Effect of Aerosol on the Clear-Sky Radiation Regime As Derived from Zvenigorod Aerosol–Cloud–Radiation Experiments

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Received June 3, 2004; in final form, February 1, 2005

Abstract—Clear-sky measurements conducted during the 2001 and 2002 Zvenigorod aerosol–cloud–radiation experiments are used to estimate the short- and longwave aerosol radiative forcing on the Earth’s surface, at the top of the atmosphere, and in the entire depth of the atmosphere. The sensitivity of the aerosol radiative forcing to variations in the aerosol optical thickness, the single-scattering albedo, the asymmetry factor of the aerosol phase function, and the surface albedo are analyzed.

INTRODUCTION

The influence of aerosol on the climate is manifested through its influence on the radiation regime of the atmosphere. Variations in the atmospheric radiation fluxes lead to variations in the atmospheric thermal stratification and to variations in the Earth’s surface temperature. Associated with greenhouse gases, present-day warming in the Northern Hemisphere can be reduced nearly by half owing to aerosol effects [1–3]. As a rule, the aerosol effect counteracts the greenhouse effect of carbon dioxide and other atmospheric greenhouse gases. An integral estimate for the radiative influence of aerosol is difficult to obtain because of the uncertainties associated with the spatiotemporal variability in aerosol loadings, optical and physical properties, chemical composition, and various features of the hydrological cycle (including cloud-formation processes). An adequate description of actual aerosol can be constructed only by using its various characteristics measured in complex experiments, such as concentration, particle size distribution, refractive index, wavelength dependence of the vertical profile of optical thickness, single-scattering albedo, and the phase function, in combination with characteristics of aerosol-involving climatic processes [4, 5]. Field radiation experiments aimed at the determination of tropospheric-aerosol optical characteristics are carried out in many regions all over the globe [6–9]. A network of automated ground-based radiometers for measuring the atmospheric transparency and sky brightness (Aerosol Robotic Network—AERONET) is available and being developed in a number of the countries, including Russia.

Since 2000, the Obukhov Institute of Atmospheric Physics, Russian Academy of Science (IAP RAS), has carried out annual cloud–aerosol–radiation experiments (ZCAREX) at the Zvenigorod Scientific Station (ZSS) (55°42’ N, 36°46’ E). The ZCAREX-2001 was performed in February–March 2001. The ZCAREX-2002 had two active phases: springtime in March–April and summertime in July–September. The 2002 summer in the Moscow region was characterized by a high inflow of smoke aerosol resulting from peatbog and forest fires. The effect of aerosol on the radiation conditions in the atmosphere is estimated quantitatively by using the aerosol radiative forcing (ARF). ARF(z) is defined as the difference between the net solar-radiation fluxes at the level z calculated with and without aerosol.

In this study, the data measured during the ZCAREX-2001 and ZCAREX-2002 are used to estimate the radiative aerosol forcing on the Earth’s surface ARF(0), at the top of the atmosphere ARF(TOA), and over the entire depth of the atmosphere (atmospheric absorption) (ARF(TOA) – ARF(0)) under clear-sky conditions. Instantaneous values of ARF(z) (z is height) are calculated for shortwave and longwave spectral ranges. In this paper, we analyze the sensitivity of ARF(z) to variations in aerosol optical parameters, such as the optical thickness, single-scattering albedo, and the asymmetry factor of the phase function (the mean cosine of the phase function), as well as to variations in the albedo of the Earth’s surface. The influence of aerosol on the radiation regime of a cloudy atmosphere is much weaker than that on the radiation regime of a clear-sky atmosphere if the role of aerosol in cloud formation is not taken into account.
METHODS FOR CALCULATING INTEGRAL SHORT- AND LONGWAVE RADIATION FLUXES USED TO ESTIMATE AEROSOL RADIATIVE FORCING

Integral fluxes in the shortwave spectral range (Δλ = 0.2–0.4 µm) (solar-radiation fluxes) were calculated using the technique described in detail in [10]. This technique takes into account multiple reflections from clouds and the Earth’s surface; molecular scattering; scattering and nonselective absorption by aerosol particles and clouds of various phase compositions; and selective absorption by atmospheric H₂O, CO₂, O₃, and O₂. The spectrum of solar radiation splits into eight intervals: 0.2–0.4, 0.4–0.52, 0.52–0.69, 0.69–0.75, 0.75–0.87, 0.87–1.19, 1.19–2.38, and 2.38–4.00 µm. It is assumed in the technique that selective absorption by the atmospheric gases, nonselective absorption and scattering by aerosol and cloud particles, and molecular scattering are separated. In this regard, integral solar-radiation fluxes are calculated in two stages. The δ-Eddington method is used first. Next, the atmospheric integral transmitance function (ITF) is introduced to take into account the selective absorption of solar radiation by atmospheric gases, such as H₂O, CO₂, O₃, and O₂. The atmosphere is divided into 18 horizontally homogeneous layers. When the atmospheric characteristics in a layer are determined, the corresponding data for all the sublayers of this layer are taken into account (the number of sublayers in each of the 18 layers is determined by the vertical resolution of the initial data). Integral solar-radiation fluxes in a clear-sky atmosphere are calculated with an error of about 1% [10].

Integral fluxes and estimates of the aerosol radiative forcing ARF(z) in the shortwave spectral range under clear-sky conditions are also determined from the ZCAREX-2002 data by using the online Calculator of Integral Solar Fluxes (CSIF), which is accessible on the Internet (www1.imp.kiae.ru/csif). The CSIF code calculates solar fluxes by interpolation over preliminary chosen reference values. The technique for obtaining the latter is described in detail in [11]. Monte Carlo computations are used in combination with a line-by-line procedure for taking into account the absorption spectra of atmospheric gases. In contrast to [11], the algorithm takes into account the NO₂ absorption of solar radiation near 0.4 µm. Additionally, new values of the molecular-scattering optical thickness and the solar constant are used. The aerosol models and the types of surfaces used in reference calculations are described in detail in the reference section of the code.

