SOME ENGINEERING APPLICATIONS OF LIQUID SCINTILLATION COUNTING

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THE main difference between the research scientist and the research engineer is that the former, if he is a 'pure' scientist, works at a task he has chosen that will give a maximum amount of significant new information for the amount of work expended; whereas a research engineer starts from a practical problem (usually a pressing one) and, by reviewing and combining knowledge in a number of fields, works towards a practical solution. Of course a large number of those trained as scientists are actually doing engineering research; a few engineers have become scientists. The separation line is not distinct, especially in a rapidly-growing field like liquid scintillation counting: few scientists are completely disinterested in practical application and very few engineers are uninspired by scientific advances, no matter how impractical they appear. A review of possible engineering applications of liquid scintillation counting would cover a large portion of all of the papers published in the field—even many of those in medicine, if one would accept the point of view that much of medical practice is a form of engineering. I shall instead limit my discussion to liquid scintillation work in progress at the Northwestern Technological Institute and not risk invading the review of industrial applications of liquid scintillation counting by Dr. Guinn. I shall review or mention work in progress on (a) tidal estuary models;[1] (b) high velocity combustion (aerothermochemistry);[2] (c) study of the fate of radioactive wastes in surface waters;[3] (d) diffusion in fluids flowing through porous media;[4] and (e) tracer study of an insect repellant in the milk and tissue of a cow.[5]

TIDAL ESTUARY MODEL STUDIES

Some of the more complicated problems in fluid mechanics encountered by the civil engineer are those related to tidal estuaries, including sea water intrusion, bed load movement, and the dispersion of injected wastes. Attempts at completely analytical solutions for practical problems often prove unrewarding. The analogue computers actually used take the form of elaborate models that reproduce the complicated delta geometry and have similitude involving the Froude and Reynolds criteria, which in the case of practical models are contradictory and must be brought into agreement through considerable ingenuity. The model must be verified at many points with its prototype over a range of conditions of tide, river flow, and salt water concentration.

Two of the problems that cause difficulty in estuary model study of dispersion with tracers are the density effect of the injected tracer and its chemical reactions. As tritium water appears to overcome both problems, it was chosen for use on a tracer experiment in April, 1956, at the Waterways Experiment Station, U.S. Corps of Engineers, Vicksburg, Mississippi. After the model of the Savannah Tidal Estuary had been brought to equilibrium conditions that had been verified with the prototype, approximately 1c of

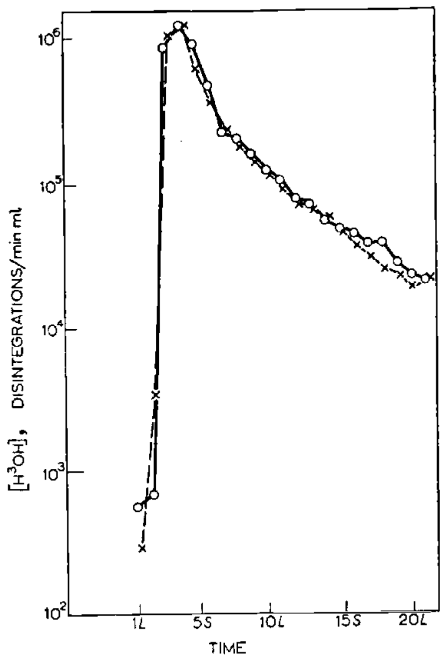

Fig. 1. Savannah Estuary model. Distribution of tritium water concentration at Station 163, Mile 6, AEC Northwestern University Project at (11-1)-353.

tritium water was injected instantaneously at Mile 26 (some 35 prototype miles from the ocean). The model was run for 21 prototype lunar days (about 8 hr of model time), and 1300 samples were collected at stations from the upper reaches of the model to its 'ocean'. Fifty distribution concentration curves were obtained. Figure 1 shows the slug passing one station (logarithm of tritium water concentration at high and the following low tide vs. time in number of tidal days after the start of the experiment). Figure 2 is similar except it shows the slug spread along the estuary at each of two times (the logarithm of tritium water concentration of all samples collected at one high tide and the following low tide vs. river miles from the ocean). Despite the complicated flow regime, the concentration distributions are in qualitative agreement with the Gaussian types derived by G. I. Taylor for steady, uniform, turbulent flow in axially symmetric pipes, which is a much simpler case.

