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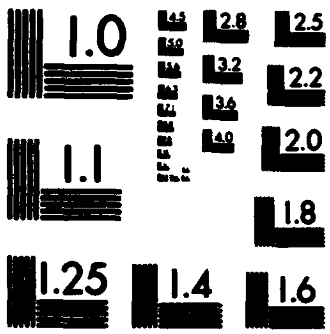

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FINAL ANNUAL REPORT
TO THE
OFFICE OF NAVAL RESEARCH

SOURCES & TRANSPORT MECHANISMS OF SEDIMENTS IN THE OCEANS

CONTRACT: N00014-75-C-0355
ENDING DATE: 31 MARCH 1982

By

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INTRODUCTION

The proposed continuing research has the overall scientific objective of elucidating the sources and transport mechanisms of ocean sediments. A quantitative approach to delineating the sources of sediments and the processes that are responsible for their transportation and alteration is critical. To accomplish these broad long-term objectives, a number of projects are underway. All of these are aimed at investigating fine-grained material since these materials comprise the vast majority (>90%) of the sediments in the oceans. Over the years a specialized group of equipment and specialized procedures have been assembled to facilitate working on these materials both in the field and in the laboratory.

Research on transport of fine-grained material has evolved because of the discovery of the importance of the flocculated state of much of this fine-grained material in the ocean. It has been very difficult to evaluate the importance of flocs in the ocean because of their fragile nature. Most sampling and analysis techniques in current use break the flocs and therefore do not permit us to observe them. Further, flocs are a product of their environment because they are at a dynamic equilibrium with it and can form and break up as turbulence levels change. Therefore my research on fine-grained material is directed toward various aspects of flocs in the ocean. The studies involve evaluating existing, and developing new, sampling and analysis techniques in order to find accurate techniques for studying flocs. Later studies will then utilize these proven techniques to study the mechanisms that produce, transport, break up, and deposit the flocs in the ocean.

ACCOMPLISHMENTS

During the past year I completed and have had published in Geology the article, "Site of river-derived sediments in the ocean". This paper shows that river-derived sediments are presently being deposited on shelf areas off 22 large rivers and that vast submarine fans are relics produced during a lower stand of sea level. A copy of the paper is included in the Appendix to this proposal.

During my studies of the shelf and slope area off the Amazon River over the past few years, I discovered an excellent example of upwelling that had never been reported before. The article reporting this, entitled "Wind- controlled coastal upwelling in the western equatorial Atlantic Ocean" was completed this past year and was published in Deep Sea Research. A copy of this paper is included in the Appendix to the present proposal.

Floc Study Techniques

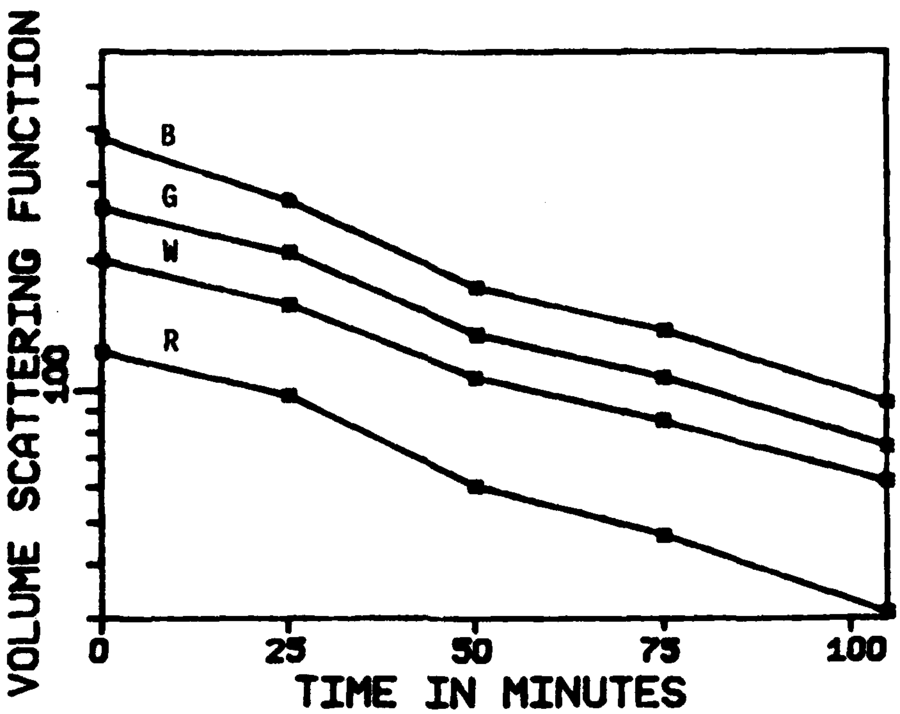
During the past year my research group has been involved in a number of studies evaluating existing, and developing new, sampling and analysis techniques in order to determine techniques that will allow us to obtain accurate information on flocs. While, intuitively, many researchers consider flocs to be fragile, there are no quantitative studies that test what procedures do and do not break flocs during sampling and analysis.

Pumps. This past year we tested the effects of three standard pumps on flocs during handling and sampling. This work showed that all pumps allowed severe floc breakage and that even a slow peristaltic pump causes breakage. The results of this work will be published in the June issue of the Journal of Sedimentary Petrology. A copy of this paper is included in the Appendix to this proposal.