Integral fluxes in the longwave spectral range (Δλ = 4–40 µm) (longwave-radiation fluxes) are calculated in the same manner as integral shortwave-radiation fluxes by using the ITF method. The ITF takes into account absorption by H₂O (including continuum absorption in the 8–12 µm window), CO₂, O₃, aerosol, and clouds of various types [12–14]. A comparison of reference fluxes with radiation fluxes calculated by the technique of [13] for a standard set of clear-sky atmospheric situations [15] and actual data [12] shows that the error in the calculated integral longwave fluxes is within 2%.

INITIAL DATA USED IN THE ESTIMATION OF AEROSOL RADIATIVE FORCING

The radiation fluxes are calculated using a plane-parallel model of the atmosphere. This involves the ground-based values of temperature, humidity, and pressure at the site of the experiment; the atmospheric moisture content w (g/cm²); and the vertical profiles of temperature, humidity, and pressure obtained by aerological sounding of the atmosphere at Dolgoprudnyi (55°45’ N, 37°57’ E) at 12:00 UTC (http://weather.uwyo.edu/upperair/sounding.html). It follows from [16] that aerological sounding data obtained at Dolgoprudnyi can be used for calculating radiation fluxes from observations made at the ZSS if the synoptic conditions at both sites coincide.

In this study, with allowance for the times of satellite flybys and radiosonde launches, the estimates of ARF(z) obtained from the ZCAREX-2001 measurements are given for afternoon times. The estimates of ARF(z) based on the ZCAREX-2002 data are obtained for the periods listed in Table 1 under the assumption that the atmospheric moisture content w (g/cm²) varies insignificantly over the daylight hours under clear-sky conditions.

A semiempirical model (SEM) is used to describe aerosol. The continental aerosol model cont-1 [17] is used in the atmospheric layer from 0 to 12 km, except for the boundary layer. The model contains the vertical profiles of the aerosol optical thickness at a wavelength of 550 nm, the spectral extinction coefficients, the aerosol single-scattering albedo, and the asymmetry factor of the aerosol phase function. In the atmospheric boundary layer, the aerosol absorption and scattering coefficients coincide with those of surface aerosol, which are measured directly. The monitoring of atmospheric radiation characteristics has been carried out at the ZSS since 2001. The spectral values of total and scattered solar radiation at the Earth’s surface are determined from clear-sky measurements in the visible range (λ = 414.5, 497.3, 613.5, 671.6, and 868.8 nm) performed with a Multi-Filter Rotating Shadowband Radiometer (MFRSR). The magnitudes of these solar-radiation fluxes and w (g/cm²) are used to determine the spectral aerosol optical thickness τₐ(λ), the Angström exponent α in the spectral power-law relation τₐ(λ) = τₐ(λ₀)(λ/λ₀)ᵃ (λ₀ = 550 nm) [18, 19], and the aerosol single-scattering albedo ωₛ(λ) over the entire depth of the atmosphere [20]. The Angström exponent α is calculated from the spectral aerosol optical thicknesses by using the relation α = ln(τₐ(414.5)/τₐ(868.8))/ln(868.8/414.5). It is assumed that variations in the total aerosol optical thickness (in particular, under variations in smoke aerosol loading) result from variations in the aerosol content in the atmospheric boundary layer. It was shown in
that solar-radiation fluxes calculated with and without this assumption are virtually identical. In the calculations, the height of the boundary layer is set equal to 2 km for smoke aerosol and to 1 km for background aerosol.

The absorption and scattering coefficients of surface aerosol at the wavelength \( \lambda_0 = 550 \text{ nm} \) must be measured simultaneously (whenever possible). The absorption coefficient \( \sigma_\alpha (\text{km}^{-1}) \) in the atmospheric surface layer are determined by illuminating aerosol samples collected during the daylight hours [21]. The wavelength dependence of the absorption coefficients is specified in the small-particle approximation: \( \sigma_\alpha (\lambda) = \sigma_0 (\lambda) (\lambda_0/\lambda) \alpha \) (where \( \alpha \) is the Angstrom exponent). If \( \sigma_\alpha (\lambda) \) and \( \sigma_\omega (\lambda) \) are known, we can find the spectral extinction coefficients \( \sigma_e (\lambda) \) and the single-scattering albedo \( \omega_0 (\lambda) \) for surface aerosol. In the near-infrared range, the spectral profiles of the scattering and absorption coefficients are determined using the corresponding power-law parameters indicated above. According to [25], the accuracy of the estimated ARF(z) virtually does not degrade when approximate aerosol optical parameters are specified in the near-infrared spectral range (the aerosol optical thickness in this region is small). If the type of aerosol is unknown, its optical thickness (extinction coefficients) in the boundary layer can be determined using measured values of the aerosol optical thickness over the entire depth of the atmosphere and the vertical profile of the optical thickness in the continental aerosol model cont-1 [17]. The spectral parameters \( \sigma_e (\lambda) \) and \( \sigma_\omega (\lambda) \) are used to calculate \( \sigma_\alpha (\lambda) \) and \( \omega_0 (\lambda) \). Given \( \sigma_\alpha (\lambda) \) and \( \sigma_\omega (\lambda) \), the extinction coefficients \( \sigma_e (\lambda) = \sigma_\omega (\lambda) + \sigma_\omega (\lambda) \) and the aerosol single-scattering \( \omega_0 (\lambda) \) are determined in eight intervals of the shortwave spectral range with a corresponding solar-constant weight. The mean cosine of the aerosol phase function (or the asymmetry factor) \( g(\lambda) \) is estimated using statistical and microphysical models of the aerosol optical characteristics constructed from the measurements conducted at the ZSS [26]. It is assumed that the wavelength dependence of the asymmetry factor is neutral; i.e., \( g(\lambda) = \text{const} \) within 0.2–4.0 \( \mu \text{m} \).

