

Complex Index of Refraction Between 300 and 700 nm for Saharan Aerosols

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Both the imaginary part (n_{IM}) and the real part (n_{RE}) of the complex index of refraction for Saharan aerosols have been determined as a function of wavelength between 300 and 700 nm: n_{RE} was determined by means of an immersion oil technique, and n_{IM} was determined from measurements of the total diffuse reflectance of the aerosol by means of an analysis using the Kubelka-Munk theory. No significant differences in the optical properties of the aerosol were seen among the samples collected at sampling sites on both sides of the Atlantic. Comparison of our results with others based on measurements of the ratio of the direct to the diffuse radiation shows that the two independent methods give remarkably similar results.

Concern has been expressed in recent years about the possible climatic effects of aerosols [Bryson, 1968; SCEP, 1970; SMIC, 1971]. In order to assess these possible effects a number of radiation models have been developed which describe radiative energy exchange in the earth-atmosphere system and the possible effects of aerosols on this exchange [Atwater, 1970; Ensor *et al.*, 1971; Shettle and Green, 1974; Chylek and Coakley, 1974]. There have also been a number of experimental studies which have attempted to relate radiation measurements to simultaneous measurements of the aerosol properties, for example, the Complex Atmospheric Energetics Experiment (Caenex) [Kondratyev *et al.*, 1974] and the Global Atmospheric Aerosol and Radiation Study (Gaars) [De Luisi *et al.*, 1976]. According to Kondratyev *et al.* [1976] the study of the Saharan aerosol layer is of great interest from the point of view of the 'climate and aerosol' problem. Accordingly, there was also an extensive program of measurements during Gate (the Garp Atlantic Tropical Experiment) in 1974 on the radiative properties and effects of the Saharan aerosol layer.

The Saharan aerosol layer consists of crust-derived aerosols which are generated by erosion processes in northwest Africa and are then transported by the global circulation westward across the Atlantic. Mass concentrations for this aerosol are high, as are atmospheric turbidities associated with the layer. In addition, measurements over the Atlantic far from the source of the aerosols enable the study of a reasonably well mixed aerosol layer. The measurements during Gate sought to provide the information needed to parameterize the effects of the layer. These measurements included radiation measurements from aircraft above and below the layer [Kondratyev *et al.*, 1976; Ellingson *et al.*, 1975], surface measurements of direct and total solar radiation [Carlson and Caverly, 1977], aerosol phase function measurements [Grams *et al.*, 1976], and aerosol physical properties measurements [e.g., Savoie and Prospero, 1976a]. In addition, Rahn *et al.* [1976] have reported measurements of the elemental composition of Saharan aerosols during 1973. The radiation measurements have shown that the Saharan aerosol layer can have a significant effect on the radiation balance; so accurate knowledge of the parameters of the layer which affect the interaction of the aerosols with the radiation field is needed.

In general, the optical effects of the aerosols are determined by the optical properties (expressed as a complex index of refraction), the size distribution, and the shape of the aerosol

particles. The complex index of refraction $n = n_{RE} - in_{IM}$ consists of a real part n_{RE} , which is the ratio of the speed of light in a vacuum to the speed of light in the material, and an imaginary part n_{IM} , which is an absorption parameter characterizing the material.

For a bulk material the ratio of the intensity I at some point in the material to the incident intensity I_0 is given by

$$I/I_0 = e^{-kx} \quad (1)$$

This equation is known as the Bouguer-Lambert absorption law; k is the Bouguer-Lambert absorption coefficient, which is characteristic of the bulk material, and x is the path length in the material. The units for k are the inverse of those for x . For the bulk material, n_{IM} is related to the Bouguer-Lambert absorption coefficient k by means of

$$n_{IM} = k\lambda/4\pi \quad (2)$$

where λ is the wavelength of the incident light. The units for λ are the inverse of those for k .

For particles in the size range of the optically important atmospheric aerosols, $0.1 < r < 10.0 \mu\text{m}$, the interaction of an individual particle with the radiation field is generally described by Mie theory. The Mie theory, however, assumes that the particles are spherical; so the observed scattering from nonspherical aerosol particles will differ by some unknown amount from that predicted by Mie theory. A more exact solution for nonspherical particles must be obtained by some modification of Mie theory, such as that proposed by Chylek *et al.* [1976], or by a numerical method, such as that proposed by Ward [1975]. The Mie theory describes the scattering in terms of the ratio of the particle radius r to the wavelength of the incident light, usually designated by the Mie parameter $\alpha = 2\pi r/\lambda$, and the complex index of refraction. For widely separated aerosol particles in an optically thin layer the total scattering is the sum of the individual scattering; for optically thick layers, multiple scattering must be considered. The final description of the atmospheric aerosol layer as it affects the radiation field involves the absorption cross section σ_A and the scattering cross section σ_S ; σ_S is determined by the scattering phase function, in which the scattering is given as a function of scattering angle for the incident light. The total extinction σ_E is just the sum of σ_S and σ_A . The parameters that are significant in radiation models may be determined from a knowledge of σ_A and the phase function, which can be calculated if the optical properties are known.

