

CHAPTER 36

WASTE DISPOSAL IN MARINE WATERS

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Waste disposal in marine waters is of importance to coastal engineers because of the ever-increasing requirements for an effective means of dilution of both municipal and industrial wastes. There are many considerations that enter into a satisfactory waste disposal method by dilution, but to the engineer two major considerations must be made. These are, first, the rate of diffusion and, second, the level of dilution necessary to effect disposal. These considerations become quite involved because of the varying conditions which influence dilution or diffusion processes and the uses of the water, which determines the criteria for a satisfactory dilution level. Thus, the dilution levels necessary will be dependent on the nature of the material and the area of dispersal.

Generally, however, it might be considered that adequate dilution has occurred when the disposal of waste at the site of interest does not interfere with any recreational or commercial use of the area in question. Usually in the marine area the standard is set at such a level that the biological organisms endemic to the area are not harmed by the waste material.

Methods of measuring rate of diffusion have been under study for a number of years. Munk, Ewing and Revelle (1949) investigated diffusion in the Bikini Lagoon in which the radioactivity caused by the atomic explosion was used as a tracer of the water mass. Recently Seligman (1955) reported results obtained by the use of fluorescein in estimating diffusion in the Irish Sea. This work was carried out in a littoral region in order to evaluate a disposal method for dispersion of radioactive wastes from the Harwell Atomic Energy Plant. Ketchum and Ford (1952) studied the diffusion behind a barge at sea in which the ferrous ion was used as an indicator. In these experiments the peak concentration of the waste was difficult to determine because only discrete samples were obtained.

Recently radioactive isotopes, bacteria and dyes, have been used as tracers to measure diffusion rates for various applications. Ely (1957) reports on the use of Sc^{46} in following dilution of a sewage field in Santa Monica Bay. Cochrane (1956) applied P^{32} to trace sewage

TABLE I
TRACERS USED FOR DIFFUSION STUDIES

| <u>Tracer</u> | <u>Means of Detection</u> | <u>Limits of Detection</u> | <u>Hazard</u> | <u>Adaptability for Continued Analyses</u> | <u>Quantity/10⁶ M³</u> | <u>Expendables Cost/10⁶ M³</u> |
|------------------------|---------------------------|-----------------------------|---------------|--|--|--|
| ⁴⁶ Sc | Scintillation | 1.2 x 10 ⁻⁴ C/L* | Hazardous | Good | 0.12 C | \$360.00 |
| Cs ¹³⁷ | Scintillation | 2.4 x 10 ⁻⁴ C/L | Hazardous | Good | 0.24 C | 24.00 |
| ⁷⁵ Se | Scintillation | 1.2 x 10 ⁻⁴ C/L | Hazardous | Good | 0.12 C | 120.00 |
| H ³ | Scintillation | 5.0 x 10 ⁻² C/L | Low Hazard | Poor | 13.5 C | 1,350.00 |
| Fe ⁺⁺ | Orthophenanthroline | 20µg/L | No Hazard | Poor | 44 lbs. | 44.00 |
| NH ₃ | Nesslerization | 20µg/L | Little Hazard | Poor | 44 lbs. | 8.80 |
| Lithium | Flame Photometric | 3 mg/L | No Hazard | Fair | 6,600 lbs. | 53,000.00 |
| Fluorescein | Fluorimeter | 1µg/L | No Hazard | Good | 2.2 lbs. | 5.50 |
| <u>Serratia indica</u> | Plate Count | 10 ³ C/L | No Hazard | Poor | 10 ³ L | Ca 350.00 |

*Based on requirement of 0.25 DPM/ml

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movement. He also used an alternate method in which Serratia indica was added to the sewage and subsequently sampled and cultured to determine the rate of dilution. Serratia indica was also used by Robson (1956) to trace sewage pollution. Moon, Bretschneider, and Hood (1957) have reported on a method for measuring eddy diffusion in coastal embayments in which fluorescein dye was used as the tracer, and a continuous reading fluorescent meter was used as a detector. Hood, Stevenson, and Jeffrey (1957) used similar methods for detection of diffusion behind a barge at sea. Stommel (1947) has treated the topic theoretically.

If horizontal diffusion is to be known under the conditions of the environment of interest an experiment must be conducted which will allow diffusion to be directly observed. The most direct method appears to be one of addition of an easily observable tracer to the water mass, followed by detection of the tracer as an indicator of dilution.

SELECTION OF A TRACER

The selection of a tracer for use in diffusion studies is based on several criteria. These are (1) the tracer must be one which is, in a chemical state, compatible with the conditions existing in the study area; (2) the tracer must be easily detectable to a high degree of precision, preferably by a method adaptable to continuous measurement; (3) the tracer must not be hazardous to the environment or people working with the material while conducting the experiments; (4) the cost of the tracer must be reasonable; (5) the cost of capital equipment necessary for the dispersal measurement must also be reasonable.

