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**TRANSPORT OF
RADIONUCLIDES
IN FRESH WATER
SYSTEMS**

*Report of A Working
Meeting Held At
University of Texas
January 30 -
February 1, 1963*

United States Atomic Energy Commission
Division of Technical Information

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TID - 7664

TRANSPORT OF RADIONUCLIDES IN FRESH WATER SYSTEMS

A Working Conference
held in
Austin, Texas

30 January - 1 February 1963

Sponsored by:

Department of Civil Engineering
(Environmental Health Engineering)
University of Texas

with

Division of Reactor Development
and
Division of Biology and Medicine
U. S. Atomic Energy Commission

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PREFACE

An essential phase of the Atomic Energy Commission radioactive waste disposal development program is the dissemination of technical information through the conduct of several small working group meetings each year. In this manner, an opportunity is provided to engineers and scientists in the field to informally exchange ideas and information on mutual problems and specific areas of interest. A meeting of this type was the Conference on Transport of Radionuclides in Fresh Water Systems, held at the University of Texas in Austin on January 30, 31 and February 1, 1963.

It is our belief that if there is to be a growing and developing nuclear energy industry, it will always be necessary to use the diluting power of the environment to some extent in the management of low level waste. Satisfactory dispersal operations require careful effluent control and a detailed knowledge of the behavior and distribution of specific radionuclides in the particular environment under consideration. Development of the necessary environmental data requires a spectrum of technical and scientific disciplines - biology, chemistry, ecology, hydrology, mineralogy, radiochemistry, sanitary engineering in order to achieve program objectives.

This meeting provided a unique opportunity for these scientists and engineers to analyze and evaluate what has been done in stream studies and to discuss which areas need further study. The Conference was organized by the Environmental & Sanitary Engineering Branch of the Division of Reactor Development in cooperation with the Division of Biology & Medicine. Representatives from Hanford, Oak Ridge National Laboratory, the Universities of California, Harvard, Louisville, Michigan State and Texas, U. S. Geological Survey, U. S. Public Health Service and the U. S. Atomic Energy Commission participated.

Excellent meeting facilities and typical Texan hospitality was provided by the University of Texas. A note of appreciation and gratitude is extended to Dr. Earnest Gloyne and his staff for their untiring and special efforts in making this meeting a success.

Walter G. Belter

Walter G. Belter, Chief
Environmental & Sanitary Engineering
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U. S. Atomic Energy Commission

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OPENING REMARKS

PROFESSOR E. F. GLOYNA
University of Texas

I trust that everyone is well rested this morning so that we can welcome you properly to the University of Texas. We arranged the weather just for the Washington, D. C. participants so that it would be typical. We will provide a tour for those who wish to visit any part of the campus.

You are now in Room 202 of the Texas Union Building and all of our meetings will be held here; our meals will be taken at a Faculty Club which is on this floor. Tonight we are guests of the Dean of Engineering for dinner at the Forty Acres Club; the building is located approximately three blocks from the University.

We would also like to know how many of you would wish to make a field trip to the Marine Institute of the University of Texas which is located at Port Aransas. We can make reservations and arrange transportation for those of you who would like to visit this installation on Friday afternoon. We have noted that some have indicated an interest in this trip.

I will now turn the meeting over to Mr. Walter Belter, Chief, Environmental and Sanitary Engineering Branch, Division of Reactor Development, Atomic Energy Commission, who will act as moderator for our 3-day meeting.

W. G. BELTER, USAEC
Division of Reactor Development

In looking at the tentative schedule for this conference that Dr. Gloyna has prepared I note that our fellow workers from Hanford Atomic Products Operations at Hanford are not here. As you realize, they have a very important part to play in this meeting; I understand their transportation has been delayed, but that they will arrive shortly.

Speaking on behalf of the Division of Reactor Development, at this time every year, our preliminary planning commences for research and development programs that are to be conducted approximately 18 months from now. In other words, we are starting to think of plans for Fiscal Year 1965 for program work that would start July 1, 1964.

A Division like ours in the AEC, as you might possibly surmise, is a research and development Division, with emphasis on engineering development work, we are primarily in the business of developing various types of nuclear reactors and their supporting systems. Since radioactive waste management is a very important part of reactor development and since many of the radioactive wastes emanate from parts of the fuel cycle it is appropriate that our branch should be connected with the Division of Reactor Development.

In waste management and in effluent control we are always looking for new and improved methods of waste treatment and in connection with this work we are very much interested in what effect do processes and waste effluents have on our various types of environments such as air, water and ground which then could alter treatment plant design. We are interested in the physical, chemical, and biological fate of various radionuclides in our environments. How do they accumulate in the environment? Are there any release mechanisms? If so, how, then, would these affect plant design? These are then some of the questions we are continuously being asked.

We all know there has been considerable investigation done in this area. In the AEC itself, we have studied the Mohawk River, below the Knolls Atomic Power Laboratory, the Columbia River below Hanford, and the Savannah River below the Savannah River Plant. We are currently studying on a cooperative basis with other government agencies the Clinch River below Oak Ridge. Several of you here today have been intimately connected with these studies. Some of these studies have been extremely comprehensive in nature. For instance, the Clinch River program, which is now completing its third year, has involved extensive sampling of water, suspended and bottom sediments, aquatic biota, fish, etc. This work is entering its completion phase. Several tracer runs have been performed on the Clinch River to measure the vertical, horizontal, and longitudinal dispersion in the river. The distribution of various nuclides, which have been discharged from the Oak Ridge National Laboratory, has been studied to determine the material balance of radioactivity which now exists in this river system. We are now trying to define or delineate some of the mechanisms which determine the fate of radionuclides which have been released.

Principally, we are interested in: 1) dispersion of wastes; 2) sorption by suspended sediments; 3) what reactions are involved that may form settleable precipitates; and 4) retention or release of activity from bottom sediments or aquatic biota. Also, we have been looking at the mineralogy of bottom sediments and the size distribution of these sediments. From measurements of stream velocities and dispersion, calculated areas, and bottom sediment properties, it is hoped that the rates of mass sediment movement can be defined.

With this information and data we believe a proper or appropriate safety analysis of the Clinch River system can be made.

The assimilative capacity of the stream has then been quantitatively determined and criteria for waste treatment and disposal systems for this river could then be specifically delineated. Hopefully, some of this information and scientific data would have transfer value to other river environments below various nuclear facilities.

We are now posing the question, "How much more do we have to know in stream studies to provide for a continued, reasonable utilization of this environment for waste disposal purposes?" Do we need to have more comprehensive studies of the Clinch or Columbia River types for all major nuclear facilities? For instance, whenever we build a power reactor on a given river must we have the type of study that we have had on the Clinch River and the Columbia River?

With these questions in mind, it was decided in cooperation with our Division of Biology and Medicine, to call together a small working group - to include biologists, ecologists, chemists, hydrologists, sanitary engineers, and other disciplines. This has been done with the idea of considering several points. What do we know now about stream studies? What areas still require further study? Are we weak in certain aspects of this work or can we reduce our efforts in others? What has been done which can be applied to other stream environments? Does any of the data or information which is going to be reported here in the next two and one-half days have any transfer value to other streams? To answer these questions, we hope to utilize background and experience of each of you to assist us in planning our future programs in the Division of Reactor Development and also in the Division of Biology and Medicine. Also, we would not expect that up-to-date reporting of stream data, per se, is the major item of interest at this meeting. In fact, we believe this should be of a secondary nature. Our interest is in what each of you have done in analysing and evaluating the results of data that have been collected. After you have presented this analysis we would propose to open the meeting for discussion.

There will be no time limit on the speakers or the participants. You may wish to take one-half hour, one hour, or two hours for your presentation. If discussion of one paper takes one or two hours we are prepared to stay as long as necessary to give everyone as much time as needed.

We are pleased to have Dr. Morgan and Messrs. Jamison and Kornegay of VMI on the Johns Hopkins Contract to help us in recording this conference. They will prepare a rough draft of the comments and remarks made here, will forward them to you for your consideration, and ultimately, will prepare the final conference report.

In summary, we hope there will be a free exchange of information, a hair-letting-down session, and that none of us will have any inhibitions about asking questions or candidly answering them. At this time, I would like to call

on Dr. Hooper of the Division of Biology and Medicine for a few comments.

FRANK HOOPER, USAEC

Division of Biology and Medicine

We appreciate that Mr. Belter has dignified our participation by being a co-sponsor; I do not think that we deserve this. It is a very unique occasion when a sanitary engineer will invite a biologist to one of their sacred meetings. I just want Mr. Belter to know that we appreciate it. All I can say is that Mr. Belter's office must be in very bad shape, they must be desperate, to invite us.

Seriously, in the past there has been a serious lack of communication between these two disciplines. Everyone knows that communication is desirable but no one does much about it. Well, here is a chance to do something about it. We have the opportunity to communicate and I for one appreciate this a great deal. I am also heartened by Mr. Belter's plea for informality and I hope that this will be the key-note of the meeting. The biologists are out numbered about six to one and so I realize that some biological blood will be spilled but we expect to get a lot out of this. Thank you again, Mr. Belter, for inviting the Division of Biology and Medicine to co-sponsor this meeting.

W. G. BELTER, USAEC

Division of Reactor Development

Thank you, Dr. Hooper, for your kind remarks. I do not believe the biologists are out numbered in this meeting. If we look at the disciplines involved here we have at least one-half dozen in the biology and ecology line, a half-dozen engineering types, some chemists, some hydrologists, and other associated specialities. Therefore, in this respect, the biologists are not out numbered. I certainly feel that the relationship between the biologists and the engineers, and I am speaking about the Reactor Development and Biology and Medicine Divisions within the AEC, and their respective environmental branches, has been particularly good, especially during the past year. I believe much of the credit for this can go to men like Dr. Hooper, Mr. Vince Shultz, and others. We are now having excellent communication between the two Divisions. Meetings of this type will greatly increase the flow of information from one Division to the other.

At this time I would like to introduce Mr. Hal Bernard, of our Branch.

HAL BERNARD, USAEC

Division of Reactor Development

You may have gotten the impression from the great emphasis placed on helping us in the AEC in our future planning; this is not entirely the case. After all, it is you who are doing the work and we hope that in this meeting we will all be open to suggestions, and that there will be cross-fertilization of ideas. Some of these ideas may germinate into projects in areas that need additional emphasis; suggestions could result in modification of existing projects to reap benefits to a larger cross-section of researchers. That there has been much work on the many facets of stream studies has been aptly described by Mr. Belter, but the areas in which we need more research, I hope, will result from this meeting. Obviously, every stream is somewhat different from every other stream biologically and chemically as well as physically. Therefore, if you want accurate detailed results of the fate of radionuclides in streams, we must naturally run a detailed survey of the particular stream. However, for safety analyses, wherein we utilize broad assumptions involving maximum credible accidents for release of radioactivity into streams, it would be well if the engineer who is concerned with the safety analysis had the use of several mathematical models with which he could determine the ultimate fates of radionuclides released to streams. Will the radionuclides disperse into the sea, end up in a delta or in farm land?

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PRELIMINARY SAFETY ANALYSIS OF RADIONUCLIDE RELEASE TO THE CLINCH RIVER

by K. E. COWSER, W. S. SNYDER, and M. J. COOK

INTRODUCTION

When radioactive material is released to a body of water, there is a complex network of mechanisms by which the material can be transmitted from one component, animate or inanimate, to another. At each point in the network or chain of transmission, humans or other life forms may be subject to some degree of radiation exposure.

The probability that human exposure will occur and the degree of exposure depend upon many interrelated factors. These include: the physical, chemical and biological conditions in the body of water, the number of people exposed and their habits which may influence the nature and extent of exposure, the sources, types, quantities, and distribution of radioactive contaminants released to the water; and the adequacy of control measures to keep the levels of contamination within safe limits. Definitive information about these and other pertinent factors is necessary in order to make realistic estimates of the potential exposures and to evaluate their significance.

In the Clinch River Study, safety evaluation depends primarily upon descriptive and analytical data needed to define exposure factors. Criteria of permissible radiation exposures, adopted by national and international agencies, are accepted as guides. On these bases conservative estimates of human exposure that may result from Clinch-Tennessee River contamination can be made and conclusions reached regarding their importance.

This paper summarizes some of the progress made in assessing the degree of safety of radionuclides released to the Clinch River as practiced up to 1961.

OBJECTIVES

The immediate objective is to evaluate the potential contribution of each relevant pathway in causing radiation exposure to man. The most direct means of evaluating internal exposures is to determine the amounts of radioactive material in the bodies of exposed members of population groups, for example, by whole-body counting or excretion analyses. Lacking such data at present,

Fig 1 Lower Clinch River Basin

TABLE I

UNCLASSIFIED
ORNL-LR-DWG. 77401RYEARLY DISCHARGES OF RADIONUCLIDES TO CLINCH RIVER (CURIES)^a

Year	Gross Beta	Cs ¹³⁷	Ru ¹⁰⁶	Sr ⁹⁰	Tre(-Ce)	Ce ¹⁴⁴	Zr ⁹⁵	Nb ⁹⁵	I ¹³¹	Co ⁶⁰
1949	718	77	110	150	77	18	180	22	77	
1950	191	19	23	38	30		15	42	19	
1951	101	20	18	29	11		4.5	2.2	18	
1952	214	9.9	15	72	26	23	19	18	20	
1953	304	6.4	26	130	110	6.7	7.6	3.6	2.1	
1954	384	22	11	140	160	24	14	9.2	3.5	
1955	437	63	31	93	150	85	5.2	5.7	7.0	6.6
1956	582	170	29	100	140	59	12	15	3.5	46
1957	397	89	60	83	110	13	23	7.1	1.2	4.8
1958	544	55	42	150	240	30	6.0	6.0	8.2	8.7
1959	937	76	520	60	94	48	27	30	0.5	77
1960	2190	31	1900	28	48	27	38	45	5.3	72
1961	2230	15	2000	22	24	4.2	20	70	3.7	31

^aValues calculated from data supplied by Applied Health Physics Section, ORNL.

exposures are being calculated from measurements of the amounts of radioactive material in the various environmental media, and by the use of assumptions as to the fraction of this material that may affect the exposed population.

The long-range objectives are evaluation of the total potential of radioactivity in this river system environment in causing exposures and delineation of exposure pathways so as to estimate the prevailing levels of safety for animal and plant life and understand the potential for exposure of each such pathway in the future. Hopefully, the study will establish parameters of downstream safety for river disposal which will be applicable to the environment, as well as to man, and under many combinations of conditions.

RADIONUCLIDES RELEASED AND CONCENTRATIONS IN THE RIVERS

Virtually all radioactivity emanating from the Laboratory and reaching the Clinch River passes through White Oak Creek. The final control point for waste water released to the river (Clinch River Mile 20.8) is at White Oak Dam (Figure 1).

The flow of water through White Oak Dam has been determined by two methods.^{1, 2} During the period, 1953-1955, while White Oak Lake was still impounded, a gaging station at the dam was used. After 1955, when the lake was drained and the gaging station inactivated, flow was calculated by summing the separate measurements of flow in White Oak Creek and Melton Branch which are the principal surface streams draining the basin. The gaging station at the dam was reactivated in 1960 and has been used for flow measurements since that time.

Until 1948, daily radiation measurements were made and periodic samples were collected at White Oak Dam. Gross-beta activity was determined; and, coupled with daily flow, the number of beta curies released each day was calculated. Beginning in 1949, monthly composite samples were also analyzed radiochemically for cesium, ruthenium, strontium, cobalt, trivalent rare earths (TRE), cerium, zirconium, niobium, and iodine; and the curies released each year were calculated (Table I). The increased quantity of Cs^{137} released in 1955 was due to the draining of White Oak Lake. Subsequent reduction in release of this nuclide is associated with treatment of process water and partial reimpoundment of the lake. The increase in Ru^{106} released is associated with operation of the waste pits, while the decrease in Sr^{90} released can be related to the operation of the Process Waste Water Treatment Plant. Currently, calculations are in progress to estimate at least the total strontium released during the period, 1943-1948.

Estimates of the mean annual concentrations of radionuclides in the Clinch and Tennessee rivers were based on dilution ratios and the fact that White Oak Creek effluent is completely mixed with river water after 3 to 5 miles of flow downstream from the mouth of the creek. This was shown by three tracer tests in the Clinch River in 1958, 1961, and 1962 (ref 3, 4, and 5). The concentration values derived in this way are conservative, since no allowance was made for decreases of the radioactivity in the water due to radioactive decay or removal by suspended sediments.

Four downstream locations were selected for the evaluation analyses (Figures 1 and 2), namely: (1) Clinch River Mile (CRM) 14.5, which is 6.3 miles downstream from the White Oak Creek discharge at CRM 20.8; (2) CRM 2.6, downstream from mouth of Emory River near Kingston Steam Plant; (3) Tennessee River Mile (TRM) 529.9, Watts Bar Dam; and (4) TRM 465.5, at Chattanooga water supply intake and 5.5 miles downstream from Chickamauga Dam. At CRM 14.5 and in the vicinity of CRM 2.6, water supplies taken from the river are used by the Oak Ridge Gaseous Diffusion Plant and the Kingston Steam Plant, respectively, for sanitary and industrial purposes. There are downstream recreational areas at the Kingston waterfront, at Watts Bar Dam, and at numerous places along Watts Bar reservoir. There are large recreational areas along Chickamauga reservoir, notably just above Chickamauga Dam (CRM 471.0). The first large population center (Chattanooga, Tennessee) is located a few miles downstream from Chickamauga Dam (TRM 471.0) and is served by a public water supply taken from the Tennessee River at TRM 465.5. In addition, CRM 14.5, TRM 529.9, and TRM 471.0 are stations in the basic water-sampling network of the Clinch River Study. The quantity of water passing each location annually was calculated from average flow values (Table II). The average concentrations of radionuclides at each location was determined from the curies released and the total flow for each year. In this report the calculated concentration values for two of the locations are given, namely: CRM 14.5 and TRM 465.5 (Tables III and IV).

AVENUES OF HUMAN EXPOSURE

The potential avenues of human exposure resulting from release of radioactivity to the environment are many and complex. Parker has conceived of a number of exposure pathways and has indicated those which he believed to be of major consequence.⁶ From radioactive wastes in rivers, streams, lakes, or reservoirs, he emphasizes the hazards related to use as drinking water, immersion in the water, close approach to the water (including contaminated mud and vegetation), use of water for irrigation, uptake by biological chains, industrial processes, sewage disposal, and atmospheric discharges.

The list is well conceived but, unfortunately, includes many avenues for which data are not currently available. A complete estimate of human exposure is not available now, and probably will not be for many years to come. However, based on present experience, the avenues of human exposure considered in this report are believed to include most of the significant or potentially significant mechanisms of exposure resulting from radionuclide discharge to the Clinch River.

CRITICAL ORGANS CONSIDERED

For a detailed estimate of exposure to radioactive material in the environment, it is necessary to calculate the dose for these organs for which the dose may reasonably be expected to be a maximum or to be in excess of the prescribed limits. To reduce the number of calculations, an insight concerning the potentially critical organs may be obtained by considering the type and

Figure 2 Network Water Sampling Stations.

TABLE II

UNCLASSIFIED
ORNL DWG. 63-179RMEAN ANNUAL FLOW IN CLINCH AND TENNESSEE RIVERS
(CUBIC FEET PER SECOND)

YEAR	CRM 14.5 ^a	CRM 2.6 ^a	TRM 529.9 ^b	TRM 471.0 ^b
1943	5310	6620	26130	32330
1944	4800	6870	25690	32290
1945	4940	7020	26490	32270
1946	5150	6880	29100	38540
1947	4420	5720	24040	31190
1948	4290	6480	26370	34360
1949	5460	7560	33300	43630
1950	6630	9360	34240	44030
1951	6170	8760	28070	36560
1952	4570	5770	22470	29770
1953	4340	5710	22160	28130
1954	2990	4730	20480	26050
1955	4850	6610	23790	30530
1956	5040	7340	24750	30990
1957	6350	9300	36310	45250
1958	5560	6880	27780	34330
1959	3490	5260	23760	29000
1960	4460	6200	25150	31010
1961	4780	7110	29520	37430

^aVALUES FURNISHED BY THE UNITED STATES GEOLOGICAL SURVEY – ESTIMATED ON BASIS OF DISCHARGE RECORDS FOR THE GAGING STATION ON CLINCH RIVER NEAR SCARBORO AND INTERVENING INFLOW.

^bVALUES FURNISHED BY THE TENNESSEE VALLEY AUTHORITY.

TABLE III

UNCLASSIFIED
ORNL-LR-DWG. 77402RCALCULATED MEAN ANNUAL CONCENTRATION OF RADIONUCLIDES
AT CLINCH RIVER MI. 14.5(UNITS OF 10^{-9} $\mu\text{c}/\text{ml}$ or $\mu\mu\text{c}/\text{liter}$)

Year	Gross Beta	Cs ¹³⁷	Ru ¹⁰⁶	Sr ⁹⁰	Y ⁹¹	Ce ¹⁴⁴	Zr ⁹⁵	Nb ⁹⁵	I ¹³¹	Co ⁶⁰
1949	150	16	22	30	0	3.7	36	4.6	16	
1950	32	3.2	3.9	6.5	0		2.5	7.2	3.2	
1951	18	3.6	3.2	5.2	0		0.82	0.40	3.2	
1952	53	2.4	3.6	18	0	5.6	4.7	4.4	4.8	
1953	78	1.7	6.8	35	0	1.7	2.0	0.93	0.54	
1954	140	8.2	4.2	51	11	8.9	5.2	3.5	1.3	
1955	100	15	7.1	22	13	20	1.2	1.2	1.6	1.5
1956	130	38	6.5	23	7.6	13	2.6	3.4	0.78	10
1957	70	16	11	15	5.5	2.2	4.0	1.3	0.21	0.85
1958	110	11	8.4	30	18	6.0	1.2	1.2	1.7	1.8
1959	300	25	170	19	11	16	8.7	9.5	0.16	24
1960	550	7.7	480	6.9	5.1	6.7	9.3	11	1.3	18
1961	520	3.5	480	5.2	0.35	0.98	4.6	17	0.87	7.3

TABLE IV

UNCLASSIFIED
ORNL-LR-DWG. 77404RCALCULATED MEAN ANNUAL CONCENTRATION OF RADIONUCLIDES
AT TENNESSEE RIVER MI. 465.5^a(UNITS OF 10^{-9} $\mu\text{c}/\text{ml}$ or $\mu\mu\text{c}/\text{liter}$)

Year	Gross Beta	Cs ¹³⁷	Ru ¹⁰⁶	Sr ⁹⁰	Y ⁹¹	Ce ¹⁴⁴	Zr ⁹⁵	Nb ⁹⁵	I ¹³¹	Co ⁶⁰
1949	18	2.0	2.7	3.7	0	0.46	4.5	0.57	2.0	
1950	4.9	0.49	0.60	0.98	0		0.38	0.11	0.49	
1951	3.1	0.61	0.54	0.87	0		0.14	0.068	0.54	
1952	8.1	0.37	0.56	2.7	0	0.86	0.72	0.68	0.74	
1953	12	0.26	1.1	5.4	0	0.27	0.30	0.14	0.083	
1954	17	0.94	0.48	5.8	1.2	1.0	0.59	0.40	0.15	
1955	16	2.3	1.1	3.4	2.0	3.1	0.19	0.21	0.26	0.24
1956	21	6.2	1.1	3.8	1.2	2.1	0.42	0.55	0.13	1.7
1957	9.8	2.2	1.5	2.1	0.78	0.31	0.56	0.18	0.030	0.12
1958	18	1.8	1.4	4.8	3.0	0.97	0.20	0.20	0.32	0.28
1959	36	2.9	20	2.3	1.3	1.9	1.1	1.1	0.02	2.8
1960	79	1.1	67	1.0	0.73	0.96	1.3	1.6	0.19	2.6
1961	67	0.47	61	0.67	0.05	0.13	0.59	2.1	0.11	0.93

concentration of radionuclides released, the maximum permissible concentration in water (MPC_w) for these radionuclides, the potentially significant avenues of exposures, and the type of individual under consideration. Based upon these considerations, the organs selected for analyses in this report include bone, gastrointestinal tract, thyroid, gonads, and total body. The bone and total body are reasonable selections when Sr^{90} and Cs^{137} are considered and when dose by immersion in contaminated fluids is possible. The increased quantity of Ru^{106} , entering the surface water in 1960 and 1961, suggested the immersion dose and the GI tract. The genetic dose is of particular concern for exposure of a population and, therefore, is included, although it can be estimated only approximately as equal to the total body dose; that is, equal to the average dose in other soft tissues. Finally, the release of I^{131} implicates the thyroid, especially when the child is considered.

The fraction of MPC_w attained for the case of internal dose was calculated according to the recommendation of the ICRP.⁷ For a mixture of invariant composition and based on a particular organ, x , the fraction of MPC_w that is attained is given by:

$$\sum_i \frac{P_{wi}}{(MPC)_w^x} \quad (1)$$

where

P_{wi} = the concentration of the particular radionuclide, i , in water, and

$(MPC)_w^x$ = the maximum permissible concentration of the particular radionuclide in water for the organ and individual of interest and for continuous exposure to radionuclide, i .

When the value of expression 1 is less than or equal to 1, the exposure is not in excess of permissible limits. This formulation neglects the dose due to external sources, but the external dose will be estimated separately in this report.

The values of P_{wi} are to be average values, the period of averaging being 1 year according to the recommendations of ICRP, NCRP, and FRC. All MPC_w values used for data relating to the Clinch River are taken as one-tenth the occupational MPC_w values for continuous exposure. To obtain MPC_w values relating to the Tennessee River, the MPC_w for continuous occupational exposure has been reduced by a factor of 1/100 for whole body as critical organ and by 1/30 with thyroid, bone, and GI tract as the critical organs. These values are suggested by ICRP for application to exposure of people living in the neighborhood of the plant and for the average exposure of the population-at-large, respectively. However, the values are intended only as interim guides for nonoccupational exposure, since they neglect several important considerations, particularly differences between children and adults.

If the fraction of MPC_w calculated from equation 1 is multiplied by the appropriate annual dose rate permitted in the particular organ of interest, an annual dose rate is obtained. A careful interpretation of such values is necessary, since the calculated dose only applies to a long-term and stable situation. The occupational MPC_w values are set by the requirement that the dose rate (rems/week) after 50 years of exposure shall not exceed a recommended limit. During a 50-year exposure period, equilibrium is reached by most of the radionuclides, because their effective half life is short compared to 50 years. However, in the case of Sr^{90} , the allowable annual dose is reached only after 50 years of continuous exposure to the MPC. For this reason, calculation of the actual dose received by ingestion of known concentrations of radionuclides is desirable.

As a second interpretation, the same numerical value calculated as a dose rate above may be considered also as a "dose commitment," meaning the dose in rem that will be received during the next 50 years due to an exposure of 1 week with P_{wi} determined for that period. Actually, the dose delivered after various times following the intake period depends upon the effective half life of the isotope involved.

Because the MPC's which enter into the calculations have been estimated on the basis of so-called "standard man," the dose really represents only that which would be received by a person of physical characteristics and habits resembling standard man. Thus, the doses estimated should be considered as average values for typical adult individuals. Very little is known at the present time concerning differences in metabolic rates or processes of children and adults as they relate to important radionuclides. Dose correction factors that take into account differences due to intake and organ size can be estimated, 8, 9, 10, 11 but in this preliminary report it is not possible to make any adjustment on this basis.

In the case of the gastrointestinal tract, the calculation of the MPC is based on the assumption that the wall of the tract will receive 50% of the beta-gamma dose and 0.5% of the alpha particle dose delivered to the contents of the tract. To a very large extent this dose will be proportional to the concentration of the radionuclide in the contents of the tract, but it will not vary greatly with the mass of the contents or with the diameter of the tract. Thus, no very significant correction is necessary so far as the masses of the organ or contents are concerned. Assuming the tract is always full and that the residence time is short compared to the half life of the radionuclides of interest, the dose received will not be changed significantly as residence time varies. This leaves the concentration of the radionuclide in the contents of the tract and, hence, the dietary composition as the only variable of significance.

The ratio,

$$\left(\frac{\text{Intake of Water}}{\text{Weight of Contents of GI Tract}} \right)_{\text{age}} \bigg/ \left(\frac{\text{Intake of Water}}{\text{Weight of Contents of GI Tract}} \right)_{\text{standard man}}$$

would seem to be the appropriate correction factor to apply here. No data have been found on the variation of the weight of the contents of the GI tract with age. It is hoped that these data will be found before completion of the report.

As mentioned above, one can interpret the annual dose rates obtained (Table V) as a dose commitment for the future; that is, dose that will be delivered sometime during the next 50 years if life extends that long. (The unit is, then, rem to be delivered during the next 50 years per year of occupancy, not rem/year.) Since the MPC values are based on data for a "standard man," either interpretation should be considered to represent the exposure of an "average" or typical adult. Both the FRC and the ICRP allow a factor of 3 as a practical factor to use to provide for the variation of dose in a homogeneous population group. That is to say, even among adults, or children of like age, exposed to the same environmental situation, there will be a considerable spread of doses actually received; and it is assumed that only a small fraction of the group will receive more than three times the average value.

ESTIMATED RADIATION DOSAGES

DRINKING WATER

Estimates of the fraction of maximum permissible dosages received from drinking Clinch River and Tennessee River water are based on concentrations of radionuclides in the raw water. This approach is conservative, because it assumes that there will be no reduction of radionuclides in the water by water treatment before drinking, and it makes no allowance for the small portions of the radionuclides released that are in the bottom sediments and would not be expected to enter raw-water intakes. Future calculations may consider radionuclide removal by water plants and bottom sediments, but the data now available do not warrant this refinement.

The fraction of MPC_w of the river water can be calculated by using the average concentration of the various radionuclides for each year where such data are available. As explained previously, all MPC_w values used for data relating to the Clinch River are taken as one-tenth of the occupational MPC_w values for exposure during the entire week (168 hours). To obtain MPC_w values relating to the Tennessee River, the MPC_w for continuous occupational exposure (168 hours/week) has been reduced by a factor of 1/100 for whole body as critical organ and by 1/30 for thyroid, bone, and GI tract as the critical organs. If the fraction of MPC_w calculated by equation 1 is multiplied by the appropriate maximum permissible dose rate for the particular organ, an annual dose contribution to an individual is obtained. However, it must be kept in mind that in the case of the radionuclides of long effective half life, this annual dose contribution will be attained only if occupancy continues for many years.

To represent estimated dosages for the Clinch and for the Tennessee rivers, two reference stations were selected; namely, CRM 14.5 and TRM 465.5. The fraction of MPC_w that would be attained by drinking Clinch River water and Tennessee River water from these two locations is given in Table 5.

TABLE V

FRACTION OF MPC IN WATER FROM CLINCH AND TENNESSEE RIVERS

Year	Clinch River Mi 14.5				Tennessee River Mi 465.5			
	Bone	G.I. Tract	Total Body	Thyroid	Bone	G.I. Tract	Total Body	Thyroid
1949	0.30	0.0043	0.076	0.021	0.11	0.0016	0.094	0.0077
1950	0.065	0.0022	0.016	0.0043	0.029	0.0010	0.025	0.0021
1951	0.052	0.0017	0.013	0.0038	0.026	0.00087	0.022	0.0019
1952	0.18	0.0015	0.044	0.0098	0.081	0.00069	0.068	0.0045
1953	0.35	0.0018	0.087	0.015	0.16	0.00053	0.13	0.0053
1954	0.51	0.0032	0.13	0.022	0.17	0.0011	0.15	0.0074
1955	0.22	0.0037	0.055	0.0099	0.10	0.0019	0.086	0.0047
1956	0.23	0.0042	0.060	0.010	0.11	0.0020	0.097	0.0051
1957	0.15	0.0024	0.037	0.0063	0.062	0.00099	0.052	0.0027
1958	0.30	0.0031	0.075	0.013	0.14	0.0015	0.12	0.0077
1959	0.20	0.021	0.050	0.0084	0.070	0.0075	0.060	0.0030
1960	0.070	0.050	0.018	0.0037	0.030	0.021	0.026	0.0016
1961	0.053	0.048	0.014	0.0027	0.020	0.019	0.017	0.0010

Values at CRM 14.5 represent the fraction of the continuous nonoccupational MPC_w for persons living in the plant vicinity that would be attained, and values at TRM 465.5 represent the attained fraction of MPC_w for the average population-at-large. Inherent in the calculation of these fractional values is the assumption that exposure is continuous for a period of 50 years to the mixture of radionuclides that is present during the particular year. For these particular mixtures of radionuclides in the raw water, estimated dose to the bone constitutes a greater fraction of the maximum permissible limit than does the calculated dose to the other body organs. This is attributable to the Sr^{90} that is released. The largest fraction of bone dose attained was 0.51 (51%) for the 1954 concentrations, assuming that the same concentrations continued for 50 years. Applying the most restrictive FRC limit of thyroid dose (for the average child of the population-at-large which is one six-hundredth of the continuous occupational exposure), the fraction of MPC_w that would be attained at CRM 14.5 during 1961 is less than 0.03 (3%). The increase in internal dose to the GI tract for 1960 and 1961 is due to the increased release of Ru^{106} . A mathematical model is being developed that will allow calculation of dose received by ingestion of known concentrations of radionuclides.

