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A MEASUREMENT OF THE ABSORPTION
COEFFICIENT OF ATMOSPHERIC DUST

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Army Electronics Command
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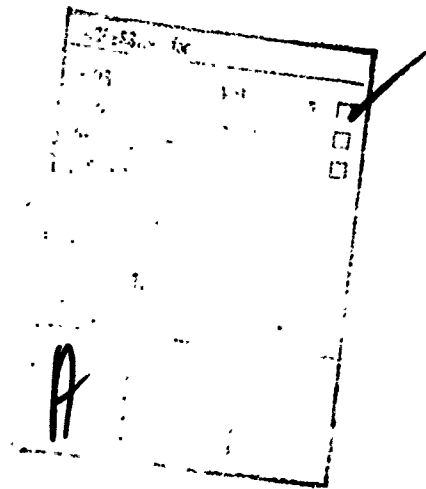
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A method developed by previous workers for measuring the absorption coefficient of strongly absorbing powdered materials has been applied to samples of atmospheric dust in the 0.3 to 1.1 µm wavelength interval. This work, which is based on the Kubelka-Munk theory of diffuse reflectance, provides an estimate of the optical absorption coefficient. The corresponding imaginary refractive index is calculated from this value. Results are given for several samples of dry atmospheric dust collected in the desert of southern New Mexico. A

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typical value for the imaginary refractive index was found to be 0.007 at 0.6 μm, with little dependence on wavelength in the spectral range investigated. These results are found to be in good agreement with those of other workers obtained by different methods.

CONTENTS

INTRODUCTION

The quantitative measurement of the absorption coefficient of atmospheric dust is a problem made difficult by the complicated nature of the material. Since dust samples consist of mixtures of finely powdered, highly scattering materials, conventional transmission spectroscopy is not directly applicable. The basic problem is to determine what fraction of the incident light attenuated by a sample of dust is lost due to scattering, and how much is absorbed by the particles themselves. The measurement of absorbed energy must then be related to the basic optical absorption coefficient of the sample. This latter problem is not trivial since the Bouguer-Lambert law, which normally solves this problem for nonscattering media, is not directly applicable here.

In spite of the difficulties in making such measurements, several workers have devised methods for solving the problem, with varying degrees of success. Fisher [1] has obtained results based on a diffuse transmittance technique and the assumption that the Bouguer-Lambert law can be used. Volz [2, 3] has applied variations of the potassium bromide pressed disk method, commonly used for qualitative work in the infrared region, to gain some information about the absorption coefficient of dust. Grams et al. [4] have estimated the imaginary refractive index of airborne fly ash by inferring the value needed to account for their measured values of laser radar backscatter cross section and particle size distribution. Previous work in this laboratory [5] has developed a somewhat complicated method for applying the Kubelka-Munk theory of diffuse reflectance to the problem. This report describes a straightforward method of obtaining the absorption coefficient from a single diffuse reflectance measurement. The final section is a discussion of the results obtained here in comparison to those obtained by other workers.

THEORY

In this work we regard a sample of atmospheric dust as a powdered material of unknown shape, size distribution, refractive index and absorption coefficient. The object is to infer the absorption coefficient, or equivalently, the imaginary part of the complex refractive index of the material, from measurement of some optical property of the sample.

The most straightforward measurement that contains information about the absorption coefficient of a powder is a measurement of its diffuse reflectance, R_∞. It is well known that the diffuse reflectance of a powder is highly dependent on the absorption coefficient. R_∞ is high when the absorption is low, and of course is low for strongly absorbing materials.

For this reason the diffuse reflectance of a powdered sample of a solid material plotted as a function of wavelength has the character of a transmission spectrum of the material itself. The resemblance is only qualitative, however.

To obtain quantitative information from a diffuse reflectance measurement, it is necessary to have some analytical relationship between the diffuse reflectance and the desired optical property of the powder, in this case the absorption coefficient. Several theories of diffuse reflectance provide such a connection. Some are very satisfactory quantitatively, but require considerable a priori knowledge about the sample, such as size or real refractive index. For the work reported here the Kubelka-Munk theory was used, since it requires no other knowledge about the sample at all. The price of this convenience is the assumption that the powder is an isotropically scattering medium, a point we will come back to later in this section, and a definition of absorption coefficient slightly different from that commonly used in the Bouguer-Lambert law.

The Kubelka-Munk theory as it is applied in this work is well known and will not be discussed in detail here. The reader is referred to two excellent books [6, 7] on the subject for details on its origin and derivation. The equations used here and the notation, as much as possible, are taken from the work by Kortum [7]. The notation is summarized in Table I.

