Direct radiative forcing and atmospheric absorption by boundary layer aerosols in the southeastern US: model estimates on the basis of new observations

Shaocai Yu\textsuperscript{a,*}, Charles S. Zender\textsuperscript{b}, V.K. Saxena\textsuperscript{c}

\textsuperscript{a}Nicholas School of the Environment, Duke University, Durham, NC 27708, USA
\textsuperscript{b}Department of Earth System Science, University of California, Irvine CA 92697-3100, USA
\textsuperscript{c}Department of Marine, Earth and Atmospheric Sciences, North Carolina State University, Box 8208, Raleigh, NC 27695-8208, USA

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Abstract

In an effort to reduce uncertainties in the quantification of aerosol direct radiative forcing (ADRF) in the southeastern United States (US), a field column experiment was conducted to measure aerosol radiative properties and effects at Mt. Mitchell, North Carolina, and at an adjacent valley site. The experimental period was from June 1995 to mid-December 1995. The aerosol optical properties (single scattering albedo and asymmetry factor) needed to compute ADRF were obtained on the basis of a procedure involving a Mie code and a radiative transfer code in conjunction with the retrieved aerosol size distribution, aerosol optical depth, and diffuse-to-direct solar irradiance ratio. The regional values of ADRF at the surface and top of atmosphere (TOA), and atmospheric aerosol absorption are derived using the obtained aerosol optical properties as inputs to the column radiation model (CRM) of the community climate model (CCM3). The cloud-free instantaneous TOA ADRFs for highly polluted (HP), marine (M) and continental (C) air masses range from 20.3 to 24.8, 1.3 to 10.4, and 1.9 to 13.4 W m\textsuperscript{-2}, respectively. The mean cloud-free 24-h ADRFs at the TOA (at the surface) for HP, M, and C air masses are estimated to be 8 ± 4 (33 ± 16), 7 ± 4 (−13 ± 8), and −0.14 ± 0.05 (−8 ± 3) W m\textsuperscript{-2}, respectively. On the assumption that the fractional coverage of clouds is 0.61, the annual mean ADRFs at the TOA and the surface are 2 and 7 W m\textsuperscript{-2}, respectively. This also implies that aerosols currently heat the atmosphere over the southeastern US by 5 W m\textsuperscript{-2} on annual timescales due to the aerosol absorption in the troposphere. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Aerosol radiative forcing; Column radiation model; Southeastern US; Observation; Aerosol absorption

1. Introduction

Eastern China, south central Europe and the eastern United States (US) are regions where the radiative forcing of anthropogenic sulfate aerosols supersedes that of anthropogenic greenhouse gases, manifesting itself in terms of cooling of the surface-air temperature (IPCC, 1995; Kiehl and Briegleb, 1993). Saxena et al. (1997) and Saxena and Yu (1998) have verified this model prediction in the southeastern US by finding the existence of aerosol cooling in the southeastern US in the temperature records of the surface air during 1949–1994. The direct aerosol effect refers to scattering and absorption of radiation by the aerosol particles themselves (Charlson et al., 1991; Penner et al., 1994; Kiehl and Briegleb, 1993; IPCC, 1995; Saxena and Yu, 1998). The conclusion of IPCC (1995) is that global mean direct forcing resulting from anthropogenic sulfate may range from −0.25 to −0.9 W m\textsuperscript{-2}, with substantial uncertainty; and biomass burning aerosol forcing may range from −0.05 to −0.6 W m\textsuperscript{-2}. The low confidence in the estimates of aerosol radiative forcing is due to highly non-uniform compositional, spatial, and
temporal distributions of tropospheric aerosol on regional scales owing to their heterogeneous sources and short lifetimes (Schwartz and Andreae, 1996). The shortwave absorption in Earth’s atmosphere due to the absorbing aerosols is also one of the least quantified properties of the climate system (Zender et al., 1997). The comparison of signals of regional aerosol-induced radiative forcing in eastern China and the southeastern US indicates that the high concentration of absorbing aerosol over eastern China was one of the major reasons for the warming trend during 1951–1994 (Yu et al., 2001).

