Reprinted from JOURNAL OF THE ATMOSPHERIC SCIENCES, Vol. 36, No. 6, June 1979
American Meteorological Society
Printed in U. S. A.

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

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(Manuscript received 29 August 1978, in final form 12 February 1979)

ABSTRACT

A hemispherical radiometer has been used to obtain spectrally narrow-band measurements of the downward hemispheric diffuse and total (global) flux densities at varying solar zenith angles on 14 days over Tucson. Data are presented which illustrate the effects of temporally varying atmospheric conditions as well as clear stable conditions on the ratio of the diffuse to direct solar radiation at the earth's surface. The ground albedo and the effective imaginary term of the complex refractive index of atmospheric particulates are derived from the diffuse-direct ratio measurements on seven clear stable days at two wavelengths using the statistical procedure described by King and Herman (1979). Results indicate that the downwelling diffuse radiation field in the mid-visible region in Tucson can be adequately described by Mie scattering theory if the ground albedo is 0.279±0.100 and the index of absorption is 0.0306±0.0082.

1. Introduction

A hemispherical radiometer designed by Huttenhow (1976) has been used in order to obtain spectrally narrow-band measurements of the downward hemispheric diffuse and total (global) flux densities at Tucson, Arizona. By combining the global (diffuse plus direct) flux density with the diffuse flux density, the diffuse-direct ratio can be computed. Data have been collected for 14 days between 5 May and 16 June 1977 at four wavelengths spaced throughout the visible region. The diffuse-direct ratio at each wavelength was measured during the course of each day, resulting in data for a range of solar zenith angles. Measurements have been collected for not only clear and stable atmospheric conditions but also for days during which the atmosphere was unstable and cloudy.

In Part I of this series (King and Herman, 1979) a statistical method was presented whereby measurements of the diffuse-direct ratio as a function of solar zenith angle can be analyzed to assess the magnitude of both the ground albedo and the index of absorption (imaginary part of the complex refractive index) of atmospheric particulates. In this paper a description is given of the hemispherical radiometer used to make the measurements. Data are then presented of the diffuse-direct ratio as a function of solar zenith angle and wavelength in order to illustrate some of the measurement sensitivities observed. Finally, data for days during which the atmosphere was clear and stable are analyzed and the optimum values of the ground albedo and the index of absorption of atmospheric particulates are presented.

2. Description of hemispherical radiometer

The basic requirement of the radiometer is to provide accurate narrow-band measurements of the hemispheric flux densities, both diffuse and global, received at the earth's surface. Fig. 1 is a schematic illustration of the hemispherical radiometer used in the present investigation. A plastic diffuser material has been selected for the first optical element since it is capable of producing a diffuse radiation field below the element independent of viewing direction. After passing through the diffuser element, the radiant energy within certain selected wavelength intervals is isolated by transmission through narrow-band interference filters. The distance between the diffuser element and interference filter is dictated by the maximum permissible angle of incidence of radiation on the filter ($\sim 7°$). After passing through the filter, the radiant energy is detected by a photodiode which produces an output current proportional to

$$F^{-}_{\mathrm{meas}}(\tau_t)=\int_0^{2\pi}\int_0^{\pi/2} I(\tau_t,\,-\theta,\,\phi)f(\theta)\,\sin\theta d\theta d\phi, \quad (1)$$

where $I(\tau_t,\,-\theta,\,\phi)$ is the intensity of light propagating in the downward direction at the level τ_t, a function

[1] The research reported in this article was supported by the National Science Foundation under Grant ATM75-15551-A01 and the Office of Naval Research under Grant N00014-75-C-0370.

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0022-4928/79/061072-12$07.00

of zenith angle θ and azimuth angle ϕ; τ_t is the total optical depth of the atmosphere; and $f(\theta)$ is the optical response function of the sensor. In order for $F^-_{\mathrm{meas}}(\tau_t)$ to agree well with the downward hemispheric diffuse flux density $F^-(\tau_t)$, it is necessary for $f(\theta)$ to equal $\cos\theta$.

Since there is a dielectric boundary between the atmosphere and the diffuser, some reflection occurs at the surface due to a discontinuity in the refractive index across the boundary. This dielectric discontinuity results in Fresnel reflection at the surface of the diffuser element which varies with polar angle θ. Any physical diffuser element will have an optical response function other than $\cos\theta$ as a direct result of reflection at the surface. It is therefore necessary to design a corrector plate to surround the diffuser element such that the combination of diffuser plus corrector will cause $f(\theta)$ to be nearly equal to $\cos\theta$. Since reflection can be thought of as reducing the effective area of the sensor element, it is necessary for the effective area of the sensor normal to the given beam of radiation to vary as a function of θ in such a way as to compensate for Fresnel reflection losses.

Huttenhow (1976) investigated several designs for the geometrical shape of the diffuser element and corrector plate. He concluded that measurement errors less than 2% could be attained by using a cylindrical diffuser element surrounded by a stepped corrector plate with only two steps, as illustrated in Fig. 1. It is possible to empirically determine the radial positions and heights where baffles can be placed in order to obtain $\cos\theta$ compensation for a range of polar angles. The more steps there are in the external corrector plate the more polar angles there are where $f(\theta)=\cos\theta$.

The housing for the hemispherical radiometer was constructed out of brass with the interior painted with a flat black paint to reduce the effects of reflected light from the sides of the housing. The radiometer contained a drawer port for interchanging the interference filters. The amplifier assembly was contained within the housing of the radiometer with the output fed into an external digital voltmeter to be recorded. The radiometer was mounted on a tripod for mobility and easy leveling. The tripod assembly contained a semi-circular metal slide ring on which was mounted a small brass ball whose location was adjustable along the ring. This ball and slide ring were used for occulting the direct sun while still allowing the maximum amount of diffusely scattered light from the solar aureole to be measured. Due to this requirement, an occulting ball was selected of sufficient diameter to block only the direct sun. With the ring lowered below the level of the table, global (diffuse plus direct) flux density measurements were obtained. Since a relative measurement between the diffuse and direct