A number of tracers have been contemplated for use and/or used, in diffusion studies. Table I gives a comparison of certain important considerations applicable to tracers for use in diffusion measurements. The radioactive tracers, which are gamma emitters which are found most useful for diffusion studies are Sc^{46} , Cs^{137} , and Se^{75} . These are all easily detected by scintillation counting with a water proof detector that can be towed to any desired depth in the sea. The limit of detection of all three is good, and the quantity of material necessary to label an area one square kilometer by a meter deep is not excessive. All of these elements are present only in trace quantities in the sea. The amount added is significant to the total amount present and, therefore, some consideration must be given to insure that absorption does not remove them from the water mass. Ely (1957) found that the addition of sodium versenate to Sc^{46} tracer minimized adsorption. The maximum level of a radioactive tracer that may be added to the water is the maximum tolerance level permitted by

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AEC regulations. Assuming a detection level when diluted 1 part in 10,000 a top practical limit is then placed on the amount of water that can be labeled from a single discharge site. These isotopes are gamma emitters and are, therefore, hazardous to handle, particularly before addition to the water mass.

Tritium, which can also be detected by liquid scintillation methods, would be an excellent isotope for use in the form of tritiated water. Since total water present is infinitely greater than the amount of the water added, one would expect this tracer to follow the water mass without special removal by any other material and/or chemical process existent in the water. The chief disadvantage of this isotope, besides being somewhat more expensive, is that the technique for detection on a continuous basis has not been worked out. Other isotopes such as Na^{24} or Cl^{37} would be suitable tracers except for the difficulty of obtaining short half-life isotopes.

A number of chemical indicators are also feasible. Ferrous ion has been used as an indicator by Ketchum and Ford (1952). It may be detected rather satisfactorily with either $\alpha\alpha$ -dipyridyl or orthophenanthroline with a sensitivity which is adequate for the purpose. The cost of the test ion is low, but its use is difficult, since it is not easily adaptable to continuous measurement. Ammonium ion is very easily detected by means of Nessler reagent and is also relatively inexpensive, but is of questionable value because of the very short half-life of this material in a biologically active community. Lithium represents an ion that could possibly be used in sea water, but the amount required for significant analytical evaluation is excessive, and the method of determination is not particularly good for continuous measurement.

Fluorescein, uranin, has been used on many occasions to trace water masses. It can be detected in very low concentrations, is adaptable to continuous analysis, and the cost of the chemical is very low. Recently Robson (1956), and Cochrane (1956) employed the use of the bacteria, Serratia indica, in tracing sewage effluents in the open sea. The organ is then plated out, cultured on peptone medium, and the number of fluorescent pink colonies developing indicates the number of organisms present in the water. The cost of adding sufficient quantities of these organisms to represent a reasonably good dilution factor is not excessive; however, assay method will not lend itself to continuous analysis. Each of the above tracers may have specific uses in particular situations. In general however, fluorescein seems to be as suitable as the other tracers and has considerable advantage in that its use entails no hazard, and the cost of the tracer is considerably less.

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TABLE II

CAPITAL EQUIPMENT

| <u>Tracer</u> | <u>Equipment</u> | <u>Approximate Cost</u> |
|--|--|-------------------------|
| Sc ⁴⁶ , Co ¹³⁷ , Se ⁷⁵ | Scintillation crystal, scaler, recorder, and handling equipment | \$4,000.00 |
| H ³ | Liquid scintillator, scaler, and sampler | 6,000.00 |
| Fe ⁺⁺ | Colorimeter and sampler | 500.00 |
| NH ₃ | Colorimeter and sampler | 500.00 |
| Lithium | Flame photometer and sampler | 2,000.00 |
| Fluorescein | Fluorescent meter, recorder and sampler | 1,800.00 |
| <u>Serratia</u> <u>indica</u> | Chemicals, sampler, and glassware | 300.00 |

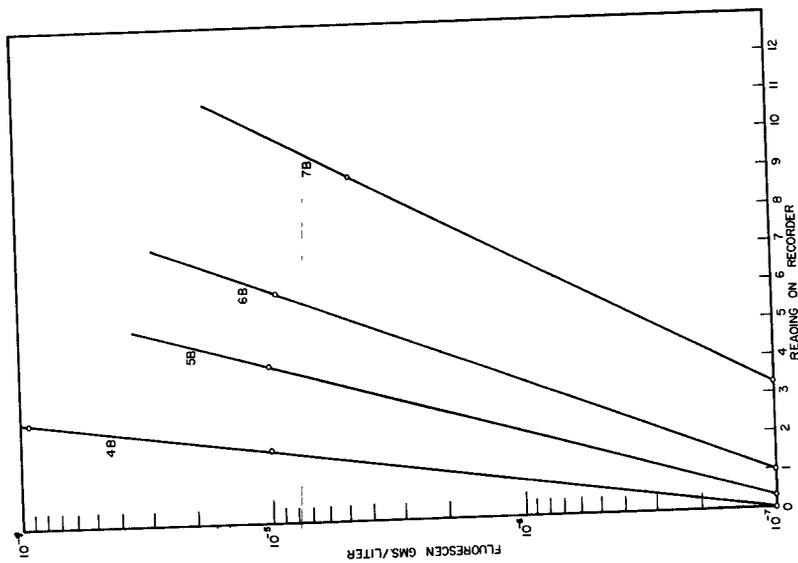


Fig. 1

Fig. 1. Concentration of fluorescein vs. scale reading at different instrument ranges. Solutions made from dilution of concentrate of fluorescein to proper value. Gulf of Mexico sea water obtained from surface at 400 fathom curve.