IMMERSION IN CONTAMINATED WATER

Due to the presence of radionuclides, the river will act as a source of radiation to persons engaged in swimming, boating, fishing, and water skiing. Since direct measurements of immersion dose rate are unavailable, it is necessary to calculate the dose rate by considering the radionuclide composition of the water.

The immersion dose calculation assumes the body is in the center of a sphere and receives equal quantities of radiation from all directions. The external exposure from beta radiation may be calculated by means of an equation recommended by Morgan.¹²

With certain reasonable assumptions this expression in units of rad per day is simplified to:

$$\text{Beta Dose Rate} = 51.2 \text{ QE}_i \quad (2)$$

where

$Q = \mu \text{ c/g of water and}$

$E_i = \text{effective absorbed energy per disintegration.}$

An empirical formula was used to estimate the average effective absorbed energy of a beta disintegration.⁷

$$E_i = 0.35 E_m f \left(1 - \frac{\sqrt{Z}}{50} \right) \left(1 + \frac{\sqrt{E_m}}{4} \right) \quad (3)$$

where

E_m = maximum energy of type considered,

f = fraction of disintegrations at a particular energy, and

z = atomic number.

The penetration distance in water of the most energetic beta particles from the radionuclides involved is about one centimeter. Therefore, the beta radiation at the surface of a body immersed in the contaminated water is effectively one-half of that calculated by equation 2. The external exposure from gamma radiation may be calculated by means of a similar formula,¹² for which the simplified expression in units of rad per day is:

$$\text{Gamma Dose Rate} = 51.2 Q E_m f \quad (4)$$

in which the terms Q , E_m , and f are the same as in equations 2 and 3. In each instance where some latitude is allowed in the assumptions, a conservative approach is taken. Therefore, the computed dose rates would be expected to be an overestimate.

Where the water contains a mixture of radionuclides, it is necessary to calculate the dose rate associated with each radionuclide. The total dose rate is simply the sum of the individual dose rates. Decay schemes presented by Blomeke and Todd are used in the calculations.¹³ The dose-rate values for beta and gamma radiation, respectively, are one-half the beta dose rate (equation 2) and the total gamma dose rate (equation 4).

The immersion dose rates due to beta and gamma radiation at the two reference stations are listed in Table VI. A maximum dose rate of 0.027 mrad per day of exposure at CRM 14.5 (1960) is calculated. The dose rate is a function of nuclide type and concentration. Until 1958, the largest fraction of beta dose was associated with Sr^{90} and the largest gamma dose was generally due to Cs^{137} . Since then, Ru^{106} has accounted for about 75% of the total immersion dose.

CONTAMINATED BOTTOM SEDIMENTS

Radionuclides associated with solids that have settled to the bottom of the river can be expected to contribute to the total radiation dose received by man. Although earlier calculations assumed complete dilution of fission products in the river, annual surveys made by the Oak Ridge National Laboratory, Applied Health Physics Section, have shown that some of the radionuclides are retained by the bottom sediments.¹⁴ Average concentrations of specific radionuclides in bottom sediments were calculated by averaging all values for the entire study reach of the Clinch River and of the Tennessee River. Cesium-137, cerium-144, cobalt-60, and, more recently, ruthenium-106 were found to be the principal radio-

TABLE VI

UNCLASSIFIED
ORNL DWG. 63-180RIMMERSION DOSE RATES IN CLINCH AND TENNESSEE RIVERS
(UNITS OF 10^{-4} mrad/24-hr EXPOSURE)

YEAR	CLINCH RIVER Mi 14.5			TENNESSEE RIVER Mi 465.5		
	BETA	GAMMA	TOTAL	BETA	GAMMA	TOTAL
1949	19	16	35	2.4	2.0	4.4
1950	3.4	5.2	8.6	0.5	0.79	1.3
1951	2.7	2.1	4.8	0.46	0.35	0.81
1952	8	5.0	13	1.3	0.77	2.1
1953	13	2.7	16	2.0	0.41	2.4
1954	20	7.2	27	2.3	0.82	3.1
1955	18	9.9	28	2.8	1.6	4.4
1956	16	18	34	2.6	2.9	5.5
1957	10	9.9	20	1.4	1.4	2.8
1958	16	8.4	24	3.1	1.6	4.7
1959	71	67	140	8.5	8.0	17
1960	170	95	270	25	14	39
1961	160	79	240	21	10	31

nuclides associated with these sediments. Reasons for such selectivity are enumerated elsewhere.¹⁵ Major changes in the radioactivity of the bottom deposits were found, but the details of sediment transport must be delineated before conclusions can be drawn about observed changes in radionuclide concentrations.

The "Flounder" is used principally to furnish qualitative information on the build-up of gamma-emitting radionuclides in sediments. Construction of the device makes it insensitive to beta radiation. Although the "Flounder" is calibrated routinely with a sealed radium source (as a stability or sensitivity check), the complex spectrum of gamma rays from both the contaminated sediments and the radium source prevents a direct determination of exposure dose by use of this instrument. Estimates of exposure dose can be made, but the limitations of such data must be recognized (Tables VII and VIII).

In a general way, the measurements in the Clinch River reflect the quantity of Cs^{137} and Co^{60} released each year. Maximum readings in the Clinch River (generally at CRM 8.3) were larger than the average readings by a factor of 1.9 ± 0.09 ; similarly, the ratio in the Tennessee River was 1.8 ± 0.2 . For the purpose of estimating the radiation dose to man, calculations of dose were made by using the average radionuclide composition of the sediments. It was assumed that this average composition was distributed uniformly in an infinite source. To assume an infinite source containing the maximum concentration of radionuclides observed seems overly conservative. Further, it was assumed that the individual would be exposed to one-half the submersion dose of beta particles and to one-half the submersion dose of gamma emission (that is, from one-half a sphere). Such an assumption is reasonable, since the individual receiving the dose is likely to be standing on or floating above the contaminated sediments. Normally, only the feet would be subjected to the total beta and gamma dose rate.

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TABLE VII

ESTIMATED RADIATION DOSE RATES FROM CONTAMINATED SEDIMENTS
IN TENNESSEE RIVER

Measured ^a (10 ⁻² mr/24-hr)			Calculated (10 ⁻² mrad/24-hr exposure)			
Year	Average	Maximum	Beta	1/2 Gamma ^c	Total	Attenuated ^b 1/2 Gamma ^c
1951	13					
1952	22					
1953	23					
1954	19	30	22	50	72	3.0
1955	26	43	60	68	128	4.2
1956	36	69	65	110	175	6.1
1957	33	58	37	80	117	4.2
1958	35	63	55	62	117	3.5
1959	30	63	48	56	104	3.1
1960	33	49	75	61	136	3.3
1961	26	48	95	54	149	2.8

^aIn units of 10⁻² mr/24-hr exposure as measured by the "Flounder."

^bAttenuation through 3 ft of water.

^cOne-half of total gamma dose from infinite source.

TABLE VIII

UNCLASSIFIED
ORNL-LR-DWG. 77409RESTIMATED RADIATION DOSE RATES FROM CONTAMINATED SEDIMENTS
IN CLINCH RIVER

Measured ^a (10 ⁻² mr/24-hr)			Calculated (10 ⁻² mrad/24-hr exposure)			
Year	Average	Maximum	Beta	1/2 Gamma ^c	Total	Attenuated ^b 1/2 Gamma ^c
1951	39			90 ^d		
1952	88			320 ^d		
1953	53			160 ^d		
1954	57	110	60	160	220	9.5
1955	60	110	130	180	310	11
1956	130	260	300	630	930	35
1957	96	180	180	460	640	24
1958	100	200	210	360	570	19
1959	160	280	450	710	1160	39
1960	150	280	510	460	970	25
1961	95	170	530	290	820	15

^aIn units of 10⁻² mr/24-hr exposure as measured by the "Flounder."^bAttenuation through 3 ft of water.^cOne-half of total gamma dose from infinite source.^dEstimated from correlation relationship.

Figure 3 Attenuation of Bottom Sediment Dose in Clinch River and Tennessee River.

Calculated dose rates from bottom sediments in the Clinch River and Tennessee River are listed in Tables VII and VIII. The beta dose rate was taken as one-half the value determined by use of equation 2 and the gamma dose rate by use of one-half the calculated value of equation 4. Since the source is not infinite in extent, the calculated values give a larger estimated dose rate than that actually available. Accordingly, the largest bottom sediment dose rate of 12 mrad per day of exposure would have occurred in 1959, and was divided as 40% beta and 60% gamma radiation. The percentage contribution of specific radionuclides to the beta and gamma dose rates shows that the total rare earths, Cs^{137} , and, more recently, Ru^{106} are the principal contributors to beta dose rates, and Co^{60} and Cs^{137} account for the largest fraction of gamma dose rates.

Since bottom sediments are generally covered by water, the gamma dose rate to the gonads of an individual standing on the river bottom would be reduced by attenuation. An average attenuation coefficient for water was calculated by weighing both the fraction of time a photon of a given energy was emitted by a particular radionuclide and the fraction each radionuclide contributed to the total loading of the bottom sediments. The fraction of dose remaining is graphed as a function of the depth of water shielding (Figure 3). The estimated gamma-dose rates, after attenuation through 3 feet of water, are listed in Tables VII and VIII.

The other avenues of potential exposure previously mentioned are being investigated. Although irrigation is not practiced along the Clinch River and is of almost negligible extent along the Tennessee River, calculations have been made of radionuclide transfer from soil to crop to man for hypothetical situations. Laboratory and field experiments are currently in the planning stage to elucidate this potential vector of contamination. No apparent difference exists in external radiation in water treatment plants using Clinch River water from points above and below the confluence of White Oak Creek. A large fish sampling program continues and, accompanied by results of a survey of fish consumption, will allow estimates to be made of dose due to contaminated fish. Whenever opportunity permits, direct measurements of dose received or dose rate will be made.

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DISCUSSION

PARKER: The values quoted by Mr. Cowser are the actual discharges divided by the flow, taking into account the local inflows as one goes down stream. The measured concentrations are considerably less than these. I have the most recent report of the Water Sampling Analysis Group of the Clinch River Study, of which Dr. Friend is a member, and looking at these we find that the highest value measured in the Clinch are at the most 4×10^{-8} $\mu\text{c/ml}$ for Sr^{90} . The usual values are lower than this. These values are per ml on a weekly sampling basis over a period of one year.

COWSER: Let me make one further point. I have been talking about an analysis of past history and thus far I have applied the recommended values that ICRP and NCRP have published. We shall shortly become involved in considering any differences in FRC values. The USPHS publication, the Recommended Guide, is fairly new. The 0.5 MPC_w that I mentioned occurred in 1954.

BELTER: Would you compare this with Handbook 52 values which were used at that time.

COWSER: I did not compare it with Handbook 52 but have made my comparison with the values in NCRP Handbook 69 and ICRP recommendations. Perhaps, this was an arbitrary decision on my part but you can then take the values that I present to you and apply any other recommended guides to them.

FREDERICK: Could I have your definition of MPC?

COWSER: I think you must recognize that MPC is really a secondary standard when we talk about MPC's published by ICRP and NCRP. The primary standard is dose rate or dose received. MPC is a facility to allow you to make calculations more rapidly. MPC is that concentration of a nuclide in water consumed by a standard man that would give a dose to a particular organ not exceeding a specified limit.

PARKER: One other point, the ICRP apparently at its last meeting recommends that they do drop the MPC_w values and use the body burden as the ultimate standard as it is now.

KAUFMAN: The ICRP merely calculated the MPC values for water as examples of how one might obtain the radioactivity if the only source was water. Because of the misinterpretation of the MPC as a permissible receiving water concentration.

COWSER: This has been based on a total body burden of so many microcuries in the organ of interest. It is two microcuries of strontium-90 distributed uniformly in the bone.

Dr. Friend, do you wish to give a definition of MPC?

FRIEND: I don't have any. You are the expert on this. This is not my field at all. One thing does concern me however. Why are you using MPC values rather than FRC values?

COWSER: There is nothing underhanded about this. I chose to use those in the beginning because I had MPC values for all the critical organs for the types of population I was interested in. We do not have this information yet from FRC for different types of populations. However, we will make calculations based on FRC dose rate guides. I suspect that any differences will be small.

BERNARD: Was your figure showing the number of millirads per day in the Clinch River based on the assumption of a person swimming in the river for 24 hours?

COWSER: This is only an assumption; that is, a person is in contact with the bottom sediments for a 24-hour period. When one gets more than a centimeter away from the bottom sediments the betas will be eliminated by various adsorption processes. We are concerned, then, with the gamma dose rate. I have made some calculations on gamma ray attenuation coefficients which would allow one to predict the dose at so many feet above contaminated bottom sediments when they are covered by water. For example, three feet of water will cut the bottom sediment dose rate to 0.39 mrad/day in the Clinch River and in the Tennessee River will cut the dose rate from 1.8 to .06 mrad/day of exposure.

BERNARD: A person swimming for a half-hour would then receive between 1/20 or 1/50 of the dose rates?

COWSER: Well, recall now when you are swimming you would be subjected to the immersion dose rate and a bottom sediment dose rate, too. Of course, you would postulate that the individual is some distance from the bottom sediment. These doses are additive in the final analyses. Some day we must add all of these up. It would be nice to know how long swimmers would be submerged three feet below the surface of the water. I know we are not going to be able to come up with that number based on a study and we will necessarily make some estimate.

BERNARD: Did the assumption that you make for grain foods include the processing of the grains?

COWSER: No, it did not include processing. I take no credit for anything being removed from the grain as a result of its being boiled in water or anything else. I say this was on the grain and any benefits derived from further processing have not been considered.

BERNARD: Where does the problem with strontium come in?

COWSER: It appears that the largest problem may be associated with the transfer of strontium from contaminated soil to the crop. Here there is a time function one must consider. If we are thinking about putting water on at a rate of 2 feet a year the nuclides will decay and it will be necessary to consider the decay term in calculation. For cesium, there seems to be no problem. At least, based on the calculations I have made, I cannot demonstrate that it will be a problem.

KAUFMAN: I would like to suggest that you try calculating the dose from an infinite plane resulting from irrigation with water containing cesium where there is a considerable concentration on the surface of the soil making reasonable assumptions. I think the greatest hazard from irrigation waters, where you are using a mixture of cesium and strontium, will be the build up of cesium concentration on the soils and the subsequent dose to a person walking or working in the field. I think if you calculate this you will find that the dose may be 100 times the dose to the person drinking the water. This would be based on a 40 hour per week occupancy.

COWSER: How thick is the infinite plane source?

KAUFMAN: Consider the plane of infinite thickness and infinite areal extent. Land irrigated for a few years should be contaminated to a depth of perhaps one foot and in this case such a depth is nearly equivalent to an infinite depth. There is no question but that it will be ten times of the dose from drinking water.

COWSER: I think it is a function of how fast we are going to apply water to the soil. I have assumed two feet of water per year, you might assume four. In the case of the soils that we have, and at the rate of application of two feet a year it looks like it would take fifty thousand years to reach equilibrium with the soil due to a sodium-potassium-cesium complex in our water. So, for that time period, the equilibrium value obtained without considering decay and by considering decay is considerably different. It is a fraction of about .03 per cent when you throw in the decay factor. However, Dr. Kaufman, this is a very interesting point and one frankly that I have not thought much about.

KAUFMAN: The air attenuation is relative small. I can give you an example. We were getting 30 mr/hr around a pump that was leaking on the ground

in which the water being pumped by the pump was $1.5 \times 10^{-4} \mu\text{c/ml}$.

PARKER: How long a time was this pump in operation?

KAUFMAN: For about six weeks.

BELTER: Was ruthenium a problem in all of this work?

COWSER: I do not know how to calculate ruthenium soil loadings. We just don't know enough about the chemistry of ruthenium in various waste solutions to make reasonable calculations. Experimentally, one would determine the effect of contacting the soil with a typical solution.

THE AQUEOUS BEHAVIOR OF STRONTIUM - 85, CESIUM - 137, ZINC - 65, AND COBALT - 60 AS DETERMINED BY LABORATORY TYPE STUDIES

by ALBERT G. FRIEND

The Cooperative Studies Unit, Radiological Health Research Activities, at the Robert A. Taft Sanitary Engineering Center is studying the "fate" of radionuclides discharged to natural fresh-water systems. In the course of this study, about 17 field trips have been made to streams located below atomic energy installations that discharge liquid radioactive wastes. In samples of aquatic media, the man-made radionuclides ruthenium-106, cesium-134, -137, zirconium-95, niobium-95, zinc-65, cobalt-60, cerium-141, -144, strontium-89, -90, as well as daughter products of uranium-238 and thorium-232, have been identified.

Data resulting from these field trips show the distribution of these radionuclides in the various segments of the aquatic environment. It will be necessary, however, to carry out complementary studies in the future in environments less complex and more closely controlled than the natural one to determine mechanisms, reaction rates, and other physical, biological, and chemical factors which control this distribution.

The Cooperative Studies Unit has initiated two such studies, one in a completely artificial aquarial environment, and the other in a semi-natural pond situation.

The aquarial studies were factorially designed investigation to determine the effects of the major cations in natural water systems, i. e., calcium, magnesium, sodium, and potassium, on the uptake of strontium, cesium, zinc, and cobalt by small bluegill fish. The two studies completed to data show that over the range 0.2 to 20 mg/liter, only calcium has any effect upon strontium uptake, and that this uptake is uniformly depressed as the aqueous concentration of calcium is increased.

The semi-natural study was carried out during the summer and fall of 1962 by injecting multiple radionuclides into a shallow fresh-water pond. The distribution of these radionuclides was followed in the various segments of this environment by analyzing samples collected at periodic intervals.

The pond measured about 50 x 70 feet in surface area, with a depth ranging from 0 to about 3 1/2 feet. Because the pond was underlain by deep sand and gravel beds, it was lined with a one-piece sheet of 8 mil black polyethylene to retain the added activity and to make possible a radionuclide mass balance at the termination of the experiment.

After the liner was in place, approximately 30,000 gallons of spring water was added. The chemical composition of the pond water during the course of the experiment is shown in Table I. The water level in the pond was checked after 2 to 3 days to see that the liner had remained intact. About 14 tons of sand was then added to the pond; washed sand was specified, but subsequent analyses indicated that the "washed sand" consisted of about 90% sand ($> 50 \mu$), 8% silt (2μ - 50μ), and 2% clay ($> 2 \mu$). Unfortunately, the percentages of clay and silt were higher than desired for this particular experiment, inasmuch as these soil components have a high affinity for some of the added radionuclides.

Immediately prior to adding radionuclides, samples of montmorillonite and kaolinite clays and an illitic clay shale were placed in specially constructed plastic containers in both the shallow and the deep end of the pond. The cation exchange capacity of some of the montmorillonite and kaolinite samples were saturated with calcium or sodium in order to check on the effect of clay form on the uptake of these minerals.

Biota used included two species of fish and clams and single species of snails and tadpoles. These were added to the pond as secured. Except for the clams and one species of fish (bluegill), all the biotic media were taken from ponds fed by the same spring water so that they were in reasonable equilibrium with the test pond environment.

About one month after the sand and clays and the biota had been added to the pond, 4 millicuries each of cobalt-60, cesium-137, and zinc-65 and 3 millicuries of strontium-85 were mixed with about 50 gallons of water and were distributed over the surface of the pond by means of a pump and hose to minimize "hot spots." Thermal stratification in the pond during the test was prevented by a pumping system which removed water by means of an intake manifold extending completely across the bottom of the deep end of the pond and reintroduced it by a similar manifold at the surface of the shallow end.

Samples of the physical media were collected in duplicate and biotic media were collected in triplicate 2, 4, 8, 12, 16, 24, 38, 52, 66, and 80 days after addition of the radionuclides. A few extra water samples were collected in the early stages of the experiment and analyzed in order to follow more precisely the initial distribution of the radionuclides in this medium. Radionuclides were added on July 30 and the final samples were collected on October 18, 1962. At each sampling date, water samples were collected for water chemistry, as noted above, and for plankton count.

On December 8, fifty days after termination of the experiment, and prior

TABLE I

Results of Chemical Analyses of Water Samples--Pond Experiment No. 1

Date, 1962	Time from dosing days	Concentrations in milligram/liter								Alkalinity, as CaCO_3
		Calcium, as Ca^{+2}	Magnesium, as Mg^{+2}	Iron, as Fe	Manganese, as Mn^{+2}	Ammonia, as NH_3/N	Sulfate, as SO_4^{-2}	Chloride, as Cl^-	Total phosphate, as PO_4^{-3}	
June 20	--	33	23	0.20	0.00	--	42	11	0.36	114
Jul. 5	--	19	20	0.08	0.0	0.00	40	23	0.03	90
Jul. 29	--	18	19	0.36	0.04	0.11	33	13	0.07	82
Aug. 1	2	22	18	0.45	0.2	0.35	35	12	0.0	106
Aug. 3	4	23	16	0.56	0.20	0.55	35	12	0.1	112
Aug. 7	8	22	18	0.44	0.20	0.47	34	12	0.1	112
Aug. 11	12	29	18	0.36	0.11	0.15	34	13	0.25	130
Aug. 15	16	25	19	0.32	0.09	0.15	32	13	0.05	130
Aug. 23	24	25	20	0.32	0.1	0.10	37	14	0.11	126
Sep. 6	38	22	19	0.34	0.1	0.2	32	14	0.7	120
Sep. 20	52	21	19	0.28	0.11	0.20	32	14	0.13	118
Oct. 4	66	27	18	0.68	0.08	0.12	32	10	0.19	122
Oct. 18	80	24	22	0.12	0.06	0.15	31	8	0.13	116

to release to the environment, the pond water was decontaminated by a mobile water treatment unit developed by the U. S. Army Corps of Engineers. This equipment consists of a mixing chamber and upflow clarifier in a single unit (ERDLATOR) followed by a diatomaceous earth filter, a weak acid cation exchange unit (H^+), and a strong base anion exchange unit (OH^-) in that order. Sodium hypochlorite as a disinfectant, ferric chloride as a coagulant, and powdered limestone are added at the mixing chamber, and diatomaceous earth is added between the clarifier and the filter. The pond water was processed at a rate of 25 gpm, the rated capacity of the water treatment unit.

RESULTS AND DISCUSSION

WATER

The suspended solids were separated from each water sample collected during the pond experiment and treated as separate samples. The separation of these suspended solids was accomplished with a Type HA Millipore filter (0.45 μ pore size). The material retained on the filter defines the term suspended solids as used in this discussion.

Data on the specific activities associated with the water samples are shown in Table II. These data are plotted in Figures 1 and 2.

Figure 1 shows that the cesium, cobalt, and zinc disappear from the liquid phase rapidly. Calculated concentrations of these radionuclides per liter of water, based on 4 mc diluted to 30,000 gallons, give values of about 35,000 pc/liter. If averages for the 1.04, 1.18, and 1.23 days are taken as the mean concentrations of the cobalt-60, zinc-65, and cesium-137 in solution one day after dosing, then about 73%, 60%, and 47% of these radionuclides, respectively, were lost by the water to other pond media. By the end of the fourth day these losses increased to about 92%, 96%, and 95%, respectively.

In contrast to the behavior of the cobalt, zinc, and cesium, strontium-85 showed little loss from solution, its measured concentration being above the calculated value of about 26,000 pc/liter for the first 14 days. Such behavior of the strontium might have been predicted in view of the high calcium concentrations in this system and the chemical similarity of these elements.

Figure 2 shows the plotted radionuclide data on the suspended solids from the water samples. The suspended solids take up the cobalt and zinc very rapidly, the activities in this fraction of the water sample being two to three times that in the dissolved state one day after dosing. Less cesium than cobalt and zinc is taken up, but the maxima of all appear at about the same time. This rapid reaction between the suspended solids and the cobalt, zinc and cesium in great part accounts for the loss of activity from the water phase, since these suspended solids, in settling, carry these radionuclides to the bottom sediments.

Strontium again appears to behave quite differently from the cobalt, zinc,

TABLE II

Radionuclide Concentrations in Water Samples--Pond Experiment

July 30 to October 18, 1962

Time from dosing, days	Fraction ^a	Radionuclide concentration, pc/liter			
		Co ⁶⁰	Zn ⁶⁵	Sr ⁸⁵	Cs ¹³⁷
0	SS (1.75)	0	--- ^b	10	13
	DS	12	---	39	131
1.04	SS (25.3)	10,000	8,300	200	2,700
	DS	14,700	33,000	63,000	27,000
1.18	SS (21.0)	16,900	12,500	400	2,600
	DS	7,900	3,800	31,000	11,000
1.23	SS (18.5)	17,100	13,000	500	4,000
	DS	6,100	5,600	31,000	10,400
2.25	SS (18.4)	15,600	9,100	450	2,100
	DS	4,900	4,600	31,000	4,400
3.25	SS (8.8)	13,500	5,600	400	700
	DS	4,600	1,100	29,000	2,600
4.25	SS (14.4)	11,500	4,200	350	600
	DS	2,900	1,300	27,000	1,700
8.0	SS (28.1)	8,400	2,800	300	650
	DS	2,600	1,200	28,000	800
14.0	SS (9.4)	2,700	850	150	125
	DS	1,500	1,000	29,000	600
16.0	SS (3.4)	2,400	450	150	150
	DS	1,300	200	23,000	500
24.0	SS (17.3)	1,400	800	200	250
	DS	675	50	18,000	400
38.0	SS (28.0)	400	750	100	350
	DS	600	---	18,000	225
52.0	SS (23.1)	200	450	75	200
	DS	350	50	13,000	100
66.0	SS (20.1)	125	300	50	150
	DS	225	50	9,800	100
80.0	SS (17.6)	125	250	50	100
	DS	100	50	3,100	50

^a SS = suspended solids; DS = dissolved solids. Numbers in parentheses indicate weight of suspended solids in mg/liter.

^b Concentration below limits of detectability.

and cesium, only small percentages of this radionuclide becoming associated with the suspended solids.

The behavior patterns of the radionuclides in the pond are quite similar to those described in reports of field studies in which strontium-90 was found to be little affected by the stream environment and moved along with the water for long distances practically undiminished in concentration except by dilution; cobalt-60, zinc-65, and cesium-137 or 134 were rapidly lost to the surrounding environment after a relatively short distance of flow.

At the completion of the above uptake experiments, the pond water was decontaminated by use of the Army mobile treatment unit previously described. During the processing, hourly samples were taken of the raw water and of the water after each of the treatment processes, i. e., coagulation filtration, cation-exchange, and anion-exchange.

At the beginning of the decontamination run the water was withdrawn from the pond with as little disturbance as possible, and since it had been quiescent for about 50 days, it was fairly clear. The raw water entering the treatment unit had a turbidity of about 60 Jackson units.

After about half the pond water had been withdrawn, the recirculation pump was started and the plastic pipe to the return manifold was disconnected and used as a hose to agitate the bottom sediment. Many of the lighter sediments were resuspended and the water appeared quite turbid, showing turbidity readings of about 1000 Jackson units.

Table III gives the percentage removals of each radionuclide during the water treatment process. In water treatment, the main purpose of coagulation and filtration is to remove particulate matter of colloidal size and larger from the water.

The cation and anion exchangers form a demineralizing unit. The cation exchanger (H^+ form) removes the positively charged ions from the water and causes some molecules to dissociate, thus forming more cations, which are also removed. Hydrogen ions (H^+) replace the cations removed from the water. The anion exchanger removes the anions issuing out of the cation exchanger, and replaces them with hydroxyl ions (OH^-), which combine with the hydrogen ions to produce water molecules. In actual operation, either of these units will remove small charged colloids as well as ions.

With these precepts in mind, the coagulation and filtration data in Table III indicate that in the undisturbed pond water, about 40% of the cobalt, 82% of the zinc, and 47% of the cesium are in particulate form or are associated with particulate matter in the water, while only 4.2% of the strontium is so found.

The corresponding cation exchanger data indicate that about 95% of the strontium, 35% of the cobalt, 18% of the cesium, and none of the zinc exist as

TABLE III

Percentage Removal of each Radionuclide during Water Treatment Processes				
Condition A ^a	Removals, %			
	Cobalt-60	Zinc-65	Strontium-85	Cesium-137
Coagulation & filtration	39.9	81.6	4.2	46.6
Cation exchanger	35.0	0	95.1	18.3
Anion exchanger	12.1	12.5	0.2	17.8
To waste	13.1	5.8	0.4	17.3
Condition B ^b				
Coagulation & filtration	74.8	92.4	11.3	97.5
Cation exchanger	21.8	6.6	87.7	0.5
Anion exchanger	1.3	0	0	0.4
To waste	2.3	1.0	1.0	1.6

^a Condition A - After addition of radionuclides, water in pond kept in gentle motion (Ca 3.5 ft/hr) for 80 days and then allowed to be quiescent for 50 days. Turbidity about 60 on Jackson scale.

^b Condition B - Some bottom sediments resuspended by agitating pond water with recirculation pump and hose. Turbidity about 1000 on Jackson scale.

free cations or in molecules dissociated by the action of the hydrogen-form resin.

The anion exchanger data show that about 16% of the cobalt and zinc, 18% of the cesium, and only 0.2% of the strontium are removed by this treatment process. Illite, the predominant clay mineral in the pond, sorbs cesium irreversibly. As a colloid and at the high pH range existing in the anion exchanger, this silicate would carry a strong negative charge. It may then be assumed that this reduction in cesium concentration by the anion exchanger is due to the removal of the silicate colloids. The cobalt, zinc, and strontium reductions are incident to this colloid removal.

The radionuclides going to waste are presumed to be leakages through the filter and exchangers, and may in the case of cobalt, for example, represent a strongly complexed non-ionic form of material.

Where most of the activity of each radionuclide in the water is associated with particulate matter (Table III, Condition B), a greater percentage removal is effected by coagulation and filtration, as would be expected, and a lesser percentage is removed by the cation and anion exchanger.

CLAY MINERALS AND SEDIMENTS

Radionuclide analyses were made of each of the pure clay samples immersed in the pond, and of the sediment samples collected. The uptakes of cesium-137 by three of these clay samples are shown in Figure 3. Table IV summarizes some of the data relative to maximum uptakes determined from these samples.

The most surprising aspect of the data shown in Table IV is that of the four nuclides, strontium-85 reached the highest level of uptake. Generally, field data on river sediment have shown strontium to be in the smallest concentration of any of the identifiable man-made radionuclides. This apparent contradiction may be explained by the fact that in the field projects, montmorillonite was not present in the sediment whereas it was in the pond study and it was by this clay that the high strontium-85 uptake was achieved. This finding indicates a promising line of research.

Uptake of all radionuclides by samples placed in the shallow end of the pond was greater than the uptake by equivalent samples from the deep end. This, we believe, was due to the circulation pattern of the water in the pond. The samples from the deeper end of the pond were from a relatively stagnant area. This stagnant area gradually accumulated a sizeable deposit of clayey sediment, washed from other areas of the pond; as the clayey sediment settled, it removed a large quantity of activity from the water phase. A single water sample taken on the eighth day from the deeper part of the pond showed significantly lower levels of cobalt-60 and zinc-65 than a similar sample taken from the circulating pump output.