The optical properties of the aerosol may be determined in a number of ways. Inversion techniques use data on the scattered radiation to infer optical properties. For example, if the size distribution and real refractive index are known, the ratio of the direct to the diffuse illumination from the sun may be used to infer an imaginary index of refraction by a technique described by *Herman et al.* [1975], or the scattering phase function may be used to infer a value for n_{IM} [*Grams et al.*, 1974]. Alternatively, the aerosol particles may be collected and analyzed by a laboratory method of measuring aerosol absorption from which n_{IM} may be determined. Such measurements have been reported by *Fischer* [1970, 1973], *Lin et al.* [1973], and *Lindberg and Laude* [1974]. Generally, n_{RE} is measured in the laboratory by use of an immersion oil technique.

The advantage of the laboratory measurements is that they avoid the questions associated with inversion of the scattered radiation from nonspherical particles by Mie theory. The potential disadvantage of the laboratory techniques is that the processes of collecting and preparing the sample for measurement may change the sample, thus affecting the measured optical properties. For crust-derived mineral aerosols, such as Saharan aerosols, we would not expect the samples to be affected by the collection and preparation procedure.

We have made a series of measurements of the complex index of refraction for Saharan aerosols collected at island stations on both sides of the Atlantic Ocean during the Gate program as well as for one sample collected on board a ship during November 1973. The purposes of these measurements have been threefold: (1) to determine values for the complex index of refraction as a function of wavelength for these aerosols from the various sites, (2) to determine whether there is any systematic variation in the measured optical properties that may be related to the transport of the aerosols across the Atlantic, and (3) to compare our laboratory measurements with measurements obtained on the basis of the radiation data in order to obtain some estimate of the effect of nonsphericity on the measurements. We have measured both n_{RE} and n_{IM} in the laboratory. We have measured n_{RE} by a standard immersion oil technique, the Becke line technique as described by *Wahlstrom* [1974]; we have measured n_{IM} by an integrating sphere technique by means of an analysis using the Kubelka-Munk theory. We chose this method of measuring n_{IM} because the analysis is not dependent on the shape of the particles, being completely independent of the usual Mie theory analysis, and because it is possible to determine a value for the imaginary index of refraction from the laboratory measurements in a straightforward manner by using the Kubelka-Munk theory.

KUBELKA-MUNK THEORY

The Kubelka-Munk (K-M) theory is a phenomenological theory that relates the total diffuse reflection from a material to the effective scattering and absorbing properties of the material. As was discussed by *Kortum* [1969], the K-M theory is a two-flux theory in which the radiation is assumed to be composed of two oppositely directed radiation fluxes through a continuous medium. This material is assumed to have an absorption coefficient per unit distance, k , and a scattering coefficient per unit distance, s , the term scattering referring only to radiation redirected into the backward hemisphere. The K-M theory assumes diffuse radiation onto the surface of the material as well as diffuse radiation within the material, which results in an increased effective path length within the material. The scattering and absorption coefficients are gener-

ally redefined as $S = 2s$ and $K = 2k$ to take into account this increased path length. *Gate* [1974] has shown that the K-M constants S and K are related to the usual scattering and absorption cross sections σ_s and σ_A and are defined for radiation transfer calculations by

$$K \approx 2\sigma_A \quad (3)$$

and

$$S = \frac{3}{2}\sigma_s \quad (4)$$

which are valid for isotropically scattering, weakly absorbing materials ($K/S < 0.01$). These relations have been verified experimentally [*Gate*, 1971; *Brinkworth*, 1971] and have been found to be valid within approximately 5%.

Since the definitions $K = 2k$ and $S = 2s$ have been made, these results imply that the quantity k is the same as the usual Bouguer-Lambert absorption coefficient, although $s = \frac{3}{2}\sigma_s$. The difference in the scattering coefficients arises because s is defined as scattering into the backward hemisphere. Since k is the usual Bouguer-Lambert absorption coefficient appropriate to a bulk material, the values for n_{IM} may be determined from k by using (2). Because of the correspondence between k and σ_A , our discussion of the K-M theory will be in terms of the constants s and k rather than the more common S and K .

According to K-M theory R_∞ , the total diffuse reflectance from a semi-infinite thickness of material, will depend only on the ratio k/s by means of

$$k/s = F(R_\infty) = (1 - R_\infty)^2/2R_\infty \quad (5)$$

$F(R)$ for any reflectance is called the Kubelka-Munk function for R . *Kortum* has shown that the K-M scattering coefficient S may be determined from measurements of the reflectance of a thin layer of material against a dark background if the layer thickness d and R_∞ for the material are known. The equation that describes this relation, written in terms of s , is

$$s = \frac{1}{2d} \frac{R_\infty}{(1 - R_\infty^2)} \ln \frac{(R_\infty R - 1)(R_g - R_\infty)}{(R_\infty R_g - 1)(R - R_\infty)} \quad (6)$$

where R is the total diffuse reflectance of the shallow layer of material and R_g is the reflectance of the background; all measurements are expressed in terms of the absolute values of the reflectances. The absorption coefficient k may then be determined from a measurement of R_∞ if the scattering coefficient has been measured.

