Refractive Index and Size Distribution of Aerosols as Estimated from Light Scattering Measurements

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ABSTRACT

Angular distributions of the intensity of light scattered by airborne particles were measured for both parallel and perpendicularly polarized components. A precision polar nephelometer was constructed for use of this study. The data were analyzed using a newly developed inversion library method to give a simultaneous estimate of the complex index of refraction $m = m_r - m_i i$ of aerosols and their size distribution, where m_r is the real part and m_i the imaginary part of the refractive index. Results of 302 measurements show that the monthly mean values of m_r and m_i vary within the range 1.47–1.57 and 0.009–0.037, respectively, depending upon the relative humidity.

The size distribution of aerosols can be approximated by the log-normal distribution function; the mean radius and the standard deviation are found to be $r_g = 0.138$ μm and $\sigma_g = 2.56$, respectively, as geometrical mean values for the whole data.

1. Introduction

Atmospheric aerosols play an important role in the heat balance of the earth-atmosphere system. Increases in aerosol loading of the atmosphere can lead either to an increase or a decrease in the mean global temperature of the earth. The sign of their effect depends on the values of their optical constant (i.e., complex index of refraction) in the visible and infrared portions of the spectrum as well as on the size distribution of aerosols and the albedo of the ground surface (Yamamoto and Tanaka, 1972: SMIC, 1971). In this connection, general attention has been attracted to the estimation of optical characteristics of aerosols and their influence on climate.

An important achievement of a number of experiments carried out in recent years is the complex character of aerosols; they have revealed the strong variability of aerosol concentration field in the free atmosphere, as well as large changes in the aerosol optical characteristics. According to these studies, the real part m_r of the index of refraction of aerosols falls in a rather narrow range of 1.45–1.55, while the imaginary part m_i varies greatly in the range 0–0.1. It has not yet been concluded that aerosols falling in this range of absorption index do so as a result of their own properties or an inadequacy in measurement techniques. The uncertainty in the value of m_i is one of the most important obstacles to our understanding of the effect of aerosols on the heat balance of the earth. Of primary importance, therefore, are investigations of their absorption index.

The methods of determining the value of the imaginary part of the index of refraction of aerosols are divided into three classes; transmittance measurements, diffuse reflection measurements and differential scattering cross section measurements. Voltz (1972a, b) and Fischer (1970) have measured the values of m_i for precipitation residues and filtered aerosol samples, respectively, by means of the transmittance method. The diffuse reflectance method based on Kubelka-Munk theory (Kortüm, 1969; Wendlandt and Hecht 1966) have been employed by Lindberg and Laude (1974) and Patterson *et al.* (1977) to determine the value of m_i. A characteristic of these two methods is that the value of m_i can be obtained as a function of wavelength without knowing the size distribution of the aerosols. The reliability of the results, however, depends on a number of assumptions and, especially, on the change in optical constants due to collection of aerosol samples. The method based on light scattering measurements has also been employed by several authors. Eiden (1966) has attempted to estimate the values of optical constants of aerosols from measurements of the ellipticity of scattered light. The same attempt has been made by Grams *et al.* (1974) but from measurements of the intensity of the perpendicularly polarized component of scattered light. In this method, it is a great advantage that we do not need to take into consideration the change in optical constants of aerosols due to sample collections. On the other hand, accurate information of particle sizes is indispensable for this

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of every day in the period 14 March–7 May 1976. The climatic conditions in this period changed pe-

method, since the scattered light strongly depends on the size distribution of aerosols as well as on their optical constants. In this connection, Grams *et al.* have measured the size distribution of aerosols simultaneously with scattering measurements, but Eiden has assumed Junge's distribution in his analysis.

In this study, we also attempt to estimate the values of the optical constants of aerosols from measurements of the scattered light. Angular distributions of the intensity of light scattered by aerosols were measured for both parallel and perpendicularly polarized components within the range of scattering angles from 7° to 170° by use of a precision polar nephelometer. These data were analyzed by an inversion library method developed for this study, and the maximum likelihood values of the optical constants of aerosols were estimated simultaneously with their size distribution.

