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Determination of the Ground Albedo and the Index of Absorption of Atmospheric Particulates by Remote Sensing. Part I: Theory[1]

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ABSTRACT

A statistical technique is developed for inferring the optimum values of the ground albedo and the effective imaginary term of the complex refractive index of atmospheric particulates. The procedure compares measurements of the ratio of the hemispheric diffuse to directly transmitted solar flux density at the earth's surface with radiative transfer computations of the same as suggested by Herman *et al.* (1975). A detailed study is presented which shows the extent to which the ratio of diffuse to direct solar radiation is sensitive to many of the radiative transfer parameters. Results indicate that the optical depth and size distribution of atmospheric aerosol particles are the two parameters which uniquely specify the radiation field to the point where ground albedo and index of absorption can be inferred. Varying the real part of the complex refractive index of atmospheric particulates as well as their vertical distribution is found to have a negligible effect on the diffuse-direct ratio. The statistical procedure utilizes a semi-analytic gradient search method from least-squares theory and includes a detailed error analysis.

1. Introduction

Within the past decade there has been an increasing concern about the observed decline in the mean temperature of the Northern Hemisphere which begain in the 1940's. Because this decline followed a steady rise in mean temperature which began at the end of the last century, there have been countless attempts at explaining this behavior. Bryson (1968) discusses the various factors which could alter the basic climate of of the earth. These factors have included a change in sunspots, an increase in the carbon dioxide content of the atmosphere due to the consumption of fossil fuels, and a change in the properties and number of particulates in the atmosphere. All of these factors influence the complicated energy budget of the earth-atmosphere-ocean system.

Solid and liquid particles suspended in the atmosphere affect climate through two major processes. They directly affect the transfer of radiant energy in the clear atmosphere as well as affecting the optical and microphysical properties of clouds (Twomey, 1974, 1977; Ackerman and Baker, 1977). The first of these processes will be considered in some detail in this series of articles.

The first successful applications of radiative transfer models to the problem of actual particles in the atmosphere were made by Rasool and Schneider (1971) and Yamamoto and Tanaka (1972). The latter-named authors have computed the global albedo which results as particles of varying values of the imaginary index of refraction (index of absorption) are introduced into a model atmosphere. They conclude that an increasing turbidity will always lead to an enhanced global albedo (i.e., cooling) if the index of absorption is less than 0.05.

Since Yamamoto and Tanaka only considered surfaces with a ground albedo less than 0.15, Wang and Domoto (1974) and Herman and Browning (1975) extended the analysis to include surfaces with greater reflection. Both sets of authors found that heating can result if the surface reflectivity exceeds about 0.4, regardless of the index of absorption of the particulates. These studies address a very important point, that both ground albedo and index of absorption are important in assessing the effect of aerosol particles on the earth and its atmosphere.

It has been suggested in a preliminary study by Herman *et al.* (1975) that the ratio of the hemispheric diffuse to the directly transmitted solar flux density at the earth's surface be used as an indirect means of inferring the index of absorption of the atmospheric particulates. The diffuse radiation field in the earth's atmosphere at the shorter wavelengths (visible and ultraviolet) is complicated by its dependence on such things as the vertical distribution, optical depth, size distribution and index of refraction of the efficiently scattering atmospheric particulates as well as the position of the sun and the reflectivity of the earth's surface. In order to apply measurements of the diffuse radiation

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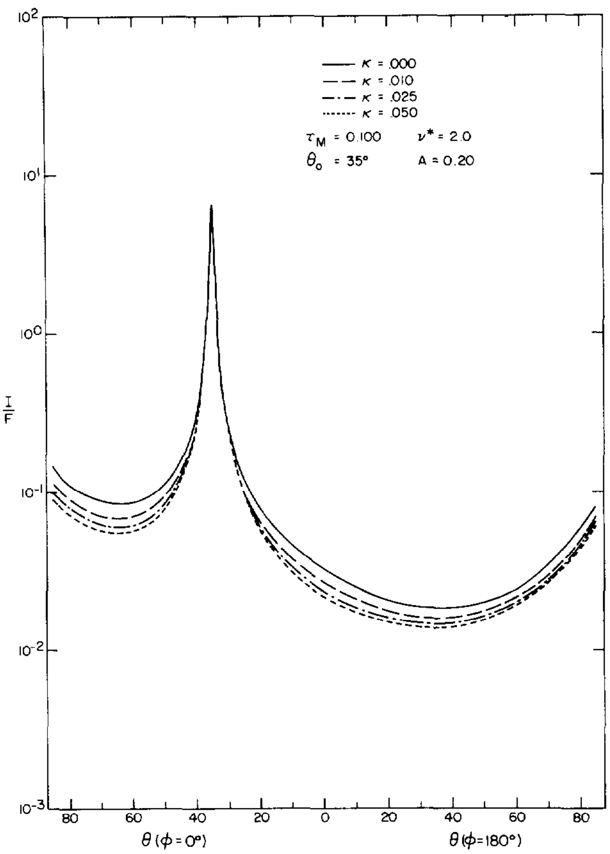

FIG. 1. The ratio of the transmitted intensity to directly transmitted solar flux density for $n=1.54$ and for four values of the imaginary part of the complex refractive index.

field to theoretical calculations of the same, as Herman *et al.* (1975) have suggested, it therefore becomes necessary to establish the relative importance and sensitivity of the radiation field to all of the radiative transfer parameters.

This paper considers the sensitivities and assumptions of primary importance for a successful application of theoretical calculations to real data in a real atmosphere. It will be demonstrated that the ratio of the hemispheric diffuse to directly transmitted solar flux density at the earth's surface can be measured as a function of solar zenith angle and, with certain *a priori* information about the Mie optical depth and aerosol size distribution, compared to theoretical calculations to assess the magnitude of both the ground albedo and the index of absorption of atmospheric particulates. An inversion procedure is developed which makes use of the laws of diffuse reflection and transmission together with a gradient search method from least-squares theory. Formulas are derived for estimating the magnitude of the errors in both ground albedo and the imaginary index of refraction of the particles.

In Part II of this series (King, 1979) measurements of the diffuse-direct ratio which have been collected in Tucson are presented and discussed. Data for days during which the atmosphere was clear and stable are analyzed and the optimum values of the ground albedo and index of absorption of atmospheric aerosol particles are presented.

2. Theoretical sensitivity of the diffuse radiation field

To assess the sensitivity of the diffuse radiation field to the many radiative transfer parameters mentioned above, computations were made for 364 model atmospheres illuminated from above by the sun at five solar zenith angles. The method used to calculate the diffuse radiation field is described by Herman and Browning (1965). This solution to the equation of radiative transfer consists of a Gauss-Seidel iteration technique and many numerical integrations over optical depth (τ), zenith angle ($\theta=\cos^{-1}\mu$) and azimuth angle (ϕ). In all calculations to be presented below, the bottom boundary of the atmosphere is assumed to reflect radiation according to Lambert's law with some albedo A. The computations are made for a plane-parallel, vertically inhomogeneous atmosphere with the dust vertically distributed according to Elterman (1968). In all calculations the wavelength of incident illumination is $\lambda=0.5550$ μm which, at the pressure level of Tucson, has a Rayleigh (molecular) optical depth $\tau_R=0.0860$. The phase matrix for the particles is computed from Mie theory [see van de Hulst (1957) for details] and is thus a function of the complex refractive index and size distribution of the particulates, the shape having been assumed spherical. Absorption by atmospheric ozone is neglected in all computations for reasons discussed below.