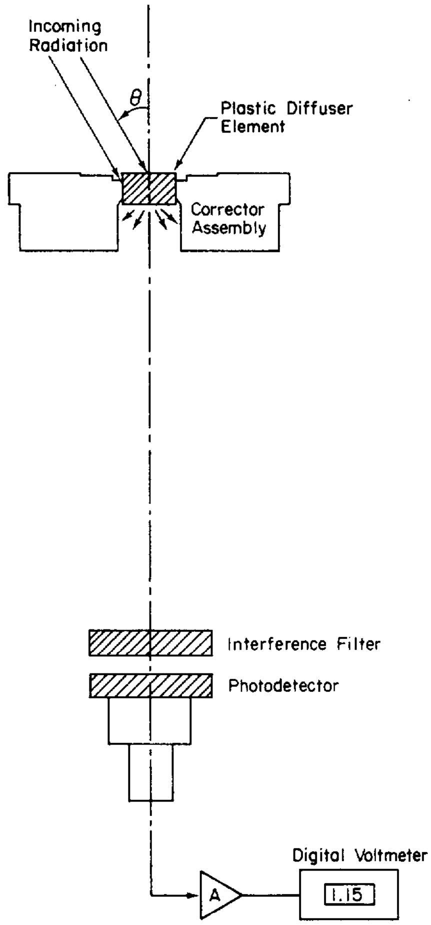

FIG. 1. Schematic illustration of the hemispherical radiometer.

flux densities was of interest, it was not necessary to calibrate the instrument in an absolute sense.

It is exceedingly critical that the top surface of the diffuser element be at the same level as the top level of the corrector assembly in order to assure that $f(\theta)$ approaches zero when $\theta=90°$. Any height difference in excess of about 0.025 mm can produce substantial errors in the response function for large polar angles.

3. Measurement sensitivity of the diffuse radiation field

In order to obtain estimates of the index of absorption of atmospheric particulates and the reflectivity of the earth's surface, measurements have been collected of the diffuse and global flux densities using the hemispherical radiometer described in the preceding section. These measurements have been made from the roof of the Civil Engineering Building (altitude 762 m MSL) on The University of Arizona campus (32°14'N, 110°57'W) because of the relatively clear and unobstructed view it affords of the entire upward hemisphere. Simultaneous to the collection of the flux density measurements, the directly trans-

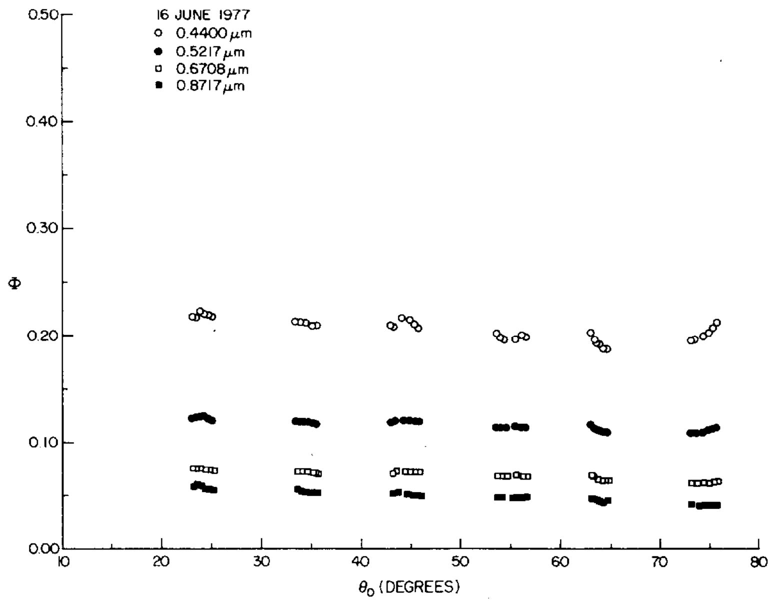

FIG. 2. Diffuse-direct ratio measurements for 16 June 1977 at four wavelengths.

mitted solar radiation was measured during the course of the day with a multi-wavelength solar radiometer described by Shaw *et al.* (1973). The diffuse-direct ratio Φ, as defined by Herman *et al.* (1975) and King and Herman (1979), is obtained from the diffuse $F^{-}_{\mathrm{meas}}(\tau_t)$ and global $G_{\mathrm{meas}}(\tau_t)$ flux density measurements once the solar zenith angle θ_0 is computed.

Under the assumption that $f(\theta_0)=\mu_0$, where μ_0 is the cosine of the solar zenith angle θ_0, it follows that the diffuse-direct ratio is given by

$$\Phi=\mu_0\frac{F^{-}_{\mathrm{meas}}(\tau_t)}{G_{\mathrm{meas}}(\tau_t)-F^{-}_{\mathrm{meas}}(\tau_t)}. \tag{2}$$

Due to the deviation of the instrumental response function from $\cos\theta$, however, it can be shown that

$$\frac{\Phi-\hat{\Phi}}{\hat{\Phi}}=\frac{\mu_0}{f(\theta_0)}\frac{F^{-}_{\mathrm{meas}}(\tau_t)}{F^{-}(\tau_t)}-1, \tag{3}$$

where $\hat{\Phi}$ and $F^{-}(\tau_t)$ are the measurements which would be obtained if $f(\theta)=\cos\theta$.

Measurements of the angular response function $f(\theta)$ have been obtained in the laboratory and used to simulate errors in the diffuse-direct ratio for a number of intensity distributions representative of six different atmospheric models and five solar zenith angles. Results of this investigation indicate that the Φ measurements are systematically higher than theory when $\theta_0 \lesssim 70°$ and systematically lower than theory when $\theta_0 \gtrsim 70°$ for all models. The maximum systematic errors ($\sim 2\%$) occur when $\theta_0 \approx 55°$, the angle where the largest error in the instrumental response function occurs. Over the range of solar zenith angles $35° \leqslant \theta_0 \leqslant 75°$, $\Delta\Phi/\hat{\Phi} \approx 0.7\%$ for a Mie (particulate) optical depth τ_M (0.5550 μm) $=0.05$. The magnitude of this systematic error varies somewhat with the atmospheric model and decreases slightly as the Mie optical depth increases.