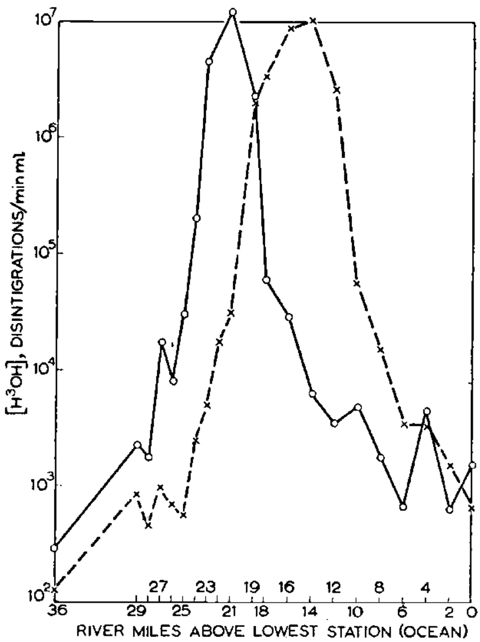

Fig. 2. Savannah Estuary model. Distribution of tritium water concentration in the model at various stations, collected simultaneously tides SL, 2S.

The samples were counted using Hayes' dioxane formula. No problems with spurious counts or non-reproducibility in counting were encountered. Calibrations were made with internal standards. The NaCl used in the model ocean water had a definite quenching effect on the count rate; efficiency dropped sharply by about 30% at NaCl concentration of 10 g/l. and then gradually by about 10% at 35 g/l. (the approximate concentration in both the prototype and model oceans). At the same time the tritium was injected, a rather large concentration of methylene blue was released nearby into the model river. When the center of the slug had reached the ocean part of the model, the methylene blue had been dissipated until it could not be detected by eye. Whether or not the model study can give reliable indications of the fate of injected wastes in the prototype will depend on future studies. The

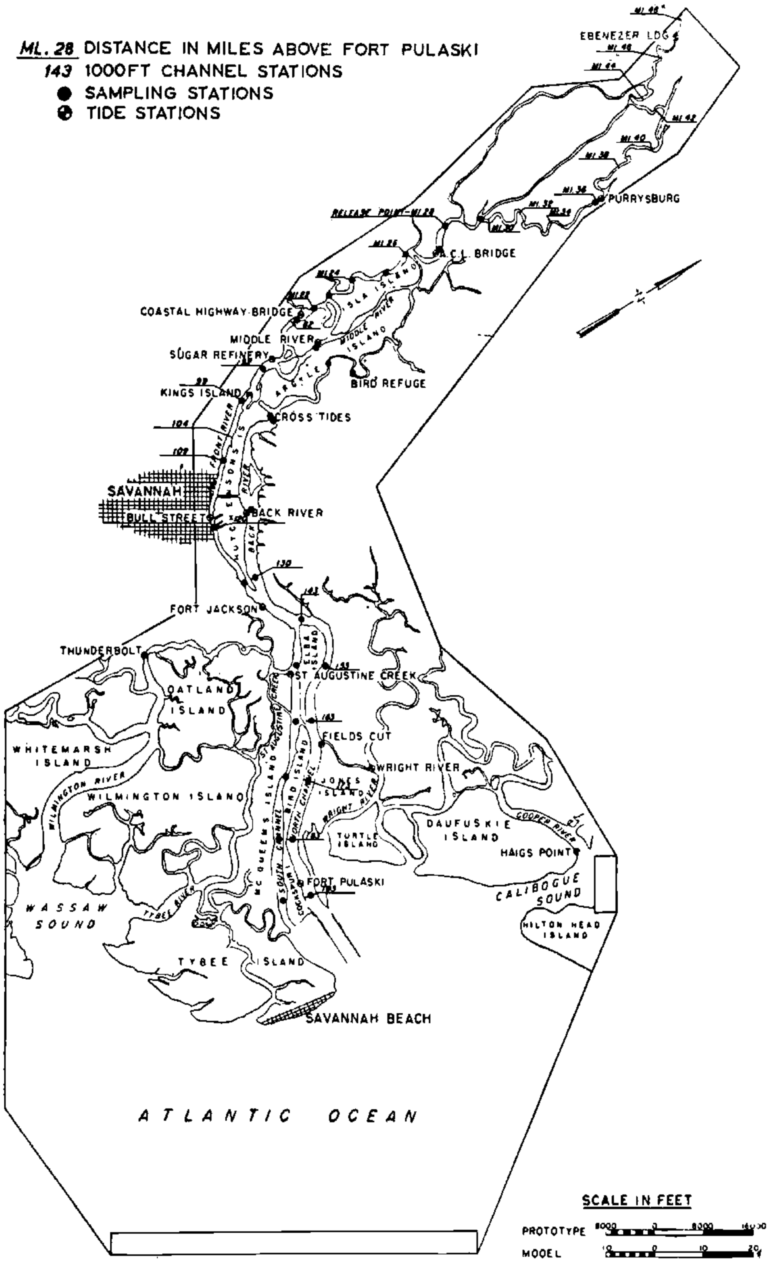

Fig. 3. Savannah Estuary model. Location of sampling stations.

usual prototype–model comparisons for sea water intrusion studies, although apparently necessary as part of the confirmation studies for the problem of injected wastes, are not known to be sufficient. It would be of much interest to make an injection into the Savannah Estuary and then program the flow conditions and tides on the model to see if the prototype results could be confirmed. If confirmation were possible, a large number of model studies could be made in anticipation of future prototype injections quickly, cheaply, and safely.

