Ground-based Spectral Measurements of Solar Radiation (I)

—Extinction and Size Distribution of Aerosol Particles in the Atmosphere—

by

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Abstract

A comprehensive measurement of solar radiation at the ground level was carried out by using a spectro-pyrheliometer, spectro-pyranometer and some ordinary pyranometers. Data have been accumulated since 1967 on direct solar radiation, solar aureole intensity, global and scattered sky radiation. These data were analysed to obtain some understanding of the optical properties of aerosols as one of the components of the short wave radiation balance in the atmosphere.

The extinction coefficients measured in Tokyo show the trends of gradual decreases after 1970. The amount of decrease is larger in the shorter wavelength region than in the longer. The size distributions of aerosols determined from the spectral distributions of extinction coefficients vary from case to case. The amount of particles in smaller size ($r \leq 0.3 \mu m$) gradually decreases from year to year and that in the larger ($r \geq 0.6 \mu m$) shows a slight increase in the earlier stages of the period and a rapid decrease in the later.

1. Introduction

The studies of solar radiation are recently carried out by many investigators as one of the important components affecting the long range climatic changes. In this connection, change in aerosol particles is considered as the cause of change in the solar radiation budget in the atmosphere. Lettau (1969) estimated the effect of aerosols on the solar radiation budget in the atmosphere in urban (Kew) and desert (La Joya and O'Neill) areas. His calculation indicates that the increase of the aerosols by a factor two brings a significant effect in the patterns of the solar radiation budget for each station. This effect arises from differences in the optical properties of aerosols represented by the ratio of absorption to scattering efficiency of particles. Mitchel (1971) proposed a generalized model of the effect of aerosols on the solar radiation budget and applied it to the estimation of the change of temperature near the earth's surface due to absorption by particles.

Besides the above investigations, there are a lot of studies about the effects of aerosols on climatic changes on the basis of solving the transfer equation for the model atmospheres including the aerosol particles (see e.g., Halpern and Coulson, 1976; Liou and Sasamori, 1975; Wang and Domoto, 1974; Braslaw and Dave, 1973; Yamamoto and Tanaka, 1972). The results of these calculations show that the flux divergence in the lower atmosphere is highly
dependent on the solar zenith angle, the imaginary part of the refractive index, the height and size distribution of aerosols and the characteristics of the ground surface.

On the other hand, observational studies of the absorption of solar radiation in the atmosphere have been performed by several investigators since 1960. Robinson (1962) obtained the absorption of solar radiation by atmospheric aerosols from an analysis of the surface measurement data on solar radiation. In 1964, Robinson (1966) also made aircraft measurements of the upward and downward solar radiation over the English Channel. He showed by comparing the data with the calculation that the indirect estimation of the absorption from the surface measurements are in agreement with the direct aircraft measurements.

In recent years, the solar radiation budget in the earth-atmosphere system was directly measured in several big projects which aimed at complete radiation measurements. In the USSR, Complete Atmospheric Energetics Experiment (CAENEX) was carried out in 1972 and the results of the experiment were reported by Kondratyev et al. (1973). In the U. S. A., Barbados Oceanographic and Meteorological Experiment (BOMEX) was done in 1969 and its results were reported by Drummond (1974) and Reynolds et al. (1975). The newest project of this kind is the Global Atmospheric Aerosol and Radiation Study (GAARS) conducted by NCAR, NOAA, NASA and other cooperators. The preliminary study of GAARS was undertaken in 1972 and its results were reported by DeLuisi et al. (1976). All of the above experiments included measurements of the upward and downward solar radiation by aircraft and directly obtained the flux divergences in the atmosphere.

The inherent shortcoming of the direct estimation of the fluxes with ground and aircraft measurements is the restriction of the area and the period for obtaining statistical information on variations of aerosol effects. The indirect methods, on the other hand, are free from this defect because of the wide network of measurements and the continuous measurement data. Herman et al. (1975) tried to determine the imaginary part of the refractive index of the aerosols by comparing the measured flux at the ground level with the calculated fluxes. They extended this analysis to a practical method for obtaining the optical properties of the aerosols by means of rather simple radiation measurements.