Kortum has shown that there are deviations from K-M theory under conditions of high absorption but that the K-M theory is applicable when the highly absorbing material whose absorption is to be measured is diluted with an excess of a white standard. These are the same conditions under which the relationships discussed by *Gate* are valid. Under these conditions also, directed incident radiation may be used in place of the diffuse incident radiation, and the applicability of the K-M theory is maintained. In practice, the substance whose absorption is to be measured is diluted with a large excess of the diluent, so that the scattering of the mixture is just the scattering of the diluent.

When the dilution method is used, the total K-M function calculated from the observed absolute total diffuse reflectance of the mixture of sample and standard, $F_{st+sa}(R_\infty)$, is the sum of two individual K-M functions:

$$F_{st+sa}(R_\infty) = F_{st}(R_\infty) + F_{sa}(R_\infty) \quad (7)$$

Here the subscript st refers to the standard, and sa refers to the sample. F_{st} is just the K-M function for the pure standard, and

so F_{sa} may be determined by subtracting the K-M function for the mixture from the K-M function for the pure standard:

$$F_{sa}(R_{\infty}) = F_{st+sa}(R_{\infty}) - F_{st}(R_{\infty}) \quad (8)$$

which may be written as

$$k_{sa}/s_{st} = (k_{sa+st} - k_{st})/s_{st} \quad (9)$$

since the scattering of the mixture is assumed to be the scattering of the diluent or standard. The absorption of the sample, k_{sa} , may then be determined by multiplying the difference in K-M terms by the scattering of the standard if the K-M functions are determined from absolute values of the reflectance. Since the intrinsic absorption as well as the scattering of the diluent must be taken into account in determining the absorption of the sample, both the s and the k of the standard must be known.

The absorption of the sample is k_{sa} , but when the sample is diluted with the standard, the sample occupies only a small fraction of the total sample volume. Kortum has shown that the value of k_{sa} measured for such a dilute sample is proportional to the concentration of the sample whether the dilution is achieved with a standard or with air. The value of k appropriate to a bulk material, k_{bulk} , is that value of k which would be measured if the sample completely filled the sample volume. The value for k_{bulk} may then be determined from the measured value for k_{sa} by multiplying the measured value k_{sa} by the ratio V_0/V :

$$k_{bulk} = k_{sa}(V_0/V) \quad (10)$$

where V_0 is the total sample volume and V is the volume occupied by the sample, which may be determined from the known weight if the sample density is known. Equations (8), (9), and (10) may be combined to give a value of the absorption coefficient for the bulk material:

$$k_{bulk} = (V_0/V)s_{st}[F_{st+sa}(R_{\infty}) - F_{st}(R_{\infty})] \quad (11)$$

which may be rewritten by using (2) as

$$n_{IM} = (\lambda/4\pi)(V/V_0)s_{st}[F_{st+sa}(R_{\infty}) - F_{st}(R_{\infty})] \quad (12)$$

The method described is similar to that used by Lindberg and Laude [1974] to measure n_{IM} for atmospheric dust collected over the southwestern United States.

AEROSOL COLLECTION AND SAMPLE PREPARATION

Saharan aerosol samples were collected from sites on three Atlantic islands during the summer of 1974 as a part of the Gate program as well as aboard the German research vessel *Meteor* during a November 1973 voyage. The sampling locations are shown on the map in Figure 1; the island sites were Barbados, West Indies (13°10'N, 59°25'W), Sal, Cape Verde Islands (16°45'N, 22°57'W), and Tenerife, Canary Islands (28°17'N, 16°39'W), and the *Meteor* was at approximately 17°N, 26°W during the collection of the sample analyzed here. The Barbados sampling site was located on the eastern coast of the island, approximately 20 m above the surface of the ocean. Local contamination was avoided because of the location of the sampling site and the persistent easterly flow. The Sal Island site was similarly situated for the same reason; each of these sites was in the same location as sampling sites operated by the University of Miami. The Tenerife samples were collected near the top of Pico de Teide, the highest point in the Canary Islands, at an altitude of approximately 3.7 km above sea level.

The aerosol samples were collected onto Delbag Micro-

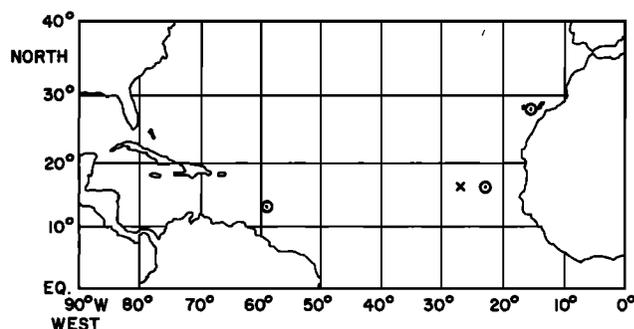


Fig. 1. Collection locations for Saharan aerosols for this study. The circles denote the island sites: Barbados (13°10'N, 59°25'W), Sal (16°45'N, 22°57'W), and Tenerife (28°17'N, 16°39'W). The cross is the approximate location of the research vessel *Meteor*.

poran polystyrene filters by using high-volume samplers operated at a sampling rate of approximately $2.8 \text{ m}^3 \text{ min}^{-1}$. The individual filters were run for approximately 24 hours; in order to obtain a sufficient sample for analysis, several partial sections of filters were grouped as one sample. The samples were separated from the filter material by placing the exposed filters in toluene to dissolve the filters. The aerosol material was not soluble in the toluene and so could be sedimented out; the toluene and dissolved filter material were poured off. The remaining material was washed several times with additional toluene to remove any remaining plastic material completely; then the toluene remaining after the last wash was removed by evaporation, leaving the aerosol sample.