The aerosol size distribution for the sensitivity tests was assumed to be that proposed by Junge (1955), given as

$$n(r)=Cr^{-(\nu^*+1)}, \qquad (1)$$

where C and ν^* are constants and $n(r)$ is the number density of particles per unit interval of radius r. All model calculations presented below are for Junge distributions where the radius r extends from 0.01 to 10.01 μm.

Fig. 1 illustrates the sensitivity of the transmitted intensity field in the $\theta=0°$ and 180° plane (i.e., the vertical plane containing the sun) to the imaginary term κ of the complex refractive index of the aerosol particles. The refractive index $m=n\text{-}\kappa i$ is a complex number; the real part n is the ordinary index of refraction of the material, while the imaginary part κ determines the absorption of electromagnetic radiation. All intensities are plotted relative to the directly transmitted solar flux density at the earth's surface, given by the Lambert-Beer law as

$$F=F_0e^{-\tau_t/\mu_0}. \qquad (2)$$

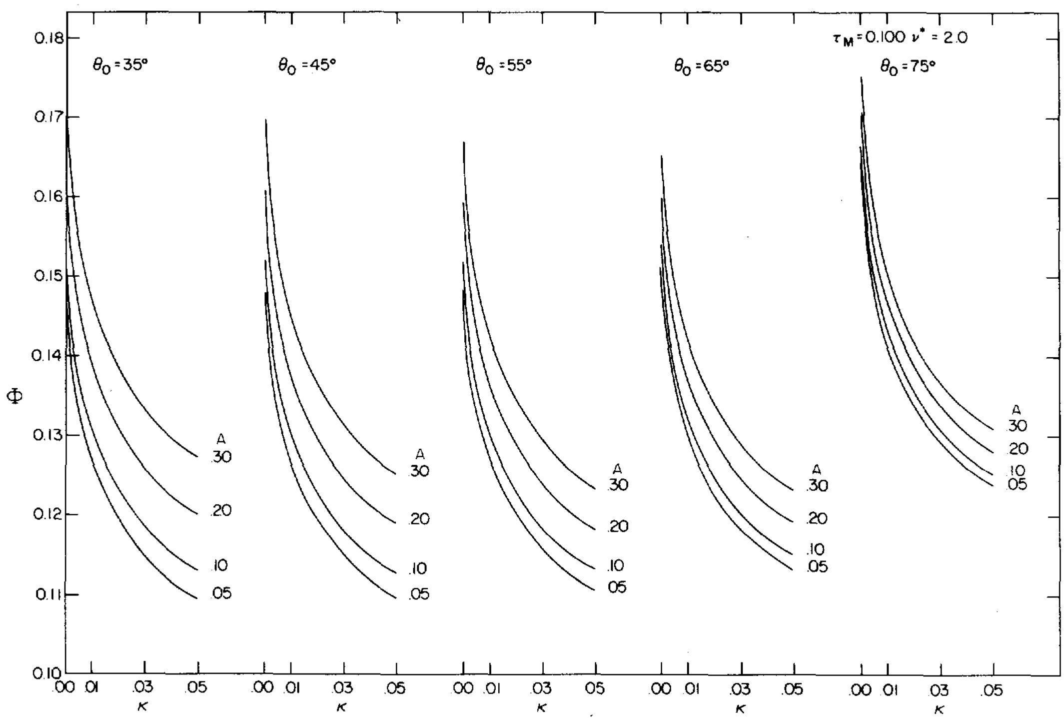

FIG. 2. The ratio of the hemispheric diffuse to directly transmitted solar flux density as a function of index of absorption, ground albedo and solar zenith angle.

In this expression F_0 is the solar flux density incident on the top of the atmosphere ($\tau=0$ level), μ_0 the cosine of the solar zenith angle θ_0, and τ_t the total optical depth of the atmosphere. From measurements of the directly transmitted solar radiation at wavelengths for which absorption due to molecular species other than ozone can be neglected, it is possible to determine the optical depth τ_M due to particulates alone, known as the Mie optical depth (King and Byrne, 1976).

Although the particular model atmosphere illustrated in Fig. 1 has $\tau_M=0.100$, $\nu^*=2.0$, $\theta_0=35°$, $A=0.20$ and $n=1.54$, the obvious sensitivities in the figure are similar for all other model atmospheres investigated, models which had values of A from 0.05 to 0.30, κ from 0.00 to 0.20, τ_M from 0.00 to 0.30 and ν^* from 2.0 to 4.0. In general, the solar aureole (the strong intensity region in the vicinity of the sun) is very insensitive to κ, being primarily produced by Fraunhofer diffraction around the particles, while the intensity at larger scattering angles shows a marked sensitivity to absorption.

Computations of the intensity field similar to Fig. 1 have been compared to measurements of the intensity by Herman *et al.* (1971) where theoretical calculations were made using measured values for τ_M and θ_0, while assumed values were used for the index of refraction, Junge size distribution parameter ν^* and ground albedo. Although no attempt was made to optimize the assumed values of ν^*, A and m, the agreement between measurement and theory was considered quite acceptable, and demonstrated for the first time that inclusion of aerosol particles into a Rayleigh atmosphere yields better agreement between observation and theory. It is possible, however, to optimize the determination of the index of refraction and ground albedo by examining the diffuse radiation field as a function of solar zenith angle. The method by which this is accomplished is presented in Section 4.

The hemispheric diffuse flux densities at the earth's surface are given by

$$F^{\pm}(\tau_t)=\int_0^{2\pi}\int_0^1 I(\tau_t,\pm\mu,\phi)\mu d\mu d\phi, \tag{3}$$

where the $+(-)$ sign refers to intensities propagating in the upward (downward) direction at the level τ_t. If the downward hemispheric diffuse flux density is calculated using (3), it follows that $F^-(\tau_t)$ should monotonically decrease as κ increases since the intensity field has this behavior for all observation angles (μ,ϕ) as seen upon examination of Fig. 1. Fig. 2 presents a family of curves for the diffuse flux density versus imaginary index of refraction κ (hereafter referred to as the index of absorption) where again the diffuse flux density is plotted relative to the directly transmitted solar flux density. This ratio,

$$\Phi=F^-(\tau_t)/F, \tag{4}$$

shows the required sensitivity to κ as well as sensitivities to θ_0 and A. Yamamoto and Tanaka (1972) investi-

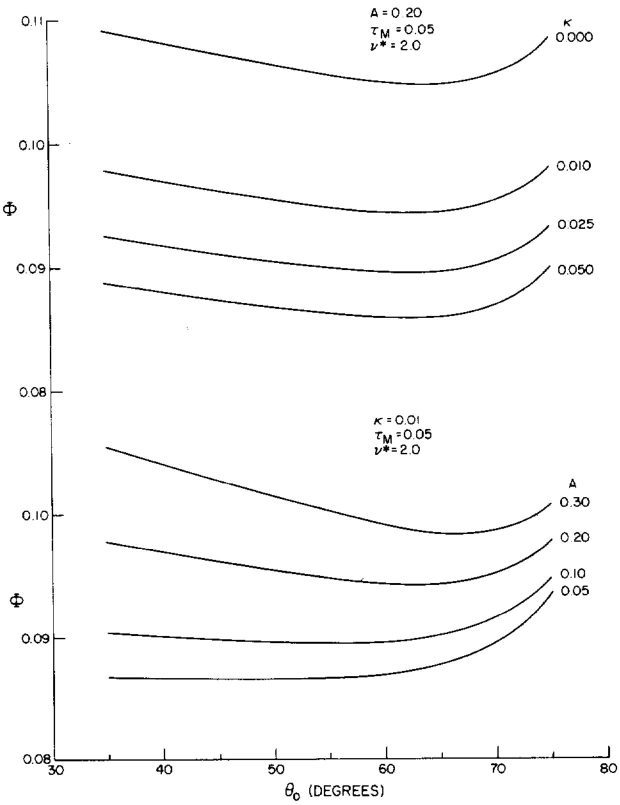

FIG. 3. The diffuse-direct ratio as a function of solar zenith angle for $\tau_M=0.05$, $\nu^*=2.0$, and for different values of κ and A.