No differences were observed in the uptake by the calcium, sodium, or

TABLE IV

Kinetics of Uptake of Radionuclides by Silicate Minerals*--

Pond Study, July 30 to October 18, 1962

Radionuclide	Maximum Rate of Uptake	Maximum Uptake	Maximum Concentration at end of test
Cobalt-60	I	I (7,000)**	I
Zinc-65	K	K (3,000)	---***
Strontium-85	I	M (12,000)	M
Cesium-137	K	I (11,000)	I

* I = Illite
K = Kaolinite
M = Montmorillonite

** Figures in parentheses are pc/sample. All samples have equal surface area exposed and are 10 ± 0.5 gms in weight.

*** Data not available.

natural forms of kaolinite. The uptake by calcium-montmorillonite, however, was greater in most cases than the uptake by the natural (Na) form. This is believed due to the improvement in the physical state of the calcium-clay paste, which made it slightly more permeable to diffusing ions.

A number of the clay mineral samples, and clay samples from the pond sediments, were treated chemically to determine the "state" of the radionuclides that had been sorbed while in the pond. The treatments used were as follows: (1) leaching with ammonium chloride to remove those radionuclides held on the clay by adsorption and non-specific ion exchange; (2) leaching with ammonium chloride plus stable species of the radionuclides (which reduces resorption of the radioactive species) to remove ions held at specific sites; (3) leaching with ammonium acetate plus stable species buffered at pH 5 to remove radionuclides held by phosphates, carbonates, and other acid soluble materials; (4) digesting in hydrogen peroxide plus stable species to solubilize organic matter; and (5) digesting with dithionite plus stable species to solubilize such substances as iron oxide.

The illite samples tested to date indicate that the major portion of the cobalt-60 was in the acid soluble or the reductant soluble form; that up to 60% of the zinc-65 and 75 to 85% of the cesium were not removed by any of the treatments and that these radionuclides were presumed to be irreversibly "fixed" in the clay mineral; and that roughly 85% of the strontium-85 was in an easily exchangeable form.

On the basis of these fractionation studies, it is suspected that in the pond experiment the removals of the radionuclides from the water are attributable to specific components of the pond sediments; zinc-65 and cesium-137 by the illitic clays, strontium-85 by the carbonates and oxides, and cobalt-60 by both the acid soluble carbonates and oxides and by the reductant soluble iron oxides.

BIOTA

The two species of fish used in the investigation were carp and bluegill. The accumulation of radionuclides by the carp was consistently greater than by the bluegills, due undoubtedly to their feeding at different trophic levels.

Cobalt-60 and zinc-65 were taken up rapidly by both fish, reaching maximum uptake in about 2 days and maintaining these values to the end of the experiment, i. e., about 80 days. Cesium-137 was taken up much more slowly than the zinc and cobalt. After reaching a maximum value, the cesium concentration gradually decreased in the latter stages of the test. In contrast to zinc, cobalt, and cesium, strontium-85 concentrations continually increased in the flesh and bone fraction of both species of fish. This was probably due to the higher strontium-85 levels maintained in the water and to rapid growth of bone tissue by the fish during the warm season.

Concurrent with the pond investigation, fish of both species were

maintained in holding tanks adjacent to the pond, through which filtered pond water was continuously circulated. It was hoped by this arrangement to measure the uptake via the natural food chain, since the fish in the pond would maintain themselves on the flora and fauna of the pond and those in the tanks would be fed a commercial fish food, their only source of radionuclides being by cutaneous absorption from the water. Unfortunately, there were very unequal growth rates in the fish from the two environments, more or less negating comparison of these data, curiously enough, however, the tank fish, which demonstrated the smaller growth rate, concentrated cobalt-60 to a greater degree than did the pond fish.

The uptake of the four radionuclides by clams followed the exact pattern demonstrated by fish. As an added refinement, however, some of the clams that had been in the pond for the duration of the experiment were removed, placed in fresh pond water for 3 weeks, and then analyzed for radionuclide content. There was no significant loss of cobalt-60, zinc-65, or strontium-85 from the clams, and cesium appeared to follow the same gradual lessening of activity as in the fish.

In summary, it is felt that the field studies taken to date by the Public Health Service give a fairly good overall picture of the distribution of specific radionuclides in streams located below atomic energy installations; as an example, field data indicate that the strontium-90 discharged to flowing streams persists in the water for appreciable lengths of time. However, specificities must be defined by simpler and more refined techniques than can be exercised in the field. Laboratory studies in a completely artificial environment may define trends but the application of these trends to events which may occur in the field is difficult, and in some cases, completely impossible.

The pond investigations were planned to assist in the correlation of certain data resulting from the natural and artificial studies and the preliminary results of the first such study seem to indicate they will materially assist in this difficult task.

DISCUSSION

PATTERSON: How do you explain the disappearance and re-occurrence of zinc in the water on Figure 1?

FRIEND: We are down to about $50 \mu\text{c}/1$ and I think it can be explained by counting. The computer that we used does not give us plus or minus readings. It only gave us the mean value. There are, as you know, a considerable number of counting errors at low concentrations.

BERNARD: What was the temperature gradient?

FRIEND: I don't have that information with me. We did monitor the temperature continuously as well as continuous monitoring of light intensity. We tried to set this up as complete an experiment as we could.

PARKER: Was the pond exposed to the wind very much?

FRIEND: Yes, it was an open pond. We had to add about three inches of make-up water over the experiment. It became quite dry.

PARKER: Did you do any sampling before the Army worked on the samples?

FRIEND: Yes.

PARKER: Was there any pH change or did the pH stay the same?

FRIEND: Really, I do not remember what it was at the present time.

KAUFMAN: With reference to Figure 3 and the behavior of three types of clays; were the samples all of the same weight?

FRIEND: They were about 10 grams in weight plus or minus 0.5 grams. The surface area of all of them were the same. There was probably very little penetration into the crystal lattice structure.

KRUMHOLZ: You said there was more activity in the shallow end. What was the difference in depths between the shallow and the deep end?

FRIEND: About one foot in the shallow end and three and one-half in the deep end.

BERNARD: With the continuous circulation wouldn't you expect that some particles would settle in the shallow end?

FRIEND: These were prepared clay samples. There was no silt in the pond. This was all removed at first.

NELSON: Did your fish grow any?

FRIEND: Yes. We had some tanks by the pond in which we kept fish that we circulated filter pond water continuously through. We tried to get some measure of the uptake of the fish through their feeding habits other than those in the pond who were eating fauna and flora. The fish in the pond increased, I think, somewhat over 200%.

KRUMHOLZ: Was this in weight or in length?

FRIEND: In weight. The ones we held in the tank did not. They just about maintained their weight and length. They did not grow as well as the ones in the ponds. This also points up the fact that we have to do these experiments in the field and not only in the laboratory.

AUSTIN: Were the calcium and magnesium levels comparable in the aquarium studies? How high did your magnesium and calcium concentrations go?

FRIEND: During the course of the experiment the calcium varied from about 19 mg/l to about 33 mg/l; magnesium varied from about 16 to 23 mg/l.

AUSTIN: Were the fish in the aquarium taking up the radionuclides directly from the water?

FRIEND: These were completely unfed in the pond during the whole course of the experiment.

THOMAS: At the end of the experiment in which you put the fish in fresh spring water the water was pretty cold, wasn't it? What about the metabolic rate? It was pretty low, wasn't it? What conclusions do you draw from this? Would the same thing happen then that happened during the month of July?

FRIEND: I have not made any conclusions, yet. I have not had a chance to look at all the data yet.

NELSON: As far as the effect of strontium, anyway, in both the clams and the fish you would expect a great deal of the activity to be tied up in the shell of the clams and in the bone of the fish and there is a very slow metabolic

turnover in these tissues, in fact, you would expect none in the shell of the clam.

FRIEND: We separated the clams into the soft part and the shell when we counted them. There was much more uptake, I think on a per gram dry weight basis, in the fleshy part.

NELSON: You got a high concentration in the fleshy part?

FRIEND: Yes, a high concentration in the fleshy part or in the soft part of the clam than we did in the shell of strontium.

NELSON: Yes, this is obvious. The shell did not have enough time to deposit much tissue so you would not get as much in there and the tissue would turn it over more quickly and take it up.

FRIEND: We only carried this on for eighty days. It was more or less a pilot study.

NELSON: Did you actually test shell that was deposited during the eighty day experiment or test the whole clam shell?

FRIEND: Well, we had some algae growing on the clam shells and we do not know how much of this contributed. We tried to remove this but as you know you cannot remove all of it. What we tested was the shell of the clam. I am sure that some of the activity was on the outside of the shell.

HOOPER: Do you have any idea of the amount of stable material in these systems compared with the isotopes added? In other words what is the budgeted amount in the whole pool? Was the cobalt you added a measurable part of the total cobalt in the pool or pond? Have you made any calculations of this sort?

FRIEND: We did measure the concentration of the stable elements with the exception of cesium. I do not recall at this moment what they were; the data are available. I presume there must be very little of this available in the environment because it disappears so fast.

KAUFMAN: Did you add any carrier cesium?

FRIEND: No we did not want to upset the system by adding stable elements. We considered this.

KRUMHOLZ: On the fish, what kind of dissecting did you perform?

FRIEND: We separated the fish into three fractions: the flesh, the bone and the fissile and counted each one separately.

KRUMHOLZ: Where did you find most of the cobalt-60?

FRIEND: Initially, I recall it was mostly in the fissile section. We found appreciable amount in the bone but the maximum I believe was in the fissile.

FOSTER: Did you run a separate assay on zinc in the gills? The reason I mention this is because we have been doing some zinc-65 studies at Hanford and there is usually a high zinc concentration in the gills themselves.

FRIEND: We found the highest concentration of zinc in the Blue Gills in the gills.

HETEROGENEOUS EQUILIBRIA IN SEDIMENT UNDER TRANSPORT

by W. F. BRADLEY

You are soon going to join me in wondering why I have been asked to speak. It appears that I always fall into a deal where the audience spreads out in a spectrum from the people who know more about the subject than I do to those whose interests are diverse enough that they do not care to know any more than they do already. The diversity does have some nice points. People who have had quite different training can make substantial gains out of communicating with each other. On this campus there are those who have interests in ordinary water chemistry, some in hydraulics or hydrology and some in the kind of mineral grains that get buffeted about by the water agencies, and there are some who have interest in vegetables, and others in bugs and beasts that grow in the water. It is a kind of comfort to know that in spite of all these diversities the same kind of physical principles govern what can take place.

My own interests have been in mineralogy, in the constitution of minerals, and is one of long standing. These permit a sort of "quick and dirty" appraisal that one can make of primary rocks. The heterogeneous compositions include ions of varying valences, coordinated in varying degrees of tightness of bonds, and these impose the property they have incongruent solubilities.

Some kinds of ions leave the solids more freely and move into the liquid phase, especially the things like alkalis and some alkaline earths. Having left the solid, they constitute a deterioration of it, and are subject to freer transport by wind, water and weather. The ions that go to the liquid move eventually to the ocean, and the deteriorated solid products move eventually to the ocean; and in this new environment where the ions which had high solubility have been accumulating, their activities are such that when their own solid parents finally catch up with them some activity is driven back into those solids. One nice abundant type of silicate crystallization is the layer silicate that includes clays and, of course, micas and illites and with a little tolerance they have aspects in common with chains like the palygorskites and sepiolites so that a person can take a look at the nature of layer crystallization and settle down on firmly pertinent observations with respect to them.

Figure 1 shows a micro-structure of a muscovite. Pictures of this sort, of course, are always details in an infinitely repeating motif. The actual crystal extends as much as 10^6 times side-ways and up-and-down. There are

Figure 1

The structure of muscovite

Figure 2

Diffractometer record of a swollen monomineralic oriented aggregate of montmorillonite.

certain pertinent features. One is bound to notice the entities are bunched rather closely together at particular spots, moderately close together at others, and rather sparse at others. The closely knit levels are the silica sites, the less closely knit the aluminum and magnesium sites, and finally the thinly populated levels are the alkali sites.

The patterns in which closely knit regions can articulate with each other, such as in chains or cages rather than as the nets shown, are a control over whether the structure wants to remain as grains or whether it wants to disperse into thin layers that maintain great lateral dimension. Dispersals in consequence of depletion of population of these thinly occupied levels do not deteriorate to any great extent the urge that the whole complex has for holding itself together laterally. So, clay minerals, as illite and montmorillonite, which have already turned up in the discussion, are examples of this sort of crystallization. The handiest study that one makes of these structures is by X-ray diffraction character and without trying to do anything rigorous about it.

Figure 2 shows a typical collection of diffraction data. An aggregate of layers is arranged with the layers all parallel; and with continuously increasing angle of scan, a set of diffraction maxima of varying intensities is observed. Each individual maximum is displaced from the preceding by an exact equal increment of $\sin \theta$. The recorded trace of data of this nature permits analysis of relative electron density at each of the several levels similar to those pointed out on the first slide, i. e., closely bunched levels are represented by high electron densities and sparsely populated levels by commensurately lower electron densities.

Figure 3 shows relative electron density data for some examples which exhibit the layered nature we have been discussing. The densities are high in the articulated silicate skeletons and lower in the interlayered space. As dispersion is initiated to the closely controlled degree illustrated here it is notable that the high density features related to the closely articulated levels in the layer structure are essentially unchanged, but that the densities in and the volumes occupied by interlayer matter are variable.

These illustrations are instances in which a deliberate control insured that each layer level in a meticulously repeating array was attacked equally to produce a new meticulously repeating sequence. In nature, under field conditions, when minerals deteriorate, a grain does not deteriorate uniformly throughout the grain, but instead is spotted at specific levels with the result that originally meticulous repetitions are no longer meticulously repetitive.

Failures in repetitive nature have the effect of broadening and diffusing the several diffraction maxima, and as failures become increasingly frequent, the spacing of features at exact increments of $\sin \theta$ is lost. Such a diffraction record is no longer subject to the niceties of interpretation which are possible from better organized systems, and the actual imperfect record illustrated here as Figure 4 is far superior to records customarily obtainable from

Figure 3

Relative electron density plot for vermiculite in three controlled states of hydration

Figure 4

Diffractometer record for a oriented aggregate, in natural and swollen state, for two kinds of layers in near equal abundance but in random sequence

Figure 5

Diffractometer record for an oriented aggregate of a nearly regularly alternating double-layer structure, swollen and natural with a comparison plot of the calculated curve for the natural case

contemporary sediments or collected stream loads.

To obtain any information at all it is necessary to seek graded examples of only slightly inferior quality and then extrapolate to poorer and poorer qualities. To this end, I illustrate one case as Figure 5 in which alteration has effected an almost exact one half of an original sequence to produce an array like A B A B A B ---. The quality of this record is between those from good crystals and those just shown. The records with which your investigators are required to work are poorer than any of these examples.

In spite of these inherent difficulties, many of you have already made careful individual studies and have related exchange phenomena to individual minerals. I do not do that sort of stuff myself. I would rather like to tell you the way in which I feel that the character of the crystals permits a unified expression of the behavior of these materials in dispersions.

The persistent articulated masses have the character of great polyanions. As anionic functionality declines, these great laterally expansive layers can be dispersed from each other and they have the property, then, of exterior surfaces. From dispersions of this material, one can formulate that there exists a colloid and the colloid retains some negative charge. The colloid with the negative charge has metal ions associated with it, and the colloid has also a great expanse of surface available to solvation. In nature, the solvator is preponderantly water. It does not have to be. Identity of the ion need not be specified. Most of the references today have been mainly to cesium and strontium; nature turns up more frequently sodium, potassium, and calcium. Identity is not important to the exposition of principles.

The body of the colloid in dispersion is essentially a solid but the liquid that bathes it may have ionic species in it such that a metal of one kind in a solution that suspends a solid which is populated with a metal of the other kind will do the ordinary inorganic exchange. The cations are not constrained to be inorganic. An organic radical, R, and a functional group, A, with a proton, H^+ will make a cation, and in reaction with a negative colloid the organic base can release inorganic metal. The equilibria pertinent to the colloid behavior can be summarized in one schematic diagram in which each reaction is subject to ordinary mass action. The radical and its functional group are in equilibrium with the ionic species of the base according to the pH. The neutral entities are in equilibrium as neutral solvating molecules on the surface so that as surfaces are solvated with neutral organic molecules, that is, a radical with a functional group, water is released. The bottom situation is a solid and the top situation is a solid and the middle situation is the constitution of the liquid.

The hydrolysis reaction of the organic base in solution fixes its partition into neutral and ionic species, and the ionic species conforms to its exchange equilibrium equation. The activity of water in the liquid over the activity of neutral organic material in the liquid is proportional to H_2O solvating the colloid over the RH solvating colloid (the latter two being in the solid state). These

represent the same things that you were talking about as partition in Freundlich experiments. If, for example, a count of one entity in the liquid is proportional to a count in the solid, the same thing is true for some competing entities. So, these are the same equations that you are using; applying to any one absorption reaction what might also be applied to the other.

This kind of data has been collected by prominent colloid chemists farther back than I can remember. In spite of the fact the data is excellent and abundant, in a very few instances were actual constants quoted for either neutral solvation pairs or for any particular ion pair. It was the custom instead to report the ion results as in terms of lyotropic series; i. e., "A" is preferred to atom "B" is preferred to atom "C," and so on. The mass action formulation is one that could be made and would be much more descriptive. For example, if sodium and cesium are in competition with one another it comes out that the cesium to sodium ratio in the liquid is only about one-tenth the cesium to sodium ratio in the solid for Wyoming Bentonite. Actually, from among clay materials considered, Wyoming Bentonite is probably the poorest selector for cesium. So, this means that the clay is as good as a 10 to 1 collector and can go from there on up.

I have tried to take this opportunity to develop the impression that there is a thread that runs all of the way through from the rocks to their geochemical behavior. I enjoy doing the type of work I have presented and I am very happy that you gentlemen are collecting the data.

Summary of Equilibria

neutral equilibria

$$\left(\frac{\alpha_A}{\alpha_B} \right)_{\text{soln.}} = K \left(\frac{N_A}{N_B} \right)_{\text{solid}}$$

Uni - univalent exchange

$$\left(\frac{\alpha_{\text{Me}_1^+}}{\alpha_{\text{Me}_2^+}} \right)_{\text{soln.}} = K \left(\frac{N_{\text{Me}_1^+}}{N_{\text{Me}_2^+}} \right)_{\text{solid}}$$

DISCUSSION

SERATA: What kind of treatment are you using?

BRADLEY: The treatment was a solvation; a natural-water system equilibrated with ethylene glycol as the neutral organic liquid. It is more voluminous than water molecules. So, having made the equilibration, all of those places originally separated by water before must now be separated by a greater space. That is the sort of criterion that permits observation that only part of the layers are subject to the solvation reaction and the others did not move. When crystals are not meticulously repeated the quality of the data falls and with the lesser quality of the data one does well to look more than once in order to arrive at a judgment.

KAUFMAN: Do you feel that the water competes with the organic molecule more than it competes with the metallic molecule?

BRADLEY: Yes, the neutral entities are one batch of competitors and the charged entities are another batch of competitors.

KAUFMAN: Do they compete with each other as well?

BRADLEY: No, the system must remain neutral.

KAUFMAN: I was thinking of resin blending by organics in the demineralization of polluted waters. Sometimes this can be rather nasty and reduce the capacity of the resin for a metallic ion.

BRADLEY: One is required to have pH control. Protons are competitors with charge entities. So, running the pH down defeats the absorption of cations.

BELTER: In our stream studies we are trying to determine how much effort should go into clay mineralogy. Is it necessary to characterize the sediments in terms of illitic or kaolinite clays or categorize by particle size distribution? Would you care to comment on how much effort should be expended on the clay mineralogy aspects in a stream study?

BRADLEY: People do that on a personal preference basis. My personal preference is that: "If it is kaolinite it is not useful, and the others are close

enough in the family to be as well characterized by exchange capacities as is necessary." These little fellows derived from weathering have no common character and are repeated in each of several grades; they are heteorgeneous. The more they become heteorgeneous the more freely one can talk.

FRIEND: Would you comment a little more on your second equation?

BRADLEY: It is a very simple expression of exchange. Actually, I am cheating here because there has not been a firm demonstration of how one handles the divalent ions. I am inclined to vote on how I think one handles the divalent ions but no one has set out their data in form from which I could figure out whether I was right or not. I am not about to take any of my own data. I think the divalent ion deal follows this general equation:

uni - divalent exchange

$$\left(\frac{a_{\text{Me}^{++}}}{a_{\text{Me}^+}^2} \right)_{\text{soln.}} = K \left(\frac{N_{\text{Me}^{++}}}{N_{\text{Me}^+}} \right)_{\text{solid}}$$

This is only saying that the activity of the divalent in the liquid over the square of the activity of the univalent in the liquid is proportional to the mol fraction of the divalent over the mol fraction of the univalent. I wish there were data available somewhere to straighten that out. My feeling can make a difference between liquid character and solid. Mass action in liquids is something that one cannot argue with. Activity coefficients and power loss for valences are nicely substantiated. In the solid state, although we call crystals ionic crystals, they are not ions in the sense that they are full-powered ions. Both univalent and divalent ions in the solid state have electron deficiencies of the magnitude of only about 1/3 or 1/2 of an electron and the electrons that belong to them honestly are still close in the environment so that the valency powers are no longer applicable. Each of these abundances might be subject to some fractional exponent, but, as the fractional exponent declines, it comes to a matter that it would have to decline an awful lot, before it would become apparent that the ratio was any different from mol fractions.

SPOFFORD: Why don't you use the mol fraction in the liquid phase as well as in the solid phase?

BRADLEY: They set up a law against that when I was going to school and I don't believe it has ever been repealed.

SPOFFORD: It seems to me, that if you used the mol fraction in the liquid

phase as you have done in the solid phase, the actual concentrations of ions in each phase would be more consistent with the theoretical concentrations.

BRADLEY: Yes, that would be true there. There is a artificiality in great valences like 8 or 7. That entity never has that charge as an isolated individual in the solution. It always has a lesser charge with the glob that is coordinated with it. So, big powers, like 7 or 8, are fanciful and mol fractions are certainly superior to them.

SPOFFORD: I didn't have in mind valences that high; just bivalent and trivalent ions.

BRADLEY: The matter of ionic state in the crystal is less definite than in liquid, that is for sure.

MINERALOGY AND EXCHANGE CAPACITY OF MODERN FLUVIAL SEDIMENTS

by VANCE KENNEDY

In late 1959 the U. S. Geological Survey began a study of the variation of mineralogy and exchange capacity of stream sediments as related to grain size, stream discharge, time, geologic terrane, and geographic location. A progress report on that study will be presented and then some of the implications of the data and suggestions for further work will be considered.

In conducting the study, an effort was made to obtain five samples of both suspended sediment and bed material from 21 streams located in various parts of the United States as shown in Figure 1. One sample set was to be gotten during low flow, one at median flow, and three during different periods of high flow. Sample sites were restricted to locations where sediment investigations were already being made by Survey personnel. However, within this limitation, an effort was made to study as great a variety of geologic and climatic conditions as possible.

In a few streams sampling problems or poor flow conditions allowed us to get only one or two sample sets. During low to median flow periods only bed material samples were collected from most streams, because suspended sediment concentrations were very low. Nevertheless, I believe that the data obtained will give a reasonably representative picture of the conditions to be expected in our modern streams.

Both suspended and bed samples were divided into clay ($< .004$ mm), silt (.004-.061 mm), sand (.061-1.0 mm) and, if necessary, gravel (> 1.0 mm) sizes. Mineralogical analyses and exchange-capacity determinations were made upon the clay, silt, and sand fractions.

Information on mineralogy and exchange capacity of stream clays is summarized in Table I. Exchange capacities were determined using radioactive cesium. Despite all the numbers, I think you can see that there are definite trends present in the mineralogy, and this, in turn, is reflected in exchange capacity. You will remember that on Figure 1 the numbers increased in a westerly direction. In this Figure the eastern streams (first 5 numbers) appear to be characterized by kaolinite, illite, and dioctahedral vermiculite. The term kaolinite as used here refers to the group of minerals with a basal spacing of about 7 angstroms. Illite refers to a nonexpanding, essentially noncollapsing, clay mineral with a basal spacing of about 10 angstroms. Dioctahedral

Figure 1

MAP OF SEDIMENT SAMPLING SITES

TABLE I

Mineralogy and exchange capacity of stream clays (< 004 mm)

Name	Minerals (parts in ten)										Exchange Capacity meq / 100 g
	Kaolinite	Montmorillonite	Illite	Chlorite	Diocahedral Vermiculite	Mixed Layer Clay	Quartz	Carbonate	Amorphous SiO ₂ Al ₂ O ₃	Other	
1 Brandywine Cr Del	2 5		2	Tr	3		1		1	5	20
2 Juniata R Pa	1 5		4	Tr	1	Tr	2		5	1	21
3 Yadkin R N C	4		1 5	Tr	2		1		1	5	14
4 Licking R Ky	1 5		3 5	Tr	1 5	5	2		5	5	23
5 Green R Ky	1		3		2	Tr	3		5	5	21
6 Mississippi R Mo	5	2 5	2	Tr		2 5	1 5		5	5	45
7 Kansas R Kans	5	1 5	2			4	1		5	5	55
8 Elk Cr Okla		1	4	5		2 5	1		5	5	46
9 Colorado R Tex	5	4	1			2 5	1	5	5	5	58
10 Pecos R N Mex	5	5	2	5		4	1 5		Tr	1	37
11 Rio Grande N Mex	1	3	1	Tr		5	1		Tr	1	43
12 San Juan R (Bloomfield) N Mex	1	1 5	1	Tr		4	1 5		Tr	1	38
13 Animas R N Mex	1	1	1 5	Tr		4	1 5	Tr	5	5	36
14 San Juan R (Shiprock) N Mex	1	2 5	1	Tr		5	1 5		Tr	5	--
15 Colorado R Utah	1	1 5	2	5	Tr	2	1 5	5	Tr	1	30
16 Cheyenne R S Dak	1 5	1 5	1	Tr		2	3	Tr	Tr	1	--
17 Big Horn R Wyo	1 5	2	2	Tr		2	1 5		Tr	1	41
18 Cosumnes R Calif	2 5	1 5	1			4	Tr		5	5	32
19 Cache Cr Calif	1	1	1 5	1 5	1	2	1		Tr	1	33
20 Mad R Calif			2 5	3 5		1 5	1		5	1	18
21 Crooked R Oreg		4		Tr		3 0	5		1 5	1	63

vermiculite is characterized by a basal spacing of about 14 angstroms which does not expand appreciably on glycollation but collapses gradually to 10-11 angstroms when heated over a range of several hundred degrees. The exchange capacity of the minus 4 micron material in 4 of the 5 streams is about 20 milliequivalents per hundred grams. The Yadkin River clay has considerably more kaolinite and less illite than the other streams and, hence, has a lower exchange capacity.

As one moves westward little, if any, dioctahedral vermiculite is found and the kaolinite content decreases. In place of these minerals montmorillonite and mixed layer montmorillonite-illite are prominent constituents and the exchange capacity rises accordingly. One exception to the trend is the clay suite in the Mad River near the coast in northern California. There, chlorite and illite are the predominant clay minerals and the exchange capacity is about 20 meq/100 g. Another point that should be made is that the Crooked River clay does not collapse well at 300° or at 550° C, indicating that chlorite-montmorillonite mixed layer material may be present.

It should be emphasized that the numbers shown in the Figure are intended to be only estimates of the amounts of the various clay minerals. The presence of three to four different clay minerals plus some mixed layered minerals in a single sample causes some analytical problems, particularly since poor crystallization is also a complicating factor.

In Table II the exchange capacities of the various size fractions are shown. You will note that the percentage variation in exchange capacity of the clays is considerably less than that of the silts or sands. The Crooked River clay at 63 meq/100 g is about 4.5 times that of the Yadkin River clay. But the Crooked River silt is almost 8 times that of the Yadkin River silt and the Crooked River sand is about 40 times that of the Yadkin River bed sand. You will also note that the exchange capacity of suspended sand or silt tends to exceed that of similar sizes in the bed material. This is partly due to the fact that the average size of bed silt or sand tends to be considerably larger, and thus has less surface area, than that of suspended silt or sand. The greater exchange capacity of the suspended sand and silt may also be due in part to a difference in mineralogy or organic content.

The exchange-capacity data presented are averages and so do not show variations between samples from any one stream. The available information suggests that, as a general rule, one sample from a stream will give a good idea of the mineral suite to be expected at any time. In many streams there do not appear to be great variations in exchange capacity or in relative quantities of the various minerals in any of the size fractions. However, it is not unusual to find wide variations in the proportions of the various minerals when a stream traverses regions of widely varying geology or drains areas of widely varying climate.

Not all of the analytical information on the mineralogy of the sand and silt fractions is available as yet, but some tentative conclusions can perhaps be drawn from available data. You may have noted that some stream sands

TABLE II

Exchange capacity of various components of stream sands

Sampling site	High-iron fraction		Rock-fragment fraction		Quartz-feldspar fraction		Percent of total exchange capacity due to rock-fragment fraction
	Exchange capacity (meq/100 g)	Weight (per-cent)	Exchange capacity (meq/100 g)	Weight (per-cent)	Exchange capacity (meq/100 g)	Weight (per-cent)	
Juniata River Newport, Pa.	4.9	1.2	6.1	20.5	0.2	78.3	85
Licking River McKinneysburg, Ky.	11.1	2.1	6.5	52	.2	45.9	91
Kansas River Wamego, Kans.	4.9	.8	2.7	13.7	.4	85.5	49
Elk Creek Hobart, Okla.	1.8	.3	5.0	15.2	.4	84.5	66

have amazingly high exchange capacities. Such exchange capacities appear to be related to the percentage of fine-grained rock fragments in the sand and to the clay minerals present in those rock fragments. This is illustrated in Table III. The separations were made using a Frantz magnetic separator. The high-iron fraction was composed primarily of magnetite, ilmenite, and iron-stained rock fragments containing these highly magnetic minerals. The rock fragment portion ranged from heavily iron-stained mudstones to quartz or feldspar aggregates cemented with iron-stained clay. The quartz-feldspar fraction contained relatively clean grains having very little iron-stained clay as a cement or coating.

It is apparent that the rock fragment portion of the stream sand is responsible for much of the exchange capacity.

The high exchange capacity of some of the silts indicates that grain aggregates are common in this size range, also.

You have seen data indicating the range of clay mineralogy found and the effect of grain size on exchange capacity in modern streams. Some idea of the importance of this exchange capacity in controlling the chemical composition of stream water can be gotten by comparing the amount of cations in solution to those adsorbed on suspended sediment or on bed materials as indicated in Table IV.

In this Figure I've considered two flow conditions. The upper half of the Figure pertains to the ratio of adsorbed ions to dissolved ions under high flow, high suspended sediment conditions. The lower half indicates the thickness of bed sediment required to make the adsorbed cations equal to those in solution in the overlying water under conditions of average flow.

In the case of each of the five streams, I went through our published and unpublished records to find a period of high flow when suspended sediment concentration, particle-size distribution, and chemical composition or specific conductance were known. If specific conductance instead of chemical composition was known, a plot of equivalents per million versus specific conductance was used to estimate cations in solution. Using our exchange-capacity data for the sand, silt, and clay fraction the exchangeable cations adsorbed could be calculated. The ratio of adsorbed to dissolved cations is shown in line three. Although the suspended sediment concentrations were high under these conditions, they were not the highest on record, because in no case did we have chemical and/or particle-size information during periods of record high sediment concentrations. If we assume, however, that particle-size distribution of suspended sediment and chemical composition of stream water was about the same at the time of record sediment concentration as at the time referred to in lines 1 and 2, we can estimate the ratio of adsorbed to dissolved ions under record conditions. These ratios are shown in line four.