To reduce the uncertainty in the climate effect of tropospheric aerosols in the industrial pollution plumes that are transported from the east coast of the US over the Atlantic Ocean, the tropospheric aerosol radiative forcing observational experiment (TARFOX) was carried out on the eastern coast during July 10–31, 1996 (Russell et al., 1999; Redemann et al., 2000). In two case studies, Redemann et al. (2000) found that the instantaneous shortwave aerosol radiative forcing was of the order of $-36 \text{ W m}^{-2}$ at the top of the atmosphere (TOA) and about $-56 \text{ W m}^{-2}$ at the surface. However, the TARFOX datasets do not overlap geographically with the southeastern US nor is it of sufficient length to estimate the regional annual average aerosol direct radiative forcing (ADRF). The objectives of this study are to use tropospheric aerosol measurements to estimate the instantaneous and annual mean ADRF at the surface and TOA, and aerosol absorption over the southeastern US with column radiation model (CRM) of the NCAR (National Center for Atmospheric Research) community climate model (CCM3) (Kiehl et al., 1998; Briegleb, 1992). The atmospheric shortwave absorption due to the aerosol layer was determined as the difference between the TOA ADRF and surface ADRF. Since the ADRF is not determined by surface aerosols only but by total aerosols within the atmospheric column, it is very useful to obtain the average aerosol radiative properties for the atmospheric column, for example, through a column experiment. One of our field sites (Mt. Mitchell) is at a remote and elevated location that is influenced by air masses arriving from marine, continental and polluted sectors. It stays frequently in the free troposphere due to decoupling with the surrounding inversion layer. The data sets are considered to be regionally representative of the southeastern US (Yu et al., 2000; Wenny et al., 1998). We also compare our in situ results with the estimates from other global models for the aerosol radiative forcing in the southeastern US.

2. Retrieval of aerosol radiative properties in the southeastern US

A detailed description of the instrumentation, methodology, data quality assurance and quality control of the data has been given by Wenny et al. (1998) and Yu et al. (2000). Here a brief description is presented. The research sites are a mountain top station located on the peak of Mt. Gibbs (35.78°N, 82.29°W, 2006 m MSL) in Mt. Mitchell State Park, and a valley station located adjacent to the Burnett Reservoir near the town of Black Mountain, North Carolina (35.66°N, 82.38°W, 951 m MSL) (for details see Yu et al., 2000). The two sites are separated horizontally by 10 km and vertically by 1 km. The experimental period was from June 1995 to mid-December 1995. The aerosol optical depths (AOD) at the three operational wavelengths (415, 500 and 673 nm) are determined on the basis of the direct components of solar irradiance measured by the multi-filter rotating shadowband radiometer (MFRSR) (Yu et al., 2000). Table 1 lists the mean total AOD and standard deviation as measured from the valley and mountain sites and mean 1-km layer AOD between the two sites as a function of air mass type for cloud-free days during the experimental period. The clear-sky intervals were determined by a full-sky imaging camera. Total 34 cloud-free days of measurement data were available because of the bad weather (cloud and rain) situation and instrumental problems during the field experiment period. Fig. 1 shows the time series of AOD at 500 nm for cloud-free periods during the experiment. The air masses are classified (Yu et al., 2000) as highly polluted (HP), marine (M) and continental (C) based on the SO$_2$ and NO$_x$ emission inventory of the US Environmental Protection Agency. It is clear from Table 1 that the HP air masses exhibit the largest average AOD, compared to M and C air masses at the three operational wavelengths. The mean total AOD at 500 nm at the valley site was $0.68 \pm 0.33$, $0.29 \pm 0.12$, and $0.10 \pm 0.04$ for HP, M, and C air masses, respectively. As mentioned by Yu et al. (2000), there are two reasons that the mean AOD of marine air masses is higher than that of continental air masses at the research site. First, the marine classification for the sampling site does not imply pure marine air, but rather a modified marine air caused by additional influence from continental and polluted sources between the ocean and the site (the shortest distance between the site and the ocean is about 290 km). Second, the AOD is typically very low during winter months (Peterson et al., 1981). In this study period, the majority of the marine air mass cases occurred during September and early October and the majority of continental air mass cases occurred during late October and November. Overall, the ratios of mean 1-km layer AOD to total mean AOD from the valley site for HP air mass were 73%, 73% and 79% for 415, 500 and 673 nm, respectively. This indicates that the major portion of atmospheric aerosols at our sites, which make an important contribution to the AOD, is located in the lowest 1-km boundary layer of the troposphere. This is in reasonable agreement with that of Hegg et al. (1997),
who found that AOD for the lowest 4 km of the
troposphere constituted over 90% of the total column
AOD off the mid-Atlantic coast of the US.