gated some of the sensitivities of Φ, including its dependence on κ and λ, and came to the same conclusions for the sensitivity of Φ to absorption. However, Yamamoto and Tanaka made no attempt to investigate this sensitivity as a potential remote sensing means of inferring κ and A directly. Herman *et al.* (1975), on the other hand, show curves similar to Fig. 2 and suggest using the sensitivity of Φ, termed the diffuse-direct ratio, to infer the index of absorption of the atmospheric particulates.

In addition to the obvious instrumental advantage of making a relative (ratio) measurement in lieu of an absolute flux density measurement, there is a theoretical advantage associated with formulating the ratio of diffuse to direct solar flux density. Since ozone has a very broad absorption feature extending from about 0.4 to 0.9 μm, known as the Chappuis continuum, ozone must be considered when calculating the diffuse radiation field at the earth's surface having wavelengths in this region. Since most of the absorption by O_3 occurs above 20 km while the majority of scattering by molecules and particles is confined to heights well below 20 km, O_3 serves mainly to attenuate the direct solar beam before the radiation can interact with the scattering atmosphere. It is expected, therefore, that O_3 absorption will affect the diffuse and direct flux densities by nearly the same fraction such that the diffuse-direct ratio will be largely insensitive to absorption by O_3. Since approximately 7.5% of the total ozone content of the earth's atmosphere is located within the troposphere (Elterman, 1968), however, there is an imperfect separation with height. As a direct consequence of the presence of tropospheric O_3 one would expect to see some minor sensitivity to O_3 absorption, particularly for wavelengths in the heart of the Chappuis band (centered near 0.6120 μm). This subject will further be addressed in part II of this series (King, 1979) where the experimental results are presented.

The sensitivity of the diffuse-direct ratio to solar zenith angle and ground albedo is also illustrated in Fig. 2, where it is apparent that the affect which a changing solar zenith angle has on Φ depends upon the values of κ and A. For small indices of absorption and large values of surface albedo (e.g., $\kappa=0.01$, $A=0.30$), Φ decreases slightly with θ_0 until about 65° after which point it starts to increase. For smaller values of the surface reflectivity, however, Φ increases with zenith angle for all $\theta_0>45°$, regardless of the index of absorption. Fig. 3 explicitly illustrates the combined effects of κ and A on the zenith angle dependence of the diffuse-direct ratio for a model atmosphere having $\tau_M=0.05$ and $\nu^*=2.0$. The family of curves for a fixed ground albedo (top portion of the figure) shows that the predominant effect of the index of absorption is to alter the *magnitude* of Φ with very little effect on its functional relationship with respect to solar zenith angle. Ground albedo, on the other hand, affects both the magnitude and *shape* of Φ as a function of θ_0. This is readily seen upon examination of Fig. 3 for the family of curves having a fixed value for the index of absorption of the atmospheric particulates (bottom portion of the figure). This suggests that a measurement of the diffuse-direct ratio versus solar zenith angle contains enough information to determine both the imaginary index of refraction of the atmospheric particulates as well as the reflectivity of the earth's surface, given a knowledge of the aerosol size distribution and Mie optical depth, provided a measurement accuracy on the order of 1% can be achieved. The instrumentation used to make the measurements was capable of this requirement (King, 1979).

Up to this point little mention has been made of the sensitivity of the diffuse-direct ratio to Mie optical depth and aerosol size distribution. This is because these parameters are not variables to be determined from the diffuse flux density measurements but are, instead, input parameters to be determined *a priori*. Since an error in either τ_M and/or $n(r)$ leads to errors in the determination of κ and A, the Mie optical depth and aerosol size distribution play a very important role in the successful application of the diffuse-direct technique to real data in a real atmosphere.

Fig. 4 shows Φ as a function of τ_M for two refractive indices and three Junge size distributions. As the Mie optical depth increases, the diffuse-direct ratio increases for aerosol particles having a fixed refractive index and size distribution, a feature which can also be deduced from the results of Yamamoto and Tanaka (1972). It is obvious from this figure that any uncertainty in τ_M leads to an uncertainty in the determination of κ (and A), even if the aerosol size distribution is known. It is similarly apparent that any uncertainty in the aerosol size distribution will lead to incorrect assessments of the value for κ, even if the Mie optical depth is known with a high degree of accuracy. In general, neither τ_M nor $n(r)$ are known precisely, and thus they both must be estimated from alternative measurements.

The method developed for obtaining accurate Mie optical depth measurements (King and Byrne, 1976) optimizes the determination of the ozone optical depth, the major uncertainty in obtaining τ_M from measurements of the total optical depth. By making use of the spectral variation of Mie optical depth it is possible to obtain an estimate of the columnar aerosol size distribution $n_c(r)$, defined as the number of particles per unit area per unit radius interval in a vertical column through the atmosphere (King *et al.*, 1978). This work further demonstrates that the index of refraction has very little effect on the size distribution determined from spectral attenuation measurements (at least for $1.45 \leqslant n \leqslant 1.54$ and $0.00 \leqslant \kappa \leqslant 0.03$), thus allowing a determination of $n_c(r)$ essentially independent of κ (and of course A). By making use of the optical depth and size distribution of the atmospheric particulates thus obtained, it is possible to make explicit radiative transfer calculations for a particular day at a given wavelength similar to the set presented in Figs. 2 and 3. In this manner errors in the determination of κ and A are minimized since the need to interpolate in wavelength, size distribution and Mie optical depth can thus be eliminated.

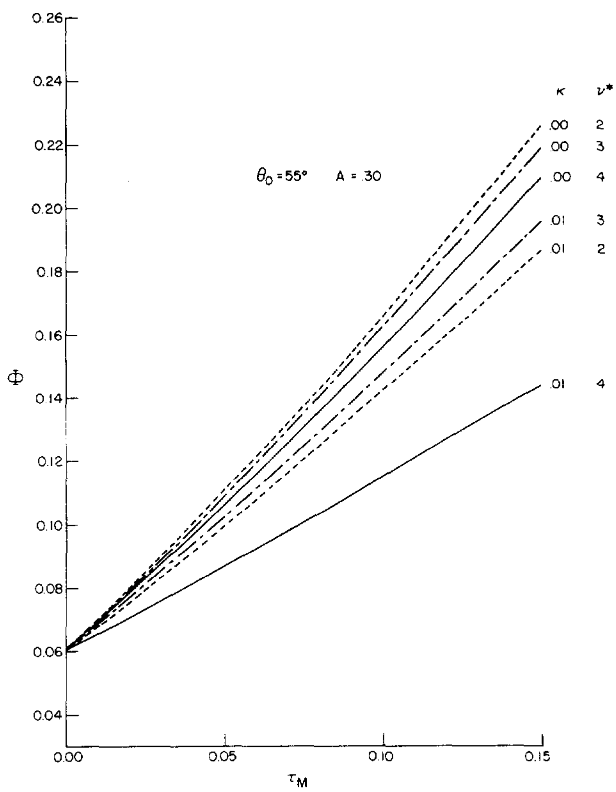

FIG. 4. The diffuse-direct ratio as a function of Mie optical depth for $\theta_0 = 55°$, $A = 0.30$, and for different κ and ν^* values.

