The Particulate Matter in the Sea as Determined by Means of the Tyndall Meter

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Abstract

The Tyndall scattering has been studied for different material occurring in the sea, such as minerogenic matter, calcareous matter, and some marine organisms. It is shown that the scattering is proportional to the total surface of the particles provided that they are larger than $2\,\mu$. This relationship is practically independent of the concentration. The size distribution of particles in marine waters is discussed with a view to estimate the amount of matter removable by sedimentation.

The use of the Tyndall meter offers a simple and quick procedure for current studies of the amount of suspended matter in natural waters (JERLOV, 1953). A parallel Tyndall beam from an electric lamp is projected into the water sample contained in a small glass flask which is placed in a chamber filled with water. The intensity of the light scattered in a forward direction through an angle of 45° is determined by means of a photomultiplier attachment. During the measurement the flask is kept rotating in order to stir up big particles from the bottom of the flask. The particles are maintained in random orientation by the stirring.

The standardization of the method aimed at ascertaining whether the Tyndall reading really does represent the scattering over all angles except a certain minimum angle in the forward direction. The experimental tests furnished sufficient evidence for this; the ratio between the Tyndall reading and the scattering coefficient as derived from measurements with the transparency-meter showed small variations with the particle concentration.

ATKINS and POOLE (1952) recently published an experimental study of the scattering of light by natural waters. Their findings on the angular distribution of the scattered light indicate that the particle scatter through 45° is nearly proportional to that between 20° and 145° for the different water samples (it was not possible to include angles near 0° and 180°). This should support the result of the above-mentioned standardization but it must be observed that much scattering is limited to small angles, less than 20°.

From a physical point of view it appears satisfactory to express the Tyndall reading I_s in terms of a scattering coefficient s. Properly the relation should be

$$e^{-s} = 1 - k\, I_s,$$

where the constant k is found by the standardization. For small values of s the equation is reduced to

$$s = k\, I_s$$

There is a tendency in favour of this latter relationship even for fairly large values of I_s.

Too much emphasis should not be placed on the absolute values of s, which ultimately depend on the standardization of the transparency-meter (JERLOV, 1953). The minimum angle mentioned is not defined for the transparency-meter and the s-values are thus dependent on its construction (cf. GUMPRECHT and SLIEPCEVICH, 1953).

Particle Size

The scattering by particles small compared with the wave-length is given by the Rayleigh equation, the scatter term of which may be written

$$I_s = \text{const.}\ C\ D^3\ \lambda^4 \qquad (1)$$

where D is the diameter of the particles and C their concentration in the medium.

Rayleigh's Law has been extended to larger particles by MIE (1908) and by JOBST (1925). Their assumption that the particles are homogeneous spheres of known refractive index does not apply exactly to the particulate matter in the sea.[1] It may be inferred, however, that for particles sufficiently large to act as true reflectors, the scattering should be proportional approximately to the square of the diameter. For a dilute system of large particles the scattering may be expressed by the equation

$$I_s = \text{const.}\ ND^2, \qquad (2)$$

N being the number of particles per unit volume.

As the concentration C is proportional to the volume and so to ND^3, equation (2) can be transformed into

$$I_s = \text{const.}\ C/D \qquad (3)$$

This approximate relationship has been verified especially by the experiments of TOLMAN *et al.* (1919).

The Rayleigh scattering according to equation (1) occurs for particles of a diameter less than about one-tenth of the wave-length of light. With increasing particle size the power of the diameter decreases from 3 over 0 to —1 until equation (3) is valid. This implies that the scattering for a given concentration passes through a maximum.

Experiments have been conducted to bridge the gap between equations (1) and (3). STUTZ (1930) has studied suspensions of zinc oxide, and ANDREASEN and his collaborators (1939) have investigated suspensions of solid material in water. As anticipated, these measurements establish the fact that for white suspensions the light scatter has a maximum at grain sizes of the same order as the wave-length of the incident beam.