The purpose of this paper is to analyse the measurements of the solar radiation at the surface for the past decade, for the purpose of obtaining the year to year changes of the extinction coefficients and the size distributions of the aerosols, and to study the other optical properties such as solar aureole intensity and aerosol absorption. In Part I, the instrumentation and results of the measurements of direct solar radiation are described. In Part II, the analysis of the measurements of the solar aureole and diffuse sky radiation will be described.

2. Instrumentation

Several kinds of instruments were prepared for the spectral measurement of direct solar and diffuse sky radiation at the ground level. The oldest one is the spectro-rheliometer designed to measure the spectral intensities of direct solar and aureole radiation. It is composed of a telescope, a double monochromator and an electric system. The field of view angle of the telescope is less than one degree, so that we can measure the aureole radiation coming from the sky area about one degree from the center of the sun. A photo-electric sun seeker is attached to the telescope to follow the direction of the solar beam. The double monochromator contains two quartz prisms to avoid the stray light in the monochromator. The photomultiplier is used to detect the spectral intensities of ultra violet and visible radiation. The beam collimated by the telescope is chopped by the rotating sector in the front of the entrance slit of the monochromator and 400 cps. alternative
output photocurrent is fed by the detector to the amplifier and recorded on the chart paper of the electronic recorder. Besides the telescope, an integrating sphere can be set on the monochromator to measure the spectral intensity of global and diffuse sky radiation. The quantity of light incident on the entrance slit is controlled by the diaphragm and the neutral density filters which are attached to the top of the telescope and the front of the entrance slit, respectively. The instrument is shown in Photo. 1.

A newly designed instrument is the spectro-pyrheliometer without the collimating telescope. It is mounted on the equatorial mounting so that the entrance slit is directly exposed to the solar beam. The primary spectrum is obtained by a diffraction grating and the secondary by a quartz prism. The wavelength range we can measure is between 0.30 and 2.0 μm, and the sampling interval of wavelength is variable from 5 to 500 Å. The time needed for a scanning of the whole range is about 10 min. in the case of sampling interval of 100 Å in UV region, 250 Å in visible and 500 Å in the near infrared region, respectively. In order to see the structure of the water vapor absorption in the near infrared region, the sampling interval less than 50 Å is necessary and the time needed for the scanning of the whole range exceeds about 30 min. Although this scanning duration seems a bit too long for accurate measurement, we can get stable measurement around the time of solar culmination in a clear day.

The change of the sensitivity of the instrument is examined by the reference lamp to which 12 or 24 stabilized d.c. voltages is supplied. The output photocurrent corresponding to the solar radiation intensity is corrected by the variation of the output current for the reference lamp. In order to get the absolute values of the measured intensities, we prepared a standard lamp whose spectral values of emitted flux are calibrated by comparison with the international standard lamp. Data arrangement is performed by using the measured photocurrent relative to the extraterrestrial values which are extrapolated through the so-called long method. The extraterrestrial values thus obtained are checked by the standard lamp described above. Views of the instrument and the optical system for the measurement of the standard lamp are shown in Photos. 2 and 3.

The third equipment is a spectro-pyranometer for the measurement of global and diffuse sky radiation. The monochromator used is the same type as the former. An
integrating sphere of 15 cm. dia. is attached to the entrance slit of the monochromator to provide the receiving surface of the flux. The entrance slit faces the small area of the integrating sphere, and the inside of the sphere is coated with white paint mainly composed of barium sulfate to ensure sufficient diffuse reflection. In order to separate the global radiation into the direct and the diffuse components, a shade disk is installed. The disk is driven by a motor so as to be in the solar beam impinging on the sphere and it is easily deviated about 10 degrees from the beam for the instances needed. Photo. 4 shows the view of the spectro-pyranometer.

Besides the instruments described above, a pyrheliometer which employing interference filters, one using glass filters, and the ordinary pyranometers are prepared for supplementary spectral measurement. In cloudy cases, the amount of cloud was measured by a camera equipped with fish-eye lens.

