Seasonal Variation of Aerosol Direct Radiative Forcing and Optical Properties Estimated from Ground-Based Solar Radiation Measurements

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ABSTRACT
The surface direct radiative forcing and optical properties of aerosols have been analyzed from a ground-based solar radiation measurement, which was made under clear-sky conditions in Tsukuba, Japan, over two years from April 1997 to March 1999. The global and diffuse irradiances in the total and near-infrared (NIR) solar spectral regions were simultaneously measured by using two sets of the total-band and NIR-band pyranometers, respectively. The visible (VIS)-band irradiances were estimated by taking differences between the total-band and NIR-band irradiances. Spectral aerosol optical thicknesses (AOTs) in the air column were also measured, using a sun photometer. By combining the spectral AOTs and the surface diffuse irradiances, a retrieval algorithm for simultaneously estimating the effective aerosol size distribution and imaginary index of refraction ($m_i$) was developed. Seasonal variations of the broadband surface radiative forcings and retrieved optical properties of the colunm aerosols have been studied. A close correlation was found among these parameters with similar features of seasonal variations. In winter the columnar aerosols exhibit the minimum surface radiative forcing and a minimum AOT, but the maximum $m_i$ value of 0.04. The opposite is true in summer, when the minimum $m_i$ value of 0.02 was estimated. The surface radiative forcing in the VIS band was estimated to be almost 4 times larger than in the NIR band. The total-band aerosol forcing efficiency is defined as the change in the surface radiative forcing in the total band due to a unit increase of AOT at 500 nm. This has its largest magnitude of $-219$ W m$^{-2}$ in winter and its smallest magnitude of $-150$ W m$^{-2}$ in summer. The results suggest that the correlated seasonal variations between the aerosol radiative forcing and the optical properties may result from seasonal changes in the dominant aerosol components.

1. Introduction
Atmospheric aerosols substantially affect the radiation budget of the earth–atmosphere system in both direct and indirect ways. The direct effect is directly related to scattering and absorption of solar radiation by aerosol particles (e.g., Charlson et al. 1992; Kiehl and Briegleb 1993). The indirect effect is seen in the way aerosols influence optical properties and the lifetime of clouds through cloud formation processes (e.g., Twomey 1977; Albrecht 1989). In the present paper, we shall concentrate on aerosol direct effects on the solar radiation on the earth's surface. Radiative forcing is an important parameter in assessing the aerosol direct effect on the radiation budget, and consequently many investigators have made estimates of aerosol radiative forcing. These estimates are generally made by model simulations (e.g., Charlson et al. 1992; Kiehl and Briegleb 1993; Mitchell et al. 1995). Recently, extensive and sophisticated surface aerosol-radiation networks such as AERONET (Aerosol Robotic Network: Holben et al. 1998) have been deployed worldwide. Further, several intensive aerosol-radiation field experiments such as the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia; Russell et al. 2002) and the Asian Atmospheric Particulate Environment Change Studies (APEX; Nakajima et al. 2002) have been or are being carried out in the various regions of the globe. The products from these radiation networks and field campaigns will be promising and useful to improve the knowledge on aerosol radiative effects. However, those sophisticated networks and intensive field campaigns are expensive to operate and do not enough yet cover the wide variability of tropospheric aerosols. For example, only a few estimates of aerosol radiative forcing have been made from surface radiation measurements, especially, in the Asian region. From the solar irradiance measurements carried out during the Indian Ocean Ex-
periment (INDOEX) campaign, several authors (e.g., Jayaraman et al. 1998; Meywerk and Ramanathan 1999; Conant 2000; Rajeev and Ramanathan 2001) made estimates of the aerosol forcing efficiency ($\beta$) as well as the aerosol radiative forcing in the Indian Ocean region. The forcing efficiency $\beta$ is defined as the change in radiative forcing per unit change in aerosol visible optical thickness, say, at a wavelength of 500 nm ($\tau_{500}$). It is a useful parameter for more directly assessing the aerosol direct radiative effect. On the other hand, Takayabu et al. (1999) estimated monthly and annual mean values of aerosol radiative forcing from ground-based solar radiation observations. They analyzed the radiation data for the year 1996 routinely measured at the Tateno Aerological Observatory (TAO) of the Japan Meteorological Agency (JMA) located at 36.05°N, 140.13°E in Tsukuba, Japan. They estimated the annual-mean surface radiative forcing to be $-18 \text{ W m}^{-2}$, which corresponds to about 6% of the insolation at the top of the atmosphere. Here, we propose a method to estimate the direct radiative forcing as well as aerosol optical properties by combining the surface solar irradiance observation and sun photometer measurements.

The size distribution and complex refractive index ($m = m_r - im_i$) of aerosols are indispensable parameters to compute their single scattering properties from Mie theory and then to simulate the aerosol effects on solar radiation budget. The imaginary index of refraction ($m_i$) is a key parameter to represent radiant absorptivity of aerosols, and it can influence the sign (heating or cooling) of solar absorption effects by aerosols in the earth–atmosphere system (e.g., Yamamoto and Tanaka 1972; Herman and Browning 1975). However, these parameters have not yet been adequately well documented from field observations because the size distribution and complex refractive index of tropospheric aerosols are extremely variable, both temporally and spatially (e.g., Fitch and Cress 1981, 1983; Tanaka et al. 1983; Shibara et al. 1991; Hayasaka et al. 1992). It is important to document long-term variations such as seasonal and annual variations of the aerosol optical properties at various places.

In this paper, we develop a method to infer the direct radiative forcing and optical properties of aerosols from a ground-based solar radiation measurement and discuss the results. We estimate the surface direct radiative forcing and derive the size distribution and wavelength-mean values of the imaginary index of refraction of aerosols in a vertical air column. It should be noted that the estimated refractive index is an optically equivalent (or effective) value that can reproduce the observed solar diffuse irradiances together with the estimated aerosol size distribution, but it may not necessarily represent the true value of the complex refractive index of aerosol materials. This study has three advantages: first, we estimate the aerosol radiative forcing not only in the total solar spectral region but also in the visible (VIS) and near-infrared (NIR) regions. The solar irradiance measured in the total band can be strongly affected by water vapor absorption that occurs mainly in the NIR region. It is therefore useful to measure separately the VIS-band and NIR-band solar irradiances, in order to discriminate between effects on the surface solar radiation due to water vapor and aerosols. Second, the dataset from the 2-yr-long observation enables us to investigate seasonal variations of the estimated aerosol parameters. Here we focus on seasonal variations, from a statistical point of view, of typical aerosol features in the Tsukuba area. Third, by combining the measurements made with four pyranometers as well as a sun photometer we can estimate not only the aerosol radiative forcing but also such optical properties as the effective size distribution and imaginary index of refraction. Thus, we can investigate the relationship between the estimated surface direct radiative forcing and optical properties of aerosols. Further, the present method has a potential applicability to routinely operated surface radiation measurements at worldwide stations with an extension of spectral aerosol optical thickness (AOT) measurements.