2. Instrument, calibration and method of analysis

The methods of observation and analysis are briefly summarized in this section, as they have been already described elsewhere (Takamura and Tanaka, 1978; Tanaka *et al.*, 1982). A precision polar nephelometer used in this work has been equipped with an argon-ion laser (λ = 514.5 nm, light output 750 mW) as a light source. The polarized plane of the source beam is set at a tilt angle of 45° from the scattering plane to measure both perpendicular and parallel components of the scattered intensity. Two head-on photomultiplier tubes are adopted as detectors: one is a scanning detector and the other a monitoring detector fixed at a scattering angle of ~30°. The scanning detector, which consists of a telescope, an analyzer and a photomultiplier tube (HTV. R374), has a scattering volume of about 1.1 cm^3 at the scattering angle of 90° and covers the range of scattering angles from 7° to 170°. The intensities of scattered light polarized perpendicularly and parallel to the scattering plane have been measured at the scattering angles Θ = 7(1)10(2.5)25(5)170° for about half an hour. The total (i.e., unpolarized) intensity of the scattered light received by the monitoring detector was measured simultaneously with each angular datum. A photon counting technique was used in processing photoelectron pulses from the detectors, so as to detect very weak signals such as found in a dust-free atmosphere with sufficiently large S/N ratio.

Fluctuations of sample concentration and intensity of the light source are unavoidable during the angular scan. Most of the data were corrected by use of the monitoring signal. Since large fluctuations of the monitored intensity may be caused by fluctuations not only of aerosol concentration but also of aerosol characteristics, i.e., the complex index of refraction and the size distribution, it is not always expected that the influence of sample fluctuations can be reduced correctly by such a procedure. In fact, several examples showed such serious fluctuations that any reasonable corrections could not be made. However, most of the observed angular distributions of scattered light can be reconstructed from inferred values of the complex index of refraction and the size distribution, with root mean square errors not exceeding a few percent, assuming that the corrected data were no more influenced by sample fluctuations than they would be under usual weather conditions.

The instrument was absolutely calibrated from measurements of the Rayleigh scattering by N_2 gas, whose angular distribution law has been firmly established (e.g., Goody, 1964). The calibration constants (or the conversion factor) was estimated for each measurement by comparing respective polarizations of observed Rayleigh scattering with their known values by the least-square method. Precise positions of the polarization axis of the analyzer and the polarized plane of the incident laser beam as measured from the scattering plane were also examined simultaneously. Such careful measurements of the Rayleigh scattering by N_2 contributed greatly to substantiate the reliability of the data of Mie scattering by aerosols. Neither systematic nor random errors were found to seriously affect the final results.

In the analysis, we assumed that aerosols consist of homogeneous spherical particles with the complex index of refraction m ($=m_r - m_i i$) and the size distribution function $n(r)$, where $n(r)dr$ is the number density of aerosols of radii between r and $r + dr$. The differential scattering cross sections of aerosols (or phase functions) can be expressed (van de Hulst, 1957) as

$$\beta_j(\Theta_i) = \int_{r_{\min}}^{r_{\max}} i_j(\Theta_i, r, m)n(r)dr, \qquad (1)$$

where $i_j(\Theta_i, r, m)$ is the differential scattering cross sections of a single particle with radius r and the complex refractive index m for light polarized either perpendicularly (j = 1) or parallel (j = 2) to the scattering plane, and $r_{\min}$ and $r_{\max}$ are radii of the smallest and the largest particles, respectively. Our problem is how to determine the most plausible values of the complex index of refraction m and the size distribution $n(r)$ from measurements of $\beta_j(\Theta_i)$ (j = 1, 2; i = 1, . . . ; N = 39).

The methodology we adopted is outlined as follows:

1) A set of discrete values of m (i.e., discrete values of m_r and m_i) is chosen so as to cover an expected range of the complex refractive index of aerosols with proper resolution.

2) The size distribution function $n(r)$ is determined for a certain value of m among the above set by inverting the measured differential scattering cross sections $\beta_j(\Theta_i)$, i.e., the above integral equation.

3) The differential scattering cross sections are reconstructed for the above pair of m and $n(r)$, and the root-mean-square deviation between reconstructed and measured differential scattering cross sections σ is estimated.

4) After repeating 2) and 3) for all values of m given in 1) a pair of m and $n(r)$ which minimizes the σ-values is adopted as the most plausible one, i.e., the solution of our problem.

Clearly, one of the most important and difficult points of our methodology is in step 2), i.e., determination of the size distribution of aerosols from differential scattering cross section measurements. Details of the method have been described elsewhere (Tanaka *et al.*, 1982).

3. Results and Discussion

a. Measurements

Extensive measurements of the differential scattering cross section of aerosols have been made for visible light with a wavelength of 514.5 nm on the roof of a university building in the suburbs of Sendai, Japan. A total of 52 and 250 samples were collected for the periods 11 January–28 April 1977 and 7 February–7 November 1978, respectively. In climatic conditions, a dry and cool continental air mass covers the area including the observation site in winter, while a wet and hot oceanic air mass prevails in summer.

Meteorological variables such as air temperature and relative humidity have also been measured at the observation site simultaneously with each scattering measurement, and tabulated for respective periods in Takamura and Tanaka (1978) and Takamura (1979). Monthly mean values of air temperature and relative humidity are shown in Fig. 1, comparing with climatological values for the period 1941–70 at Sendai Meteorological Observatory.