The integral albedo of the Earth’s surface \( A \) is determined from satellite data. The technique for determining surface albedo from first-channel AVHRR/NOAA measurements is described in [27]. The estimates of ARF(z) based on the ZCAREX-2001 data are calculated for \( A \) equal to 0.40. According to aerological sounding, the mean air temperature in March 2001 was below 0°C during daylight hours and did not exceed −9°C at night. The snow cover was observed nearly everywhere at the ZSS. The monthly mean air temperature in March 2002 was higher by more than five degrees than that in March 2001. The estimates of ARF(z) based on the ZCAREX-2002 data were calculated for \( A \) equal to 0.15 or 0.20.

Aerosol absorption in the longwave range is taken into account according to [28]. It follows from [28] that the longwave aerosol radiative forcing (ARF1) in the middle latitudes increases with the aerosol integral absorption coefficient \( a_{\Delta \lambda} (\text{km}^{-1}) (\Delta \lambda = 4–40 \mu \text{m}) \) in the infrared spectral range (in particular, on the Earth’s surface). The influence of aerosol is considerable in the atmospheric boundary layer, where the aerosol concentration is maximal. The values of ARF1 at the top of the atmosphere virtually coincide in the aerosol models under consideration (0.0457 ≤ \( a_{\Delta \lambda} \) ≤ 0.115) [28]. The integral longwave radiation fluxes are determined using an aerosol model that differs from cont-1 [17] only in that \( a_{\Delta \lambda} \) is equal to 0.0500 (\text{km}^{-1}) in the former, while \( a_{\Delta \lambda} = 0.0435 (\text{km}^{-1}) \) in the latter. The estimates of

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Table 1. Minimum and maximum aerosol optical thickness at 550 nm (\( \tau_{\min}(550 \text{ nm}) \), \( \tau_{\max}(550 \text{ nm}) \)) in indicated observation periods

<table>
<thead>
<tr>
<th>Date</th>
<th>Time*</th>
<th>( \tau_{\min}(550 \text{ nm}) )</th>
<th>( \tau_{\max}(550 \text{ nm}) )</th>
<th>( \alpha )</th>
<th>( \omega_{0, 550} )</th>
<th>( g_{550} )</th>
<th>( w )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mar. 1, 2001</td>
<td>14:00, 14:30, 15:00</td>
<td>0.12</td>
<td>0.20</td>
<td>1.60</td>
<td>0.87</td>
<td>0.57</td>
<td>0.28</td>
</tr>
<tr>
<td>Mar. 6, 2001</td>
<td>14:00, 14:30, 15:00</td>
<td>0.04</td>
<td>0.05</td>
<td>1.40</td>
<td>0.87</td>
<td>0.59</td>
<td>0.33</td>
</tr>
<tr>
<td>Mar. 26, 2001</td>
<td>14:00, 14:30, 15:00</td>
<td>0.08</td>
<td>0.10</td>
<td>2.10</td>
<td>0.86</td>
<td>0.53</td>
<td>0.35</td>
</tr>
<tr>
<td>Mar. 19, 2002</td>
<td>13:30–16:00</td>
<td>0.03</td>
<td>0.20</td>
<td>2.00</td>
<td>0.87</td>
<td>0.52</td>
<td>0.48</td>
</tr>
<tr>
<td>July 20, 2002</td>
<td>9:00–18:00</td>
<td>0.09</td>
<td>0.18</td>
<td>1.36</td>
<td>0.87</td>
<td>0.61</td>
<td>2.1</td>
</tr>
<tr>
<td>July 30, 2002</td>
<td>9:00–18:00</td>
<td>0.85</td>
<td>1.45</td>
<td>1.65</td>
<td>0.96</td>
<td>0.58</td>
<td>2.4</td>
</tr>
<tr>
<td>Aug. 15, 2002</td>
<td>9:00–18:00</td>
<td>0.30</td>
<td>0.57</td>
<td>1.68</td>
<td>0.94</td>
<td>0.57</td>
<td>1.9</td>
</tr>
<tr>
<td>Sept. 3, 2002</td>
<td>13:00–17:00</td>
<td>0.14</td>
<td>0.32</td>
<td>1.70</td>
<td>0.97</td>
<td>0.56</td>
<td>1.7</td>
</tr>
<tr>
<td>Sept. 6, 2002</td>
<td>13:32–14:10</td>
<td>0.87</td>
<td>1.02</td>
<td>1.53</td>
<td>0.95</td>
<td>0.59</td>
<td>2.5</td>
</tr>
</tbody>
</table>

Note: \( \alpha \) is the Angstrom exponent, \( \omega_{0, 550} \) is the aerosol single-scattering albedo, \( g_{550} \) is the asymmetry factor of the aerosol phase function, and \( w \) (g/cm²) is the atmospheric moisture content.

