

# The Determination of the Index of Refraction Distribution of Oceanic Particulates

J. RONALD V. ZANEVELD, DAVID M. ROACH, AND HASONG PAK

*School of Oceanography, Oregon State University, Corvallis, Oregon 97331*

A method is described for determining the index of refraction distribution and the particle size distribution of suspended particles. The distributions are obtained by breaking down an observed volume scattering function into its contributing components. The component scattering functions are calculated using Mie theory. The component functions include all size distributions and indices of refraction that can be expected to be present. The method was applied to a volume scattering function observed in the Sargasso Sea. Forty components were used with five different indices of refraction and eight different particle size distributions. The resultant index of refraction distribution was bimodal. Components with indices of 1.05 and 1.15 dominate the calculated volume scattering function. The calculated particle size distribution falls within experimentally determined limits for the size distribution.

The parameters that determine the light scattering and attenuation properties of a particle suspended in the ocean are shape, size, and internal distribution of the index of refraction. Light scattering properties of the individual particles are difficult to measure. Techniques for studying large numbers of particles simultaneously have thus been developed as a necessity. Optical measurements are particularly well suited for the study of particles in the ocean, since there is no destructive interaction between the particles and the light. Light scattering by oceanic suspensions is usually described in terms of the volume scattering function  $\beta(\theta)$  defined by

$$\beta(\theta) = \frac{dI(\theta)}{E dV}$$

where  $dI(\theta)$  is the radiant intensity scattered at an angle  $\theta$  with respect to the incident beam from the volume element  $dV$  when it is illuminated by a monochromatic irradiance  $E$ . Except for the extremely near-forward angles ( $\theta < 1^\circ$ ) the volume scattering function can readily be determined. Determination of the particle characteristics from the volume scattering function is extremely difficult owing to the great variety of shapes, sizes, and indices of refraction. It is necessary to make several simplifying assumptions if any particle properties are to be calculated from observed optical parameters.

The theoretical link between observed and calculated scattering functions is provided by Mie's [1908] theory. By using Mie's theory it is possible to calculate the light scattering characteristics of a single spherical particle with a uniform index of refraction. The light scattering characteristics of a collection of spheres can then be obtained by summing the contributions of the individual spheres provided they are separated by a distance of about three radii or more [Van de Hulst, 1957]. The scattering calculations have been extended to include other simple shapes and layered spheres. It has been shown, however [Holland and Gagne, 1970; Plass and Kattawar, 1971], that when one considers any size distribution of randomly oriented particles, the same size distribution of spherical particles will give comparable volume scattering functions, particularly in the forward hemisphere, when the other particle parameters are similar.

Nonuniformity of the index of refraction within a particle will cause the light scattering characteristics to differ markedly from those for a particle with a uniform index [Mueller, 1973].

Calculation time increases dramatically for multilayered spheres, however.

For a collection of spheres one can define a particle size distribution  $f(D) dD$  giving the number of particles per unit volume with diameters between  $D$  and  $D + dD$ . For nonspherical particles the definition of size becomes more difficult. Since particle size distributions are usually obtained with an instrument that measures the volume of a particle, the diameter of a nonspherical particle will be taken to be the diameter of a spherical particle with the same volume as the observed particle. The instrument most commonly used for measurements of the particle size distribution is the Coulter counter. In our theoretical considerations of light scattering it is not necessary to know the size distribution beforehand.

The index of refraction of the suspended particles is still largely unknown, although great progress in this area has been made in recent years. The index of refraction of particles may become a valuable indicator of the nature of the suspended particles. Already a qualitative correlation has been found between dynamic features in the ocean and the distribution of the index of refraction [Pak and Zaneveld, 1973].

Even if a given suspended particle is homogeneous and can be characterized by one index of refraction, a size distribution of particles can only be characterized completely by a two-dimensional index of refraction distribution, since for each size of particle there exists a distribution of indices of refraction. It is this two-dimensional distribution that one must ultimately obtain in order to fully understand light scattering by particulate matter in the ocean. The present paper is a step toward obtaining methods for the determination of such a distribution.

Earlier investigations [Sasaki et al., 1960; Kullenberg, 1972; Gordon and Brown, 1972; Zaneveld and Pak, 1973; Carder et al., 1972; Morel, 1973a] have dealt with the determination of one value of the index of refraction that would be representative of the entire collection of particles. The value of the index of refraction chosen is the value that best reproduces an observed set of light scattering characteristics, given an observed or postulated particle size distribution. Table 1 summarizes the results of the various authors and shows that the average indices of refraction calculated by various authors fall either in the range 1.15–1.25 or 1.01–1.05. This result indicates that particle distributions are either dominated by inorganic and organic skeletal material with a high index of refraction or by the watery protoplasm of organic matter with a low index.

