

The Effect of Particles on Light Scattering in the Sea*

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Abstract: Relationships of light scattering to particle concentration and size distribution were experimentally determined to follow theoretical findings, both in laboratory suspensions and in a variety of conditions in the natural marine environment. Confirmation was obtained by electronic particle counts paired with scattering measurements that light scattering provides a measure of concentration of particle cross-section in the sea. The use of light scattering as a measure of particle bulk concentration is shown to require constancy of the distribution of particle size, which is met under prescribed circumstances in the upper part of the sea. A simple test can indicate the constancy of the size distribution. A tentative estimate is made of the effect particles have on the inherent properties of open ocean water.

1. Introduction

Although interactions between light and particle ensembles have stirred interest in a wide variety of the physical and biological sciences, light scattering measurements coupled with determination of particle characteristics in the sea have been limited. In particular, work on size and character of optically active particles has been restricted by the ephemeral nature of marine particles and by the lack of non-interfering methods of measurement. Recent refinements of electronic counting have been made possible limited work at sea on particle size distributions but there is still no direct *in situ* method available. Optical methods, on the other hand, offer the possibility of non-interfering, *in situ* measurement of some aspects of particle incidence in the sea. The purpose of this work is to derive through physically and oceanographically defined measurements the relationship of light scattering to size distribution and concentration of natural ensembles of particles and to specify conditions under which light scattering can be used to measure particle concentration.

A few investigators who have worked with both optics and particle characteristics (JERLOV,

1955; PICKARD and GIOVANDO, 1960; OCHAKOVSKY, 1966; TUCKER *et al.*, 1969; BEARDSLEY *et al.*, 1970; CARDER, 1970; CARDER *et al.*, 1971; PAK *et al.*, 1971) reach conflicting conclusions on the optical predominance of particle size classes. Of these, only OCHAKOVSKY combined optical measurements (volume attenuation rather than scattering) with bulk measures of particle load as well as size distribution measurements.

2. Theory and assumptions

The volume scattering function and its integral, the volume scattering coefficient, comprise one of the three inherent optical properties of water, the others being the volume absorption and volume attenuation coefficients.

Nearly every observation of light scattering in natural waters suggests control by particles of refractive index close to that of water and of dimensions large compared to the scattered wavelengths: the scattered light exhibits an intense forward component and independence of wavelength over much of the visible spectrum. VAN DE HULST (1957) who has summarized scattering theory in his classic reference work, refers to scattering from particles with these characteristics as "anomalous diffraction." The character of equations derived by VAN DE HULST from the MIE (1908) solution for scattering of a monochromatic plane wave incident on a homogeneous, isotropic sphere shows that

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the low scattering efficiency of particles below about $0.5 \mu\text{m}$ radius renders them optically ineffective at refractive index of 1.1 and measurement wavelength of 500 nm. Theoretical and experimental work by PENNDORF (1953), HODKINSON (1963), and HOLLAND and GAGNE (1970) shows that for polydispersed suspensions of large particles the optical cross section is directly related to geometric cross-section, even for irregularly-shaped particles oriented at random. This suggests that an effective area coefficient can be applied to measurements on natural particle ensembles.

For any given distribution of particle size, the area coefficient for the particle ensemble, $\bar{Q}$, takes on a constant value and may be taken outside the integral to be used as a bulk coefficient. The equation relating scattering to particle cross-section then becomes

$$s = \bar{Q} \int_0^\infty N \pi r^2 dr \quad (1)$$

Under polydisperse conditions $\bar{Q}$ is dependent only on changes in refractive index and on changes of the size distribution below particle radii of about $2 \mu\text{m}$ under normal measurement conditions.

The same argument can be used to justify the substitution of a composite or "effective" index of refraction for the individual index when particles having a variety of indices are under consideration, provided that such an effective index does not vary with particle size. Although some previous results are consistent with an increase in composite refractive index with depth in the sea over a large depth range (BURT, 1958; PAK *et al.*, 1971), the restricted depth range used in the present work (0 to 100 m) is unlikely to span any significant range of refractive index values. Further, the work was mainly conducted offshore where more constant proportions of materials of differing origin diminishes the effect of refractive index on variation of scattering.

The refractive index of individual components of natural particle ensembles may not vary widely in the sea. BURT (1952) indicated that the relative indices of both organic and in-

organic materials of Chesapeake Bay water clustered closely around 1.15. While natural particles can have relative indices varying from 1.0 to 1.4 (JERLOV, 1968), the relative index for mineral substances has been estimated at a constant 1.17 by PAVLOV and GRECHUSHNIKOV (1966) and that of biological materials ranges rather narrowly between 1.03 and 1.06 for bacteria, plant and yeast cells (WYATT, 1968; LATIMER *et al.*, 1968; BRYANT *et al.*, 1969) and 1.10 for spores (LEWIS and LOTHIAN, 1954).

It seems clear from these considerations that the effect on light scattering from particle concentration and size distribution will be greater than the effect of variations of the composite refractive index over the restricted depth interval considered here. On this basis, the composite refractive index of particles under consideration is here assumed to be close to that of sea water and constant over the water masses where measurements were made.

The volume scattering coefficient s , defined in terms of particle ensemble characteristics by Equation (1), can also be defined optically summation of the scattered fluxes in every direction (4π steradians) from the irradiated volume containing the particle ensemble:

$$s = \int_0^{4\pi} \sigma(\theta) d\Omega$$

The term $\sigma(\theta)$ is the volume scattering function, defined by the equation

$$\sigma(\theta) = \frac{I_e(\theta)}{E_e V}$$

where $I_e(\theta)$ is the intensity of scattered light the direction θ , E_e is the irradiance on the scattering element of volume V . The volume scattering function thus describes the flux scattered through a unit solid angle Ω in the direction θ per unit of incident irradiance, per unit of incident irradiance, per unit volume of the scattering element.