The Kubelka-Munk theory treats a powder sample as a continuous, turbid medium. In a layer of infinitesimal thickness, fractions s and k per unit path length of the incident light are scattered and absorbed, respectively. The quantities s and k, in units of cm^{-1}, are called the Kubelka-Munk scattering and absorption coefficients It is known that the coefficient k is very closely related to the absorption coefficient in the familiar Bouguer-Lambert law. Discussions of this point are available in the references [6, 7, 8]. The object of the work reported here is to determine the value of k for a sample of atmospheric dust, and to infer the imaginary refractive index from it.

One of the most important accomplishments of the Kubelka-Munk theory is an equation relating the diffuse reflectance of a powder to the scattering and absorption coefficients:

$$\frac{(1 - R_\infty)^2}{2R_\infty} = \frac{k}{s} . \tag{1}$$

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TABLE I

DEFINITIONS OF SYMBOLS

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c Concentration, in moles per liter, of a pure powder sample

c^* Concentration, in moles per liter, of sample in a mixture of sample and diluting agent

d Thickness, in cm, of a sample layer

k Kubelka-Munk absorption coefficient in cm^{-1}

k^* Kubelka-Munk absorption coefficient in cm^{-1} for a mixture of sample and diluting agent

n' The imaginary part of the complex refractive index of atmospheric dust

R Diffuse reflectance of a layer of powder of thickness d

Rg Diffuse reflectance of the flat black paint used to coat the bottom of a shallow aluminum sample dish

R_∞ Diffuse reflectance of a layer of powder with a thickness large enough that no light passes through

R_∞^* Diffuse reflectance R_∞ measured for a mixture of sample and diluting agent

s Kubelka-Munk scattering coefficient in cm^{-1}

s^* Kubelka-Munk scattering coefficient in cm^{-1} for a mixture of sample and diluting agent

W The weight of pure atmospheric dust sample required to completely fill the volume of the sample dish

W^* The actual weight of atmospheric dust in the sample dish when R_∞^* is measured

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This expression is known as the Kubelka-Munk remission function. This provides a determination of the ratio k/s from one measurement of reflectance, R_∞. In order to arrive at values of k and s separately, more information is required.

The reflectance R_∞, by definition, is the diffuse reflectance of an "infinitely thick" layer of the powder; this means a layer so thick that no detectable light is transmitted through the sample. In practice this turns out to be a thickness of a few millimeters, depending on the value of k and s for the material. Suppose that we measure R_∞ on a suitably thick layer of sample, and then also measure R, the diffuse reflectance of a much thinner layer of thickness d. This can be done by placing the powder in a shallow dish of depth d. Suppose also that we have made the bottom of the dish nearly black, and have previously measured its diffuse reflectance Rg. Kortum [7, 9] has show from the Kubelka-Munk theory that s, R_∞, R, Rg, and d are related by the expression

$$\ln\left[\frac{(R - 1/R_\infty)(Rg - R_\infty)}{(Rg - 1/R_\infty)(R - R_\infty)}\right] = 2sd(1/R_\infty - R_\infty), \qquad (2)$$

and that s can be determined satisfactorily from measurement of these quantities. Having obtained s in this manner, one can then solve for k in Eq. (1). This provides a means of obtaining the absorption coefficient of a powder from three reflectance measurements and one thickness measurement.

The method described above has been used successfully to obtain k and s for very weakly absorbing (nearly white) powders. But it is not a satisfactory solution to the problem of interest here for three reasons. One is that for a sample of atmospheric dust, the absorption coefficient is relatively high, and therefore even a thin layer appears "infinitely thick," making it difficult to determine R, R_∞, and d to a satisfactory degree of accuracy. Second, it is not easy to collect a sufficiently massive sample of atmospheric dust to carry out these kinds of measurements. The third problem arises from the assumption in the Kubelka-Munk theory that the sample powder scatters light isotropically. Experiment shows that weakly absorbing powders are isotropic scatterers to a very good degree of approximation. But this is not necessarily true for strong absorbers like atmospheric dust.

The difficulties mentioned above arise from the fact that atmospheric dust is a relatively strong absorber of light. Therefore, they can be avoided by diluting the dust sample with some other powder which has a

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trivially low absorption coefficient. One can then apply Eqs. (I) and (2) to the diluted sample, and obtain an absorption coefficient for the mixture. The remaining problem is to infer the absorption coefficient of the pure dust sample from that of the mixture and knowledge of the dilution ratio.