Fig. 2 illustrates a typical data set of Φ vs θ_0 which has been obtained for Tucson at four wavelengths (0.4400, 0.5217, 0.6708 and 0.8717 μm) on 16 June 1977. For all the filters reported here the full bandwidth at half-peak transmittance was less than or equal to 0.012 μm. From the directly transmitted solar radiation measurements the Mie optical depths $\tau_M(\lambda)$ were obtained by the method described by King and Byrne (1976). This day was cloud-free and stable with Mie optical depths ranging between 0.0500 ($\lambda=0.4400$ μm) and 0.0356 ($\lambda=0.8717$ μm). Because the wavelength dependence of $\tau_M(\lambda)$ was relatively small, a log-normal type of aerosol size distribution was obtained [see King *et al.* (1978) for details]. Examination of Fig. 2 suggests that Φ decreases slightly with θ_0 until a certain zenith angle is reached after which point Φ starts to increase. The increase of Φ at larger solar zenith angles is the most evident for the shortest wavelength, becoming less evident the longer the wavelength. The solar zenith angle dependence of the diffuse-direct ratio is in essential agreement with the theoretical computations presented by King and Herman (1979, Fig. 3), a case for which $\lambda=0.5550$ μm and $\tau_M=0.0500$.

On occasions when the occulting ball is incorrectly positioned, a diffuse flux density in excess of the proper amount is recorded. As a consequence of this error, the measured value of the diffuse-direct ratio is systematically higher than it should be [see Eq. (2)]. Although it is not always possible to identify such systematic errors with absolute certainty, the more obvious ones are evident when repeated measurements are made over a small range of solar zenith angles. Fig. 2 suggests that a few such errors did occur, mostly notably at $\lambda=0.4400$ μm and $\theta_0\approx 45°$. In these situations the appropriate index of absorption κ and ground albedo A should be determined by analyzing only the more accurate data points. After elimination of the data for the times having occulting errors, the majority of the measurements yield characteristics similar to the theoretical computations illustrated in Part I (Fig. 3).

It sometimes happens that the total optical depth of the atmosphere varies with time during the course of a single day. As pointed out by King and Herman (1979), a fluctuation in τ_M is sufficient to alter the magnitude of the diffuse-direct ratio. A particularly dramatic illustration of the effect of temporal and spatial fluctuations in the atmosphere can be seen from the diffuse-direct ratio data for 16 May 1977 (see Fig. 3). On this day measurements were collected at four wavelengths during both the morning and the afternoon. As the solar zenith angle decreased throughout the morning from an initial value of 54°, the diffuse-direct ratio decreased. Had the atmosphere been horizontally homogeneous with a fixed Mie optical depth and aerosol size distribution throughout the day, the diffuse-direct ratio would have been a function only of λ and θ_0. Instead, the afternoon measurements did not repeat those of the morning but were noticeably lower in magnitude for a fixed solar zenith angle and wavelength. Although this day was cloud-free, it was visibly very turbid. Noticeable streaks were visible in the sky during the morning which appeared very similar to the stratospheric dust striations which occurred following the eruption of Volcán de Fuego in October 1974. By the afternoon all visible dust striations were gone but the late afternoon sky in the vicinity of the sun was very white suggesting a still quite appreciable dust content in the atmosphere. Data collected with a multi-wavelength solar radiometer on 16 May 1977 show very clearly that the total (and hence Mie) optical depth of the atmosphere was steadily decreasing throughout the day.

Garrison *et al.* (1978) report observations of the diffuse-direct ratio in the near ultraviolet for a similar situation in which the diffuse-direct ratio differed appreciably between morning and afternoon. They similarly attribute these fluctuations to variations in the optical depth of the atmosphere.

Since the Mie optical depth is a very important atmospheric parameter determining the magnitude of the diffuse-direct ratio, it is of interest to compare the magnitude of Φ for various days having different Mie optical depths. In order to compare measurements of the diffuse-direct ratio for several different days, estimates of Φ were obtained for $\lambda=0.5217$ μm and $\theta_0=55°$ by assuming that Φ can be approximated by a linear function of θ_0 for data collected at zenith angles near 55°. The results, including statistical error bar estimates for both Φ and τ_M, are presented in Fig. 4 for seven days between 6 May and 16 June 1977. As the Mie optical depth increases, the diffuse-direct ratio generally increases as expected from the computations illustrated by King and Herman (1979, Fig. 4) for $\lambda=0.5550$ μm. The magnitude of Φ does not increase monotonically as τ_M increases, however, simply due to daily differences in the aerosol size

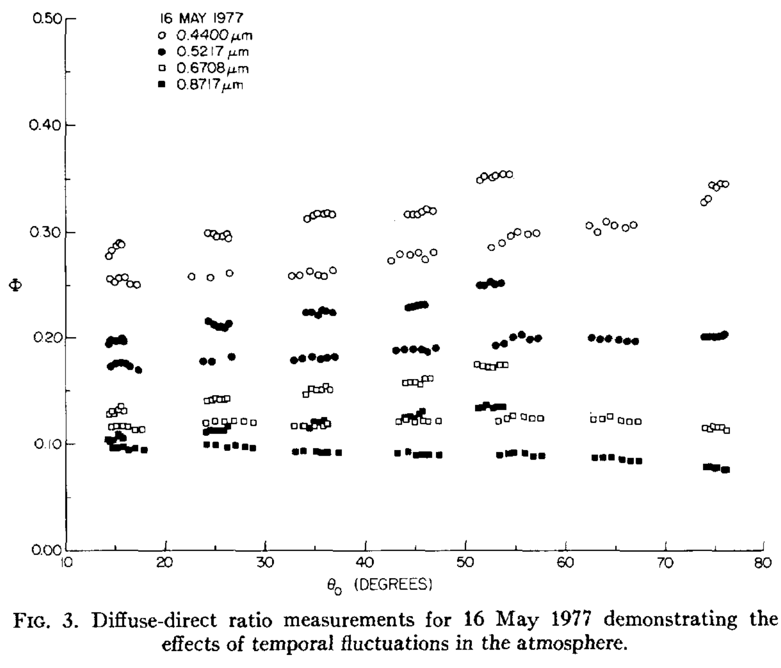

FIG. 3. Diffuse-direct ratio measurements for 16 May 1977 demonstrating the effects of temporal fluctuations in the atmosphere.