2. Observation and data sampling

Since 1997, the Meteorological Research Institute (MRI), located at 36.05°N, 140.13°E in Tsukuba, Japan, has conducted solar radiation measurements with ground-based pyranometers and a sun photometer. Here we use only the observational data obtained under completely cloudless conditions for time intervals longer than a half day during the period from April 1997 to March 1999. From March to August 1998, the measurements suffered from instrument problems and cloudy weather conditions, and we had only a few favorable days. We analyzed the datasets obtained for a total of 68 days and classified the observational data into four seasons: spring from March to May (13 days), summer from June to August (5 days), autumn from September to November (27 days), and winter from December to February (23 days).

Four ventilated, broadband pyranometers (Kipp and Zonen CM21) were used to measure the surface global and diffuse solar irradiances in the total band (305–2900 nm) and the NIR band (715–2900 nm), respectively, with a sampling rate of one measurement every 10 s. The total-band and NIR-band irradiances were measured by the pyranometers with transparent (Schott WG305) and red (Schott RG715) filter domes, respectively. For the measurement of the diffuse solar irradiances, two (total band and NIR band) pyranometers, installed on an automatic sun-tracking mount, were shaded with shadowing disks. The direct solar irradiances were obtained as the difference between the global and diffuse irradiances measured with the unshaded and shaded pyranometers, respectively. The global, direct and diffuse irradiances in the VIS band (305–715 nm) were obtained from the difference between the corresponding irradiances measured in the total band and the NIR band.
The pyranometers were calibrated once a year by a side-by-side comparison with a standard pyranometer (a Kipp and Zonen CM21) at the JMA Meteorological Instrument Calibration Center in Tsukuba. In the calibration of the NIR-band pyranometers, the RG715 domes were replaced by the WG305 domes, and then the spectral transmittance of the RG715 domes was corrected for the NIR-band irradiance measurement. The spectral and temperature dependence of the filter domes may be a factor causing measurement errors. According to Schott, the temperature dependence of spectral characteristics of the domes is generally very small; however, the edge wavelength ($\lambda_c$) of the RG715 dome is said to shift to longer wavelengths with a rate of $\Delta \lambda_c/\Delta T = 0.15$ (nm K$^{-1}$). The temperature dependence might yield biases of, at most, 5 W m$^{-2}$ in the measured NIR irradiances.

It is well known that broadband pyranometers using thermopile detectors suffer measurement errors and biases due to their cosine-law response (incident-angle dependence), temperature dependence, and the so-called thermal offset (zero offset) (e.g., Bush et al. 2000; Ji and Tsay 2000; Haeffelin et al. 2001; Dutton et al. 2001). We experimentally investigated the characteristics for the cosine-law response and the temperature dependence of the CM21 pyranometers. We found that the errors due to the cosine-law response and temperature dependence were fairly small, and almost the same for the four pyranometers. The cosine response errors were less than 0.5% for incident zenith angles ($\theta_o$) less than 70°, and the temperature dependence errors were at most 1% for the daytime air temperature range of 0°–30°C that was encountered in the observation period. Regarding the zero-offset bias, quite large nighttime biases, sometimes exceeding 10 W m$^{-2}$, have been reported for several Eppley Precision Spectral Pyranometers (PSPs) (Bush et al. 2000; Ji and Tsay 2000; Haeffelin et al. 2001). Haeffelin et al. (2001) and Dutton et al. (2001) showed that clear-sky daytime biases could be larger, as much as double, than the nighttime values for the diffuse irradiances measured by few PSPs. In addition, Dutton et al. (2001) suggested an effectiveness of forced ventilation systems to reduce nighttime and daytime offsets, and they also suggested that daytime offsets of even unventilated CM21 pyranometers could be less sensitive to net thermal infrared exchanges between the detector and domes than those of the ventilated PSPs. Actually, we found much smaller nighttime negative outputs of 2–3 W m$^{-2}$ for all of the ventilated CM21 pyranometers.

Since any further quantitative feature of daytime offsets of CM21-type pyranometers was not available in this study, we suppose that daytime offsets of the present CM21 pyranometers might be not so large and less than 5 W m$^{-2}$.

By considering the aforementioned uncertainties, we estimated the overall relative accuracy of our solar irradiance measurements by using the CM21 pyranometers to be within 2% (at most $\pm 15$ W m$^{-2}$ for the total-band global irradiances) for $\theta_o < 70°$. However, for the irradiance components (direct components and/ or VIS-band components) derived by taking differences of outputs of two corresponding pyranometers, the uncertainty could be reduced by partial cancellation of errors due to the almost identical characteristics and zero-offset biases of the two pyranometers. In particular, the measurement errors for the VIS-band diffuse irradiances could be greatly reduced because of negligibly small cosine response errors for the diffuse irradiance measurements by the shaded pyranometers, as well as a cancellation of zero-offset biases of the pyranometers for measuring the total-band and NIR-band diffuse irradiances.

The sun photometer (EKO Instruments MS-115) was used to measure aerosol optical thicknesses (AOTs) at six wavelengths ($\lambda = 369, 499, 675, 778, 862,$ and 1050 nm), except for the period between August and October 1997, when another sun photometer was used to measure at only five wavelengths ($\lambda = 368, 502, 676, 864,$ and 1050 nm). The sun photometer data were sampled at a rate of one point every 10 s. The calibration constants of the sun photometers were determined every month by comparison with a reference spectroradiometer (Opt Research MSR-7000), which was calibrated once a year at Mauna Loa, Hawaii, by means of the Langley method. The sun photometer calibration constants could involve a relative error as large as 2% in all channels except the 1050-nm channel. This error might introduce an uncertainty of, at most, $\pm 0.02$ in the optical thickness. Since the 1050-nm channel was rather unstable during the period from January to February 1998, the AOT at a wavelength of 1050 nm was estimated from the approximate expression of Angström (1961), using data measured at the other channels during this period.