This view is supported by the fact that low index dominated particulates were mostly observed in regions of high productivity.

It has usually been assumed that a low index of refraction is associated with particulates of organic origin and a high index of refraction with an inorganic origin. This assumption has not been thoroughly tested. Organic particles such as phytoplankton frequently contain skeletal material with a high index of refraction, so that assigning extremely low indices (1.01–1.02) to organic matter is probably incorrect.

#### THEORETICAL CONSIDERATIONS

If a volume element contains  $N$  particles, each with a scattering function  $\beta_i(\theta)$ , then the scattering function for the volume element is given by

$$\beta(\theta) = \sum_{i=1}^N \beta_i(\theta) \quad (1)$$

The problem to which we direct ourselves is, given  $\beta(\theta)$  (but not necessarily the size distribution of the  $N$  particles), to obtain maximum information concerning the indices of refraction of the  $N$  particles and the size distribution of the particles if it is not known. If we suppose that the index of refraction of all the particles is the same, then

$$\beta(\theta) = \int_{D_1}^{D_u} A f(D) \beta(\theta, D, m) dD \quad (2)$$

where  $f(D) dD$  is the normalized size distribution (one particle per unit volume) of the particles,  $A$  is the total number of particles per unit volume,  $m$  is the index of refraction of the particles relative to water, and  $\beta(\theta, D, m)$  is the scattering function for a particle with diameter  $D$  and relative index of refraction  $m$ . The  $D_1$  and  $D_u$  are the smallest and largest diameters, respectively, of particles that are present or from which a contribution to the scattering function is expected. The  $\beta(\theta, D, m)$  functions are obtained from Mie theory. By performing the integration for several values of the index of refraction relative to water  $m$ , one may choose that value of  $m$  for which the experimental volume scattering function  $\beta(\theta)$  most closely approximates the theoretical one. This approach is the classical one for which the results are shown in Table 1. An improvement over assuming that all particles have the same index of refraction is to postulate that the particles consist of inorganic

and organic material. We then have

$$\beta(\theta) = \int_{D_{11}}^{D_{1u}} A_1 f_1(D) \beta(\theta, D, m_1) dD + \int_{D_{21}}^{D_{2u}} A_2 f_2(D) \beta(\theta, D, m_2) dD \quad (3)$$

and

$$f(D) dD = f_1(D) dD + f_2(D) dD$$

where subscripts 1 refer to organic matter and subscripts 2 refer to inorganic matter. One may vary the indices  $m_1$  and  $m_2$  as well as the limits  $D_{1u}$ ,  $D_{11}$ ,  $D_{2u}$ , and  $D_{21}$  until again the experimental curve most nearly matches the observed one. In the past this has been done by trial and error; a more deterministic approach will be presented in this paper.

It is natural to try to expand the model to contain a large number of components. We can group particles according to size distribution and index of refraction given

$$\beta(\theta) = \sum_{i=1}^N \sum_{j=1}^M \int_{D_{1j}}^{D_{1u}} A_{ij} f_{ij}(D) \beta(\theta, D, m_j) dD \quad (4)$$

and

$$f(D) dD = \sum_{i=1}^N \sum_{j=1}^M A_{ij} f_{ij}(D) dD \quad (5)$$

In many cases the total size distribution  $f(D) dD$  is not known, in which case (5) is not a condition that needs to be satisfied. As only  $f(D) dD$  and  $\beta(\theta)$  or  $\beta(\theta)$  alone are known, it is clearly impossible to determine the  $f_{ij}(D) dD$  without a number of approximations, and even then one cannot be assured of obtaining unique solutions, since the shape of the volume scattering function is only a weak function of the size distribution and index of refraction.