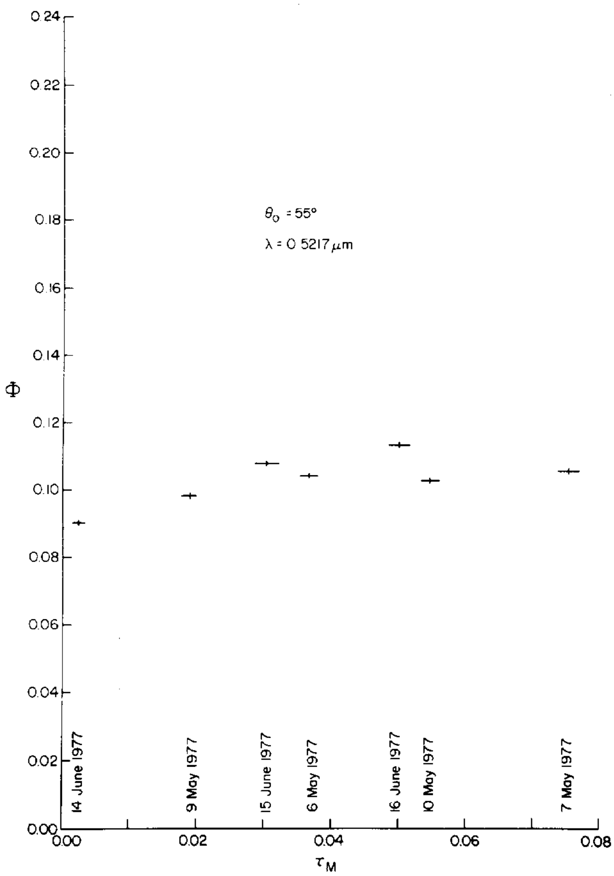

FIG. 4. The diffuse-direct ratio for seven days of varying Mie optical depths for $\lambda = 0.5217\ \mu m$ and for $\theta_0 = 55°$.

distribution. The rate of increase of Φ as a function of τ_M is less than any of the cases illustrated by King and Herman (1979). Although the wavelengths are slightly different, it will be seen in the next section that the primary reason for the smaller change of Φ with increasing τ_M for the data presented here is due to an index of absorption κ larger than 0.01.

Illustrations similar to Fig. 4 have been constructed for $\theta_0 = 55°$ at each of the other three wavelengths (0.4400, 0.6708 and 0.8717 μm). Data for the two longer wavelengths are qualitatively in agreement with the results presented in Fig. 4 with Φ tending to increase with increasing τ_M. The results for $\lambda = 0.4400\ \mu m$ tend to be less predictable due to the increased statistical and systematic errors characteristic of this wavelength.

Fig. 5 illustrates Φ as a function of λ for four days between 9 May and 16 June 1977 at a fixed solar zenith angle of 55°. The values of the diffuse-direct ratio presented here were obtained in the same manner as those of Fig. 4, by making a linear least-squares fit to Φ as a function of θ_0 for data collected around $\theta_0 = 55°$. All days for which data have been collected, including the ones presented here, have diffuse-direct ratios which monotonically decrease with wavelength and exhibit slight positive curvature.

In assessing the agreement between the measured wavelength sensitivity and theoretical expectations, it becomes necessary to perform radiative transfer calculations for the specific aerosol size distribution and optical depth applicable to a particular day and wavelength. Since computations are required only of the transmitted hemispheric flux density at the earth's surface and not the intensity field in any particular direction, considerable computational time can be saved by expressing the elements of the intensity vector and phase matrix in Fourier series in $(\phi' - \phi)$, the difference between the azimuth angles of the meridian planes containing the directions of incidence and scattering. This procedure has been adopted by Dave (1970) and by Herman and Browning (1975) using Gauss-Seidel iteration. Though the Fourier analysis procedure can readily be used to calculate the complete intensity field, one of its main advantages when particulates are present in the atmosphere is in computing the hemispheric flux densities since only the constant, azimuth-independent terms are required (Herman and Browning, 1975). Theoretical computations of the diffuse-direct ratio have been performed for three wavelengths (0.5217, 0.6708 and 0.8717 μm)

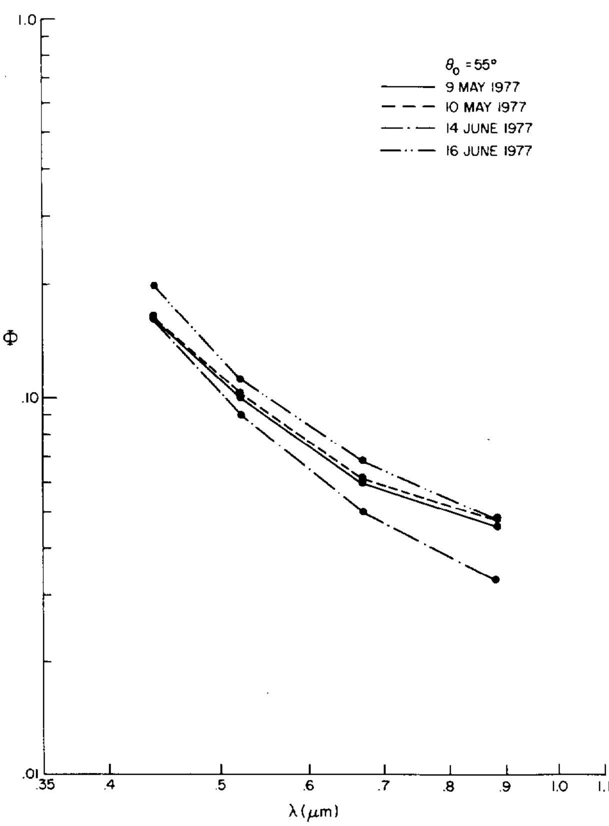

FIG. 5. The diffuse-direct ratio as a function of wavelength for $\theta_0 = 55°$ and for four different days.

on five days for which the Mie optical depths and aerosol size distributions are known. The theoretical results (assuming κ and A to be wavelength independent) are in close agreement with those of Fig. 5 in having Φ decrease monotonically in λ with slight positive curvature. Since both the theoretical and experimental sensitivities of Φ as a function of λ are in close agreement, having no anomalously high or low values at wavelengths in the ozone Chappuis absorption band (0.5217 and 0.6708 μm), it appears that the sensitivity of the diffuse-direct ratio to total ozone content is quite small as suggested by Herman *et al.* (1975) and King and Herman (1979).