The significance of the Tyndall reading is realized if equation (2) or (3) is given in the form

$$I_s = K\,S, \qquad (4)$$

where S is the total surface of the particles. The Tyndall reading for large particles thus gives a measure of particle surface (cf. DALLA VALLE, 1948, p. 339). The constant K includes the optical properties of the material (reflectivity, absorption, refraction etc.).

The conclusive results from the experiments mentioned demonstrate that equation (4) is valid down to a size of about 2 μ. Below 2 μ a departure appears which increases toward smaller sizes, and, in addition, the scattering becomes more and more selective.

Particle Colour

The individual particles in a suspension partly scatter, partly reflect, and partly absorb light. These processes exert some selective action on the incident light. The Rayleigh scatter is highly dependent on wave-length [equation (1)]. For large particles the selectivity is chiefly due to their colouring. This effect may be exemplified by some minerogenic suspensions having a high grade of monodispersity (JERLOV and KULLENBERG, 1953). In Table 1 it is shown that the Tyndall reading with red light is 16 % higher than that with blue light for particle sizes 3—12 μ whereas a

Table 1. Ratio of Tyndall reading for red to that for blue. Minerogenic suspensions

Size	I_{red}/I_{blue}
12 μ	1.15
9	1.18
7	1.18
3	1.14
1	0.99

[1] Quite recently, BURT (Tellus, *6*, p. 229) has obtained results with the Mie theory which compare favourably with experimental results for the minerogenic suspensions discussed in this paper.

superposed effect of size appears for particles of one μ.

The selectivity due to colour is generally not important. Thus, the existence of brown or red particles in the sea as indicated by a higher scattering for the red than for the blue is a rare phenomenon.

Distribution of Particle Size in the Sea

So far the development concerns uniform particulate matter. The large range of particle size and the variety in particle material in the sea confronts us with new problems. The suspended matter is minerogenic as well as organogenic. The latter component is to a large extent made up of detrital matter, the living fraction being only some percent of the total organic substance (ARMSTRONG and ATKINS, 1950; FOX, ISAACS and CORCORAN, 1952).

Our knowledge of the distribution of particle size in the sea is rather limited but it is clear that the small sizes predominate in number. This is illustrated by a typical distribution in the surface water found at Bornö Station in the Gullmar Fjord on 1 September 1950 (Table 2). The distribution obtained was a

Table 2. Size distribution of particles in the Gullmar fjord on 1 September 1950

Diameter $D\,\mu$	Number N	Surface ND^2	Volume ND^3
< 2	7,800	8	8
2—4	4,600	40	100
4—10	1,900	90	600
10—20	790	200	3,000
20—40	280	300	8,000
40—100	87	400	30,000
100—200	2	50	7,000
		1,088	48,708

result of microscopic counts performed down to 0.5 μ. In spite of their high number, the contribution of particles below 2 μ to the total surface is negligible.

GOLDBERG, BAKER, and FOX (1952) have studied size distribution of plankton as measured on a molecular filter. From their data it may be derived that sizes of 2 μ contribute less than 1% of the total surface of the plankton. JENKINS and BOWEN (1946) made observations on scattering particles in ocean water and found that most of the light cannot be scattered by particles much smaller than the wave-length of the light. Another significant piece of information is furnished by ATKINS and POOLE (1952). They state: "The measurement with filtered samples show that most of the scattering is caused by particles larger than 1 μ, a small part only being due to particles smaller than 0.2 μ, and relatively little to those of intermediate size."

Similar experiments with filtrations through collodion membranes have been conducted at Bornö Station. The results in Table 3 illustrate

Table 3. Tyndall scattering of unfiltered and filtered samples of surface water at Bornö Station on 20 October 1954

Sample	Tyndall reading
Surface water	840
do, filtered, pore diam. 1.5 μ	52
do, refiltered, p. d. 0.5 μ	60
surface water, filtered, pore diam. 0.5 μ	45

the low percentage of scattering due to particles smaller than 1.5 μ. Filtering must be carried out with utmost care in order to avoid, or to surpress as far as possible the effect of contamination, in particular dust from the vessels and from the air.