3. Method of analysis

a. Radiative transfer calculations

1) COMPUTATIONAL SCHEME

For radiative transfer calculations in the atmosphere with and without aerosols, we employed an improved version of the radiative transfer computing scheme originally developed by Asano and Shiobara (1989). The band-by-band calculations of solar irradiances were carried out by means of the doubling and adding method (Lacis and Hansen 1974) assuming plane-parallel atmospheres. The solar spectrum between 300 and 2900 nm was divided into 50 intervals, and the model atmosphere was divided into 31 layers from the surface up to the altitude of 50 km. The gaseous absorption by water vapor, carbon dioxide, oxygen, and ozone were considered. The correlated k-distribution coefficients for water vapor, carbon dioxide, and oxygen molecules were computed by line-by-line calculations (Uchiyama 1992) from the high-resolution transmission molecular ab-
Briegleb et al. (1986) that gave a surface albedo value as a function of solar zenith angle may be within 0.02. This was estimated from the equation used in Frohlich and Shaw (1980) with the depolarization correction by Young (1981). The ground surface was assumed to be Lambertian.

2) Input Data for Molecular Atmospheres

In order to calculate the surface solar irradiances for molecular atmospheres without aerosols, we used the vertical profiles of pressure, temperature, and humidity as measured by radiosondes launched at 0000 UTC from the TAO, located in the neighborhood of the MRI. To take into account time variations of precipitable water vapor, we relied on the sun photometer measurement at the 938-nm water vapor channel. Shiobara et al. (1996) developed a method to estimate water vapor amount from the sun photometry. They estimated the amounts of precipitable water vapor from the sun-photometer-measured transmittance at the 938-nm channel, for which they determined the calibration constant by a modified Langley method (Shiobara et al. 1996). On the other hand, we determined the calibration constant from the output voltages of the sun photometer measurements at 0000 UTC by comparing the measured 938-nm transmittance with that calculated from the humidity data observed by radiosondes launched at 0000 UTC. For ozone, we used the monthly mean total ozone amounts measured with a Dobson spectrophotometer at the TAO. The vertical ozone profile was approximated by a formula of Green (1964).

Since we did not measure the upward solar irradiances, which properly represent the surface albedos of the area, we adopted the values of 0.09 for the VIS band, 0.19 for the NIR band, and 0.14 for the total band from Asano and Shiobara (1989). They carried out aircraft measurements over the Tsukuba area on several fine days in winter and obtained surface albedo values at solar zenith angles ($\theta_0$) around 60° from airborne solar irradiance measurements. In addition, we assumed that these surface albedo values did not vary significantly through the observation period. While it is known that the surface albedo varies temporally and seasonally due to changes of the solar zenith angle as well as surface conditions (e.g., Kondratyev 1969; Li et al. 2002), we believe that the seasonal variation of the surface albedo is not very large in the Tsukuba area. For example, from the vertical profiles of the downward and upward solar irradiances measured by radiometersondes, Asano et al. (1997) estimated the summertime surface albedo in the area and reported a value of 0.14 for the total band. Furthermore, when we restrict our analysis to data obtained at $\theta_0 < 70°$, the possible range of surface albedo change as a function of solar zenith angle may be within ±0.02. This was estimated from the equation used in Briegleb et al. (1986) that gave a surface albedo value as a function of $\mu = \cos \theta_0$. Thus, uncertainties as large as ±0.02 might be involved in the assumed surface albedo values of 0.09 for the VIS band and 0.19 for the NIR band.

3) Vertical Profiles of Aerosols

In the radiative transfer calculation for cloudless atmospheres with aerosols (or turbid atmospheres), such single scattering properties of aerosols as extinction coefficient, single scattering albedo, and phase function must be known. The single scattering properties were calculated from Mie theory for aerosols with the retrieved values for the size distribution and the complex refractive index. Since there was no information available about vertical distribution of aerosols from the present measurements, we relied on the study of Hayasaka et al. (1998) for the vertical distribution of the extinction coefficients, or aerosol concentrations. Note that the extinction coefficient integrated over the whole altitude should be consistent with the AOT calculated from the retrieved size distribution and complex refractive index. Here, the retrieved size distribution and complex refractive index of aerosols were assumed to be constant throughout the aerosol layers. That is, the single scattering albedo and the phase function were assumed to be constant throughout the air column.

b. Estimation of the surface radiative forcing

The surface radiative forcing is usually defined as the difference between the surface net (downward − upward) solar irradiances measured for turbid atmospheres and calculated for corresponding aerosol-free atmospheres. Since we did not measure the upward solar irradiances, we calculated the aerosol net radiative forcing at the surface in the spectral $l$ band, $AF_{net}(l)$, from the following equations:

$$AF_{net}(l) = [1 - \alpha(l)]AF_{vis}(l), \quad (1)$$

$$AF_{obs}(l) = F_{obs}^{↓}(l) - F_{mol}^{↓}(l). \quad (2)$$

In Eq. (1), $\alpha$ denotes the surface albedo, and $AF_{obs}$ is the aerosol radiative forcing defined in terms of the surface downward solar irradiances by the relation of Eq. (2). In Eq. (2), $F_{obs}^{↓}$ is the surface downward irradiance measured for turbid atmospheres, and $F_{mol}^{↓}$ denotes the surface downward irradiance computed for the corresponding molecular atmospheres without aerosols. The signs of the irradiance are defined such that downward is positive. The surface albedos of $\alpha$(VIS) = 0.09 and $\alpha$(NIR) = 0.19 were assumed as discussed above. The uncertainty of ±0.02 in the surface albedo may lead to a relative error of about 2% in the net radiative forcing estimated from Eq. (1).
c. Estimation of the microphysical properties of aerosols

1) RETRIEVAL ALGORITHM

We have developed a simultaneous retrieval method of effective aerosol size distributions and imaginary indices of refraction by the combined use of the spectral AOTs and the VIS-band diffuse irradiances. The retrieval method is schematically illustrated in Fig. 1. Assuming that aerosols are homogeneous spherical particles, the size distributions of aerosols in a vertical air column can be retrieved from spectral AOTs measured by the sun photometer using the so-called inversion method (e.g., Yamamoto and Tanaka 1969; King et al. 1978). The present inversion code is the same as that developed by Asano et al. (1985, 1993) to estimate the size distributions of volcanic aerosols. In the present case, however, the particle size range was limited between 0.05 and 3.0 μm: the range of sizes was divided into eight bins of equal width in units of log(radius). Among various choices of size limits and number of bins, the above values were selected as suitable for the present input data. From several simulation calculations, the reliable range of sizes of the retrieved size distributions was estimated to be between 0.1 and 1.0 μm for this inversion scheme.