The monthly mean values of the normalized phase function $P_{ave}(\Theta)$ and degree of polarization $p(\Theta)$ are shown respectively in Figs. 2a and 2b for the period from February to November, 1978. In Fig. 2a, the horizontal lines near the scattering angle of 60° show the levels where $P_{ave}(\Theta)$ takes the value of unity for corresponding months, and the vertical segments at the scattering angle $\Theta = 120°$ or $\Theta = 125°$ indicate the standard deviations from the monthly mean at designated scattering angles. Similarly, the vertical segments in Fig. 2b indicate the standard deviation of the degree of polarization at respective designated angles. The standard deviation of data presented without error bars is, in general, much smaller than that designated. From the figure, we can see that the monthly mean of $P_{ave}(\Theta)$ varies logarithmically at all observation angles within the limits of 4–10%. While month-to-month variation of the $p(\Theta)$, as well as the day-to-day variation, is considerably larger than that

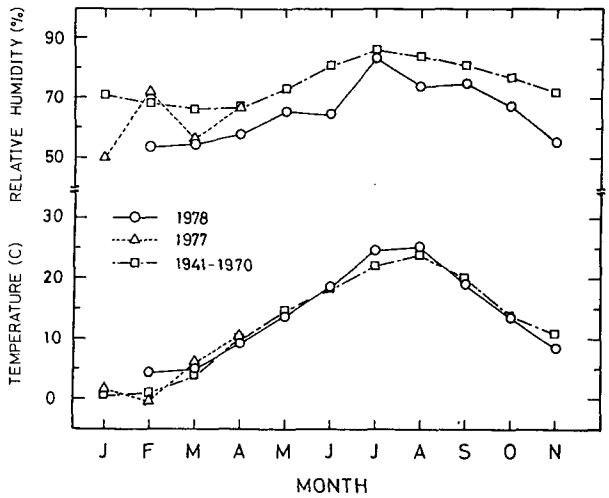

FIG. 1. Seasonal variation of the monthly mean values of air temperature (lower) and relative humidity (upper).

in $P_{ave}(\Theta)$, amounting to 15–30% in relative values and 0.05–0.15 in absolute values. It is expected from such comparisons that more information on the optical characteristics of aerosols is involved in the degree of polarization than the single phase function. Already, we have shown that polarization measurements are essential for reliable estimation of the index of refraction and the size distribution (Tanaka *et al.*, 1982).

b. Results of analysis

1) COMPLEX INDEX OF REFRACTION

The above 302 samples have been analyzed by our inversion library method (Tanaka *et al.*, 1982). The optimum value of the complex index of refraction is selected from a set of discrete values covering an expected range of the index so as to minimize rms deviation of reconstructed phase functions from measured ones. In this sense, the inferred value should be regarded as an optically equivalent index of refraction. If there is no tabulated value of the index of refraction just coincident with the true value, a mesh-point closest to the true value is simply selected as the optimum value. Inadequacy in selection of the complex index of refraction is more or less compensated by a simultaneously inferred size distribution but the extent of compensation is usually restricted within narrow limits, which makes possible a reasonable determination of the complex index of refraction. The discrete values of m_r and m_i thus obtained are averaged arithmetically and geometrically, respectively, in order to know their monthly mean values.

Seasonal variations of the monthly mean values of m_r (below) and m_i (above) are shown in Fig. 3. In the