It would appear that in the upper strata of the sea the small sizes, below one μ, play a relatively small part in the total surface of the suspended matter. As regards the deep sea the available information on the size distribution is so far quite imperfect but there are several indications that the particles are on the whole smaller than those in the surface layers. From the numerous Tyndall measurements made on deepsea samples the blue scattering by particles comes out 7 % higher than the red, or about the same as for surface samples. This fact alone proves that the selective scattering, which as mentioned begins at a size of 2 μ, is on the whole not significant.

It may be added that some direct observations of the scattering particles in the sea have been made. EMERY (1952) found at middepths in the ocean a considerable number of scattering objects as counted from photographic negatives. Most of them were too small for identification. Similar photographic studies by NISHIZAWA, FUKUDA, and INOUE (1954) revealed the presence of many large objects, mostly above one mm, in coastal waters. They suspect that a disintegration of particles might occur during the process of water sampling.

Correspondence between Tyndall Reading and Particle Distribution in the Sea

Obviously, it is not always possible to include in the Tyndall reading the scattering by occasional large particles. Sometimes a large piece of organic material causes a momentary lighting up in the field of view which effect is not taken into account. Samples from coastal waters often contain relatively large particles to such an extent that an incessant fluctuation appears in the scattering which renders the measurement uncertain, though not impossible. As a rule, the Tyndall reading for oceanic waters gives a satisfactory representation of the average particle content in the flask volume (60 mL) and ultimately in the water bottle (1.2 L) provided that this, when brought onboard, is turned upside down repeatedly, and that the samples are drawn off immediately.

GOLDBERG *et al.* (1952) emphasize that the mass of marine phytoplankton as well as of inorganic suspended material is inhomogeneously distributed in the sea. "The amount of substance collected over a short time interval from a unique location will not necessarily represent conditions in the water region; rather, they may reflect the layering of the water masses or the turbulence of the area."

The discontinuous vertical distribution of particles, which has also been recorded in many transparency measurements, may often find a plausible explanation, say in density variations. Nevertheless it is an indisputable fact as asserted by GOLDBERG *et al.* that irrelevant fluctuations in space and time occur. They suggest that larger samples or integrating collecting devices be utilized to overcome these difficulties.

Considering the Tyndall reading as representative of the particle conditions in the sea the following may be stressed. In studies of the suspended matter its volume or mass is usually concerned. It is obvious that a small number of large particles, even a single large particle, can contribute a considerable part of the total volume (cf. GOLDBERG *et al.*, Table 2). The Tyndall scattering, which records the total surface, is less dependent on size distribution and therefore gives a statistically more satisfactory picture of the particle content in the sea. Thus, the Tyndall method has some advantages which tend to suppress the stated fluctuations in the distribution of the suspended matter.

It is further pointed out that more reliable values are secured by taking average values of readings from various levels in the sea. For instance, the mean particle content between surface and 50 m depth found from Tyndall measurements in the Pacific shows a distribution which is remarkably consistent with the dynamical features (JERLOV, 1953). Another alternative to arrive at a more correct representation would be to repeat the routine water sampling and the Tyndall tests.

Determination of Total Particle Surface

It is of interest to test the validity of the approximate equation (4) for suspensions of different material present in marine waters. This involves a determination of the total surface of the particles which is preferably performed by microscopical studies.

A graded sample of a material is examined under the microscope. Let the number of particles in one litre with the diameter (projected diameter) D_1 be N_1, that of particles with diameter D_2 be N_2 etc. If α_s is the surface shape factor the total surface of the material in one litre will be

$$S = \alpha_s \Sigma N_i D_i^2 \quad (5)$$

Marine particles of a definite shape, for which the shape factor α_s was readily obtained, or else particle samples for which reliable data of α_s were available, were primarily chosen for the preparation of suspensions. The factor α_s generally keeps within the range 2.0—2.9 (for spherical particles $\alpha_s = \pi$). In the samples the number of particles having a certain projected diameter was determined by direct microscopic measurement and counting after which the surface was evaluated according to equation (5). As a rule, particles down to 0.5 μ were visible in the microscope.