The constancy of the slope of the forward part of the volume scattering function and the absence of extremums of the function in the upper layers of the sea has become apparent through

the work summarized by DUNTLEY (1963), and through individual investigations of JERLOV (1961), HINZPETER (1962), SASAKI *et al.* (1960), OCHAKOVSKY (1966), and SPILHAUS (1968). These suggest that scattering at any of a wide range of forward angles can be specified from that at a single angle, and that the total volume scattering coefficient, s , can be similarly approximated. The latter has been started as a hypothesis by JERLOV (1951) and suggested by the results of a number of studies (JERLOV, 1961; TYLER, 1961; HISHIDA, 1966). Until recently the scarcity of adequate scattering data for the important extreme forward angles in sea water had not permitted a test of the hypothesis. SPILHAUS (1968), using observations from 30° to 130° estimated that the partial scattering coefficient over that interval could be estimated by $\sigma(45^\circ)$ or $\sigma(30^\circ)$ with less than 6% error. MORRISON (1970a, b) has supplied the necessary data at extreme forward angles to provide a test of the adequacy of representing s by measurement at a single forward angle. Using a co-axial scattering meter based on the experimental design used by DUNTLEY (1963) in a New Hampshire lake, MORRISON (1970a, b) measured scatterance at 0.20° , 0.62° , and 1.26° and, with another instrument, from 25° to 140° . His test of the hypothesis confirms that of SPILHAUS (1968): very high correlation coefficients of the regression of s on $\sigma(30^\circ)$ or on $\sigma(45^\circ)$, both greater than 0.97, indicate the adequacy of representing s by any value of $\sigma(\theta)$ for the range $20^\circ \leq \theta \leq 60^\circ$.

The shape of Morrison's $\sigma(\theta)$ function is substantially that given by DUNTLEY (1963) who combined his own low angle lake data with that of KOZLYANINOV (1957) from the East China Sea. Because the integrated function (yielding s) frequently was much larger than expected from Morrison's independent estimates of volume absorption and attenuation, however, the exact relationship between s and $\sigma(\theta)$ at a given value of θ still cannot be specified even though the relationship was shown to be close.

Through theoretical considerations and the substantiating observations noted above, the following assumptions and simplifications can

be made to validate light scattering as a measure of particle cross-section concentration.

That single scattering only need be considered in the sea over path lengths of less than 1 m and that scattering effects are additive.

That an area coefficient $\bar{Q}$ relating particle area to light scattering can be considered a constant.

That particles having irregular shapes scatter as spheres having the same cross-section and equivalent radius or volume.

That a constant, composite value of relative refractive index can be assigned to particle ensembles in the upper layers of the sea.

That scattering at a given forward angle represents total scattering, and hence concentration of particle cross-section as well.

3. Scattering by experimental suspensions

Because the scattering measurement is potentially diagnostic of particle size distribution and of particle concentration, laboratory experiments were conducted to separate these two variables as well as to confirm by observation and extend to other angles Deirmendjian's (1963) derivation from theory that scattering at $\theta=45^\circ$ is directly proportional to the particle cross-section concentration of polydispersed ensembles. Cross-section concentration is the combined expression of size distribution and volume concentration. It was to be hoped that the light scattering meter would be sensitive to both parameters the range of environmental variations to be encountered.

Aqueous suspensions were required in which particle size distribution could be varied while maintaining a constant value of particle concentration, and in which concentration could be varied without changing size distribution. Controlled artificial suspensions were suggested because size distribution, if not concentration, would have been difficult to manipulate using natural water suspensions. An additional requirement was that these suspensions be optically polydispersed *i.e.*, that a sufficient range of size present in such quantities as to produce a smooth, monotonic scattering function. If particle size fractions were to be changed to

satisfy the separation of volume and projected area variables, it would also be necessary that Mie lobes, produced by scattering from monodisperse suspensions, be absent because the scattering signal at a given angle would otherwise depend more on the projected area of one size fraction than on that of the ensemble.

These requirements were met through the use of polystyrene latex sphere suspensions. The spheres are of nearly uniform, single sizes and are commercially available (Dow Chemical Corp.) Their uniformity facilitates analysis of concentration because only numbers per unit volume of suspension need be assessed.

Stock suspensions were made up for each of five sphere sizes (1.10, 1.86, 5.13, 7.25, and $15.95\text{ }\mu\text{m}$ diameter) by dilution of the original concentrate with filtered, deionized water. This size range was intended to provide sufficient polydispersion, when combined, to satisfy the optical requirement stated above. Aliquots of these stocks were analyzed in a Coulter equipped with a $30\text{ }\mu\text{m}$ -diameter orifice. Counts obtained were converted to volume concentration and to concentration of projected area.

Given those stock concentrations, combinations were made to yield four working suspensions having equal volumes of particles with as wide a variety of total particle cross-sections as possible while maintaining polydispersion. The working suspensions were then used to determine the response of the σ -meter to variation in particle cross-section.

The working suspensions each contained a volume concentration of $32.01 \times 10^3\text{ }\mu\text{m}^3/\text{cm}^3$ and projected area concentrations of 15.87×10^3 , 10.37×10^3 , 8.03×10^3 , and $5.65 \times 10^3\text{ }\mu\text{m}^2/\text{cm}^3$. Polydispersion was enhanced because particle counts indicated that some of the latex spheres were in doublet and triplet form; these were accounted for in the calculations for preparation of the working suspensions.

The working suspensions were submitted within a short time after their preparation to the same light scattering meter, referred to as the σ -meter, as was used at sea. The σ -meter (described in the next section), which measures at 20° off the forward axis, was fitted with a closed cuvette to contain the suspensions.

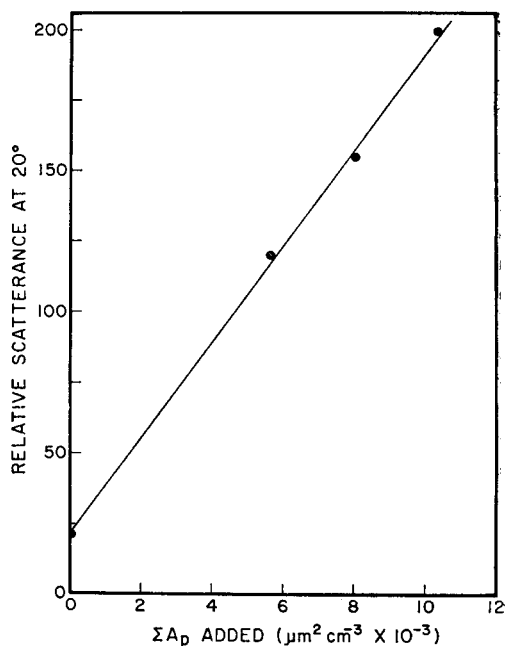


Fig. 1. Response of the σ -meter to variation of particle cross-section concentration at constant particle volume concentration.

Previous use of the cuvette showed that stray light was not detectable, as both projector and receiver were aimed into blind light traps beyond the sensed volume element of the projector beam. The σ -meter was standardized and operated in the same way in the laboratory as at sea. Results of these determinations are shown in Figure 1.