4. Experimental results

The statistical method for inferring the ground albedo and the index of absorption of atmospheric particulates described by King and Herman (1979) has been applied to diffuse-direct ratio data collected on seven days during May and June 1977. After determining the spectral Mie optical depth values and columnar aerosol size distribution for a particular day, radiative transfer computations were performed for the two wavelengths 0.5217 and 0.6708 μm. The methods which have been adopted for estimating the Mie optical depth and aerosol size distribution are described by King and Byrne (1976) and King *et al.* (1978), while the method used to compute the hemispheric flux density and diffuse-direct ratio is described by Herman and Browning (1975). The real part of the complex refractive index of atmospheric particulates is assumed to be 1.45 both for the determination of the aerosol size distribution and for the application of the diffuse-direct radiation method. Fortunately, neither of these techniques is very sensitive to the real part of the particle refractive index (King *et al.*, 1978; King and Herman, 1979).

The values of the Mie optical depth, index of absorption, ground albedo and mean diffuse-direct ratio for 10 data cases are presented in Table 1. In view of the amount of scatter in the values of the ground albedo A, it does not seem justified to separate the data taken at two different wavelengths (0.5217 and 0.6708 μm) and seek a wavelength dependence. The standard deviations in the individual values of τ_M, κ and A shown in Table 1 have been derived by standard error propagation methods for each data set and thus reflect the combined effect which each individual data point has on the determination of the regression coefficients τ_M, κ and A [see King and Byrne (1976) and King and Herman (1979) for details]. The magnitude of the ground albedo presented here shows a relatively large day-to-day variability which is due, in part, to uncertainties in the aerosol size distribution and Mie optical depth. The analysis procedure which we have used has not attempted to incorporate information on the uncertainties of either the Mie optical depth or aerosol size distribution. The mean values of the imaginary index of refraction and ground albedo, weighted by the reciprocal of the variances, are found to be 0.0306 and 0.279, respectively, with corresponding standard deviations of 0.0082 and 0.100 (see Table 1). Considering the magnitude of the ground albedos and Mie optical depths obtained for these days, this amount of absorption on the part of the atmospheric particulates implies an absorption by the atmosphere on the order of 3% or less of the radiation incident at small solar zenith angles, increasing to 5% at zenith angles of 65° on 10 May 1977.

By comparing the data of Fig. 4 with the theoretical computations presented in Part I of this series (Fig. 4, applicable to $\lambda=0.5550$ μm) it is apparent that the tendency for very little increase of the diffuse-direct ratio measurements with Mie optical depth is

TABLE 1. Summary of the Mie optical depth, index of absorption, ground albedo and mean diffuse-direct ratio obtained for seven days during 1977.

Date	Wavelength (μm)	Mie optical depth	Index of absorption	Ground albedo	Mean diffuse-direct ratio
6 May	0.5217	0.0367±0.0014	0.0330±0.0048	0.325±0.004	0.1078
6 May	0.6708	0.0341±0.0014	0.0074±0.0051	0.469±0.064	0.0645
7 May	0.6708	0.0629±0.0010	0.0336±0.0033	0.289±0.013	0.0671
9 May	0.5217	0.0189±0.0012	0.0334±0.0090	0.394±0.007	0.0990
10 May	0.5217	0.0548±0.0014	0.0333±0.0031	0.123±0.006	0.1049
10 May	0.6708	0.0589±0.0012	0.0340±0.0074	0.173±0.011	0.0643
14 June	0.5217	0.0025±0.0010	0.0334±0.0091	0.370±0.012	0.0859
15 June	0.5217	0.0304±0.0019	0.0354±0.0048	0.236±0.015	0.1005
16 June	0.5217	0.0501±0.0016	0.0367±0.0037	0.199±0.011	0.1164
16 June	0.6708	0.0445±0.0013	0.0233±0.0036	0.348±0.025	0.0695
Weighted mean			0.0306	0.279	
Standard deviation			0.0082	0.100	

Note: The observed Mie optical depths and estimated aerosol size distributions for 6 May and 15 June 1977 are illustrated in King *et al.* (1978).

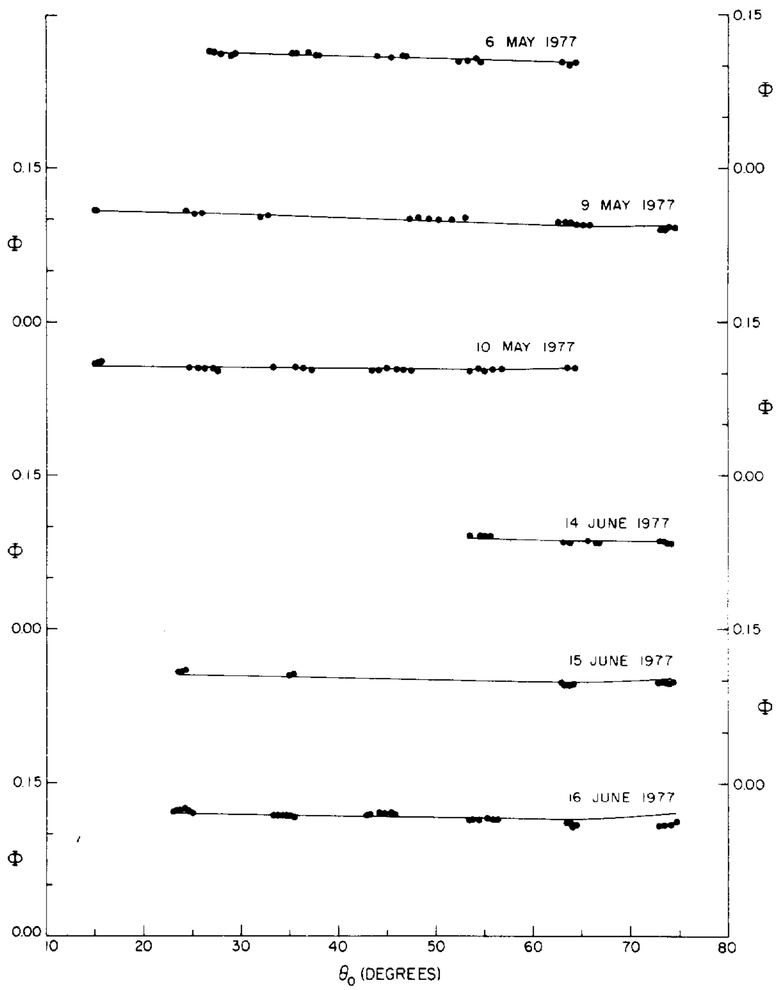

FIG. 6. Observed and computed diffuse-direct ratio versus solar zenith angle for six days at $\lambda=0.5217$ μm. The ordinate scale to which each curve refers is repeated alternately on the left and then the right side of the figure in order to separate the individual data cases.

consistent with a relatively large value of 0.0306 for the index of absorption. In addition, the theoretical computations for 6 May and 15 June require the diffuse-direct ratio to be less on 6 May for fixed values of κ and A simply due to differences in the aerosol size distributions. The reduced magnitude of 10 May over 16 June is similarly due to a difference in aerosol size distributions and is to be expected theoretically (see Fig. 4).