For the minerogenic suspensions mentioned a shape factor of 2.2 was used (cf. HERDAN, 1953, p. 232).

Furthermore, two calcite suspensions were prepared from shell fragments in a sediment core which were selected by sedimentation analysis. These suspensions were to some degree graded, the mean diameter being 30 μ and 10 μ respectively. On account of the high flakiness of the particles a fairly low surface factor, 2.0, was estimated.

The behaviour of the marine organisms as scattering objects was studied in some cases. Organisms were selected with a regular shape preferably spherical so that their surface was readily evaluated. It was aimed at representative selection of the material, calcium carbonate, silica, cellulose etc., with which the species are constructed. Two samples of foraminifera, *viz. Pulleniatina obliquiloculata* and *Orbulina universa*, and one sample of radiolaria were obtained by sieving from sediment. The diatom, *Coscinodiscus*, was a living population collected in the sea whereas other suspensions were cultures.

Finally an attempt was made to study a natural suspension, surface water at Bornö Station on 16 June 1953. The constituents were largely minerogenic due to land drainage after heavy rain, but also planctonic and detrital. The large range of sizes rendered the microscopical examination rather laborious. A considerable amount of the minerogenic particles were found to have a size near one μ but their part in the total surface was not important. Some doubt arose as to the plausible shape factor which was eventually estimated at 2.2. In consequence, the evaluated surface value is open to some uncertainty.

Tyndall Measurements

The Tyndall effect was determined for red light (615 mμ) which shows smaller variations with particle material than does the blue light, and which practically does not include any scattering by the water itself.

Most suspensions were gradually diluted with distilled water in order to comprise a scale of different concentrations in the study. The distilled water was produced with due precautions to avoid contamination so that its Tyndall effect was as low as 20. The samples of foraminifera and radiolaria were examined in a single concentration adequately chosen. There was some difficulty in stirring up these big particles even by violent rotation and the readings for them are somewhat uncertain.

The Tyndall effect being read, the counted particles were allowed to settle and the residual scattering due to fine particles or dust was observed. This correction to be deducted from the Tyndall reading was unimportant except for the surface water mentioned. In this case the correspondence between settling time and the lower limit of particles size (0.7 μ) was possibly somewhat misinterpreted.

Relationship between Tyndall Reading and Total Particle Surface

Derived values of total particle surface and Tyndall reading for the uniform minerogenic suspensions are plotted in fig. 1. It is noticed that the curves for sizes 3—12 μ keep together whereas the anticipated departure for the one μsize is considerable. Fig. 1 also shows that the calcite particles produce a relatively more intense Tyndall effect due to their high reflectivity. This is more pronounced for the 30 μ size than for the 10 μ size, the latter particles being to some extent contaminated by a black cover, probably of manganese.

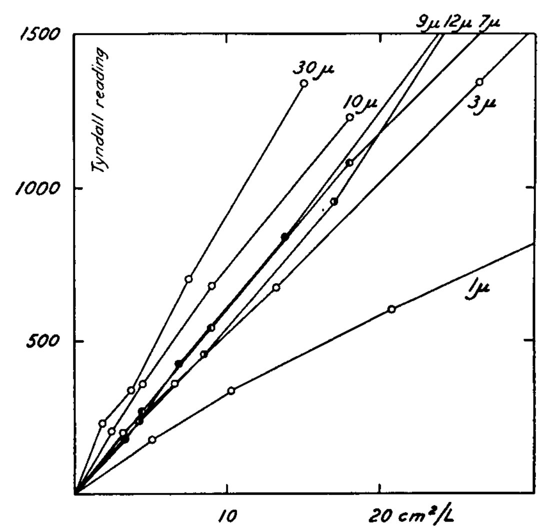

Fig. 1. Tyndall reading and total surface of suspended matter (cm²/L). Uniform minerogenic suspensions with average diameters of 1, 3, 7, 9, and 12 μ, and calcareous suspensions (to the left) with average diameters of 10 and 30 μ.