* Moscow time.
**Table 2.** Integral fluxes of shortwave radiation $Q_{↓}(z), \quad Q(z) = Q_{↓}(z) - Q_{↑}(z)$ (W/m²) and longwave radiation $F_{↓}(z), \quad F(z) = F_{↓}(z) - F_{↑}(z)$ (W/m²). Measured fluxes $Q_{↓}^{\text{meas}}(0)$ (W/m²) and $F_{↓}^{\text{meas}}(0)$ (W/m²) at the Earth’s surface. Aerosol radiative forcing: shortwave ARF$_1(z)$ (W/m²), longwave ARF$_1(z)$ (W/m²), and integral ARF$_2(z) = \text{ARF}_1(z) + \text{ARF}_1(z)$

<table>
<thead>
<tr>
<th>Air mass</th>
<th>2.37</th>
<th>2.53</th>
<th>2.79</th>
</tr>
</thead>
<tbody>
<tr>
<td>Time</td>
<td>14:00*</td>
<td>14:30*</td>
<td>15:00*</td>
</tr>
<tr>
<td>$Q_{↓}(z)$</td>
<td>452.1</td>
<td>418.5</td>
<td>375.0</td>
</tr>
<tr>
<td>$Q_{↑}(z)$</td>
<td>180.9</td>
<td>167.4</td>
<td>150.0</td>
</tr>
<tr>
<td>$Q(0)$</td>
<td>271.2</td>
<td>251.1</td>
<td>225.0</td>
</tr>
<tr>
<td>$Q(z)(\text{TOA})$</td>
<td>576.4</td>
<td>538.6</td>
<td>489.2</td>
</tr>
<tr>
<td>$Q(z)(\text{TOA})$</td>
<td>213.2</td>
<td>199.9</td>
<td>182.4</td>
</tr>
<tr>
<td>$Q(\text{TOA})$</td>
<td>363.2</td>
<td>338.7</td>
<td>306.8</td>
</tr>
<tr>
<td>$F(z)(0)$</td>
<td>189.7</td>
<td>190.9</td>
<td></td>
</tr>
<tr>
<td>$F_{↓}(0)$</td>
<td>304.1</td>
<td>304.1</td>
<td></td>
</tr>
<tr>
<td>$F(z)(\text{TOA})$</td>
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<td>0.0</td>
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<tr>
<td>$F(z)(\text{TOA})$</td>
<td>213.1</td>
<td>213.1</td>
<td></td>
</tr>
<tr>
<td>$F(\text{TOA})$</td>
<td>–213.4</td>
<td>–213.4</td>
<td></td>
</tr>
<tr>
<td>$Q_{↓}^{\text{meas}}(0)$</td>
<td>435.6</td>
<td>397.2</td>
<td>352.2</td>
</tr>
<tr>
<td>$F_{↓}^{\text{meas}}(0)$</td>
<td>195.5</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Shortwave range: 0.2–4.0 µm

| ARF$(0)$ | –18.8 | –18.7 | –18.3 | –14.6 |
| ARF$(\text{TOA})$ | 1.4 | 0.8 | 0.1 | 0.9 |
| ARF$(\text{TOA}) - \text{ARF}(0)$ | 20.2 | 19.5 | 18.4 | 15.5 |

Longwave range: 4.0–40.0 µm

| ARF1$(0)$ | 1.2 | |
| ARF1$(\text{TOA})$ | 0.3 | |
| ARF1$(\text{TOA}) - \text{ARF1}(0)$ | –0.9 | |

Whole spectrum: 0.2–40.0 µm

| ARF2$(0)$ | –17.6 | | –13.4 |
| ARF2$(\text{TOA})$ | 1.7 | | 1.2 |
| ARF2$(\text{TOA}) - \text{ARF2}(0)$ | 19.3 | | 14.6 |

Note: $\tau_{550}$ is the aerosol optical thickness at a wavelength of 550 nm averaged over the period from 14:00 to 15:00, $\tau_{550} = 0.119$ is the average aerosol optical thickness at 14:00, $\sigma_s$ and $\sigma_{\alpha}$ (km$^{-1}$) are the respective scattering and absorption coefficients, $g_{550}$ is the asymmetry factor at 550 nm for surface aerosol, and $w$ is the moisture content (g/cm²). The integral surface albedo is $A = 0.40$.

* Moscow time.

ARF1$(z)$ obtained using this aerosol model are overestimated as compared with the estimates of ARF1$(z)$ based on cont-1 [17].

**AEROSOL RADIATIVE FORCING DERIVED FROM THE ZCAREX-2001 DATA**

Table 1 presents measured values of atmospheric parameters and optical characteristics of surface aerosol and the periods of clear-sky observations. The aerosol radiative forcing is calculated using these data.

The ZCAREX-2001 data were used to choose the days best covered with information. Clear-sky conditions occurred on March 1 and 6. A cloud cover of 1 was observed on March 26. During the ZCAREX-2001, the aerosol background optical thickness $\tau_{550}$ at a wavelength of 550 nm did not exceed 0.20. The results presented in Tables 2–4 are obtained from the measure-
ments conducted on March 1, 6, and 26, 2001, respectively. The tables list the integral shortwave \( Q_{\downarrow\uparrow}(z) \) and longwave \( F_{\downarrow\uparrow}(z) \) radiation fluxes and the effective fluxes \( Q(z) = Q_{\downarrow}(z) - Q_{\uparrow}(z) \) and \( F(z) = F_{\downarrow}(z) - F_{\uparrow}(z) \) (W/m\(^2\)) on the Earth’s surface \((z = 0)\) and at the top of the atmosphere (TOA) \((z = 100 \text{ km})\). The fluxes \( Q_{\downarrow\uparrow}(z) \) were calculated at the moments 14:00, 14:30, and 15:00 (Moscow time) with \( A = 0.40\). The tables also present the measured radiation fluxes at the Earth’s surface. The integral solar-radiation fluxes were measured using an Eppley Precision Pyranometer. The total measurement error of this instrument does not exceed 3%. The integral longwave fluxes were measured with the help of an Eppley Precision Infrared Pyrgeometer. The maximum error in the measured longwave fluxes is about 5% [29]. The tables also list the estimated aerosol radiative forcing (shortwave (ARF), longwave (ARF1), and in the whole spectrum (ARF2)) on the Earth’s surface and at the top of the atmosphere and atmospheric absorption.