Niskin Bottle. Because the majority of samples in oceanographic research are taken using Niskin bottles, the effect they have on flocs was tested. In our study, we found that significant floc breakage occurred when the samples were withdrawn from the spigots at the bottom of the Niskin bottles. This work has been written up and submitted to Estuarine and Coastal Marine Sciences for publication. A copy of the manuscript is included in the Appendix to this proposal.

Pipetting. In the study of flocs a pipette is commonly utilized either to take the sample to place it into an analytical instrument or to measure a known volume of sample material for the procedure. In the laboratory we tested the effect of pipettes with various tip openings on handling flocs. It was found that the all normal commercially available pipettes severely broke flocs. This work has been written up and submitted to Environmental Science and Technology for publication. A copy of the manuscript is included in the Appendix.

Light Scattering and Transmission. Experiments are underway in our laboratory to determine the effect flocs have on light scattering and transmission. These experiments are designed to obtain a series of light scattering and transmission readings at various floc sizes from 2 to 200μm at a constant total suspended material concentration. The experiments this far show a striking effect of flocs on the optic readings. As the suspended material flocculates the optical readings change many orders of magnitude (Figure 1) from readings for the same concentration of suspended material that has not flocculated.



Light scattered at 15° by white, blue, green, and red light from natural sediments. Time zero is 2μm particles and time 100 min. is 200μm flocs.

The light scattering and transmission experiments will be discussed further in the section on Proposed Research.

<u>Holography</u>. Our experimental holographic system has been tested on a variety of flocs under both quiet-settling and open-flow conditions. The system, similar to Carder's set-up, is only useful under quiet settling of limited settling velocities. Under conditions of open flow similar to that of the ocean the system is not capable of stopping the motion of the particles and therefore produces poor particle resolution. Since our objective is to obtain a system that can obtain images of flocs in the open ocean, modification of the system is necessary. The most promising change will be incorporation of a pulsed laser that will permit high light intensity for very short time durations. Development will be discussed under Proposed Research.

Turbulence System. One of the important variables in the flocculation (and break up) of suspended material is the turbulence of the environment. To measure this turbulence we have continued to improve our hot-film anemometry system during the past year. The system consists of a TSI parabolic hot-film probe, a DISA constant-current anemometry system, a PDP-11-03 minicomputer, and a digital printer. We will be able to obtain a real-time turbulence spectrum and, as a result, our flocculation experiments can be based on a scientifically sound turbulence measurement.

Acoustic Projects. This past year we started two acoustic projects, both involved with the effects of flocs on acoustic signals. One project is in collaboration with the Naval Research Laboratory and the second with Dr. Marshall Orr of Woods Hole, related to HEBBLE. For both projects preliminary feasibility experiments have been conducted. The details of the projects are presented in Proposed Research.

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FLOC BREAKAGE BY PUMPS

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INTRODUCTION

Pumps are convenient and inexpensive devices for sampling suspended material in water from various depths in water bodies. The pump can be used above water, sucking the water sample up through the hose and the pump; or the pump can be lowered into the water, with the water sample being pushed through the pump and then up the hose. In both cases the suspended sediment passes through the pump and through a length of hose. The objective of the work reported here was to investigate to what extent flocs of the suspended material are broken by pumps. While, intuitively, many researchers consider flocs to be fragile, there are no quantitative studies that test what procedures do and do not break flocs during sampling and analysis.

The existence of flocs in the environment has been observed by Berthois (1961), Biddle and Miles (1972), Edzwald (1972), Gibbs (1977), Kranck (1975), Krone (1962), Schubel and Kana (1972), Sheldon (1968) and Zabawa (1978). Because the process of flocculation is critical to our understanding of the transportation and deposition of suspended material, it is important to have accurate and precise methods for sampling flocs.

Three types of pumps were selected for testing, in order to cover the range of pumps utilized by researchers. Both the centrifugal and impeller pumps (those most popularly used in sampling suspended material) produce high shear in a sample as it passes through the pump. A third type of pump, the peristaltic pump, was selected for testing as being the type producing the lowest amount of shear among all pumps presently on the market. The peristaltic action, that is, progressively squeezing the tube through which the water being sampled passes, thus forcing portions of water along the tube, was found to offer the best chance of not breaking flocs in the sample.

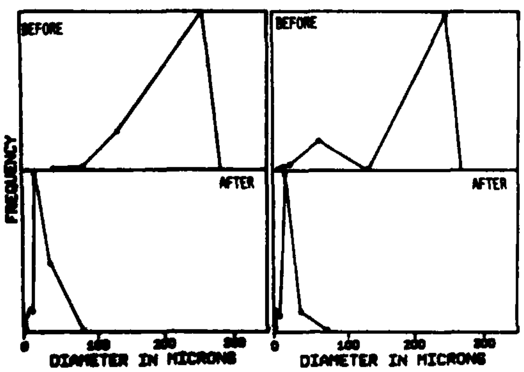

FIG. 2.—Floc size distribution of a) natural samples and b) kaolinite samples before and after samples were passed through a centrifugal pump.