Table 2 lists the experimental results of AOD, and
retrieved columnar lognormal size distribution (number
concentration ($N$), number median radius (geometric
mean radius, $r_g$) and geometric standard deviation
($s_g$)) by the search-graph method. Table 2 also lists the
aerosol radiative properties (imaginary part, asymmetry
factor and single scattering albedo ($\omega$)) and ground
albedo obtained by using a procedure involving a Mie
code and a radiative transfer code in conjunction with
the retrieved aerosol size distribution, AOD, and diffuse-
direct ratio (DDR) (Yu et al., 2000). A detailed
description of the search-graph method and the proce-
dure has been given by Yu et al. (2000). Table 2 shows
that $N$, $r_g$ and $s_g$ are in the range of 12 to 600 cm$^{-3}$,
0.03 to 0.54 \( \mu m \) and 1.12 to 2.95, respectively. The
asymmetry factors at 500 nm are in the range of 0.61 to
0.77. $\omega$ was in the range of 0.72 to 0.97 and the
imaginary part of refractive index was in the range of
0.005 to 0.051. Reported values of $\omega$ in the scientific
literature show $\omega$ to be 1.0 for sulfate and pure marine
aerosols and range from 0.5 to 0.9 for desert dust and

Table 1

<table>
<thead>
<tr>
<th>Air mass</th>
<th>$\tau_{V}$ at the valley site</th>
<th>$\tau_{M}$ at the mountain site</th>
<th>$\tau_{L}$ for the intervening layer</th>
<th>Ratio ($\tau_{L}/\tau_{V}$)</th>
<th>415</th>
<th>500</th>
<th>673</th>
</tr>
</thead>
<tbody>
<tr>
<td>HP</td>
<td>0.26 ± 0.04</td>
<td>0.23 ± 0.04</td>
<td>0.19 ± 0.03</td>
<td>0.75 ± 0.04</td>
<td>0.73 ± 0.12</td>
<td>0.73 ± 0.12</td>
<td>0.73 ± 0.12</td>
</tr>
<tr>
<td>M</td>
<td>0.36 ± 0.06</td>
<td>0.33 ± 0.06</td>
<td>0.30 ± 0.05</td>
<td>0.69 ± 0.09</td>
<td>0.69 ± 0.09</td>
<td>0.69 ± 0.09</td>
<td>0.69 ± 0.09</td>
</tr>
<tr>
<td>C</td>
<td>0.68 ± 0.10</td>
<td>0.65 ± 0.10</td>
<td>0.58 ± 0.08</td>
<td>0.90 ± 0.02</td>
<td>0.82 ± 0.02</td>
<td>0.82 ± 0.02</td>
<td>0.82 ± 0.02</td>
</tr>
</tbody>
</table>

Fig. 1. The time series of aerosol optical depth (AOD) at
500 nm at valley site (A), for 1-km layer between valley and
mountain sites (B), and at mountain site (C) for cloud-free
period during the experiment from June to mid-December 1995.
Table 2

The measured optical depth (τ) at 415, 500, 673 nm, columnar size distribution inferred by the search-graph-method, single scattering albedo (ω), asymmetry factor, refractive index, zenith angle, air mass types and instantaneous aerosol direct radiative forcing (ADRF) at the top of the atmosphere (TOA ADRF) and surface (Sfc ADRF) calculated by CRM model. The atmospheric absorption was calculated as the difference between TOA ADRF and Sfc ADRF.

<table>
<thead>
<tr>
<th>Day (1995)</th>
<th>Time (EST)</th>
<th>Zenith angle</th>
<th>Air mass</th>
<th>τ measured</th>
<th>Search-graph-method</th>
<th>ADRF (W m⁻²)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>415</td>
<td>500</td>
<td>673</td>
<td>415</td>
</tr>
<tr>
<td>249(9/5)</td>
<td>10:58</td>
<td>35.5</td>
<td>M</td>
<td>0.3</td>
<td>0.23</td>
<td>0.14</td>
</tr>
<tr>
<td>272(9/29)</td>
<td>8:32</td>
<td>66.9</td>
<td>M</td>
<td>0.25</td>
<td>0.19</td>
<td>0.12</td>
</tr>
<tr>
<td>272(9/29)</td>
<td>8:59</td>
<td>61.9</td>
<td>M</td>
<td>0.24</td>
<td>0.18</td>
<td>0.11</td>
</tr>
<tr>
<td>274(10/1)</td>
<td>8:31</td>
<td>69.6</td>
<td>M</td>
<td>0.12</td>
<td>0.1</td>
<td>0.07</td>
</tr>
<tr>
<td>274(10/1)</td>
<td>9:01</td>
<td>62.1</td>
<td>M</td>
<td>0.11</td>
<td>0.09</td>
<td>0.07</td>
</tr>
<tr>
<td>274(10/1)</td>
<td>9:15</td>
<td>59.6</td>
<td>M</td>
<td>0.11</td>
<td>0.1</td>
<td>0.08</td>
</tr>
<tr>
<td>275(10/2)</td>
<td>9:32</td>
<td>56.9</td>
<td>M</td>
<td>0.2</td>
<td>0.16</td>
<td>0.11</td>
</tr>
<tr>
<td>275(10/2)</td>
<td>10:06</td>
<td>51.5</td>
<td>M</td>
<td>0.21</td>
<td>0.18</td>
<td>0.12</td>
</tr>
</tbody>
</table>