3. Measurements and data

Spectral measurements of solar radiation have been carried out in the case of clear sky since 1967. Most of the measurements were performed on the roof of the Meteorological Research Institute in Tokyo, the rest at the following places, which are far from the contaminated area (Tokyo): Karuizawa about 200 km NW, Yagisawa about 150 km N., Hachijo Is. about 300 km S., and Tateno about 50 km NE. from Tokyo. Almost all of the data were obtained in autumn and winter, since the measurements had to be carried out under clear skies, which are most frequent in these seasons in the areas concerned.

In the early stage, from 1967 to 1970, the measurements were limited to the spectroscopy of the aureole radiation along with the direct solar radiation. The scattered radiation coming from the small area of the sky 1 to 15 deg. from the center of the sun was measured in the vertical plane containing the sun and the zenith. The instrument used was the oldest one, and 14 wavelengths were selected for the measurements in the range from 0.35 to 1.6 μm. Since 1974 we started to measure the global and the diffuse sky radiation by using the ordinary pyranometer with the thermopile. At the same time, the spectral measurements of the global and the diffuse sky radiation with the integrating sphere were commenced. The spectral region measured, however, was limited to the UV and visible region, because of the insufficient sensitivity of the instrument in the near infrared region. Corresponding to these measurements, the cloud amount and type were recorded on film by means of a camera with a fish-eye lens.

Since precise measurements by the spectrometer are rather complicated and time consuming, a simple method of mea-
Measurement with several glass filters is desirable in network observations of solar radiation. We prepared a pyrheliometer with the thermopile as detector and three glass filters and ten interference filters. Measurements of direct solar radiation by the filter method were carried out at the same time with spectral measurements in the last three years, and the data obtained were compared with each other.

The new type of instrument for spectral measurements of direct solar radiation started its operational work in 1974, and was operated in parallel with the older instrument for getting reliable data in the beginning of the operations. The sampling interval of wavelength is much smaller than for the older one, so we can detect a more detailed molecular absorption, especially the absorption by water vapour. In order to get the spectral absorption of the atmosphere, more than 30 min. is necessary for a scanning of the whole wavelength region. Consequently such a measurement is performed only around the solar culmination during which the variation of the solar height is small.

4. Optical extinction and size distribution of aerosol particles

The monochromatic intensity of direct solar radiation reaching the ground, \( I(\lambda) \), is represented by

\[
I(\lambda) = I_0(\lambda) \exp \left[ -\tau(\lambda)m \right]
\]

where \( I_0(\lambda) \) is the intensity at the top of the atmosphere, \( m \) the optical air mass \((=\sec z, z: \text{ solar zenith angle})\). \( \tau(\lambda) \) is the atmospheric extinction coefficient and is represented by the sum of three components: the molecular scattering \( \tau_R(\lambda) \), the molecular absorption \( \tau_w(\lambda) \) and the scattering and absorption by aerosols \( \tau_M(\lambda) \). That is,

\[
\tau(\lambda) = \tau_R(\lambda) + \tau_w(\lambda) + \tau_M(\lambda)
\]

\( \tau_R(\lambda) \) is calculated from the Rayleigh scattering cross section and the molecular density of the atmosphere. Among the numerical values calculated by several authors, we took the values published by Elterman (1968). \( \tau_w(\lambda) \) contains the absorption by water vapor and ozone and other minor absorption by atmospheric gases. In order to get the values of \( \tau_w(\lambda) \), we used the table published by Wyatt et al. (1964) for the absorption coefficient of water vapour and the values obtained by Inm and Tanaka (1953) for ozone. The values of the water vapour content at the time of the radiation measurement were determined from radiosonde data at Tateno, about 50 km NE from Tokyo. The values of the total ozone were determined from the data of the measurement with the Dobson spectrometer at Tateno. Although \( \tau_M(\lambda) \) should be separated into the components of the absorption and the scattering to investigate the physical and optical characteristics of the aerosols, we have no clue to detect these components individually in the data afforded by our measurements.