AEROTHERMOCHEMISTRY

As the majority of the fuels used in high-speed thermodynamic problems connected with rocket and jet propulsion has been made principally of hydrocarbons, it would seem that those doing radiotracer tests would naturally find the liquid scintillation counting technique useful. The work I will mention was done by V. Kopytoff as a MS. thesis under the advice of Professor A. B. Cambel and the author. It involved the study of the backflow or

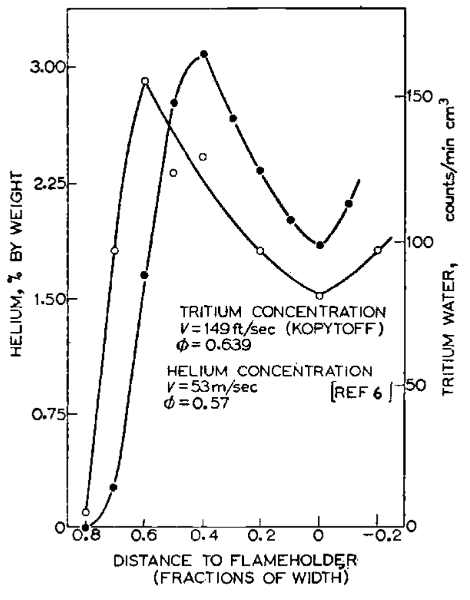

Fig. 4. Comparison of the tritium and helium techniques in measuring recirculation behind a V-gutter flameholder (V. Kopytoff)

recirculation zone existing behind a V-gutter flameholder. Tritium water vapor was added as a tracer at a constant low rate. About 15 mc of tritium water in 50 ml of water was found sufficient for a run. Commercial propane was used as a fuel and samples were collected through a probe into evacuated bottles. Samples were processed on a Los Alamos type tritium gas–water converter and counted in the dioxane solution. The method compares quite favorably with a helium tracing technique used by others.[6] Concentration distribution at various positions near the flameholder are compared by Kopytoff in Fig. 4 with the helium technique. The single study reported is only an indication of the many possible radiotracer studies in high speed combustion. A simple example of a reasonable extension of the work here mentioned would involve the use of labeled fuel in similar experiments.

INJECTION OF RADIOACTIVE WASTES INTO SURFACE WATERS

The Technological Institute has a contract with the A.E.C. (Reactor Development Division) to study the fate of radioactive wastes in surface waters. Preliminary studies were conducted on a small creek flowing through the Joliet Arsenal of the U.S. Army near Joliet, Illinois. The first injection consisted of approximately 0.3 c of yttrium-90 and the second of 2 c of tritium water and 0.1 c of both cesium-137 and cerium-144. Several hundred samples were collected in these experiments and analyses are not yet complete. The objective is to inject several nuclides simultaneously into the stream and to measure, by separation through chemistry or counting technique, relative concentrations in each of the samples—thus circumventing a portion of the complicated and unpredictable fluid mechanics effects. Tritium water was chosen as a reference material and other nuclides were selected as representatives of the eleven species (from six chemical families) that dominate fission product radioactivity between 6 months and 5 years of cooling time.

Following the preliminary Joliet injections, a large tracer study has recently been made by the project with the help of a number of government agencies in which approximately 1000 c of tritium water and 20 c of scandium in a concentrated acid solution were injected simultaneously into a large stream having excellent geometric properties and a regulated flow of approximately 3000 ft^3/sec. Although tritium water measurements on this study are still preliminary, good distribution curves appear measurable at least 15 miles below the injection point, and traces have been detected at 25 miles even though the flow passes through a lake with a capacity of 2×10^6 ft^3 located 12 miles below the injection point. Figure 5 is a preliminary plot of a few of the tritium water measurements taken. The scandium has been flocculated from the samples with alum and lime and will be counted using a large NaI (Tl) crystal inside the Institute's recently-constructed steel room.

An additional test on the same stream at the same point is planned for the late spring of 1958. Approximately 4000 c of tritium water will be injected simultaneously with several gamma-emitters chemically kin to the hazardous fission product species. It is hoped that measurements of the relative gamma-emitter concentrations can be obtained using crystal spectrometry and a 256 channel analyzer. Thus, it is hoped that some insight can be obtained on the fate of fission products (diffusion rates, sorption, flocculation) following injection at one point at one time into a stream.