The integral

$$I_{ij} = \int_{D_{1j}}^{D_{1u}} f_{ij}(D) \beta(\theta, D, m_j) dD \quad (6)$$

has been shown to be [Morel, 1973b] a smooth function of  $C$  for the range of values expected in the ocean when  $f_{ij}(D) dD$  is Junge-type distribution  $f_{ij}(D) = ND^{-c} dD$ . Similarly,  $I_{ij}$  is a smooth although possibly double-valued function of  $m_j$ . It is thus reasonable to assume that a limited number of these  $I_{ij}$  components can be used to reconstruct observed scattering functions that are also reasonably smooth. We may then choose the appropriate  $m_j$  and  $f_{ij}(D) dD$  in (4). The values of  $m_j$  must span the range of indices that one expects to find; this range is 1.00–1.20 relative to water. Ideally, one would construct the  $f_{ij}(D)$  to be very narrow distributions, since each size range has its own index of refraction distribution;  $N$  would then be the number of size steps. Another less general but more tractable approach is to let the  $f_{ij}(D)$  be Junge-type distributions. The limits of integration also influence the shape of the scattering function; the summation in (4) thus may include the same size distribution but with various upper and lower limits of integration.

When the particle size distributions appropriate to the problem under consideration have been chosen as well as the indices  $m_j$ , one can then determine the index of refraction distribution by minimizing the difference between the experimentally observed volume scattering function and the sum

TABLE 1. Calculated Relative Indices of Refraction of Suspended Particulate Matter

Location or Material	Calculated Relative Index of Refraction	Reference
Japan Trench		
600 m	1.25	Sasaki et al. [1960]
1500 m	1.25	Sasaki et al. [1960]
2650 m	1.25	Sasaki et al. [1960]
3000 m	1.20	Sasaki et al. [1960]
Sargasso Sea	1.20	Kullenberg [1972]
Mediterranean	1.15 to 1.20	Kullenberg [1972]
Baltic Sea	1.04 to 1.05	Kullenberg [1972]
Phytoplankton	1.02 to 1.04	Carder et al. [1972]
Sargasso Sea	1.05 - 0.01i	Gordon and Brown [1972]
Eastern equatorial Pacific	1.02 to 1.05	Zaneveld and Pak [1973]
Sargasso Sea		
Diameter of $\leq 2.5 \mu$	1.01 - 0.01i	Brown and Gordon [1973a]
Diameter of $> 2.5 \mu$	1.15	
Numerous areas	1.03 to 1.05	Morel [1973b]
Tongue of the ocean	1.15 to 1.20	Brown and Gordon [1973b]

of the chosen components. That is, if the observed volume scattering function is given by  $\beta_e(\theta)$  and  $L$  is the number of data points, the quantity

$$E = \sum_{k=1}^L \left[ \sum_{i=1}^N \sum_{j=1}^M \int_{D_{i1}}^{D_{iu}} A_{ij} f_i(D) \beta(\theta_k, D, m_j) dD - \beta_e(\theta_k) \right]^2 \quad (7)$$

is minimized by adjusting the coefficients  $A_{ij}$ . The technique employed is a rapidly converging descent method described by *Fletcher and Powell* [1963]. The determination of the coefficients  $A_{ij}$  then constitutes a determination of the index of refraction distribution as well as the particle size distribution by means of (5).

#### APPLICATION

A volume scattering function measured by *Kullenberg* [1968] in the Sargasso Sea at a wavelength of 632.8 nm has been analyzed extensively by *Gordon and Brown* [1972] and *Brown and Gordon* [1973a]. In the earlier paper these authors concluded that the best fit (average error of about 20%) to the scattering function was obtained by a distribution of

$$f(D) dD = 3.3 \times 10^4 D^{-4} dD$$

and an index of refraction of  $m = 1.05 - 0.01i$ . The size limits considered were 0.08 and 10  $\mu$ . In the later paper the authors concluded that a best fit (average error of about 19%) was obtained with

$$f(D) dD = 4.83 \times 10^4 D^{-4} dD$$

The indices of refraction used were  $1.01 - 0.01i$  from 0.1 to 2.5  $\mu$  and 1.15 from 2.5 to 10.0  $\mu$ . *Kullenberg's* scattering function thus appeared ideal to test our method, since it would permit comparison with an earlier determination of an index of refraction distribution.

The Mie scattering curves were obtained from an excellent set of tables computed by *Morel* [1973a]. The tables contain volume scattering functions for Junge distributions with parameters  $C$  of 3.1, 3.3, 3.5, 3.7, 3.9, 4.2, 4.5, and 5.0 and for indices of refraction relative to water of 1.02, 1.05, 1.075, 1.10, and 1.15. The integrations in the table are carried out between limits of  $\alpha = 0.2$  and  $\alpha = 200$ , where  $\alpha = 2\pi D/\lambda$ . These values represent diameters of  $D_1 = 0.04 \mu$ m and  $D_u = 40 \mu$ m at the wavelength under consideration. Although these tables do not permit us to vary the limits of integration (which do have a slight influence on the shape of the scattering function) and also do not contain imaginary components of the index of refraction, the choice of scattering functions still permits an excellent fit to the experimental distribution.