On any given day and wavelength it is not always possible to determine values for κ and A due to uncertainties in either the diffuse-direct ratio, the Mie optical depth or the aerosol size distribution. Temporal and spatial fluctuations in the atmosphere can also make it not feasible to analyze a particular day or wavelength, as is apparent upon examination of Fig. 3, since it is necessary to measure Φ over a range of solar zenith angles while the atmosphere remains constant. Although 7 May appeared very clear at the time of the observations, subsequent examination of the solar radiometer data indicated an appreciable time variation of optical depth in the late morning and afternoon. The hemispherical radiometer data were fortunately collected in the early morning on this day but some time variations in optical depth were nevertheless detectable at 0.5217 μm (but not so severely at 0.6708 μm). The analysis for 7 May 1977 at $\lambda=0.5217$ μm has not been included in Table 1 for this reason. It is desirable, particularly at the longer wavelengths, to have data for days exhibiting a relatively large Mie optical depth since the larger the optical depth the greater the sensitivity to A and κ (King and Herman, 1979). The days presented in Table 1 at $\lambda=0.6708$ μm were the four days which had the largest Mie optical depths at this wavelength.

Due to the normally small Mie and Rayleigh optical depths at wavelengths in the near-infrared, sensitivities to κ and A are so greatly reduced that the diffuse-direct technique is normally not a viable remote sensing method for deriving these parameters. A preliminary analysis of some of the data sets at 0.8717 μm indicates that the measurements may have been systematically higher than theory at this wavelength (but the general lack of sensitivity to κ and A for the Mie optical depths measured at 0.8717 μm make it difficult to say whether these errors are significant). The near-ultraviolet wavelength region, on the other hand, does seem attractive for applying the diffuse-direct technique since the Mie optical depths are normally larger at these wavelengths. No attempt has been made in the present investigation to analyze the measurements at 0.4400 μm since the relatively small measurement signal contributed to larger statistical and systematic errors (see Figs. 2 and 3) and because the Mie optical depths were particularly small during this period at 0.4400 μm. Since there is negligible ozone absorption at $\lambda=0.4400$ μm, it would be a good wavelength to investigate further in the future if measurements can be made with a sufficient degree of accuracy.

Figs. 6 and 7 present the diffuse-direct ratio data and results of the fitting procedure for all 10 cases of Table 1. The solid curves represent the regression fit to the data points using the optimum values of the coefficients κ and A. Most hemispherical radiometer measurements were made only in the afternoon but the data for 7 May (Fig. 7) were collected in the morning while the data for 6 and 9 May include both morning and afternoon observations. The atmosphere on 14 June was incredibly clean with Mie optical depths of about 0.01 at most wavelengths. Although it was clear throughout the day, large systematic occulting errors occurred at small solar zenith angles thus limiting the range of useful solar zenith angles on 14 June to $\theta_0 \geqslant 53.56°$. By comparing Figs. 6 and 7 it is clear that theory requires Φ to increase slightly toward large solar zenith angles at $\lambda=0.5217$ μm, while not at all at $\lambda=0.6708$ μm. Had the optical depths been much larger on these days, as on the morning of 16 May (see Fig. 3), the diffuse-direct ratio would have increased monotonically in zenith angle for all wavelengths. The sample standard deviation of the data points about the regression fit is typically 0.0028 at $\lambda=0.5217$ μm and 0.0016 at $\lambda=0.6708$ μm, representing random fluctuations on the order of 2.5% at both wavelengths. These errors are larger than those attributed solely to measurement error (~0.7% as discussed in Section 3) and thus reflect the combined effect of measurement errors, atmospheric fluctuations and the accuracy of the determination of the Mie optical depth and aerosol size distribution.

The best months in Tucson for obtaining clear skies and stable atmospheric conditions for a long enough period of time to obtain good optical depth data are May, June, October and November. Due to the small solar declination angles in the fall, however, a restricted range of solar zenith angles is available ($\theta_0 \gtrsim 40°$) thus making May and June the best time to collect diffuse-direct ratio data in Tucson. In addition to the large range of solar zenith angles which are available in the spring, there is normally a secondary peak value of Mie optical depth at this time of year (with the absolute peak usually occurring in July or August). Unfortunately, the optical depths during May and June of 1977 were smaller than seasonally expected from previous years and thus the sensitivity of the diffuse-direct ratio measurements to A and κ was not as large as anticipated.

Although the measurements reported in this investigation were obtained near the center of the city of Tucson, the mean value of the ground albedo is in close agreement with the results obtained elsewhere in the southwest. Since the surface albedo determined by the diffuse-direct radiation method represents an area averaged albedo consistent with the transfer of radiation in the earth's atmosphere, the most representative values to be compared with it are other area-averaged radiation measurements such as those

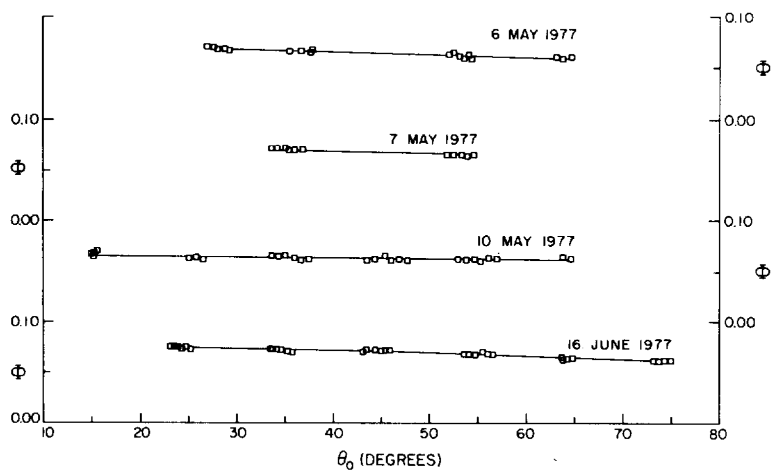

FIG. 7. Observed and computed diffuse-direct ratio versus solar zenith angle for four days at $\lambda=0.6708$ μm. The ordinate designation is the same as Fig. 6.

obtained by low flying aircraft with upward and downward pointing radiometer systems. Kung *et al.* (1964) and Griggs (1968) obtained values ranging between 0.22 and 0.25 for the surface albedo in southern Arizona using measurement systems having a very wide wavelength response. DeLuisi *et al.* (1976) found values ranging between 0.20 and 0.35 for Quartzite, Arizona, where crude wavelength resolution was obtained by attaching Schott glass cutoff filters to their aircraft mounted Eppley pyranometers.