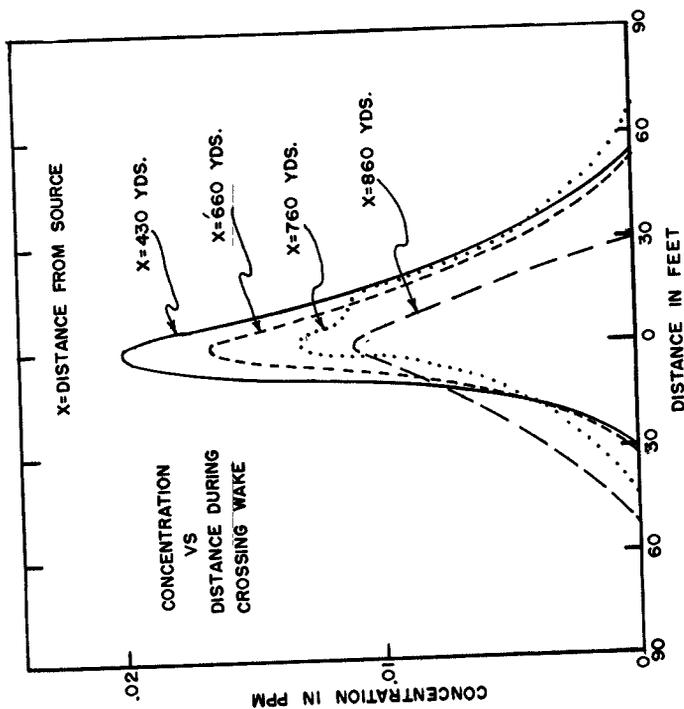


Fig. 2

Fig. 2. Concentration at various distances from point of introduction. Moon, et. al.

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Fluorescein is known to decay in sunlight but for short period measurements, which are often adequate for diffusion studies, this decay would not be significant. At night, the compound is quite stable. Whether the difficulties encountered through the decay of fluorescein are greater than those incurred in some metallic ions through absorption or precipitation in sea water systems is at the present time indeterminant.

A comparison of costs for capital equipment to detect the various tracers discussed above are presented in Table II. In general, the methods which are applicable to continuous recordings require a greater capital investment. However, this difference in cost is usually offset by the decrease in personnel time required to obtain the data. The data obtained are also much more readily used in calculations than are those which are derived from discrete sampling. In view of the above considerations, studies which we have undertaken, to the present time, have all been conducted with the use of fluorescein.

DIFFUSION MEASUREMENTS

Instrumentation

A continuous recording fluorescent meter was designed and constructed for these studies. A detailed description is given elsewhere (Huebner and Hood, 1957, and Moon, 1955). In general, it consisted of a filtered ultraviolet light source which was directed to a low, fluorescent glass cell into which the sample was continuously pumped. The fluorescent light, caused by the ultraviolet absorption of the sodium salt of fluorescein, passed through glass filters (600-625 m μ) and activated a photomultiplier circuit. The signal was amplified and fed to an Esterline-Angus Recorder. The instrument was standardized against sea water solutions of the sodium salt of fluorescein. The standard curves obtained in one environment are shown in Figure 1.

Field Work

A number of experiments have been conducted in different locations and under different environmental conditions to test the method. The first of these was a typical marine coastal region near Port Aransas, Texas; second, in the Gulf of Mexico in about 100 fathoms of water; and third, in the wake of a barge pumping black liquor wastes at the 400 fathom curve in the Gulf of Mexico.

In the first study, fluorescein was dispersed from a point source and the concentration of fluorescein downstream was determined in right angle crossing of the envelope at varying distances from the point of

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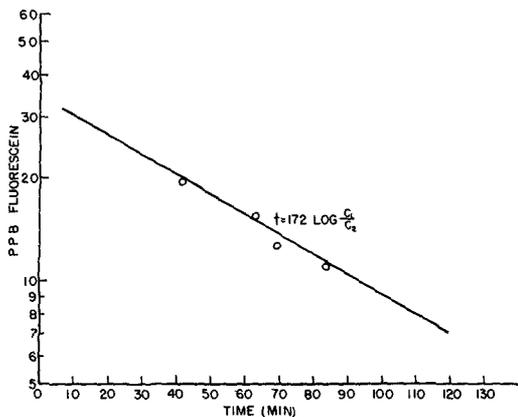