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soot aerosols (Lacis and Mishchenko, 1995). Wenny et al. (1998) reported that $\sigma$ at 312 nm varied from 0.75 to 0.93 for aerosols at our research site. Russell et al. (1999) found that the estimated column single scattering albedos for the ambient TARFOX aerosol was $\sim 0.9$ at 550 nm. The values of $\sigma$ from this study are consistent with these values given by other authors. Lacis and Mishchenko (1995) showed that the asymmetry factor of soot and large desert aerosols was about 0.9, but for sulfate, marine and smaller dust aerosols the asymmetry factor ranges from 0.65 to 0.8. The values of asymmetry factor reported here are close to those for sulfate, marine and smaller dust aerosols. Since the size distribution was inferred from the total AOD, the aerosol radiative properties derived by this method represent a weighted average over the entire column. Yu et al. (2000) found that ground albedo is 0.19 at 0.10 at our research site. The mean ground albedo (0.19) was used in the calculation of this study.

### 3. Calculation of aerosol direct radiative forcing and aerosol absorption

The NCAR CCM3 CRM (Kiehl et al., 1998; Briegleb, 1992) is used to estimate the TOA and surface ADRF and aerosol absorption for the measurement sites. The CCM3 CRM uses an $\delta$-Eddington approximation with 19 spectral intervals (7 for O$_3$, 2 for the visible, 7 for H$_2$O, and 3 for CO$_2$) spanning the solar spectrum from 0.2 to 5.0 $\mu$m. A detailed description of all the physical and numerical methods used in CCM3 CRM is given in Kiehl et al. (1998) and Briegleb (1992). The atmospheric initial conditions of Kiehl and Briegleb (1993) for mid-latitude summer vertical profiles of temperature, pressure, ozone and H$_2$O vapor mixing ratios were used in the CRM. The mass extinction coefficient, single scattering albedo and asymmetry factor at 19 spectral interval wavelengths calculated by a Mie code with the retrieved columnar size distribution and refractive index, and ground albedo (0.19) were input into the CRM. The density of aerosol particles was assumed to be 1.86 $\text{g cm}^{-3}$. The ADRF is obtained as the difference in shortwave net radiative fluxes at the TOA (or at the surface) between CRM simulations with and without aerosol mass loading. We also infer the atmospheric absorption due to boundary layer aerosols as the difference between the TOA ADRF and surface ADRF.

Table 2 lists the examples of the instantaneous ADRF for each cloud-free time during the experiment. The zenith angle, air mass types, AOD at the three wavelengths, size distribution, retrieved aerosol radiative properties and refractive indices are listed in Table 2. Hansen et al. (1997) indicated that the ADRF is very sensitive to single scattering albedo (or imaginary part of refractive index). Unfortunately, it is very difficult to