King and Herman (1979) demonstrated that spectral values of the imaginary index of refraction for columnar aerosols, together with the surface albedo, could be estimated from the ratio of the values of spectral diffuse irradiance to direct irradiance, measured at the surface. Extending the King and Herman method to direct and diffuse broadband irradiances measured in the 300–4000-nm region, Nakajima et al. (1996) estimated wave-length-mean values of the imaginary index of refraction of aerosols in the Iranian region after the Gulf War in 1991. In the present study, by modifying the method of Nakajima et al. (1996), we tried to estimate band-mean values of the imaginary index of refraction from the diffuse irradiances measured in each spectral band. Here we assumed that surface diffuse irradiance may be primarily determined by the imaginary index of refraction of aerosols, as discussed by Nakajima et al. (1996), under given AOTs and water vapor amount. We used the measured direct irradiances as a performance check of the data analysis.

The data analysis method takes the following steps shown in the flowchart of Fig. 1, including an iteration procedure shown in Fig. 2. First, the complex refractive index of the aerosols was assumed to be constant in each spectral band, and the real index of refraction was fixed at 1.52. The issues due to this assumption are discussed in the next subsection. By presetting seven values for the complex refractive index $m_j$ ($j = 1$ to 7) as $m_1 = 1.52 - 0.00i$, $m_2 = 1.52 - 0.01i$, $m_3 = 1.52 - 0.02i$, $m_4 = 1.52 - 0.03i$, $m_5 = 1.52 - 0.05i$, $m_6 = 1.52 - 0.07i$, and $m_7 = 1.52 - 0.10i$, the size distribution $s_j$ ($j = 1$ to 7) are derived for each value $m_j$ of the complex refractive index.

Next, we calculate the downward diffuse irradiance for each set of $(m_j, s_j)$. By comparing the computed diffuse irradiances with the measured ones, we select two preset complex refractive indices (e.g., $m_2$ and $m_3$).
of \( j = 2 \) and \( 3 \), as shown in Fig. 2) that give the two closest values to the observed irradiance: one (e.g., \( m_2 \)) gives a larger diffuse irradiance and the other (e.g., \( m_3 \)) gives a smaller irradiance than the observed one, respectively. The first approximation of a true refractive index value \( m_{i1} \) can be linearly interpolated from the two preset complex refractive indices (\( m_j \); e.g., \( j = 2 \) and 3 in Fig. 2).

Using the estimated true refractive index \( m_{i1} \), we estimate the corresponding size distribution \( s_{i1} \) from the sun-photometer-measured spectral AOTs, and recalculate the diffuse irradiance for the set of \((m_{i1}, s_{i1})\). When the newly computed diffuse irradiance for \((m_{i1}, s_{i1})\) is smaller than the measured one, another set of the complex refractive indices of \( m_{i1} \) and \( m_{i2} \) can be used to interpolate a better true refractive index value \( m_{i2} \). A similar procedure can be repeated to find the best combination of complex refractive index and size distribution that satisfied the convergence criterion,

\[
|F_{\text{obs},df} - F_{\text{est},df}^0| < \delta. \tag{3}
\]

In the above, \( F_{\text{obs},df} \) denotes the diffuse irradiance computed for the effective set of \((m, s), F_{\text{est},df}^0 \) is the observed diffuse irradiance, and \( \delta \) is a certain threshold.

2) Error estimation

From the ground-based measurements of the spectral AOTs and the VIS-band diffuse irradiances, we have simultaneously estimated the best combination of size distribution and imaginary index of refraction of aerosols that can reproduce the observed diffuse solar irradiance and aerosol optical thicknesses at the selected wavelengths, under the assumption of the fixed values of \( m_r \), and surface albedo. King and Herman (1979) and Nakajima et al. (1996) studied the sensitivity of the surface diffuse irradiance to the complex refractive index and concluded that the diffuse irradiance is strongly sensitive to the imaginary index of refraction \( m_i \), but rather insensitive to the real index \( m_r \). From the observational studies such as King (1979), Tanaka et al. (1983), and Hayasaka et al. (1992), it is known that values of \( m_i \) and \( m_r \) of most tropospheric aerosols generally fall in the ranges \( 1.47 < m_i < 1.57 \) and \( 0.001 < m_r < 0.1 \), respectively, in the solar spectral region. We investigated uncertainties involved in our estimation of the effective value of \( m_i \) when \( m_i \) changed within a range of \( 1.47 < m_i < 1.57 \) instead of the fixed \( m_i = 1.52 \). The results revealed that only small variations in the computed diffuse irradiances within a range of \( \pm 3 \) W m\(^{-2} \) occurred for different values of \( m_i \), and that the variations of the computed diffuse irradiances may lead to an uncertainty of, at most, \( \pm 0.005 \) in the estimate of effective \( m_i \) value. Since the uncertainty was rather small, we assumed the fixed value of \( m_i = 1.52 \) throughout the present analysis.

In the convergence criterion Eq. (3), we set \( \delta = 1 \) W m\(^{-2} \) for the VIS-band retrieval because the VIS-band surface irradiances could be measured quite accurately within 1% (relative error), and the maximum and minimum values of the measured VIS-band diffuse irradiances were 240 and 40 W m\(^{-2} \), respectively, throughout the observation period. However, since \( F_{\text{est},df} \) might involve a larger uncertainty of several W m\(^{-2} \), we investigated the sensitivity of the \( m_i \) retrieval to a larger convergence threshold by supposing \( \delta = 5 \) W m\(^{-2} \). We found that uncertainties involved in the estimated effective \( m_i \) values depended on AOT and the \( m_i \) value itself with larger uncertainties for smaller AOT and larger \( m_i \) values, and vice versa. For an average case of \( \tau_{000} = 0.3 \) and \( m = 0.03 \) in the observation period, the uncertainty could be as large as \( \pm 0.012 \).