Since sea salt aerosol was known to be a constituent of several of the samples and since sea salt is not soluble in toluene, portions of the aerosol from each of the island sites were also washed with water to remove the sea salt aerosol. This sea salt aerosol was a major constituent of the aerosol samples collected at Barbados according to data of Savoie and Prospero [1976b] on the relative concentrations of soluble and insoluble aerosol material; it appeared to be a minor constituent of the aerosol collected on Sal. The *Meteor* sample was not washed to remove the water-soluble aerosol, but the relative concentrations of Saharan aerosol and sea salt aerosol were determined from an elemental concentration analysis of the sample performed by K. Rahn (private communication, 1976).

The elemental concentrations were determined by using neutron activation analysis techniques; the measured elemental iron concentration was then used to infer a mineral aerosol concentration, as was discussed by Rahn *et al.* [1976]. The measured soluble sodium concentration in the sample was first corrected for the presence of soluble sodium in the crystal aerosols to determine the sodium concentration due to sea salt; the total salt concentration was then inferred from the soluble Na concentration by using data on the relative elemental composition of seawater presented by Goldberg [1965]. For our sample the sea salt concentration was determined to be approximately $14 \mu\text{g m}^{-3}$, a figure slightly smaller than but consistent with the mean value of approximately $20 \mu\text{g m}^{-3}$ reported by Savoie and Prospero [1976b] for their measurements at several stations during the Gate program. The concentration of Saharan mineral aerosol for this sample was approximately $76 \mu\text{g m}^{-3}$, so that the sea salt aerosol represented approximately 16% of the total aerosol by weight. This value was used to adjust the measured value of n_{IM} for the *Meteor* sample to obtain the actual value characteristic of the Saharan mineral aerosol.

MEASUREMENTS OF n_{RE}

Measurements of n_{RE} were made by using the Becke line technique for samples collected at Barbados and Tenerife as well as on the *Meteor*. The particles were immersed in oils whose indices of refraction and dispersion were known; no single-particle analysis was done, but average values of the real refractive index for clusters of particles were determined. Measurements were made by using both white and red (633 nm) illumination. According to Wright [1913], when white light is used to determine n_{RE} , the effective wavelength at which the index is determined is 550 nm, the wavelength of peak sensitivity of the eye in photopic vision.

The n_{RE} for the majority of the particles was bounded above and below by two index oils differing in n_{RE} by 0.004 for white light. Since a white Becke line was observed in each case, our estimate of the best value for n_{RE} for the particles is the value of n_{RE} midway between the values for the two oils anywhere in the visible. Considered alone, this bounding above and below would lead to an uncertainty of ± 0.002 in n_{RE} , but we are looking at average values of n_{RE} for clusters of birefringent particles that have three principal indices of refraction in general and are oriented in random directions. For this reason we feel that our uncertainty at each point is larger, and we have estimated the uncertainty at ± 0.004 in n_{RE} . When light with $\lambda = 633$ nm was used for illumination, the n_{RE} for the majority of the particles was bounded by the same oils as bounded it for the white light illumination.

Each of the samples was composed primarily of brownish aggregates of birefringent crystals, which appeared to be clay-coated particles. These aggregates generally had radii greater than $0.3 \mu\text{m}$. The *Meteor* sample also included some clear isotropic crystals having n_{RE} values of 1.548 ± 0.002 at 550 nm and 1.544 ± 0.002 at 589 nm. The crystal appearance and optical properties are appropriate for halite crystals, which we attribute to the sea salt aerosol. The rest of the *Meteor* sample consisted of the brown aggregates. Between 85 and 90% of the aggregates had a value of $n_{RE} = 1.558 \pm 0.004$ at 550 nm, and between 10 and 15% had a slightly higher value, $n_{RE} = 1.562 \pm 0.004$ at 550 nm.

The Tenerife sample was very similar. It contained brown aggregates in the amount of approximately 85% by volume, having an average n_{RE} value of 1.558 at 550 nm; the other 15% consisted of slightly larger aggregates with a slightly higher n_{RE} , 1.562 ± 0.004 at 550 nm. These were the same values determined for the *Meteor* sample. As was expected, no halite crystals were found in the Tenerife sample, since the sampling site on Tenerife was 3.7 km above sea level, above the marine boundary region in which sea salt aerosols would be expected to be a significant constituent of the atmospheric aerosol.

The Barbados sample consisted of the brown aggregates with $n_{RE} = 1.558 \pm 0.004$ at 550 nm. None of the material with the slightly higher index of refraction was observed in our Barbados sample.