The calculated scattering function was thus given by

$$\beta_c(\theta) = \sum_{i=1}^8 \sum_{j=1}^5 A_{ij} \beta_{ij}(\theta)$$

where  $\beta_{ij}(\theta)$  represents a tabulated volume scattering function. The  $\beta_{ij}$  were normalized to  $10^4$  particles/ml.

In our minimization approach for this particular problem, no conditions were imposed on the size distributions. We did not assume that a size distribution for the particulate matter was known. A condition such as that shown in (5) was there-

fore not imposed. The program was free to choose the  $A_{ij}$  that minimize the square of the difference between the observed and calculated volume scattering functions. The program not only determines the index of refraction distribution but also the resultant particle size distribution. The resultant particle size distribution and its components are shown in Figure 1. The components as well as the result of the 40-component fit using the Fletcher-Powell minimization technique are shown in Table 2. Although all 40 components were found to be present, only 7 contribute significantly to the calculated volume scattering function. These are components with a relative index of refraction of 1.05 and exponents of 3.5, 3.7, and 3.9, components with an index of 1.075 with exponents of 3.7 and 3.9, and components with an index of 1.15 with exponents of 3.5 and 3.7. Components with indices of 1.02 and 1.10 were essentially not present. The calculated and observed volume scattering functions and the components of the calculated scattering function are shown in Figure 2. The average error was only 13.4%, and the maximum error was 28.3%. The experimental error was estimated to be 30% by *Kullenberg*.

The major contribution to the calculated volume scattering function is due to the particles with an index of refraction of 1.15, especially at the larger angles. At the smaller angles the contribution of the 1.05 component rapidly increases. The 1.075 component contributes but little at any angle, and ignoring this component would not greatly affect the calculated scattering function. The most striking result is that so few components are chosen by the minimization program, es-

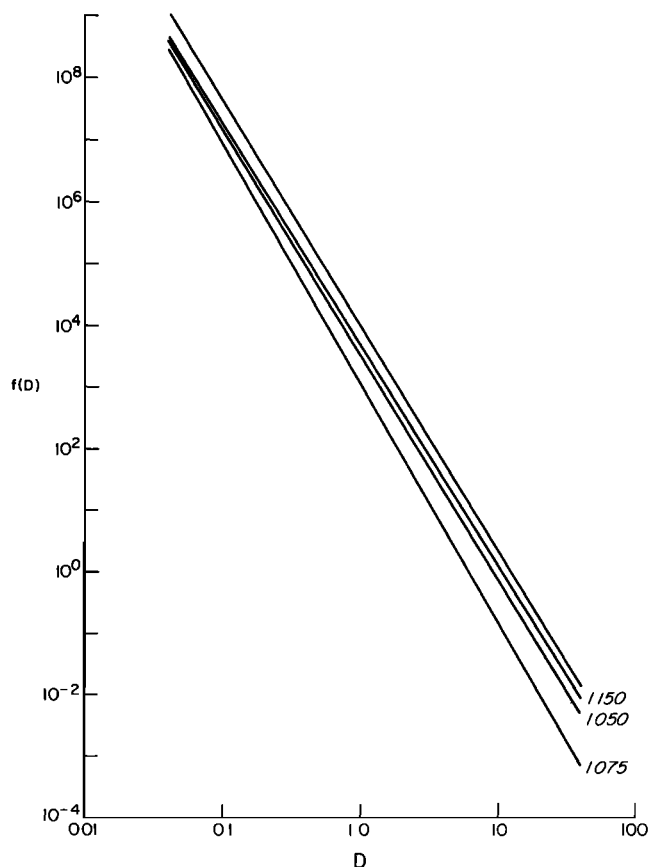


Fig. 1. The calculated total particle size distribution (uppermost curve) for the scattering function shown in Figure 2 and the particle size distributions for the components with indices of refraction of 1.050, 1.075, and 1.150.