Table 2 lists the radiation fluxes calculated from the measurements performed on March 1, 2001. The calcula-
lations were carried out for two values of $\tau_{550}$. Specifically, $\tau_{550} = 0.119$ was recorded at 14:00 Moscow time, and $\tau_{550} = 0.154$ was obtained by averaging the optical thickness over the period 14:00–15:00 (Moscow time). In this time interval, $\tau_{550}$ ranged from 0.119 to 0.190 and the surface aerosol single-scattering albedo at 550 nm was $\omega_{0,550} = 0.87$. The asymmetry factor of the aerosol phase function was $g(\lambda) = 0.57$. Table 2 shows that the measured and calculated solar-radiation fluxes at the Earth’s surface are in better agreement when the integral radiation fluxes and spectral transparency used to determine the aerosol optical thickness are measured at the same time.

Table 3 lists the estimated values of ARF($\tau$) at the moments 14:00, 14:30, and 15:00 (Moscow time) on March 6 obtained for $\tau_{550}$ equal to 0.045, 0.048, and 0.057, respectively. Here, $\omega_{0,550} = 0.87$ and $g(\lambda) = 0.59$. The small values of ARF($\tau$) are explained primarily by the low aerosol content in the atmosphere.

Table 4 presents the results calculated from the measurements performed on March 26 at 14:00, 14:30, and
The degree of the aerosol effect on the atmospheric radiation regime (characterized by ARF(z) at all the levels) is determined primarily by the amount of solar radiation arriving at the top of the atmosphere, i.e., primarily by the sun’s altitude. Tables 2–4 show that ARF(0) in the shortwave spectral range is negative and ranges from –18.8 to –5.7 W/m², while ARF(TOA) ranges from 0.1 to 3.5 W/m². The aerosol layer impedes the flow of solar energy to the Earth’s surface because part of the energy is reflected from the aerosol layer (albedo effect). In the longwave spectral range, the aerosol layer absorbs part of the longwave radiation emitted by the Earth’s surface and the atmosphere. As a result, the Earth’s surface and the atmospheric layer below the aerosol are heated (greenhouse effect). Tables 2–4 show that ARF1(0) ranges from 0.2 to 1.4 W/m², while ARF1(TOA) ranges from 0.1 to 0.2 W/m². When we use the aerosol model with the only difference from cont-1 [17] being that the former uses a higher value of the integral absorption coefficient ($a_{350} = 0.0500$ km⁻¹), the influence of aerosol on longwave radiation is considerably weaker than that on shortwave radiation. The radiation effect of aerosol in the whole spectrum depends on the ratio between the albedo and greenhouse effects. On the whole, the ZCAREX-2001 data suggest that the atmosphere–surface system is heated: $0.4 \leq$ ARF2(TOA) $\leq 3.7$ W/m².

### AEROSOL RADIATIVE FORCING DERIVED FROM THE ZCAREX-2002 DATA

Test computations were performed to obtain more reliable estimates of ARF(z) in the shortwave spectral range. The radiation fluxes were determined under clear-sky conditions by using the ITF and CSIF techniques for the standard midlatitude winter atmosphere [15] and from the ZCAREX-2002 data. The results are listed in Tables 5 and 6. The discrepancies between the calculated fluxes are within 1–2%.

During the ZCAREX-2002, the data of the ZCAREX-2001 were supplemented with the aerosol single-scattering albedo in the entire depth of the atmosphere [30]. In the case of the SEM aerosol model, this characteristic is determined primarily by the single-scattering albedo in the boundary layer. The aerosol single-scattering albedo in the entire atmospheric depth was estimated using the D–D (diffuse–direct) method [31] from clear-sky direct and diffuse shortwave radiation fluxes measured with an MFRSR instrument at the five wavelengths indicated above. Thus, an opportunity appeared to compare the aerosol single-scattering albedo inferred from radiation measurements with that derived directly by measuring the aerosol optical parameters. The discrepancies between these are about 0.01–0.03. The single-scattering albedo inferred from optical measurements is generally lower than that derived by the D–D method.

The aerosol optical thickness $\tau_{550}$ derived from the clear-sky measurements of spectral transparency performed in the 2002 spring ranges from 0.03 to 0.32. Table 6 lists the radiation fluxes on the Earth’s surface and at the top of the atmosphere as derived from the measurements performed on March 19, 2002 (14:30 Moscow time). Here, the integral longwave-radiation fluxes were calculated according to [13] and the integral shortwave-radiation fluxes were calculated using the CSIF and ITF techniques. The shortwave-radiation fluxes at the Earth’s surface calculated by the ITF and CSIF techniques differ by 7–8 W/m², while the difference between them and the measured fluxes amounts to 20–25 W/m². The discrepancies between the measured and calculated fluxes lie within the measurement and calculation errors. The differences between the calculated and observed integral longwave fluxes at the Earth’s surface are 6–13 W/m². In the shortwave spectral range, the values of ARF(z) obtained by using the two techniques virtually coincide.

In March 2002, under clear-sky conditions, the values of ARF(0) on the Earth’s surface ranged from –46 to –20 W/m². (The surface was cooled.) However, the entire thickness of the atmosphere was heated: 10 W/m² ≤ (ARF(TOA) – ARF(0)) ≤ 29 W/m². The values of ARF(TOA) ranged from –18 to –2 W/m². On the whole, the atmosphere–surface system was cooled [32, 33]. The calculations based on the measurements performed in March 2001 suggest that the atmosphere–surface system was heated [34]. Note that $\omega_{0, 550}$ ranged from 0.86 to 0.87 on March 1, 6, and 26, 2001, and from 0.87 to 0.91 on March 17–19, 2002. The calculations showed that, under cloud-free conditions, the heating or cooling of the atmosphere–surface system (caused, among other things, by the aerosol effect) is determined by the surface albedo and the aerosol single-scattering albedo.