TABLE III

Sampling Site	Cation exchange capacities of some modern stream sediments					
	Average exchange capacity in milliequivalents per 100 grams					
	Suspended Sediment			Bed Material		
	Clay <.004 mm	Silt .004-.061 mm	Sand .061-1.0 mm	Clay <.004 mm	Silt .004-.061 mm	Sand .061-1.0mm
Brandywine Cr., Del.	19	10	6	22	8	2
Juniata R., Pa.	15	11	5	22	7	3
Yadkin R., N. C.	15	9	6	14	4	0.3
Licking R., Ky.	19	11	-	28	7	6
Green R., Ky.	20	11	-	22	7	1
Mississippi R., Mo.	51	19	3	40	11	1
Kansas R., Kans.	56	21	2	52	10	0.6
Elk Cr., Okla.	46	26	3	46	10	0.6
Colorado R., Tex.	64	26	1	52	17	0.4
Pecos R., N. Mex.	41	15	4	32	7	1
Rio Grande R., N. Mex.	43	28	3	--	--	1
San Juan R., (Bloomfield) N. Mex.	38	17	2	39	15	1
Animas R., N. Mex.	27	18	6	50	15	3
San Juan R., (Shiprock) N. Mex.	40	19	3	--	18	1
Colorado R., Utah	29	17	3	31	8	2
Cheyenne R., S. Dak.	--	--	-	25	--	1
Big Horn R., Wyo.	40	14	3	42	13	2
Cosumnes R., Calif.	27	11	7	34	15	4
Cache Cr., Calif.	--	--	-	33	13	4
Mad R., Calif.	17	12	6	18	9	6
Crooked R., Oreg.	--	--	-	63	31	12

You will note that although the dissolved cations ranged from 0.44 to 28 milliequivalents per liter in line one (a factor of increase of about 60) the estimated maximum ratio of adsorbed to dissolved ions ranged from 0.2 to 0.7, a factor of only 3.5. There appears to be a tendency for the maximum ratio of adsorbed to dissolved ions to be relatively constant under conditions of widely varying climate and geology. This is probably due to the fact that streams in a moist climate tend to be low both in dissolved solids and in suspended sediment. Also, the exchange capacity per unit weight of the suspended sediment is low. As one goes to more arid areas all three factors tend to increase, thus holding the ratio of adsorbed to dissolved ions relatively constant.

Under average to low flow conditions, suspended sediment concentrations are usually very low, and the ratio of adsorbed to dissolved ions is probably less than .001. Thus, suspended sediment would be expected to have very little effect on water quality under these conditions.

The bed material, however, will become relatively more important in buffering water composition as stream flow and suspended sediment concentrations decrease. An effort has been made to show the importance of bed sediment under conditions of average flow in the lower half of Table IV. In Brandywine Creek, for example, there would be approximately 6.3 milliequivalents of cations in solution above 100 square centimeters of streambed. If we assume a porosity of 50 percent in the bed sediment, then a layer of clay 0.9 centimeter thick would hold about 6.3 milliequivalents of ions adsorbed. If the bed sediment were of silt size its exchange capacity per unit weight would be less and a thicker layer would be required, in this case, about 2.7 centimeters. Sand, with an even lower exchange capacity, would have to be 10 centimeters, or about four inches thick. In the Colorado River near Cisco, Utah, both the depth of water at average flow and the concentration of dissolved ions are greater than in the Brandywine, and greater thicknesses of sediment are required to make adsorbed ions equal to those in solution. One should realize, of course, that the depth of flow is less than that at average discharge for well over half the year.

The information just presented is intended to give you an idea of the relative amounts of adsorbed and dissolved cations under varying conditions of flow. Many of the complications in this picture have been ignored.

First, mention should be made of the adsorption selectivity which certain minerals exhibit. Such selectivity is a well known phenomenon although much work remains to be done in determining what minerals are selective for what elements. The research that has been done to date has tended to concentrate on relatively pure and well crystallized clay minerals. Although such minerals do occur in stream sediments, and this is a logical place to start, it should be noted that poorly crystallized clays, mixed layer clay minerals, amorphous material, and, in the eastern United States, dioctahedral vermiculite may comprise a major portion of the stream clays. Investigation of the adsorption selectivity of these materials should properly be included in further

TABLE IV

Comparison of adsorbed and dissolved cations

			Sampling sites				
			Brandywine Cr. Del.	Yadkin R. N. Car.	Licking R. Ky.	Colorado R. Utah	Crooked R. Oreg.
High flow conditions	Dissolved cations (meq/liter)		0.71	0.44	1.5	28	1.7
	Cations adsorbed on suspended sediment (meq/liter)		0.086	0.15	0.14	6.2	.12
	Calculated Ratio $\frac{\text{adsorbed cations}}{\text{dissolved cations}}$		0.12	0.34	0.095	0.22	.073
	Estimated maximum Ratio $\frac{\text{adsorbed cations}}{\text{dissolved cations}}$		0.2	0.7	0.4	0.6	0.2
Conditions	Cations in solution above 100 sq cm of bed (meq)		6.3	4.2	34.3	107	8.3
at average flow	Thickness of sediment required to make cations adsorbed on bed equal to those in solution above (cm)	Clay <.004 mm	.9	.2	.9	2.7	.1
		Silt .004 - .061 mm	2.7	.8	3.8	10	.2
		Sand .061 - 1.0 mm	10	11	4.4	40	.5

studies.

Particulate organic material was included in our samples and no effort was made to destroy or separate it. Thus, a part of the exchange capacity reported is undoubtedly due to this organic material. Further investigation of the importance of this stream-borne organic matter in transport of various elements seems necessary.

A significant part of the exchange sites on stream sediments may be occupied by ionized organic compounds, so that the actual exchange capacity for cations in dilute solutions may be considerably less than that indicated in the Figures. We should know more about the ratio of inorganic to organic ions held on stream sediments.

Judson and others (1959) reported that slow precipitation of iron hydroxide carries down essentially all the fission products except cesium from a neutralized solution from the Purex plant at Hanford. Such slow precipitation of iron hydroxide may occur in streams and be an effective method of removing some trace elements from solution. When the streamflow decreases following a rise, commonly some sediment will be deposited on the bed along with organic material. If the permeability of the sediment is relatively low an anerobic condition develops beneath a thin skin of oxidized sediment. The ferrous iron content of the interstitial waters may increase and reach 10 ppm or more. When the next heavy runoff occurs this sediment will be thrown into suspension and the ferrous iron mixed with the stream water. Oxidation and precipitation of this ferrous iron can then proceed. The significance of this mechanism in removing trace elements from solution is not known, but it may be quite effective near the front of an advancing flood wave.

There is the possibility that radioactive elements may be held on organic or inorganic sediment and then released to solution when deposited and exposed to a reducing environment. This reducing environment can also cause removal of iron coatings on clays and perhaps make exchange sites more accessible.

In the early part of this discussion reference was made to the thickness of bed sediment required to hold adsorbed ions equal in amount to those in solution in the water above. The implication was that the adsorbed ions could interact freely with those in solution to buffer water quality. Such an implication may not be justified, however. If there is vertical movement of water through the bed material, then ionic diffusion into or out of the bed will be hampered. Commonly, a stream will tend to lose or gain water from the water table, and exact equilibrium with the water table would seem to be a condition which couldn't persist for very long periods.

If this is so, there will be a tendency to flush water out of the streambed into the stream or to flush interstitial water down through the bed sediment. If the exchange capacity of the bed material is to act as a buffer on the chemical composition of stream water, ions must be able to migrate both into and

out of the bed. When the rate of vertical flow (either up or down) exceeds the rate of ionic diffusion, the bed material below the zone of active sediment transportation will have little, if any, effect on the chemical composition of the stream water. In fact, if inflow to the stream is relatively rapid and bed material movement slow, upper layers of the stream water may be separated fairly effectively from interaction with any bed material.

It would appear quite desirable to learn more about the actual ability of the stream water to contact the bed sediment before any definite conclusions can be drawn regarding the probable effect of bed sediment on water quality.

In summary, the data presently available indicate that under appropriate circumstances, the exchange capacity of stream sediment can be quite important in controlling water composition. It appears that one or two samples of suspended sediment concentration and particle-size distribution will permit a rough estimate of the amount of adsorbed ions transported by a stream. If chemical data are also available, the ratio of adsorbed to dissolved ions can be calculated.

Very little information is available regarding the mineralogy and size distribution of bed material over a reach of stream, although some data are available at bridge sections. Even with such information over a stream reach, it would be impossible to estimate the probable effect of vertical water flow through the bed sediment, the effect of mineral selectivity, and the effect of organic material without considerably more knowledge than we have at hand now.

It seems to me that a logical next step is to select a very few streams which demonstrate considerable variation in factors likely to affect the interaction between water and sediment. These streams should be studied in great detail so that we can understand the relative importance of these various factors. Only then can we state what measurements are necessary when we wish to learn how radioactive nuclides will be transported in a previously unstudied stream.

Although I have not mentioned the fact previously, it seems obvious that biological and biochemical studies should be tied in closely with the mineralogical and chemical work.

REFERENCE

Judson, B. F., Moore, R. L., Van Tuyl, H. H., and Wirts, R. W., 1959, Recovery of radioactive cesium at Hanford: Chem. Eng. Progr. 55, Symposium Ser. No. 23, p. 1-4.

DISCUSSION

BELTER: Is the Crooked River analyses in Oregon representative of what you would expect of the Columbia River?

KENNEDY: I don't believe so; the Crooked River drains an arid environment where the rocks are almost entirely tuffaceous or basaltic. Although there would be a fair amount of this material in the Columbia drainage, I think the Crooked River represents an extreme case. We were fortunate, I think, that we sampled the Crooked River sediments. We had a limited number of streams for which we had a sediment record, so we had to work with those. Fortuitously, we have the Crooked River at one extreme of sediment mineralogy and exchange capacity, and the Yadkin River near the other extreme.

KAUFMAN: Why did you use the cesium method in determining the exchange capacities? What was the basis for selecting this method?

KENNEDY: The major reason that we used it was that we were working with another Survey group which has a set-up for making exchange determinations by the cesium method in large numbers. Thus, the cesium method was most convenient for us.

KAUFMAN: How do you feel these numbers would compare, say, with exchange capacity numbers determined by ammonium acetate or exchangeable calcium?

KENNEDY: I suspect that there would be some considerable amount of variation, depending on the amount of illite, for one thing. I might emphasize that this is a progress report. We are hoping to get some exchange capacity determinations by the ammonium acetate method so that a comparison can be made.

BERNARD: Would you say that if you took a number of samples of the bottom sediment at one time you could get sufficient statistical data so you wouldn't have to return during high and low flows for additional samples?

KENNEDY: Well, the point I was making is that you can get an idea of the kinds of minerals that you are dealing with, but not of the relative amounts over an extended period of time. In general, there is some homogenization of

sediments because a part of the sediment carried during one storm period will be buried in the bed until the next high-flow period. As discharge increases again, sediment from the bed will be picked up and mixed with the sediment derived from outside the stream channel. The net result is that, at any one time, you can get some idea of what minerals are present but cannot predict accurately the relative amounts of these minerals during some other period of time.

BERNARD: This is one of the points. When you take several samples during this period you actually obtain a range of values. Now wouldn't you observe a similar range of values by taking enough samples at one time as you would by taking a small number of samples at several flow conditions?

KENNEDY: If you did this at one site, I don't think so; but if you were to sample all the major tributaries at one time, I think you could establish the kinds of materials that would be dumped into the stream by these tributaries. If the sediments from the tributaries were essentially constant in their mineral composition, then I wouldn't hesitate to predict the mineral composition of sediments in the main stream. In Brandywine Creek, for instance, we obtained one sample from each of the major tributaries. Then, we mounted the clay minerals on tiles and ran off a series of x-ray patterns. It's apparent just from glancing at the patterns that they are all similar. I would guess, therefore, that in Brandywine Creek we wouldn't get much variation in clay mineralogy from time to time. Now, on the other hand, when we sampled the San Juan River at Bloomfield and the Animas River at Farmington, N. Mex., right after a high-flow period, we got an exchange capacity of about 37 meq for the clay being carried in the stream itself, but a sample of clay from moist sediment along the bank had an exchange capacity of about 60 meq. X-ray diffraction showed a greater proportion of montmorillonite in the sample with the greater exchange capacity.

In the San Juan River at Shiprock the exchange capacity of clay in both suspended and bed material was about 60 meq/100 g. A stream rise had begun perhaps 12 hours before we did our sampling on the Animas at Farmington, and the data suggest that the clay minerals carried during the early part of the rise were different from those in suspension when we did our sampling. Inasmuch as the distance from Farmington to Shiprock is about 30 miles, there would have been time for the high-exchange-capacity clay from the Animas River to enter the San Juan River and travel to Shiprock before we collected our Shiprock sample. This is a tentative interpretation of the situation.

BERNARD: Was it a low flow condition?

KENNEDY: Well, commonly the material here has an exchange capacity in the neighborhood of 40 meq/100 g. But, as you follow the Animas River upstream to Durango, you go through an area that differs considerably in climate and geology from the area upstream from Durango. Therefore, I would expect to get variation in mineralogy and exchange capacity of stream sediments depending upon what part of the drainage basin supplied the sediment being transported. In the Brandywine, on the other hand, the geology is quite varied, but

you are dealing with soils that are moderately well developed so there is a tendency to homogenize the clay mineralogy.

BELTER: Could you put Figure 5 back on? I know you have other ones all lined up but there was one question I had on that last figure of yours.

PARKER: Would you say why you used the 4 micron rather than the 2 micron size?

KENNEDY: Yes, when separating clay from silt geologists usually use 4 microns, whereas soil scientists use 2 microns. However, that wasn't our main reason. We wanted to use past records as much as possible, and many of the field offices make their break at 4 microns. In order to make use of their records we separated clay and silt at 4 microns.

BELTER: In a Kentucky stream and the Colorado stream both of those factors are a couple of orders of magnitude greater than Brandywine Creek. What is the difference in the sediment mineralogy and also the chemical characteristics of the streams which might cause that great a difference?

KENNEDY: The Colorado is a deeper river and has a higher dissolved salt content than the Licking River. You multiply the dissolved salt content by the depth of the water to estimate the cations in solution above 100 sq. cm. of bed. Therefore, under average flow conditions the Colorado River has a higher amount of dissolved salts above a unit area of streambed than does the Licking River. Brandywine Creek is shallower and has a lower dissolved content than either the Colorado or Licking Rivers. In Delaware, you have a much more humid environment than in the Colorado River drainage basin. The dissolved salts are usually low under such humid conditions. In the case of the Colorado River, however there is a moderate amount of runoff coming in downstream from Palisades that would be high in dissolved salts. There is also an inflow from irrigated areas with a high salt content.

KAUFMAN: Would you venture a guess as to what the radiological significance of that line of numbers is in terms of potential hazards of one sort or another, say strontium, cesium, loading in the river? Do they infer anything? You have derived a rather complex number, presumably as an index of the radiological problem?

KENNEDY: I simply wanted to point out the thickness of bed sediment that is required to hold as many ions adsorbed (in terms of milliequivalents) as you have in solution above the streambed. The sediment thicknesses calculated are of the same order of magnitude that you might expect to find in some natural streams. For instance, I wanted to point out that, in the Brandywine, only 9 millimeters of clay are required to hold adsorbed as many cations as would be in solution above the bed under average flow conditions. This suggests that the bed material is a significant factor in acting as a buffer on the water composition. This is the only point to be made from this. To make this quantitative

I think you have to go into more detailed studies than what we have done. These are just gross exchange capacities. For instance, in the Colorado River, I think it is apparent that we are considering about 40 centimeters, or 12 to 14 inches, of sand. Under average flow conditions the sand waves may be about 20 centimeters high. The lower half of the 40 centimeters required may be below the zone of sediment movement. This is where the question of which way the ground water is moving comes into consideration. If you have water coming up through the bed, you are going to prevent interaction between the flowing water and the bed material that is below the zone of sediment movement.

SAYRE: With respect to the question of interaction between the water and the sediment I think that in many of the sand-bed streams it is somewhat unrealistic to think of the sediment as being down there and the water as being up here because a significant proportion of the bed material will spend some time in suspension and will come back to the bed again.

BRADLEY: Doesn't this say that every time there is a condition of high flow that there is effective complete sediment turnover in any one of the streams?

KENNEDY: Well, I would guess that you could interpret it this way because the full 40 centimeter thickness of bed material presumably would be moved during periods of high flow in the stream.

BRADLEY: Even over on the Colorado River?

KENNEDY: Yes. I do not know how deep the bed is cut or scoured under high flow conditions, but it seems reasonable that scouring would be greater than 40 centimeters.

KAUFMAN: Has anyone made any determination of the interstitial water velocities in these beds?

KENNEDY: Not to my knowledge, but that doesn't prove that it hasn't been done. One of the things that I think we could consider doing would be to add some material, perhaps a dye, to a stream for a day or two and see how deeply this penetrates into the bed. One would then stop adding the dye and wait for a day or two to see if the dye moved back out of the streambed. This could vary with grain size or the velocity of the water coming in.

SAYRE: In some of the experimental work that has been done with fine sediments at Colorado State University it has been found that the concentration of fine sediment, say bentonite, in the pore water in the bed is just about the same as the concentration of bentonite in the water flowing above the bed. Therefore, when the concentration, let's say in the flow is reduced, the concentration in the pore-water goes right down with it. It is a pretty accurate indicator of the concentration in the flow because quite thorough mixing takes place between the pore-water and the water in the flow.

KENNEDY: There is no vertical movement, though, in these circumstances?

SAYRE: In that particular set of circumstances there was no vertical movement.

KENNEDY: I think if you can assume that there is no vertical movement in these beds then there is no reason to assume that, given sufficient time, you couldn't get ionic diffusion down to this point (40 centimeters below surface of streambed) and back up. If you were to change the chemical composition of the water, this sediment would act as a buffer over the whole depth of ionic movement. It is this question of water movement in the bed that could complicate things.

FOSTER: Does anyone have information for specific ions on the rate of exchange between water and the sediments. In other words, if one considers the sediments as a reservoir for hold-up what would be the residence time of, say, a cesium atom in going into and coming back out again.

KENNEDY: I doubt that you could make a general statement on this because, in the case of clays, some of the people who are working on our ion exchange project thought that 99% of the total exchange (as distinguished from uptake in the mineral lattice) takes place in a few minutes in the case of finely divided clays. One of the questions that we haven't explored yet is that of finding out how deeply into the sand grains the replacing ions penetrate. I can easily imagine a situation where you would attempt to saturate a sand made up of rock fragments and it would take several hours before you got complete penetration. Maybe it wouldn't even occur in 10 hours. We don't know this yet. On the other hand, if one changed the composition of water in contact with this rock fragment, it's going to take awhile for ionic diffusion to equilibrate the material adsorbed in the inner part of the rock fragment with that in solution. I would presume the time required would be measured in hours in the case of sand-size rock fragments.

FOSTER: I'm thinking here of the situation where you have a high discharge of waste into a stream. Do you have a situation when you first start this discharge in which the bed material is rapidly saturated with the newly added material and then get virtually no exchange thereafter. I usually envision a continual ion exchange phenomena which has some implication in relation to the discharge of the shorter-lived materials. If you have a shorter-lived nuclide, you perhaps get the effect of a retention reservoir in the sediments in which the material moving down the stream might be moved into the sediment bed and back so you could take advantage of some depletion by virtue of the delayed transport.

BRADLEY: Mr. Bob Garrells made some studies on exchange rates by using ground muscovite. By moving to a mineral where the process (in his case, hydrolysis) was slowed up enough so that times were measurable, he got a system which netted him periods like hours in which to make measurements. In material like we deal with here I think the exchanges run too quickly for study.

KENNEDY: Yes, in the case of the clays; but I don't know about the sands.

BRADLEY: Well, the sands have some muscovite flakes in them too.

KENNEDY: Yes, on the surface. The question concerns the material inside.

BERNARD: The percentage of the exchange capacities of the clay to the total exchange capacity is very high. The effect of the uptake by the sand grains should be insignificant.

KENNEDY: Well, in one type of stream this is true, but this isn't a generalization. In the Crooked River there is relatively little clay carried by the stream, but there is quite a bit of sand which has a high exchange capacity. In the case of the Yadkin River your statement is probably true because the exchange capacity of the sand is only 0.3 of a milliequivalent/100 g.

BERNARD: To what do you attribute the high capacity of the sands in the Crooked River?

KENNEDY: The Crooked River sand is made up of about 70% rock fragments. Not only that, but the rock fragments probably have montmorillonite coatings. We may have a high-exchange-capacity clay forming coatings and, in some cases, also penetrating the rock fragments themselves. It's a situation where you have two conditions which are favorable to a high exchange capacity.

KRUMHOLZ: Have you done anything at all with organic detritus in these streams?

KENNEDY: We have not.

KRUMHOLZ: Are you planning on it or is anyone planning on it?

KENNEDY: Well, I suggest that we definitely look at this if further work along this line is done. I personally feel that this is an important omission. We can see particulate organic material in some sand fractions, and one of the things that I have been doing is counting organic material along with the mineral grains in thin section. There hasn't been much evidence for tearing up of the organic material during sectioning; I don't see holes, for instance, with little bits of organic material at the edges. We can get some information regarding the variation of organic matter in sands from one particular stream or from different streams, so we may be able to get at this question a little bit. But as far as separating out organic matter and measuring it, this hasn't been done and it won't be done on this project.

KRUMHOLZ: The reason I mentioned this is that we have very good evidence that this is a very important part of the food chain.

BELTER: Is this for just specific isotopes maybe like cobalt or is it for gross activity?

KRUMHOLZ: This is for gross activity. The reason I mention this is that we had good information from Oak Ridge that a live plant accumulated certain radionuclides and then when this live plant died and fell in the water it acted more or less like a sponge and contained more radioactive material than the live plants simply because it was indiscriminate in the material attached to it by adsorption.

NELSON: Were you ever able to discriminate between bacteriological activity on this and microbiological uptake in contrast to your absorption?

KRUMHOLZ: No, we did not study that phase of the metabolism of the community.

KENNEDY: We were dealing with exchange capacities and we didn't really concern ourselves with the ability of streambed materials to precipitate certain elements. I think that this is a factor that certainly should be considered. For example, we get carbonates in some of these stream sediments. There is a possibility that elements such as strontium may be precipitated on the carbonate. The content of carbonates in stream sands is usually less than what we can pick up easily in thin sections. You might expect elements like radium to be precipitated out on some carbonates. In work that I did about ten or eleven years ago in Wisconsin, I found a spring containing one part per million zinc. The water flowed about 105 feet down through an area of organic matter mixed with silt and lost about 99-1/2% of its zinc in that distance. The zinc was removed in a matter of about twenty minutes; it was an extremely effective method of removal. In that case the bed sediment contained 3 to 4% zinc.

NELSON: This is probably a chemical precipitation and the zinc is a carbonate rather than associated with the plant material. This phenomenon is pretty well known in England where there are a number of mines and they find a gradual influx of zinc to streams but the zinc does go down and become a part of the bed load in an insoluble form.

KENNEDY: Well, I presume that this is a possibility. The greatest concentration occurred within a distance of about 15 or 20 feet of the spring.

BRADLEY: Mr. Kevin Tiller, at Cornell, with Rockefeller support, made a study of plating out of cobalt on particular microcrystals of vermiculite. Of the cobalt that those crystals took, what plated out at faults in the actual crystal was in the nature of a precipitate and not subject to exchange by material in a solution that bathed it. After the preparation was made some cobalt could be moved off by an exchange process and the rest had been incorporated in the faults in the actual crystal. It took a substantial acid extraction to recover what was on the grain boundaries. He monitored his job with autoradiographs. These consist of contact exposures, following which he could strip back his emulsion

and develop it and get a pattern of spots at which cobalt was concentrated. Both his and the other precipitation processes can be active and differ several magnitudes in degrees of difficulty of subsequent extraction.

FOSTER: This was possibly work done in the chemical effluent technology group at Hanford.

THOMAS: Several years ago I made some preliminary studies on a flume with dyes and sands run in the water in such a way so that sand waves and ripples would form. My recollection is that with ripples, the velocity difference at the top and the bottom of the sand undulations were of a sizeable order. Dye was drawn into the sand by the Bernoulli effect. This effect is of at least the same order of importance as a ground water drainage and recharge. There is much more conduction in the granular media than one would expect. This movement together with the ground water interchange are much more important than molecular diffusion in the interstices of the bottom sediments.

FRIEND: It seems like every conference I go to someone wants to talk about activity. You know you can fall into fallacious reasoning. I think you have to be specific. In one plant effluent we monitored cesium was normally discharged as 137 but about four years ago they had a spill of 134. And we can still find 134 in this sediment, much of it in the same place, in the same small stream so to say that it goes in and comes back out is not quite true. You have to specify what element, what isotope. As far as I am concerned this cesium-134 is going to be there until it decays unless there is a big flood which scours it all out.

KENNEDY: I would presume that this might vary somewhat with the element that is involved; cesium, for instance, might be taken up and held in the lattice and no longer be available.

FRIEND: Very true. You must say what you are speaking of and not activity.

PARKER: While we are talking about cobalt, Dr. Tamura at Oak Ridge has done some work on cobalt and a lot of the uptake that we have attributed to organic detritus doesn't appear to be solely that but uptake by attachment to any surfaces that are available.

FREDERICK: Do you propose to do any further studies?

KENNEDY: My supervisor is trying to settle this for me while I summarize this phase of the project in a report. The nature of further work is still uncertain.

COWSER: It seems to me that you have a valuable interesting series of sediment samples from various streams in now of the United States. With your records you have a knowledge of the chemical quality of the water in the streams. It appears that one might be interested in making a calculation of the total amount

of radionuclides that one would expect in suspended materials if these nuclides for some reason get into this water. With information, say on distribution coefficients, you could make this type of calculation.

KENNEDY: Well we considered this in some of our earlier samples, and it became obvious that we had sample preservation problems. By the time the samples arrived at the laboratory I had no way of knowing whether I had the same distribution as was present in the stream. We did make some measurements of absorbed ions, but I would hesitate to publish them because I do not know what they represent. My strong feeling in this regard is that we must improve our sample preservation procedures a great deal before I would want to go into studies of this particular type.

KAUFMAN: I don't think we need any more rate studies in short and long columns in the laboratory concerned with the exchange between various ions and natural exchangers until we know what to do with these rate data in the field. We already have an enormous amount of data on rates of exchange under highly idealized column systems. However, we still don't know what to do with it in a natural environment.

KENNEDY: The approach which I've been using is essentially an empirical one. There is another Survey group in Denver which has been concentrating on a laboratory study of ion exchange. Presumably, if you do the field studies, you have some idea of what system your laboratory studies will ultimately have to explain. Thus, you can guide your research in the laboratory using data obtained in the field. We have been trying to cooperate in this fashion.

BEHAVIOR OF RADIONUCLIDES IN THE COLUMBIA RIVER

by J. M. NIELSEN

I might mention, in case it does not become apparent, that I am a radiochemist, actually a physical-colloid chemist, and my interest and the interest of my group are in radiochemical techniques as they can be applied to trace radionuclides in the environs.

Figure 1 shows the Hanford environs and that is where we are doing most of our study. Actually, this is in the southeast corner of the State of Washington. The Columbia River arises in Canada and flows southerly until it enters the Hanford reservation where it makes a large bend to the north and back to the south. It continues south, curves to the west and ultimately becomes the boundary between the States of Washington and Oregon, flowing almost directly westwardly until it discharges into the Pacific Ocean. The Hanford reservation is 600 square miles in extent. There are reactor areas along the river; the reactors are actually stretched out about 12 miles along the river banks.

The Hanford reactors use river water which has been treated by a process which is almost the same as a conventional water treatment process for municipalities. This water is used to cool the reactor and after the reactors are cooled the water, being thermally hot, is discharged into basins and then released to the river. The water is released to the river at many points along the 12 mile stretch and is discharged through large pipes to the center of the river and current. In the course of going through the reactors, many of the salts become activated giving many radionuclides, in fact, since uranium is an element in the water we get all of the fission products. To give an example, we have separated, identified, and measured over 100 specific radionuclides formed in this process and released to the river. Although Hanford releases something like 2,000 curies a day of radioactive material, the Columbia River is large and the flow rate is something like 60,000 cfs so that by the time the water gets down to Pasco, Washington, partly due to decay and partly due to dilution, the concentration of radionuclides there is such that if you could count all of the disintegrations in one milliliter of water there would be about 20. Remember, however, that this represents some 100 radioisotopes. To measure a specific radionuclide we must use quite sensitive techniques, large volumes of water, or both.

Hanford is not desirous of releasing many radionuclides to the environs; therefore, much effort has gone into reducing the releases so this complicates the problem of using the radionuclide tracers introduced into the stream to

Figure 1

study the movement of the radionuclides to the environs.

One of our first studies was to determine the distribution of radionuclides released 38 river miles upstream from Pasco, Washington, the first city below the reactors to use Columbia River water for its domestic water supply. You will notice that Priest Rapids Dam which is just upstream from the Hanford reservation has been operating for about one year. It controls the river so severely that it can change the amount of water in one day going by Hanford by as much as a factor of 4 at certain times of the year. The dam is for power production and draws its power when needed. Note that the Snake River enters the Columbia just below Pasco. Also notice the location of the Yakima River. I also direct your attention to McNary Dam, the pool of which was formed in 1954.

Figure 2 may take some explaining; it was difficult to get all of the information I wanted on the single figure. I tried to bar graph the ionic constituents of the water. The first bar represents 100% and I further expanded these areas from one to another. There is quite a wide range of concentration of constituents in the water. The last two bars indicate those radioisotopes which contribute to radiation exposure to the people at Pasco. This graph was based on records obtained several years ago and the concentration varies depending upon the radioisotope release. I want to point out that arsenic in the water represents the 9/10,000 of a percent of the salts and yet the radionuclide, arsenic-76, represents the major hazard. If you consider the eating of fish then the most important radionuclide is P^{32} and phosphorus represents less than 2/10 of a percent of the total salts.

We can take samples of the material released to the river. During the course of the 38 miles to Pasco the river is mixed. We can sample at Pasco and knowing the volumes of water in the river, the volumes of waste released to the river, the rate of flow, and the time that it takes to reach Pasco I can determine how much of each radionuclide gets to Pasco. In Figure 3, I have plotted how much does not get to Pasco. The samples were taken over a period of years, mainly in the winter, although one was taken during October when the water is quite warm, the biotic life is active and I was interested in seeing if the active biotic life would change the removal. It changed it very slightly. For chromium-51, which is the major radioisotope in the water below Hanford on a dpm basis, about 15% does not get to Pasco. Half of the sodium-24 does not get to Pasco and that is rather interesting because you think of it as being in solution. About 30% of neptunium-239 does not get there, 57% of the copper, 40% of the arsenic, and so forth.

Figure 4 shows the percentage depletion of radioisotopes. You will notice that somewhat between 0 and 65% does not reach Pasco, at least we cannot find it in the water.

Figure 5 shows that we actually continue the measurement of some of these radioisotopes, the longer lived radioisotopes, downstream from Hanford.

REACTOR EFFLUENT RADIOISOTOPES FROM PROCESS WATER SALTS

Figure 2

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APPROX 0 67

NATURAL DEPLETION (EXCLUDING DECAY)
BETWEEN REACTOR OUTFALLS AND PASCO, WASHINGTON

Figure 3

Figure 4

**PERCENTAGE DEPLETION OF MAJOR
RADIOISOTOPES IN THE COLUMBIA RIVER
FROM THE REACTORS TO PASCO, WASHINGTON**

<u>SAMPLE</u>	<u>RADIOISOTOPE</u>							
	<u>Cr⁵¹</u>	<u>Na²⁴</u>	<u>Np²³⁹</u>	<u>Cu⁶⁴</u>	<u>As⁷⁶</u>	<u>Ga⁷²</u>	<u>Zn⁶⁵</u>	<u>P³²</u>
NOVEMBER 14, 1956	21	47	24	56	42		58	51
NOVEMBER 15, 1956	0	39	17	49	54		19	57
NOVEMBER 16, 1956	25	54	30	60	32		67	52
MARCH 7, 1957	10	50	19	60	36		24	0
MARCH 8, 1957	26	47	46		30		21	11
MARCH 9, 1957	29	61	37	52	45		36	13
JANUARY 21, 1958	8	51	29	59	25	6	28	37
OCTOBER 9, 1958	<u>3</u>	<u>49</u>	<u>28</u>	<u>62</u>	<u>50</u>	<u>—</u>	<u>59</u>	<u>36</u>
AVERAGE (With Standard Deviation)	15 ^{±11}	50 ^{±6}	29 ^{±9}	57 ^{±5}	39 ^{±10}	6	39 ^{±19}	32 ^{±21}

Figure 5

*DEPLETION OF CERTAIN RADIOSOTOPES
FROM THE COLUMBIA RIVER BETWEEN
ARLINGTON AND VANCOUVER, WASHINGTON*

<u>RADIOISOTOPE</u>	<u>HALF LIFE</u> (DAYS)	<u>DEPLETION</u> (PER CENT)
Cr ⁵¹	27	21
Np ²³⁹	2.33	< 25
p ³²	14.5	22
Zn ⁶⁵	245	48

I chose Arlington because it is below McNary Dam and Vancouver is about at the upper end of the estuary of the Columbia River. It is almost the lowest point that you can sample and feel that you are sampling the Columbia River without effect of the ocean.