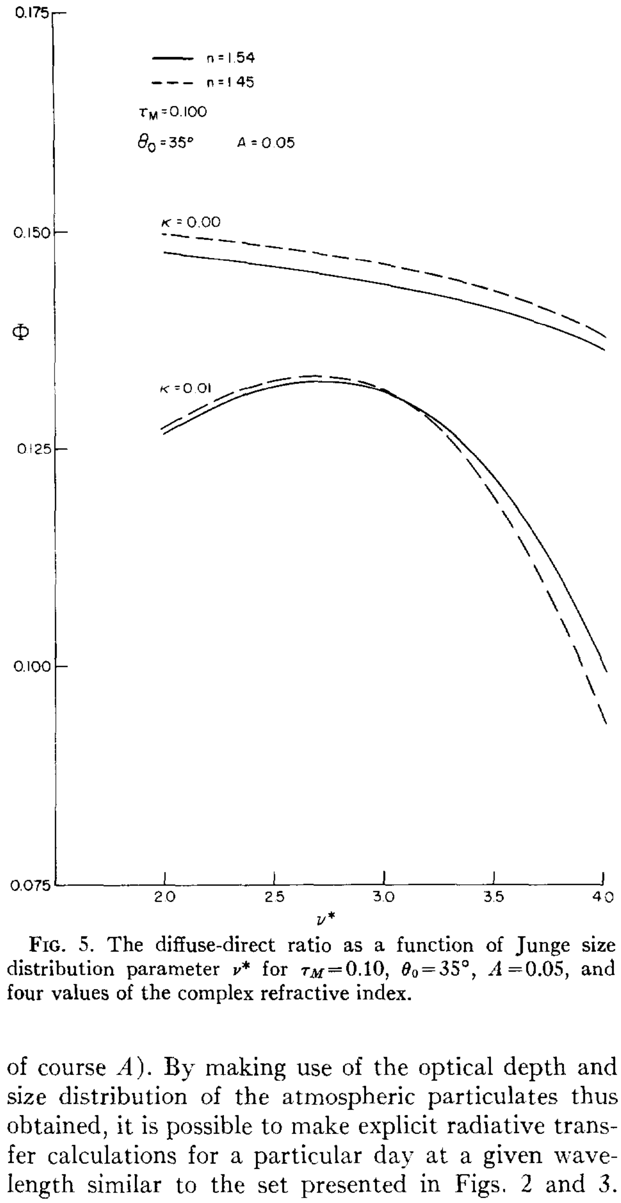

FIG. 5. The diffuse-direct ratio as a function of Junge size distribution parameter ν^* for $\tau_M = 0.10$, $\theta_0 = 35°$, $A = 0.05$, and four values of the complex refractive index.

The sensitivity of the diffuse-direct ratio to Junge size distribution parameter ν^* is also illustrated in Fig. 4 where it is apparent that the effect which a changing Junge parameter has on Φ depends upon the value of κ. For $\kappa = 0.00$, Φ decreases monotonically with ν^* regardless of the Mie optical depth while for larger values of the index of absorption Φ is nonmonotonic in ν^*, having the largest value for $\nu^* \approx 3.0$. Fig. 5 explicitly illustrates Φ as a function of ν^* for four values of the complex refractive index. In addition to the sensitivities of the diffuse-direct ratio to ν^* just mentioned, the same sensitivities are evident for both real indices of refraction illustrated, *viz.*, 1.45 and 1.54.

TABLE 1. Values of $\bar{s}(\tau_t)$ and $t(\tau_t,\mu_0)$, at $\mu_0=0.819$ and 0.259 and at $\lambda=0.5550$ μm, for a variety of atmospheric models.*

τ_M	κ	ν^*	$\bar{s}(\tau_t)$	$t(\tau_t,0.819)$	$t(\tau_t,0.259)$
0.00			0.07478	0.04061	0.03619
0.05	0.000	2	0.08788	0.07852	0.06012
0.05	0.000	3	0.09137	0.07693	0.05794
0.05	0.000	4	0.09608	0.07380	0.05604
0.05	0.025	2	0.07693	0.06658	0.05190
0.05	0.025	3	0.08261	0.06729	0.05126
0.05	0.025	4	0.07408	0.05114	0.04053
0.10	0.000	2	0.09989	0.11384	0.07901
0.10	0.000	3	0.10643	0.11065	0.07514
0.10	0.000	4	0.11535	0.10452	0.07179
0.10	0.025	2	0.07863	0.09026	0.06327
0.10	0.025	3	0.08923	0.09152	0.06239
0.10	0.025	4	0.07357	0.06035	0.04313

* All calculations are for Junge distributions where the radius extends from 0.01 to 10.01 μm.

It is evident from this figure, as well as from computations made for other values of τ_M, θ_0 and A, that the effect of the real term of the complex refractive index on the magnitude of the diffuse-direct ratio is quite negligible compared to the much greater effect of absorption. According to Gebbie *et al.* (1951), Eiden (1966) and Hänel (1968), natural haze has a real refractive index lying somewhere between 1.33 and 1.57; the former value corresponds to that of water droplets and the latter value to several salts, sulfates and silicates arising from natural and man-made sources. In desert regions like Tucson, it is reasonable to expect n to lie in the region between 1.45 and 1.54 due to the sources of the particulates and the relatively low humidity of the atmosphere.

The main significance of Fig. 5 is to illustrate the relative insensitivity of the diffuse-direct ratio to the real, as opposed to imaginary, part of the complex refractive index of the suspended atmospheric particulates. From inversion of spectral optical depth data in order to infer the columnar aerosol size distribution, King *et al.* (1978) have suggested that scarcely 20% of all days over Tucson have aerosol size distributions which can be adequately described as Junge. Most days appear to have a combination of a Junge plus a log-normal type of distribution. Although the actual size distribution to be used in analyzing the diffuse-direct ratio measurements on a particular day is the one obtained by inverting the spectral Mie optical depth measurements, Fig. 5 nevertheless indicates that the diffuse-direct technique is largely insensitive to the real part of the complex refractive index, regardless of size distribution.

Since spectral optical depth measurements are capable of sensing only the columnar aerosol size distribution, a logical concern is the sensitivity of the hemispheric diffuse flux density to the vertical distribution of the particles. By assuming that the shape of the size distribution is height independent, Herman and Browning (1975) performed computations of the reflected flux density out of the top of an atmosphere having an Elterman (1968) height profile for the particulate concentration as well as two perturbed Elterman profiles simulating pollution conditions under an inversion layer. Their results indicate that the total Mie optical depth is the principal factor determining the diffuse flux density with little effect attributed to the vertical distribution. Similar computations for the transmitted diffuse flux density at the earth's surface indicate that errors less than 0.75% arise if $\tau_M=0.10$. The relative insensitivity of the diffuse-direct ratio to the height profile of the aerosol particles is a fortunate result since obtaining that additional information would require another instrument such as a monostatic lidar [see Spinhirne (1977) for details].