Under smoky conditions, in particular, during the daylight hours on July 30, 2002, the influence of aerosol on the radiation balance at the boundaries of the
atmosphere was 3–4 times higher than that of background aerosol in March 2002 [32]. Recall that a poorly absorbing aerosol ($0.93 \leq \omega_{0.550} \leq 0.98$) [21] was contained in the atmosphere during the smoky period (July–September, 2002).

Figure 1 shows the values of $\tau_{550}$ calculated from the clear-sky measurements of spectral transparency on March 19, July 20 and 30, August 15, and September 3 and 6, 2002. On July 20 during daylight hours, $\tau_{550}$ ranges from 0.80 to 0.195. Such values are characteristic of the background aerosol contents at the ZSS. The differences between the values of the optical thickness from 13:50 to 16:00 (Moscow time) on July 20 and March 19 are no larger than 0.01. During the periods of highest smoke aerosol loadings in July–August and during the smoky period at the beginning of the fall (September 6), the aerosol optical thickness exceeds its background value ($0.50 \leq \tau_{550} \leq 1.45$) by several times. In the second half of the day on July 30, the aerosol optical thickness increases by a factor in excess of 1.5 as compared with its minimum value observed on that day.

Table 6. Integral shortwave-radiation fluxes $Q_{\downarrow}(z)$, $Q(z) = Q_{\downarrow}(z) - Q_{\uparrow}(z)$ (W/m²) computed following the ITF [10] and CSIF [11] methods and the integral longwave-radiation fluxes $F_{\downarrow}(z)$, $F(z) = F_{\downarrow}(z) - F_{\uparrow}(z)$ (W/m²). Measured integral fluxes $Q_{\downarrow}^{\text{meas}}(0)$ (W/m²) and $F_{\downarrow}^{\text{meas}}(0)$ (W/m²) at the Earth’s surface. Aerosol radiative forcing: shortwave ARF(z) (W/m²), longwave ARF1(z) (W/m²), and integral ARF2(z) = ARF(z) + ARF1(z)

<table>
<thead>
<tr>
<th>Mar. 19, 2002, 14:30*</th>
<th>ITF ($\sigma_s = 0.139$, $\sigma_a = 0.018$)</th>
<th>CSIF</th>
</tr>
</thead>
<tbody>
<tr>
<td>Air mass = 2.03</td>
<td>no aerosol ($w = 0.48$)</td>
<td>aerosol (SEM) ($\tau_{550} = 0.13$)</td>
</tr>
<tr>
<td>$Q_{\downarrow}(0)$</td>
<td>525.8 (524.1)</td>
<td>500.2 (498.4)</td>
</tr>
<tr>
<td>$Q_{\uparrow}(0)$</td>
<td>105.2 (78.6)</td>
<td>100.0 (74.8)</td>
</tr>
<tr>
<td>$Q(0)$</td>
<td>420.6 (445.5)</td>
<td>400.2 (423.6)</td>
</tr>
<tr>
<td>$Q_{\downarrow}(\text{TOA})$</td>
<td>673.4</td>
<td>673.4</td>
</tr>
<tr>
<td>$Q_{\uparrow}(\text{TOA})$</td>
<td>143.8 (125.4)</td>
<td>152.8 (131.5)</td>
</tr>
<tr>
<td>$Q(\text{TOA})$</td>
<td>529.6 (548.0)</td>
<td>520.5 (541.9)</td>
</tr>
<tr>
<td>$F_{\downarrow}(0)$</td>
<td>239.9</td>
<td>241.1</td>
</tr>
<tr>
<td>$F_{\uparrow}(0)$</td>
<td>368.0</td>
<td>368.0</td>
</tr>
<tr>
<td>$F(0)$</td>
<td>-128.1</td>
<td>-126.9</td>
</tr>
<tr>
<td>$F_{\downarrow}(\text{TOA})$</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>$F_{\uparrow}(\text{TOA})$</td>
<td>251.5</td>
<td>251.2</td>
</tr>
<tr>
<td>$F(\text{TOA})$</td>
<td>-251.5</td>
<td>-251.2</td>
</tr>
<tr>
<td>$Q_{\downarrow}^{\text{meas}}(0)$</td>
<td>475.0</td>
<td></td>
</tr>
<tr>
<td>$F_{\downarrow}^{\text{meas}}(0)$</td>
<td>251.5</td>
<td></td>
</tr>
</tbody>
</table>

Note: $\sigma_s$ and $\sigma_a$ (km⁻¹) are the respective scattering and absorption coefficients of surface aerosol at 550 nm, and $w$ is the moisture content (g/cm²). The integral surface albedo is $A = 0.20$ (the values computed at $A = 0.15$ are given in parentheses).

* Moscow time.
As $\tau_{550}$ increases, the absolute value of aerosol radiative forcing at the considered levels increases as well. The value of ARF(0) ranges from –180 to –27 W/m$^2$, and ARF(TOA) varies from –95 to –8 W/m$^2$. The absorption of the atmosphere under the influence of aerosol ranges from 13 to 85 W/m$^2$.

SENSITIVITY OF:AEROSOL RADIATIVE FORCING TO VARIATIONS IN AEROSOL OPTICAL PARAMETERS AND SURFACE ALBEDO

The variability in ARF(0), ARF(TOA), and ARF(TOA) – ARF(0) under variations in the aerosol spectral optical thickness in the shortwave spectral range is illustrated in Fig. 5. It shows the possible changes in ARF($z$) if $\tau_{550}$ increases from 0 to 4.5. The extremely high upper value of $\tau_{550}$ was chosen deliberately to reveal the asymptotic behavior of the dependence for values of $\tau_{550}$. The single-scattering albedo and the asymmetry factor of the aerosol phase function were determined from the measurements performed on March 19, 2002. In this case, the Angstrom exponent is $\alpha = 2.00$ and the surface albedo is $A = 0.15$. Figure 5 displays the logarithms of the absolute values of ARF($z$). It can be seen that the sensitivity of the radiative forcing to variations in $\tau_{550}$ for background values of $\tau_{550}$ is higher than that for higher values of $\tau_{550}$. Moreover, the sensitivity of the aerosol radiative forcing to variations in the aerosol optical thickness is much higher at the Earth’s surfaces than at the top of the atmosphere or over the entire depth of the atmosphere.