The spectral intensities of the direct solar radiation, \( I(\lambda) \), were measured at several values of optical air mass between 1.0 and 4.0 on each day. Although the instrumental factor converting an output photocurrent to an absolute value of intensity is not established, the output current corresponding to the intensity at the top of the atmosphere can be determined by the extrapolation of the output current measured by means of Bouguer-Langley method, on the occasion of a clear sky. According to the value thus obtained, the spectral extinction coefficient at any time is obtained from the measurement of direct solar radiation. And the extinction coefficient due to aerosols is calculated by,

\[
\tau_M(\lambda) = \tau(\lambda) - \tau_R(\lambda) - \tau_w(\lambda)
\]

The spectral distributions of the extinction coefficient of aerosols thus obtained are shown in Figs. 1 through 4. Selected curves are chronologically described in the figures. We can see from the figures that there are few curves represented by the power function of the wavelength and that deviation from the power function is rather complex.

The spectral distribution of the aerosol
Fig. 1. Spectral distributions of the extinction coefficient of aerosols measured at several places in 1967.

Fig. 2. Same as Fig. 1 but in 1970 and 1971.

Fig. 3. Same as Fig. 1 but in 1972.

Fig. 4. Same as Fig. 1 but in 1973, 1974 and 1975.
Fig. 5. Size distributions of aerosols corresponding to the measured extinction coefficients represented in Fig. 1.

Fig. 6. Same as Fig. 5 but corresponding to Fig. 2.

Fig. 7. Same as Fig. 5 but corresponding to Fig. 3.

Fig. 8. Same as Fig. 5 but corresponding to Fig. 4.
The extinction coefficient is related to the size distribution of aerosol particles as follows:

$$\tau_M(\lambda) = \pi \int_r r^2 K \left( \frac{2\pi r}{\lambda} \right) n(r) dr$$

where \( r \) is particle radius, \( n \) refractive index of a particle, \( n(r) \) the number of particles per cubic centimeter per unit radius interval at radius \( r \), and \( K \) the ratio of the scattering cross section to the geometrical cross section of a particle. From the measured values of \( \tau_M(\lambda) \), we can infer the size distribution of the particles by means of the inversion technique. In practice, we used the technique developed by Yamamoto and Tanaka (1969), which is based on the method of Phillips (1962) and Twomey (1965). The size distributions obtained by this technique from the measured extinction coefficients are represented in Figs. 5 to 8. In this calculation, \( \bar{n} \) is assumed to be 1.50.

The curves represented in the figures correspond to the curves of \( \tau_M(\lambda) \) described in Figs. 1 to 4. The curves of size distributions show slight deviations from the power law distribution curve reflecting the characteristics of the spectral distributions of \( \tau_M(\lambda) \). The general features of the size distribution represented in the figures are (1) that the variation of the particle amount is larger in the smaller size region than in the larger, (2) that meandering of the curve frequently appears between 0.1 and 1.0 \( \mu \)m region.

5. Year to year variations of the extinction coefficient and size distribution

The extinction curves selected from the accumulated data are shown in the figures of the previous section as the typical representation of the year and the places. All of \( \tau_M(\lambda) \) measured in Tokyo since 1967 are plotted for five wavelengths in Figs. 9 through 13 in order of the date of measurement to see the year to year variation of the extinction. Unfortunately in 1968 and 1969 there are no data because of the malfunction of the instrument, and the data plotted in the figures are limited to those obtained in autumn and winter but we can see the variation with the years throughout the

![Fig. 9. Year to year variation of aerosol extinctions in Tokyo, for wavelength 0.35 \( \mu \)m.](image-url)
Fig. 10. Same as Fig. 9 but for 0.45 $\mu$m.

Fig. 11. Same as Fig. 9 but for 0.60 $\mu$m.
period considered.