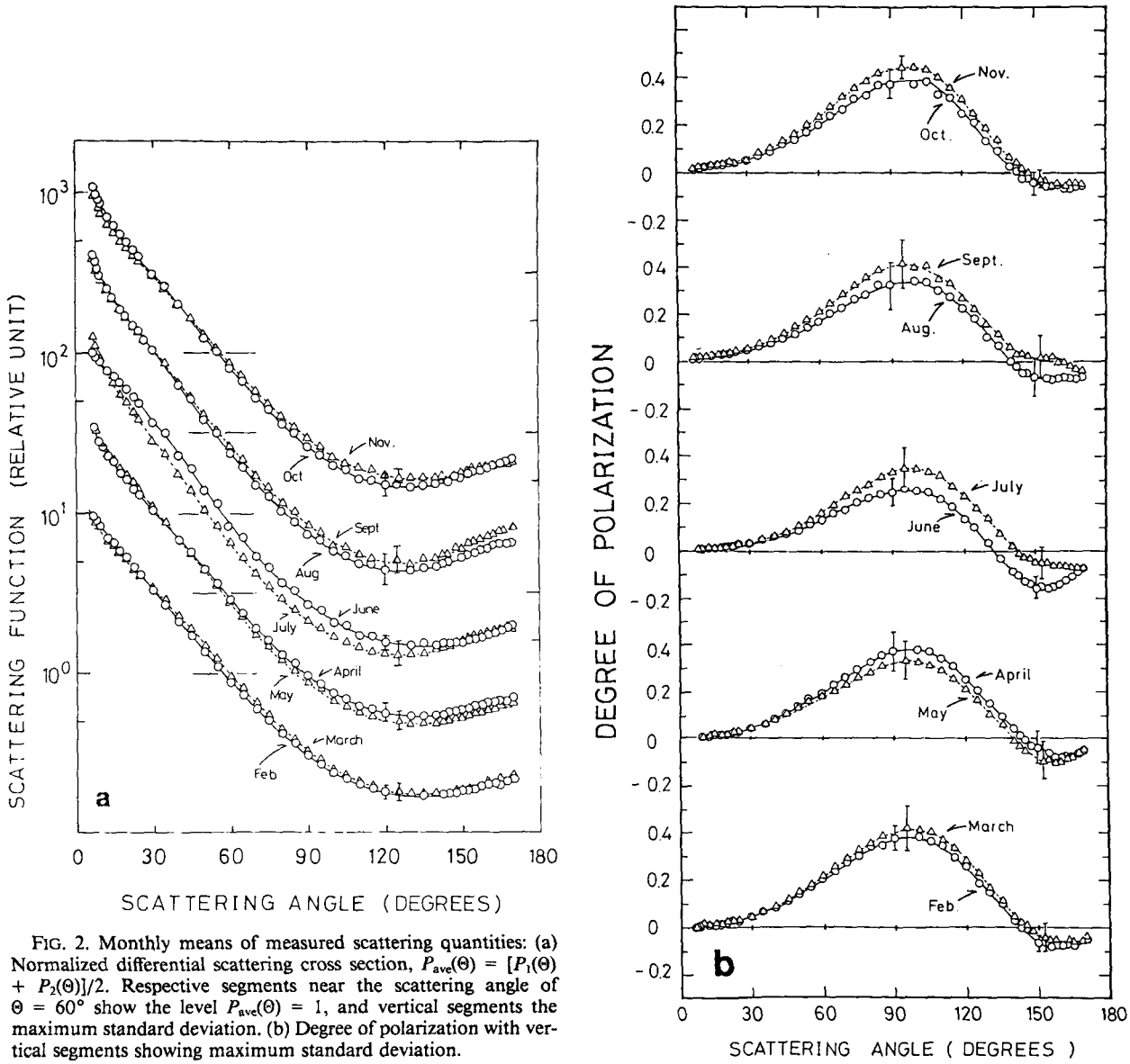

FIG. 2. Monthly means of measured scattering quantities: (a) Normalized differential scattering cross section, $P_{ave}(\Theta) = [P_1(\Theta) + P_2(\Theta)]/2$. Respective segments near the scattering angle of $\Theta = 60°$ show the level $P_{ave}(\Theta) = 1$, and vertical segments the maximum standard deviation. (b) Degree of polarization with vertical segments showing maximum standard deviation.

figure, solid and dashed lines indicate the results in 1978 and 1977, respectively. The abscissa is the month from January to November and ordinates for m_r and m_i are indicated in the linear and logarithmic scales, respectively. We can see that the values of m_r and m_i vary in the ranges from 1.47 to 1.57 and 0.009 to 0.037, respectively. It is noticeable that the values of both quantities are smaller in summer than in spring and fall. Differences in atmospheric conditions and the origin of air masses seem to be responsible for this seasonal variation. In this respect, it is noted that the monthly mean values of the complex index of refraction are highly correlated with corresponding values of the relative humidity shown in Fig. 1. Suposing that aerosol particles are composed of water soluble materials, their average optical characteristics are expected to depend upon the thermodynamic equilibrium between the particle surface and the ambient moist air, and are changeable with atmospheric moisture, as pointed out by Hänel (1968, 1976). The relation between the optical characteristics and the relative humidity will be discussed in detail in the succeeding paper (Takamura *et al.*, 1983).