PROCEDURES

The flocs used in testing the three types of pumps were produced from pure, coating-free kaolinite (API standard #3), size-fractionated into the 1- to 4-μm class. This material was selected for comparison because this, or similar kaolinite, is extensively used in laboratory studies (Whitehouse et al., 1960; Edzwald, 1972). To study the effect of pumps on samples taken in natural environments, an untreated sample of suspended sediment from the Amazon River, obtained from the Atlantic Ocean area off the mouth of the Amazon River, with natural organic and iron coatings, was utilized. The sample was not treated in any way and was not permitted to dry out. The composition of this natural sample was 40 percent kaolinite, 40 percent illite, and 20 percent montmorillonite. In order to eliminate the effect of large particles that may not be involved in flocculation (including plankton, quartz and feldspar grains) these coarser particles were eliminated, leaving only material <4μm in diameter.

The samples were flocculated in a reaction chamber using stirring blades similar to those utilized in the work of Edzwald (1972) and Eppler (1975). The turbulence produced in the reaction chamber (a shear rate, G, of 20 sec^{-1}) would represent the turbulence of an environment having above-average turbulence, that is, higher than most parts of estuaries and oceans. The concentration of the suspended sample was 75 mg/l of suspended solids; the various salinities used in testing were obtained by mixing seawater and river water from the Delaware River, both filtered through 0.45 μm filters. The flocs in the samples were permitted to reach an equilibrium size distribution that showed no further change with time and were then permitted to age for an additional 8 hours. These reasonably high shear conditions, and permitting the flocs to reach equilibrium and then age, would permit the development of strong flocs. The floc samples created in this way were then passed through a centrifugal pump (TEEL IP760A), through an impeller pump (TEEL IP866), and through a peristaltic pump (Sigmamotor T8).

Before and after they passed through each type of pump, the suspensions were sampled using a 2.5 μm-diameter glass tube. These samples were each placed on a glass slide having a 400 μm-deep well. The flocs were then sized using an optical microscope having a magnification of 450.

RESULTS AND DISCUSSION

During the impeller pump study, the flocs of the natural sample material were, initially, nearly 80 μm in diameter, but after passing through the impeller pump the flocs were broken down to less than 20 μm diameter (Fig. 1a). Flocs of the kaolinite sample were, for the most part, about 220 μm in diameter before being passed through the impeller pump, but after passing through the pump the majority of flocs were less than 20 μm, with a minor amount about 80 μm in diameter (Fig. 1b). The impeller pump is extremely destructive to both types of flocs.

In the test using the centrifugal pump the flocs of the natural sample were initially about 250 μm in diameter and, after passing through the centrifugal pump, the majority of flocs were less than 20 μm in diameter, or only 1/12th their size before passing through the pump (Fig. 2a). Flocs in the kaolinite sample were initially about 250 μm in diameter, but after passing through the centrifugal pump the remaining flocs were also about 20 μm in diameter (Fig. 2b). Therefore, the centrifugal pump significantly broke flocs of both the natural sample and the kaolinite sample.

In the study using the peristaltic pump, the natural sample flocs were 80 to 150 μm in diameter and, after passing through the peristaltic pump at high speed (1300 RPM) the majority of the flocs remaining were about 20 μm in diameter (Fig. 3a). In order to determine if flocs would be broken using a low-speed peristaltic pump (300 RPM), with much lower shear, a test was conducted using natural samples. Before the low-speed test the flocs attained a diameter of 80 μm. After passing through the peristaltic pump at low speed, the majority of the flocs were only 20 μm in diameter (Fig. 3b). Therefore, using even the pump with the lowest shear, operating at low speed, significant floc breakage occurred. In the kaolinite sample, flocs were originally between 100 and 250 μm in diameter, but after passing through the peristaltic pump at 1300 RPM, the flocs were between 20 and 50 μm in diameter or 1/5 the floc size originally present.

The use of pumps to sample suspended material from various environments is fairly common practice. Based on testing the effect of impeller, centrifugal, and peristaltic pumps on flocs that were stronger than those encountered in research, it was determined that floc breakage occurs, with all three types of pumps, in flocs of both natural samples and kaolinite. The breakage that occurs produces flocs 1/4 to 1/12th the size flocs had attained before being passed through the pumps. It is obvious that use of pumps during sampling of suspended material for floc size is not an acceptable technique.

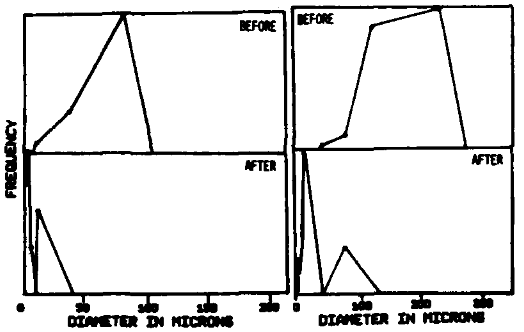

FIG. 1.—Floc size distribution of a) natural samples and b) kaolinite samples before and after samples were passed through an impeller pump.