The samples are very similar, and we have taken the value of n_{RE} exhibited by the vast majority of the aerosol particles as our estimate of n_{RE} for the Saharan aerosol. Including the few particles with the slightly larger index of refraction would not significantly change the estimated values. This estimated value of n_{RE} is 1.558 ± 0.004 at 550 nm; use of the red illumination results in a value of 1.552 ± 0.004 at 633 nm. These values are shown in Figure 2 as the circles with error bars at 550 and 633 nm. In this figure we have also shown a dashed line that is determined by the dispersion curves of the index matching oils and is the value halfway between the index values at each

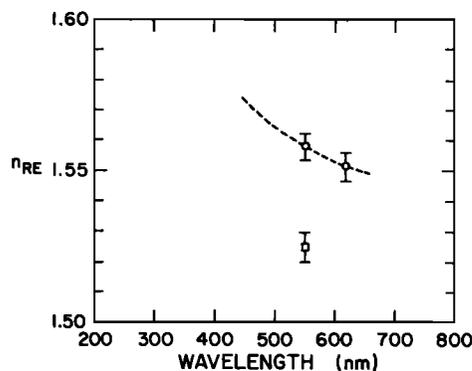


Fig. 2. Values of n_{RE} for soil-derived aerosols. The circles denote the measured values for Saharan aerosols at 550 and 633 nm. The dashed line is our estimate for the behavior of n_{RE} with wavelength for Saharan aerosols. The square is an average value measured for soil-derived aerosols over the southwestern United States. See text for details.

wavelength for the two oils that bounded n_{RE} above and below for these particles.

The dispersion curve of the particles appeared to match that of the oils between 550 and 633 nm, and no color effects indicating a crossing of the dispersion curves were observed. Thus we feel that a useful estimate of n_{RE} is given by the curve in Figure 2, although the extrapolation to wavelengths shorter than 550 nm is less certain than our measured values. An average value was determined for similar aerosols collected in rural areas of the southwest United States [Grams *et al.*, 1974]; the value of 1.525 ± 0.005 , determined for these aerosols by similar methods, is slightly lower than that determined for the Saharan aerosols.

MEASUREMENTS OF n_{IM}

All of the reflection measurements were made with a Cary-14 spectrophotometer equipped with a 15-cm integrating sphere coated with barium sulfate paint. In order to obtain absolute values for the reflectance, each reflectance measurement was referenced against a plate whose absolute reflectance had been determined by the method described by Goebel *et al.* [1966]. Basically, the absolute reflectances of the plate as a function of wavelength were determined by measuring the relative reflectances of the plate and of the opening in an auxiliary sphere coated with the same material as coated the plate. The absolute reflectance of the coating material was determined from the relative geometries of the auxiliary sphere and of the flat plate.

The dilution method described earlier was used so that the reflectance measurements would be in the range in which the Kubelka-Munk theory is most accurate. A highly refined barium sulfate powder, Eastman white reflectance standard (Eastman Co., Rochester, New York), was used as standard and diluent. The amount of powder used in a total sample volume of approximately 19 cm^3 was approximately 34 g, giving a density of approximately 2 g cm^{-3} , a value similar to that of Lindberg and Laude [1974].

We have previously measured the absorption and scattering properties of this barium sulfate powder [Patterson *et al.*, 1977]. The measurement procedure involved the determination of R_∞ as a function of wavelength for pure barium sulfate and for mixtures of the barium sulfate and aerosol having various concentrations of aerosol; each determination of R_∞ was the result of averaging several individual measurements.

For each concentration the K-M function was calculated from the measured value for absolute reflectance; the K-M function for the pure barium sulfate was subtracted from the K-M function for the mixture; the aerosol volume was calculated from the known mass of aerosol and an assumed density of 2.5 g cm^{-3} ; and n_{IM} was calculated as a function of wavelength by using (12). The measurements with different aerosol concentrations were made to check the validity of our data, since the absorption must be proportional to concentration for the dilution method to be valid; for our data the aerosol concentration dependence was found to be valid for aerosol concentrations between 4 and 24 mg per sample.

The uncertainties in our data fall into two categories. In the first are the random errors such as spectrophotometer noise, slight variations in the surface that can affect the measured values of reflectance, and weighing uncertainties in the sample. These random errors appear to be of the order of 20%. There are also systematic uncertainties that increase the total measurement uncertainty but do not affect the relative values for the samples. Such uncertainties are those due to uncertainties in the scattering, which we estimate to be approximately 10%, and uncertainty in the aerosol density, which we feel is less than 5%. There is uncertainty in the Kubelka-Munk theory itself as it is applied to our measurements. This uncertainty is of course harder to estimate quantitatively, but the comparison with the work of Gate discussed earlier indicates that there is no significant systematic discrepancy greater than about 5% between the value of k inferred from the Kubelka-Munk theory and the value of k measured for a bulk material. We will take 5% as the estimate of the uncertainty in the correspondence of the k , so that our total uncertainty in the measurement is approximately 40%.