If one does tracer work on a stream having a 3000 ft³/sec flow, dilution soon reduces concentrations so that even a kc injection does not raise the sample count rate far above background levels, except near the peak of the slug. A large portion of the background in tritium counting by liquid scintillation is caused by the potassium-40 in the sample bottle. One of three 60 ml

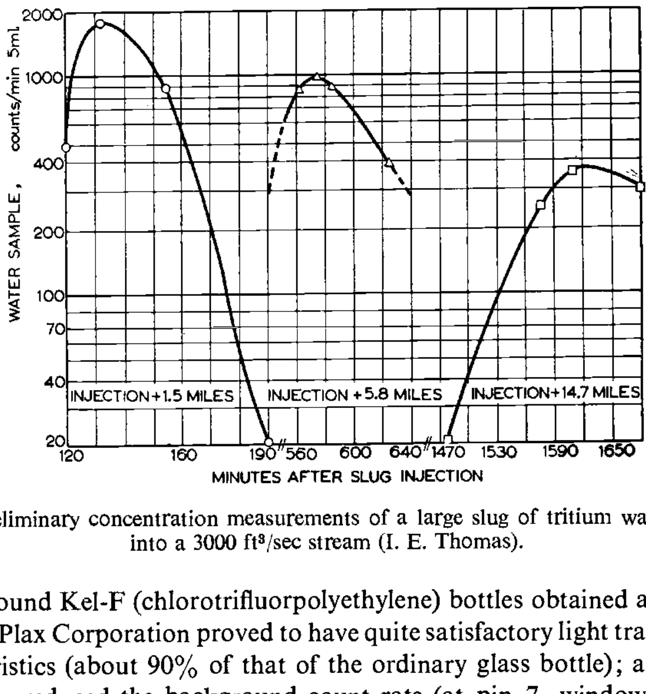

Fig. 5. Preliminary concentration measurements of a large slug of tritium water injected into a 3000 ft³/sec stream (I. E. Thomas).

Boston round Kel-F (chlorotrifluorpolyethylene) bottles obtained as samples from the Plax Corporation proved to have quite satisfactory light transmission characteristics (about 90% of that of the ordinary glass bottle); also use of this bottle reduced the background count rate (at pin 7, window 10-50 V) from 140 to 40 counts/min. In addition, spurious counts created by light activation of glass were apparently eliminated. (The other two Kel-F bottles had a brown tint and were found quite unsatisfactory.) The milky color of the good Kel-F bottle does not seem to hurt the count rate, and preliminary tests indicate that the now generally discarded light-coupling technique using silicone oil might here be advantageous despite its inconvenience.

Another problem faced in the direct measurement of tritium in natural waters is the light-emitting reaction, apparently between impurities of supposedly high quality commercial dioxane and the dissolved matter in natural waters. A possible explanation of this reaction can be shown by placing a small crystal of pyrogallic acid (which is an antioxidant that could be used in dioxane) into a counting bottle of dioxane–natural water counting solution. The acid was found in one test to raise the coincidence count rate from 140 to 100,000 counts/min. An occasional bad bottle of commercial dioxane can have the same effect. The cause does not seem related to the peroxide content. Hydroquinone helps reduce the spurious count rate but also introduces some quenching; Versene seems to have no effect in reducing the spurious counts. As observed by HAYES,[7] acidification of the sample greatly reduced or eliminated the spurious counts.

TRACERS IN POROUS MEDIA FLOW

The civil engineer and geologist are still in search of an effective ground water tracer. At one time it was thought that tritium water would be almost ideal, but apparently it is preferentially sorbed over ordinary water as it moves through the medium, probably in exchange with the bound water at the interface. It has been shown by others that tritium water is not as good a tracer as, for example, the chloride ion. Although the Northwestern University laboratory experiments (using glass spheres as media and methylene blue tracer) that have been conducted up to the present were quite successful, sand media tests have indicated appreciable sorption. Tests are planned using tritium water and, if exchange is principally a mass effect, the addition of D_2O carrier could make tritium water a useful porous media flow tracer, at least in laboratory studies.

TRACER STUDY OF AN INSECT REPELLANT IN ANIMAL TISSUE

A study was undertaken by RAPPORT[5] and the author on the measurement of small concentrations of di-*n*-butyl succinate (made by esterifying succinic-2C14 acid with butyl alcohol) in cow's milk, lean meat, kidney, liver and fat. The results of the work are typical of many investigations using weak beta-emitting tracers: measurements that were crude or insufficiently sensitive with the usual G-M tube technique became, after a short development period, quick, sensitive, and reliable when using the liquid scintillation counter. In preparation of samples from the milk and tissues there was a light-quenching yellow color produced by extractives in the final toluene solution. This effect, which greatly reduced the count rate, was overcome by prior treatment of the extract with activated charcoal. After treatment it was possible in all cases to make a determination of the labeled di-*n*-butyl succinate in a concentration of 1 p.p.m. with a net rate of over 100 counts/min above background of 26 counts/min (counter efficiency of 67%).

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