Fig. 3

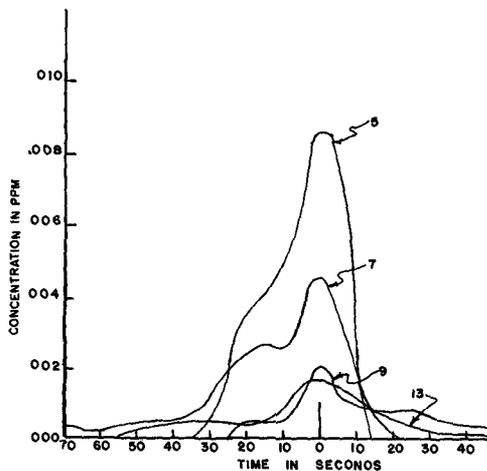


Fig. 4

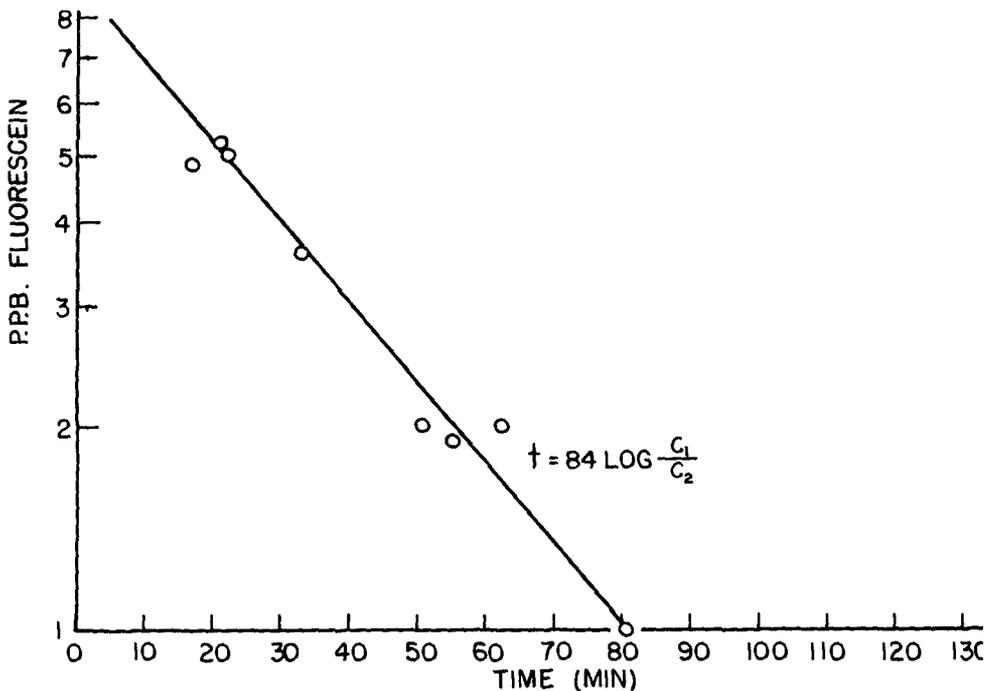


Fig. 5

Fig. 3. Rate of diffusion in tidal channel. Moon, et. al. (1957).

Fig. 4. Fluorescein concentration at various crossings of established wake. Sea Temperature, 82°F; thermocline at depth of 175 feet; wind speed greater than 2 knots; sea surface, slick; location 27° 50' N, 30' W.

Fig. 5. Rate of diffusion in an established wake. Same as Fig. 4.

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introduction. Figure 2 shows the distribution of fluorescein across the envelope at various distances. It will be seen that the curves are fairly symmetrical. The log of the concentration plotted against time from dispersal gives the relationship indicated in Figure 3. The details of these experiments are reported by Moon, et al. (1957).

The second experiments were conducted in 100 fathoms of water in the Gulf of Mexico. Three pounds of fluorescein dissolved in 20 gallons of sea water were dispersed over a distance of 1250 feet behind the R/V A. A. Jakkula in a manner so as to minimize the effect of ship motion on the tracer. Floating buoys were placed on either side of the labeled water, and the concentration of fluorescein in different cross sections was obtained at various times. During these studies, a current of approximately 1 knot was observed, but the sea surface was almost completely devoid of waves or swell. A portion of concentration profiles observed during this study are indicated in Figure 4. The peak concentration shown in this figure, plus other peak concentrations observed in the same study, are plotted against time in Figure 5.

The third study was conducted in connection with the deep sea disposal of a barge load of black liquor waste originating from the Champion Paper and Fiber Company of Pasadena, Texas. A barge containing 265,000 gallons of waste was labeled with 200 pounds of the sodium salt of fluorescein. Black liquor waste was loaded into the barge at a temperature 165°F and the fluorescein was added through the manholes in such a fashion as to assure uniform mixing with the entire content of the barge. At the time of addition of fluorescein to the barge, a sample of the black liquor was heated to 165° in the laboratory and suitable concentration of fluorescein dye was added. This, then, was retained as a standard for subsequent measurements at sea. The waste was pumped at the rate of 1.56×10^3 gm/cm on a course due south of the 400 fathom curve. Using the fluorescein as a guide, the A. A. Jakkula cruised in the center of the wake at various distances behind the barge while continuous fluorescein analysis was conducted. A copy of the traces obtained on the recorder of the fluorescent meter at 300 and 1,000 feet are shown in Figures 6 and 7, respectively. Reference to Figure 1 and Figure 6 indicates a range of concentration between about 10 and 92 ppb within a distance of approximately ~~ten~~ meters of boat travel (speed of vessel was 200 cm sec^{-1}). Because of a time lag in the sampling system of about 5 seconds and hold time in the sample cell of about 2 seconds, the extremes have probably been modified to some undetermined degree.