The suspensions were also submitted to a general-angle laboratory scattermeter made available by the Visibility Laboratory, Scripps Institution of Oceanography, which was capable of measurement at angles from $12\text{--}1/2^\circ$ to 160° off the beam axis. The optical geometry of this instrument is not very well defined, due in part to the uncertain optical quality of the beaker used to contain the sample, through which the instrument must both illuminate and sense the volume element. The filter combination used resulted in a spectral sensitivity approximately equivalent to that of the $\sigma(20^\circ)$ meter. These measurements were made at 5° intervals from 15° off the forward

axis of the beam to 100° to check the smoothness of the scattering function, *i.e.*, to check that no pronounced Mie lobes were present which would vitiate the test of scattering response to particle cross-section.

Results of these measurements, shown in Figure 2, indicate that no pronounced Mie lobes were present in the suspensions—maximums and minimums were not detected. There was, however, a change in the slope of the $\sigma(\theta)$ function that shows that the dispersion of particle sizes was not broad enough to generate a perfectly smooth function. These may be seen to be approximately equivalent for each of the suspensions so that the test of this hypothesis remains valid.

Plots of scatterance at various measured angles are shown in Figure 3 as a function of particle cross-section. At most angles the relationship is linear and slopes are equivalent within instru-

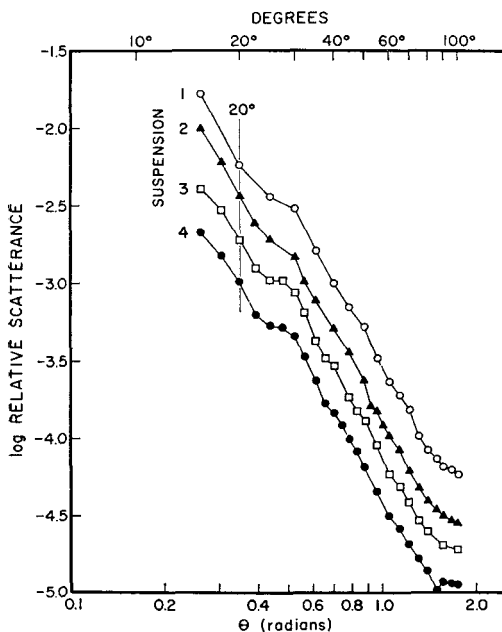


Fig. 2. Light scattering function for each of four known polydispersed suspensions. Suspensions 1 to 4 respectively contained 15.87×10^3 , 10.37×10^3 , 8.03×10^3 and $5.65 \times 10^3 \mu\text{m}^2$ of particles per cm^3 of suspension.

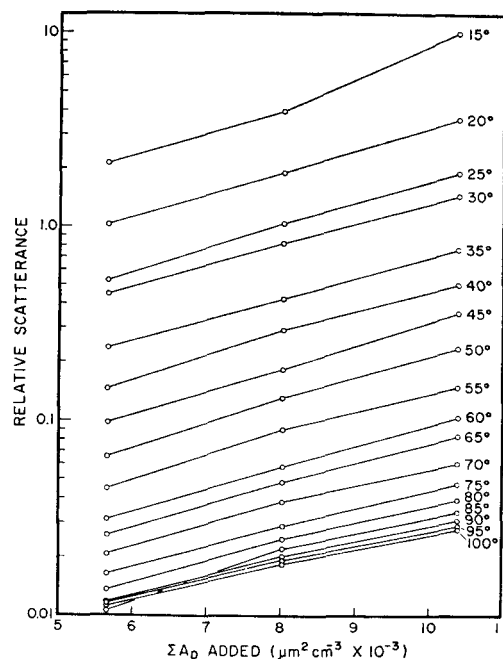


Fig. 3. Light scatterance as a function of particle cross-section concentration at angles shown.

ment error despite the change in slope noted above. One of the working suspensions (containing $15.87 \times 10^3 \mu\text{m}^2/\text{cm}^3$) was subsequently found to have become contaminated by rod-shaped bacteria. Data for this suspension consequently were discarded although its scattering function is shown in Figure 2 as suspension 1.

This determination of the effect of total particle cross-section on scattering constitutes a test of the CLEWELL (1941) hypothesis that at constant mass (or volume) concentration, scattering from suspensions of particles large compared to the scattered wavelength will be directly proportional to the equivalent reciprocal diameter of the particle ensemble. The linear relationship observed between scattering at 20° (as well as at other angles) and total particle cross-section at constant particle volume constitutes confirmation of the hypothesis, as this is equivalent to the reciprocal diameter relationship.

The determination also expands Kalle's (1939)

hypothesis, and Deirmendjian's (1963) derivation from scattering theory which state that scattering at 45° is directly proportional to particle cross-section concentration. Direct proportionality at other angles, including 20° , also occurs whenever polydispersed suspensions cause the scattering.

The concentrations of particle cross-section in the working suspensions used in these tests were three to 10 times more dilute than those observed at sea over a comparable range of particle sizes (OWEN, 1972). These latter concentrations will not give rise to appreciable multiple scattering, however, and it is therefore apparent that the laboratory results apply also to field measurements. The conclusion may be drawn that the scattering measurement is sensitive to environmental changes in particle cross-section concentration.

Testing the sensitivity of the measurement to particle mass or volume concentration was somewhat less complicated. The test required that volume of particles be varied without changing the size distribution, which could be accomplished by serial dilution of an initial suspension of particles having any given distribution of particle sizes.

An initial suspension was constructed using seven different sizes of the polystyrene latex spheres to insure a smoothed $\sigma(\theta)$ curve. The initial concentration of particle volume was well above that observed on the particle-rich stations of Cruise 48. The water that was to be used to dilute the initial suspension was filtered twice through membrane filters having $0.3 \mu\text{m}$ pore diameters. A small amount of the initial suspension then was added to the diluting water to mask the optical effects of any particles still present after filtration so that the size distribution of particles in both the initial and final suspensions would be optically identical.