3. Diffuse reflection and transmission

It is sometimes convenient to express the diffusely reflected light at the top of the atmosphere and the diffusely transmitted light at the bottom of the atmosphere in terms of a reflection function $S(\tau_t;\mu,\phi;\mu_0,\phi_0)$ and a transmission function $T(\tau_t;\mu,\phi;\mu_0,\phi_0)$. In terms of these functions, the reflected and transmitted intensities from a horizontally homogeneous atmosphere illuminated from above by a parallel beam of radiation of incident flux density F_0 may be expressed in the forms

$$I(0,+\mu,\phi)=\frac{F_0}{4\pi\mu}S(\tau_t;\mu,\phi;\mu_0,\phi_0), \qquad (5)$$

$$I(\tau_t,-\mu,\phi)=\frac{F_0}{4\pi\mu}T(\tau_t;\mu,\phi;\mu_0,\phi_0). \qquad (6)$$

Eqs. (5) and (6) can be viewed as defining the functions S and T for the case of zero ground reflectivity. The advantage of including the factor of $1/\mu$ in the expressions for the emerging intensities given above is that the reflection and transmission functions obey the Helmholtz principle of reciprocity as discussed in detail by Chandrasekhar (1960).

In order to examine the effect which a non-zero ground reflectivity has on the downward hemispheric diffuse flux density $F^-(\tau_t)$, and hence on Φ, it is necessary to consider an extension of (6) to include not only plane-parallel illumination from above the atmosphere but also diffuse radiation from below. The inward intensity at the level τ_t, for cases when the surface is assumed to reflect radiation according to Lambert's law with some albedo A, is given by (Chandrasekhar, 1960)

$$I(\tau_t,-\mu,\phi)=\frac{F_0}{4\pi\mu}\left[T(\tau_t;\mu,\phi;\mu_0,\phi_0)+\frac{4A}{1-A\bar{s}(\tau_t)}\mu\mu_0\gamma_1(\tau_t,\mu_0)\frac{s(\tau_t,\mu)}{\mu}\right]. \qquad (7)$$

In this expression,

$$s(\tau_t,\mu)=\frac{1}{4\pi}\int_0^{2\pi}\int_0^1 S(\tau_t;\mu,\phi;\mu',\phi')d\mu'd\phi',$$

$$t(\tau_t,\mu)=\frac{1}{4\pi}\int_0^{2\pi}\int_0^1 T(\tau_t;\mu,\phi;\mu',\phi')d\mu'd\phi',$$

$$\bar{s}(\tau_t)=2\int_0^1 s(\tau_t,\mu)d\mu,$$

$$\gamma_1(\tau_t,\mu)=e^{-\tau_t/\mu}+\frac{t(\tau_t,\mu)}{\mu}. \quad (8)$$

In order to determine Φ, it is necessary to evaluate the downward hemispheric diffuse flux density $F^-(\tau_t)$ for application of (4). From definitions, it follows that

$$F^-(\tau_t)=\int_0^{2\pi}\int_0^1 I(\tau_t,-\mu,\phi)\mu d\mu d\phi$$

$$=F_0\left[t(\tau_t,\mu_0)+\frac{A}{1-A\bar{s}(\tau_t)}\mu_0\gamma_1(\tau_t,\mu_0)\bar{s}(\tau_t)\right]. \quad (9)$$

Substituting (9) into (4) and recalling the definition of F, it follows that

$$\Phi=e^{\tau_t/\mu_0}\left[t(\tau_t,\mu_0)+\frac{A\bar{s}(\tau_t)}{1-A\bar{s}(\tau_t)}\mu_0\gamma_1(\tau_t,\mu_0)\right], \quad (10)$$

which, upon rearranging the order of terms, can be shown to yield

$$\Phi=\frac{t(\tau_t,\mu_0)e^{\tau_t/\mu_0}+A\mu_0\bar{s}(\tau_t)}{1-A\bar{s}(\tau_t)}. \quad (11)$$

This expression for the diffuse-direct ratio gives its dependence explicitly upon ground albedo, solar zenith angle and optical depth in terms of the reflection and transmission functions for zero ground reflectivity. The dependence of Φ on the index of refraction and size distribution of the atmospheric particulates is, of course, through the functions $t(\tau_t,\mu_0)$ and $\bar{s}(\tau_t)$. These functions may be determined for specific values of κ and $n_c(r)$ through solutions of the transfer equation for zero ground reflectivity. The diffuse-direct ratio may then be determined for any ground albedo through application of (11).

For a combination of optical depth and ground albedo such that $A\bar{s}(\tau_t)\ll 1$, it follows from (11) that Φ is very nearly a linear function of A with an intercept of $t(\tau_t,\mu_0)e^{\tau_t/\mu_0}$ and a slope of $\mu_0\bar{s}(\tau_t)$. This result will be employed in the next section so that the lengthy calculations required to determine $\bar{s}(\tau_t)$ may be avoided. The function $\bar{s}(\tau_t)$ physically represents the value of the spherical albedo, i.e., the ratio of the total radiation reflected from a spherical atmosphere to the total radiation incident from a distant source when the surface albedo $A=0.00$. For small optical depths $\bar{s}(\tau_t)$ is proportional to τ_t where the proportionality constant is a function of the single scattering phase function. Values of $\bar{s}(\tau_t)$ and $t(\tau_t,\mu_0)$, for $\mu_0=0.819$ ($\theta_0=35°$) and $\mu_0=0.259$ ($\theta_0=75°$) and for $\lambda=0.5550$ μm, are presented in Table 1 for a variety of atmospheric models. When $\kappa=0.00$, $\bar{s}(\tau_t)$ increases monotonically with τ_t to a limiting value of 1.0 when $\tau_t\to\infty$. Since the slope of Φ as a function of A is approximately equal to $\mu_0\bar{s}(\tau_t)$ in the clear atmosphere, it follows that the slope decreases with increasing zenith angle (due to μ_0) and increases with increasing Mie optical depth [due to $\bar{s}(\tau_t)$]. The first of these sensitivities can readily be seen upon examination of Figs. 2 and 3.

For the case in which polarization is included, as it has been in the radiative transfer computations presented above, the principle of reciprocity is complicated somewhat, and the reflection and transmission functions become four-vectors. Chandrasekhar (1960) derives a relationship analogous to (7) for the case of scattering according to a phase matrix. The general conclusions and functional relationships derived in this section remain essentially unaltered, however, so nothing is gained by the increased complexity of the mathematical formulation.

4. Determination of ground albedo and index of absorption

The reflection and transmission functions in a planetary atmosphere depend on the size distribution and index of refraction of the atmospheric particulates as well as the optical depth and solar zenith angle explicitly indicated (see Table 1). For the cases in which the Mie optical depth and aerosol size distribution can be determined, the functions $t(\tau_t,\mu_0)$ and $\bar{s}(\tau_t)$ depend primarily on solar zenith angle and index of absorption. From (11) it therefore follows that the functional form of the diffuse-direct ratio may be written as

$$\Phi(\theta_0,\kappa,A)=\frac{a(\theta_0,\kappa)+b(\theta_0,\kappa)A}{1-c(\kappa)A}, \quad (12)$$

where

$$a(\theta_0,\kappa)=t(\tau_t,\mu_0)e^{\tau_t/\mu_0}, \quad (13)$$

$$b(\theta_0,\kappa)=\mu_0\bar{s}(\tau_t), \quad (14)$$

$$c(\kappa)=\bar{s}(\tau_t). \quad (15)$$

Since $c(\kappa)A\ll 1$ for Mie optical depths at least as large as $\tau_M=0.30$, a point at which $c(\kappa)=\bar{s}(\tau_t)\lesssim 0.14$, the observations of Φ may be fitted to a straight line of the form

$$\Phi(\theta_0,\kappa,A)=a(\theta_0,\kappa)+b(\theta_0,\kappa)A \quad (16)$$

in order to determine the index of absorption κ and ground albedo A.