An alternative type of surface albedo determination in the southwestern United States has been obtained by Otterman and Fraser (1976) by examining the earth-atmosphere system reflectivities obtained from the Landsat satellite. By modeling the scattering properties of the atmospheric particulates they inferred values for the surface albedo of 0.264 in the wavelength range of 0.5 to 0.6 μm with little sensitivity to the particulate model. The mean value of 0.279 found in this investigation is in close agreement with that of these investigations. The variability of the values of ground albedo presented in Table 1 is, however, quite large. In order to better establish the ground albedo and its daily and spectral variability, it is necessary to collect more observations on days for which the Mie optical depths are large since the sensitivity of the diffuse-direct ratio to ground albedo increases in direct proportion to τ_M. As pointed out earlier, the Mie optical depths during May and June 1977 were smaller than normally expected for that time of year.

Since there are very few assessments of the imaginary index of refraction of naturally suspended atmospheric aerosol particles which have been obtained by examining the scattered radiation field, the number of studies which can be compared on an equivalent basis with the present one are few. DeLuisi *et al.* (1976) examined the percent absorption by the atmosphere between two different heights using as inputs previously determined values of the Mie optical depth and surface reflectivity. They reported values of κ on the order of 0.013 for Quartzite, Arizona, but with error bar estimates extending to 0.028. Kuriyan *et al.* (1979) examined the intensity and degree of polarization of the sunlit sky as a function of azimuth angle in the Los Angeles area. They found that during days having dry continental air masses the radiation data could best be ascribed to a low Mie optical depth ($\tau_M \approx 0.05$) and large index of absorption ($0.02 \leqslant \kappa \leqslant 0.05$). They felt that the relatively large values for the index of absorption (roughly consistent with those of Table 1) were due to the low humidity of the continental air mass.

Eiden (1966) compared theoretical and measured ellipticities of light scattered by a volume of atmospheric air and obtained an index of absorption κ ranging between 0.01 and 0.1 at Mainz. He also pointed out that an increased humidity decreases the effective value of κ supporting the conclusions by Kuriyan *et al.* (1979).

Since Eiden (1966) considered only three different Junge models for the aerosol size distribution, a better estimate of the size distribution should in theory lead to a more accurate determination of the imaginary index of refraction of the aerosol particles. Grams *et al.* (1974) measured the angular variation of the intensity of light scattered from a collimated beam by the atmospheric aerosol while simultaneously collecting the particles for size distribution analysis. With an accurate determination of the aerosol size distribution thus obtained, theoretical angular scattering intensity computations were performed and compared to simultaneous measurements of the same in order to determine the index of absorption of the atmospheric particulates. Results of this procedure applied to 23 data sets obtained at Big Spring, Texas, indicate that κ ranged between 0.0011 and 0.0214 with a geometric mean value of 0.0050.

Although Bergstrom (1973) and Eiden (1971) have pointed out the problems inherent in using bulk indices of refraction of collected samples of atmospheric dust in computing the radiation field of the free atmosphere, it is nevertheless of interest to compare the values obtained in the present investigation with laboratory measurements of the bulk index of absorption. Fischer (1970) has collected samples of aerosol particles with a jet impactor and, by means of an integrating sphere to collect the scattered light, inferred the index of absorption of the particulates. Assuming that the density of the aerosol particles is approximately 2.0 g cm^{-3}, Fischer's (1970) values for the imaginary index of refraction lie in the range $0.010 \lesssim \kappa \lesssim 0.045$ in the mid-visible wavelength region. More recently Fischer (1973) examined specific particulate samples from both urban and rural locations from which he found that the index of absorption is typically 0.010 in rural locations while being more typically 0.028 in urban locations.

Lin *et al.* (1973) used opal glass instead of a sphere to integrate the light scattered by collected samples of New York City particulates. They determined that the index of absorption ranged between 0.028 and 0.050 with a mean value of 0.040. Using the Kubelka-Munk theory of diffuse reflectance Lindberg and Laude (1974) inferred values for the index of absorption of samples of New Mexico dust lying between 0.007 and 0.008.

One of the limitations of the present procedure for inferring the ground albedo and the index of absorption of atmospheric particulates is the sensitivity of the diffuse-direct ratio to particles $\lesssim 0.10$ μm in radius. Since these particles are very difficult to sense from spectral Mie optical depth measurements (King *et al.*, 1978), the effects of small particles on the values of κ and A inferred by the diffuse-direct technique were considered. This was accomplished by comparing results obtained assuming a Junge (1955) size distribution for the aerosol particles with one slope ($\nu^*=3.0$) but with two different radii ranges (0.01–5.0 μm and 0.10–5.0 μm). Results indicate that the effect of small particles is insignificant in the determination of the surface reflectivity though it may affect significantly (factor of $\sim$2.5) the value of the index of absorption inferred. The fact that the presence of small particles can have a large effect on the value of κ inferred by the diffuse-direct technique may readily be understood. Since absorbing particulates $\lesssim$0.10 μm in radius are more efficient absorbers than scatterers (i.e., $Q_{sca}\propto r^4$ and $Q_{abs}\propto r$, where Q_{sca} and Q_{abs} are the Mie efficiency factors for scattering and absorption, respectively), the neglect of these particles in computing the diffuse radiation field in the earth's atmosphere affects the theoretical values of the diffuse-direct ratio. For a fixed Mie optical depth of the atmosphere, the presence of a significant number of small absorbing particles decreases the single-scattering albedo and consequently the computed diffuse-direct ratio over those values computed if only particles larger than 0.10 μm are present. Since these computations are to be compared with measurements of Φ at a particular value of τ_M, the inclusion of a significant number of particles less than 0.10 μm in radius tends to *decrease* the inferred value of κ over the value determined if only particles larger than 0.10 μm are included. As a consequence of this effect, one should consider the values reported in the present investigation as being an upper limit of the true index of absorption. Most of the aerosol size distributions obtained for the seven days analyzed in Table 1 were log-normal in character, however, and thus showed little tendency to have very many particles smaller than 0.10 μm in radius. These types of distributions are typical of days for which the Mie optical depths are small (King *et al.*, 1978). The observed Mie optical depths and aerosol size distributions for 6 May and 15 June 1977 are illustrated by King *et al.* (1978, Figs. 4 and 5).