Scattering by the two suspensions was measured shortly after their preparation, again employing a cuvette fitted to the σ -meter to contain the samples. A series of eight sequential dilutions was then prepared by volumetric combinations of the two suspensions and submitted to the σ -meter, rinsing the cuvette between each measurement. The results of these measure-

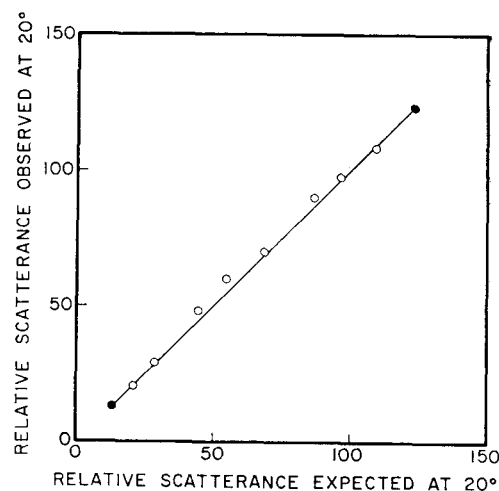


Fig. 4. Observed vs. expected scatterance, $k\sigma(20^\circ)$ from serial dilutions of an initial suspension (upper solid circle) by a dilute suspension (lower solid circle).

ments are shown in Figure 4.

The terminal solid circles represent the observed scattering values from the initial suspension and the diluting suspension. In the absence of multiple scattering, the expected scatterance is that which would result from measurement of any given combination of these two suspensions if the σ -meter response is linear with change in volume concentration of particles. The fit of the observed values, plotted as open circles, indicates that multiple scattering effects were negligible over the range of concentrations used, and that the σ -meter response to change in volume concentration is in fact linear.

The range of concentrations more than encompassed that observed in the ocean, in terms of $k\sigma(20^\circ)$ values. It may therefore be stated that the σ -meter responds linearly to particle volume concentration at constant size distribution as well to cross-section concentration and is a valid tool for detection of variation of either as they occur in the sea.

4. Location and methods of field measurement

Light scattering was measured at a single forward angle using a simple, submersible instrument, the σ -meter, which consists of a

projector, receiver, and depth transducer mounted on a rigid "optical bench" in a way that the receiver unit senses an optically defined, homogeneously irradiated 0.6 cm^3 segment of the collimated light beam of the projector at an angle of 20° off the forward direction of the beam. This angle was chosen to provide maximum light flux for the detector while still maintaining a well-defined forward angle. The solid angle of the projector beam is $3.58 \times 10^{-4} \text{ sr}$, and that of the receiver beam is $6.83 \times 10^{-4} \text{ sr}$. The light source of the projector is a tungsten-iodide cycle lamp operated at a color temperature of about $2,900^\circ \text{K}$ and is unfiltered.

A monitor cell housed in the projector provides information for regulating lamp flux. The receiver is optically filtered to yield a response peaking at $492 \mu\text{m}$ with half-band width of $37 \mu\text{m}$. The light sensor is a multiplier phototube. Power and signals were set between the ship and the σ -meter via multi-conductor cable so that continuous measurement of scatterance was possible as the σ -meter was lowered through the upper 100m of the sea.

Absolute calibration of the σ -meter has not yet been obtained, so that 20° -scatterance values are reported here in relative units as $k\sigma(20^\circ)$. The constant k , relating the σ -meter signal to scatterance, $\sigma(20^\circ)$, was evaluated at sea. Nearly simultaneous vertical profiles of scatterance at 20° were obtained at two locations in the San Pedro Channel off California using the σ -meter and a carefully calibrated scattering meter described by PETZOLD (1970). Comparison of the two sets of profiles yielded a value for k of $6.82 \times 10^2 \text{ mv m}^{-1} \text{ sr}^{-1}$. Further details of this calibration are provided by OWEN (1972).

Particle mass, or PM, was determined by a modification of the method described by BANSE *et al.* (1963), in which the dry mass of particles retained on cellulose membrane filters is determined by the weight change accompanying filtration of known volumes of sea water. The method differs from that of BANSE *et al.* in that control filters subjected to the same environment as sample filters (except for filtration) are used as counterweights on a Cahn "Gram" Electrobalance in determining filter mass both before and after filtration, so that

mass difference is measured rather than total filter mass or filter plus particle mass. This means that the filter mass is very nearly cancelled by the control filter counterweight; a low weighing scale can thus be used to assess particle mass, resulting in greater sensitivity. Also, because less water need be filtered, the filter size could be reduced. Precision at the 95 % level was found to be $\pm 6.2 \%$ of the mean, based on replicate sample determinations (OWEN, 1972).

Particle counts for measuring dispersion of particle size were made with the Coulter Counter (Model B, Coulter Electronics) using the general procedures of SHELDON and PARSONS (1967) for calibration and operation. This instrument is, in principle, capable of measuring differential particle size distributions in single water samples by repeated counts at predetermined threshold control settings. Its use in this work, however, was relegated to the secondary role, that of providing a dispersion measure that could demonstrate the uniqueness of the effective radius, for two reasons: counts in the important small size categories could not be separated from electronic noise and, second, the range of sizes that could be counted at sea was small.

Sensitivities and threshold settings of the counter were chosen to obtain particle counts in sea water samples in size categories that would give both arithmetic and logarithmic distributions according to particle cross-sectional area, the particle parameter of most direct physical influence on light scattering. Distribution of counts on the arithmetic scale was measured at $1.0 \mu\text{m}^2$ intervals over the range from 2 to $16 \mu\text{m}^2$ (1.6 to $4.5 \mu\text{m}$ equivalent diameters). On the geometric scale, counts were made in intervals of $2^n \mu\text{m}^2$ starting at $n=1$ through $n=10$ to measure over the range from 2 to $1,000 \mu\text{m}^2$ (1.6 to $36 \mu\text{m}$ equivalent diameters). A $50 \mu\text{m}$ diameter orifice was used to count particles up to $6.4 \mu\text{m}$ diameter and a $100 \mu\text{m}$ orifice was used thereafter.

Water samples for PM determinations and particle counts were obtained from several depths predicated on the variation of the scattering profile, which was obtained minutes

before, to characterize optically homogeneous layers and to sample the major scattering maxima and minima encountered. Water samplers were cleansed Lexan (polycarbonate) bottles. Subsequent manipulation of the water consisted of withdrawal into clean glass containers in a gentle, bubble-free flow through a 500 μm pre-filter screen.

Matching particle samples from discrete depths with corresponding values on the vertical profile of scatterance obtained up to 20 minutes previously appeared to have created error because the time intervals between observations represent at least part of the period possible for internal wave displacement under the stability conditions computed from measurements of temperature and salinity profiles. Väisälä frequency, representing the highest frequency possible for stable (non-breaking) internal waves, ranged on the examined stations from 0.0140 to 0.0356 sec^{-1} in the most stable part of the water column. The internal wave period indicated was thus short enough to have changed the geometric depths sampled between casts.