Figure 6 shows that we have filtered the samples with a millipore filter which should take out about everything above 0.5 microns. We analyzed what was left on the filter and what's in the filtrate and summed them to get the information. Figure 6 shows what we got on the filter. If the materials were on the filter as suspended sediments and flowing down the river as suspended sediments perhaps this indicates the radioisotopes did adsorb on the larger sediments which were lost to the river and that is perhaps where our loss came from. To compare this, the chromium on the filterable solids was about 3%, and we lost about 15% on the way there as was shown in Figure 3. Only about 1% of the sodium was on the filterable solids and yet we lose about 50% to Pasco; neptunium shows on solids 5% as compared to 29% lost, and so forth. This led us to believe that about the only place the material is going is to sediments which are no longer available for sampling at least by the methods we were using at Pasco.

Figure 7 shows the radioisotopes in McNary Reservoir sediments. We tried to sample the bottom sediments at many locations on the river, but the Columbia River is a fast flowing river and we damaged many sampling devices trying to get sediments. There just isn't much, except in the slack water portions. The first major slack water segment below Hanford is the McNary Dam Reservoir. In the McNary Dam Reservoir, using a coring device, we were able to get cores between 2 and 3 feet long, that is the radioactivity extends in depth 2 or 3 feet in the cores. This represents some 8 or 9 years because the reservoir was not put into operation until 1954, and I presume before that the bottom of that stretch was scoured like it is now in the rest of the streams. Taking a series of cores and measuring the radioisotopes present in them we then calculated how many curies there were per square mile of these radioisotopes in the sediments. If radioisotopes are lost by this mechanism to the bottom then they represent a reservoir of radionuclides which can by other processes get back into circulation and they might be important from the radiological point of view. For instance, the reservoir may be scoured during times of high water flow. We have not seen much evidence of this and we are making an effort to get some of this information next year. All of the radioisotopes shown in Figure 7 with the exception of cesium and cobalt have short half-lives, at least they are short with respect to the length of the time that we have been collecting the sediments. Since the reactors have been putting out these radioisotopes at a reasonably constant rate, the values given (I left off the P^{32} value which is 0.5) represent equilibrium values. They will maintain that value as long as conditions stay the same; they will not build up, except for cobalt-60 which is roughly three-quarters of the way to equilibrium and cesium of course which is quite a way from equilibrium.

Figure 8 shows typical data from an 18" core. The core was sectioned by taking off the top inch and the second inch and thereafter by 2" sections. The cores were counted for several radioisotopes but I have shown only cobalt-60

Figure 6

**PERCENTAGE RADIOISOTOPES ON FILTERABLE
SOLIDS COMPARED WITH AVERAGE DEPLETION**

SAMPLE	RADIOISOTOPE						
	<u>Cr⁵¹</u>	<u>Na²⁴</u>	<u>Np²³⁹</u>	<u>Cu⁶⁴</u>	<u>As⁷⁶</u>	<u>Zn⁶⁵</u>	<u>p³²</u>
JANUARY 27, 1957	3.9		6.5	59		62	86
NOVEMBER 10, 1958	3.3	0.9	1.4	15	3.6	67	43
OCTOBER 21, 1959	3.3	0.6	1.4	15	3.7		
FEBRUARY 16, 1962	3.3	1.7	8.0	16	4.7	31	
FEBRUARY 17, 1962	<u>3.4</u>	<u>1.2</u>	<u>7.1</u>	<u>22</u>	<u>6.4</u>	<u>33</u>	<u> </u>
AVERAGE (With Standard Deviation)	3.4 ^{±.3}	1.1 ^{±.3}	4.9 ^{±3}	25.4 ^{±19}	4.6 ^{±1.3}	48.2 ^{±18}	64.5 ^{±30}
DEPLETION	15	50	29	57	39	39	32

Figure 7

*RADIOISOTOPES IN
MCNARY RESERVOIR SEDIMENTS*

<u>RADIOISOTOPE</u>	<u>HALF-LIFE</u>	<u>CURIES PER SQUARE MILE</u>
Zn ⁶⁵	245 d	230
Cr ⁵¹	27 d	62
Co ⁶⁰	5.2 y	10
Sc ⁴⁶	85 d	9
Mn ⁵⁴	300 d	6
Cs ¹³⁷	30 y	4
p ³²	14.5 d	

Zn^{65} AND Co^{60} IN TYPICAL SEDIMENT SAMPLE

Figure 8

and zinc-65. Note that the 0 should be 10 on the log plot. Since near the bottom of the core the radioisotope concentration is about 1 d/m/g it is sometimes necessary to take a large number of cores and composite them and then employ the lowest level techniques we have to count them which involves large sodium-iodide crystals. In most of the cores we wanted to point out that there is a place where activity appears to increase with depth instead of decreasing with depth. If you plot the cobalt-zinc ratio you find a break. What we wanted to do was use the ratio of the different radioisotopes to determine how old the sediments were with depth. We have not released cobalt-60 and zinc-65 at the same rates in the time that we are discussing. However, we do have values for the ratio and therefore we can calculate what the age should be. The reason that there is a break is because the ratio between the two change markedly in one year. I think that year was 1958 and so it provides a marker that at this particular depth occurred in 1958. Where we can no longer detect sediments we assume that it was 1954. So, in a sense, we do have a dating mechanism for this as we go down in depth.

Using the information in Figure 8 I have plotted the two sediment cores that we picked. This one is 22" long and the deepest section was roughly 5 years old. This Figure 9 itself is a year or so old.

Often when one thinks of radionuclides in the stream absorbing on sediments you think of them as being in solution, ions absorbing on sediments, but this is not necessarily true. How did we introduce them to the stream? When it came out of the reactors the P^{32} was a few percent particulate and that is what we spiked the river with. Later in the river, some 30 miles downstream, 40% of the P^{32} was on filterable material-on solids. So you actually see that P^{32} is going to the solids. Scandium-46, largely particulate when we released it, perhaps has done nothing but flow downstream as a particulate. Copper was largely particulate when introduced. One of our aims at first was to determine the chemical forms of these materials so we could study the uptake. It turns out that so many of these are present in such large quantities in a physical form that it means that if it is present on particulate and if it is going to stay on that particulate it does not make any difference whether it is arsenate or arsenite or what chemical form because it is probably particulate and will move downstream.

We have taken a sample of McNary sediment and have tried to fractionate it by particle size; the fractions are not clean; they are somewhat rough fractionations. Nevertheless, what they give is from a small fraction of up to 5 microns, the second fraction goes up to 20, then to 40, then to 60 and finally to 90 microns. We found that there is more radioactivity, more radionuclides, on the fine fractions per gram than there are on the larger fractions per gram. This measurement was actually made on zinc and cobalt radionuclides. It was quite apparent that there was organic material present so we made a separation of organic to inorganic material. I am not proud of the separation; we actually did it with carbon tetrachloride. We have tried the sucrose solutions that Oak Ridge is using; I should say we are still looking for the best material. At least it shows that the organic material contains most of the radioactivity on this larger fraction. We have not

**SEDIMENT AGES AS DETERMINED
BY THE Zn^{65} / Co^{60} DATING METHOD**

Figure 9

carried this study to other fractions so I do not know whether this is due largely to organic or the inorganic material.

I mention that we did do some chemical form studies, and it may not have been apparent but the isotopes we are mostly interested in this case are anions. I am talking about arsenic, phosphorus, and chromium which are present as anions. Those materials which are in solution we determined by a paper electrophoresis method. In the chemical form we find that phosphorus is present as the ortho-phosphate, the manganese is present as the manganous ion, the arsenic is present as both the arsenite and the arsenate ions, sometimes in different samples there is more arsenate than arsenite, and vice versa, and the chromium is present as chromate. The farther we go downstream more and more of it becomes the chromium-III ion.

I plotted one other curve which I could not make a figure of, and I doubt if it would show up too well on the chart, but I did want to mention it. We have obtained a series of membrane filters with different pore size. We made a chain of these filters, some 8 filters long, with the top filter stopping all particles above 5 microns and the bottom filter had at least a nominal particle size of about 10 millimicrons, or 0.01 microns. According to our measurements it was more like 0.03 microns. After we set up the chains we actually made colloids, gold colloids for instance, so that we could look at them under a microscope and determine the particle size. We then filtered the colloids through the columns. We would find that the material would go through the filters until it came to the one that had smaller pore openings than the particle size and they would be removed on this particular filter. We took some of the sediments, the finer fractions, I assume that which you would call the washload, which were moving with the water, put it on the column and fractionated it. In this particular one we chose a sediment in the water above Hanford which contained no radioactivity. We performed neutron activation on each filter and so far I have only the results for P^{32} . Some 65% of the material which on activation forms P^{32} , (usually phosphorus but sulphur can contribute by the n, p reaction), is on the particles which are 5 microns or larger. The next filter was at 3 microns and it held some 6% more, the next filter was at 1.2 microns and it held 5% more. Then the next 4 filters had nothing on them. 17% of the P^{32} came out on the 0.1 micron filter, then we had 2 filters which also picked up nothing and then 10% of the P^{32} containing the material was found in the filtrate. In these figures that I have showed you prior to this we have used a filter which takes out only about 0.5 microns or larger. The data described above was for four samples. If these four samples are representative then we would say some 30% of the P^{32} parent material would be in solution as dissolved salts if 0.5 μ filter used, whereas actually only 10% was, and the rest was on particles which were too small to be held by the filter. This particular range is about right for some bacteria. I cannot see why we should have a fixed size colloid unless it is bacteria. I could be wrong.

The biologists who are sampling the plankton of the river are using a No. 12 plankton net which effectively removes everything above 5 microns.

The biologists look at these materials under the microscope and identify the various diatoms, and so forth, but when they count the radioactivity they count the whole sample and they call that the activity of the net plankton. This is the same material that we filter and we are calling sediments. The biologists pass this through the millipore filter and this is their nanno-plankton. This again points out the importance of the organic (including biological) material problem.

DISCUSSION

THOMAS: In reference to the absorption of copper on particulates, in so far as chemistry is concerned, is it true for copper if you had put ionic copper in there, that you might still have gotten that much absorption build-up? The particulate that you do release might have dissolved and have been absorbed in that physical state.

NIELSEN: Exactly or these particulates could have settled out and then the ions absorbed on the particles.

AUSTIN: Is it necessary when you filter the small amounts of sediment on the membrane filters to distinguish the activity that may be on the sediments and that which may be taken up by the membrane? Do any ions sorb on the membrane?

NIELSEN: No, they do not. These are cellulose acetate or nitrate membranes and we run many things through them and we find that they do not stick to the membranes.

AUSTIN: Do you rinse the sediments after filtration to wash out the water left in the pores of the deposit and the membrane?

NIELSEN: Yes. I might mention that on this filtering we have found that we can pass a material through a filter and later take the same filtrate and pass it through a filter again and get more out on the filter. But, if you pass it through the filter again immediately you do not get any more out on the filter, but if you wait awhile you get more out, so it's a dynamic thing. More things are going onto particles or condensing to particle sizes that can be filtered. This is why in all our samples we are now filtering right at the site when we take them.

HOOPER: I wondered if you have done anything with the material that passed through all of the filters? We did the filter stunt that you talked about and in addition we took the filtrate and extracted it with an organic solvent to see whether we had any activity. We made a study of this fraction and found some activity.

NIELSEN: We are doing this right now. I am interested to hear that you tried it. We haven't finished it.

HOOPER: In our series of filters, which were comparable to yours, all of our phosphorus came out into the diatom range and not much in the bacteria range of pore size. It came out in the range of rather small diatoms. We were dealing with a small stream and not a large river.

NIELSEN: You actually added P³²?

HOOPER: Yes we spiked it with P³².

NIELSEN: In our case we did not. This is just phosphorus and it may be that whatever contains the phosphorus in the low range will not pick it up if we add P³². This study was done for another purpose and it just happened to fit in with this.

FRIEND: We have always had trouble trying to filter through the millipore filters; it just takes too long. Have you ever tried centrifugation to do this separation?

NIELSEN: If our budget permits we will next year. We like the filter because we are afraid that when we take the time to transport from the river into the laboratory and into the centrifuge there will be many changes we are not interested in. We have purchased pressure filtration apparatus and we carry gas bottles with us and actually force the material through the filters. We have found it best to use two filters; one to take the coarse material off and then try to filter the fines or you would never get the mass through the finer filter. I suppose that's a typical observation.

FRIEND: We have tried to take a split sample of Clinch River water and filter it. Depending on the time it takes to put it through you will find a lot more of your activity retained on the filter. If you take 24 hours to put it through, then take 12 hours to put it through then, you will find quite a bit of difference in the activity of the material retained on the filter.

NIELSEN: Now we're filtering on the rate of say 30 minutes to get everything through but we need large samples, as you can guess; actually we are not spiking the river to a high enough level to do real good studies, so we have to strain to do these analyses. But it appears to me that many measurements that are being made on radioactivity in sediments, where you have just separated the sediments strictly by filtration or centrifugation, without taking into account organic material or biological material. I do not know what the data means or represents and I'm rather loath to do a lot of this until I see. It looks to me like you have to make at least one simple separation of organic and inorganic before you do anything. This bothers me also in the particle size fractionation. Many people interested in sediments can determine the particle size range of the sediments and that's all they need, the curve. But we have to separate it because we want to know the activity on it. So it's one thing to determine it and it's another thing to separate it into fractions. We now have a Coulter counter connected to a multichannel analyzer so it will print out the data very fast of

any fraction we are interested in. So, we feel now that we can seriously process the fractionating of these materials so we get the finer sediments, recognizing, as you pointed out, that the time that it takes and the manipulations that we do on it are going to change it somewhat, but I don't know how else to get at it.

FRIEND: We finally went to a sort of a supercentrifuge; putting 5 gallons of water through a millipore filter is a chore as you probably know.

NIELSEN: Well we are doing 3 gallons routinely. I know it's a chore and I am quite interested in trying this new centrifuge that spins a coil of plastic tubing. Has anyone tried this?

FRIEND: No, we have seen it but have not used it.

HOOPER: Have you tried any dialysis techniques on your fractions from water?

NIELSEN: Actually, we did try once but we got what we felt was precipitation during the course of the dialysis. The water comes in and seems to be clear and seems to be staying in suspension then as soon as we start to manipulate it gets cloudy and we see material that makes us mistrust the sample so we haven't done any more of the dialysis.

BERNARD: In using centrifugation wouldn't you be concerned that the small particles you want to keep separate would coagulate or conglomerate?

NIELSEN: I would; however, I think if you would use the microcope you could see if it had happened; if it had not maybe you would feel safe using that.

KAUFMAN: You still feel that the millipore filter determines size by straining or by simply by giving the opportunity to establish for contact absorption of the particles passing through the membrane. In other words is this a realistic size in the screening sense?

NIELSEN: We have made colloids of different sizes and checked fairly well what their sieves size is.

KAUFMAN: The gold colloid and the ones with which you are dealing in the river might be considerably different in surface properties.

NIELSEN: That's right, and we do measure the zeta potential of these materials. Most of the materials in the river have about a 30 mv zeta potential and we tried to make iron colloids with a negative zeta potential. By the way, if we made them positive they would get stuck on everything and they wouldn't go through anything.

GLOYNA: Did I understand you to say that you had reached equilibrium with the sediment load of the Columbia River at McNary Lake?

NIELSEN: For those short life isotopes as long as our load output remains at the same level and the sedimentation occurs at the same rate by the same process then technically we have.

GLOYNA: If you changed either the reservoir complex or the organic load coming into a stream, do you think you could predict what changes would occur in the sediment liquid loads?

NIELSEN: No, I could not. I will have to plead two things; one, I don't think we have enough data, and then I am a radiochemist. But what leads me to say this is we have taken core samples across the McNary Reservoir from one bank to the other back up to where the Snake River comes in. The Snake River is normally about one-third the size of the Columbia River but it peaks sooner than the Columbia and sometimes its flow is slightly greater than the Columbia for a particular time. When it comes in it's dirty and you can see the difference, you can see on the Snake side of the Columbia River it's brown and on the other side it's nice and clear. So, it seems to me that there should be great differences in the sediments as you go across the river and yet we didn't find that. We didn't even find a factor of two difference in the sediments across the river.

GLOYNA: Perhaps this represents or reflects the separations techniques that you have used.

NIELSEN: Well, we just took a core and counted them. There is no separation here, this is just the sediment taken out and determined how much zinc is in it all across the river. Although I didn't mention it on many of those depletion runs the dissolved solids contents of the Columbia River varied over a factor of 100 and yet the depletion is about the same. I don't understand this.

FRIEND: Did I understand you to say that across the Columbia River below the mouth of the Snake River you find the same zinc concentration?

NIELSEN: Within a factor of two. We actually do see it higher on the Snake River side, maybe one and a half times as high. There may be two factors here. The Snake River does not have the radioactivity in it you see, it's a clean stream and yet it has more sediment and sometimes it's relatively unmixed by the time it gets to the dam. The operators there say they can look back up the lake and see the Snake on one side and although it doesn't have the radioactivity in it it must get it by the exchange with the Columbia River and apparently it doesn't do that too fast. It is one and one-half times at most on the Snake side.

THOMAS: My memory of your annual pattern of flow there is a little hazy but it seems to me that during the time you obtained those samples between from November to January, the river is either in a falling stage or bottoming out. Maybe part of your missing 40% is going by with the first wave of that snow melt in the spring.

NIELSEN: Well, remember for these depletions we measure what we are putting into the river one day and calculate say 24 hours to get at Pasco and then we run down there and 24 hours we withdraw our samples.

THOMAS: And you are still missing 40%?

NIELSEN: Yes.

THOMAS: Now if you had taken the samples when the river was rising or at a higher flow it might well be different.

NIELSEN: That's right. Now many things have determined when we did the depletion studies and there are more we need to do. We did the one in October because the temperature is still high in the river. In the first place when the Columbia River peaks it is something like six times the normal volume so we have one-sixth the activity. So, we are straining for counting again. That particular time is the most difficult time to measure depletions because the concentration is lower.

THOMAS: Yes, but you have a factor of 5 or 10 to work against but it's the 40% you are looking for.

NIELSEN: Right. Well I think we can do it. But, it's getting now so that that may be the only time we can do the depletion studies. The Priest Rapids Dam that I mentioned is controlling the river so markedly that over a daily basis that we don't know the travel time down to Pasco well enough to do them. They are sending waves down the river and we don't know how to determine the flow time or we haven't determined the flow times yet.

PARKER: Have you tried to do a mass balance yet by intergrating the flow times the concentration over a long period of time and getting the depletion in an actual case?

NIELSEN: The river and the reactor effluent water analyses are made by separate groups and not by our group. We are essentially a research group. They do not try to hold to the limits that are necessary to make this reasonable. In other words, 10% may be perfectly adequate for those measurements. We do not have the charter nor the personnel to take that many samples unless that appears to be something that really should be done.

NELSON: To get back to the suspended organic and inorganic materials again and centrifuge separation, TVA has had an on-loan employee with us to develop a technique and he has used a continuous flow centrifuge operating about 30,000 g's to pull out the suspended particulate material then he has taken these particulates and re-suspended them and layered them out on inverse sugar solutions of varying densities and then put them in an ordinary bucket centrifuge and throws them down and the silt very nicely goes to the bottom and the organic matter stays up at the top.

NIELSEN: Is this Lamner's work?

NELSON: Yes.

NIELSEN: We're doing that. That's what I meant when I said we are trying to use gradient densities to separate.

NELSON: Well he's going to develop another chemical gradient. I believe it's a hydrocarbon with a lot of bromine on it and has a density of about 1.6 to 1.8.

NIELSEN: He discussed one that wasn't miscible.

NELSON: Well he has another one now, but I've forgotten exactly what it is.

NIELSEN: His curves are somewhat disturbing because the clay fraction extends up in density as far as the organic fraction and if the so-called organic fraction contains diatoms, say, that is not really too surprising because they would have densities approaching silt.

PARKER: How did you actually take your core samples?

NIELSEN: We bought our core samples from the GM Manufacturing Company but it's one that has a trip and then it drops some 20 feet and puts the core in a plastic tube which we immediately freeze and take back to the laboratory and saw into sections.

KENNEDY: What effect does the freezing have upon the exchangeability of the elements in the core? Do you have any idea?

NIELSEN: We haven't been measuring the exchangeability of these. We have just been sawing them and determining the activity; it may well have some effect. The reason for freezing is to keep the biological and living material from crawling around, and also water from running down the sides of the tube. The water problem isn't very serious because the water contains very little cobalt and zinc and some of these things we are measuring compared to the amount in the sediments.

FOSTER: I might point out that our regular surveillance program on the Columbia River provides some clue to the retardation of isotopes in the sediments based on total inventories. There are some analytical problems to be solved but our regular annual reports include these kinds of numbers as best we can estimate them. Data are obtained at two places within the Hanford reservation, at Pasco, at the Vancouver bridge. These values are based on grab samples taken at intervals of about two weeks. It is not exactly the same water mass that is sampled as it moves down. I can show you some of the numbers from a different kind of sampling method that Dr. Nelson speaks of.

NIELSEN: There is a paper - I think it's by Harris at the Taft Engineering Center - where he calculated from his total beta measurements how much is lost in this stretch of the river. It is not too far off for some of the material.

SORPTION AND RELEASE OF RADIONUCLIDES BY SEDIMENTS

by UEL S. CLANTON

The purpose of this investigation was to evaluate the uptake of radionuclides by sediments in the fluvial and lagoonal environments. The area selected for this investigation was the Guadalupe River drainage basin. This drainage basin was selected because of its freedom from large-scale municipal and industrial waste, its ideal location, and environmental conditions.

A geologic map of Texas shows that the general outcrop pattern parallels the present coast line. The Guadalupe River drainage basin has an aerial extent of about 6,000 square miles. Rainfall in the area averages from 28 inches per year in the headwaters to 34 inches per year near the coast. Generally, the heaviest rainfall periods are October through December and April through June.

The Guadalupe River headwaters in the Comanche Series of the Cretaceous which forms the Edwards Plateau, essentially a high limestone plain but now eroded into rough and broken land.

The Edwards Plateau is bounded to the southeast by the Balcones Fault System which also parallels the coast line. Accompanying this topographic change, the section changes to the Gulf series of the Cretaceous which forms the "Black Belt" of Texas. This area is the most intensely cultivated region of Texas and is a major source of sediment to all streams crossing the area.

The Tertiary crops out in two broad bands, the Eocene and the undivided Pliocene, Miocene, and Oligocene. The band nearest the coast includes the undivided Quaternary and Recent.

The Comanche Series is essentially flat lying while the Gulf series forms a series of lowuestas dipping gently to the southeast. The Tertiary and Quaternary sediments dip only a few feet per mile to the southeast so that a map view gives an exaggerated thickness even to the thinnest units.

Figure 1 is a map of the Guadalupe River drainage basin showing sample locations. The original idea was to collect sediment samples each time the river crossed a major geologic unit. Only after the collection had been completed and most of the samples identified by X-ray diffraction did we realize that the clay sediment load of the Guadalupe River was almost completely soil derived and that almost all traces of the parent clay material had been destroyed

during the weathering processes.

The collected samples were dried, ground, sieved, and then fractionated by centrifugation to obtain the clay-sized fraction of the sediment. Most of the sediment load of the Guadalupe River is sand and silt. Only by fractionating could we obtain a reasonable amount of clay to work with.

Figures 2, 3, and 4 illustrate the material with which we have been working. "Q" indicates quartz, "C" calcite, and "D" dolomite. The various peaks which reflect the clay contribution have been left undesignated. These include the maxima in the 15.5 - 12 Å range, discrete features 10 Å and 7 Å, and the composite clay intensities observed at 4.4 Å and 2.5 Å.

The clays contained in the sediment samples from the Guadalupe River are weathered material which has been transported into the stream system after developing by subaerial weathering of the parent rock in the soil zone. The progressive alteration under the influence of the weathering mechanisms has destroyed the well crystallized clay material and left a residual debris which can be best described as a heterogeneous clay mineral complex which has some expandable layers within the crystallites.

The X-ray diffraction patterns of this material cannot be considered to be characteristic of a single clay group, i. e., kaolinite, illite, chlorite, or montmorillonite, but rather a composite diffraction pattern. The basal reflection which is usually considered to be the most distinguishing feature of a clay group, appears as a low diffuse reflection which usually covers the entire 10 - 15.5 Å spacings with more or less equal intensity. Some of the samples have superimposed on this composite basal reflection a minor feature or features which can be attributed to a 10 Å mica or a 7 Å kaolinite. These minor features also show rather broad diffuse reflections rather than the sharp narrow reflections which are expected from well crystallized material.

The asymmetry of the basal reflection and the generally broad character of the other reflections suggests that this material can be considered as a mixed-layered clay mineral complex of poor crystallinity. The structure is probably best described as randomly stacked degraded clay minerals which are also randomly disposed in the C crystallographic direction.

The percentage of non-clay minerals was determined by comparing the X-ray diffraction intensities of quartz and calcite standards with the X-ray diffraction intensities observed from the sediment samples. A log scale corresponding to a full scale chart deflection of 1500 cps was used to record all X-ray diffraction data.

Figure 5 is a plot of the non-clay mineral percentage determinations. All of the samples contained quartz and calcite in addition to the clay minerals. In addition, the first few samples contained a small amount of dolomite. Calcite is the dominant non-clay constituent in all samples except C-45, with quartz

following in a secondary role. The points plotted on the graph are observed values. The possible variation from the true value is considered to be plus or minus ten per cent of the reported value. Two general trends are apparent. The calcite content decreases toward the coast - the quartz content increases toward the coast.

Ion exchange capacity measurements were determined by the barium saturation technique after the organic material present on the clays had been removed using sodium hypochlorite. This technique for removing organic material has the distinct advantage over other techniques in that it will oxidize organic material in the presence of carbonates and does not degrade the clay structure. A series of barium standards were made in the range of 0-80 meq barium/100 grams clay and compared with the barium saturated sediment samples using X-ray fluorescence.

Figure 6 shows two curves - the lower one is a repeat of the total non-clay mineral content of the previous figure, and the upper curve is the exchange capacity of the fractionated sediment. Considering the material involved, there appears to be a reasonable correlation of non-clay mineral content and ion exchange capacity. Some of the irregularities can be smoothed somewhat when the minor features of the X-ray diffraction patterns are considered -- the 7 Å kaolinite and the 10 Å illite.

Using the data from this graph, it is possible to calculate the average exchange capacity of a clay with properties similar to the one observed for a particular sample. The second line from the bottom on Figure 7 is the calculated average exchange capacity. Values range from 20 to 73 meq barium/100 grams for the heterogeneous clay which is characteristic of the sample.

The top curve of this figure reflects the relative amounts of clay present in a sample based on peak-area calculations for the composite 4.5 Å spacing and for the 001 basal spacings for the various clay minerals. The right half of the top curve fits nicely with the trends shown by the lower curves. The left half of the top curve, however, leaves something to be desired. This perhaps shows best the almost complete loss of crystalline character of the degraded clays which make up the clay-sized sediments being transported in that portion of the Guadalupe River.

Sr^{89} and Cs^{137} sorption studies were made on selected samples in order to determine the effects of mineralogy and exchange capacity on radionuclide uptake. Each sample was divided into two parts, one was treated to remove the organic material present, the other remained untreated. Master solutions containing 5×10^{-10} mg/l Sr^{89} and 5×10^{-7} mg/l Cs^{137} without carrier were prepared. Fifty milligrams (50 mg) of sediment was suspended in 100 ml of the master solution and agitated by a mechanical shaker. After a period of one hour, one day, three days, and seven days, the bottles were centrifuged and one ml. aliquots removed for counting. The technique then determines the amount of radionuclide remaining in solution.

Cs^{137} SORPTION FOR 30 SAMPLES, UNTREATED

<u>1 Hour</u>		<u>7 Days</u>
91.4 %	Average	92.6 %
89.4 %	Minimum	91.3 %
94.5 %	Maximum	95.7 %

Cs^{137} SORPTION FOR 30 SAMPLES,
ORGANIC MATERIAL REMOVED

<u>1 Hour</u>		<u>7 Days</u>
88.2 %	Average	89.9 %
85.0 %	Minimum	85.9 %
91.7 %	Maximum	92.4 %

Figure 8

Sr⁸⁹ SORPTION FOR 25 SAMPLES, UNTREATED

<u>1 Hour</u>		<u>7 Days</u>
52.8 %	Average	55.4 %
46.7 %	Minimum	50.2 %
58.0 %	Maximum	61.6 %

Sr⁸⁹ SORPTION FOR 25 SAMPLES,
ORGANIC MATERIAL REMOVED

<u>1 Hour</u>		<u>7 Days</u>
42.8 %	Average	45.9 %
34.0 %	Minimum	36.1 %
48.1 %	Maximum	54.1 %

Figure 9

Figure 8 shows in summary form the average Cs^{137} sorption for samples with organic material removed and untreated samples. The data indicate:

- 1) Sorption is highest in untreated samples. 91 - 93% untreated vs 88 - 90% treated
- 2) Sorption gradually increases during the seven day period.
- 3) The greatest rate of sorption occurs during the first few minutes of contact.
- 4) The organic material sorbed on the clay surface increases the overall sorption capacity of the sediments.

Figure 9 shows similar data for Sr^{89} . The most noticeable difference is the lower sorption values obtained with this cation. Untreated samples removed on the average of 53 - 55% of the Sr^{89} while the treated samples removed 42 - 46% of the Sr^{89} .

At these levels of radionuclide concentration, there appears to be no correlation with radionuclide sorption and exchange capacity determinations. Another mechanism as yet undetermined and presently poorly understood must be envisioned to explain much of the apparent anomalous data relating radionuclide sorption and ion exchange capacity measurements.

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SORPTION AND RELEASE OF RADIONUCLIDES BY SEDIMENTS

by TOM D. REYNOLDS

The dispersion of a particular radionuclide in a stream may be expressed in a simplified form by the following radioactivity budget:

$$R + r_n = [r_w + r_s + r_o + r_{od} + r_d] e^{\lambda t}$$

where:

R = radioactivity discharged into the stream at time $t = 0$

r_n = background radioactivity in the stream at time $t = 0$

r_w = radioactivity in solution in the water at time t

r_s = radioactivity sorbed by stream sediments at time t

r_o = radioactivity sorbed by biological systems in the stream at time t

r_{od} = radioactivity sorbed by organic debris at time t

r_d = radioactivity discharged from the watershed at time t , i. e.,

$$\int_0^t r(t) e^{\lambda t} \text{ where } r(t) \text{ is the radioactivity discharged from the watershed as a function of time}$$

The complexity of stream transport can be recognized immediately since many of the variables in the equation are not entirely independent of one another. For instance, the amount of activity sorbed by the sediments will be dependent upon the activity in solution. Also, it is dependent upon the sediment volume, the amount of sediment in suspension and other factors which are related to hydraulic and hydrological considerations. At the present, data and knowledge are inadequate for evaluating the use of the radioactive budget for a particular stream; however, the budget does illustrate the factors involved and may be

possibly used once more data are available.