The above data demonstrate the stirring and mixing processes in incompressible fluids described by Eckhart (1948). The rapid fluctuations

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observed when samples were taken along the axis of the wake at constant time with respect to the introduction of the tracer are evidence of sharp gradients between the interfaces of the waste and the water. The energy dissipated by the tug and also by the barge and by pumping the waste is probably a major factor in creating the distorted masses of the two fluids. The 1,000 foot data shown in Figure 9 shows a decrease in the extreme of concentration gradients in the liquids. A difference of about 100 seconds in time of mixing is represented by the two traces. Continuous traces were also taken at other distances behind the barge. From these, the average values of concentration in the center of the wake were computed by graphical integration of several minutes of continuous record at each of the distances. A plot of the log of concentration against time for these data is presented in Figure 8.

There is some indication that the straight line plot of the data would tend to increase in slope as zero time is approached. This would be expected because of the greater energy being dissipated immediately at the site of pumping. However, more data will be necessary to determine the validity of this observation.

After observing concentration in the middle of the wake for some period of time, a floating buoy was placed immediately behind the barge in the center of the wake, and a study was initiated on the diffusion within a given water mass. These studies were conducted after dark. The data were obtained while crossing the wake of the barge at right angles the vicinity of the floating buoy. Two representative traces observed when crossing the wake are shown in Figures 9 and 10. The dispersion across the wake was such that peak concentrations, as used in previous experiments, did not yield satisfactory data for predicting the maximum concentration in the wake at any given time. Inadvertently, the analysis equipment ceased to function before a sufficient number of crossings could be made to complete the study of the diffusion pattern which occurred.

A total of fourteen crossings were made which extended over a period of two hours and twenty-four minutes. The area beneath each of the curves was estimated by a planimeter and the values obtained are reported in Table III and Figure 11. The data show that of the useful information obtained in these crossings the area beneath the trace on seven approached the value of 1.8 square inches. Three values were quite high and one very low. The indications are, however, that the fluorescence concentration in the wake at any time was constant, indicating little horizontal diffusion or decay of the fluorescein molecules after the initiation of the experiment.

It was not possible in these experiments to get accurate data on

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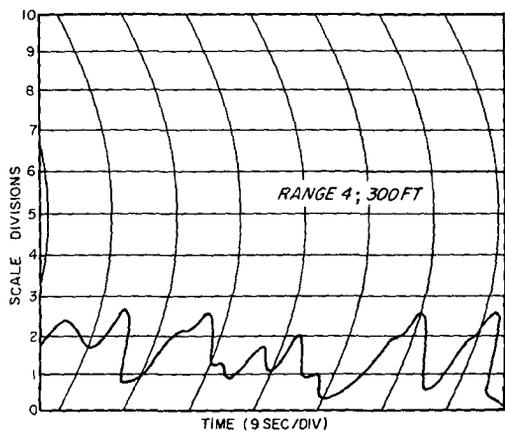


Fig. 6

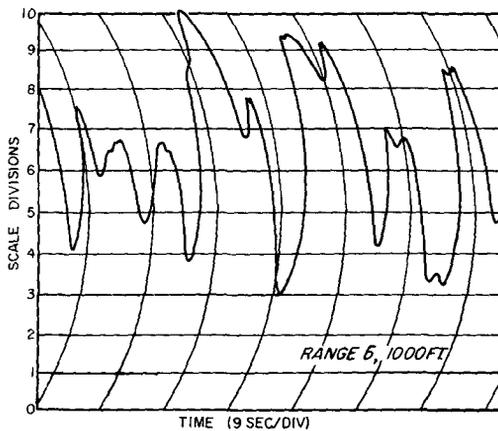


Fig. 7

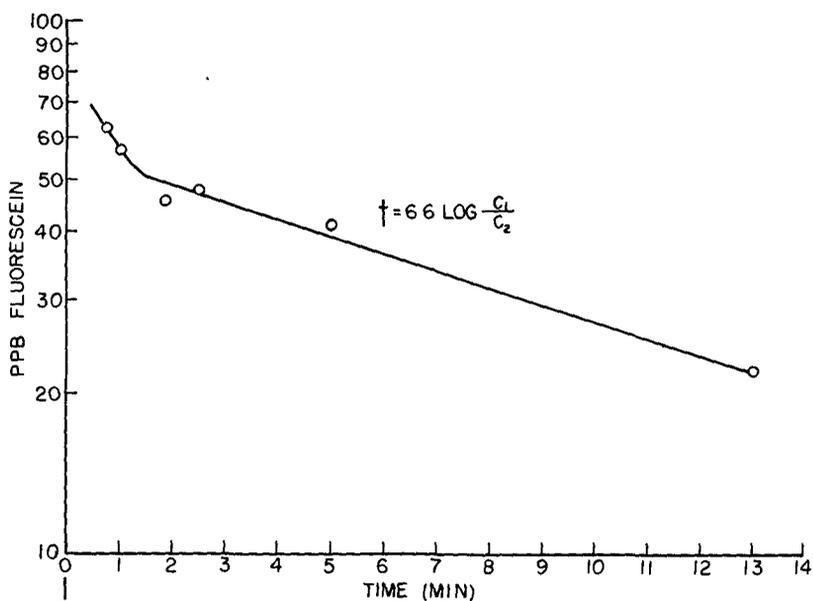


Fig. 8

Fig. 6. Concentration of fluorescein in center of wake. Distance, 300 feet aft of barge; pumping rate: of waste, 1.56×10^3 gm/cm; of fluorescein, 1.23×10^{-1} gm/cm; sea surface temperature, 78°F ; strong thermocline at depth of 150 feet; wind speed, 10 knots; wave height, 4 feet; location, $27^\circ 30' \text{ N}$, $94^\circ 40' \text{ W}$.