On the diurnal variation of the aerosol characteristics, Whitby *et al.* (1972) have found in the Los Angeles region that the volume concentration of aerosols in the size range 0.1–1 μm in diameter increased noticeably during the daytime without changing either the shape of the size distribution or the mode diameter, synchronizing with the solar radiation. The

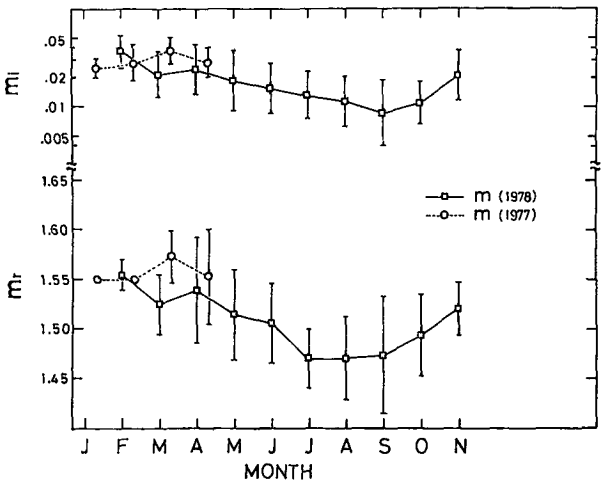

FIG. 3. Seasonal variation of the monthly mean values of the real and imaginary parts of the complex index of refraction, m_r and m_i. The imaginary part m_i is shown in the upper part of the figure in logarithmic scale, and the real part m_r is shown in the lower part in linear scale.

change in concentration in this size range is likely due to condensation, as pointed out by Husar *et al.* (1972). Moreover, the urban atmosphere would be affected by man's activities such as exhaust fumes from automobiles, combustion products, industrial waste and so on. To examine these effects, nephelometer observations were made at midnight and noon of every day in the period 14 March–7 May 1978. The climatic conditions in this period changed periodically by alternation of migratory highs and lows. The sources of primary particles were therefore expected to change considerably with time, but such effects were supposed to be smoothed out by averaging over the whole period independently of either daytime or nighttime. Contrary to this expectation, some systematic discrepancies were found between the optical characteristics of aerosols at midnight and noon. The values of the complex index of refraction m, the total scattering cross section C_{sca} and the albedo for single scattering ω_0 averaged over the whole period are shown in Table 1. The values of both real and imaginary parts of the index of refraction decrease in the nighttime corresponding to an increase of the relative humidity, while the value of C_{sca} shows a tendency to increase in the daytime. Corresponding volume spectra are shown together with C_{max} values in Fig. 4. Since the particle growth due to condensation of water vapor results in a decrease of the refractive index and an increase of the total scattering cross section, the above result is not consistent with the behavior of hygroscopic particles. The influences of man's activities, coagulation of photochemical reaction products, and the stability of atmospheric stratification, all dependent on the time of day, are expected to be responsible for the above result. Discussions on such details will also be given in the succeeding paper (Takamura *et al.*, 1983).

TABLE 1. Relative humidity and optical characteristics of aerosols at noon and midnight, averaged over the period of 14 March–7 May 1978.

	Noon	Midnight
RH (%)	48.7	61.4
m	$1.544-0.026i$	$1.527-0.019i$
C_{sca} (cm^{-1})	8.7×10^{-7}	7.7×10^{-7}
ω_0	0.77	0.77
cases	48	49

We assumed that aerosols consist of homogeneous dielectric spheres with the same refractive index $m = m_r - m_i i$. Validity of this assumption has become one of the most interesting problems in atmospheric optics in the last decade (Powell *et al.*, 1967; Holland and Gagne, 1970; Chylek *et al.*, 1977; Heintzenberg, 1977; Heintzenberg and Welch, 1982; and other). Although the general characteristics of the difference between the scattering pattern of randomly oriented nonspherical particles and that of spheres are still open to discussion, it may be that the backward portion of the phase function of nonspherical particles depends less on the scattering angle than it does for spheres. Then, as pointed out by Grams *et al.* (1974), the value of the imaginary part of the refractive index inferred from observation of the phase function must be regarded as the upper limit of the true value when

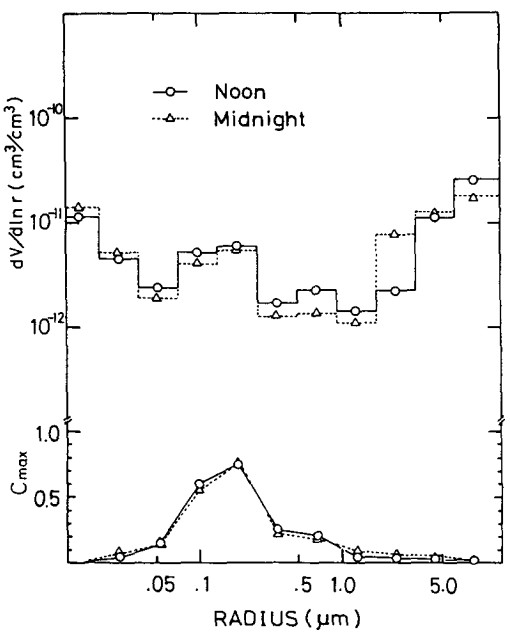

FIG. 4. Aerosol volume spectra at noon (solid line) and midnight (dotted line) averaged over the period from 14 March to 7 May 1978. Corresponding values of C_{max} are shown in the lower part of the figure.