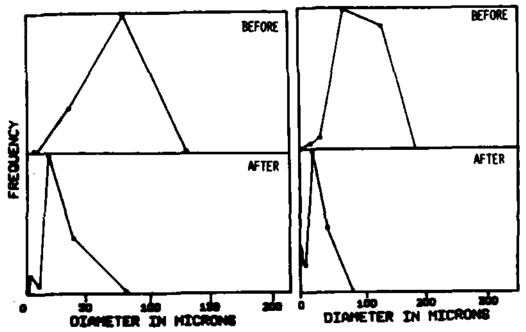

FIG. 3.—Floc size distribution of natural samples before and after samples were passed through a peristaltic pump at a) high speed and b) low speed.

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Sampling of Mineral Flocs Using Niskin Bottles

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■ The effect of using a Niskin sampling bottle to obtain water samples for size and concentration analysis of coagulated suspended material was determined by experimentation. Flocs of kaolinite and of an untreated sample with natural organic and iron particle coatings were produced in a Niskin bottle containing mixing blades. Samples of kaolinite withdrawn at different rates of flow from the outlet valve at the bottom of the bottle contained flocs that were $^1/_5$–$^1/_3$ the average size of the flocs inside the bottle. Samples of untreated natural sediment withdrawn at different rates of flow from the bottom outlet contained flocs $^1/_2$–$^1/_8$ the size of the flocs inside the bottle. This extensive floc breakage indicates that samples withdrawn from the bottom outlet of Niskin-bottle samplers are not acceptable for size and concentration studies of flocs.

Introduction

The existence of flocs have been observed in brackish and marine waters by Berthois (*1*), Biddle and Miles (*2*), Gibbs et al. (*3*), Kranck (*4*, *5*), Krone (*6*), Owen (*7*), Schubel and Kana (*8*), Sheldon (*9*), and Zabawa (*10*). The significance of the coagulation process to our understanding of the transportation and deposition of suspended material has been recognized by these researchers. While, intuitively, many researchers consider flocs to be fragile, there are few quantitative studies that test what procedures do and do not break flocs during sampling and analysis.

Procedures utilized to obtain water samples with unbroken flocs for later size and concentration analysis are difficult, and their reliability has not been tested. The effect of using pipets for sampling or handling has been studied by Gibbs and Konwar (*11*), who found significant floc breakage with commercially available pipets. However, to obtain samples from various levels in the water column, either sampling bottles or pumps are employed. It is not surprising that pumping techniques would be considered sufficiently harsh as to break flocs (*12*). Therefore, sampling bottles are used to obtain samples for floc studies, but no studies have been made to determine if these procedures deliver samples for analysis with flocs remaining in an unbroken state. During the counting analysis of flocs, breakage has been demonstrated to occur by utilizing the Coulter counter (*13*) and the HIAC light-blocking instrument (*14*). The objective of the study was to evaluate the degree of floc breakage that occurs during use of Niskin sample bottles (*15*).

Procedures

Hydrodynamic consideration of the shear that flocs would be subjected to during the various stages of sampling using a Niskin bottle indicated that, as the bottle is lowered downward in the water column, shear would be generally equal to or lower than that of the environment to which the flocs were previously exposed. In removing portions of the samples from the Niskin bottle, the greatest shear occurred at the discharge valve at the bottom of the bottle where water passed through the few-millimeter orifice in the valve at a high veloicty and through a tube into a sample receptacle.

We decided, therefore, to study this critical aspect of the sampling procedure in greater detail. The objective was to produce flocs inside a Niskin bottle and to study the changes in their size distribution that accompany flow through the discharge valve into a breaker.

Flocs were produced in a 5-L Niskin bottle by stirring mixtures of material and seawater at 50 rpm (equalling a shear of 10 s^{-1}), with a series of four propellers mounted on a vertical shaft running the length of the Niskin bottle. Stirring promoted flocculation until a state of equilibrium with regard to size distribution was reached. Stirring also maintained uniform distribution of the suspended material within the Niskin bottle. Samples of the flocs were obtained with a glass tube having a 2.5-mm i.d. using a slow filling velocity (< 0.5 m/s). The samples were then carefully placed on microscope slides having a 400-μm-deep well, and size distribution was counted by using a microscope at 450 magnification.

The sample materials used for these experiments were 2–4-μm size fractions of pure, coating-free kaolinite (American Petroleum Institute standard no. 3) and an untreated sample of Amazon River suspended material from the Atlantic Ocean area off the mouth of the Amazon River, with natural organic and iron particle coatings. The composition of this natural sample was 40% kaolinite, 40% illite, and 20% montmorillonite. The natural sample was kept wet and was thus an example of environmental flocs. Kaolinite was used because of the extensive flocculation studies on this material and to permit comparisons of floc strength.

Two experiments were conducted and duplicated on the kaolinite sample by using sediment concentrations of 15 mg/L, salinity of 15‰, temperature of 20 °C, and stirring time of 7 h. Two experiments were conducted and duplicated with the natural sediment sample material using sediment concentrations of 20 mg/L, salinity of 15‰, temperature of 20 °C, and reaction times of 1 and 5 h.

Results and Discussion

The kaolinite flocs developed to almost 200 μm in diameter at equilibrium (Figure 1) in the Niskin bottle but were reduced to about 70 μm in diameter, only 35% of the original size, when the floc sample was allowed to run gently down the inside wall of the beaker through a tube connected to the outlet valve of the Niskin bottle. A more striking difference was produced when the floc sample was taken from the Niskin bottle in the normal manner, which involves a stream of water free falling 5 cm into the beaker from the outlet valve. With this method, the flocs were broken down to a dominant size of 25 μm and a secondary size of 7 μm, representing only $^1/_8$ and $^1/_{35}$ of the original floc size.