One phase of the transport project at the University has been devoted to stream sediment studies in order to learn more of their role in the uptake phenomena. The water bodies selected for study as shown in Figure 1 were the Guadalupe River and its receiving bodies of water, Guadalupe Bay and San Antonio Bay; Lake Austin on the Colorado River and Lake Falcon on the Rio Grande. The greater portion of the radioactivity found in the watersheds comes from fallout and natural activity present in the watersheds since there are not any appreciable releases of radionuclides in this area.

Samples collected included bottom sediments, suspended sediments, and water samples, most of which were collected in the spring of 1962. The bottom samples were collected either by an Ekman dredge, a Peterson dredge or a tube type bottom sampler, which, when working properly, collected a 2 1/4 inch diameter sample in a plexiglass tube. The Ekman dredge was found to be the most versatile sampler and was useful in muddy and silty samples. The tube-type sampler was only effective in clayey materials since silty and coarse-grained materials fell out of the tube while it was being hoisted. Samples collected by the Ekman dredge were usually the top 10 or so inches of material, whereas, some of the tube samples contained as much as 21 inches of material.

The investigation included: (1) sorption and release studies using the isotopes Cs^{137} , Sr^{89} , Ru^{103} , Cr^{51} , Fe^{59} , and I^{131} ; and (2) gross beta counting, Sr^{90} , and radiocesium analyses to determine activity levels existing presently in the water bodies. All sediment samples were sieved and separated into the fractions less than 140 mesh and greater than 140 mesh. The fraction less than 140 mesh closely represents the material that would be in suspension during high flow conditions since it includes the size ranges for the clays, silts, and very fine sands.

The sorption and release studies employed slurry techniques usually using a sediment to water concentration of 500 mg/l. Other concentrations were sometimes used; however, concentrations were always kept within a range that would appear as feasible to exist in actual stream conditions. In the slurry technique, master solutions were made of known counts, then 100 ml portions were placed in 4 oz. bottles and known amounts of sediment added. The bottles were shaken and after a time duration of one hour, one, three, and seven days the bottles were centrifuged and one ml aliquots taken for counting.

Initial studies investigated certain factors affecting Sr^{89} and Cs^{137} sorption such as sediment concentration, carrier concentration, competing cation concentration, pH, particle size and time.

As the sediment concentration increased, the per cent of Cs^{137} activity sorbed by the sediment increased as indicated in Figure 2. The Figure shows the plots of the values from the tests and the average curve for these values. Also shown is a curve constructed from the average of the distribution

LAKE AUSTIN WATER,
SEDIMENT SAMPLE 1-a

Figure 2 **SEVEN DAY Cs^{137} SORPTION VERSUS
SEDIMENT CONCENTRATION.**

coefficients or K_d values from the tests. The K_d value being:

$$K_d = \frac{fc/m}{fs/v}$$

Where:

fc = fraction of activity sorbed by the sediment

fs = fraction of activity in solution

m = mass of sediment

v = volume of solution

The K_d value is derived from the mass action equilibrium equation with the assumption that the exchanged ion is in very low concentrations, which is the case for many radionuclides in streams. Since the curve from experimentation closely approaches the curve derived from the average K_d value, it shows that the distribution coefficient fairly accurately represents the distribution of Cs^{137} activity between the suspended sediment and the water.

The sorption of Sr^{89} was found to increase as the sediment concentration increased, as shown in Figure 3, and the distribution coefficient, K_d , was found to give a fair representation of the distribution of the activity between the sediment and the water.

Cs^{137} sorption was found to be relatively insensitive to the pH of water, whereas, Sr^{89} sorption was very sensitive, see Figure 4, particularly in the pH range of natural waters. The effect of pH is of consideration in southwestern streams since high pH variations are known to occur. pH values in Texas streams may range from pH 5.5 to pH 10.

The K_d values for the size fractions less than 140, 200, 270 and 325 mesh did not vary appreciably. Even the material collected by decantation of suspended material in water showed only a slightly greater sorption than the fraction less than 140 mesh.

Cs^{137} and Sr^{89} sorption was found to be slightly time dependent since the seven day sorption was slightly greater than the one hour sorption.

The sediments from the entire length of the Guadalupe River did not vary appreciably in their mineralogical composition although the river begins in a predominately limestone region and traverses through sandy and clayey formations on its way to the Gulf. The sediments from the entire river length did not vary appreciably in their percentile sorption and K_d values when tested in portions of the same water sample. The most significant factor affecting the sorption of Cs^{137} was the concentration of competing cations. The monovalent

Figure 3 SEVEN DAY Sr^{89} SORPTION VERSUS
SEDIMENT CONCENTRATION.

DISTILLED WATER, SEDIMENT SAMPLE 1-a,
SEDIMENT CONCENTRATION = 500 mg/l

SEVEN DAY Cs^{137}
SORPTION VS. pH.

SEVEN DAY Sr^{89}
SORPTION VS pH.

Figure 4

cations, in particular K^+ , were most effective in inhibiting Cs^{137} sorption as shown by Figure 5. Both competing cation concentration and pH were found to be significant in Sr^{89} sorption. The divalent cations indicated by Figure 6 were most effective in inhibiting Sr^{89} sorption.

The cation concentration of the Guadalupe River water increases only slightly as the water proceeds downstream; however, an appreciable change in competing cation concentration does occur at the mouth of the river where the river water diffuses out into the Guadalupe and San Antonio Bays. See Figure 7. There, within a length extending from several miles above the mouth of the river to the southern end of Guadalupe Bay, the dissolved solids content increases from several hundred parts per million to approximately 30,000 ppm. The sorption of Cs^{137} was found to decrease greatly as the salinity of the water increased. See Figure 8. The sorption in fresh water at a sediment concentration of 500 mg/l was about 92 per cent, whereas, in salt water of 30,000 mg/l salinity the sorption was reduced to about 7 per cent. Strontium-89 sorption in salt water was also found to be less than in fresh water; however, the difference was not as great as for Cs^{137} . The sorption of Sr^{89} in fresh water was about 9 per cent compared to 0.2 per cent in salt water of 16,530 mg/l salinity.

Since sorption is principally an ion exchange process in which mass action and reversibility hold true, it was expected that sediments which had sorbed activity in a fresh water environment would release a portion of the sorbed activity when transferred to salt water because of the increase in competing cation concentration. It was found that about 44 per cent of the sorbed Cs^{137} activity was released when such transfers were made into bay water of 16,530 mg/l salinity as shown by Figure 9. An increase in salinity resulted in an increase in release of sorbed Cs^{137} activity which is indicated in Figure 10.

Similarly, a portion of sorbed Sr^{89} activity was released when sediment which had sorbed the activity in fresh water was transferred to bay water. See Figure 11. In the case of Sr^{89} about 43 per cent of the sorbed activity was released. The release was due to both the lower pH of the bay water and the increase in competing cation concentration.

Radiocesium and Sr^{90} radiochemical analyses were performed on the sediment samples taken from the length of the Guadalupe River and from the western side and center of San Antonio Bay. The Guadalupe River discharges into Guadalupe Bay, a small narrow body of water north of San Antonio Bay, and salinity measurements have shown that the river water passes through Guadalupe Bay and diffuses out into San Antonio Bay along its western side. Consequently, samples were taken from the western side of the bay in addition to the center of the bay.

Figure 5 **SEVEN DAY Cs¹³⁷ SORPTION VERSUS
COMPETING CATION CONCENTRATION.**

DISTILLED WATER, SEDIMENT SAMPLE 35,
SEDIMENT CONCENTRATION = 500 mg/l

Figure 6 SEVEN DAY Sr^{89} SORPTION VERSUS
COMPETING CATION CONCENTRATION.

Figure 7

SAN ANTONIO BAY AND VICINITY

GUADALUPE RIVER WATER, SAN ANTONIO BAY WATER,
SEDIMENT SAMPLE 35,
SEDIMENT CONCENTRATION = 500 mg/l

SEVEN DAY Cs^{137} SORPTION VERSUS SALINITY.

Figure 8

GUADALUPE RIVER WATER, SAN ANTONIO BAY WATER,
(SALINITY = 16,530 mg/l), SEDIMENT SAMPLE 35,
SEDIMENT CONCENTRATION = 500 mg/l

**SORPTION OF Cs^{137} IN FRESH WATER
AND ITS RELEASE IN SALT WATER.**

Figure 9

GUADALUPE RIVER WATER, SAN ANTONIO BAY WATER,
SEDIMENT SAMPLE 35,
SEDIMENT CONCENTRATION = 500 mg/l

Figure 10 SEVEN DAY RELEASE OF SORBED Cs^{137}
VERSUS SALINITY.

GUADALUPE RIVER WATER, SAN ANTONIO BAY WATER
(SALINITY = 16,530 mg/l), SEDIMENT SAMPLE 35
SEDIMENT CONCENTRATION = 500 mg/l

Figure 11

**SORPTION OF Sr^{89} IN FRESH WATER
AND ITS RELEASE IN SALT WATER.**

PROFILE OF RADIOCESIUM ACTIVITY IN THE GUADALUPE RIVER AND SAN ANTONIO BAY.

Figure 12

PROFILE OF STRONTIUM-90 ACTIVITY IN THE
GUADALUPE RIVER AND SAN ANTONIO BAY.

Figure 13

In general, the river sediments were found to have a higher radiocesium activity than the bay samples as shown in Figure 12. The average activity in the river sediments was $1.69 \mu\mu\text{c/gm}$ which was 48 per cent greater than the average activity of $1.14 \mu\mu\text{c/gm}$ found in the bay sediments. The cesium activity in the water itself was found to be greater in the bay waters than the river waters.

Similarly, the Sr^{90} activity in the river sediments was found to be greater than in the bay sediments. See Figure 13. The average activity in the river sediments was $0.15 \mu\mu\text{c/gm}$, which was 275 per cent greater than the average activity of $0.04 \mu\mu\text{c/gm}$ in the bay sediments. The Sr^{90} activity in the bay waters was found to be generally higher than in the river waters.

The lower activities in the bay sediments may probably be attributed to one or both of two causes. First, a portion of the sorbed activity in the river-borne sediments is released once they are deposited in marine water. Second, the original bay sediments sorb less activity than river sediments. The sorption and release studies performed in the laboratory showed that these two causes could exist either singly or jointly, and that both are due to the effects of competing cations.

Some contributions produced by this research effort are:

- 1) It gives information on radionuclide uptake by typical southwestern streams.
- 2) It adds knowledge to the release of radionuclides by riverborne sediments that are deposited in marine waters.

DISCUSSION

ODUM: When samples reach San Antonio Bay, do you visualize a displacement of major cations?

CLANTON: Yes. Some of the cations which have been stripped from the day lattice during weathering, for example potassium, are abundant in sea water. When the degraded clay particles are transported into the marine environment these ions again become available and are reconstituted as part of the crystal lattice.

ODUM: Do you think it has anything to do with the changes that take place in the clays in their process of going to the sea and deposition there?

CLANTON: Whitehouse at Texas A and M has made electron micrographs of well defined clay samples which have been placed in sea water and left to react for several years. These are laboratory studies so that most of the variables are known. Progressive electron micrographs show that in time the original clay morphology is modified by additional crystal growth.

ODUM: Do you think that magnesium and potassium are particularly involved in this particular immediate action when it first gets to the Bay or do you think its just a general action of some cations?

CLANTON: Electron micrographs by Whitehouse show the development of brucite crystallites in the interlayer position of the expanding clay minerals. In the original reaction then, magnesium would be important in the development of the brucite ($\text{Mg}(\text{OH})_2$) structure. Magnesium is also being supplied with the sediment from the dolomites in the Edwards Plateau region.

PARKER: Did you get any agglomeration of clays when they entered the marine environment?

CLANTON: The clays will flocculate in a marine environment.

THOMAS: From an engineering viewpoint one of the questions that I would like to ask is about the major station near the downstream end of the test reaches. Did you make an estimate of all the activity that goes by in a year; could one compute the proportion that is absorbed relative to the

proportion that is not? This is an important engineering question, and though it may not be essential for your purpose, would your data permit you to make such a calculation?

REYNOLDS: Our sampling was only on one occasion; as to what would occur throughout a year we don't have enough data on this.

THOMAS: You might be able to put it in though.

REYNOLDS: Yes, we could make an estimate of the amount of radiocesium and radiostrontium that would be discharged from the watershed during a year and the proportion of the total activity that would be associated with the sediments and the water. Also, an estimate could be made of the amount of radioactivity released by the sediments when the sediments are discharged into San Antonio Bay. In this respect it would be valuable to do a radioactivity budget study on the watershed over a period of time, for instance a year or so.

CLANTON: One thing that perhaps adds some confusion to a generality here is the drainage area involved. The upper half of the Guadalupe River drainage basin is in the Edwards Plateau. Because of faulting and the natural drainage profile, this portion of the Guadalupe River has a much higher stream gradient than is observed in the lower portion of the drainage basin. Also there is a three to five hundred foot difference in topographic relief at the Balcones Fault Zone which borders the Edwards Plateau. The soil developed on the limestone of the Edwards Plateau is thin. At certain times during the year there may be five inches or more of rainfall in a short period of time. During a period of brief but extensive rainfall there would be considerable run-off because of the topography and the thin soil cover. In such a situation there is extensive transport of the sediments in the stream bed. Any radionuclides sorbed by the sediments would be picked up and flushed down the river, eventually into the marine environment.

THOMAS: That's a point of complication, but it's also a very important point, too.

KENNEDY: I was wondering about the variation in mineralogy with increased size. There is some evidence that as you get into larger sized particles the variation in mineralogy in a stream system like this may greatly increase. Do you have any evidence for this?

CLANTON: When we started this program we were using some of the published data from Oak Ridge as a pattern. We thought that we too could use the less than 140 mesh material for our experiments. However, with this size fractionation there was not enough clay to work with. Even after fractionating to obtain a sediment size of about 10 microns only half of the sample is made up of clay minerals. Some of the samples were fractionated to obtain a greater than 0.5 micron size and a less than 0.5 micron size separation. X-ray diffraction patterns of the less than 0.5 micron still showed the presence of

calcite and quartz. Since the x-ray diffraction patterns were so similar to the x-ray patterns which were shown in Figures 2, 3 and 4, the effort expended in the second fractionation did not seem to justify the slight increase in data quality.

KENNEDY: Well, I was really interested in the variation in mineralogy of the larger sizes and not the variation in mineralogy of clay mixed with the larger sizes. I was wondering if you had done any work on this.

CLANTON: No we did not. Originally we started looking at all material which was less than 140 mesh in size. When we found that the relative clay percentage was so low, the separation technique was changed so that the finer particles were selectively concentrated. No attempt was made toward identifying the mineral composition of the larger particles and comparing these.

GLOYNA: We can make a pretty fair approximation of the concentration of fallout (radioactive) in San Antonio Bay.

ODUM: You proved it wasn't in the sediments so where else can it be? That's the question.

GLOYNA: Well, I think we know how much is going into the fish and this sort of thing.

FRIEND: From the bottom sediments you mean?

GLOYNA: No, probably from the water.

ODUM: It would be good to try.

GLOYNA: Haven't you done this experiment for strontium?

ODUM: Not for this particular Bay, no. We have done it for a grass flats. We take a system where the light comes to the bottom and the bottom has a very heavy vegetation, whereas in the San Antonio Bay you get the plankton plus oyster least in the upper end.

GLOYNA: From the upper part of San Antonio Bay? At any rate I think we can make an estimate.

ODUM: Were you able to eliminate the possibility that it was a direct action of salt exchange rather than the clays going through the animal stomachs and coming out modified?

GLOYNA: Our field experiments were repeated in the laboratory under controlled studies.

REYNOLDS: We used both clays from the river and the Bay and they both

showed the same tendency in uptake of the radionuclides. I think the same mineralogical identification of the material in the Bay showed it was the same material as in the river.

CLANTON: As you proceed down the river the closer you approach San Antonio Bay the better crystallized the clay appears. Part of the problem is lithology; the Edwards Plateau is composed of dolomite, limestone and marl which really don't furnish much clay material. The material that is furnished is badly weathered and degraded.

The Tertiary section near the Gulf Coast is mostly sand and shale. Transport time and distance to the Gulf is relatively short compared to the Plateau derived sediments. A combination of factors then is responsible for this apparent increased crystallinity as one approaches the marine environment.

The pattern that is obtained by the x-ray diffraction technique is dependent on the repeat sequence of the crystal structure. When the units are repeated one gets diffraction patterns which are characteristic of a particular mineral species. But in the material which we describe the individual layers are poorly crystalline and randomly stacked units from the several clay mineral species. The x-ray diffraction pattern of this material is not characteristic of a single clay mineral species but rather a poorly crystalline heterogeneous clay mineral complex which has spacings covering the entire 10 to 15.5 Å range.

NATURE AND DISPOSITION OF SEDIMENT SORBED RADIONUCLIDES ENTERING SAN FRANCISCO BAY

by WARREN J. KAUFMAN and PETER C. KLINGERMAN

The transport of radionuclides through estuaries poses diffusional as well as chemical and biological problems of considerable complexity. Radiocontaminants may enter estuarine waters by the contributions of rivers containing fallout, natural radio-elements, or wastes from nuclear operations. With the increasing number of nuclear-powered ships, concern has been expressed for the disposition of fission products released in reactor start-up or in the event of an accident involving such vessels. Transport of water miscible contaminants from an estuary is dependent on displacement by fresh water input at the estuary head and by longitudinal eddy diffusion bringing about fresh-salt water exchange. However, in the event the radiocontaminants become associated with the estuarine biota or sediments, the rate of transport to the sea may be greatly reduced. It is to this aspect of sea disposal of radioactive wastes that the remainder of this discussion will be devoted.

STUDIES OF SEDIMENT-SORBED FISSION PRODUCT TRANSPORT IN SAN FRANCISCO BAY

Studies of sediment transport in the San Pablo Bay portion of San Francisco Bay system have been in progress since 1957 by the Sanitary and Hydraulic Engineering Laboratories of the University of California, Berkeley (1). The investigation had the broad objective of determining the mechanisms of sediment re-suspension and transport within estuaries. Of specific interest was the improvement of dredging operations conducted by the Corps of Engineers and the Mare Island Naval Shipyard to maintain shipping channels and dock areas. As an aid to the investigation sediment labelled with gold-198 was used under various circumstances and traced along the bottom by a sled-mounted underwater scintillation detector. As only limited amounts of radioactive tracer could be used, frequent background surveys of the bottom sediments were found to be necessary. These surveys suggested that a correlation might exist between bottom background variations and the seasonal discharge of the Sacramento River, especially during the spring months following periods of weapons testing. Furthermore, gamma radiation spectral analyses of bottom samples indicated that the radioactivity increments accumulated during heavy runoff were probably of recent fission origin.

The study was continued in the late winter and spring of 1962 with the support of the California State Water Pollution Control Board.

During this latter phase of the study special emphasis was given to the amount and nature of radioactive material transported into San Pablo Bay by the Sacramento River and to the disposition of this material within the Bay. It was thought that the annual sediment load might be sufficiently labelled with fall-out from the Russian 1961 tests to examine sediment transport within the entire Bay rather than only in a single segment as had been previously possible with artificially applied tracers. The objectives were two-fold; to investigate the transport of a natural waste product, suspended sediment, through an estuarine system and to determine the nature and movement of sediment-sorbed fission products in such a system. A secondary objective was to investigate the radioecology and lithography of benthic deposits as they relate to sediment-sorbed radionuclides.

Bottom sampling stations and traverse lines were established as shown in Figure 1. These stations were selected to provide an index of changes in bottom radioactivity and were not intended to serve to measure the total fission products deposition in the Bay. Periodic cruises were made beginning on January 14, 1962, with the most recent on May 29, 1962. In situ bottom measurements were made at individual stations and along traverse lines by utilizing the sled-mounted scintillation probs. Bottom core samples were also taken at each station and a surface layer one centimeter in thickness assayed for total beta, total gamma and for the gamma spectrum.

RADIOACTIVITY INPUT TO THE SAN FRANCISCO BAY SYSTEM

In order to estimate the nature and amount of radioactive materials entering the Bay system a sampling station was established on the Sacramento River at Rio Vista, approximately 40 miles east of Station TR-1 in San Pablo Bay. Due to tidal effects it was not possible to gauge the river discharge at Rio Vista, but rather at the city of Sacramento, some 30 miles further upstream. The discharge hydrograph for the river at Sacramento, together with the concentrations of beta and gamma radioactivity are shown in Figure 2.

The water samples analyzed were composited from 15 portions taken at three depths along 5 vertical sections. Samples of about 30 liters were flocculated with alum and the suspended solids deposited on millipore filters. The gamma radioactivity reported is that in excess of 0.10 Mev, while the beta contribution was determined with a thin-window low-background flow type Geiger Mueller Counter. All counting efficiencies were referenced to a cesium¹³⁷ standard and the reported radioactivity concentrations may be considered as "cesium¹³⁷ equivalent" values.

It is not possible to make an accurate calculation of the input radioactivity due to the inadequacy of sampling during late February and early March. However, as the concentration of suspended river sediments was relatively small by March 15, it may be assumed that radioactivity concentrations were probably down to nearly their April value by this date. As a further source of error, the Yolo By-Pass channel carried considerable flow around the Sacramento gauging

FIGURE 2. WATER AND RADIOACTIVITY INPUT TO SAN FRANCISCO BAY FROM SACRAMENTO RIVER, SPRING 1962

FIGURE 3. GAMMA RAY SPECTRUM AND DISSOLVED SOLIDS IN SACRAMENTO RIVER WATER, RIO VISTA, CALIFORNIA, FEBRUARY 17, 1962

station and this discharge does not appear in Figure 2. These flows exceeded 1,000 cfs from February 10 to 27, exceeded 10,000 cfs from February 12 to 22, and reached a peak of 69,800 cfs on February 17. The By-Pass flow dropped to below 1,000 cfs on February 27. From the limited data available it is estimated that about 500 curies of sediment-sorbed radioisotopes, largely of fission origin, entered San Pablo and Suisun Bays during the month of February. As shown in Figure 2, the dissolved radioactive material was far exceeded by the sorbed material.

The distribution of suspended load and radioactive material with river depth is shown in Table I. Surface samples were taken at a depth of five feet, while bottom samples were taken at three feet above the bottom. It is noteworthy that although the peak discharge occurred on February 17, the concentration and specific activity ($\mu\mu\text{c}/\text{gram}$) of the sediment-sorbed radioactivity was appreciably greater on February 11, i. e. during the rising limb of the runoff hydrograph, than on the 17th. As might be expected, the near surface suspended solids concentration was less than that at the bottom but the specific activity was greater.

TABLE I
SUSPENDED SOLIDS DISTRIBUTION OF SORBED RADIOACTIVITY
IN THE SACRAMENTO RIVER. FEBRUARY 1962

Depth	Susp. Solids Conc. mg/l	Sorbed Beta Radioactivity*	
		$\mu\mu\text{c/liter}$	$\mu\mu\text{c/gram}$
<u>Feb. 11:</u>			
Surface	347	139	399
Mid-depth	462	146	317
Bottom	607	203	319
<u>Feb. 17:</u>			
Surface	591	117	198
Mid-depth	713	150	211
Bottom	761	161	211

* Cs^{137} equivalent radioactivity

The characteristics of the sediment-sorbed and dissolved radioactivity are illustrated by the gamma radiation spectra shown in Figure 3. It is again

evident that a major fraction of the radioactive materials were associated with the suspended solids. For the sample shown in Figure 3 the total counting rate per liter dropped from 137 counts per minute on March 12 to 42 counts per minute on June 8, indicating the short half-life and recent origin of the sorbed radioactivity.

DISTRIBUTION OF SEDIMENT-SORBED RADIOISOTOPES IN SAN PABLO BAY

As noted earlier, the space and temporal distribution of deposited sediment-sorbed radioactivity were determined by bottom sampling and traversing with a sled-mounted scintillation detector. As illustrative of the field measurements data, a 2.5 mile traverse is shown in Figure 4 for an area north of the main shipping channel. The January 14 traverse data are typical of the pre-runoff situation, while the April and May data are typical of the period following the heavy February runoff. A major portion of the radioactivity labelled sediment was within one-half and one mile of the shipping channel on April 13. However, by May 29 active deposits were found immediately adjacent to the channel. From earlier studies it is believed that this material resulted from the re-suspension of previously deposited sediment and that the area north and within 0.5 miles of the channel was prone to late spring and summer shoaling.

The results of the analyses of core samples at four stations in San Pablo Bay are shown in Table II. A composite sample of the top one centimeter prepared from four to eight three-inch diameter cores was analyzed in each instance. Analyses were completed within one week of sampling.

TABLE II
DISTRIBUTION OF SEDIMENT-SORBED RADIOACTIVITY
IN SAN PABLO BAY

January - May 1962

Sampling Date	Sediment-Sorbed Beta Radioactivity*			
	$\mu\text{mc/gram}$			
	Sta. TR-2	Sta. TR-3	Sta. TR-4	Sta. SB-2
Feb. 8	56	59	63	50
Feb. 15	67	62	60	46
Mar. 15	73	127	127	87
April 12	73	67	136	150

*Cs¹³⁷ equivalent radioactivity

FIGURE 4. AREAL AND TEMPORAL VARIATION IN BOTTOM RADIOACTIVITY
SAN PABLO BAY, SPRING 1962

FIGURE 5. VARIATION IN NATURE AND AMOUNT OF RADIOACTIVITY WITH TIME
STATION TR-4, SAN PABLO BAY, SPRING 1962

Samples at Station TR-2, located one mile north-east of Station TR-1 (the terminus of the traverse shown in Figure 4), increased by only about 50 per cent from February 8 to March 15 and had returned to pre-runoff levels by May 24. On the other hand, bottom activity at Station TR-4, located three miles north-east of TR-1 and further into a shallow area of deposition, continued to increase until April 12, but also dropped sharply by May 24. A similar delayed build-up was found at Station SB-2 located in shallow water south of the shipping channel. Since it is believed that little suspended solids sorbed radioactivity entered the Bay during March and April, the changes observed at Stations TR-3, TR-4 and SB-2 were probably due to the translocation of previously deposited material. The reduction in radioactivity at any point may be due to a combination of decay and transport. Export of active sediments or the import of non-active material, thus shielding earlier deposited active material, may both serve to reduce the measured surface activity of the bottom sediments.

Gamma radiation spectra of surface sediment samples taken at Station TR-4 on four dates, February 8, February 15, March 15 and April 12, are shown in Figure 5. Since a 20-channel spectrometer rather than a 200-channel instrument was employed for this analysis, the peaks are not as well defined as for the spectrum of suspended river solids (see Figure 3), but it is evident that the sediment radioactivity has its origin in the Sacramento River and that the zirconium⁹⁵ radioisotope was retained on the particulates during transport and deposited in the salt water environment. An analysis of sediment samples at several depths from Station TR-4 on April 12 showed the upper centimeter total gamma radioactivity to be three and four times that in the second and third centimeters of deposited material, respectively.

SUMMARY AND DISCUSSION

Mr. Whipple has presented a general discussion of the problems of radioactive waste discharged into coastal waters and has dealt specifically with certain aspects of dispersion of these wastes. Acting to retard and reverse the processes of dispersion are various biological and chemical sorptive reactions that may concentrate radionuclides. The sorption of trivalent radiocations on suspended river solids, especially the clay minerals, is the first step in the concentration of these radioisotopes in certain estuaries. The suspended colloidal material, which has the greatest tendency to sorb radionuclides, is flocculated by the high salinity and gentle agitation encountered in the estuary and subsequently settles to the bottom. Through a combination of wind induced wave action and tidal currents, these materials are brought into suspension and re-deposited in areas liable to seasonal shoaling. Such deposits could become areas of relatively high radioactive contamination and under some circumstances comprise the critical point in the disposition of radioactive wastes into rivers and estuaries. By following the movement of sediment-sorbed fall-out of weapons test origin it should be feasible to study sediment transport throughout an entire estuarine system and to thus appraise the potential hazard from this particular reconcentration phenomenon.

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DISCUSSION

FRIEND: Did I understand you to say that the cesium is still tied up in the soil that has not reached the rivers?

KAUFMAN: Well, the fact that we did not find it in the river and that represents a significant portion of the unfractionated fission products suggests that it is still there.

NELSON: Strontium, too?

KAUFMAN: We haven't analyzed for strontium; so, I should not say that would be true for strontium.

FRIEND: How did you separate zirconium from the others?

KAUFMAN: By gamma spectrometry at selected time intervals. We did not make chemical separations.

NELSON: Why didn't the clays wash off to the stream?

KAUFMAN: We must assume that the clay present in the river did represent an inwashing of surface material. Furthermore, cesium should have accompanied these clays into the river. I think we can accept the fact that such elements as zirconium and lanthanum will remain on clays in salt water. We probably should not expect them to show the characteristics that the earlier speakers note for cesium arriving in salt water because the mechanism of association of trace trivalent ions with clays is not simple ion sorption. But where is the cesium? It did not appear in the river so we can only postulate that it is still on the land. But how the others were washed in, the rare earths, and the cesium was not and yet the cesium is still with the clays is difficult to explain. Further analysis of the data will be necessary.

FRIEND: Do you think it is covered up by the zirconium-niobium spectrum?

KAUFMAN: Perhaps, although the gamma energies differ by about one Mev, the higher energy Zr-Nb peak may obscure the Cs¹³⁷. We have yet to strip the spectra. Moreover, at 100 days Cs¹³⁷ represents only about 0.2% of the fission product total radioactivity. This could easily be missed.

PARKER: Did you compute the amount of cesium that might be deposited directly on the water surfaces?

KAUFMAN: No. It would be rather small though. We do not know whether the cesium has a strong affinity for the clays in the Sacramento River water. It may have been in the aqueous phase but below the limits of detectability.

SAYRE: Briefly, what sort of information were you able to collect about the sediment transport in the drainage system?

KAUFMAN: In one area adjacent to the Mare Island Shipyard there is a series of piers which seem to serve as flocculating basins. A great deal of dredging is required there. We find a buildup in that particular area. Furthermore, in these low lying areas where the water is relatively quiescent, the so-called mud flats, there was a significant buildup of the radioactive materials and we also noted there is a sediment buildup. But, curiously enough, in calculating the amount of sediment coming down into San Pablo Bay it doesn't seem to result in a rate of buildup in these flat areas that one would expect so we feel that the buildup may be a temporary one and there is a subsequent transport of the sediment out to sea.

SAYRE: Do you have any idea of the mean residence time of sediment in the Bay?

KAUFMAN: No, we do not have that information. One of the problems is that we are sampling a relatively small portion of the Bay.

SAYRE: What about the water phase; does the concentration stay about the same?

KAUFMAN: We did not analyze the radioactivity in Bay waters. We are only determining the changes in counting rate on the bottom. We have observed the counting rate in deep water and several feet from the bottom to be quite constant. We corrected for this by extrapolating the cosmic radiation contribution and subtracting it from the natural radioactivity of the sediments.

NELSON: I may have missed your last part of the redistribution mechanism. What do you mean by wave action or wind action?

KAUFMAN: Wind action in shallow water produces suspension and if it coincides with tidal movement, transportation of the resuspended materials occurs.

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CLINCH RIVER STUDIES

by FRANK PARKER

Figure 1 shows a map of the Clinch River system which has its headwaters in Virginia. It joins the Tennessee River about twenty miles below where the Oak Ridge National Laboratory discharges into the Clinch River. There are some minor uses of this water previous to that. I should now like to go over a bit of the study of the Clinch River with which most of you are familiar. You may not realize what a large administrative job it is.

The Clinch River Steering Committee consists of representatives of the Laboratory, U. S. Geological Survey, U. S. Public Health Service, Tennessee Valley Authority, Tennessee Fish and Game Commission, Tennessee Department of Public Health and Stream Pollution Control Board, and as ex officio members, AEC representatives from the Oak Ridge Operations Office and the Divisions of Reactor Development and Biology and Medicine in Washington.

The five major objectives of the Clinch River study are:

1. Determine the fate of radioactivity materials currently being discharged;
2. Determine and define mechanisms of dispersion of radionuclides released;
3. Evaluate direct and indirect hazards of current disposal practices;
4. Evaluate the capacity of the river for radioactive waste disposal; and
5. Recommend long term monitoring procedures.