Further, the surface albedo can also affect the surface diffuse irradiances. However, as already discussed by King and Herman (1979) and Nakajima et al. (1996), the sensitivity of diffuse irradiance to the surface albedo is rather weak compared to the sensitivity to \( m_i \). In the present study, we assumed a constant surface albedo of \( \omega_{\text{VIS}} = 0.09 \) and \( \omega_{\text{NIR}} = 0.19 \) as stated above. When an uncertainty of \( \pm 0.02 \) was introduced into the VIS-band surface albedo, that is, \( \omega_{\text{VIS}} = 0.09 \pm 0.02 \), the surface albedo uncertainty could introduce rather small uncertainties of, at most, \( \pm 0.003 \) into the estimation of effective \( m_i \) value.

All the aforementioned sensitivity tests and uncertainties involved in the estimation of the effective \( m_i \) values are case dependent, having larger uncertainties for those cases with smaller AOT and larger \( m_i \), and vice versa. In the Tsukuba area, the former cases were more general in winter, and the latter cases in summer. Thus, the overall uncertainties in the retrieved VIS-band \( m_i \) values (see Fig. 9) can be estimated to be about \( \pm 0.017 \) in winter and \( \pm 0.008 \) in summer.

4. Results

a. Radiative forcing on the surface solar radiation

Figure 3 compares the measured direct and diffuse solar irradiances \( F_{\text{obs}} \) with those calculated for the corresponding aerosol-free atmospheres \( F_{\text{mol}} \), as a function of \( \mu = \cos \theta_s \) with solar zenith angle \( \theta_s \), for the whole observation period. Here \( F_{\text{mol}} \) was calculated using the datasets taken every hour from 2100 to 0900 UTC and at \( \theta_s < 70^\circ \), having carried out the radiative transfer calculation under the assumption of plane-parallel atmospheres. The figure shows that the direct components of \( F_{\text{mol}}^d \) in both the VIS band and NIR band linearly increase with increasing \( \mu \). However, \( F_{\text{mol}}^d \) for the NIR band displays a larger dispersion than that for the VIS band. The large dispersion in the NIR band might have resulted from variations in the amount of precipitable water vapor for which the estimated values varied between 0.3 and 4.5 g cm\(^{-2} \) during the observation period.

The aerosol radiative forcing defined by Eqs. (1) and
Figs. 3 and 4 show three evident features about the aerosol radiative forcing. First, the magnitudes of radiative forcing in the VIS band are larger than those in the NIR band. The ratio \([AF_{\text{vis}}(\text{NIR})/\mu]/[AF_{\text{vis}}(\text{VIS})/\mu]\) averaged over the observation period is 0.29 (0.17) for the global component, 0.43 (0.14) for the direct component, and 0.58 (0.16) for the diffuse component. Here, the value in parentheses indicates the standard deviations. For the net radiative forcing, the averaged ratio of \([AF_{\text{net}}(\text{NIR})/\mu]/[AF_{\text{net}}(\text{VIS})/\mu]\) is 0.26 (0.15); the surface net radiative forcing in the VIS band is almost 4 times larger than that in the NIR band. This larger VIS-band radiative forcing is primarily due to larger AOTs in the VIS region than in the NIR region. Figure 6 shows the time variation of the aerosol optical thickness of
Fig. 5. As in Figure 4 but for the global component of the normalized net radiative forcing (AF$_{net}$/µ) in the VIS band and NIR band.

Fig. 6. Temporal variation of the daily-mean aerosol optical thickness (τ) at wavelengths of 500 and 1050 nm. The τ values at λ = 500 nm are those measured at λ = 499 or 502 nm.

τ$_{500}$ and τ$_{1050}$ were measured at λ = 1050 nm. In the figure, τ$_{500}$ and τ$_{1050}$ are daily-mean values obtained over the whole period. The ratio of τ$_{1050}/τ_{500}$ was 0.41 (0.10) averaged over the period. The value in parentheses again indicates the standard deviation. A further contribution to this larger VIS-band radiative forcing may be the difference of strength of aerosol absorption in the VIS band and NIR band. The absolute ratio of the aerosol radiative forcing for the downward diffuse irradiance to that for the direct irradiance is called the effective scattering efficiency $e$, where $e = |AF_{dw,d}/AF_{dw,dr}|$. This is an important parameter that represents the strength of aerosol absorption (Jayaraman et al. 1998). The effective scattering efficiency is about 0.85 for nonabsorbing aerosols at $θ_u < 60°$ (Braslau and Dave 1973) and has smaller values for more-absorbing aerosols. The present value of 0.48 in the VIS band is smaller than the value of 0.70 in the NIR band; this implies that the effect of aerosol absorption on radiative forcing is stronger in the VIS band than in the NIR band. In addition, since water vapor may saturate the attenuation at some wavelengths in the NIR region, there may be no sensitivity to aerosols at those wavelengths.

The second feature shown in Figs. 4 and 5 is that the surface radiative forcing generally depends nonlinearly on τ$_{500}/µ$, but it has almost linear dependence to τ$_{500}/µ$ when τ$_{500}/µ$ is smaller than about 0.8. Such a linear relationship between aerosol radiative forcing and AOT has also been reported from other studies (e.g., Jayaraman et al. 1998; Meywerk and Ramanathan 1999), and the aerosol forcing efficiency $β$ has been introduced to represent the magnitude of aerosol effects on the radiative forcing as the slope of a linear regression equation. The aerosol forcing efficiency at the surface represents the change of the clear-sky surface irradiances for a unit increase of aerosol optical thickness. Jayaraman et al. (1998) estimated $β$ values by restricting their cases to τ$_{500} < 0.4$ and $θ_u < 60°$. They suggested that an exponential equation according to the Bouger–Beer–Lambert law may be more appropriate to fit data up to larger τ$_{500}/µ$ values than is a linear equation. In the present study, for τ$_{500}/µ < 0.8$, we estimated $β$ values from a linear fitting as

$$\frac{AF}{µ} = β \frac{τ_{500}}{µ}. \quad (4)$$

where AF represents AF$_{dw}$ or AF$_{net}$. Further, we fitted all of the radiative forcing data to the following exponential equation as