To reduce these matching errors, alternate values of scattering were computed by moving the scattering profile vertically (as would happen over an internal wave cycle) to obtain the closest agreement between extremes of the scattering and particle profiles in the density discontinuity layer. This adjustment was made only when there was a consistent mismatching of the profiles, and the resulting "best-fit" light scattering values have been used separately as alternatives.

5. Results and conclusions

The field program was conducted in part to determine the relationship of light scattering to cross-section concentration of natural particles, to define the variability of scattering, and to determine independently the size and mass of optically-active particles. Measurements were made to 100 m depth at several stations in each of four areas of the Pacific: off Baja California; off La Jolla, California; off Oahu, Hawaii; and in the mid-North Pacific along 140°W to provide a variety of oceanographic conditions. Cruise and stations are

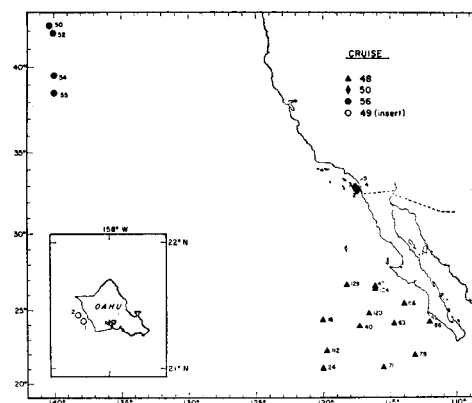


Fig. 5. Cruise and station locations of measurements of light scattering and particle characteristics.

shown in Figure 5.

Confirmation of laboratory results that light scattering at 20° provides a linear measure of particle cross-section concentration was sought under field conditions by comparison of scatterance from σ -meter profiles with particle cross-section concentrations as measured over a truncated size range by the Coulter Counter, as described above. Parallel work (OWEN, 1972) has shown that the frequency distribution of particle size in the open sea is approximately steady, resulting in a quasi-stationary distribution (QSD). This condition implies that size distributions determined over a limited range of sizes are directly proportional to the total, and therefore that particle cross-section evaluated over a truncated interval is proportional to total cross-section concentration.

The degree to which field measurement of $\sigma(20^\circ)$ represents cross-section concentration of natural particles is shown Figure 6. Data pairs are from observations at Cruise 48 stations off Baja California (Figure 5). Regression analysis was performed separately on data pairs from the upper mixed layer and from the underlying pycnocline layer because small differences in the QSD of each layer might have given somewhat different scattering responses. The analysis indicates, however, that the two layers are indistinguishable by these data and that the relationship is described by the solid line of

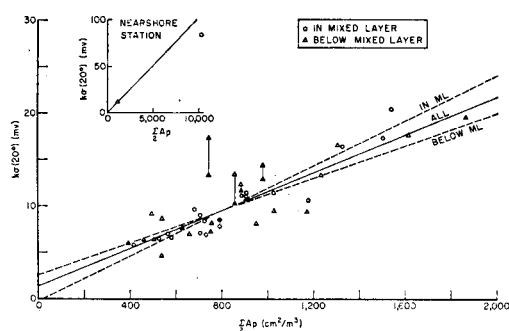


Fig. 6. Response of $k\sigma(20^\circ)$, measured *in situ*, to concentration of particle cross-section, $\sum_{i=1}^{1000} A_{p_i}$, off Baja California. The computed regression equation for all points except those shown in the insert is $k\sigma(20^\circ) = 0.01016(\sum A_p) + 1.42$; the correlation coefficient is 0.898. Solid symbols represent scattering values adjusted by the amount shown to reduce profile matching error described in text.

Figure 6. The computed correlation coefficient of 0.898 connotes a high degree of correlation, considering the error of measurement of cross-section concentration and the profile matching error discussed previously.

Light scattering had a characteristic vertical profile that was somewhat predictable from the physical structure of the water column. In each of the four areas, scatterance typically was nearly constant with depth over the upper mixed layer (defined by isopycnal conditions), increased to a major maximum in the upper part of the pycnocline, then decreased more gradually with depth. At the greatest depths usually measured (about 100 m), scatterance was less than that in the mixed layer.

Within the mixed layer at each location, the observed variance of scatterance was less than 5% of the mean on most stations and exceeded 10% on only three stations out of 28—one station off La Jolla, California and two stations off Oahu, all near land.

The major maximum, usually accompanied by extremums of lesser magnitude, was variable in its magnitude and its relation to other σ -profile features. It was present at 18 of the 24 stations occupied for routine data collection. Where present, the major maximums ranged

in magnitude between 1.1 and 2.3 times the mixed layer scatterance, averaging about 1.6. This maximum exhibited the most variation of any segment of the profile. The first or uppermost maximum was also the major maximum on 16 of the 20 stations exhibiting maximums. The depth to the major maximum was variable, apparently depending on the depth of the upper pycnocline; in no case was the maximum located in the mixed layer but always in the upper part of the pycnocline.

The deepest part of the measured layer (90 to 100 m), well within the pycnocline, exhibited considerably less scatterance variation than either mixed layer or upper pycnocline although part of the deep variation probably was due to the varying proximity of the overlying pycnocline to this fixed depth. Considering also that the euphotic layer is the principal and original source of particles in the open sea, it is to be expected that variation would decrease with depth below it, but it is surprising to see so striking a decrease of the variation so close to the source layer.

Both direct and inverse scattering problems may require knowledge of the connection between light scattering and bulk characteristics of the particle ensembles that occur in natural waters, either to use scattering to measure particle load or vice versa, or to use their ratio, specific scatterance, as a parameter of size distribution or of composite refractive index of scatterers. This knowledge is needed for an important class of problems in oceanography concerned with physical and biological effects of suspended particles.

That scatterance is directly related both in theory and by observation to cross-section concentration implies the sensitivity of scatterance to bulk concentration and to size distribution of the scatterers. Thus the degree to which scatterance expresses particle mass or volume concentration is the degree to which the size distribution is invariable from sample to sample.