Fig. 9 represents the variation of UV extinction coefficient measured in Tokyo. We can see from the figure that the aerosol extinction plotted is widely scattered in the range between 2.25 and 0.54 in 1967, and there are a rather large number of points whose values are larger than 1.0. On the other hand, since 1972 almost all points are plotted in the domain smaller than 1.0 and both the maximum and minimum values are smaller than those in the period from 1967 to 1971. In the visible region the features of the variation are similar to those of the UV region as seen in Figs. 10 to 12. In Fig. 13 is shown the variation of the extinction in the near infrared region with the years; the features of variation are rather different from the preceding. We can not detect the distinct variation of the extinction with the years in this wavelength region. The annual mean values of extinction co-
efficient for each wavelength are shown in Fig. 14. The figure shows that the annual mean values for 0.35 and 0.45 μm decrease rather rapidly with the years, that those for 0.60 and 0.70 μm, showing no remarkable changes in the early stage, decrease in the later, and that the values for 1.00 μm slightly increase until 1972 and diminish to very low values after 1973.

The size distributions determined from the measured extinction coefficients show wide variations in their shapes, corresponding to the variations of the spectral distributions of the extinction coefficients. The amount of particles for various radii is plotted against the year of measurement. The ordinate in Fig. 15 describes the amount of particles contained in a vertical column from the surface to the top of the atmo-

![Fig. 14. Variation of the annual mean values of aerosol extinction in Tokyo. Open circles indicate the mean values of aerosol extinction measured in non-urban areas. T: Tateno, K: Karuizawa, and H: Hachijo Is.](image)

![Fig. 15. Amount of aerosol particles in a vertical column of atmosphere for various radii. Open circles represent the annual mean values of the amount of particles.](image)
sphere. The figure shows (1) that the amount of particles in the smaller size region, 0.06, 0.13 and 0.3 \( \mu m \) shown in the figure, decreases with the years and (2) that the amount in the larger does not show monotonous variation as does that in the smaller size region. The annual mean values of the amount of particles for each particle size are shown in Fig. 15 with open circles. The annual mean values represent a rather distinct decrease with the years in the size region smaller than 0.3 \( \mu m \) and those in the region larger than 0.64 \( \mu m \) show the maximum in 1971 or 1972. Thus the year to year variations of the aerosol amount are remarkable in the smaller size region, corresponding to the variations of the extinction in the shorter wavelength region, while the variations are indistinct in the larger size region.

6. The extinction coefficients measured in some non-urban areas

For a comparison of atmospheric tur-

Fig. 16 a. Aerosol extinction coefficients for various wavelengths obtained in non-urban areas.
bidity between the urban and the non-urban area, the spectral distributions of the extinction coefficients were measured at several places distant from Tokyo. These places are Tateno 50 km NE from Tokyo, Karuizawa 150 km NE, Yagisawa 150 km N, and Hachijo Is. 300 km S. Although the serial data of $\tau(\lambda)$ for each place are not available, it is possible to compare them with each other. Some examples of the spectral distribution of $\tau(\lambda)$ for the above places are presented in Figs. 1 to 4, and the size distributions of particles are shown in Figs. 5 to 8. The measured values of $\tau(\lambda)$ for each place are plotted in Fig. 16a and b. From these figures, we see that the aerosol extinctions, in general, are smaller in the four distant places indicated above than in Tokyo, and that the differences of $\tau(\lambda)$ between Tokyo and the other places are pronounced in the shorter wavelength region than in the longer. Since we have no successive measurements available in the four places, we cannot find any trend in the change of $\tau(\lambda)$ at any of them. By taking a glance at Fig. 14, however, the annual mean values of the extinction coefficient of Tokyo approach those of the other places, which are shown in the figure by open circles, in the later years of the period. That is, the difference of the atmospheric turbidity between Tokyo and the non-urban areas is diminishing with the years in the period.

7. Year to year variation of visibility

As one of the meteorological parameters affected by the optical property of the atmosphere, we picked up horizontal visibility. The frequency of appearance of visibilities classified by range was studied by Nomoto (1976) for the following three cases: visibility in Tokyo at noon is (1) less than 3 km, (2) less than 5 km and (3) more than 10
km. In Fig. 17, the frequency of appearance only in autumn and winter is shown, since the extinction coefficients were principally measured in these seasons. From the figures we see that the frequency of appearance of the visibility less than 5 and 3 km diminishes with the years after 1969, while that of visibilities more than 10 km increase after the same year.