$$\frac{AF}{µ} = γ \left[ 1 - \exp \left( - \frac{τ_{500}}{µ} \right) \right]. \quad (5)$$

where AF again represents AF$_{dw}$ or AF$_{net}$. The curves in Figs. 4 and 5 indicate the fitted exponential curves. The estimated $β$ and $γ$ values are summarized in Table 1. The larger magnitudes of $γ$, compared to $β$, are due to a rather strong nonlinear dependence of the surface solar irradiances on the normalized AOTs. The $β$ value of $-220$ W m$^{-2}$ for the global component in the total band means a decrease of $22$ W m$^{-2}$ in the normalized downward global irradiance with an increase of 0.1 in the normalized midvisible AOT; that is, Δτ$_{500}/µ = 0.1$. The decreased amount is the result of a partial compensation by an increase (26 W m$^{-2}$) in the normalized diffuse irradiance to the larger decrease (48 W m$^{-2}$) in the direct irradiance following the increase of Δτ$_{500}/µ$ to 0.1. For the surface net radiative forcing, the VIS-band forcing efficiency of $β = -162$ W m$^{-2}$ is almost 5 times larger than that in the NIR band ($β = -34$ W
discuss the seasonal variation of aerosol optical properties scattering strength in the visible radiation. We will further VIS band in the summer season due to their effective efficiency and larger optical thickness (see Fig. 6) in the winter season. The particles may bring smaller forcing in the dominant aerosol components with season over the optical properties could be different due to differences autumn. The seasonal variations suggest that the aerosol to spring and the minimum during the period summer to with the maximum magnitude during the period winter is less evident and differs from that of the VIS band, is featured for the total-band forcing efficiency. (b) Seasonal variation of aerosol optical properties in the next section. Note that the dispersion seen in Figs. 4 and 5 is little due to the normalization method using solar zenith angle by \( \Delta \tau / \mu \). Through the model calculations, we investigated effects caused by the normalization method applied to the VIS-band and NIR-band radiative forcing. In the simulation, the aerosol single scattering albedo and phase function were fixed to the mean values estimated for the whole observation period, and the values of \( \Delta \tau / \mu \) were limited between 0.3 and 0.8. We compared the normalized radiative forcing calculated, by changing the solar zenith angles, for the fixed values of aerosol optical thickness with those calculated, by changing optical thickness, for the fixed values of air mass (solar zenith angle). The result showed that the differences between the computed normalized radiative forcing were less than \( 8 \, W \, m^{-2} \) in the VIS band and less than \( 5 \, W \, m^{-2} \) in the NIR band. Therefore, the normalization method could not cause such large scatter of data points as seen in Figs. 4 and 5.

### b. Seasonal variation of aerosol optical properties

The size distributions and the band-mean imaginary indices of refraction \( m \) in the VIS band were retrieved for aerosols in a vertical air column, every hour from 2100 to 0900 UTC. In the present analysis, we limited the retrieval of \( m \) to the VIS band because the reconstructed AOT in the NIR region was overestimated. This overestimation introduced a significant underestimation of the computed direct solar irradiances in the NIR band compared to those observed, as shown in Fig. 7. The main cause of AOT overestimation in the NIR region might be a rather unstable calibration constant of the sun photometer for the 1050-nm channel as well as a lack of measurement of AOT in the 1050–2900-nm spectral region. The overestimated AOT in the NIR region could affect the retrieved size distribution.
AOT in the NIR region may be effective in the retrieval of larger particles but is less sensitive to smaller particles. Actually, the calculated and observed values of direct irradiances in the VIS band, where AOT is mainly affected by small particles, shows excellent agreement within small rms error of 6.6 W m\(^{-2}\), which is well within the aforementioned measurement accuracy of the CM21 broadband pyranometers.

Figure 8 shows the mean volume size distributions of the aerosols, obtained by averaging many retrieved size distributions in each season. As mentioned in many other published studies (e.g., Fitch and Cress 1981; Shiobara et al. 1991), tropospheric aerosols generally exhibit a bimodal size distribution having an accumulation mode with a mode radius of order submicron, and a coarse mode with a mode radius of order micron. In Fig. 8, the aerosol concentration in the size distribution was minimum around the radius \(r = 0.4\ \mu m\), except for the summer case, for which the minimum appears around \(r = 1.0\ \mu m\). In summer, the concentration in the accumulation mode around \(r = 0.2\ \mu m\) was higher compared with that in other seasons: the accumulation mode concentration was lowest in winter. On the other hand, the coarse-mode concentration was somewhat smaller in summer than in other seasons.

Figure 9 shows the seasonal variation of the daily-mean imaginary index of refraction \(m_i\) in the VIS band. It is shown that the \(m_i\) values were maximum in winter and minimum in summer. This is similar to that reported by Tanaka et al. (1983) and Hayasaka et al. (1992) for aerosols in the Sendai area (38.25°N, 140.83°E); they estimated the \(m_i\) values at a midvisible wavelength using a polar nephelometer. The present seasonal-mean value of \(m_i\) was estimated to be 0.03 in spring, 0.02 in summer, 0.035 in autumn, 0.04 in winter, and 0.035 over the whole period. On the other hand, from airborne mea-
Fig. 8. Seasonal-mean volume size distributions of aerosols in a vertical air column. The vertical error bars indicate the standard deviations for each season average.

Fig. 9. As in Fig. 6 but for the VIS-band-mean imaginary index of refraction ($m_i$).

Measurements of the vertical profiles of the upward and downward solar irradiances in the total band in the lower troposphere, Asano (1989) estimated $m_i = 0.03 \pm 0.02$ for aerosols over the Tsukuba area in the winters of the mid-1980s.