We also estimated the sensitivity of ARF($z$) to variations in $\omega_{0}(\lambda)$ and $g(\lambda)$. The variations in $\omega_{0}(\lambda)$ and $g(\lambda)$ were chosen so as to cover with excess their ranges observed during the experiments. The results displayed in Fig. 6 were obtained from the measurements performed on March 26, 2001 (14:00 Moscow time, $\cos \theta = 0.451$, $\tau_{550} = 0.090$, $\omega_{0.550} = 0.86$, and $\alpha = 2.1$). (Here, $\alpha$ is the Angstrom exponent and $\theta$ is the solar zenith angle.) The values of $\omega_{0}(\lambda)$ varied from –11 to +11%. The value of ARF(TOA) was calculated for three values of $A$: 0.2, 0.3 and 0.4. Figure 6 shows that the atmosphere–surface system is heated more strongly when the absorption of aerosol is higher (i.e., $\omega_{0}(\lambda)$ is lower). When the surface albedo increases, the heating of the system increases as well. The figure also presents the regression relations between ARF(TOA) and variations in $\Delta g(\lambda)$ (%) and $\Delta \omega_{0}(\lambda)$ (%). The dependence between ARF(TOA) and $\omega_{0}(\lambda)$ is virtually linear. The correlation coefficient between these quantities is roughly equal to unity.

Figure 7 shows ARF($z$) versus $\Delta g(\lambda)$ as derived from the measurements performed on July 30, 2002 (14:00 Moscow time, $\tau_{550} = 1.2$, $\omega_{0.550} = 0.96$, and $g_{550} = 0.58$). The values of $g(\lambda)$ ranged from –50 to +50%. The variability of the aerosol radiative forcing under variations

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**Fig. 1.** Aerosol optical thickness $\tau_{550}$ at the wavelength $\lambda = 550$ nm on March 19, July 20 and 30, August 15, and September 3 and 6, 2002.

**Fig. 2.** Aerosol radiative forcing at the Earth’s surface ARF(0) (W/m$^2$) on March 19, July 20 and 30, August 15, and September 3 and 6, 2002.

**Fig. 3.** Aerosol radiative forcing at the top of the atmosphere ARF(TOA) (W/m$^2$) on March 19, July 20 and 30, August 15, and September 3 and 6, 2002.
in $g(\lambda)$ on the boundaries of the atmosphere is stronger than that over the entire depth of the atmosphere. The regression relations between the aerosol radiative forcing at the considered levels and the asymmetry factor of the aerosol phase function (Fig. 7) suggests that these parameters are related by a linear dependence in the range of actual values of the asymmetry factor. A 1% increase in $g(\lambda)$ yields a roughly 1.3% increase in $\text{ARF}(0)$ and an increase in $\text{ARF(TOA)}$ slightly in excess of 1%. The value of $(\text{ARF(TOA)} - \text{ARF}(0))$ changes roughly by 0.2% when $g(\lambda)$ changes by 1%. Thus, in the case of the SEM aerosol model, the sensitivity of the aerosol radiative forcing on the Earth’s surface to variations in the asymmetry factor is higher than the sensitivity of the aerosol forcing at the top of the atmosphere or over the entire depth of the atmosphere.

Table 7 presents the solar-radiation fluxes and the estimated $\text{ARF}(z)$ as functions of $g(\lambda)$, with cont-1 [17] used as an aerosol model. The results in the table are
In the former case, we used the asymmetry factor of the aerosol optical thickness at an effective wavelength spectral ranges and for the whole spectrum separately. The aerosol radiative forcing in the shortwave spectral range ARF(z) = Q^↓(z) – Q^↑(z) (W/m²) as a function of the asymmetry factor of the aerosol phase function g(λ).

<table>
<thead>
<tr>
<th>No.</th>
<th>Asymmetry factor</th>
<th>z</th>
<th>Q^↓(z) (Q^↑(z))</th>
<th>ARF(0)</th>
<th>ARF(TOA)</th>
<th>ARF(TOA) – ARF(0)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>cont-1[17]: g(λ)</td>
<td>0</td>
<td>460.5 (513.3)</td>
<td>391.4 (436.3)</td>
<td>-44.9</td>
<td>-17.2</td>
</tr>
<tr>
<td>2</td>
<td>g(λ) – 0.1g(λ)</td>
<td>0</td>
<td>456.5</td>
<td>388.0</td>
<td>-48.3</td>
<td>-20.4</td>
</tr>
<tr>
<td>3</td>
<td>g(λ) + 0.1g(λ)</td>
<td>0</td>
<td>464.4</td>
<td>394.9</td>
<td>-41.4</td>
<td>-13.9</td>
</tr>
<tr>
<td>4</td>
<td>g(λ) = g550</td>
<td>0</td>
<td>460.2</td>
<td>391.2</td>
<td>-45.1</td>
<td>-17.4</td>
</tr>
</tbody>
</table>

Note: τ550 = 0.320 is the aerosol optical thickness at a wavelength of 550 nm. The Angstrom exponent is α = 1.95. The air mass is 2.02.

The moisture content is w = 0.84 g/cm². The integral radiation flux measured at the Earth’s surface is Q^↓(0) = 461.8 W/m². The integral surface albedo is A = 0.15.

* Moscow time.