First we look at the problem of obtaining the absorption coefficient of a mixture of atmospheric dust and some other powder with low absorption coefficient used as a diluting agent. We can then write Eq. (I) in the form

$$\frac{(1 - R_\infty^*)^2}{2R_\infty} = \frac{k^*}{s^*} , \tag{3}$$

where the asterisk indicates that we are dealing with the properties of the diluted sample. Kortum has shown that if the sample is highly diluted, the scattering coefficient s of the mixture is not affected by the presence of the sample, so for a value of s^* we can use the scattering coefficient for the pure dilutant. This quantity can easily be determined by measuring R_∞, Ro, Rg, and d, then applying Eqs. (I) and (2) as discussed above, since the diluting agent is not a strong absorber like the pure dust sample. We can measure R_∞^* directly for the diluted dust sample, and determine k^* from Eq. (3).

Now we have a valid measurement of k^*, the absorption coefficient of the diluted dust sample. What we really want, however, is the absorption coefficient of the pure dust sample, undiluted by air spaces or other white diluting agents. Previous work [7, 9] has shown that for strongly diluted samples, the absorption coefficient k^* is directly proportional to the sample concentration, in moles per liter, over several orders of magnitude of concentration. This is analogous to Beer's law in conventional transmission spectroscopy of solutions. For the case of powdered materials, derivations from linearity occur when the sample is not sufficiently diluted, and the assumption that the scattering coefficient of the mixture is that of the pure diluting agent is not valid. Nonlinearities also occur for extremely weak dilutions because the absorption coefficient of the mixture is so small that the intrinsic absorption coefficient of the diluting agent can no longer be considered trivial. In practice for the kinds of materials dealt with here, the linear region extends typically over a dilution range of one part sample in 10^2 parts diluting agent to one part sample in 10^5 parts diluting agent.

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We now assume that we have measured R_∞^* for a diluted sample of concentration which falls in the linear region, and have calculated the corresponding value of k^*. Then we can obtain the absorption coefficient k for a pure sample, free from dilution by any agent, from the expression

$$k = k^* \frac{c}{c^*},\qquad(4)$$

where c^* is the concentration in moles per liter of sample in one dilution, and c is the concentration, in moles per liter, the sample would have if it were free of air spaces or other diluting agents. Since concentration in moles per liter is directly proportional to the weight per unit volume of sample, we can replace the ratio c/c^* in Eq. (4) with the ratio W/W^*, where W^* is the weight of atmospheric dust in our sample volume, and W is the weight of dust that would be required to fill the sample volume completely with the dust. Making this change in Eq. (4) and substituting for k^* from Eq. (3), we have

$$k = \frac{s^*(1 - R_\infty^*)^2\, W}{2 R_\infty^*\, W^*}.\qquad(5)$$

This expression gives the Kubelka-Munk absorption coefficient for the pure atmospheric dust sample as a function of one reflectance measurement, a weight measurement, and a predetermined scattering coefficient for the diluting agent.

MEASUREMENTS

All of the diffuse reflectance measurements were made with a Cary 14 spectrophotometer equipped with the manufacturer's 25 cm integrating sphere, as described by Hedelman and Mitchell [10]. In such an instrument the quantity measured is the relative reflectance, that is, the reflectance of the sample divided by the reflectance of some material used as a reference standard. Thus, all measurements must be corrected by multiplying the measured reflectance by the absolute reflectance of the reference material to determine the absolute reflectance of the sample. In the work done here, highly refined $BaSO_4$ [11] was used as a reference standard, and the absolute reflectance of this material was obtained from the work of Grum and Luckey [12].

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As a diluting agent, the same $BaSO_4$ used as a reference standard was chosen. A shallow aluminum dish a fraction of a millimeter deep, painted with flat black enamel, was used to measure R, the reflectance of a thin layer of the white powder. Rg, the reflectance of the black dish, was also measured. The scattering coefficient was determined from Eqs. (1) and (2) and used as the value of s*, the scattering coefficient of the diluted samples.

The dust samples used here were collected on the surface of a 0.45 μm pore size membrane filter at the input of a laboratory vacuum pump. Each sample was collected over a period of ten days, from a location 3 meters above the ground in a desert basin in southern New Mexico. The dust collected on the filter was gently scraped off, and an appropriate quantity was mixed with 34 grams of the $BaSO_4$ diluting agent [13]. Mixing of the $BaSO_4$ and dust was done in a polystyrene vial in a vibrator mixer for a period of 5 minutes. The mixture was then pressed into a 1 cm deep by 5 cm diameter lucite dish using a clean glass plate. The reflectance, R_∞^*, was measured directly, and the weight, W*, of dust in the lucite dish was recorded. The quantity W, the weight of dust that would completely fill the dish, was calculated from the known volume of the dish and the assumption that the average specific gravity of atmospheric dust materials is 2.4, as reported by other workers [14].