We also estimated the seasonal variation of single scattering albedo of aerosols. The single scattering albedo is another important parameter to represent solar absorption effects of aerosols. However, it may not be practical to directly infer the single scattering albedo only from the ground-based measurements of spectral AOTs and VIS-band diffuse irradiances. This is because the surface diffuse irradiances are affected not only by single scattering albedo but also asymmetry factor of aerosols, even for an aerosol layer with a given AOT. Figure 10 shows the seasonal variation of the daily-mean single scattering albedo at the wavelength of 500 nm, which was calculated from the retrieved size distribution and imaginary index of refraction. The uncertainties in the retrieved values can be estimated to be about $\pm 0.07$ in winter, $\pm 0.04$ in summer, and $\pm 0.06$ for an average case of $\tau_{500} = 0.3$ and $m_i = 0.03$ in the
observation period. The feature of seasonal variation for the single scattering albedo is similar, but opposite, to that for the imaginary index of refraction, that is, minimum in winter and maximum in summer. The seasonal-mean value of the single scattering albedo was estimated to be 0.79 in spring, 0.87 in summer, 0.77 in autumn, 0.69 in winter, and 0.75 over the whole period. These values are rather small, compared to the values estimated from the AERONET data for several key aerosol types (e.g., Dubovik et al. 2002), and the INDOEX campaign for aerosols in the tropical Indian Ocean area (Satheesh et al. 1999), and the APEX campaign for aerosols at Amami-Oshima island (24.4°N, 129.7°E) in the southern East China Sea (Ohta et al. 2001). However, Hayasaka et al. (1992) reported similar values of 0.80 in spring, 0.93 in summer, 0.75 in autumn, 0.69 in winter for the seasonal-mean single scattering albedo at λ = 632.8 nm for aerosols in the Sendai area. These results suggest that aerosols in the Japan area might be more solar absorptive, compared to aerosols in the other regions; however, more comprehensive studies will be necessary before a concrete conclusion on this issue can be reached.

5. Discussion

a. Comparison of the surface radiative forcing

In this section, we compare the present surface radiative forcing, in particular aerosol forcing efficiency, with that obtained from the other observational studies. Only a few studies have estimated the radiative forcing of aerosols in Asia from ground-based solar irradiance observations. In the INDOEX campaign, Jayaraman et al. (1998) estimated the forcing efficiency β in the spectral band of 280–780 nm for τ₅₅₀/µ < 0.8 from shipboard observations over the Indian Ocean and the Arabian Sea (−5°–20°N, 60°–76°E), through January and February 1996. For the global component of the normalized surface downward radiative forcing AFₘₛ₋ₐ/µ, they obtained β values of −285 W m⁻² per unit increase of τ₅₅₀/µ, near the coast of India, and −80 W m⁻² in the central region over the Arabian Sea and the Indian Ocean. They also estimated effective scattering efficiencies of e = 0.5 near the coast of India and e = 0.77 for the interior ocean region. From the lower values of e near the coast of India (representative of very turbid atmospheres), compared to those in the open ocean region (representative of clean maritime atmospheres), they pointed out that the coastal aerosols might be more absorbing compared to aerosols in the open ocean region. We obtained β = −178 W m⁻² for the VIS-band global component and e = 0.48 for the VIS-band downward radiative forcing. The present value of e is close to that obtained by Jayaraman et al. (1998) near the coast of India. However, the magnitude of the present β value is much smaller than that which they obtained in the same region. This is due mainly to the narrower wavelength range of the VIS-band irradiances measured in the present study (λ = 300–715 nm) compared to those used by Jayaraman et al. (1998), (λ = 280–780 nm); the VIS-band insolation at the top of the atmosphere at θₕ = 0° in the present study is about 90 W m⁻² smaller than that for Jayaraman et al. (1998).

Further, Conant et al. (2000) estimated the surface radiative forcing in the spectral band 400–700 nm from surface solar irradiances measured during February and March 1998 at the Kaashido Climate Observatory (4.96°N, 73.47°E), the Republic of Maldives, during the INDOEX campaign. They estimated the mean value of e = 0.7 averaged over the observation period. These results suggest that aerosols over Kaashido Island were less absorptive for visible solar radiation than were the aerosols near the coast of India and over the Tsukuba area.

In the Japan area, the work by Takayabu et al. (1999) is, at present, the only published estimation of the surface radiative forcing of aerosols (but without the forcing efficiency) obtained from routine solar radiation measurements. They estimated a value of −18 W m⁻² for the year-averaged, daily-mean net radiative forcing. Unfortunately, however, the present results could not be directly compared with their estimation because the present aerosol radiative forcing was evaluated from the limited number of clear day measurements, so it was hard to accurately estimate a year-averaged, daily-mean value.

b. The seasonal variation of aerosol components

The present study suggests that the seasonal variations of the estimated aerosol optical properties and surface radiative forcings were correlated and that they...
The seasonal variation of aerosol optical properties estimated in the present study could result from differences in the dominant aerosol components over the Tsukuba area, depending on the season. It is also of interest to know what aerosol components are dominant in each season and what processes can affect the seasonal variation of the dominant aerosol components. In this section, we discuss the seasonal variation of aerosol components over the Tsukuba area from the point of view of the seasonal features of the aerosol optical properties estimated in the present study.

The meteorological data such as wind direction, wind speed, and relative humidity can help the discussion. Figure 11 shows the distribution of wind direction and wind speed observed in each season, and Table 3 gives the seasonal mean values of the wind speed and relative humidity. The surface meteorological data were obtained at the MRI.

The highest particle concentration in the accumulation mode in summer could be partly caused by local generation of small particles due to the gas-to-particles conversion process under the summer weather conditions of abundant sunshine and high relative humidity (see Table 3). Further, Hayasaka et al. (1998) pointed out that the spreading or transport of hazy atmospheres from the Tokyo metropolitan area might enhance the aerosol loading in the summer season over the Tsukuba area. This is because the MRI is located about 50 km
northeast of the Tokyo metropolitan area and the prevailing wind is generally southerly in summer, as shown in Fig. 11. Since most of particles transported from the Tokyo metropolitan area are considered to be anthropogenic aerosols, which are generally dominated in the accumulation mode, this transportation also affects the high concentration of accumulation mode particles found over the Tsukuba area. Thus, locally generated and transported small particles consist mainly of anthropogenic aerosols such as sulfate and nitrate particles as well as soot particles. The relatively smaller values of the VIS-band mean imaginary index of refraction $m_i$ estimated in summer suggest that the fractions of such weakly absorbing aerosols as sulfate and nitrate particles compared to strongly absorbing soot particles are relatively larger in summer than in the other seasons. Furthermore, water uptake by hygroscopic aerosols in accumulation mode could also contribute to the larger values of the aerosol optical thickness and single scattering albedo in summer under the higher relative humidity of 70% (Table 3).