If one assumes that each measurement is made with

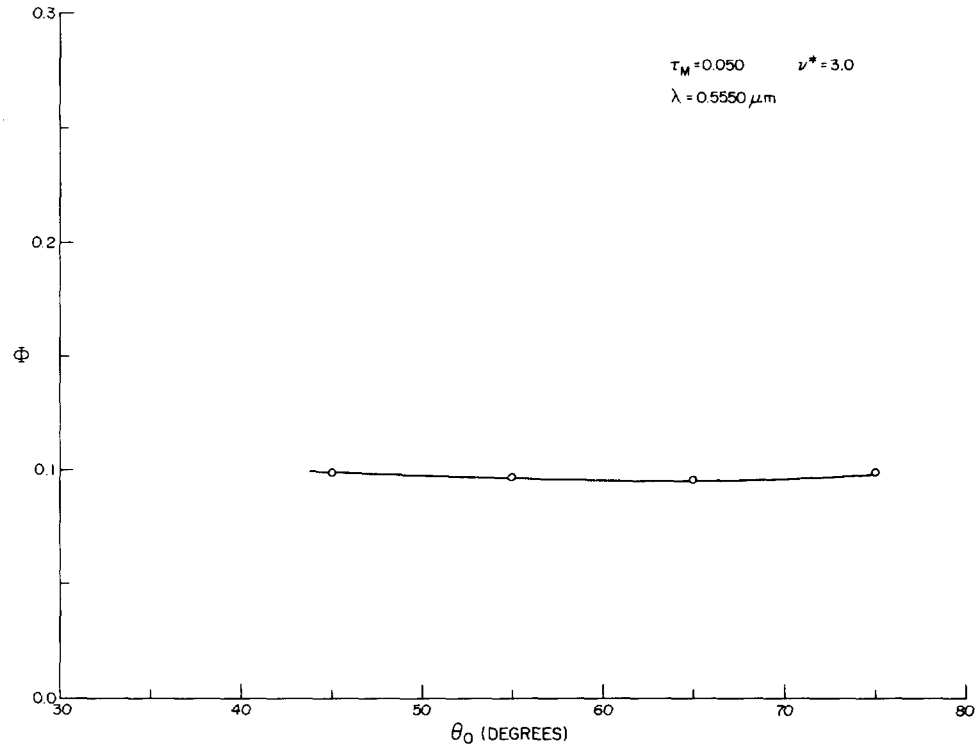

FIG. 6. The diffuse-direct ratio versus solar zenith angle for a model atmosphere having $\tau_M=0.05$ and $\nu^*=3.0$.

the same precision, Bevington (1969) shows that maximizing the probability that Φ_i observations have the functional form $\Phi(\theta_i,\kappa,A)$ is equivalent to minimizing the statistic χ^2 defined as

$$\chi^2=\sum_i [\Phi_i-\Phi(\theta_i,\kappa,A)]^2$$
$$=\sum_i [\Phi_i-a(\theta_i,\kappa)-b(\theta_i,\kappa)A]^2, \qquad (17)$$

where the summation extends over all solar zenith angles θ_i for which measurements have been made and calculations performed.

Minimizing χ^2 as defined by (17) is equivalent to making an unweighted least-squares fit to the data. The minimum value of χ^2 can be determined by setting the partial derivative of χ^2 with respect to each of the coefficients (κ,A) equal to zero. This procedure results in two simultaneous equations

$$\frac{\partial\chi^2}{\partial A}=-2\sum_i \epsilon_i b(\theta_i,\kappa)=0, \qquad (18)$$

$$\frac{\partial\chi^2}{\partial\kappa}=-2\sum_i \epsilon_i\left[\frac{\partial a(\theta_i,\kappa)}{\partial\kappa}+A\frac{\partial b(\theta_i,\kappa)}{\partial\kappa}\right]=0, \qquad (19)$$

where

$$\epsilon_i=\Phi_i-a(\theta_i,\kappa)-b(\theta_i,\kappa)A. \qquad (20)$$

Due to the complicated dependence of the functions a and b on κ, the set of Eqs. (18) and (19) is nonlinear in the unknowns κ and A. As a consequence, no analytic solution for the coefficients exists.

By combining (20) with (18), it immediately follows that

$$A=\sum_i b(\theta_i,\kappa)[\Phi_i-a(\theta_i,\kappa)]/\sum_i b^2(\theta_i,\kappa). \qquad (21)$$

For any arbitrary value of κ, however, χ^2 will not necessarily be a minimum since $\partial\chi^2/\partial\kappa$ will not be identically zero as required by (19). As κ is varied, the functions $a(\theta_i,\kappa)$ and $b(\theta_i,\kappa)$ are determined from the theoretical set of computations, from which A and χ^2 can be computed using (21) and (17), respectively. The coefficient κ is continuously varied until a minimum value of χ^2 is determined, always with the knowledge that the value of A computed using (21) is that value of A giving the minimum value of χ^2 for a given κ.

In order to see how this procedure works, consider the data set of Fig. 6 which pertains to a model atmosphere having a Junge distribution of the aerosol particles with $\nu^*=3.0$. All "measurements" of the diffuse-direct ratio are for $\theta_0=45°(10°)75°$ and $\lambda=0.5550$ μm, a wavelength for which $\tau_M=0.050$ and $\tau_R=0.086$ in this example. Since the theoretical $\Phi(\theta_0,\kappa,A)$ function can be well approximated by a linear function of A, radiative transfer computations are required for only two ground albedos at any given index of absorption and solar zenith angle. From these two computations, the coefficients $a(\theta_0,\kappa)$ and $b(\theta_0,\kappa)$ may readily be determined through application of (16). Due to the excessive computer time required to generate a set of calculations $\Phi(\theta_0,\kappa,A)$ for any given value of κ and θ_0, it is desirable to be able to interpolate in κ for fixed values of θ_0 and A and in θ_0 for fixed values of κ and A.

Fig. 2. illustrates a data set of $\Phi(\theta_0,\kappa,A)$ for values of $\theta_0=35°(10°)75°$, $\tau_M=0.10$ and $\nu^*=2.0$. Although curves for four values of A are illustrated, it is now obvious that this is redundant since computations at two values of A are sufficient to predict all other values. It is also apparent upon examination of this figure that $\Phi(\theta_0,\kappa,A)$ is a smooth, monotonically decreasing function of κ for fixed values of θ_0 and A, a feature readily lending itself to interpolation. For this purpose a spline under tension interpolation has been adopted [see Cline (1974) for details] and thus computations of $\Phi(\theta_0,\kappa,A)$ are required for a finite number of κ values only (usually four). It is similarly necessary to interpolate a set of calculations $\Phi(\theta_0,\kappa,A)$ in θ_0 because measurements are normally made at solar zenith angles θ_i other than those for which calculations are performed. Since $\Phi(\theta_0,\kappa,A)$ is a smooth function of θ_0 for fixed values of κ and A, as illustrated in Fig. 3, spline under tension interpolation has been used where computations are made for a finite number of θ_0 values.