Experiments with untreated natural sediment material produced comparable results at both reaction times. The flocs in the Niskin bottle ranged between 150 and 300 μm in diameter with a mean of about 225 μm (Figure 2). When the sample was allowed to run gently down the inside wall of the beaker through a tube connected to the outlet valve of the Niskin bottle, the majority of the flocs were about 100 μm in diameter, with none larger than 150 μm. The flocs obtained in this manner were less than about 45% of the size of the flocs inside the Niskin bottle. In the untreated natural sediment sample taken from the Niskin bottle in the normal manner, with the stream of

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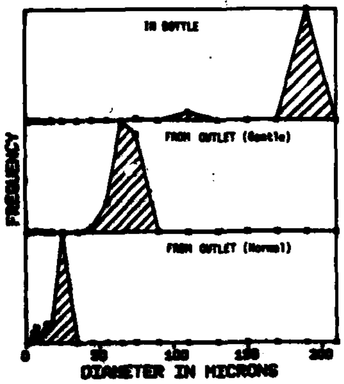

Figure 1. Size distributions for kaolinite flocs in the Niskin bottle and after removal from the outlet with gentle handling and with normal handling.

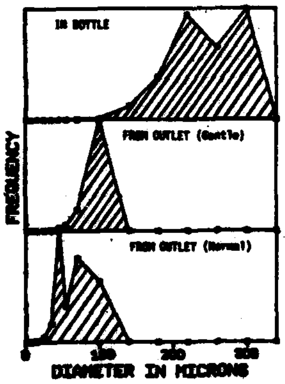

Figure 2. Size distributions for flocs of natural sample in Niskin bottle and after removal from the outlet with gentle handling and with normal handling.

water free falling 5 cm into the beaker from the outlet, the majority of the flocs were 45 μm with a small portion approximately 70 μm. This was only $^1/_5$ the size of the flocs inside the Niskin bottle.

The turbulence in the Niskin bottle created by four mixing propellers producing a G of 10 s^{-1} is comparable to that encountered in natural environments and, probably, most comparable to high-energy environments. If a slower stirring rate were used, resulting in a lower shear, the flocs would be larger and weaker, exhibiting even more dramatic breakage of flocs.

Conclusion

These controlled experiments demonstrate that significant breakage of flocs can occur when suspended material is withdrawn from a Niskin bottle in the usual manner, either allowing the water to fall freely from the discharge valve at the bottom of the bottle or allowing the water to run gently down the inside wall of a beaker through a tube connected to the outlet valve of the Niskin bottle. Alternative methods of obtaining samples from Niskin bottles for coagulation studies (such as using pipets through the top opening of the bottles) should be considered.

Registry No. Water, 7732-18-5.

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Effect of Pipetting on Mineral Flocs

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■ Flocs of kaolinite and of untreated natural suspended material were produced in a Couette-type reaction chamber using 2‰ and 30‰ seawater solutions. The diameter of the pipet tip opening directly affected the degree of floc breakage when samples were extracted. Extremely small pipet tip openings (125–400-μm diameter) disrupted nearly all flocs. In general, pipet tip openings should exceed 2000 μm in order to prevent floc breakage. Pipet openings should be larger than 3000 μm in diameter in cases of weakly bound or large flocs.

Introduction

Flocs in the marine environment have been observed by Berthois (*1*), Biddle and Miles (*2*), Edzwald (*3*), Edzwald and O'Melia (*4*), Gibbs (*5*), Kranck (*6*), Krone (*7*), Schubel and Kana (*8*), Sheldon (*9*), and Zabawa (*10*). Because flocculation affects transportational and depositional processes of materials, it is critical to have accurate as well as precise methods for measuring the flocs.

An important experimental task in the investigation of the flocculation–deflocculation phenomenon of particles is documentation of the size and/or the number of flocs. Because clay flocs are often fragile, utmost care must be taken during the measurement process. While, intuitively, many researchers consider flocs to be fragile, there are no quantitative studies that test what procedures do and do not break flocs during sampling and analysis. Another study (*11*) showed breakage when using pumps to sample flocs.

There are mainly three techniques for determining the size and the number of particles. The simplest approach is taking a sample by using a pipet and placing it on a slide for microscopic examination. A second approach for measuring the size distribution of suspended material is the Coulter-counter technique, in which the particles are passed through a tiny orifice, changing the resistance across the orifice in proportion to particle size. The effectiveness of this method in defining the size and the number of flocs has yet to be evaluated. The advantage of the Coulter counter is that large numbers of particles can be counted in a short time and a computer coupled to the counter can produce displays of size distributions rapidly. The third group of methods utilizes optical techniques in an in situ mode. Light scattering generally is considered a nonspecific technique for this purpose, but it can give some useful information (*12*). Recording the image by either photographic or holographic techniques (*13*) is useful for some studies, but it is fairly expensive and presently lacks the resolution needed for flocs smaller than 5 or 10 μm in diameter.