TABLE 2. The Values of the Coefficients of the 40 Components Providing the Best Fit to a Volume Scattering Function Observed in the Sargasso Sea

Exponent of Junge Distribution	Relative Index of Refraction				
	1.02	1.05	1.075	1.10	1.15
3.1	$1.64 \times 10^{-7}$	$1.45 \times 10^{-7}$	$4.92 \times 10^{-7}$	$2.92 \times 10^{-13}$	$9.33 \times 10^{-9}$
3.3	$5.83 \times 10^{-7}$	$1.30 \times 10^{-7}$	$1.65 \times 10^{-12}$	$1.69 \times 10^{-10}$	$1.43 \times 10^{-8}$
3.5	$1.39 \times 10^{-16}$	$4.41 \times 10^{-2}$	$2.88 \times 10^{-10}$	$4.06 \times 10^{-9}$	$1.10 \times 10^{-1}$
3.7	$7.74 \times 10^{-21}$	$6.30 \times 10^{-2}$	$6.02 \times 10^{-3}$	$3.49 \times 10^{-12}$	$6.27 \times 10^{-2}$
3.9	$2.60 \times 10^{-14}$	$4.98 \times 10^{-3}$	$3.06 \times 10^{-2}$	$2.31 \times 10^{-20}$	$2.13 \times 10^{-8}$
4.2	$1.80 \times 10^{-12}$	$1.42 \times 10^{-9}$	$9.75 \times 10^{-13}$	$1.28 \times 10^{-14}$	$6.54 \times 10^{-9}$
4.5	$1.79 \times 10^{-11}$	$1.68 \times 10^{-14}$	$7.34 \times 10^{-8}$	$4.17 \times 10^{-8}$	$3.81 \times 10^{-11}$
5.0	$4.58 \times 10^{-24}$	$7.02 \times 10^{-13}$	$3.30 \times 10^{-12}$	$3.12 \times 10^{-11}$	$8.70 \times 10^{-10}$

The coefficients are normalized to  $10^4$  particles/ml greater than  $1 \mu$  in diameter.

pecially since the shapes of the component scattering functions do not change very much when one of the parameters is varied.

Since the slope of the Junge distribution is different for the components with indices of refraction of 1.05, 1.075, and 1.15, the percentages of the total represented by a given index of refraction are a function of size. These percentages are shown in Figure 3, which indicates that the high-index particles constitute a larger percentage of the total with increasing particle size. The total particle size distribution had an exponent of about 3.6, which falls in the range (3.4–4.1) observed by *Gordon and Brown* [1972] for the particulate matter in the surface water of the Sargasso Sea. The amount of particles per milliliter larger than  $1 \mu$  was found to be  $3.2 \times 10^3$ . This quantity is a factor of 4 less than the minimum observed by *Gordon and Brown* [1972]. As the scattering function and the particle size distribution were measured neither at the same time nor at the same location, this result lies entirely within expected fluctuations in the particle concentration.

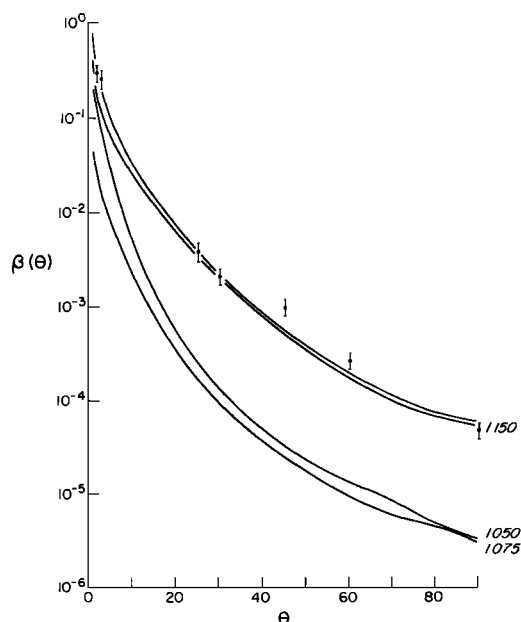


Fig. 2. The experimental volume scattering function observed by Kullenberg (dots), the calculated 40-component best fit (uppermost curve) to the experimental curve, and the contributions to the total scattering function by the 1.050, 1.075, and 1.150 index of refraction components.

The present results clearly indicate a bimodal distribution, with a 1.15 index of refraction component and a 1.05–1.075 index of refraction component; all other indices are unimportant. This result is not unexpected, since previous calculations summarized in Table 1 showed a similar distribution.

One interpretation of this observation is that the organic matter present had an index of around 1.050 and the inorganic and organic skeletal material had an index of 1.15.

Our average error was only about 13% as compared with 19% for the model of *Brown and Gordon* [1973a]. Their contention that the large particles must consist mostly of inorganic material is supported by our results. Attaching physical significance to a best fit must be done with great care, however, since the many independent parameters involved in the determinations of volume scattering functions can be combined in several ways to obtain calculated scattering functions that are similar to observed ones.