Different quantities of dust were used to insure that the concentration chosen was in the linear range as discussed above, and to determine over what wavelength range the measurement could be made. It was found that a suitable linear relationship existed for wavelengths from 0.3 to 1.1 μm. Outside of this range, the intrinsic absorption coefficient of the $BaSO_4$ was too high to be considered negligible. It was found that quantities of from 3 to 30 mg of dust in the sample dish produced satisfactory results. From measurements of R_∞^* at different wavelengths, and knowledge of W, W*, and s* as determined earlier, k was calculated as a function of wavelength from Eq. (5). Typical results are listed in Table II and discussed in the next section.

A programmable desk calculator was used to perform the calculations required in this work. By estimating the errors in each of the input measurements involved and determining the effect of each on the computed value of k, an estimate of the experimental error was made. The largest source of error was found to be in the determination of s*, because of the difficulty of preparing a uniform layer of powder a few tenths of a millimeter thick. It was estimated that s* might be in error by as much as 20%. Photometric errors, assumed to be about 2%, were found to contribute an error of about 5% to the determination of k. Since s* is not determined individually for each computation of k, its uncertainty affects the absolute accuracy of the measurement, but not the repeatability

TABLE II

ABSORPTION COEFFICIENT AND IMAGINARY REFRACTIVE INDEX FOR THREE ATMOSPHERIC DUST SAMPLES

λ, μm	Sample #1		Sample #2		Sample #3	
	k, cm^{-1}	n'	k, cm^{-1}	n'	k, cm^{-1}	n'
.35	5805	.016	6312	.018	5803	.016
.40	3298	.011	4239	.013	2837	.009
.45	2275	.008	3230	.012	1814	.007
.50	1843	.007	2432	.010	1478	.006
.55	1485	.007	2243	.010	1300	.006
.60	1328	.006	1892	.009	1047	.005
.65	1119	.006	1659	.009	837	.004
.70	1109	.006	1537	.009	968	.005
.75	1100	.006	1396	.008	725	.004
.80	933	.006	1240	.008	668	.004
.85	919	.006	1202	.008	541	.004
.90	949	.007	1098	.008	494	.004
.95	978	.007	1057	.008	476	.004
1.00	1005	.008	1058	.008	702	.006
1.05	886	.007	1045	.009	852	.007
1.10	793	.007	907	.008	780	.007

from one sample to the next. Therefore, the experimental error in this determination of the Kubelka-Munk absorption coefficient of atmospheric dust is estimated to be on the order of 25% on an absolute basis, with a precision considerably greater. This error could be reduced substantially by performing many determinations of s* and averaging the results.

DISCUSSION OF RESULTS

The measurements made here give the Kubelka-Munk absorption coefficient for atmospheric dust, in units of cm^{-1}. In practice this number is found to be nearly equivalent to the absorption coefficient as defined by the Bouguer-Lambert law, usually differing from it by a factor of about two [6, 7, 8]. In order to compare the results of the work presented here with that of previous workers, we will assume the two absorption coefficients to be equivalent, so that a corresponding imaginary refractive index, n', can be computed from the usual expression:

$$n' = \frac{k\lambda}{4\pi} ,$$

where k is the absorption coefficient in cm^{-1} and λ is the wavelength in cm. In Table II the Kubelka-Munk absorption coefficient and corresponding imaginary refractive index are listed for different wavelengths for several dust samples.

Fisher [1] has obtained a value of 0.01 for the imaginary refractive index in the visible region for atmospheric dust collected at a location in West Germany. He also reported a sample-to-sample variation of about a factor of 3, and noted that there was little dependence on wavelength in the visible spectrum. Volz [2, 3, 15] has reported 0.007 for the imaginary refractive index of atmospheric haze particles in the visible spectrum, and similar values, although with a strong wavelength dependence, for water soluble rainwater residue.

The method reported here gives results which are consistent with those of other workers. An exact comparison is of course difficult because identical samples were not used in all cases, and the measurement errors involved are not easy to assess. However, it can be concluded from this work that diffuse reflectance spectroscopy can give a useful measurement of the imaginary refractive index of atmospheric dust samples. This method clearly has the advantage of simplicity and convenience, since it consists of a routine total diffuse reflectance measurement. Most laboratory spectrophotometers are capable of making such measurements using accessories designed for this purpose.

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