The purpose of this study is to evaluate what effects, if any, pipet handling has on the flocs either when taking the sample or when diluting the sample for analysis. Various investigators use pipets of varying diameters without stating reasons for their choice, and in many cases other procedures that may have affected floc breakage have not been considered in preparing samples. This study's specific objective is to determine the relationship between floc breakage and the size of pipet tip openings and to develop a procedure that will minimize floc breakage when using pipets in handling suspended-material samples.

Procedures

The materials utilized to produce the flocs for this study were untreated kaolinite, (API standard no. 3), size-fractionated into 2–4-μm intervals, and an untreated sample of suspended sediment from the Amazon River, obtained from the Atlantic Ocean area off the mouth of the Amazon River. The 2–6-μm size fraction from the Amazon River

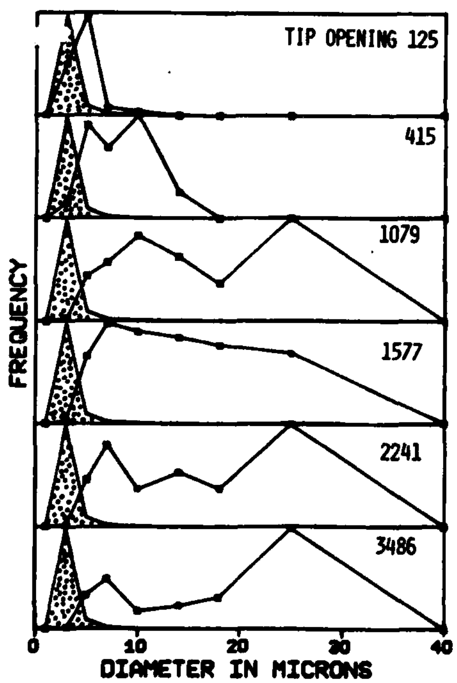

Figure 1. Size distributions obtained for kaolinite flocs by using pipets with tip openings of various diameters at 30‰ salinity. Primary particle size mode is shown by stipling.

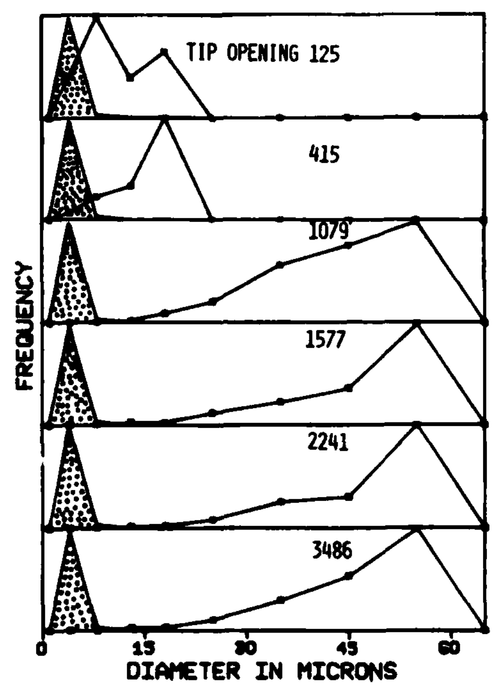

Figure 2. Size distributions obtained for flocs of natural sample by using pipets with tip openings of various diameters at 2‰ salinity. Primary particle size mode is shown by stipling.

sediment was not treated and thus retained its natural organic and iron coatings. The composition of this natural sample was 40% kaolinite, 40% illite, and 20% montmorillonite.

Flocs were produced in a Couette-type reaction chamber with a velocity gradiant of 32 s^{-1} for kaolinite and 20 s^{-1} for the untreated natural sample. The solutions were prepared by using filtered natural seawater and diluted to 2‰ with distilled water. The concentration of suspended material was maintained at 15 mg/L for all experiments, a concentration typical of many estuaries.

In an effort to eliminate human error, an automatic mechanical pipet was utilized for sampling and for placing the material on microscope slides having wells 400 μm deep. The number and the size of flocs were determined by using a microscope with 450× magnification.

Primary particle-size distributions were determined before the material started to flocculate. The material was then permitted to flocculate and sampled by using pipets with six size openings, ranging from 125 to 3486 μm in diameter. For perspective, the openings of commercially available pipets used for most analytical work range from 800 to 1500 μm in diameter. The samples were permitted to reach an equilibrium size distribution before successive sampling with the various pipets. This minimized the possibility of the sample changing during sampling. The pipetted samples were taken in rapid succession from the same position in the container.

Results and Discussion

The experiments for floc breakage of kaolinite at both 2‰ and 30‰ salinity showed similar results. A 125-μm pipet opening allows breakage of almost all of the 20–30-μm flocs (Figure 1), whereas 2241- and 3486-μm diameter pipets appear to disrupt very few flocs. The 415-, 1079-, and 1577-μm pipet openings appear to cause some breakage. The kaolinite flocs were formed under a high shear (32 s^{-1}) in the reactor, which would produce strong flocs and break weak flocs. The natural environment would have shears below 10 s^{-1}, and as such the experiments are

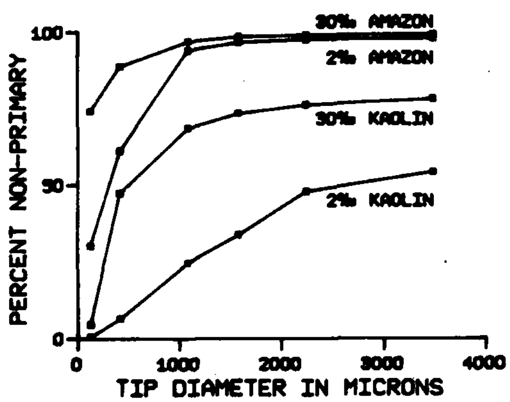

Figure 3. Percent of nonprimary particles (flocs) for pipets with tip openings of various diameters.

considered to be an extreme test for producing least breakage.