<table>
<thead>
<tr>
<th>Day (1995)</th>
<th>Time (EST)</th>
<th>Zenith angle</th>
<th>Air mass</th>
<th>AOD at 415 nm</th>
<th>AOD at 500 nm</th>
<th>AOD at 673 nm</th>
<th>Refractive index</th>
<th>Single scattering albedo</th>
<th>Asymmetry factor</th>
<th>Absorption</th>
</tr>
</thead>
<tbody>
<tr>
<td>18/7/95</td>
<td>9:47</td>
<td>53.5</td>
<td>HP</td>
<td>0.7</td>
<td>0.5</td>
<td>0.3</td>
<td>0.94</td>
<td>0.72</td>
<td>0.62</td>
<td>1.58</td>
</tr>
<tr>
<td>21/7/95</td>
<td>8:40</td>
<td>58.6</td>
<td>HP</td>
<td>0.6</td>
<td>0.4</td>
<td>0.3</td>
<td>0.87</td>
<td>0.58</td>
<td>0.63</td>
<td>1.57</td>
</tr>
<tr>
<td>24/7/95</td>
<td>9:23</td>
<td>55.3</td>
<td>HP</td>
<td>0.8</td>
<td>0.7</td>
<td>0.6</td>
<td>0.89</td>
<td>0.72</td>
<td>0.62</td>
<td>1.58</td>
</tr>
<tr>
<td>27/7/95</td>
<td>10:05</td>
<td>49.3</td>
<td>HP</td>
<td>0.6</td>
<td>0.5</td>
<td>0.4</td>
<td>0.87</td>
<td>0.58</td>
<td>0.63</td>
<td>1.57</td>
</tr>
<tr>
<td>30/7/95</td>
<td>12:12</td>
<td>40.2</td>
<td>HP</td>
<td>0.8</td>
<td>0.7</td>
<td>0.6</td>
<td>0.89</td>
<td>0.72</td>
<td>0.62</td>
<td>1.58</td>
</tr>
<tr>
<td>2/8/95</td>
<td>13:18</td>
<td>34.9</td>
<td>HP</td>
<td>0.6</td>
<td>0.5</td>
<td>0.4</td>
<td>0.87</td>
<td>0.58</td>
<td>0.63</td>
<td>1.57</td>
</tr>
<tr>
<td>5/8/95</td>
<td>15:23</td>
<td>28.6</td>
<td>HP</td>
<td>0.8</td>
<td>0.7</td>
<td>0.6</td>
<td>0.89</td>
<td>0.72</td>
<td>0.62</td>
<td>1.58</td>
</tr>
<tr>
<td>8/8/95</td>
<td>17:31</td>
<td>22.3</td>
<td>HP</td>
<td>0.6</td>
<td>0.5</td>
<td>0.4</td>
<td>0.87</td>
<td>0.58</td>
<td>0.63</td>
<td>1.57</td>
</tr>
</tbody>
</table>

* The refractive index was obtained by averaging available results (see the text for explanation).
retrieve mean single scattering albedo for the whole atmospheric column. As mentioned by Yu et al. (2000), our DDR method can only have solutions for the cases whose AOD at 500 nm was between 0.1 and 0.3. The averages of the imaginary part of refractive index for September, October and November are $-0.017 \pm 0.009$, $-0.027 \pm 0.021$, $-0.050 \pm 0.001$ on the basis of available results in Table 2. The values of $-0.017$, $-0.027$ and $-0.050$ were used for cases for which imaginary part of refractive index is not available, between July and September, in October and in November, respectively, as indicated in Table 2. Since entirely cloud-free days were very rare, we assume that the aerosol radiative properties obtained at one cloud-free time were constant to study the diurnal variation of instantaneous cloud-free ADRF. Fig. 2 shows the diurnal variations of instantaneous surface and TOA ADRF for C, M, and HP air masses, calculated on the basis of measurements at Julian Day 321 (11/17/1995, 10:27), 272 (9/29/1995, 8:59) and 227 (8/15/1995, 9:50) (see Table 2). The diurnal behavior of the TOA aerosol direct radiative forcing (ADRF) is determined by the changing ratio of aerosol forward-scattered radiation (scattering angle less than 90°) to aerosol up-scattered radiation (scattering up to space rather than down to surface) as the zenith angle changes (Wiscombe and Grams, 1976). When the sun is closer to the horizon much (up to half) of the forward-scattered sunlight corresponds to up-scattered radiation that is reflected back to space and thus cools the climate system (hence ADRF minima (more negative) at twilight). When the sun is overhead, the forward-scattered solar radiation coincides with the down-scattered radiation, i.e., it penetrates the aerosol layer where it may be absorbed by the absorbing aerosols or by the surface, warming the climate system (hence ADRF maxima (less negative) at noon). These results are in agreement with those of Russell et al. (1997). Fig. 2 also indicates that the TOA ADRF is much smaller than surface ADRF during the daytime (from sunrise to sunset), especially for highly absorbing aerosol (small single scattering albedo) and at the midday peak of incident sunlight. This is consistent with the finding of Satheesh and Ramananthan (2000) who reported the large difference in tropical aerosol forcing at TOA and the Earth’s surface due largely to solar absorption by carbonaceous aerosol.