The results of our n_{IM} measurements for aerosols collected on Tenerife are shown in Figure 3. The plotted curves for these and the other n_{IM} measurements are for wavelengths between 300 and 700 nm, the data points being spaced 10 nm apart between 300 and 400 nm and 20 nm apart between 400 and 700 nm. Figure 3 shows the results of two individual measurements of different portions of the Tenerife sample as well as the average of the two measurements. As was mentioned earlier, the optical microscopic techniques used in the determination of n_{RE} for this aerosol did not reveal the presence of sea salt

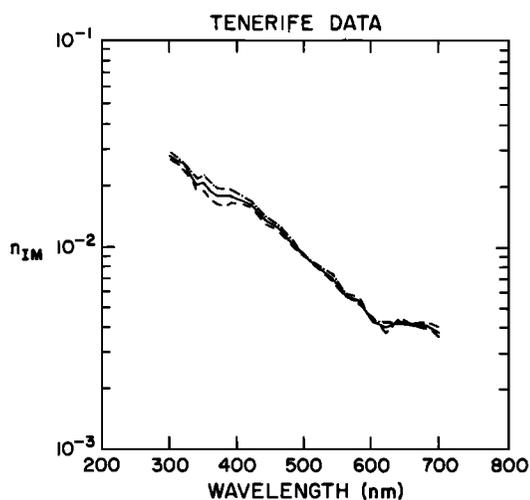


Fig. 3. Imaginary index of refraction for Saharan aerosol samples collected on Tenerife. The dashed and dashed-dotted lines represent two individual samples, and the solid line represents the average of the two samples.

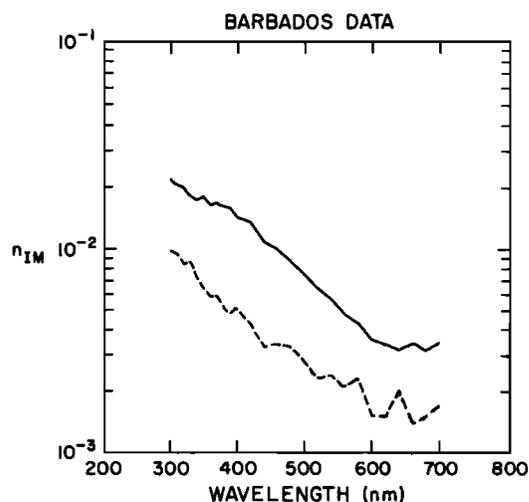


Fig. 4. Imaginary index of refraction for Saharan aerosols collected on Barbados. The dashed line represents the mixture of Saharan and sea salt aerosol; the solid line represents the pure Saharan aerosol.

aerosol in the sample. Partial measurement of a washed Tenerife sample did not show any increase of the measured value of n_{IM} over the unwashed sample, a result confirming that there was no significant amount of sea salt in the sample.

The measured values of n_{IM} for the Barbados samples, by contrast, do show the influence of the sea salt aerosol. Two measured curves of n_{IM} versus wavelength for samples collected at Barbados are shown in Figure 4. The lower curve is an average of two measurements made on an aerosol sample consisting of both the Saharan mineral aerosol and the sea salt aerosol. The upper curve is the result of measuring a different portion of the same aerosol sample after the sea salt had been removed and so is characteristic of the Saharan aerosol only. The fine structure in each of the lines is due to experimental uncertainties rather than to an actual structure in the measured spectrum.

We also measured n_{IM} as a function of wavelength for the sea salt removed from the Barbados aerosol sample. The measured values for wavelengths between 300 and 600 nm are shown in Figure 5; the values of n_{IM} for wavelengths greater than 600 nm were not determined, because the absorption of

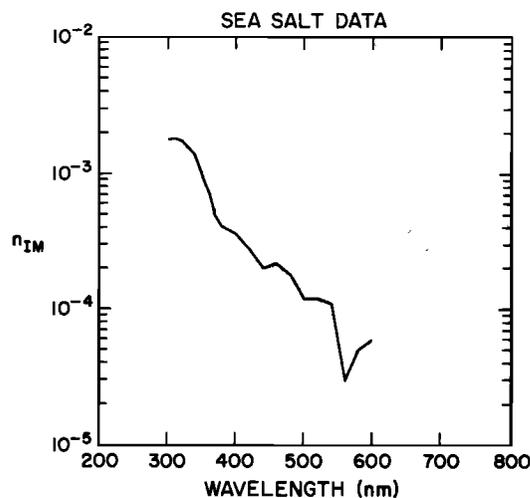


Fig. 5. Imaginary index of refraction for sea salt aerosol separated from the Barbados sample.

the sample was too small to be measured accurately. Owing to the small values for absorption and the limited sample volume available as well as the possibility of small admixtures of other water-soluble aerosols that can influence the measured imaginary index, the uncertainties in the measured values of n_{IM} for the sea salt are greater than those for the Saharan aerosol; throughout the range measured, our measurements of n_{IM} are accurate only within $\pm 5 \times 10^{-6}$. We can say, however, that n_{IM} for the sea salt aerosol is less than the corresponding n_{IM} for the Saharan mineral aerosol by more than 1 order of magnitude throughout the visible. So for the relative concentrations of mineral and sea salt aerosols seen in our samples and for the accuracy of our measurements we may consider the sea salt to be a nonabsorbing diluent for the Saharan crust-derived aerosol, effectively reducing the measured value of n_{IM} by some constant factor below the actual value.