The untreated natural material was allowed to grow to an equilibrium size distribution at a lower shear, representative of that found in estuaries. The results (Figure 2) show that the flocs grew to over 50 μm in diameter in both the 2‰ and 30‰ salinity experiments. Results for pipets having 125- and 415-μm openings indicate significant floc breakage, whereas results for 1079-μm diameter openings indicate less breakage; larger-diameter openings do not produce significant breakage of flocs.

In an effort to quantify the degree of floc breakage as a function of the tip diameter of the pipets, the percent of nonprimary particles (flocs) was determined (Figure 3). More breakage of flocs occurred for 2‰ salinity samples than for 30‰ salinity samples, indicating the existence of weaker flocs at the lower salinity. Because the experiments on the two samples (untreated natural material and kaolinite) were conducted at two different shear rates, it is difficult to draw direct comparisons. However, judging from the progressive increase in the curve for the 2‰ salinity solution of the kaolinite sample as opposed to the more rapid rise in the curve for the 2‰ salinity sample of the untreated natural sample, the kaolin flocs appear weaker. This would be expected because of the composition of the natural sample, which would produce a stronger floc than kaolinite alone.

A pipet having a tip opening less than 2000 μm in diameter can, therefore, produce significant floc breakage; in the case of weak flocs, tip openings larger than 3000 μm in diameter are recommended. It should also be pointed out that, because the maximum floc size studied was only 65 μm in diameter, care in handling should be applied when dealing with larger flocs.

Conclusions

The diameter of the pipet tip opening used for sampling suspended material for flocculation studies directly affects floc breakage. Extremely small pipet tip openings disrupt flocs of all sizes. Therefore, pipet tip openings should be larger than 2000 μm in order to minimize floc breakage. If flocs are large and/or weak, the tip opening of pipets should be larger than 3000 μm. All commercially available pipets have unacceptably small tip openings. Studies of flocs should include measurement to show that sampling methods do not disrupt flocs.

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Floc Breakage during HIAC Light-Blocking Analysis

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■ Flocs of kaolinite and natural suspended sediment samples were measured by using an optical microscope before and after analysis with a HIAC light-blocking instrument. Extensive floc breakage was indicated for all samples after they passed through the HIAC sensor. This breakage was dominant in the upper two-thirds of the recommended size range for the HIAC sensing cell.

Introduction

Determining the size of flocs or aggregates is important in studying transportation processes in nature because flocs determine the hydrodynamic behavior of suspended solids. The existence of flocs in the environment has been observed by Berthois (*1*), Biddle and Miles (*2*), Edzwald (*3*), Edzwald and O'Melia (*4*), Gibbs (*5*), Kranck (*6*), Krone (*7*), Schubel and Kana (*8*), Sheldon (*9*), and Zabawa (*10*).

Because flocs and aggregates are often fragile, utmost care must be taken during the measurement process. While, intuitively, many scientists think flocs are fragile, there are few quantitative studies that test what procedures do and do not break flocs during sampling and analysis. Using any type of pump for sampling was shown by Gibbs (*11*) to break flocs. Pipetting samples is also a cause of floc disruption (*12*). Another common sampling technique in oceanography, using Niskin bottles, was shown by Gibbs and Konwar (*13*) to break flocs. The size analysis of flocs by using the Coulter counter had extensive error due to breakage of flocs (*14*).

The purpose of the present study is to evaluate the effectiveness of the popular HIAC light-blocking instrument in measuring flocculated material for size.

Procedures

Flocs of known size were passed through the HIAC instrument and then measured again. The materials utilized to produce the flocs for this study were coating-free, untreated kaolinite (API standard no. 3), size-fractionated to obtain the 2- to 4-μm material, and an untreated sample of suspended sediment from the Amazon River obtained from the Atlantic Ocean area off the mouth of the Amazon River. Because this 2–6-μm size fraction from the Amazon River was not treated, its natural organic and iron coatings were retained. This natural sample was composed of 40% kaolinite, 40% illite, and 20% montmorillonite. The flocs were produced in a reaction chamber using stirring blades similar to those utilized in the work of Edzwald (*3*) and Eppler et al. (*15*). The turbulence produced in the reaction chamber (a shear rate, G, of 10 s^{-1}) would represent above-average turbulence, that is, greater turbulence than found in most parts of estuaries and oceans. Sediment concentration was 50 mg/L and salinity was 2‰ for all experiments. The flocs were removed from the flocculation chamber by using a pipette having a 3-mm opening, and the sample was carefully diluted to 1/100 with filtered 2‰ salinity solution. This diluted sample was analyzed by using a HIAC instrument. Samples from before and after HIAC analyses were also taken and placed on a special slide having a well for sizing and counting using an optical microscope at 100–450 magnification. The HIAC light-blocking sensor, CMH-60, is recommended by the manufactor for use with particles having diameters up to 60 μm, so the flocs were produced to a maximum diameter of 55 μm. The HIAC instrument was set with its 12 channels ranging from 1.4- to 60-μm particle diameters. Immediately behind the sensing cell of the HIAC instrument a special large-bore outlet was placed, through which the samples were obtained for microscopic analysis. The particle-size distribution in the beaker from which the HIAC instrument draws its sample was stirred gently to maintain uniform distribution. When monitored, the sample was shown to have no effect on floc-size distribu-