The scientific investigations necessary to meet these objectives are being carried out in a co-ordinated and concentrated effort and are evaluated and directed by subcommittees on water sampling and analysis, bottom sediment sampling and analysis, aquatic biology, and safety evaluations. The extent of the co-ordinated effort is evidenced by the fact that in determining the water-borne load of radioactive materials, the sampling stations were selected by the Laboratory and the Geological Survey in cooperation with the subcommittees. The samplers were designed and built at ORNL under the direction of a Geological Survey employee on loan to the Laboratory for this study. The samples are

Figure 1

collected by Laboratory and TVA employees, mixed in accordance with river flow information supplied by the U. S. Geological Survey, gamma scanned, and strontium determinations made by employees of the U. S. Public Health Service, and the data tabulated and plotted by TVA employees and evaluated by the sub-committee. In addition, the usual stable elements are determined by employees of the State Department of Public Health and the rarer trace elements measured at ORNL. In all of the work done at the Laboratory, the U. S. Geological Survey people have been completely integrated into the Waste Disposal Research group. They are providing much of the technical effort in this project, and work reported hereafter will be that of the U. S. Geological Survey and Laboratory personnel, as well as that of other groups working on the study.

This study that we are making is not the only study that has been going on in the Clinch River System. The Applied Health Physics Group of the Laboratory has been monitoring the river practically from the day the Laboratory started, and the Waste Disposal Research Group began making some studies in 1951. Historically, the records go back to 1943. These were notes in the secret records of the Manhattan District about the effect of discharges into the Clinch River on fish population.

Figure 2 shows the waste generating areas at the Oak Ridge National Laboratory. All of the waste that originates in the Oak Ridge National Laboratory area must eventually come down over White Oak Dam as surface water where it is monitored and then flows one-half mile down into the Clinch River. The miles listed on the figure show the miles upstream from the mouth. I should point out that this is the way the river exists at this moment. However, a dam is being constructed 2.2 miles upstream and eventually we will have a situation worse than at Hanford. While Hanford will have a flow varying throughout the day as much as 4 times, we will have a flow that can vary from zero discharge to 23,000 second-feet in a matter of minutes. This will change our stream regime considerably. However, I will talk about what has happened in the last two years and by extension what we think has happened over the last twenty years.

Figure 3 shows the number of sampling stations that we have on the Clinch River. At the sampling stations we take daily samples, either grab or four-hour composite samples, and then weekly composite them on the basis of flow. On the Clinch the flows are almost completely governed by Norris Dam, at mile 79.8 upstream, and we are at mile 20.8. The headwaters from Watts Bar ordinarily come right up to White Oak Dam during the low water season, elevation at Watts Bar, 735 feet. Normal summer elevation at Watts Bar is 741 feet and then the backwaters go all the way up to about mile 28. When the new dam goes into operation we will essentially be in a pool continuously. This makes it very interesting to speculate what will happen.

Figure 4 shows the radionuclides discharged from White Oak Dam. These values were found from grab samples taken by the Applied Health Physics Group. The main contributor since 1959 has been ruthenium. The amount of strontium has decreased, in 1960 it was 27 curies, 1961 22 curies and last

Sources of Radioactive Contamination in the White Oak Creek Drainage Basin.

Figure 2

Network Water Sampling Stations

Figure 3

Figure 4

year it was about 14 curies.

Figure 5 shows the mass load of strontium-90 transported through the Clinch River system by the water and Figure 6 shows the same for cesium-137. We are dealing with extremely low concentrations. If you multiply the concentrations by the large flows then you can get some phenomenal results. The figures show the results of just the first year's sampling. The results of the second year of the study do not look as pretty yet. Some adjustment was necessary in the first year study. It is likely that the results of the second year will look similar to this. The interesting part of these curves is that if you have White Oak Creek samples and add to that the Oak Ridge Water Treatment Plant samples taken about 20 miles upstream as our background figure, the amount that comes down to the mouth of the river is about the same. We see approximately the same thing for strontium and cesium. For cesium we find that the percentage passing the downstream station is 110% of that introduced.

Figure 7 shows cumulative load of ruthenium-106 for the first year. Figure 8 shows the cumulative load of cobalt-60 for the first year and we see essentially the same thing. We could still trace the strontium and ruthenium further downstream in the Tennessee River but the cesium and cobalt in the Tennessee River were very difficult to follow. There is a tremendous increase in load at the downstream locations in the cesium curve in the second year of study and we can account for that by the pump at the sampling station at CRM 5.5. The pump was not operating efficiently and the result is that we were depositing sediment in the line. Until the pump was changed the sediments concentrated in the sample so that for a period of time we had excess sediment in the sample. By virtue of the fact that we do have six sampling stations on the river and we do have the long-term records we were able to correlate some of the stations and adjust these erroneous values.

Table I shows the comparison of material that was in suspension and what was dissolved. Roughly 90% of the cesium was suspended at Center's Ferry, but at the point of release at White Oak Dam only 63% was in suspension. In the river at Center's Ferry, 18% of the ruthenium was in suspension and at the point of release only 6% was in suspension. 12% of the cobalt in the river was in suspension and at the point of release 33% was in the suspended solids. For strontium-90, 4% was in suspended solids in the river and 3% at the point of release.

Cumulative Load of Sr^{90} in Clinch River System.

Figure 5

Cumulative Load of Cs^{137} in Clinch River System.

Figure 6

Cumulative Load of Ru^{106} in Clinch River System.

Figure 7

Cumulative Load of Co^{60} in Clinch River System.

Figure 8

TABLE I
STATE OF RADIOACTIVE ISOTOPES IN CLINCH RIVER

Isotope	White Oak Dam		Clinch River Mile 5.5	
	% Dissolved	% Suspended	% Dissolved	% Suspended
Sr ⁹⁰	97	3	96	4
Cs ¹³⁷	37	63	10	90
Co ⁶⁰	67	33	88	12
Ru ¹⁰⁶	94	6	82	18

It is interesting to notice the gamma count of the bottom silts based upon data from the Applied Health Physics section in Figure 9. This gives some indication of what is happening at the surface of the Tennessee River silt. The dosage from the silts has not increased in the Tennessee River. The rate of sedimentation in the Tennessee River and its tributaries is not great.

Figure 10 shows the gamma count at the surface of the silt of the Clinch River. We can correlate the gamma count at the surface of the Clinch River silt with the discharges from the Laboratory minus ruthenium. We can get a good indication of the total release for the previous year by measuring the gamma count of the silt.

We felt that measurement of the gamma count at the surface was not really sufficient to tell us what was going on in the system. We were interested in making an inventory of the material put into the river, the material that went down the river, the material that was stored in the bottom sediments, and material that went into the organic phase. We felt that to do this it was necessary to core sediment samples and determine the amount of radionuclides accumulated in the bottom sediments.

In 1960 we used a Pflieger Sampler to core the bottom sediments. Figure 11 is a picture of the Pflieger Sampler. We had some difficulty using the Pflieger Sampler so we investigated other types of samplers. After testing a whole series of samplers we used a piston type sampler, the Swedish Foil Sampler. Since it is only available under license we took it on a one week trial basis. It worked so successfully that we had them do the coring for us.

Figure 12 shows the places we actually collected the cores. You can see that we covered the river pretty thoroughly as well as some of the tributaries to the main stream. We feel that in this way we will get a good number for the

GAMMA COUNT AT SURFACE OF TENNESSEE RIVER SILT

Figure 9

Figure 10

Figure 11

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Sediment Core Sampling Stations

Figure 12

Figure 13

Figure 14

total inventory of radionuclides in the bottom sediments.

Figure 13 shows the Swedish Foil Sampler and Rig and Figure 14 a closeup of the Swedish Foil Sampler itself. The sampler took samples as long as 17 feet in 45 feet of water. Its capabilities are even greater than that. The best features of the sampler are the stainless steel foils on the side of the sampler so that the core itself never comes in contact with the side of the core barrel. We have taken over 600 feet of core and have built a special core scanning device. The device will move the core past a 3"x3" NaI crystal in 2" increments and will be scanned for gross activity as well as specific nuclide analyses. Figure 15 shows the log plot of core holes of number 6-C. The top of the core is the hottest and down at the bottom we find we are down to background.

This brings us up to date. When the new dam goes into operation the flows will vary all the time. We are going to change the regime in the river. This is one of the reasons we were so anxious to get the study completed this past year. We hope that on the basis of the information we now have that we can make some accurate predictions. We plan to follow the same sampling schedule with suitable variations as will be imposed by the new regime in the river and see what will happen and what the differences are at high flow rates and at high velocities in comparison to the older system. Our big problem will be getting suitable sampling devices proportional to the flow so that we can get the same sort of budget figures and have a reasonable knowledge of what is going on.

DISCUSSION

FOSTER: I don't understand the scales in Figures 5 and 6.

PARKER: This is the sum of the concentration of the weekly composite samples multiplied by the average flow during those weeks; i. e., a mass diagram, the accumulated load in curies passing that station per unit time. The abscissa is the number of weeks since the beginning of the study.

BERNARD: To what do you attribute the high jump in cesium?

PARKER: Concentration in a sample. This is at mile 5.5; we have a station at mile 14.6 and then we have a few stations further downstream at Watts Bar and at Chickamauga Dam. There is no indication either upstream or downstream that there is any concentration as great as these. We have shown it as measured and we've also shown it as correlated with the concentrations we have found at the other stations.

SERATA: What is the relative magnitude of the concentrations of Cs^{137} and Sr^{90} ?

PARKER: The average at Center's Ferry for the first year's study is 1×10^{-8} $\mu\text{c}/\text{ml}$ for Sr^{90} and for Cs^{137} at the same point 5.9×10^{-9} $\mu\text{c}/\text{ml}$.

KAUFMAN: What is your reason for showing a cumulative plot? What function does it give you that a concentration plot would not?

PARKER: Two things; first, the flows vary quite considerably in the river, the average daily flow is about 4,500 cfs, and the range for 1937-1950 is 131 and 42,000 cfs. Secondly, there is the time of travel. It takes about 1 to 3 weeks from point of introduction to Watts Bar Dam and about another week to travel from Watts Bar Dam down to Chickamauga Dam where Chattanooga is located. By integrating it you mask some of the individual errors you might have in it. If you looked at just concentrations you could not make heads or tails of it.

SERATA: Did you say Sr^{90} had a concentration of 2.4×10^{-9} $\mu\text{c}/\text{ml}$? We are finding very small concentrations so that's why I asked you.

PARKER: 1×10^{-8} .

SERATA: 10^{-9} is equivalent to $\mu\text{c}/\text{l}$. You say Cs^{137} about 60 $\mu\text{c}/\text{l}$ so you have Sr^{90} 10 and Cs^{137} about 60. Is this a reasonable ratio?

PARKER: Yes

KAUFMAN: How about variations in the silt?

PARKER: I think Dr. Friend should answer that since the variations of samples in the silts themselves are so great.

FRIEND: Yes, we tried to take a few cross-sections on the river and see. We took triplicate samples - dropped the dredge down in the same place each time and took three separate samples and analyzed them all separately. We obtained very poor results. In fact, of all of our analytical errors the biggest one is sampling.

KAUFMAN: Doesn't a dredge give you a rather mixed up sample of surface and deeper material?

FRIEND: Yes.

PARKER: You also lose fines as the samplers leak.

FRIEND: I think the best way to sample is to put skin divers down who can actually see what they're doing. Otherwise we're working completely in the dark. The biggest error we have in sampling sediments is actually collecting samples.

KAUFMAN: A detector on the bottom may give you a better gross measurement than the sample. At least it's undisturbed.

FRIEND: Yes, I'm inclined to agree with you.

BELTER: You do have increases with depth in Figure 15.

PARKER: We will have a better picture when we can scan them all.

BELTER: You haven't attempted to date those at all?

PARKER: These were just scanned this last week.

KAUFMAN: Do you have any idea how much of this you contribute to plants and how much would be there naturally? Have you got any cores in similar areas upstream?

PARKER: We do have as shown in Figure 12.

KAUFMAN: Have you scanned those yet?

PARKER: We haven't scanned all of these. I think we may have scanned about 10 cores out of 150. You can see we were scheduled and did go all the way back to Melton Dam. What we have had for background samples

is those taken at Norris Dam.

BELTER: Did you take some grab samples back from White Oak Creek up the river?

PARKER: Yes.

FRIEND: How do you calculate the amount of release to the river and on what basis?

PARKER: White Oak Dam discharges.

FRIEND: How long have you kept records on all this?

PARKER: Back to 1949 we have nuclide discharges. Previous to 1949 we kept gross counts.

COWSER: We have gross beta discharge reported prior to 1949. Currently, we are examining past records, in an attempt to estimate the strontium and cesium releases for that four or five year period.

FRIEND: When we requested values, we were told it wasn't available, that it couldn't be calculated.

COWSER: There are some records and we are attempting to come up with meaningful values.

PARKER: This is in progress reports and published.

COWSER: Prior to 1949? I think Dr. Friend was questioning what happened prior to 1949.

FRIEND: Dean Harris asked me if I could get any information on the amount of activity released to the river. I inquired and I was told these data were not available.

PARKER: Previous to 1949, the data is not good. On the basis of the activity at the Laboratory at that time and the gross count, we're trying to obtain a unified laboratory opinion of discharges.

Knowing what's gone downstream with the water and what's gone into the sediments we have to see what the organic inventory is. Trying to get information on this matter is quite difficult so we've taken an engineering type of approach to this and we've said if all the phosphorus in the river were incorporated into the bio-mass and this were contaminated to the highest degree measured in the river, we could get an outside estimate of what the total load at any one time in the bio-mass in the river could be. Since we know the amount taken up in sediment, I won't go through the same sort of thing that we went through yesterday, showing the $K_d's$ and uptake by the river sediment.

We know it's influenced, of course, by the water composition and by the type of isotopes released, the form the isotopes are in, and the velocity. We know that the phosphates are about 0.2 ppm. And if we assume that phosphorus is about 0.5 of one percent of the weight of all the organic materials in the river, and multiply that by the total volume of water in the Clinch River stretch, and if we take the most contaminated fish that has yet been taken out of the Clinch River, we find that the total strontium load at any one time in the Clinch River biomass, is about 30 millicuries. If we do the same thing for cesium with the most contaminated fish there we find the total of cesium is only about 8 millicuries. The amount in the water itself is actually quite a bit greater than that.

KAUFMAN: I do not understand how you use this 0.2 parts per million and this 0.5 percent organic to end up with a strontium load in the stream.

PARKER: If you have 0.2 ppm phosphate that's in the water, that's 0.065 ppm phosphorus in the water and knowing that the exchange of phosphorus is very rapid, therefore, we assume we are in equilibrium. Assuming that all the phosphorus in the water can be taken up and that the weight of phosphorus in the biomass is 0.5 of one percent (it varies, for man one percent and for marine animals about 0.5 percent) and multiply the ppm of phosphorus by the total amount of water in the Clinch River which is roughly about 10^9 cu. feet, and divide this by the 0.005 phosphorus, we obtain 4×10^8 grams of biomass possible in the river.

HOOPER: I still don't understand how you calculate the phosphorus pool. You would have to know something about rates. Even though this doesn't calibrate, it still doesn't show you the size of the pool unless you can determine how long it takes to pass through the pool.

PARKER: We know from previous studies that this takes place very rapidly. We assume that all the free phosphorus is taken into the biomass, though this is certainly not the case, not an actual value. I was using an engineering type approach saying that it won't be greater than this; it must be less than this.

GLOYNA: Are you making the assumption that all the phosphorus is as readily available as it was when it was initially introduced?

PARKER: I'm saying all phosphorus in the river is readily available, which isn't true either.

ODUM: Isn't it true that most of the phosphorus in the river is benthic so that if you measure it in the water you're only getting a small fraction of what's in a column. Most of what's in a column is in the organic phase. It is already in the organic phase and stays in the organic phase. The .065 ppm is what is transient between the biological cycles and is kept in the biomass of the bottom bacteria and bacterial life and so on. So the actual total content in any one column may be a factor of 10 or a 100 larger.

KAUFMAN: It doesn't make any difference.

PARKER: It doesn't make any difference in this whole approach. Let's take a hundred times more than this. Let's assume then instead of having 30 millicuries, we have 3000 millicuries. It still doesn't mean anything. That is what we are trying to show, that the total amount biologically is negligible. I see the biologists don't agree.

HOOPER: We make the biomass larger.

KAUFMAN: But really should you calculate the biomass for this kind of an estimate this way? I think this is the part that is misleading.

PARKER: How are you going to do it any other way?

ODUM: Measure it.

PARKER: How? I don't know any better way to measure the total biomass in the system.

FRIEND: Well this system is not unique. Don't oceanographers use this system?

PARKER: Yes, I think they do.

I think we can answer one of the questions that we were concerned about at the beginning of the studies. Can we really have a catastrophic situation with the radioactive material that is stored in the sediments and that is in the biomass or the organic matter in the river at any one time? I think that by using this calculation that this is an outside estimate of what could be there. If we had 3000 millicuries or 30 millicuries, it doesn't make any difference, because I think we showed we had a very small amount in the water and in the bottom sediment. I think that we don't have a hazard in the river itself. We can have a hazard and we do worry about a hazard from the use of water or the use of fish over a long period of time. The amount in the Clinch River reservoir at any one time is not large. So we don't have to worry about large scale flows or changes in pH, or oxidation - reduction potential. I think this is possibly true for other rivers. The rivers themselves do not constitute a major reservoir for radioactive material.

THOMAS: You have to be careful there; in parts of some rivers, such as the Sudd in the Nile, enormous biomasses are found.

PARKER: This is true for the Clinch River. We are looking at some of the other rivers as well.

FOSTER: Did you convert the 3×10^8 grams into grams per square foot of bottom area?

PARKER: No, this figure is for the entire Clinch River, from below the

Laboratory down to the mouth of the Clinch.

GLOYNA: How are you going to date the previous releases by looking at the present.

PARKER: We are going to see if it is possible.

GLOYNA: But you have no vast records do you?

PARKER: Before the laboratory went into operation, Watts Bar Dam was already in operation so we do have a record of zero contamination followed by an increased amount.

BERNARD: Can we go back to the first five stipulations in your paper and summarize each one for the current project?

PARKER: We don't have all the water completely analyzed nor do we have all the bottom cores analyzed yet.

BERNARD: Can you take what you have now and make some general assumptions?

PARKER: I think it is quite obvious from what we have shown so far in mass diagrams and the sediments based on the incomplete 1960 sampling plus the biological reservoir that the major part of the radioactive material passing through the Clinch River stretch is in the water at suspended sediments or as dissolved material. What happens further on downstream is a little more difficult to say as we are getting such low concentrations.

FRIEND: I think it has been demonstrated that this deposition right at the mouth of the Clinch River takes care of most of the suspended matter in the Watts Bar reservoir. I am convinced that this deposition that takes place at the mouth of the Clinch River where it enters the Tennessee River accounts for most of the cobalt. I think this is the reason you don't find it in the water from there on down.

PARKER: Even before the new dam goes into operation we are operating in a very complex hydrological system. We are in a completely controlled area. We are operating in the back waters of Watts Bar Dam. We do have a density current. The waters released from Norris Dam in the summertime are considerably colder than the waters already in the Watts Bar embayment. The cold water does underflow these higher temperature waters.

KRUMHOLZ: The cold water from Norris Dam does not warm up in the 40 to 60 miles of flow?

PARKER: It must warm up some but it doesn't take many degrees at all to have a density current.

FOSTER: Do you have any drinking water use or irrigation water use immediately downstream before it has a chance to disperse and dilute?

PARKER: We talked yesterday about the dispersion and the number of dye tracer tests. We can pretty definitely state that at almost all stages of flow that there will be good dispersion and relatively uniform concentrations by the time it gets down to our first downstream water intake which is the gaseous diffusion plant. Whether you call these people occupational or neighborhood workers I am not sure; I guess really you call them occupational workers. There is a water treatment plant there for the people who work at that plant. We have been whole body counting these people, but we haven't found anything significant.

FOSTER: My point here is that the fish, which are a dominant source of exposure for people, provide a long term integrating system. This provides somewhat less of an incentive, I think, to spread out the concentration uniformly in the river since the fish are really doing this for you over the period of several weeks.

PARKER: We have had quite a discussion about this at our sub-committee meeting on water sampling and analysis. This is essentially the reason the laboratories hold that the total amount of activity going downstream is not changed due to Melton Hill Dam. As a matter of fact it is decreasing as there have been a number of changes in laboratory procedures so that the total amount of radio-nuclides released other than ruthenium has been decreasing rather drastically. The ruthenium itself also is drastically being reduced now. So, we feel the total amount going downstream is still the same. The only thing we have to worry about is places, say like, a treatment plant or water intake. If they sampled only the water from the stagnant pools they would, of course, obtain higher values. This is not likely because the dam will be discharging during peak electrical use hours which is primarily during the daytime and this is primarily when the water treatment plant will be taking water in. Until we can demonstrate that there is an increased hazard due to present operations we are not going to make any changes in our operating system. However, we are going ahead and complete engineering plans drawn up on the changes that might be required if we find a hazardous condition developing.

KAUFMAN: Does it make any difference whether there is any dispersion following a transient release with respect to the dose to an organism in the river?

FOSTER: The actual dose that the organism will receive from an external source, say from the activity dispersed in the water, is pretty small in comparison to the dose that a person will receive from internally deposited materials which the fish have accumulated through chains. For most of the kinds of things that I am talking about here, the accumulation of particular radioelements is relatively slow. We are talking about a period of a couple of weeks or so at least for this material to build up to some equilibrium level in the fish.

PARKER: The average concentration still remains the same.

FOSTER: The average concentration which a fish sees over a two week period is the same.

KAUFMAN: The amount taken up by a fish is directly proportional to the time and concentration he is exposed to. The product of these two is independent of the dispersion.

PARKER: That's right exactly.

KAUFMAN: Why were you doing all these dispersion tests when it didn't make any difference?

PARKER: We were interested in the uniformity of concentration downstream at the water plant. We had some channeling as on the Columbia River. We were concerned since the water intake is on the same side of the river as the discharge side. We wanted to make sure that we were not channeling right into the treatment plant intake.

HOOPER: This time concentration that we are talking about is the real absorption.

KAUFMAN: It probably does not depend on the mechanism. The longer an organism is exposed, the greater the opportunity to sorb. The higher the concentration (where the isotherms absorptions are linear) the greater the uptake.

HOOPER: Do you have data to show all this?

KAUFMAN: We have some data. But no data are necessary to establish this principle.

PARKER: As long as it is linear.

FRIEND: But you are not taking elimination into account.

PARKER: We do reach a equilibrium.

FRIEND: You just don't figure you release as much cesium as you take up.

KAUFMAN: No, but the amount taken up is dependent upon the time exposed and the concentration during exposure. The product of these two is independent of dilution.

FRIEND: You have got a different situation as far as I am concerned with strontium than you have with cesium.

KAUFMAN: Not with regard to this principle! We are talking about transient of minutes, hours and a few days; not years.

FOSTER: You have some information on your Michigan P³² introduction which fits into this picture where the maximum level in the fish was reached in a few days after introduction.

HOOPER: That was more than three days or several days it was more like three months. You talking about cesium uptake I would have to see that this would really work. There are so many other complications involved.

KAUFMAN: I think it is an over-riding principal that will apply to biological or chemical systems.

PARKER: As long as it's linear. Uptake is linearly proportional to concentration.

GLOYNA: You are making the assumption that all the food chains utilize or exchange at the same rate.

KAUFMAN: All I am assuming is that the amount taken up is proportional to the concentration and to the opportunity, as expressed by time, to take it up. Certainly for the levels we were talking about in the environment the isotherms are linear.

FOSTER: This situation would be different if you have algae or single cell organisms in which the concentrations are rapidly fluctuating with that in the water because you have a very rapid exchange situation.

PARKER: I agree. The only place there could be any trouble is if this mass stays together down the Tennessee River. Industry is using the water and they are sensitive to the amount of radioactive material in it. If they were taking water in only at the time this cloud was coming by, then the concentrations could be higher. This is the only place we can foresee any difficulty right now.

NELSON: One other factor maybe you are overlooking here you must assume the uptake and excretion rate in biological materials are the same in this situation.

KAUFMAN: We know that whatever they are, they are independent of the concentrations that they are exposed to when the concentrations are this small. The rates are independent.

KENNEDY: Can you expect changes in the uptake of food with temperature differences, concentration differences, or anything of this sort?

KAUFMAN: Temperature differences, yes.

PARKER: This is where we could have a problem. If we were using the simple rule that for each 10 degrees rise in temperature we would double the metabolic rate then we would be reducing the uptake by lowering the temperature.

FRIEND: I think you would run into the same problem here talking in generalities. I don't think you can talk about cesium and strontium in one breath. We are going to have to talk about them completely separately. We can almost determine the amount of strontium in a bone from the age of the fish if he is caught say in the Clinch River.

KAUFMAN: The amount in the bone of a man say depends how many drinks he takes and what the concentration in the drink is; the fish is not unique in this respect.

PARKER: If you take a fish out of White Oak Lake and compare it with a fish in the Clinch River you cannot judge age on the basis of the amount of strontium.

FRIEND: I didn't say this. I said if caught from the same place, Mile 14.6 where most of our test fish are caught, then it is possible.

PARKER: I am not so sure that's true based on the tagging results. On the basis of the tagging some of these fish range widely and the concentrations can be considerably different. Even when they come from the same place there is considerable variability of the concentration in the fish.

KRUMHOLZ: When they come from the same place and at the same time there might be a variation of more than 300 per cent.

FRIEND: You can do this with strontium, but you cannot do it with cesium.

FOSTER: The time constants for the different nuclides are certainly going to be different dependent upon the physical and biological halflives. In the case of strontium it may take on the order of a year for the fish to reach equilibrium. There are some other materials which have some shorter time constants. Sodium is probably one of the fastest because rapid exchange occurs directly via the gills rather than through a food chain. Then the time to reach equilibrium may be a matter of hours, but you can also look for the fish to lose it rapidly through the gills.

FREDERICK: You mentioned several times that the discharge of radioactivity to the Clinch River has been drastically decreased over the past six months to a year except for ruthenium. I would like to know how this is being decreased, is it by way of waste treatment or less production of waste?

PARKER: Actually by both methods.

FREDERICK: What are you doing with that that you formerly put in the river

but you are not putting in the river now?

PARKER: Convert it to solids and putting it in the waste pits. The fuel element storage canals, for example, have been taken out of the processing waste system and put on an ion exchange system.

FREDERICK: You are putting it somewhere else then?

BELTER: We do not have the same type of operation that we did. The fuel reprocessing work has been drastically reduced.

YOTSUKURA: Do you have any information about the rate of dilution of radio-nuclides following continuous release?

PARKER: Yes. We have some tracer tests as well as the radionuclides themselves.

YOTSUKURA: Is this shown in any of the information presented here?

PARKER: Yes, it is shown in Figure 3. We have six stations downstream. I can give you the exact distances the nearest downstream station is seven miles, then sixteen miles downstream, then sixty miles downstream, then one-hundred and twenty miles downstream.

YOTSUKURA: Have you plotted the maximum concentration against the distance on the log-log paper?

PARKER: Against the dilution factor of other waters coming into the river but not just by distance itself. I don't think that's particularly meaningful. We could do it by drainage area.

YOTSUKURA: You didn't see any linear relationships?

PARKER: Well, essentially that's what the mass curves do show. If we do assume that there is very little uptake then it all goes through. We divide the concentration by the dilution. You see that the curves fall one on top of the other essentially showing the factor of dilution.

BELTER: I can see some of the wheels turning in the minds of our biologist friends - perhaps going in the opposite direction as certain statements were made by some of our engineers. I think probably this is a good time for letting the biologists speak. For continuity sake I think it is appropriate to have Dr. Nelson continue with the work at Oak Ridge and present some of the biological work that has been connected with the Clinch River.

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ROLE OF BOTTOM ORGANISMS

by DAN J. NELSON

In consideration of the role of bottom organisms in aquatic environments we should look at rate processes instead of the standing crop. This morning I am going to discuss three modes in which organisms living in a river can affect the distribution or re-distribution of radioactive material. One is on tudificid worms. Another is on clams and their large concentration factor of strontium-90. The third regards aquatic insects and the possibility of their flying out of the river and contaminating the nearby landscape.

In some of our preliminary bottom samplings on the Clinch River we found the tudificid worms were the most abundant organism in the river bottom. These worms have a unique feeding habit as shown in Figure 1. They burrow with their heads in the bottom sediments and with tails up in the flowing stream. They defecate upwards into the liquid and the fecal pellets fall back. Dr. Parker mentioned the density currents in the Watts Bar Reservoir. We wanted to measure the effect of feeding activity of these worms on the distribution of contaminated river bottom sediments as a function of the density currents. We have a height factor, a depth factor and a velocity factor. We could not measure these effects directly in the field so we did some field sampling to determine populations, brought some worms into the Laboratory, set them up, studied their feeding activities, measured the height they would stick their tails up, the depth they would burrow into the sediments, and so forth.

The model for the estimation of sediment moving by the tudificid worms is shown in Figure 2. For the conditions shown in Figure 2 we can solve all the distance downstream a pellet would fall, time it would be required to travel downstream, X . It was assumed that the worm would have to eat the volume to depth, h , until he ate the surface layer. Therefore, it was possible to determine the number of hours to move the volume in the area.

Figure 3 gives the measurement data used in the model to determine downstream transport of sediment by the tudificid worms. We found that the average distance downstream was about 9.2 cm and the time to travel this distance was about 4.2×10^{-4} hours. The time to remove the sediment from 1 cm^2 to depth h was about one-third of the year. At a population density 1 worm/cm^2 it would take three-tenths of a year to move the sediment downstream 9.2 cm. The worm population in the Clinch River is only one-tenth this number, so it can be seen that the effect of the worm populations as they are in the Clinch River are clearly

v = CURRENT VELOCITY
 a = TAIL ELEVATION
 h = DEPTH OF HEAD
 p = FECAL PELLETS

Figure 1

Model for Estimation of Sediment Movement by Tubificid Worms

Height variable, $y = a - rt$

where

a = the height of the worm tail above the sediment surface,

r = the rate of fall of fecal pellets,

t = time, and

Distance variable, $x = vt$.

where

v = velocity.

Then

$$y = a - \frac{rx}{v}.$$

a , r , and v are determined experimentally; therefore $y = 0$.

$$x = \frac{av}{r} \text{ or distance downstream.}$$

$$t = \frac{a}{r}, \text{ time to travel distance } x.$$

The worm is considered to be feeding in 1 cm^2 of bottom surface with its head at depth h .

When w is the rate of feeding in g/hr and s is the specific weight of the sediment, mg/cm^3 , it will take $\frac{hs}{w}$ hours to move the volume of the $h \text{ cm}^3$.

Figure 2

Figure 3

Measurement Data Used in Model to Determine Downstream Transport of Sediment by Tubificid Worms

	Symbol	Number of Observations	Mean Value $\pm$ S.D.**
Rate of fecal pellet fall	r	14	26.43 $\pm$ 8.56 cm/min
Rate of feeding	w	15	7.47 $\pm$ 4.36 $\times 10^{-4}$ g/hr
Height of worm tail	a	88	0.665 $\pm$ 0.309 cm
Depth of burrowing	h	15	0.827 $\pm$ 0.317 cm
Diameter of feeding worms		21	17.6 $\pm$ 4.96 $\times 10^{-3}$ cm
Mud density	s	1	2.64 g/cm ³
Current velocity	v	*	2.19 $\times 10^4$ cm/hr

* Value estimated from: A. S. Fry, M. A. Churchill, and Rex Elder, 1953, "Significant Effects of Density Currents in TVA's Integrated Reservoir and River System," Proc. Minnesota International Hydraulics Convention, pp. 335-354.

** S.D. = one sample standard deviation.

insignificant as far as the transport of sediment is concerned. However, if you consider tudificid populations, and we have a number of sanitary engineers around here who are familiar with tudificids, suppose we had a population of 100 worms/cm². Then it could be said that the head end of the worm buried into the mud might displace a lot of sediment and since you had a lot more worms you would actually move a lot more dirt. Well, the movement of the dirt at even the high populations could cause sediments to move downstream at a rate of 27.5 m/yr. If we wish to relate this to half-life of cesium-137, which is strongly adsorbed to the sediments, and measure the distance the worms would move these sediments downstream in one half-life of cesium we would come up with about 800 meters. This movement is probably significant compared to the transport of sediments by water. We do not have the exact figure for transport by water but we are waiting for a figure to make some comparisons.