Using specific scatterance, as defined by the ratio of 20° scatterance to particle mass concentration, together with independent measures of particle size dispersion, OWEN (1972) determined that ocean areas could be defined which

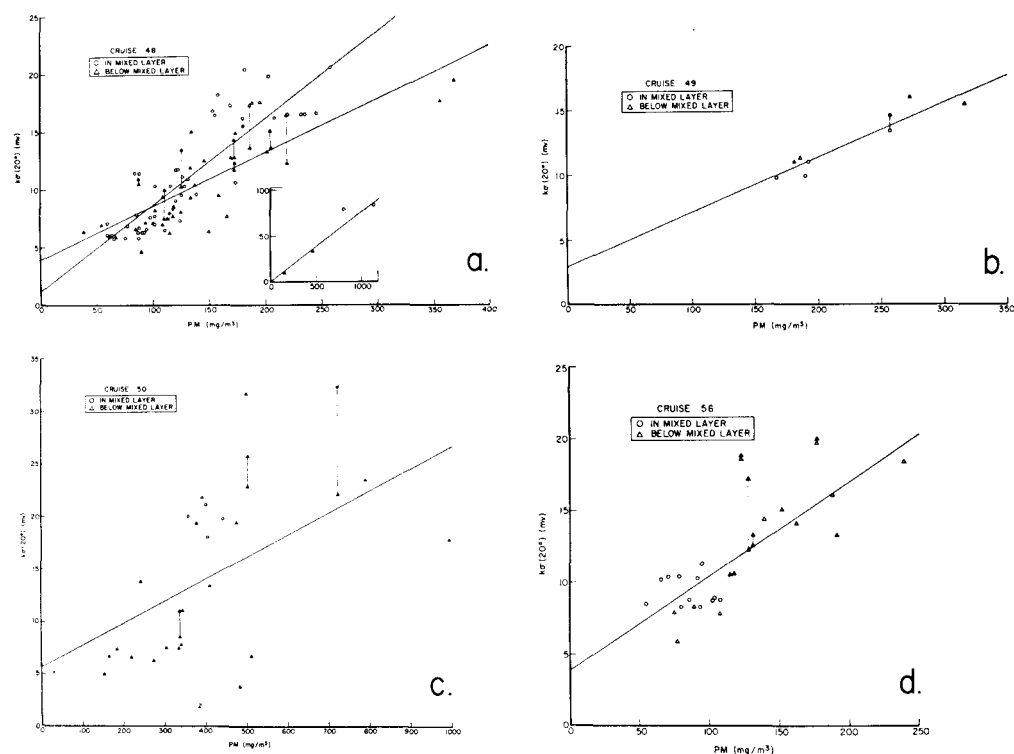


Fig. 7. Relationship of light scattering, $k\sigma(20^\circ)$, to particle mass concentration, PM, a) off Baja California, where $k\sigma(20^\circ)=0.075\text{ PM}+1.2$ and the correlation coefficient $r=0.87$ in the mixed layer, and where $k\sigma(20^\circ)=0.049\text{ PM}+3.6$ and $r=0.83$ in the pycnocline; b) off Oahu, Hawaii, where $k\sigma(20^\circ)=0.044\text{ PM}+2.6$ and $r=0.94$; c) off La Jolla, California, where $k\sigma(20^\circ)=0.025\text{ PM}+4.8$ and $r=0.57$; d) in the mid-North Pacific where $k\sigma(20^\circ)=0.067\text{ PM}+3.8$ and $r=0.77$.

exhibited quasi-stationary distributions of particle size and suggested a single QSD might apply over broad expanses of the open ocean. Near land, however, very different size distributions may occur, and these can be distinguished from one another by specific scatterance.

The relationship of 20° scatterance to particle mass (PM) is shown in Figure 7 for the four areas sampled. Off Baja California (Cruise 48, Figure 7a), nearshore and offshore areas were distinguished statistically by their scatterance: PM relationships but were sufficiently close to warrant their combination in this context. Further offshore, in mid-North Pacific along 140°W the scatterance: PM relationship (Cruise 56, Figure 7d) is similar to that off Baja California in terms both of the mean slope of the relationship and of the variation. This is

perhaps because both locations are imbedded in the same water mass of the North Pacific gyre, although it is surprising to find that the balance of processes affecting this relationship should remain consistent between these widely separated areas.

By way of contrast, the relationship differs widely in nearshore regions, where either a different balance of processes influencing particle size results in a size distribution that alters the scatterance: PM slope, as off Oahu, Hawaii (Cruise 49, Figure 7b), or where an imbalance of processes produces a variety of size distributions as indicated by the scatter of data pairs off La Jolla, California (Cruise 50, Figure 7c).

These relationships confirm that light scattering can be used to measure bulk particle concentration (or *vice versa*) in natural waters.

It is equally apparent, however, that since scattering is sensitive to particle size distribution as well as to concentration, the use of scattering in this way is limited to domains exhibiting a common QSD. Errors of 10 % to 40 % can be incurred in the estimation of particle load by scatterance alone in oceanographically dissimilar offshore areas, and analogous comparisons between nearshore and offshore waters

can err by a factor of three.

Knowledge of specific scatterance is thus required for each such area before scattering alone can be said to represent particle concentration, although only a few measurements of specific scatterance need be obtained to interpret thereafter either scatterance or PM profiles in terms of the other. It follows that previous descriptions of particle content based on scattering measurements (*e.g.*, JERLOV, 1953, 1958, 1964) might be re-examined in view of this argument.

6. Estimates of relative contribution by particle concentration to natural variation of optical properties

Particulate substances variously influence the three inherent optical properties of natural sea water-characterized by the volume scattering, absorption and attenuation coefficients.

Volume scattering is almost completely controlled by particulate substances. Molecular scattering accounts for less than 6 % of the total even in clear ocean waters (JERLOV, 1968; TYLER *et al.*, 1972), and for a great deal less of the forward component of scattered light. This means that more than 94 % of scatterance is due to particles and that any change in concentration of particles of a given size distribution and refractive index will proportionately alter the magnitude of this inherent optical property.