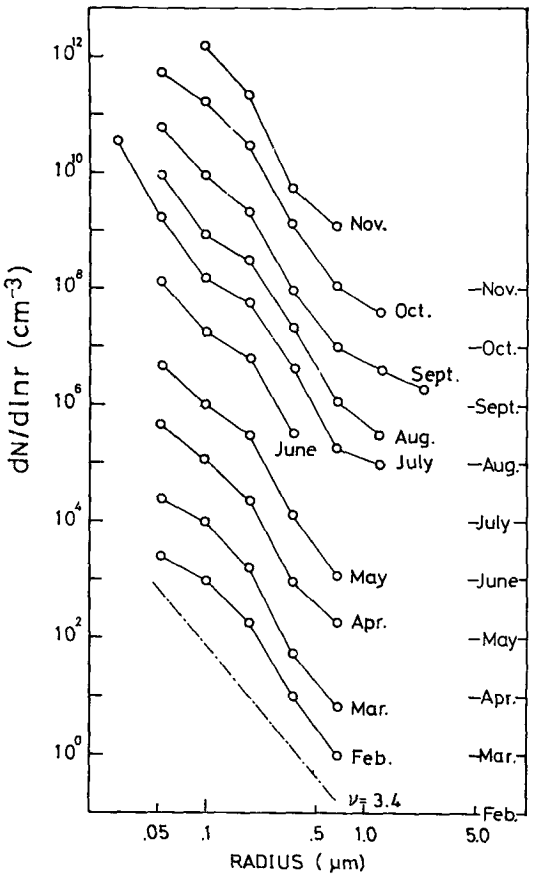

FIG. 5. Monthly means of aerosol number spectra in the effective size range for the period from February to November in 1978. Horizontal lines on the right side indicate the level $dN/d\ \ln r = 10^{-1}$ particles cm^{-3} for respective months.

the size distribution is known. In this context, Pinnick *et al.* (1976) tried to fit scattering patterns of nonspherical particles to those of spherical particles by introducing a fictitious absorption corresponding to the imaginary part of the refractive index of 0.02–0.12. We also encountered rare cases suggesting a similar effect, when yellow dust appeared. For such cases, inferred values of both real and imaginary parts of the refractive index became unrealistically large around the values of 1.70 and 0.08, respectively. Data in such rare cases were omitted from our analysis.

2) SIZE DISTRIBUTION OF AEROSOLS

The reliability of the size distribution of aerosols inferred simultaneously with the complex index of refraction can be examined from the magnitudes of the contributions of particles with sizes corresponding to observed values of $\beta_j(\Theta_i)$. The reliability of inverted size distributions is restricted within the size range where the maximum contribution of a spectral portion to any portions of the observed differential scattering cross sections C_{max} exceeds the limit of 10%, i.e., $C_{max} > 0.1$ (Tanaka *et al.*, 1982). The monthly mean values of the volume and number size spectra are shown in Figs. 5 and 6 respectively, for size ranges satisfying the above criterion. As seen in the figures, effective size ranges (solid lines) of inferring are usually limited to the range from 0.05 μm to less than 2 μm, for visible light with a wavelength of 514.5 nm. While the effective size ranges are exceptionally wide in July and September, contributions to $\beta_j(\Theta_i)$ from the ranges outside the above-mentioned range are very small. It is noted that the criterion $C_{max} > 0.1$ is applied to our data $\beta_j(\Theta_i)$ within the scattering angles $7° \leq \Theta \leq 170°$. Thus, if forward portions of the scattering data (i.e., data for $\Theta < 7°$) are added, particles with larger sizes are expected to become necessary to reconstruct the differential scattering cross section data, or equivalently, the reliability of inferring the size distribution may increase for larger sizes. Because of the restricted particle size of interest, the size distribution functions can be approximated by a power law distribution rather than a modified gamma distribution. The values of the exponent ν are shown in Table 2 for each month. The value of the exponent averaged over whole data is given as $\nu = 3.4 \pm 0.3$, which is in good agreement with the previous results. The values of $dN/d\ \ln r$ averaged over all months are shown in Fig. 7a, where the optimum spectrum shown by the solid line is given by