The entire group of curves clearly demonstrates that the actual relationship is practically independent of the concentration. Thus it is not vitiated by multiple scattering. In other words, the Tyndall reading is proportional to the number of particles per unit volume for the highest concentrations ordinarily attained in the sea.

Table 4. The ratio, *K*, of the Tyndall reading to the total surface of suspended matter

Suspension	*K*
minerogenic 1 μ	32
3	54
7	60
9	59
12	56
calcareous 10 μ	74
30	90
Orbulina universa	79
Pulleniatina obliquiloculata	73
Radiolaria	102
Coscinodiscus	98
Chlamydomonas	26
Prorocentrum micans	30
Natural suspension of surface water at Bornö Station on 16 June 1953	80

Table 4 contains the derived values of K (equation (4)), also for the biologic samples. The Tyndall effect was relatively high for particles constructed with calcium carbonate or silica whereas the green alga, which besides cellulose contains an abundance of chlorophyll, produced a low scattering. Proportionality between Tyndall reading and number of particles also holds for these biologic samples. The plotted value for the surface water though uncertain fits rather well into the scheme. These characteristics of organisms have an interest of their own, and Tyndall measurements may be useful for instance in studies of plankton cultures.

The question arises whether a mean value of the factor K could be chosen so as to obtain from the Tyndall reading the total surface of all suspended matter of the minerogenic material and the organic material, the living as well as the detrital part. This presumes that the water sample shows a normal variety of particles. One must reckon with restricted variations in such a normal distribution but situations with predominance of a certain particle type must be left out.

The large span of the curves in fig. 1 and of the K-values in Table 4 does not at first sight seem to lead to a representative average. The deviations from a mean value would be almost $\pm$ 50 %, partly variations due to size and chiefly variations due to particle material.

The presence of fine particles, which produce a lower scattering than equation (4) accounts for, would tend to the total surface becoming underestimated. The above deliberations suggest that fine particles below one μ in the sea contribute little to the total surface though doubt still exists about the size distribution in deep waters. We are thus aware that the surface as directly proportional to the Tyndall reading of a water sample may be underestimated if fine particles dominate. The method must be taken to suffer from this limitation.

As regards the constituents of suspended matter ordinarily occurring in the sea, the preponderance of forward scattering indicates a large amount of rather big, transparent particles (JERLOV, 1951; ATKINS and POOLE, 1952). Transparent is here taken in the sense that particles reflect and absorb a small part of the incident light. It is manifest that detrital matter is an important component in the sea. This is chiefly made up of resistant remains of organisms such as chitin-, cellulose-, or ligninlike substances (SVERDRUP, JOHNSON and FLEMING, 1942) which are not apt to produce a high Tyndall effect. On the other hand, ARMSTRONG and ATKINS (1950) found that the ignited residue of suspended matter in the English Channel contained on an average 40 % silica and 20 % calcium carbonate which substances show a higher scattering.

It seems reasonable to conclude that on account of the great variety of particles in an ordinary water sample the Tyndall effect produced by it would be of medium intensity. We may venture an estimate that the Tyndall reading for red multiplied with a constant factor will give the total surface of suspended matter with an accuracy of at least $\pm$ 30%. In this connection it may be mentioned that the best alternative of determining the surface would be by the laborious measurement and counting with the microscope. Even if microscopic studies are made with great care a spread in the surface value as high as 20% is obtained.

Polarization of the Scattered Light

We have seen that the Tyndall measurements give little information about the size distribution of particles apart from the fact that they are mainly large. For coastal waters a filtering technique may be resorted to in order to separate size ranges. But this is out of question for deep-sea samples which are easily contaminated. Let us for a moment consider the possibilities offered by polarization measurements.