The effect of particulates with radii $\gtrsim$4.0 μm is of little importance since the reduced number of particles at these sizes is such that their contribution to the Mie optical depth, and hence to the diffuse radiation field, is on the order of a few tenths of one percent.

Grams *et al.* (1974) investigated the effect of including particles with radii $<$0.6 μm in computing the theoretical scattering intensities, since their primary refractive index analysis only included log-normal size distributions with particle radii $>$0.6 μm. They concluded that a Junge distribution extension to smaller particles with $\nu^*=3.0$ (i.e., $dN/d\log r\propto r^{-3}$) tended to *increase* their inferred geometric mean value of the index of absorption from 0.005 to 0.008. Although no mention was made of the lower radius limit of the Junge distribution extension, it is interesting to note that the direction of the effect of small particles is just the opposite of that of the diffuse-direct technique.

5. Conclusions

For a successful application of the diffuse-direct technique described in the present investigation, it is necessary to have a clear atmosphere devoid of any cloud cover for a long enough period of time for the solar zenith angle to undergo a large change, preferably of about 40°. It is further required that accurate normal incidence and downward hemispheric flux densities be obtained over narrow bandpass wavelength intervals. A hemispherical radiometer capable of quasi-monochromatic flux density measurements has been designed by Huttenhow (1976) and used in the present investigation. From measurements of the hemispheric diffuse and total (global) flux densities, the diffuse-direct ratio is obtained after computing the solar zenith angle at the time of the measurements.

Due to fluctuations in the magnitude of the diffuse-direct ratio associated with daily differences in Mie optical depth and aerosol size distribution (see Fig. 4), a procedure for interpreting measurements of the diffuse-direct ratio has been adopted which uses as input parameters values of the Mie optical depth and aerosol size distribution determined from alternative measurements of the directly transmitted solar radiation. In addition, the complete analysis requires an accurate radiative transfer program and a statistical optimization procedure for combining these factors in order to obtain the particulate index of absorption and ground albedo.

Although the combination of atmospheric conditions and instrumentation are difficult to satisfy in most locations, they can normally be met in Tucson during the months of April, May and June. Measurements have been collected on 14 days during May and June 1977 using the hemispherical radiometer described in Section 2. The clear and stable days have been analyzed and presented in Table 1 for two wavelengths (0.5217 and 0.6708 μm), with the diffuse-direct ratio data and corresponding regression fits being illustrated in Figs. 6 and 7. The mean value of the ground albedo presented here is in agreement with other measurements obtained both from satellite (Otterman and Fraser, 1976) and low flying aircraft (Kung *et al.*, 1964; Griggs, 1968; DeLuisi *et al.*, 1976). The relatively large variability among the individual data cases presented in Table 1 is primarily a consequence of the small Mie optical depths which occurred during May and June 1977. In order to better establish the ground albedo and its daily and spectral variability it is necessary to collect more observations on days for which the Mie optical depths are large since the sensitivity of the diffuse-direct ratio to ground albedo increases in direct proportion to τ_M (King and Herman, 1979).

The values for the imaginary part of the complex refractive index of atmospheric particulates are somewhat higher than initially anticipated but not inconsistent with the results of other investigators (Eiden, 1966; Fischer, 1970, 1973; Lin *et al.*, 1973; DeLuisi *et al.*, 1976; Kuriyan *et al.*, 1979). As a word of caution, however, it is important to note that spectral Mie optical depth measurements in the visible and near-infrared wavelength regions are relatively insensitive to the aerosol size distribution at particle radii much less than about 0.1 μm (King *et al.*, 1978). This can lead to an overestimation of the imaginary index of refraction of atmospheric particulates using the diffuse-direct technique if there are an optically significant number of small (Aitken) particles present in the atmosphere. Fortunately, the aerosol size distributions obtained for most of the days analyzed in this investigation were log-normal in character with the radius of maximum concentration lying between 0.3 and 0.9 μm. These data show little tendency to have very many particles smaller than 0.1 μm in radius. Since the sensitivities of the diffuse-direct ratio to ground albedo and index of absorption increase with increasing Mie optical depth, situations for which the concentration of small particles increases, it is necessary to obtain a good estimate for the aerosol size distribution at small radii when applying the methods described in this series of articles. On high optical depth situations in which the aerosol size distribution is nearly Junge with a large value of ν^*, it is reasonably simple to extend the minimum particulate radius for inversion purposes to 0.06 μm (King *et al.*, 1978).

Because the parameters κ and A determined by the diffuse-direct technique are those giving the best agreement between measurement and radiative transfer theory, they are of the most interest to meteorologists concerned with the impact of aerosol particles on climate. As pointed out by Yamamoto and Tanaka (1972), Wang and Domoto (1974) and Herman and Browning (1975), it is both the index of absorption and the surface reflectivity which determine whether an increase in the Mie optical depth will lead to a warming or a cooling of the earth's atmosphere. Assuming that the mean values of the surface albedo and index of absorption obtained at the 0.5217 and 0.6708 μm wavelengths apply to 0.5550 μm (the wavelength for which the most extensive set of radiative transfer computations have been performed), it is readily found that the effect of particulates over Tucson is one of heating the earth's atmosphere. This is primarily a result of the relatively large surface reflectivity of the southwestern desert region since a value for the index of absorption on the order of 0.010 is sufficient to produce a net warming over a wide range of aerosol size distributions.

Acknowledgments. This study represents part of the author's Ph.D. dissertation at the University of Arizona, prepared under the direction of Professor Benjamin Herman, to whom he would like to express his deepest gratitude for the guidance and encouragement offered during his entire graduate career. The author would further like to thank Professor S. Twomey for several helpful comments during various stages of this research and Capt. Jay D. Huttenhow for designing the hemispherical radiometer used in this investigation.

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