Based on the measurements performed on March 18, 2002 (14:40 Moscow time, cosθ = 0.476, τ550 = 0.30, and ω0.550 = 0.86). We considered two variants of g(λ): a neutral wavelength dependence (i.e., g(λ) = const) and g(λ) corresponding to the continental aerosol model. In the latter case, it was assumed that g(λ) ranges within 10%. In the former case, we used the asymmetry factor g550 at a wavelength of 550 nm. Recall that g(λ) = g550 is used in the SEM aerosol model. In the case of cont-1 [17], a 10% increase (decrease) in the asymmetry factor leads roughly to a 1% increase (decrease) in the fluxes. In this case, the aerosol radiative forcing at the Earth’s surface increases (decreases) nearly by 10%, while the absorption of the atmosphere has a weak response to variations in the asymmetry factor. The values of ARF(z) for a neutral wavelength dependence g(λ) virtually do not differ from those obtained with g(λ) corresponding to cont-1 [17].

**CONCLUSIONS**

(1) The measurements conducted during the experiments ZCAREX-2001 (March–April) and ZCAREX-2002 (March–April and July–September) have been used to obtain quantitative estimates of the aerosol radiative forcing ARF on the Earth’s surface, at the top of the atmosphere, and over the entire depth of the atmosphere under clear-sky conditions. The estimates of ARF(z) were obtained for the shortwave and longwave spectral ranges and for the whole spectrum separately. The aerosol optical thickness at an effective wavelength of λg = 550 nm in March 2001 ranged from 0.03 to 0.20. These values correspond to background aerosol loadings at the IAP RAS ZSS. The first active phase of ZCAREX-2002 in March–April was characterized by background aerosol loadings, with the optical thickness τ550 ranging from 0.04 to 0.32. The second active phase of the ZCAREX-2002 in July–September fell on the period of highly smoky conditions in the Moscow region caused by peatbog and forest fires. Under clear-sky conditions, the values of τ550 ranged from 0.50 to 1.45 during this period. Based on the measurements performed in the ZCAREX-2001, the following estimates were obtained for the shortwave aerosol radiative forcing: –19 ≤ ARF(0) ≤ –6 W/m², 0.1 ≤ ARF(TOA) ≤ 3.5 W/m², and 6 ≤ (ARF(TOA) – ARF(0)) ≤ 20 W/m². The estimates inferred from the measurements conducted in March 2002 are as follows: –46 ≤ ARF(0) ≤ –20 W/m², –18 ≤ ARF(TOA) ≤ –6 W/m², and 10 ≤ (ARF(TOA) – ARF(0)) ≤ 29 W/m². During the smoky period in 2002, the aerosol influence on the radiation regime was much stronger: –180 ≤ ARF(0) ≤ –27 W/m², –95 ≤ ARF(TOA) ≤ –8 W/m², and 13 ≤ (ARF(TOA) – ARF(0)) ≤ 85 W/m². The aerosol effect on the atmospheric radiation regime in the longwave spectral range is much weaker than that in the shortwave spectral range (at least by one order of magnitude if the continental aerosol model cont-1 [17] is used in the estimation). The aerosol radiative forcing in the whole spectrum is determined primarily by the aerosol radiative forcing in the shortwave spectral range.

(2) The degree of the aerosol effect on the atmospheric radiation regime is determined by the following factors: the aerosol optical thickness, aerosol single-scattering albedo, the asymmetry factor of the aerosol phase function, surface albedo, and (primarily) the amount of solar radiation arriving at the top of the atmosphere (mainly, the sun’s altitude). The measurements conducted during the experiments ZCAREX-2001 and ZCAREX-2002 were used to obtain quantitative esti-
matters of the cooling and heating of the atmosphere–surface system for various values of surface albedo. For example, the atmosphere–surface system was cooled in the spring–summer 2002 at the surface albedo $A = 0.15$ and 0.20 and was heated in March 2001 at $A = 0.4$. Additionally, quantitative estimates were obtained for the variation in atmospheric absorption under the influence of atmospheric aerosol. The absolute values of ARF($z$) in a smoky atmosphere (the summer–fall 2002) were found to be several times larger than the corresponding values of ARF($z$) calculated for background aerosol loadings (in March 2002). Under smoky conditions, the aerosol optical thickness exceeded their background values nearly by one order of magnitude. Regression relations between ARF(TOA) and the aerosol single-scattering albedo $\alpha_0(\lambda)$ were obtained for various values of surface albedo: $A = 0.2, 0.3,$ and 0.4. As $\alpha_0(\lambda)$ increases, the effect of aerosol on the radiation balance at the top of the atmosphere weakens. As the surface albedo increases, the atmosphere–surface system is heated, and this effect is more pronounced for higher values of surface albedo.

3. Regression relations were obtained between $g(\lambda)$ (expressed in percent) and the values of ARF($z$) on the Earth’s surface, at the top of the atmosphere, and over the entire depth of the atmosphere. The highest sensitivity to variations in $g(\lambda)$ is exhibited by ARF(0). The absorption of the atmosphere responds relatively weakly to variations in $g(\lambda)$.

4. The sensitivity of ARF($z$) to variations in the optical thickness of background aerosol is much higher than its sensitivity to variations in the optical thickness of the smoke aerosol. The sensitivity of ARF($z$) to variations in the aerosol optical thickness is reduced with increasing aerosol optical thickness.

ACKNOWLEDGMENTS

We are grateful to G.S. Golitsyn for his encouragement of this study and for helpful discussions. We also thank P.P. Anikin, M.A. Sviridenkov, E.V. Romashova, A.A. Isakov, and V.M. Kopeikin for the experimental data placed at our disposal.

This study was supported by the Russian Foundation for Basic Research (project nos. 02-05-64529, 02-05-64573, 04-05-64579) and an ARM grant (contract 5102).

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Translated by I. Ruzanova