Fig. 7. Concentration of fluorescein in center of wake. Distance, 1000 feet aft of barge. Other data the same as Fig. 6.

Fig. 8. Rate of diffusion in wake of barge. Conditions the same as Fig. 6.

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TABLE III

AREA BENEATH TRACING AT EACH INTERCEPT OF WAKE

| <u>No.</u> | <u>Clock Time</u> | <u>Area, Square Inches</u> | <u>Elapsed Time, Minutes</u> |
|------------|-------------------|----------------------------|------------------------------|
| 2 | 1823 CST | 2.01 | 9 |
| 3 | 1826 CST | 1.85 | 12 |
| 4 | 1837 CST | 1.80 | 23 |
| 5 | 1908 CST | 0.49 | 54 |
| 6 | 1917 CST | 1.78 | 63 |
| 7 | 1920 CST | 1.00 | 66 |
| 8 | 1925 CST | 1.64 | 71 |
| 9 | 1928 CST | 3.60 | 74 |
| 11 | 2030 CST | 3.60 | 136 |
| 12 | 2033 CST | 6.78 | 139 |
| 13 | 2037 CST | 1.80 | 143 |
| 14 | 2040 CST | 1.74 | 146 |

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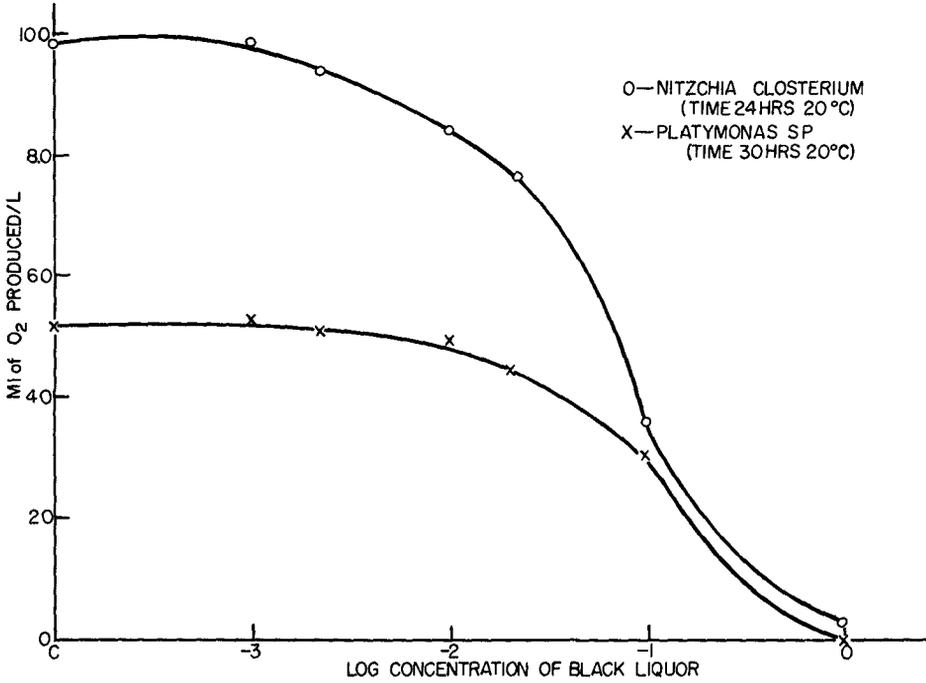


Fig. 9. Copy of pen trace of recorder (directly related to fluorescein concentration) for 6th crossing of wake of barge (1917 CST). Other data the same as Fig. 6.