Although light absorption by particles is usually treated as an imaginary component of the particle index of refraction, it is convenient here to consider its contribution to the volume absorption coefficient of natural sea water. The projected area of particles contained in unit volume of water from the upper mixed layer of the open ocean is typically 5 % of the projected area of the total (OWEN, 1972). If these particles absorb as little as 10 % of the light incident on them,* the absorption

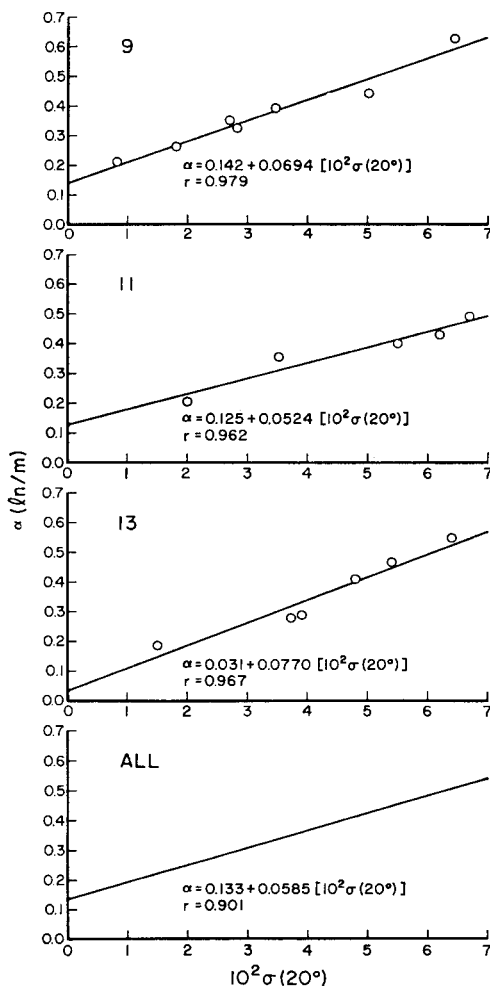


Fig. 8. Relationships of volume attenuation (α) to scatterance at one angle, $\sigma(20^\circ)$, at three stations in the San Pedro Channel. Data are courtesy of T. PETZOLD and R. AUSTIN, Visibility Laboratory, Scripps Institution of Oceanography, and were obtained with the author's help on *Jordan* Cruise 64.

* Calculations from optical cross-sections of particles presented by BRYANT *et al.*, (1969) show that chloroplasts absorb about 46 % of incident light at 480 nm, 28 % at 500 nm. The use of a 10 % value attempts to account for the unpigmented fraction of particle ensembles encountered in nature.

coefficient due to particles would be 0.005 m^{-1} , or about 13 % of observed absorption coefficients (TYLER, *et al.*, 1972) at about 500 nm, where absorption is low. This assumed value appears conservative for the present purpose of indicating the contribution of particles to absorption: *e.g.*, GORDON and BROWN (1972) find a value of 0.01 m^{-1} appropriate in their computations in a light scattering model for Sargasso Sea waters.

The third inherent optical property, the volume attenuation coefficient, is the sum of the scattering and absorption coefficients. To assess the control exerted by particles over this property it is useful to separate the effects of the water substance and its dissolved salts, which are nearly constant, from the effects of "variables" (particles and dissolved organics). At wavelengths where peak transmission occurs, the scattering coefficient accounts for about half of the volume attenuation, of which 6 % is due to water (constant) and 94 % to particles (variable) in clear ocean waters. Of the other half, due to absorption, about 50 % is due to absorption by water (constant), 13 % to particles (variable) and the remaining 37 % to dissolved organics (variable). Recombining these values results in a minimum attenuation coefficient for clear water which has a constant 27 % component due to attenuation by water, and variable components of 54 % by particles and 19 % by dissolved organics.

It is apparent from comparisons available from previous work *e.g.*, DUURSMA, 1965; MENZEL, 1970) that concentrations of dissolved organic substances are proportionately less variable than that of particulates. This qualitative statement reinforces the previous estimate that over half of the volume attenuation coefficient is controlled by particles. The observable variation of volume attenuation must therefore be even more determined by variation of particle characteristics. The partitioning of this variation between particulate and dissolved phases must be made from more definitive data, however.

The relationship of volume attenuation to scatterance at 20° is shown in Figure 8 to

demonstrate the covariation of these properties. The high correlation coefficients indicate (but do not demonstrate) the control of attenuation by scattering, hence by particulate matter. The zero intercept on the ordinate may be interpreted as an estimate of the volume absorption coefficient, although there is no *a priori* reason why absorption should not co-vary with scattering.

References

- BANSE, K., C. P. FALLS and L. A. HOBSON (1963): A gravimetric method for determining suspended matter in sea water using Millipore filters. *Deep-Sea Res.*, **10**, 639-642.
- BEARDSLEY, G. F. JR., H. PAK and K. L. CARDER (1970): Light scattering and suspended particles in the eastern equatorial Pacific Ocean. *J. Geophys. Res.*, **75**, 2837-2845.
- BRYANT, F. D., B. A. SEIBER and P. LATIMER (1969): Absolute optical cross sections of cells and chloroplasts. *Arch. Biochem. Biophys.*, **135**, 79-108.
- BURT, W. V. (1952): Scattering of light in turbid water. Ph. D. thesis, Univ. Calif., Los Angeles, 64 pp.
- BURT, W. V. (1958): Selective transmission of light in tropical Pacific waters. *Deep-Sea Res.*, **5**, 51-61.
- CARDER, K. L. (1970): Particles in the eastern Pacific Ocean: their distribution and effect upon optical parameters. Ph. D. thesis, Oregon State Univ., 140 pp.
- CARDER, K. L., G. F. BEARDSLEY JR. and H. PAK (1971): Particle size distribution in the eastern equatorial Pacific. *J. Geophys. Res.*, **76**, 5070-5077.
- CLEWELL, D. H. (1941): Scattering of light by pigment particles. *J. Opt. Soc. Am.*, **31**, 521-527.
- DEIRMENDJIAN, D. (1963): Scattering and polarization properties of polydispersed suspensions with partial absorption. In M. KERKER (ed.) *I.C.E.S. Electromagnetic Scattering* (Potsdam Conference, 1962). MacMillan Co., New York, pp. 171-189.
- DUNTLEY, S. Q. (1963): Light in the sea. *J. Opt. Soc. Am.*, **53**, 214-233.
- DUURSMA, E. K. (1965): The dissolved organic constituents of sea water. In G. RILEY and G. SKIRROW (eds.) *Chemical Oceanography*. Academic Press, New York, pp. 433-475 (total 712 pp).