The data for n_{IM} for the *Meteor* sample are shown in Figure 6. The dashed line represents our measured values of n_{IM} as a function of wavelength for the total aerosol sample consisting of Saharan mineral aerosol and sea salt aerosol. As discussed previously, the concentration of sea salt in the total aerosol for this sample was determined to be approximately 16% by weight, so that the actual concentration of the Saharan aerosol was only 84% of the total aerosol concentration. Consequently, the values of n_{IM} calculated on the basis of total aerosol were divided by 0.84 to obtain the values for n_{IM} for the Saharan aerosols. These adjusted values are shown as the solid line in Figure 6.

Our n_{IM} curves for two sets of Sal Island data are shown in Figure 7. The solid line shows the results of measurements on a washed sample of aerosol with the characteristic brown coloration of the Saharan aerosol. The dashed line, by contrast, shows the results of our measurements on a sample from which the salt had not been removed and which contained some other darker aerosol in addition to the Saharan aerosol. This additional aerosol was obtained from a filter that had a dark grey appearance after exposure on Sal rather than the characteristic brown color of the other Sal filters and the filters from the other sampling sites. The net effect of this additional aerosol is to decrease the slope of the n_{IM} line. The data are consistent with the assumption that this additional aerosol has

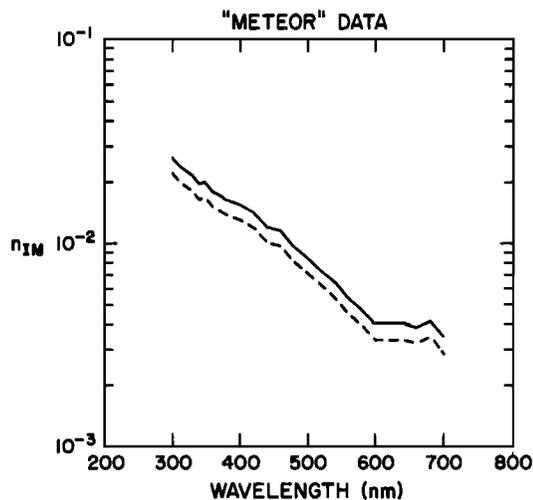


Fig. 6. Imaginary index of refraction for Saharan aerosols collected aboard the *Meteor*. The dashed line represents the mixture of Saharan and sea salt aerosols; the solid line represents our estimated value for the pure Saharan aerosol. See text for details.

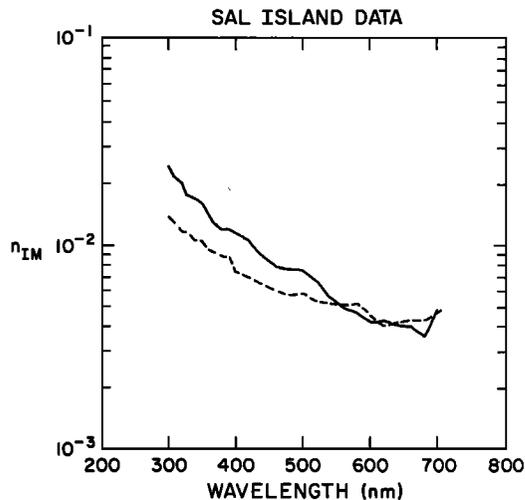


Fig. 7. Imaginary index of refraction for Saharan aerosol collected on Sal Island. The solid line represents the pure Saharan aerosol; the dashed line represents a combination of aerosols. See text for details.

a relatively constant value of n_{IM} in the visible, which is in turn consistent with the dark grey appearance of the anomalously dark filter. We did not attempt to determine the source of this dark aerosol, since it appeared on no samples other than Sal Island samples and on only a very few of the Sal samples out of a series of samples taken daily throughout the summer of 1974.

DISCUSSION OF RESULTS

In Figure 8 we have shown the four curves which we feel are the best estimates of the value of n_{IM} as a function of wavelength for each of the four sampling locations. We have included the washed Sal (dashed-dotted line) and washed Barbados (dotted line) samples, the Tenerife sample (solid line), and the adjusted *Meteor* sample (dashed line). It is apparent from the figure that the curves are very similar, and there appear to be no significant differences in the curves that can be attributed to actual differences in the measured values of n_{IM} among the various samples. It is reasonable to infer from the data that the Saharan aerosol may be described by a single set

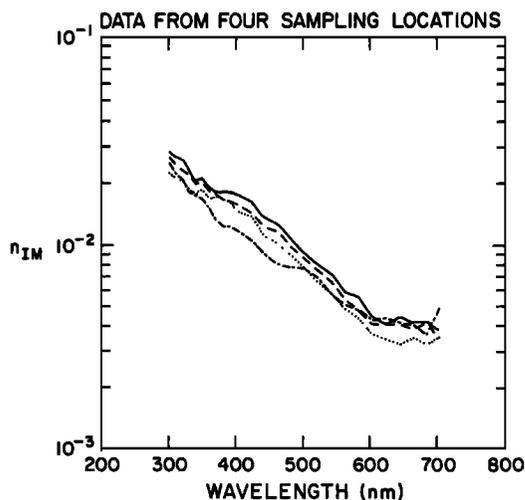


Fig. 8. Imaginary index of refraction for Saharan aerosols from each of the collection locations. The solid line represents the Tenerife sample; the dashed line, the *Meteor* sample; the dotted line, the Barbados sample; and the dashed-dotted line, the Sal Island sample.

of values of n_{IM} versus wavelength. We have determined this set of values of n_{IM} versus wavelength by averaging the four curves shown in Figure 8. The results of the averaging are shown in Figure 9 as the solid line. Our measurements indicate that the value of n_{IM} for the Saharan aerosol decreases from about 0.025 at 300 nm to approximately 0.0038 between 600 and 700 nm and then shows a slight increase with increasing wavelength.