On the other hand, the largest values of $m_i$ in winter suggest an enhancement of fractions of strongly absorbing aerosols such as dust and soot particles. Actually, Takayabu et al. (1999) estimated the weight fraction of soot particles to be about 5%–10% in summer and 10%–20% in winter for aerosols over the Tsukuba area. The local emission of soot particles might increase due to more combustion of fuels in winter than in the warmer seasons. Also, there might be a possibility of long-range transport of absorbing aerosols. Kaneyasu et al. (2000) suggested that carbon-rich aerosols frequently flow off the East Asian continent and pass over the Japanese islands in winter. In late autumn we have encountered several cases for which very large $m_i$ values, exceeding those in winter, were retrieved. This feature is consistent with the result of Takayabu et al. (1999), who reported that the soot weight fraction from October to December was larger than that from January to February 1996 over the Tsukuba area. They attributed the large soot fraction in the late autumn season to local biomass burning after the rice harvest.

The reason that the amount of coarse particles in summer was less than in other seasons can be interpreted as follows. In summer, rather calm winds and high precipitation (see Table 3) might weaken the local emission of dust particles. On the other hand, in spring the amount of coarse particles was significantly larger than in other seasons. Bimodal size distributions with enhanced coarse mode particles have frequently been observed around Japan in spring (e.g., Tanaka et al. 1989; Shibara et al. 1991; Hayasaka et al. 1998). The enhancement of coarse particles is mainly caused by local emission of dust particles and pollen, as well as by long-range transportation of wind-blown dust particles, known as the Kosa (yellow sand) event, from the East Asian continent. In the present study, a typical Kosa event was reported on 2 March, 1999, when the estimated volume size distribution in the 0.6–2.0-μm radius range was about 5 times larger than for the spring-mean size distribution. The larger amounts of dust particles together with a larger soot fraction in spring compared to summer might contribute to the higher $m_i$ values.

6. Summary

Since 1997, precise ground-based solar radiation measurements using four broadband pyranometers and a sun photometer have been carried out at the Meteorological Research Institute in Tsukuba, Japan. By analyzing the observational data obtained between April 1997 and March 1999 under completely clear-sky conditions, we have estimated the direct radiative forcing on the surface solar radiation as well as size distributions and the VIS-band imaginary indices of refraction for columnar aerosols. The seasonal variations of aerosol radiative forcing and optical properties are discussed in this paper.

The direct radiative forcing to the surface downward solar irradiances was estimated as the difference between the measured downward irradiances and those computed for the corresponding aerosol-free atmospheres at a solar zenith angle $\theta_s < 70^\circ$. In the radiative-transfer calculation, we utilized vertical temperature and humidity profiles measured from radiosondes launched at a neighboring location, as well as water vapor amounts estimated from sun photometer measurements. We also estimated the aerosol net radiative forcing to the net global irradiances at the surface by assuming constant surface albedos during the whole period. We further estimated the aerosol forcing efficiency $\beta$, which represents an increased magnitude of the radiative forcing with an increase of visible AOT, for the different spectral bands and solar radiation components. We restricted the analysis to cases with normalized visible AOTs of $\tau_{	ext{vis}}/\mu_s < 0.8$ in order to reduce the nonlinear dependence of solar radiation on $\tau_{	ext{vis}}/\mu_s$. By combining the spectral AOTs measured by the sun photometer and the measured VIS-band diffuse irradiances, we have developed an algorithm for simultaneously retrieving size distributions and VIS-band mean values of the imaginary index of refraction of columnar aerosols. Errors involved in the retrieval were discussed. The highlighted results are as follows.

1) The surface net radiative forcing in the VIS band, averaged over the whole period, was almost 4 times larger than that in the NIR band. The period-averaged aerosol forcing efficiency for the net global irradiances in the VIS band ($\beta = -162$ W m$^{-2}$) was also about 5 times larger than that in the NIR band. The sum of forcing efficiencies in the VIS band and NIR band comes to the total-band forcing efficiency $\beta = -196$ W m$^{-2}$. The VIS-band forcing efficiency has an evident seasonal variation with a maximum magnitude of $-180$ W m$^{-2}$ in winter and a minimum...
magnitude of $-134 \text{ W m}^{-2}$ in summer; the total-band forcing efficiency has a similar seasonal variation to that for the VIS band. For the NIR band, the seasonal variation of the forcing efficiency is less evident, and different from that in the VIS band, with the maximum magnitude occurring over winter and spring and the minimum over summer and autumn.

The feature of seasonal variations over the Tsukuba area suggests that the aerosol amounts and their optical properties are different due to different dominant aerosol components depending on the season.

2) The volume size distributions retrieved from the sunphotometer-measured spectral AOTs generally exhibit bimodal profiles typical of tropospheric aerosols; however, the specific size distributions differed with the season. In summer, the accumulation-mode particles were enhanced with a maximum volume concentration around 0.2 $\mu \text{m}$ in radius. The size distribution in winter showed a minimum content of accumulation-mode particles compared to that in the three other seasons. The seasonal variations of the accumulation-mode concentration can be interpreted in terms of seasonal changes in the relative contributions of various processes such as local generation of small particles by gas-to-particle conversion, biomass burning, and transport of anthropogenic aerosols from the Tokyo metropolitan area as well as from the East Asian continent. In the spring we also found spontaneous increases of coarse-mode particles due to long-range transported dust particles, called Kosa (yellow sand).

3) The estimated imaginary indices of refraction $m_i$ also showed a clear seasonal variation with the maximum seasonal-mean value of $m_i = 0.04$ in winter and the minimum value of $m_i = 0.02$ in summer. The period-mean $m_i$ value was 0.035. The relatively large imaginary index of refraction in winter suggested that the wintertime aerosols over the Tsukuba area might involve a significant fraction of soot particles.

4) We found close correlation, with similar seasonal variations, among the direct radiative forcing to the surface solar irradiances and the optical properties of the columnar aerosols. The results suggest that the correlated seasonal variations could result from seasonal changes in the dominant aerosol components over the Tsukuba area. To clarify this point, as a future study, simultaneous measurements of the surface solar radiation and aerosol compositions would be useful. It is also important to measure spectral AOTs in the near-infrared region. This will enable us to more accurately estimate aerosol size distributions for larger particle sizes, and to determine the imaginary index of refraction in the near-infrared region.

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