The light scattered from the Tyndall beam is partly polarized. The degree of polarization varies with 1) optical properties of the particle material, especially the index of birefringence, 2) particle size, 3) concentration of the suspension, 4) angle at which it is observed. HATCH and CHOATE (1930) have studied suspensions of non-uniform particulate matter as silica, granite, and calcite and found the degree of polarization to be a function of the arithmetic mean diameter of the material.

The Tyndall meter with observations through an angle of 45° from the incident beam does not present the most favourable device for polarization measurements as the degree of polarization has its maximum through 90°. Apart from this, it offers no promising chances to apply the polarization method to the mixture of particles in the sea with their large range of size. Only in cases when a certain particle type strongly predominates would it be worth while to employ this procedure. Tests have established that the polarized light as well as the non-polarized must be determined with a high degree of accuracy, otherwise the measurements will be quite useless.

Sedimentation in the Water Samples

By sedimentation of the suspended matter in the flask it would be possible to get an idea of the size distribution. The theoretical background is essentially Stokes' law of settling. GUMPRECHT and SLIEPCEVICH (1953) have made an interesting application of this law. By means of light transmission measurements combined with differential settling they studied particle sizes in polydispersed systems consisting of non-absorbing spherical particles. The method was illustrated by an analysis of an aerosol.

Here we shall only postulate that equation (4), $I_s = KS$, is valid for a suspension of a given material of different sizes. The total surface of the particles per unit volume, S, may be written as

$$S = \pi \int_0^\infty N D^2 \, dD, \tag{6}$$

if N is defined as the number of particles having equivalent diameter D per unit volume of the suspension, per unit range of particle diameter (GUMPRECHT and SLIEPCEVICH).

Thus equation (4) can be expressed in the form

$$I_s = \pi K \int_0^\infty N D^2 \, dD \tag{7}$$

The largest size, D in the Tyndall beam after an elapsed time t is given by Stokes' equation

$$D = \sqrt{\frac{18\, h\mu}{gt\,(\varrho_1 - \varrho_2)}} \tag{8}$$

where h is the settling height during the time t, ϱ_1 and ϱ_2 are the densities of the particle and the liquid respectively, g is the acceleration of gravity, and μ is the dynamic viscosity of the liquid. The settling height, h, is the distance from the upper water surface to the Tyndall beam which is assumed to be small compared with h.

Differentiating equation (7) with respect to t we get

$$dI_s/dt = \pi K N D^2 \, dD/dt \tag{9}$$

into which values of D^2 and dD/dt found from equation (8) are inserted

$$dI_s/dt = -K' N t^{-5/2} \tag{10}$$

where

$$K' = \frac{1}{2} \pi K \left(\frac{18\, h}{g\,(\varrho_1 - \varrho_2)} \right)^{3/2} \tag{11}$$

Using equation (10) the value of N at the time t is found from the observed decrease of the Tyndall reading. The corresponding value of D is obtained from equation (8). The relationship between N and D gives the size-frequency distribution. The above expressions are valid for sizes between 2 μ and 60 μ, the lower limit being set by equation (4), the upper limit by Stokes' law of settling (equation (8)).

Methods for following the sedimentation have been developed in same detail (see HERDAN, 1953, p. 445). It is particularly important to secure an undisturbed settling in order to avoid thermal convection effects.

This procedure established for material of known density does not apply to the suspended particles in the sea with their large range of density. Nevertheless there is every reason to consider the possibility of reexamining the Tyndall sample after the elapse of a certain time during which the sample is left undisturbed to settle, preferably placed in the Tyndall meter under constant temperature conditions. The time interval must be sufficiently great so that the initial disturbances due to agitation and heating are unimportant, but not so great that any decomposition occurs in the sample. It is proposed that such a time is agreed upon in order to get an idea of the matter removable by sedimentation in water sample. It must be borne in mind, however, that the tranquil conditions necessary for the sedimentation tests do not always exist at sea.

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