Our results for n_{IM} thus confirm our results for n_{RE} , namely, that there appear to be no significant differences in the optical properties of the Saharan aerosol collected from the different measuring locations, even though the sites are located on opposite sides of the Atlantic and the samples are for different seasons of different years. This result is consistent with the results of *Rahn et al.* [1976], who have shown that the aerosol is spatially uniform over several thousands of kilometers in terms of composition and aerosol concentration, and those of *Savoie and Prospero* [1976a], who have shown that the size distribution changes relatively little as the aerosol cloud moves across the Atlantic Ocean.

Since the optical properties of the Saharan aerosols appear to be relatively constant, comparisons between different measurements of the optical properties of Saharan aerosols are useful, differences in the results reflecting differences in the techniques rather than intrinsic differences in the aerosols. Our laboratory results can be most directly compared with some values for n_{IM} determined by *Carlson and Caverly* [1977] on the basis of direct-diffuse radiation measurements. These values are shown by the open circles at 375, 468, and 610 nm in Figure 9. The values of n_{IM} measured by us and by Carlson and Caverly appear to be in excellent agreement, well within the combined experimental uncertainties, across the visible spectrum. This agreement between the direct-diffuse radiation measurements of n_{IM} and our laboratory measurements appears to imply that under the conditions of the Saharan aerosol measurements the parameters determining the ratio of the direct to the diffuse radiation are not sensitive to the non-

sphericity of the Saharan aerosols. However, more work is needed to determine under what conditions such measurements are or are not sensitive to the sphericity of the aerosols.

Although the comparison is not as direct as that with Carlson and Caverly's measurements, we have also shown the results for two imaginary index determinations for crust-derived aerosols over the southwestern United States. The data point at 500 nm represents some measurements of n_{IM} by *Grams et al.* [1974], who used a polar nephelometer in a rural semiarid area. The solid horizontal line for $n_{IM} = 0.008$ between 310 and 690 nm represents some results for average values of n_{IM} determined for the same general area and type of aerosol by *De Luisi et al.* [1976] using a direct-diffuse technique similar to that used by Carlson and Caverly. Both of these sets of values appear to be consistent with the values measured by us and by Carlson and Caverly for the Saharan aerosol layer.

CONCLUSIONS

Both the imaginary and the real parts of the complex index of refraction for Saharan aerosols have been determined as a function of wavelength: n_{RE} was determined by means of an immersion oil technique, and n_{IM} was determined from a measurement of the total diffuse reflectance of the aerosol by means of an analysis using the Kubelka-Munk theory. No significant differences in optical properties were seen among the samples collected at sampling sites on both sides of the Atlantic Ocean, a result leading to the conclusion that the Saharan aerosol may be characterized by one set of optical properties whether the measurements are made near the African coast or near the Caribbean.

The homogeneity of the aerosol optical properties also indicates that differences in measurements of the optical properties are more likely to be due to differences in the measurement technique than to intrinsic differences in the aerosols, if care is taken that the aerosol measured is Saharan aerosol only. Comparison of our results with others based on measurements of the ratio of the direct to the diffuse radiation shows that the two independent methods give remarkably similar results.

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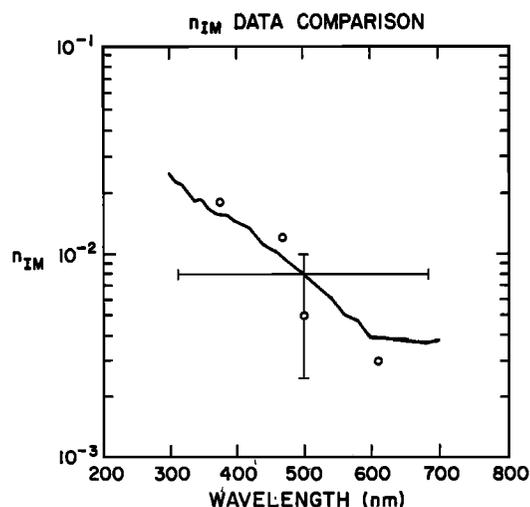


Fig. 9. Measurements of the imaginary index of refraction for Saharan and other soil-derived aerosols. The solid curve represents our best estimate of the imaginary index of refraction as a function of wavelength for the Saharan aerosols. The circles at 375, 468, and 610 nm represent measurements of Carlson and Caverly for Saharan aerosols; the circle with error bar at 500 nm represents an average of some measurements of soil-derived aerosols by Grams et al.; and the horizontal solid line at $n_{IM} = 0.008$ represents an average value measured by De Luisi et al. for similar aerosols. See text for details.

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