After computing the diffuse-direct ratio at four values of κ, two values of A, and every 10° in θ_0 for a range of zenith angles surrounding the measurements (at least four values of θ_0), it is reasonably simple to compute χ^2 at any value of κ and A, making use of spline under tension interpolation in κ and θ_0 as well as (17). Fig. 7 shows such a χ^2 hypersurface in coefficient space for the theoretically generated data of Fig. 6. The solid curves indicate the points of constant χ^2. The parameters κ and A which give the best fit of the measurements to the nonlinear function $\Phi(\theta_0,\kappa,A)$ are determined by the location of the minimum value of χ^2 in this two-dimensional space. Searching this hypersurface for the parameters which minimize χ^2 is greatly facilitated through the use of (21) since this expression gives analytically the value of A which minimizes χ^2 at any value of κ. The albedo values given by (21) are shown in Fig. 7 as a dashed line which must necessarily pass through the absolute minimum of the function χ^2.

By varying κ and following the magnitude of χ^2 along the path of lowest value of χ^2, the parameter κ may be found. The variation of χ^2 as a function of κ along the ravine (dashed line) of Fig. 7 is illustrated in Fig. 8. The value of κ at which χ^2 attains a minimum is found to be 0.0100, an index of absorption for which the corresponding value of A is 0.200 in this example. It may readily be seen upon examination of Fig. 7 that these values for the parameters κ and A correspond to the absolute minimum of the χ^2 hypersurface.

In any real experiment the absolute minimum value of χ^2 will not be identically zero as in this example due to various sources of error. After having determined the

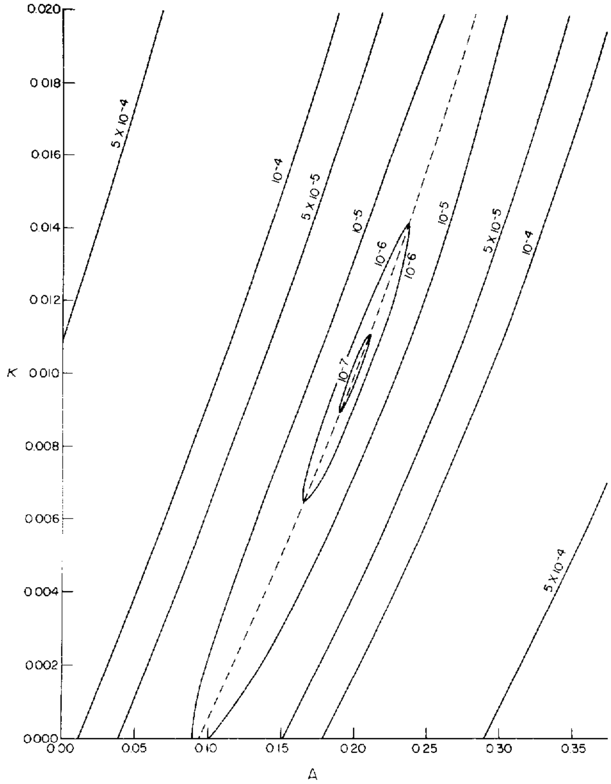

FIG. 7. χ^2 hypersurface for theoretically generated data of Fig. 6.

optimum values for the coefficients κ and A, a best fit set of $\Phi(\theta_i,\kappa,A)$ values can be computed using (16). The solid curve in Fig. 6 is the best fit to the data where $\kappa=0.0100$ and $A=0.200$. Although the effect of random errors in the Φ_i measurements was not investigated using theoretical computations, the procedure described above has been applied to measurements obtained at The University of Arizona during May and June 1977. The results of this experiment and a discussion of the various sources and magnitudes of error are presented by King (1979).

Computations similar to those used in the preceding example have been performed by assuming that the real part of the complex refractive index of the aerosol particles is 1.45, rather than 1.54. In this way, the theoretical set of computations $\Phi(\theta_0,\kappa,A)$ was altered while the "measurements" Φ_i remained the same. The results of the inversion algorithm for this situation indicate that χ^2 attains a minimum value when $\kappa=0.0099$ and $A=0.203$. This strongly supports the conclusion made previously that the diffuse-direct technique is sensitive primarily to the index of absorption and ground albedo, having substantially reduced sensitivity to the real part of the refractive index of atmospheric particulates.

Having determined the minimum value of χ^2 and the optimum values of the regression coefficients κ and A, Bevington (1969) shows that the uncertainties in

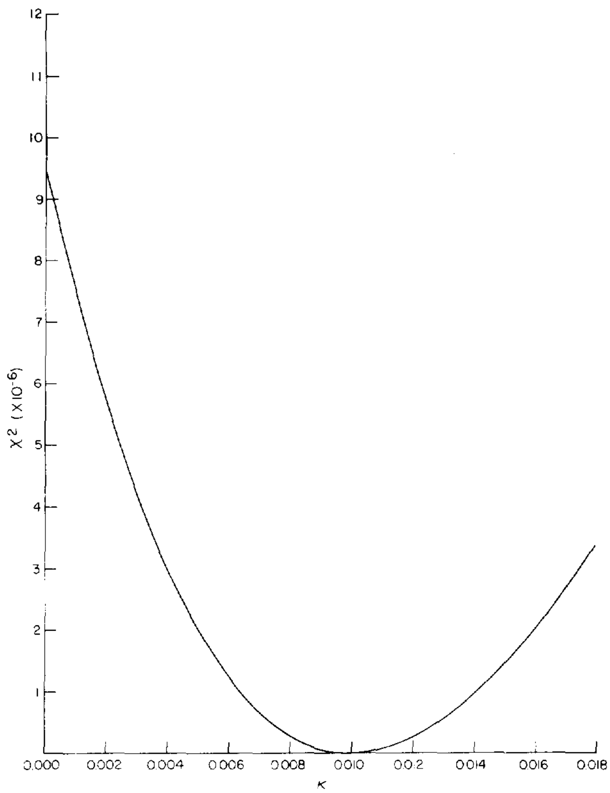

FIG. 8. Variation of χ^2 with κ along the gradient search path of Fig. 7 showing the pronounced minimum which occurs at $\kappa=0.01$.

these coefficients are related to the curvature matrix $\boldsymbol{\alpha}$, whose elements are given by

$$\alpha_{jk}=-\frac{1}{2}\frac{\partial^2\chi^2}{\partial\xi_j\partial\xi_k}, \tag{22}$$

where ξ_j represents the coefficients κ and A. The covariances $\sigma^2_{\xi_j\xi_k}$ are then obtained from the (j,k) elements of the $\boldsymbol{\alpha}^{-1}$ matrix through the expression

$$\sigma^2_{\xi_j\xi_k}\approx s^2[\boldsymbol{\alpha}^{-1}]_{jk}, \tag{23}$$

where s^2 is the sample variance for the fit, given by $s^2=\chi^2/\nu$, and $\nu=N-2$ is the number of degrees of freedom after fitting N data points with 2 parameters. The diagonal elements of (23) thus represent the variances $\sigma_A{}^2$ and $\sigma_\kappa{}^2$. The elements of the curvature matrix and the method of computation for this problem are discussed in detail by King (1977).