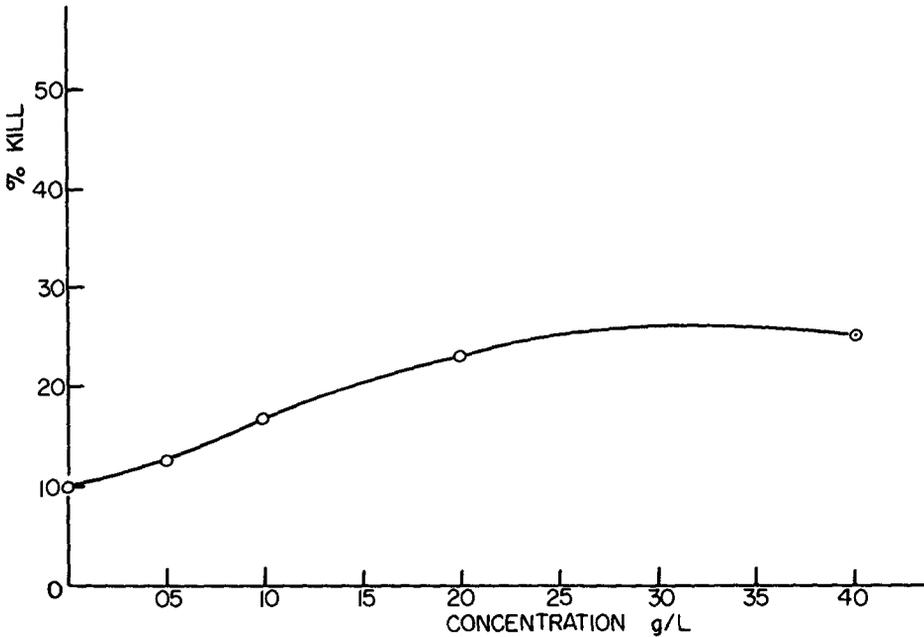


Fig. 10. Copy of pen trace of recorder for 13th crossing of wake (2037 CST). Other data the same as Fig. 6.

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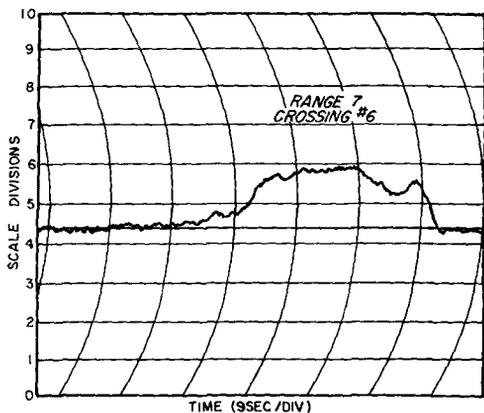


Fig. 11

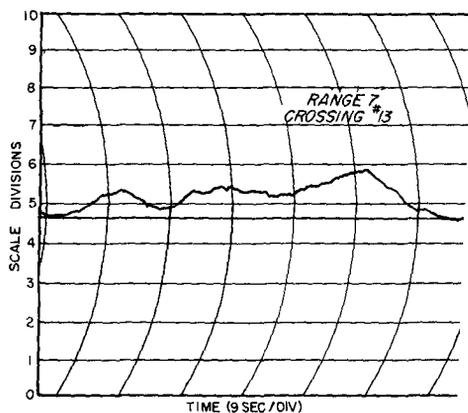


Fig. 12

Fig. 11. Area beneath chart trace at crossing vs. time after pumping 1 barge (1816 to 2040 CST).

Fig. 12. Effect of black liquor wastes on photosynthesis of two Marine Phytoplankton. Hood, et al., (1955).

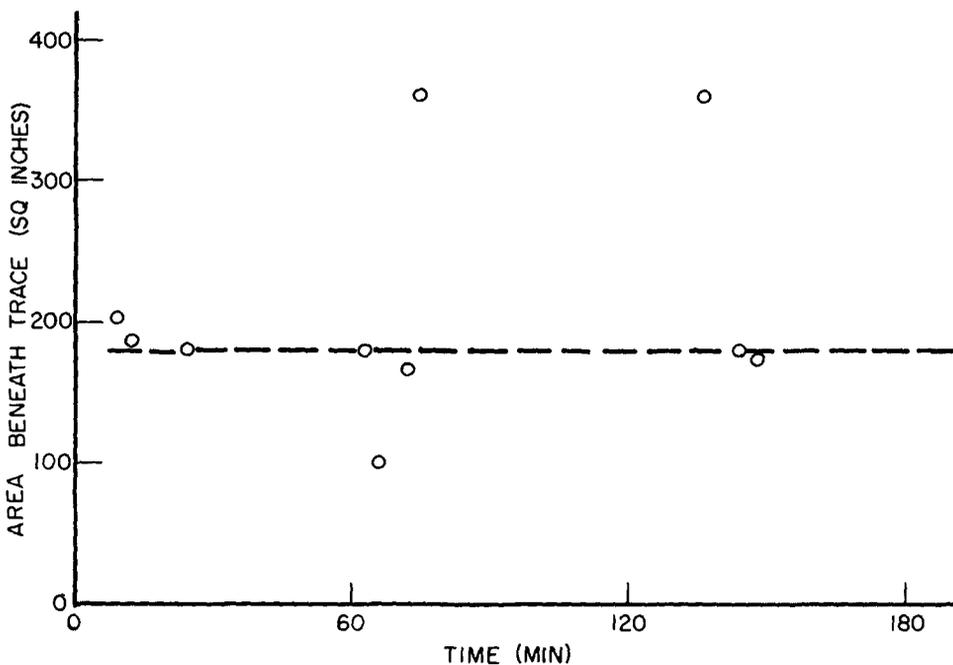


Fig. 13. Toxicity of black liquor waste to mixed zooplankton. Organisms collected from Lydia Ann Channel, Port Aransas, Texas.