Table 2 shows that the instantaneous TOA ADRF ranges from +20.3 to –24.8, +1.3 to –10.4, and +1.9 to –13.4 W m$^{-2}$ for HP, M, and C air masses, respectively. The instantaneous surface ADRF ranges from –55.3 to +156.5, +16.7 to +24.3, and +9.2 to +30.2 W m$^{-2}$ for HP, M, and C air masses, respectively. The atmospheric aerosol absorption ranges from 5.3 to 176.8 W m$^{-2}$, and is always positive. Clearly, the difference between ADRF at surface and TOA are very large, especially for highly absorbing aerosols. The inclusion of absorbing aerosols makes the ADRF at TOA less negative while making the ADRF at the surface more negative. This is in agreement with the results of Hignett et al. (1999). Fig. 3 shows the surface and TOA ADRF, and absorption as functions of aerosol optical depth and single scattering albedo. As can be seen, the surface ADRF was generally more sensitive to AOD and single scattering albedo than TOA ADRF. Note that the zenith angle (or local time) is also an important factor for ADRF as indicated in Fig. 2. Figs. 3a and b also show that both surface and TOA ADRFs are more sensitive to the AOD than single scattering albedo on the basis of the correlation coefficients. The small single scattering albedo (more absorbing) and large optical depth will greatly increase the heating rate of the atmosphere due to aerosols as indicated in Fig. 3 and
Table 2. Fig. 4 shows the statistical summaries of ranges of the AOD, asymmetry factor, single scattering albedo, surface and TOA ADRF, and absorption with their frequencies of occurrences on the basis of Table 2. As can be seen, the most frequencies of occurrences for AOD, surface ADRF, TOA ADRF and absorption are in the ranges of 0.04 to 0.30, -9.2 to -50 W m^{-2}, 0 to -10 W m^{-2}, and +5 to +50 W m^{-2}, respectively.

A sensitivity test was performed by calculating the ADRF difference between the case with our aerosol radiative properties and that with the sulfate optical properties of Kiehl and Briegleb (1993), keeping the AOD same. It is found that the instantaneous surface ADRF is -27.2 W m^{-2} on the basis of measurement of aerosol radiative properties at 10:58 AM, 11/17/1995, while the surface ADRF is -10.0 W m^{-2} on the basis of sulfate aerosol radiative properties of Kiehl and Briegleb (1993). The surface ADRF from this study is much higher. This is reasonable because the aerosol of this study is total aerosol and the imaginary part of refractive index is also considered.