The procedure described above for finding the coefficients κ and A which minimize the statistic χ^2 most nearly parallels the gradient search method from nonlinear least-squares theory. By incorporating the analytical expression for A given by (21), both parameters κ and A are incremented simultaneously such that the resultant direction of travel in parameter space is along the gradient of χ^2. This method of solving nonlinear least-squares problems which are nonlinear in only one coefficient has been developed by King and Byrne (1976) for obtaining the total atmospheric ozone.

5. Summary

A statistical technique has been developed for inferring optimum values of the ground albedo and imaginary index of refraction of atmospheric aerosol particles from surface measurements of the downward hemispheric diffuse and total flux densities. This method is based on a careful consideration of the sensitivities of the diffuse radiation field to the many radiative transfer parameters and represents an optimized extension of the diffuse-direct technique suggested by Herman *et al.* (1975). The statistical algorithm presented in this paper makes use of the laws of diffuse reflection and transmission for the planetary problem, i.e., the situation of an atmosphere resting on solid ground idealized as a surface which reflects light according to Lambert's law. As a consequence of this formulation, the ground albedo inferred by the diffuse-direct technique represents a weighted average of the albedo over the entire area which affects the transfer of radiation. A simple technique is given for estimating the standard deviations of the solution parameters κ (index of absorption) and A (surface albedo).

As illustrated by the family of curves in Figs. 2 and 3, the ratio of the diffuse to direct solar flux densities decreases with increasing absorption (κ), increases with surface reflectivity (A) and shows very little change with solar zenith angle (θ_0). Fig. 3 clearly indicates, however, that the shape of the diffuse-direct ratio as a function of solar zenith angle is the primary factor which determines the ground albedo while the magnitude determines the index of absorption. As a consequence of this sensitivity to surface albedo it is clearly desirable to obtain data on days during the spring and summer months in order to allow a large span of solar zenith angles to be examined. In Part II of this series (King, 1979) the results of such an experiment will be described.

A consideration of the sensitivities of the diffuse radiation field has led to the conclusion that several radiative transfer parameters such as the real part of the particle refractive index, total ozone content, vertical distribution of the atmospheric particulates and the law of reflection at the ground have very little effect on the diffuse-direct ratio at the ground. Koepke and Kriebel (1978) recently examined the effects of measured bidirectional reflectance characteristics of four vegetated surfaces on the radiation scattered from a realistic atmosphere containing molecules and particles. They concluded that the anisotropy of the reflection properties has negligible influence on the transmitted intensity field.

The principal assumptions upon which the technique is based are that the angular distribution of energy scattered by a polydispersion of randomly oriented particles can be adequately described by Mie theory (applicable to spheres) and that the coefficients κ and A which best describe the radiation field at the earth's surface can be determined by minimizing the statistic χ^2. Although the suspended atmospheric particulates are somewhat irregular in shape, it is the purpose here to find a consistent set of parameters which are able to predict accurately the scattered radiation field in the earth's atmosphere. Since the assumption of sphericity is not totally correct, the index of absorption and ground albedo inferred by the diffuse-direct technique may not be representative of the optical properties of the individual particles or the reflectivity of the earth's surface. They nevertheless represent a consistent set of parameters which are able to predict accurately the scattered radiation field in the earth's atmosphere. Minimization of the statistic χ^2 is a result of the principle of maximum likelihood and forms the basis of least-squares curve fitting.

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REFERENCES

Ackerman, T., and M. B. Baker, 1977: Shortwave radiative effects of unactivated aerosol particles in clouds. *J. Appl. Meteor.*, **16**, 63–69.

Bevington, P. R., 1969: *Data Reduction and Error Analysis for the Physical Sciences*. McGraw-Hill, 336 pp.

Bryson, R. A., 1968: All other factors being constant...: A reconciliation of several theories of climate change. *Weatherwise*, **21**, 56–61.

Chandrasekhar, S., 1960: *Radiative Transfer*. Dover, 393 pp.

Cline, A. K., 1974: Scalar- and planar-valued curve fitting using splines under tension. *Comm. Assoc. Comput. Mach.*, **17**, 218–220.

Eiden, R., 1966: The elliptical polarization of light scattered by a volume of atmospheric air. *Appl. Opt.*, **5**, 569-575.

Elterman, L., 1968: UV, visible, and IR attenuation for altitudes to 50 km, 1968. Rep. AFCRL-68-0153, Environ. Res. Pap. No. 285, Air Force Cambridge Research Laboratories, 49 pp.

Gebbie, H. A., W. R. Harding, C. Hilsum, A. W. Pryce and V. Roberts, 1951: Atmospheric transmission in the 1 to 14 μ region. *Proc. Roy. Soc. London*, **A206**, 87–107.

Hänel, G., 1968: The real part of the mean complex refractive index and the mean density of samples of atmospheric aerosol particles. *Tellus*, **20**, 371–379.

Herman, B. M., and S. R. Browning, 1965: A numerical solution to the equation of radiative transfer. *J. Atmos. Sci.*, **22**, 559–566.

——, and ——, 1975: The effect of aerosols on the earth-atmosphere albedo. *J. Atmos. Sci.*, **32**, 1430-1445.

——, —— and R. J. Curran, 1971: The effect of atmospheric aerosols on scattered sunlight. *J. Atmos. Sci.*, **28**, 419–428.

——, —— and J. J. DeLuisi, 1975: Determination of the effective imaginary term of the complex refractive index of atmospheric dust by remote sensing: The diffuse-direct radiation method. *J. Atmos. Sci.*, **32**, 918–925.

Junge, C. E., 1955: The size distribution and aging of natural aerosols as determined from electrical and optical data on the atmosphere. *J. Meteor.*, **12**, 13–25.

King, M. D., 1977: Determination of the complex refractive index of atmospheric aerosols by the diffuse-direct technique: A statistical procedure. Ph.D. dissertation, University of Arizona, 147 pp.

——, 1979: Determination of the ground albedo and the index of absorption of atmospheric particulates by remote sensing. Part II: Application. *J. Atmos. Sci.*, **36** (in press).

——, and D. M. Byrne, 1976: A method for inferring the total ozone content from the spectral variation of total optical depth obtained with a solar radiometer. *J. Atmos. Sci.*, **33**, 2242–2251.

——, ——, B. M. Herman and J. A. Reagan, 1978: Aerosol size distributions obtained by inversion of spectral optical depth measurements. *J. Atmos. Sci.*, **35**, 2153–2167.

Koepke, P., and K. T. Kriebel, 1978: Influence of measured reflection properties of vegetated surfaces on atmospheric radiance and its polarization. *Appl. Opt.*, **17**, 260–264.

Rasool, S. I., and S. H. Schneider, 1971: Atmospheric carbon dioxide and aerosols: Effects of large increases on global climate. *Science*, **173**, 138–141.

Spinhirne, J. D., 1977: Monitoring of tropospheric aerosol optical properties by laser radar. Ph.D. dissertation, University of Arizona, 176 pp.

Twomey, S., 1974: Pollution and the planetary albedo. *Atmos. Environ.*, **8**, 1251–1256.

——, 1977: The influence of pollution on the shortwave albedo of clouds. *J. Atmos. Sci.*, **34**, 1149-1152.

van de Hulst, H. C., 1957: *Light Scattering by Small Particles*. Wiley, 470 pp.

Wang, W. C., and G. A. Domoto, 1974: The radiative effect of aerosols in the earth's atmosphere. *J. Appl. Meteor.*, **13**, 521–534.

Yamamoto, G., and M. Tanaka, 1972: Increase of global albedo due to air pollution. *J. Atmos. Sci.*, **29**, 1405-1412.