As an example of the non-urban area, the same kinds of curves for Maebashi (about 100 km NW from Tokyo) are described in Fig. 18. We can not detect any remarkable trend in the change of visibility for the curves in the figure. A comparison of Fig. 17 with Fig. 18 reveals that the extremely hazy atmosphere, i.e. with a visibility less than 3 km much more frequently appears in Tokyo than in Maebashi before 1971, its frequency becoming nearly the same after that year.

It is interesting to compare the trends in the change of visibility with those of the extinction coefficient and of the amount of the aerosols described in the previous section. In general, the decreasing trend of the annual mean values of the extinction coefficient with the years in Tokyo corresponds to the similar one of the frequency of appearance of low visibilities. The frequency of extremely high values of extinction gradually diminishes with the years, and the plotted points of \( \tau_m(\lambda) \) in Figs. 9 to 13 are rather concentrated in the domain of smaller values of extinction in the later stage of the period. On the other hand, the variations of the amount of aerosol particles for various sizes show trends which are different from each other according to the particle radii.

In Fig. 17, the frequency of appearance of visibilities less than 5 km maintains rather high values in the period from 1967 to 1972, with the maximum value in 1969, and after 1972 it goes down to rather low values. Visibilities less than 3 km show nearly the same trend as the former. Visibilities more than 10 km show a trend opposite to the previous two cases. On the other hand, the amount of particles smaller than 0.13 \( \mu m \) gradually decreases throughout the period as shown in Fig. 15. The amount of particles larger than 0.30\( \mu m \) increases in the first stage of the period and then gradually decreases. The maximum values for each size appear.
in 1971, except for the size 6.3 \mu m, whose maximum value appears in 1972. Comparing the features of visibility with those of particle amount, we can say that the small values of the frequency of visibilities less than 3 and 5 km in the last stage of the period correspond to the small values of the amount of particles in the whole range of particle size. On the contrary, the variation of visibility in the period from 1967 to 1972 reflects the composite effects of the decrease of the amount of particles in the smaller size region and the increase of that in the larger one. Considering the size distributions of particles described in the previous section, quantity of decrease in the smaller size region is larger than that of increase in the larger one.

According to the features described above, we can say that the size distribution of the aerosol particles plays an important role in the determination of visibility. To make clear the relations between visibility and the size distribution of aerosols, we have to make precise analyses of the individual measurements of extinction and visibility.

8. Concluding remarks

The spectral extinction coefficients of aerosols were measured by the spectropyrheliometer in Tokyo and some non-urban areas in and after 1967. The extinction coefficients measured in Tokyo show a gradual decrease with the years in the period from 1967 to 1975, in the shorter wavelength region. Those in the longer wavelength region show a slight increase in the early stage and a decrease in the later stage of the period.

Comparisons of the aerosol extinctions measured in Tokyo with those in non-urban areas show that the aerosol extinctions in Tokyo are larger than in non-urban areas, especially in the early stage of the period. In the last few years of the period, the differences diminish.

The size distributions of aerosols inferred from the measured extinction coefficients by the inversion technique also change from year to year. The amount of particles in the smaller size region diminishes rather rapidly with the years, and that in the larger size region begins to decrease after 1972.

Variations in aerosol extinction and the size distribution of aerosols were compared with those of horizontal visibility in Tokyo. In general, visibility becomes better with the years, corresponding to the decrease of aerosol extinction and the particle amount.

The above comparisons show that the variation of visibility in the period from 1967 to 1972 corresponds to the composite effect of the decrease of the smaller particles and the increase of the larger particles. In the later stage of the period, the frequency of appearance of visibilities less than 3 and 5 km diminishes according to the decrease of the particles in whole range of the particle size.

In our analyses of the data, the correlation between aerosol characteristics and visibility is significant as described above, but we have to examine the data for every individual case of measurement to make clear the contributions of aerosols to visibility.

References