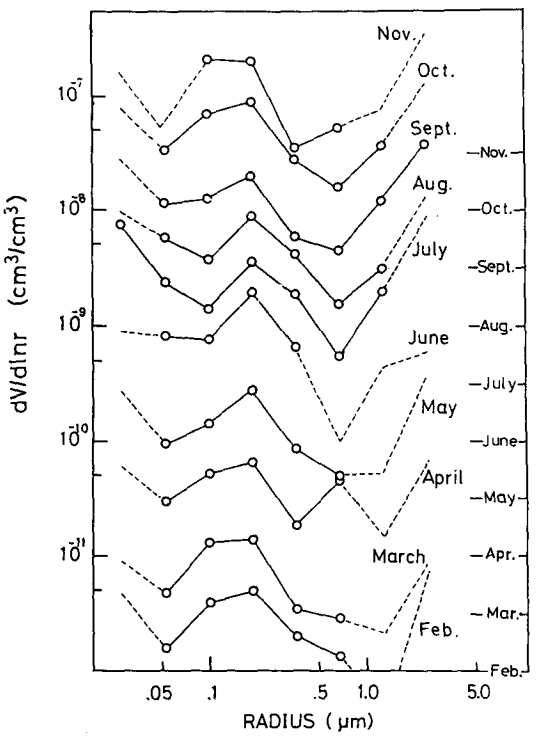

FIG. 6. Monthly means of aerosol volume spectra corresponding to Fig. 5. Horizontal lines on the right side indicate the level $dV/d\ \ln r = 10^{-12}$ cm^3 cm^{-3} for respective months.

TABLE 2. Monthly mean values of the exponent ν of the best fitting power law size distribution.

Month	ν	Cases
2	3.2	11
3	3.4	36
4	3.2	58
5	3.3	35
6	3.0	12
7	3.4	16
8	3.3	27
9	2.8	21
10	3.2	27
11	3.9	7
Average	3.4 ± 0.3	

$$dN/d\ln r = 1.65 \times 10^{-14} r^{-3.36} \text{ [particles cm}^{-3}\text{]}. \quad (2)$$

Returning to Fig. 6, it is noticeable that the maximum volume concentration appears in the size range around 0.2 μm. Since the aerosols in this size range contribute to $\beta_f(\Theta)$ most effectively, the volume concentration inferred there is most reliable. This mode of volume spectrum is recognizable in all months, being considered to be characteristic in the size distribution of aerosols in the lower atmosphere. Fig. 7b shows the volume spectrum geometrically averaged over all months. The total volume of aerosols within the optically effective size range was estimated to be 6.5×10^{-5} cm^3 m^{-3}. Many authors (e.g., Patterson and Gillette, 1977) have pointed out that the volume spectra of polydispersed aerosols in the atmosphere can be expressed by the log-normal distribution. The mean volume spectrum obtained in our measurements can also be approximated by the log-normal distribution with the geometrical mean radius $r_g = 0.138$ μm and the geometrical standard deviation $\sigma_g = 2.56$, as shown by the solid line in Fig. 7b. Comparing Figs. 7a and 7b, we can see that the log-normal approximation to the volume spectrum surpasses the power law approximation to the number spectrum in its accuracy. On the other hand, Whitby *et al.* (1972) have investigated the size distribution of smog particles over the size range 0.003–6.8 μm in diameter using the Minnesota Aerosol Analyzing System, and found a bimodal volume spectrum with the saddle point around 1–2 μm in diameter prevails universally. Patterson and Gillete (1977) have drawn a similar conclusion from extended measurements of the size distribution of soil-derived aerosols. In addition, they found that the variability in bimodal features depends upon the loading in the atmosphere. According to their notation, the size range of our inferring is in good accordance with Mode C, which appears in conditions of light aerosol loading. It has been speculated in both papers that the Mode C particles consist of secondary particles such as photochemical reaction products. It is of interest to know the composition of aerosols in this optically most effective size range in order to conjecture the complex index of refraction of individual particles. As shown in Fig. 4, there has been no appreciable difference in the volume spectra between noon and midnight. This suggests that the origin in the Mode C particles may extend widely, as compared with the observation by Whitby *et al.* (1972). Patterson and Gillette (1977) have also found that the second mode (i.e., Mode A) of the volume spectrum has its maximum around 2–3 μm in radius. The volume spectra obtained in this study also exhibit such a similar tendency, but it is difficult to identify the above conclusion on account of restricted reliability in that size range, that is, the Mode A paricles do not play an important role in light scattering phenomena in the lower atmosphere with light aerosol loading. In these respects, more complicated measurements including different particle sizing techniques are required to make such detailed analyses.