- GORDON, H. R. and O. B. BROWN (1972): A theoretical model of light scattering by Sargasso Sea particulates. *Limnol. Oceanog.*, **17**, 826-832.
- HINZPETER, H. (1962): Messungen der Streufunktion und der Polarisation des Meerwassers. *Kieler Meeresforschungen*, **18**, 36-41.
- HISHIDA, K. (1966): On the scattering and the attenuation of light in the sea water. *J. Oceanog. Soc. Japan*, **22**, 1-6.
- HODKINSON, J. R. (1963): Light scattering and extinction by irregular particles larger than the wavelength. In M. KERKER (ed.) *I.C.E.S. Electromagnetic Scattering* (Potsdam Conference, 1962). MacMillan Co., New York, pp. 87-100.
- HOLLAND, A. C. and G. GAGNE (1970): The scattering of polarized light by polydisperse systems of irregular particles. *Appl. Opt.*, **9**, 1113-1121.
- JERLOV, N. G. (1951): Optical studies of ocean waters. *Rep. Swedish Deep-Sea Exped.*, **3**, 1-59.
- JERLOV, N. G. (1953): Particle distribution in the ocean. *Rep. Swedish Deep-Sea Exped.*, **3**, 73-97.
- JERLOV, N. G. (1955): Factors influencing the transparency of the Baltic waters. *Medd. Oceanog. Inst. Goteborg*, **25**, 1-19.
- JERLOV, N. G. (1958): Distribution of suspended material in the Adriatic Sea. *Arch. Oceanog. Limnol.*, **11**, 227-250.
- JERLOV, N. G. (1961): Optical measurements in the eastern North Atlantic. *Medd. Oceanog. Inst. Goteborg, Ser. B*, **8**, 40 pp.
- JERLOV, N. G. (1964): Optical classification of ocean water. In J. TYLER (ed.) *Physical aspects of light in the sea*. Univ. Hawaii Press, Honolulu, pp. 45-49.
- JERLOV, N. G. (1968): *Optical oceanography*. Elsevier, New York, 194 pp.
- KALLE, K. (1939): Bericht 2. Teilfahrt der deutsche nordatlantische Expedition "Meteor", V. Die chemischen Arbeiten auf der "Meteor"-Fahrt, Januar bis Mai 1938. *Ann. Hydrograph. Maritimen Meteorol., Beih. Z.*, pp. 23-30.
- KOZLYANINOV, M. V. (1957): New instrument for measuring the optical properties of sea water. *Trudy Inst. Okeanol., Akad. Nauk SSSR*, **25**, 134-142.
- LATIMER, P., D. M. MOORE and F. D. BRYANT (1968): Changes in total light scattering and absorption caused by changes in particle conformation. *J. Theoret. Biol.*, **21**, 348-367.
- LEWIS, P. C. and G. F. LOTHIAN (1954): Photoextinction measurements on spherical particles. *British J. Appl. Phys.*, **5** (supplement 3), S 71-S 75.
- MENZEL, D. W. (1970): The role of *In situ* decomposition of organic matter on the concentration of non-conservative properties in the sea. *Deep-Sea Res.*, **17**, 751-764.
- MIE, G. (1908): Beiträge zur Optik trüber Medien, speziell kolloidalen Metallösungen. *Ann. Physik*, **25**, 377 pp.
- MORRISON, R. E. (1970 a): Experimental studies on the optical properties of sea water. *J. Geophys. Res.*, **75**, 612-628.
- MORRISON, R. E. (1970 b): Characteristics of the optical volume scattering function of sea water. *J. Oceanog. Soc. Japan*, **26**, 101-107.
- OCHAKOVSKY, Y. E. (1966): On the dependence of the total attenuation coefficient upon suspensions in the sea. U.S. Dept. Commerce, Joint Publ. Res. Serv. Rep., **36**, 816, 16-24.
- OWEN, R. W. JR. (1972): The scattering of light by particulate substances in the sea. Ph. D thesis, Univ. Calif., San Diego, 235 pp.
- PAK, H., J. R. V. ZANEVELD and G. F. BEARDSLEY, JR. (1971): Mie scattering by suspended clay particles. *J. Geophys. Res.*, **76**, 5065-5069.
- PAVLOV, V. M. and B. N. GRECHUSHNIKOV (1966): Some aspects of the theory of daylight polarization in the sea. U.S. Dept of Commerce, Joint Publ. Res. Serv. Rep., **36**, 816, 25-44.
- PENNDORF, R. (1953): On the phenomenon of the colored sun. Air Force Cambridge Res. Center, Tech. Rep. No. 53-7, 41 pp.
- PETZOLD, T. J. (1970): Operation manual for general-angle scattering meter. U.S. Naval Air Development Contract N-62269-67-C-0412. (Xerographed report.) Visibility Laboratory, Scripps Institution of Oceanography, 19 pp.
- PICKARD, G. L. and L. F. GIOVANDO (1960): Some observations of turbidity in British Columbia inlets. *Limnol. Oceanog.*, **5**, 162-170.
- SASAKI, T., N. OKAMI, G. OSHIBA and S. WATANABE (1960): Angular distribution of scattered light in deep sea water. *Records Oceanog. Works Japan*, **5**, 1-10.
- SHELDON, R. W. and T. R. PARSONS (1967): A practical manual on the use of the Coulter Counter in marine science. Coulter Electronics Sales Co., Canada, 66 pp.
- SPIELHAUS, A. F. JR. (1968): Observations of light scattering in sea water. *Limnol. Oceanog.*, **13**, 418-422.
- TUCKER, S. P., R. D. WAER and L. A. YESKE (1969): The correlation of oceanic parameters with light transmissivity in Monterey Bay, California, [abs.]. *J. Opt. Soc. Am.*, 1969 Spring Meeting Program,

- p. 9.
- TYLER, J. E. (1961): Scattering properties of distilled and natural waters. *Limnol. Oceanog.*, **6**, 451-456.
- TYLER, J. E., R. C. SMITH and W. H. WILSON, JR. (1972): Predicted optical properties for clear natural water. *J. Opt. Soc. Am.*, **62**, 83-91.
- VAN DE HULST, H. C. (1957): Light scattering by small particles. John Wiley & Sons, New York, 470 pp.
- WYATT, P. J. (1968): Differential light scattering: a physical method for identifying living bacterial cells. *Appl. Opt.*, **7**, 1879-1896.

海中における光散乱に及ぼす粒子の影響

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要旨: 光散乱と粒子の濃度及び大きさ分布との関係を、理論的結果を参照して、実験室内でいろいろの条件下の自然環境のもとで実験的に決めた。散乱測定と同時に粒子の個数を計測し、光散乱が粒子断面濃度の一つの尺度をあたえることを確かめた。光散乱を粒子の体積濃度の

尺度としてつかうためには、粒子の大きさ分布が定常であることが必要であるが、これは海洋上層のきまった状況下では満足されている。簡単な試みによって大きさ分布の定常性を示すことが出来る。粒子が外洋水の内部性状にあたえる影響についての暫定的な推定をした。