To estimate the 24 h and annual average ADRF for different air mass types for the whole year, it is ideal to...
<table>
<thead>
<tr>
<th>Aerosols</th>
<th>Method</th>
<th>ADRF at TOA (W m⁻²)</th>
<th>ADRF at surface (W m⁻²)</th>
<th>Regions</th>
<th>Author</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total aerosol (marine)</td>
<td>Measurement and CCM3 CRM</td>
<td>$-3 \pm 2$ (with cloud)(^a)</td>
<td>$-5 \pm 3$ (with cloud)(^a)</td>
<td>SE US</td>
<td>This work</td>
</tr>
<tr>
<td>Total aerosol (continental)</td>
<td>Measurement and CCM3 CRM</td>
<td>$-0.05 \pm 0.02$ (with cloud)(^a)</td>
<td>$-3 \pm 1$ (with cloud)(^a)</td>
<td>SE US</td>
<td>This work</td>
</tr>
<tr>
<td>Total aerosol (polluted)</td>
<td>Measurement and CCM3 CRM</td>
<td>$-3 \pm 2$ (with cloud)(^a)</td>
<td>$-13 \pm 6$ (with cloud)(^a)</td>
<td>SE US</td>
<td>This work</td>
</tr>
<tr>
<td>Total aerosol (average)</td>
<td>Measurement and CCM3 CRM</td>
<td>$-2 \pm 1$ (with cloud)(^a)</td>
<td>$-7 \pm 2$ (with cloud)(^a)</td>
<td>SE US</td>
<td>This work</td>
</tr>
<tr>
<td>Total aerosol (marine)</td>
<td>Measurement and CCM3 CRM</td>
<td>$-7 \pm 4$ (no cloud)(^a)</td>
<td>$-13 \pm 8$ (no cloud)(^a)</td>
<td>SE US</td>
<td>This work</td>
</tr>
<tr>
<td>Total aerosol (continental)</td>
<td>Measurement and CCM3 CRM</td>
<td>$-0.14 \pm 0.05$ (no cloud)(^a)</td>
<td>$-8 \pm 3$ (no cloud)(^a)</td>
<td>SE US</td>
<td>This work</td>
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<tr>
<td>Total aerosol (polluted)</td>
<td>Measurement and CCM3 CRM</td>
<td>$-8 \pm 4$ (no cloud)(^a)</td>
<td>$-33 \pm 16$ (no cloud)(^a)</td>
<td>SE US</td>
<td>This work</td>
</tr>
<tr>
<td>Total aerosol (average)</td>
<td>Measurement and CCM3 CRM</td>
<td>$-5 \pm 3$ (no cloud)(^a)</td>
<td>$-17 \pm 6$ (no cloud)(^a)</td>
<td>SE US</td>
<td>This work</td>
</tr>
<tr>
<td>Total aerosol (continental)</td>
<td>Box model</td>
<td>$-3$</td>
<td></td>
<td>SE US</td>
<td>Saxena and Yu (1998)</td>
</tr>
<tr>
<td>Total aerosol (polluted)</td>
<td>Box model</td>
<td>$-21$</td>
<td></td>
<td>SE US</td>
<td>Saxena and Yu (1998)</td>
</tr>
<tr>
<td>Anthropogenic sulfate</td>
<td>Box model</td>
<td>$-2$</td>
<td></td>
<td>E US</td>
<td>Charlson et al. (1991)</td>
</tr>
<tr>
<td>Anthropogenic sulfate</td>
<td>3-D chemical and CCM3 CRM</td>
<td>$-3$</td>
<td></td>
<td>E US</td>
<td>Kiehl and Briegleb (1993)</td>
</tr>
<tr>
<td>Sulfate</td>
<td>AGCM model</td>
<td>$\sim -0.5$ to $\sim -1.5$</td>
<td></td>
<td>E US</td>
<td>IPCC (1995)</td>
</tr>
<tr>
<td>Anthropogenic sulfate</td>
<td>radiative transfer model</td>
<td>$-3$</td>
<td></td>
<td>E US</td>
<td>Taylor and Penner (1994)</td>
</tr>
<tr>
<td>Total aerosol</td>
<td>radiative transfer model</td>
<td>$-9$</td>
<td></td>
<td>E US</td>
<td>Russell et al. (1997)</td>
</tr>
<tr>
<td>Total aerosol</td>
<td>radiative transfer model</td>
<td>$-8$ to $\sim -50$ (no cloud)(^a)</td>
<td></td>
<td>E US</td>
<td>Bergstrom and Russell (1999)</td>
</tr>
<tr>
<td>Anthropogenic aerosol</td>
<td>internal mixture</td>
<td>$-1.5$</td>
<td></td>
<td>E US</td>
<td>Haywood and Shine (1995)</td>
</tr>
<tr>
<td>Sulfate + BC</td>
<td>1-D radiative transfer model</td>
<td>$-2$ to $-3$</td>
<td></td>
<td>E US</td>
<td>Schult et al. (1997)</td>
</tr>
</tbody>
</table>

\(^a\)“No cloud” means cloud-free 24-h average, “with cloud” means that the fractional coverage of clouds is assumed to be 0.61.
measure the aerosol radiative properties and calculate
the aerosol forcing for each air mass and then average
them during the cloud-free period for the whole year.
Unfortunately, the information is very scarce. In this
study, we select the typical aerosol radiative properties
(mass extinction coefficient, single scattering albedo,
asymmetry factor) of 321 (11/17/1995, 10:27 EST, C air
mass), 272 (9/29/1995, 8:59 EST, M air mass) and 227
(8/15/1995, 9:50 EST, HP air mass) as representatives
for the C, M, and HP air masses, respectively. These
cases were selected because their AOD was close to
the mean AOD of corresponding air masses and most of
continental, marine, and polluted air masses occurred
during winter, fall and summer seasons, respectively as
shown in Tables 1 and 2. Then, the mean optical
diameter in Table 1 for three air mass types are used in the
calculation. It was found that the mean cloud-free 24-h
ADRF at the TOA (and at the surface) for HP, M, and
C air masses was estimated to be $-8 \pm 4 \ (33 \pm 16)$,
$-7 \pm 4 \ (-13 \pm 8)$, and $-0.14 \pm 0.05 \ (-8 \pm 3)$ Wm$^{-2}$,
respectively, as listed in Table 3. To assess the annual
average ADRF in the southeastern US, the relative
contributions of three air masses are needed. On
the basis of back trajectory analysis of air masses from
June 1996 to October 1996, and from March 1997 to
June 1997, Bahrmann and Saxena (1998) found that the
percentages of air masses influencing our research site
were 43.2, 22.4 and 34.4% for C, M, and HP masses,
respectively. Using these percentages, the annual cloud-
free, 24-h mean ADRF at the TOA (and at the surface)
was estimated to be $-5 \pm 3 \ (-17 \pm 6)$ Wm$^{-2}$. If the
mean fraction of the area with clear sky condition in
the southeastern US is assumed to be 0.39, which is
the globally average clear sky condition (Charlson et al.,
1991), the mean ADRF at the TOA (and at the surface)
will be $-3.2 \pm 1.5 \ (13 \pm 6)$, $-2.7 \pm 1.5 \ (-5 \pm 3)$, and
$-0.05 \pm 0.02 \ (-3 \pm 1)$ Wm$^{-2}$ for HP, M, and C air
masses, respectively. The annual mean ADRF at the
TOA (and at the surface) is $-2 \pm 1 \ (-7 \pm 2)$ Wm$^{-2}$.
Note that we assume that the ADRF is zero when clouds
are present. So our estimate of annual mean ADRF is a
lower bound. The annual mean atmospheric absorption
due to aerosol layer will be $5 \pm 3$ Wm$^2$.