3) SCATTERING CROSS SECTION AND RELATED QUANTITIES

Utilyzing the complex index of refraction m and the volume spectrum $dV/d\ln r = v$, inferred from scattering measurements, other optical characteristics of aerosols can easily be estimated. The scattering cross section C_{sca} and the albedo ω_0 for single scattering are given as

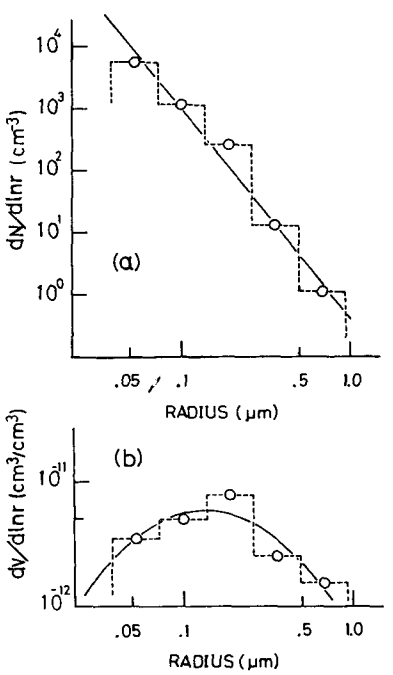

FIG. 7. Size distribution of aerosols averaged over the whole period: (a) Mean number spectrum (dotted line) as compared with the power law with the exponent of 3.4 (solid line). (b) Mean volume spectrum (dotted line) as compared with the best-fit log-normal distribution with $r_g = 0.14$ μm and $\sigma_g = 2.6$ (solid line).

$$C_{sca} = \frac{3}{4}\int \frac{1}{r^2} Q_{sca}(m, \alpha) \frac{dV}{d \ln r} dr$$
$$\approx k \sum f_{l,sca}(m) v_l, \quad (3)$$

where

$$f_{l,sca}(m) = \frac{3}{4}\int_{\alpha_{l-1/2}}^{\alpha_{l+1/2}} \alpha^{-2} Q_{sca}(m, \alpha) d\alpha, \quad (4)$$

and α, $Q_{sca}(m, \alpha)$ and k are the size parameter, the efficiency factor for scattering and the wavenumber, respectively. The extinction cross section C_{ext} is also given by substituting the efficiency factor for extinction Q_{ext} for Q_{sca} in the above equations. Thus, we have

$$\omega_0 = C_{sca}/C_{ext}. \quad (5)$$

The monthly mean values of C_{sca} and ω_0 are shown in Fig. 8. The increase of C_{sca} in the warm season is in good accordance with other observational results obtained heretofore, such as seasonal variation of the atmospheric transmission. The values of ω_0 also increase in the warm season, ranging from 0.7 to 0.88. These values are considerably smaller than those reported in the literature. This phenomenon is attributable, at least partly, to the fact that the effective size range of particles inferrable from our scattering measurements does not necessarily cover the wide range required for the typical Junge-type distribution. If we overestimate the spectral volume of particles out of the reliable size range (i.e., the ranges with $C_{max} < 0.1$), the absorption cross section C_{abs} tends to be overestimated, resulting in underestimation of the single-scattering albedo ω_0. It is difficult to measure directly the absorption cross section, C_{abs}, so that the estimated values of ω_0 cannot be confirmed from independent measurements. At present, we can only say that the estimated values of ω_0 is quite consistent with measurements of the differential scattering cross section in the range of scattering angles $7° \leqslant \Theta \leqslant 170°$.

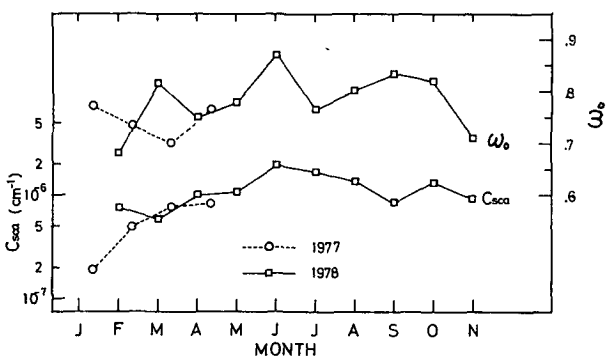

FIG. 8. Seasonal variation of the monthly mean values of the single-scattering albedos ω_0 and the scattering cross section C_{sca}.

4. Summary

The complex index of refraction and the size distribution (volume spectrum) of aerosols have been estimated simultaneously by applying the inversion library method to measured angular distributions of both polarization components of the scattered light. It was found that the monthly mean values of m_r and m_i vary within the ranges 1.47–1.57 and 0.009–0.037, respectively, showing a tendency to decrease in summer corresponding to an increase in relative humidity.

The volume spectrum of aerosols can be expressed by a log-normal distribution function with the geometrical mean radius of 0.138 μm and the standard deviation of 2.56, as an average of the whole data. If we adopt the power law for the corresponding number spectrum, the best fit is expressed as $dN/d \ln r \sim r^{-3.4}$, being in good accordance with other previous results.

To assess the effect of the non-sphericity of particles on the analyzed results is the most important problem in the future.

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