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the concentration of the fluorescein with depth in the wake. However, attempts were made to make vertical drops of the sampling device in order to determine the depth of peak concentration. Results indicated that maximum concentration appeared near the surface, or at about the depth of sampling (8 feet), but remained fairly uniform to about 150 feet or the limit of the maximum depth of sampling.

Criteria of Dilution

The second important consideration that faces the waste disposal engineer is that of determining the dilution which is necessary in order to avoid contamination or deterioration of the water mass beyond limits that will interfere with its present use. A number of techniques, and methods, have been devised for this purpose. Among these are fish toxicities, biochemical oxygen demand, chemical oxygen demand, direct chemical analysis, and other biological and/or physical means of estimating the level of pollution. It is out of the scope of this discussion to go into details of the various methods employed and the value of each. However, it seems pertinent to describe briefly the procedure that was recently developed by Hood, Stevenson, and Jeffrey (1957) for detection of low level effects to the basic members of the biological community.

It is generally accepted that the most fundamental organisms in the marine environment or any aquatic environment are phytoplankton. Upon these all other forms of life depend for food. Since these organisms are important to the economy of any marine area, it follows that adequate dilution or waste disposal treatment must of necessity take into consideration these basic organisms. To evaluate the effects of waste on phytoplankton, a technique has been devised which determines the effect of a given waste on the photosynthetic production of a number of pure cultures of phytoplankton normally found endemic to coastal regions.

The method consists of placing a uniform number of algal cells of proper age in a medium to which has been added gradient concentrations of waste material. The solution is diluted to volume in standard oxygen bottles placed at constant temperature in a lighted water bath. After the proper duration of time (about 24 hours), the amount of oxygen produced during this period is determined. The difference between the oxygen produced by organisms in solutions not containing waste and those to which wastes have been added indicate the effect of waste material on the photosynthetic process of the organisms. The results of an experiment of this type are shown in Figure 12. From these data, it may be seen that the concentration of waste which affects the photosynthetic

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process of Nitzchia closterium is about 5 ppm and a similar value for Platymonas sp. is observed. Nitzchia closterium is a diatom having wide distribution in marine areas. Platymonas sp. is a green algae which is also found in marine coastal waters.

A second community which is very important to the general ecology and food supply of organisms of any region are the small animals called zooplankton. These consist of large groups of small crustaceans known as copepods and euphausiids; also larval stages of invertebrates, fish eggs, and larvae. In addition, small forms of many other groups occur. The toxicity of the waste to these organisms is shown in Figure 13. The data obtained show only little toxicity in concentration of black liquor waste as high as 400 ppm. Greater concentrations cannot be used with the method employed for indicating the per cent kill. Attention is called to the difference between the concentration levels which affect phytoplankton photosynthesis and those which are lethal to the zooplankton. The time of exposure of the waste to zooplankton was only about one hour, and, this may account, in part, for the differences observed.

Based on the above data, a marine disposal operation which involves black liquor wastes would require a dilution of something below 5 ppm so as to avoid the impairment of the photosynthetic production of phytoplankton. It must be realized that this is only one datum point and that other considerations must be made. For example, the biochemical oxygen demand for black liquor wastes is about 50,000 ppm, and in many cases may be a limit factor in dilution.

Discussion

The above data may be used to demonstrate the effect of the environment on rate of dilution of waste dispersed in marine waters. If the diffusion curve shown in Figure 8 is extrapolated to zero time, the concentration of fluorescein in the wake immediately upon pumping would average about 56 ppb. If the assumption is made that the waste distributes in the wake the same as the fluorescein, a direct ratio may be used to compute the concentration of the black liquor in the water upon pumping. The concentration of black liquor in the sea at zero time would be 710 ppm. By substituting this value, and the 5 ppm level estimated to be non-effective to phytoplankton photosynthesis into the equation of the diffusion line, a time of about 14 minutes would be required for the black liquor waste to diffuse to a non-effective level under the conditions investigated. The same data used in estimating the time required for dilution in an established wake under the conditions studied would require about three hours. Similarly, under conditions

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existent in the tidal channel near Port Aransas, a time of about 62 hours would be required assuming adequate water for dilution was available.

These data indicate the value of dispersal of wastes under situations in which dilution occurs rapidly and under conditions where maximum mixing occurs. It is also evident that dispersal of waste in deep sea situations is much to be preferred over that in littoral or in-shore zones not only because of the greater rate of diffusion observed, but because the number of animals and plants in this area are minimal and, at the present time, are not exploited either industrially or recreationally.

By the use of the techniques described here, it becomes practical to study the problem of waste disposal in marine waters in a quantitative manner. If an estimate can be obtained with the quantity of effluent to be discharged from any point, a more or less complete evaluation of the effect of this effluent in the immediate vicinity can be determined. A rather detailed evaluation of the environmental conditions along with studies of the diffusion rate existent associated with these conditions will allow one to determine the extent of pollution which would arise from the discharge site and to estimate the total area influenced by the disposal operation.

Proper biological controls by approved methods, or by newer special methods, such as are suggested in this paper, allows a limit to be set on the quantity of effluent which can be discharged from single site under a given set of conditions. It is important that the combination of these two factors be taken into consideration in order to provide adequate waste disposal.

Acknowledgments

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