As our calculations indicate that almost no particles with an index of 1.02 were present, the choice of an index of refraction of around 1.01 for organic material is probably too low. Furthermore, organic matter contains skeletal material that has an index of refraction similar to that of inorganic matter. Our results show that the index of refraction for skeletal and inorganic material must be about 1.15 or higher. It is thus likely that a pure plankton culture would show a bimodal index of

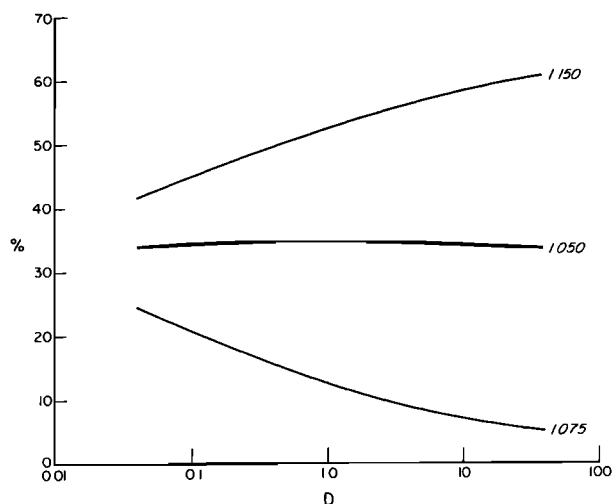


Fig. 3. The percentage of the total number of particles as a function of particle diameter in microns with the component index of refraction as a parameter.

refraction distribution, most material (watery protoplasm) having an index of refraction between 1.02 and 1.05 and a smaller fraction of the material having an index of refraction around 1.15.

#### DISCUSSION AND CONCLUSIONS

Although the principal purpose of this paper is to present a method for determining index of refraction distributions, the bimodal index of refraction distribution deserves some comment. Phytoplankton consist of hard calcareous or siliceous shells and soft watery protoplasm. The hard shells probably have a relative index of refraction in the neighborhood of 1.15, whereas the protoplasm has a relative index of refraction of 1.01–1.05. If phytoplankton are present, one might expect to find particles with an index of refraction that is some value between the indices for hard shells and protoplasm. Optical properties do not, however, average in a linear fashion. A particle that consists for 50% of material with an index of 1.04 and for 50% of material with an index of 1.10 does not have a scattering function like that of a particle with an index of 1.07. According to our present results such a particle appears to scatter more like two particles, one with an index of 1.04 and one with an index of 1.10. This hypothesis can be tested by examination of the scattering functions of phytoplankton cultures and the determination of their index of refraction distributions.

The observation that most of the larger particles appear to be inorganic or consist of phytoplankton shells is contrary to expectation. Yet the same result was obtained by *Brown and Gordon* [1973a], using a different method. These authors have also sought to explain the observation. Of all the scattering functions analyzed by using our method, the Sargasso Sea sample is the only one in which the inorganics dominate the large size ranges. No general conclusions concerning particulate matter in the ocean should thus be drawn from the analysis of the Sargasso Sea sample. The Sargasso Sea sample provides, however, a useful test of our method, since an unusual observation was confirmed.

The choice of component particle size distributions is governed by many factors. Ideally, one would choose a large number of very narrow monodisperse size distributions. Owing to the computation time involved one is forced to search for alternative but reasonable size distributions. Very dense populations of organic material, such as can be found in a plankton bloom, display a distribution that is normal or skew normal. The assumption of a Junge distribution for such a population will lead to erroneous results.

It has not been possible with present instrumentation to separate the organic from the inorganic fraction before a reliable particle count is made. If as is the case with the Sargasso Sea example the total particle size distribution is a Junge type, it is thus reasonable to assume that the components also have this type of distribution.

By means of the Fletcher-Powell minimization technique it is possible to determine the best fit to an observed volume scattering function by using a large number of component scattering functions. By judiciously choosing the component scattering functions one can interpret the fit in terms of the index of refraction distribution of the suspended particles and

the size distribution of the suspended particles. If the size distribution has also been determined experimentally, the difference between the calculated and the observed size distribution may also be minimized.

The method was applied to a volume scattering function observed in the Sargasso Sea by using 40 components covering the most likely indices of refraction and particle size distributions. The method was successful in predicting a previously observed particle size distribution and index of refraction distribution. The resultant index of refraction distribution was bimodal, with a component in the range 1.05–1.075 and a component of 1.15. The total particle size distributions had an exponent of 3.6 in the Junge distribution.

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