Table 3 lists the comparison of our estimates of
ADRF with those of other investigators for the
southeastern US or eastern US. The methods used by each
author are also listed in Table 3. As can be seen, box
models, GCM and column radiation models have been
used to estimate the ADRF on the basis of the actual
measurements or chemical simulation. The estimates of
ADRF in the eastern US (or southeastern US) have a
large variation because of different assumptions in each
model and relative uncertainty of aerosol radiative
properties. Our annual estimated TOA ADRF
($-2 \pm 1$ Wm$^{-2}$) was close to those of Boucher and
Anderson (1995), Schult et al. (1997) and Haywood and
Ramaswamy (1998), but a little lower than those of
Bergstrom and Russell (1999) show that cloud-free, 24-h
average ADRF is $-9$ Wm$^{-2}$ near the eastern US coast
in summer. Since our calculations are for total aerosols
(anthropogenic + natural) and for whole aerosol com-
ponents (sulfate + organic), our estimated ADRF is
consistent with the reported values. The major portion
of atmospheric aerosol at our sites is located in the
lowest 1-km boundary layer of the troposphere where it
exerts a negligible longwave forcing. Thus we make the
approximation that ADRF is due to shortwave forcing
alone. It is noteworthy that although there was large
negative ADRF at the surface, the atmospheric heating
rate due to aerosols is large, especially for high polluted
absorbing aerosols. As pointed out by Ackerman et al.
(2000) and Hansen et al. (1997), absorbing aerosols were
more effective than their radiative forcing due to their
cloud-burning effects. Further observations on absorbing
aerosols are obviously needed.

4. Conclusion

Using a column radiation model, the instantaneous
and cloud-free 24-h average ADRF at both TOA and
surface for C, M, and HP air masses in the southeastern
US was estimated on the basis of measurements of
aerosol radiative properties. The results show that the
annual regional cloud-free mean ADRF at the TOA
(and at the surface) is $-5 \pm 3 \ (-17 \pm 6)$ Wm$^{-2}$.
Considering the effect of cloud fraction, we estimate
that the annual regional mean ADRF at the TOA (and
at the surface) is $-2 \pm 1 \ (-7 \pm 2)$ Wm$^{-2}$. This estimate
of ADRF in the southeastern US is close to those of
other investigators (see Table 3) when they considered
both sulfate and black carbon. Since our study considers
the total aerosol (anthropogenic + natural) instead of
anthropogenic and whole aerosol (sulfate + organics)
instead of sulfate, our estimated ADRF provide an
upper bound on the direct effect of anthropogenic
aerosols. This study confirms the existence of a cooling
effect (negative forcing) due to the direct effect of aerosol
at the surface in the southeastern US as indicated by
Saxena and Yu (1998). However, the atmosphere of the
troposphere above the ground is still heated with annual
mean rate of $5 \pm 3$ Wm$^{-2}$ due to the aerosol layer.
Since the aerosol field and composition are inhomogeneous
in time and space, and our study also indicates the effect
of long-range transport of pollution from other regions,
the complexity of this aerosol puzzle seems to grow with
each new study as pointed out by Kiehl (1999).
Continuity in the long-term monitoring of the aerosol
properties, especially for absorbing aerosol, is still
crucial in improving the understanding of role of
aerosols in the climate change.
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References