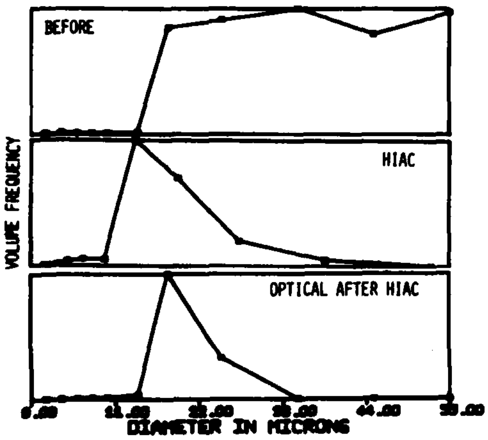

Figure 1. Size distribution for kaolinite flocs measured by using a microscope before HIAC analysis (top), by a HIAC instrument (middle), and by using a microscope after the flocs had passed through the HIAC sensor.

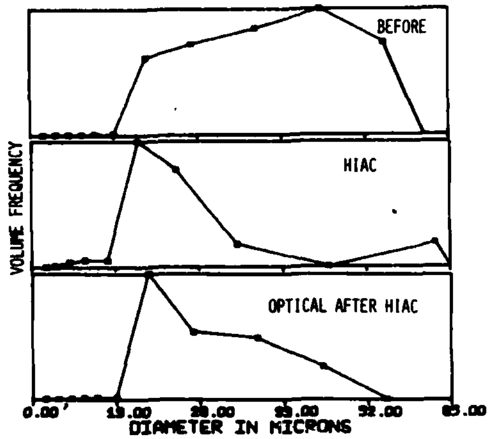

Figure 2. Size distribution for natural suspended material measured by using a microscope before HIAC analysis (top), by a HIAC instrument (middle), and by using a microscope after the flocs had passed through the HIAC sensor.

tion. The change in size of the flocs was not significant after the sample was diluted. All experiments were conducted in duplicate.

Results and Disussion

The experiments with kaolinite indicated that, before passing through the HIAC sensor, the flocs had ranged from 15 to 55 μm in diameter (Figure 1, before HIAC). For the same sample, the HIAC instrument produced results with a large mode at about 12 μm in diameter and with few flocs larger than 24 μm in diameter (Figure 1, HIAC), indicating that the flocs between 24 and 55 μm in diameter were broken by the HIAC sensor. This breakage would have occurred during high shear as the floc-laden solution passed through the 60-μm-wide cell. For further confirmation of floc activity as a result of passing through the HIAC sensor, samples taken immediately after the flocs passed through the sensor were analyzed by optical microscopic techniques. The optical analysis indicated results similar to those from HIAC analysis, supporting the idea that the HIAC sensor breaks flocs between 24 and 55 μm in diameter (Figure 1, optical after HIAC). This is especially true since the "before" and "after" samples were analyzed with the same optical procedure. The duplicate experiment with kaolinite produced almost identical results.

The flocs from the experiments on the natural Amazon sample before HIAC analysis ranged from 13 to 52 μm in diameter (Figure 2, before HIAC). Analysis of the natural sample using the HIAC instrument showed a large mode at about 15 μm in diameter with very few flocs occurring above 28 μm in diameter (Figure 2, HIAC). These results indicate that the HIAC sensor breaks flocs greater than 28 μm in diameter. For confirmation of this, samples taken from immediately behind the HIAC sensor were analyzed by optical microscopic techniques. This independent analysis showed that the floc samples leaving the HIAC senser had modes at about 15 μm in diameter, collaborating findings that the HIAC sensor breaks flocs greater than about 25 μm in diameter.

The results from analysis of both the kaolinite and the natural samples show that, for the 60-μm-wide sensor, severe floc breakage occurs above about 25 μm in diameter, which is 42% of the sensor width. A secondary problem caused by this floc breakage is that the number of particles below 25 μm is also increased because of the fragments of the larger flocs. Plots of the number frequency distributions, while shifted to the smaller sizes relative to the volume frequency distribution, still show the floc breakage.

Conclusion

The HIAC light-blocking instrument has been shown to break flocs larger than 40% of the width of the cell, obviously decreasing the number of flocs recorded above this limit, but also increasing the number of fragments of these larger broken flocs in the smaller sizes. Great caution is necessary when the HIAC instrument is used to measure the size distribution of flocculated material. We urge that an independent method be used to check for floc breakage before the HIAC instrument is used to analyze flocculated material; otherwise, it should not be utilized for analyzing flocculated material.

Acknowledgments

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