and hence are more precisely determined than by online GCM parameterizations.

The results are summarized in Table 3. In the upper part of the table we see the global total aerosol radiative effect when the same size scenario is applied to the entire globe. The greatest extinction occurs at a fine number modal dry radius around 0.04 μm. Recall that
TABLE 2. For each AERONET station the columns from left to right are station name, latitude, longitude, best fine modal radius from six size scenarios \( r_{\text{fs}} \), measured Angström exponent over all seven wavelengths (computed by averaging the five Angström exponents determined by a least-squares fit for each group of three adjacent wavelengths) \( \alpha_{\text{Angstrom}} \), difference between the modeled and measured Angström exponent and the effective radius \( r_{\text{eff}} \) computed from both the fine and coarse modes. The results are for the annual average.

<table>
<thead>
<tr>
<th>Station name</th>
<th>Lat (°N)</th>
<th>Lon (°E)</th>
<th>( r_{\text{fs}} ) (dry)</th>
<th>( \alpha_{\text{Angstrom}} )</th>
<th>Error</th>
<th>( r_{\text{eff}} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mauna Loa</td>
<td>19.539</td>
<td>122.242</td>
<td>0.010</td>
<td>3.22</td>
<td>0.69</td>
<td>0.06</td>
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<td>GSFC</td>
<td>39.030</td>
<td>128.120</td>
<td>0.035</td>
<td>1.80</td>
<td>0.36</td>
<td>0.14</td>
</tr>
<tr>
<td>Sevilleta</td>
<td>34.355</td>
<td>129.115</td>
<td>0.035</td>
<td>1.71</td>
<td>0.71</td>
<td>0.15</td>
</tr>
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<td>H.J. Andrews</td>
<td>44.239</td>
<td>127.776</td>
<td>0.035</td>
<td>1.79</td>
<td>0.48</td>
<td>0.15</td>
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<td>Cape Verde</td>
<td>16.733</td>
<td>33.065</td>
<td>0.200</td>
<td>0.40</td>
<td>0.28</td>
<td>0.89</td>
</tr>
<tr>
<td>Banizoumbou</td>
<td>13.541</td>
<td>2.665</td>
<td>0.200</td>
<td>0.44</td>
<td>0.26</td>
<td>0.84</td>
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<td>0.035</td>
<td>1.99</td>
<td>0.48</td>
<td>0.13</td>
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<td>1.87</td>
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scattering albedos, all just slightly less than the maximum at 0.05 \( \mu m \).

Figure 10 shows the total aerosol radiative clear-sky effect after adjusting the radii globally based on a best match of (a) mass burdens and fractions, and (b) just mass fractions. Both criteria show the nearly identical geographic structure. Figures 10c and 10d show the geographic distribution of the best modal radii for the fine mode. The largest particles are predicted to occur over deserts while the smallest particles are predicted over the oceans, especially over the Southern Hemisphere. Intermediate particle sizes are found over the continents.

6. Conclusions

The potential effects of aerosols, both natural and anthropogenic, depend on their chemical and physical properties. Furthermore the aerosol loading depends on the various sources and sinks for each component and their transport and conversion during its residence in the atmosphere. As air quality and climate models become more sophisticated, the treatment of aerosols should incorporate many of these properties and processes. However, the modeler is confronted with the issue of validating the results of an elaborate aerosol scheme. This represents a challenge since direct measurements of aerosol mass concentrations are limited and chemical and size observations is essentially non-existent over larger scales or prolonged times.

In this study, the aerosol predictions from a GCM were critically examined against appropriate measurements of aerosol properties that can help to assess the quality of the aerosol mass fields. Although local differences can be substantial the average over many locations do not indicate any serious bias. This finding provides some confidence that adjusting the modal radii of the assumed aerosol distribution is the best way to bring the modeled and measured optical properties of aerosols into closer agreement.

It was found that a reasonable geographic distribution...
of aerosol fine modal radii can be deduced by using the Ångström exponent curves from the AERONET stations as a way to establish a relationship between aerosol size and mass concentrations. This allows a global prediction of aerosol sizes to be made even though the surface optical depth measurements were taken at only 28 sites. It was found that the globally averaged fine modal radius was about 0.04 μm. The globally averaged direct total aerosol effect was found to be about 0.5 W m⁻² less when a geographic varying size is used compared to a constant size with the same average value.

The results and observations presented here are based on monthly averages, which will introduce some potential biases. We are using monthly averaged relative humidities to compute the water uptake on aerosols. Since the water uptake is a nonlinear function of relative humidity, using a monthly average will slightly increase the wet aerosol radius which will result in a slightly smaller predicted Ångström exponent. In this study we are focusing on demonstrating a new technique for validating GCM aerosol sizes and making adjustments to the assumed dry radii, rather than providing a definitive answer on the correct dry aerosol sizes. Further work is needed to examine and eliminate biases due to using monthly averaged quantities.

The results will only be as good as the ability of the model to correctly predict the aerosol mass concentrations. Fairly good agreement was demonstrated between average model predictions and measurements of mass concentrations; however, individual locations and times still show considerable variation, which will limit the accuracy of this method. Furthermore the validation was based only on surface concentrations whereas the aerosol optical depth and Ångström exponent is based on a column aerosols burden. If the surface aerosol type is not representative of the entire column, this will result in a source of error. Also the use of only 28 AERONET stations to represent all the possible aerosol size and mixing scenarios is not ideal. As more AERONET stations become operational and with longer observing records, this will improve. One shortcoming of this work to be addressed in future studies is to expand the number and types of aerosol size distribution scenarios in order to obtain better agreement between the measured and modeled exponent curves for the AERONET stations.
Acknowledgments. Matthew Coffin helped with the compilation of the AERONET data. Yiran Peng helped with the running of the GCM. We appreciate the work of the various AERONET principle investigators and site managers in providing the aerosol optical depth measurements. This work was funded through grants provided by NSF/CPC/CAFCAS project on Global Chemistry for Climate and SOLAS. We also thank the anonymous reviewers for their helpful comments.

REFERENCES