Sometimes it is dangerous to go from a laboratory model to a field situation but in this instance you would have to change conditions considerably before you would change the general conclusion. We were working at a fairly high oxygen concentration in water and we know when the oxygen saturation goes down the tudificid worms stick their tails out further when they feed, but this is still probably a minor factor.

I also considered studying the desorption of isotopes as they pass through the gut of worms in the digestive process; we wondered if this was taking place. But, since they were just handling so little dirt I thought I would let that problem go and come back to it when I had more time. This is another way that worms could affect the distribution of an isotope if there were enough of them.

Another possible mode in which the organisms could affect the distribution of radionuclides or other elements is through a population which has a large biomass and which concentrates the elements. Previously we used strontium-90 concentrations in clams to indicate the behavior of strontium-90 releases to the environment. During this study we came up with an average concentration factor of approximately 4×10^3 for strontium in the clams. Individual concentration factors were as low as 2.5×10^3 and as high as about 9×10^3 . The concentration factor is the concentration in the clam shell versus the concentration in water.

There is a commercial clam fishery at several locations on the Tennessee River which harvests 5,000 to 10,000 tons of shell per year. The average strontium-90 concentration in clams from Chickamauga and Wheeler Reservoirs was 7.019 $\mu\text{mc/g}$ shell. This concentration of strontium-90 in the shell and the 10,000 ton/year harvest would result in the annual removal of about 64 mc strontium-90 from the river. As was mentioned yesterday the release of strontium-90 from the Oak Ridge National Laboratory in previous years ranged from 29 to 150 curies, or roughly from 1 to 2 curies a week. Therefore the harvest of clams, an organism that concentrates strontium-90, would result in the removal of only small amounts of strontium-90 from the river when compared with average releases. Or, you can look at this situation in another way. If you

assume the harvest of clams was in equilibrium with new growth each year this would mean then that clams were depositing strontium-90 at about the rate of 64 mc per year. Fortunately, some of the biologists in the Fish and Wildlife Service have made estimates of the clam populations in this area. They actually went down with scuba equipment, made counts on the clams and arrived at a value with a range of 3.8×10^7 to 4.5×10^7 individual clams. If we take an average weight of the population of about 250 grams per clam, this might be somewhat of a high figure, we could calculate on the basis of our strontium-90 figures again a reservoir strontium-90 in this population of 79 millicuries. You realize this has been intergrated over a number of years of radioactivity release from the laboratory. Here we have an organism which concentrated a radionuclide by about 4×10^3 times but yet it cannot pick up enough to change the whole distribution pattern in the water.

One of our other problems has been the transport of radionuclides by insects. In 1959 some Russians published a paper in which they discussed the possibility of biological purification of reservoir sediments containing radioisotopes by a process of insects growing up in these reservoirs and flying away. There are a large number of insects with immature aquatic stages and many of these emerge and come to light at night and can be collected. On several nights last summer we set up a light trap on the river bank next to one of the cross-sections that the Applied Health Physics Group was sampling with the Eckman Dredge. The insects that we captured were almost exclusively chironomids; we did not identify the particular species. We captured about 8.75 grams of dried insect material; this is an awful lot of chironomids. We had the insects analyzed and compared the analyses with the analyses of the sediments taken from the same cross-section and at approximately the same time. We found strontium-90, cesium-137, and the rare earths in the insects. Ruthenium-106, cobalt-60, frequently cerium-144, and zirconium-niobium-95 are also found in the sediments. With larger samples we might have been able to detect the less common radioisotopes. In comparing these we find that there are concentration factors for Sr-90 and Cs-137 but nothing significant for TRE as shown in Figure 4.

We could consider this from another standpoint. The difficulties of sampling bottom sediments have been mentioned with respect to getting a representative sample. Perhaps for some radionuclides, strontium-90 for instance, we could consider insects as being integrating samplers over the surface area of mud. Insects do concentrate radionuclides and they are much easier to handle than mud. We might be able to set light traps on the river banks and compare activity in emergent insects with activity in mud samples.

I do think there may be some possibilities of using insects as monitors for certain kinds of radioactivity. You might even be able to pick certain habitats or environments the insects live in to find out what is in a mud bottom, a gravel bottom, what is in the algae, and so forth.

Figure 4

Radioactivity in Emergent Chironomids and Sediments from CRM 19.1

	Activity $\mu\text{c} \times 10^{-6} \text{ g}^{-1}$		
	Sr ⁹⁰	Cs ¹³⁷	TRE
Adult Chironomids	3.55 $\pm$ 1.13	139	7.82 $\pm$ 0.92
River Sediment	0.405	5.22	7.25
<u>Chironomid Concentration</u>	8.76	26.6	1.08
Sediment Concentration

Figure 5

PHOTOGRAPH FROM WORLD WIDE PHOTOS

The picture above was published in the *Chicago Daily News* on July 8, 1946. It carried the legend: "May flies stop motorists. Lawrence Rutz stops his truck on the west channel bridge at La Crosse, Wis., yesterday to clear May flies from the front of the vehicle. The insects got so thick they obstructed the view of the driver, clogged radiators and made roadway of the bridge slippery."

Figure 5 shows some of the tremendous numbers of mayflies which may emerge; a man is standing ankle deep in this Wisconsin area. Frequently, along the Mississippi River there are tremendous emergences of both mayflies and caddis flies; you can literally take a scoop shovel and shovel them up.

By comparing the quantity of strontium-90 removed from the river by emerging insects with that coming into the river from world-wide fallout, we can get an idea of the relative importance of radioactivity transported by insects. Chironomid productivity ranges widely for different habitats and may be as high as 20 g/m²/year (Teal, Root Spring). H. T. Odum observed an annual productivity of 3.6 g from Silver Springs. If we consider the strontium-90 concentrations in the Clinch River chironomids, and assume a median production of 10 g/m²/yr you can calculate that the insects would remove about 3.55×10^{-5} uc/m²/yr from the river. If we compare this with fallout figures which have been very prominent in the last few years it would be something like this. From 1959 to 1960 we had an added increment of fallout, the latest year for which data are available, of 4.2 mc/mi² (1.62×10^{-3} uc/m²/year) and the accumulated Sr⁹⁰ actively for east Tennessee was about 50 mc/mi². Currently, there is about 45 times as much fallout coming into the river as the insects carry out. Thus, it is apparent dispersal of radioactivity by emerging aquatic insects is of minor importance.

DISCUSSION

BERNARD: What is the distribution between the shell and the meat?

NELSON: I cannot even give you a good answer on that. I have not analyzed any meat samples for Sr^{90} because I did not know how to interpret the data.

BERNARD: I am trying to get some ideas on how much would actually go to a human being.

NELSON: I have some stable strontium analysis on meat; they vary by a factor of 2 to 17 in any one clam, depending upon the tissue analyzed. There is from 150 parts per million to a little over 500 parts per million of strontium in the shell and when we talk about meat we are talking about a few tenths of a part per million to 3 or 4 parts per million. There is not much strontium in the meat.

BERNARD: In essence then, would the hazard be in handling the shells and not in eating the clam meat?

NELSON: There is not that much radioactivity in the shells. We have one shell collected near White Oak Creek whose radioactivity would register on a survey meter, but this was quite different from the general situation.

PARKER: Could you make an estimate on the amount of Sr^{90} that accumulated in a clam in a year?

NELSON: No, I haven't been able to do that yet. If you assume equilibrium in the harvest and in the growth, and the biologists who made the population study by the way concluded the population was being over-harvested, we are saying we are tying up more Sr^{90} than we really are so this is a maximum figure.

PARKER: How much was that?

NELSON: 64 millicuries is the harvest which we can calculate on clams removed and their average Sr^{90} concentrations. If we assume the harvest and new growth are in equilibrium and releases were constant, but now our releases have gone down, so this is going to reduce the level somewhat.

FRIEND: This doesn't include the Asiatic migrating clams, does it?

NELSON: No. We do not have them up in the Clinch yet - I do not know why; they're every place else.

SERATA: You talked about concentrations in the body how about concentrations in the worms?

NELSON: I didn't look at radionuclide concentrations in the worms at all. When you look at radionuclide concentrations in something like a worm you have another problem. You have their gut contents plus the worm tissue itself and you start to weigh something like this and you have a little bit of worm wrapped around a lot of mud. You weigh this and you come up with a value closer to the mud concentration than the tissue concentration of the worm.

PARKER: How do you know for what period of time you would be integrating?

NELSON: It's probably pretty short. This is comparable to your time of flow; it depends on how fast they turn it over. It's probably as good as taking a sample one time a year and saying this is it for a year, it's probably better.

PARKER: Might this be of use for checking a reservoir?

NELSON: Yes. These insects have a life cycle probably anywhere from 25 to 30 days so they grow up in this period of time.

PARKER: But they tell you only what's on the surface of the muds.

NELSON: Yes, that's right but that's all you are getting out of a surface grab sample.

PARKER: How about a core sample?

NELSON: From the standpoint of a core sample you are right in that insects only sample the surface but there are two different aspects involved. If somebody did want a continuing surface grab sample you probably have some insects that are just as good, probably even better than dredge samples because they would sample a large area. If you wanted to be particular or you wanted to sample a rock or sand bottom an alga could take organisms which live in a specific habitat. For instance, some caddis flies or mayflies may have radioactivity associated with different microenvironments and feeding habits. You might even be able to convert this into a suspended solid measurement by sampling filter feeding organisms.

PARKER: How are you going to know that your river sediments are representative?

NELSON: Well, this is what we get.

PARKER: They are no better than the figures we are using anyway.

NELSON: That's right.

BRADLEY: This system of sampling consists of sampling one gram of sediment and a hundred grams of water.

NELSON: This isn't much when you dry it down.

BRADLEY: When the "bugs" sample they got about 1% of sediment and the rest of the sample was water.

NELSON: That's right.

BRADLEY: That's not very close to a sample of the sediment.

NELSON: Well, all we are looking at this thing for is as an index and a comparative value.

PARKER: The fishs' large fault is that they too can be transported out of the system.

BELTER: Do you have any quantative approximations on the fish studies as far as the populations and also some of the radiochemical analysis on them?

NELSON: No. I am satisfied to use Dr. Parker's values for awhile.

BELTER: You are not working on this phase then?

NELSON: No.

NIELSEN: On those worms you were treating them as if they were just shoveling the material downstream.

NELSON: That's right.

NIELSEN: You're sure they are doing something chemically also.

NELSON: No, I'm not sure that they are doing anything chemically. This is something I would like to investigate.

NIELSEN: Maybe I should not have said it that way. Perhaps they are putting some of these materials into the solution.

NELSON: That's right; the reason I did not pursue that study any further was the fact that they just did not handle enough mud to change the whole picture.

NIELSEN: Well, I hope we do not have them on the Columbia River.

KENNEDY: Is there any possibility that these worms might enrich the material in their bodies considerably so that when they die the remains might be carried and re-deposited in such a way that you would get local spots that were quite enriched.

NELSON: They are just not enough worms in the river bottom to change this much. However, you might get high concentrations of worms in some of these situations that the sewage plant operators are quite familiar with.

FOSTER: Another feature along this same line is that you could view this as a system in which you have a certain inventory of radioactivity materials per square foot of bottom to begin with. Then you have the worms which are eating this bottom material and this results in some concentration of the material from a square foot area into the worm which give you some little hot spots, but you have not really changed the total inventory per square foot because you have made hot spots out of the individual worms. You cannot really increase the inventory some place by moving worms.

KENNEDY: The point I had in mind is that if these things die and their remains are carried in suspension you might get highly organic layers in the area of deposition. If these remains had greatly abnormal concentrations of radioisotopes, then you may have a highly radioactive, organic-rich layer. When this organic material has an opportunity to decompose you may put the radioisotopes back into solution due in part to a great reduction in Eh.

NELSON: I guess almost all of the organic-rich layers in streams anyway come from leaf material that falls in and then the worms might then invade this later on for the organic matter which is present but in this case I guess the worms would be secondary when compared with the other organic material which is present.

FOSTER: Along this line, we made an observation at Hanford, quite a number of years ago, that involved larval insects which live on the bottom substrate. These go through a series of metamorphosis in which they shed their skins. These skins are quite light and they float to the top of the water. Sometimes these accumulate in surface scum and the radioactivity of the scum will be much higher than that of other layers. Most of the activity is associated with the molted skins.

ODUM: In an environment where you have an alternation of quiet waters in rivers (which I think this river does not have) or in deep water you have an opportunity for a ten to one ratio, between the concentration of the biologically held materials in the rapid areas to deep or quiet areas because of the rate of productivity where the physical energy of the stream provides mechanisms for greatly concentrating foods. The fouling and reef phenomena of the marine environment are similar. In comparing streams and rapids in river pools it is possible to get a 10 to 1 concentration difference. You should look for this

difference in a stream that has available current energy to supplement the productivity sources. The biological concentration provides a means of locally concentrating radionuclides which are part of the structure of the biota.

NELSON: Then you would sample these locations. Ours is a pretty homogeneous system throughout the mud bottom community.

SPOFFORD: You assume in your model that these worms do not move horizontally. Do they actually go through a life cycle in one location?

NELSON: They probably do not move very far. We know they go down into the bottom sediments and they can stir them up some and this might affect the results of core analyses if you had a big population because they will in time turn over a mass of material completely. If they are taken to the laboratory in high concentrations in enriched concentrations they will completely go to the bottom of the dish and start up again.

SPOFFORD: Then, they don't seem to travel very far in a horizontal direction?

NELSON: I don't think they travel very much. If you put them in plastic-sided containers you can see that some of them do move up and down the sides of the container so we know they move some, but I would say that this is not of any great consequence.

KAUFMAN: To what extent are they a source of food for some of the bottom feeders?

NELSON: This is probably very important. Looking at the stomachs of Blue Gills, you find remnants that maybe are worms but they break down so quickly I think they go almost as fast as they are swallowed. So, you find a little bit of tubificid material and you frequently find a lot of the larval stages of the chironomids.

KAUFMAN: In the fish food chain in the Clinch River, how important are they when compared to other sources of food for the fish?

NELSON: This is something I am not ready to say. When I said there was more of these worms down there on the bottom than any other thing this is a numbers count and I don't know how fast the worms are turning over in comparison to the chironomid larva on the bottom. If you look at "stomachs" you have to say on the basis of content you see the chironomids were more important.

KAUFMAN: In other words you wouldn't necessarily consider these a good radiological index of fish-food man chain?

NELSON: They probably enter, but to what extent I don't know.

SARATA: How deeply do the worms go?

NELSON: I have forgotten the figure we had on the Figure 3. Some of the original cores I saw had worm activity several inches down as far as the worms were concerned, but this is after they have been held in the laboratory and some anaerobic processes were apparently starting since gas bubbles were present. It's really hard to tell. The data we got in the laboratory out of deep dishes indicated they did not use the depths available.

KRUMHOLZ: On the basis of fish at Oak Ridge I don't know how accurate my data are, I'm trying to remember data from 1953 when we treated White Oak Lake with rotenone and killed the fish population; from a rough calculation I would guess that in about 17 tons of fish there was about a third of a curie of strontium.

NELSON: You are talking about a several orders of magnitude higher concentration than we are dealing with right now in the river.

KRUMHOLZ: That's right. I am talking about White Oak Lake. I am not talking about the Clinch River. What I am saying here is that this will give you an idea of how little there is in the biota as compared to what there is available in the bottom sediments.

NELSON: That's right. Also, the releases from the laboratory were higher at that time.

KRUMHOLZ: At the same time in that upper area at White Oak Lake there was a tremendous bed of Spirogyra, a filamentous alga, we figured there were about a thousand millicuries of P^{32} tied up in that one bed. The bed was about two acres in extent.

NELSON: Incidentally, I think phosphorus is probably an exception to the general rule that biological reservoirs are insignificant. Phosphorous is an element for which there is a large biotic reservoir. One of the things we are lacking on many of these elements is the turnover rate or biological half-life. So, here is an idea that maybe we ought to look at, residence time, perhaps of an element in water as much as anything to get an idea of how fast it does go through the system.

THE ELEMENT RATIO METHOD FOR PREDICTING BIOGEOCHEMICAL MOVEMENTS FROM METABOLIC MEASUREMENTS IN ECOSYSTEMS

by H. T. ODUM

In the letter that was distributed by Mr. Belter, the question was raised "how can we put together all the bits of information about various radionuclides and various components of systems; how can we get some predictability; and what do we need to study next in order to accomplish this?" The problem is one of synthesizing a hundred elements and about twenty main phases of the usual living and non-living components of the ecosystem into one picture that has quantitative predictive value. Such synthesis is ecosystem science: namely, the study of the processes of the system and their biogeochemical component cycles.

Let us consider possible extensions of the old ratio method for this purpose. For a long time in physiology, as you well know, the respiratory quotient idea has been used for relating at least two elements, namely carbon and oxygen. So, if one knows what happens to carbon, one can without knowing much else say something about what is happening to oxygen quantitatively. In oceanography, Redfield extended the idea to lesser elements, phosphorus and nitrogen.

The method of relating a trace element to the main element carbon works similarly because many element ratios of a whole ecosystem's metabolism are conservative and somewhat similar from system to system.

The general question is: How are the minor elements in waste related to the carbon which is the most abundant in the metabolic process? Can we get the ratios of these other elements to the normal process and then use the measurement of the carbon metabolism to predict the cycles of the minor elements? Can we go to an unknown situation and measure the seasonal and the spatial aspect of the carbon metabolism as a general procedure for some minor elements? Many data are now available on this from different kinds of streams, and we can do this to some extent already in the marine environment. Can we now formalize the method in terms of the metabolic circuit network, the storage quantities, and the rates? Can we provide a mechanism of presenting this to each other when we have some data and want to compare it on the basis of a common denominator? Is there a diagram we can use? Can we provide a network circuit for simulating quantitatively the temporal behavior of the system?

To get us orientated let's run through a few figures to illustrate the long-established business about RQ's except here are some examples from environments where we are not dealing with single organisms but a complex ecosystem. Figure 1 shows dual process which is involved in this system along with others. There is the photosynthetic process and the total respiratory consumption process including bacteria, animals, the plants at night and the plants in the daytime, too. We would like to be able to write a stoichiometric equation involving all of the hundred elements that have some conservative stoichiometric relationship. Which ones can not be counted upon to have any predictable relationship? We already know that phosphorus and nitrogen are related. Presumably other elements that are biologically involved and scarce in the environment do have the conservative-ratio property. We do not know about some of the others. For example, is gold involved at all?

Figure 2 is any system, here is the same photosynthetic process, the respiratory process, the storage of the organic bound materials, the storage of the inorganic bound materials, the influx of organic, the outflux of organic, influx of inorganic, and the outflux of inorganic. Any system can be classified. We can make a metabolic classification system in terms of the relative sizes of these various fluxes and internal flows. We should be able to write one of these diagrams for each element like zinc or cobalt and see if there is a conservative ratio between the circuit diagram you get for one element as compared to the one you can get for strontium. It will probably be very useful to consider biogeochemical circuits in the same sense as you do electrical circuits. For one reason you can immediately apply electrical network analyses as a number of people have started doing. So, why not start presenting our data as circuit diagrams for each element.

Figure 3 is an example from Beyers' thesis in which he reports metabolism of replicate microcosms, which are miniature systems that have the essence of natural systems. In this particular case he is plotting daytime rate of oxygen production and rate of carbon assimilation and at night the reverse. The graphs are plotted on a molar basis and carbon and oxygen do follow each other. If you had known one, you could predict the metabolism of the other in the whole system just as has long been done in single organisms.

Figure 4 shows a blue-green algal mat associated with a bleed-water pollution situation. I will use it as an example of a system in which the biological metabolic processes are so intimately coupled with the inorganic processes, that we heard about yesterday involving clay, pH changes, the effects on bonding and oxidation-reduction potential, that it is really quite impossible to separate them because the energy for driving the chemical process in this instance is apparently the biological photosynthetic process.

Figure 5, associated with that same blue-green algal mat, shows several things of interest from work by Neal Armstrong. This is a plot day and night, of the voltage. Here is platinum to platinum voltage between the top and bottom; there is a little coil of platinum a foot long just in the upper fluid over the mat

ANALOG

Figure 1

Figure 2

Figure 3

Figure 4

Photoelectric Ecosystem

Figure 5

and another one inserted just under the mat. This mat of blue-green algal has bacteria, has sulphur cycles, is self maintaining, and is a little ecosystem. The under part is the reduced and negative; it is the hypolimnion of this microstratified system. It is a photo-cell with a half volt open circuit potential. If you have thousands of acres of this - you can get as much power as you have platinum grid. We would first have to find something cheaper than platinum, if we are going to power astronauts and things with such living closed systems. The ecosystem provides a sharp diurnal chemical drive by generating the voltage diurnally across the half cm thick mat. What is happening to the radio-nuclides that might be there? What kind of effects result from electrophoresis gradients? What is happening to oxidation-reduction states? It is like Al Storey's experiment with ruthenium, in which the pH was critical in the turtle grass in summer. The diurnal metabolic change in pH and in oxidation potential are controlling the inorganic chemistry. This example illustrates the inseparable coupling of organic-inorganic processes, at least in some systems.

Figure 6 is a shallow turtle grass bed near Port Aransas, in about a meter of water with high rates of plant photosynthesis and associated animal growths. Pat Parker did his studies on the turtle grass by placing open ended cylindrical enclosures over the bed in the field and adding isotopes. He studied the normal trace concentration and then some kinetics with tracers.

Figure 7 indicates the kind of annual variation in the metabolism that you get with carbon and oxygen in the field from the Laguna Madre of Texas; in this case photosynthesis and respiration stay together. If one has such a graph as this and a ratio of the behavior of zinc to the behavior of carbon then without going out and making a separate study, one has some means for predicting the rate for cycling for different times of the year of the zinc or other elements where they have been so coupled to the biological carbon cycle.

If you could classify ecosystems in terms of their metabolism using several categories you would have some way to begin to generalize about environments. Figure 8 shows four kinds of metabolic patterns in each system. The Laguna pattern you just saw is represented by the diagram with a strong seasonal pulse and with P and R in phase. The respiration stays right with photosynthesis because of migration of the species into the area in the spring and moving out into the Gulf in winter. Also shown is the absolute steady state. (The word climax in ecological literature is synonymous with steady state, I think). Another graph represents P and R out of phase as may be characteristic of North Atlantic areas. Another graph is from a study which looks like a random noise variation pattern. In other words if you could classify systems according to carbon metabolism, the data on minor elements being gathered all over the world in great quantities might be logically classified by types for prediction purposes.

Figure 9 gives additional aspects of such a classification. Here is one in the terms of the diagram which we showed you earlier, where P and R are the only processes; that is the closed system. This is one dominated by the

Figure 6

Figure 7

Figure 8

Figure 9

photosynthetic process. To have this requires a steady input of inorganics and a steady output of organics. Another is the reverse - the pure culture type with a steady input of organics and output of inorganics, like a sewage system. The Clinch River is probably one of these mixtures but is heavier on the heterotrophic than on the autotrophic. Most streams that have been looked at, other than clear springs, are something of this type. Again if you have data on each element related to this P and R process and you have an idea of the relative ratios and magnitudes you can immediately say something predictable or if you have made a study you can show how it differs from the norm and thus begin to get some coherent knowledge. Figure 10 gives the time processes for those same types. Here are those four types we had: (1) the extreme with no energy source you simply run down, (2) $P = R$, (3) $P > R$, and (4) $P < R$. Those are the steady states we just talked about. The kinds of patterns one may predict from some initial starting state represented over on the left are given as a function of the steady state type. The ratio of the starting concentration to the steady state determines the temporal pattern. We have initially a very large organic storage, more than can be sustained in steady state. Therefore, the respiratory process is going to start off higher since it is proportional to the concentration of fuel that drives it. It is going to have a tendency to run down to a lower level which is in keeping with a lower average stock. Similarly, starting with a fertilizer content high in organic matter, the photosynthetic process is going to tend to start out higher than can be sustained by this particular steady state arrangement. Knowing your steady state and from what kind of starting condition you can classify the sequences of time approaching the steady state into these various categories. There are descending P's, rising P's, combinations with P descending and R rising and situations with both rising and settling back to an intermediate state. If we have for each element a ratio to carbon for P and R for enough systems to accept certain limits of expectation, then one might extend predictions through the schemes of classification to streams, marine bays, sewage plants, outer-space capsules, and other diverse kinds of systems. Is it possible with all of these to have some predictive basis for extending cycling rates for carbon to the lesser known elements through this kind of procedure? Can we formally unite the science of productivity studies and the biogeochemical studies that we have been hearing about this week.

Consider a specific example. The turtle grass Figure 6 merely showed the data used to get the oxygen-carbon story within a fiber glass triangle set out in the bay. Twenty-four hour sequences of oxygen, pH, and carbon were obtained. This particular graph compares data outside to that in the enclosure which isolates the cycle as a sample. We have been using the outside measurements to measure the productivity and bay respiration and have several hundred diurnal measurements up and down the Texas Coast. Before doing isotope experiments in such an enclosure you would want to be reasonably sure that the metabolic process was still continuing, and this kind of comparison is used for this verification. We computed productivity by measuring the rate on this graph after diffusion corrections were made.

HIGH ORGANIC

HIGH INORGANIC

LOW BALANCE

HIGH BALANCE

BALANCED

AUTOTROPH

HETEROTROPH

EQUILIBRIUM

HETEROTROPHIC LEAD

AUTOTROPHIC EMERGENCE

REGENERATIVE
HETEROTROPH

LOG DECAY

AUTOTROPHIC LEAD

REGENERATIVE
AUTOTROPH

**HETEROTROPHIC
EMERGENCE**

RISING BALANCE

AUTOTROPHIC RISE

HETEROTROPHIC RISE

DECLINING BALANCE

AUTOTROPHIC DECLINE

HETEROTROPHIC DECLINE

SAME AS
ABOVE

SUCCESSION IN TIME

Next, can we present our data in some kind of common diagram which can also be transferrable to electrical circuits when the number of units tend to get a little bit more involved? Can we apply the existing science of network analysis? We are in the dark ages in biogeochemical circuits as compared to electronics circuits. Why don't we short cut progress and make the language of biogeochemical circuits intra-convertable? Here in the grass flat we have several compartments, using the term compartment as does Shepperd in his book. The examples in his tracer method book concern the organs of the body but the organs of the ecosystem are just as pertinent. In the grass flat there is a water compartment, a plant compartment, a sediment-complex compartment, and an algal compartment. Each one of these can be broken down as the data become available so that you can add more and more detail. Each compartment has a storage capacity. On the electrical side of the circuit we represent it by a capacitance. Each wire is a biogeochemical conductance. If you have no animal, no physical transport of clay, no other physical process, there is no circuit. If you have a transport process then you have a conductance which may be high or low. The parameters can be determined from biomass or metabolism. Involved in this kind of approach is the assumption that there is linearity and that the analog to Ohm's Law is comparable.

Organic concentration represents voltage, metabolic flow represents amperage, and conductance represents the pathways both in the biological and electrical circuits. This concept differs from standard analog computer methods where one has operational analyzers and one adds energy at every step and without concern for current. Here we are interested in voltage, currents, resistance, and power, making all aspects of the physical circuits system analogous to the real biogeochemical circuit system.

Consider Parker's data on the zinc in the grass flat recently published (1962). Relate it to the carbon cycle. In the grass system there was about 100 grams of carbon per square meter in the water, and 1500 g/m^2 in benthic plants, and 10 gm/m^2 in the animals. Considering the sediment in the exchangeable section as a centimeter, there is about 100 g/m^2 carbon.

For the rate of flux you have 10 grams carbon per square meter per day photosynthesis and respiration. The animal flux can be determined by multiplying the biomass by the metabolism per individual animal which gives an order of one g/m^2 per day. The circuit drawing is a graphic way to try to show the comparable carbon cycle and relate it to the sun's energy and to the stock.

The time constant in any one of the phases (phases are: the plants, animals, water, and sediment complex) in contact with the system is $1/RC$ where R is the resistance of the circuit and C is the capacity, so we use the same terminology and get a biological equivalent to an electrical time constant. If there are large fish there is a big time constant; with algae, a small one. If the temperature is high, the metabolic rates are high, resistance is low which also provides a small time constant. Thus these quantities can be computed biologically to guide design of the actual electrical model. We have a simple

circuit like this that some of you have seen at some of the meetings for doing the carbon cycle. The circuit may provide a model of behavior of the element and may also show tracer behavior.

One of the points Shepperd makes very strongly is that if you add a tracer to a system which is already in steady-state, the isotope catches up with the carrier exponentially. If we have condensers with the capacity in scale and we program the pulse wave of the isotope injection, such as in a release from the Clinch River, the temporal curves in the electrical model as the condensers become charged will mimic isotope catch-up, since one has already set the resistances and the capacities on the basis of steady-state real data. On the assumptions that the fluxes are determined by the concentrations and are related linearly, the charges simulating isotope will move through this electrical circuit with the same peaks, patterns, storages, and time constants as that which will take place in the natural environment according to this theory.

The advantages of the circuit representations are: (1) present both concentration and rate data, (2) compare several systems, (3) compare elements in the same system, and (4) construct an electrical analog for prediction of temporal patterns.

What is needed to test the generality of the approach? We need to take many data on each system and put them into a circuit diagram testing the uncertainty concerning linearity and verify the behavior of the system in as many places as we can. It may be desirable to use a microcosm method with small replicate systems.

With enough knowledge of one system we can integrate circuits, species, the main compartments, the total system metabolism data, the classification typing, and the data on each radionuclide.

Suppose you want to compare, as we did yesterday, fresh water and marine systems for their handling of an element as in the case of strontium in San Antonio Bay. Strontium in water would be a very large compartment in the marine system in comparison to fresh water because of the higher concentration of strontium about 9 parts per million in the sea water. If you had set up an ecosystem circuit for fresh water and you wanted to predict what that would do in salt water you might change your condenser to a larger capacitance for strontium and see what it would do. Similarly, a circuit that is fairly general for ecosystems as a whole may be adapted to the particular case based on biomass and metabolic data.

FIGURE TITLES

- Fig. 1. Processes of photosynthesis, circulation, and respiration in a turtle grass bed expressed in three ways: (a) carbon concentrations and flow rates; (b) zinc concentrations after Parker (1962); (c) electrical analog in which coulombs represent carbon atoms.
- Fig. 2. Principal processes and fluxes in any ecosystem as defined by abstract boundaries.
- Fig. 3. Diurnal graph of oxygen and carbon metabolism in balanced aquaria from Beyers (1963).
- Fig. 4. A closed blue-green algal mat ecosystem adapted from a marine prototype situation affected by bleedwater.
- Fig. 5. Patterns of carbon metabolism, pH, and oxidation potential in the blue-green mat system in Fig. 4. Voltages are given both for an open circuit situation (b) and for an external resistance load with maximum power drain (c).
- Fig. 6. Comparison of diurnal curve for oxygen in enclosures over turtle grass with that outside after Odum and Wilson (1962).
- Fig. 7. Four year record of gross photosynthesis and system metabolism in the Laguna Madre of Texas (from Odum and Wilson, 1962).
- Fig. 8. Four types of annual metabolic patterns.
- Fig. 9. A classification of steady state ecosystem types according to combinations of boundary flux.
- Fig. 10. A classification of successional ecosystem types according to combinations of initial and final states affecting P and R.

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DISCUSSION

KAUFMAN: How large a sample are you examining for oxygen transfer?

ODUM: In the enclosure we have water one meter deep and we are taking 125 cc water samples.

KAUFMAN: There is circulation within this system?

ODUM: That's right.

KAUFMAN: Is it induced or just convection?

ODUM: In some of our smaller tanks we have added a pump to circulate it; otherwise it stratifies. In the large enclosure, and the ones in the field, there has been adequate circulation. The wind and bottom heating by the sun keep waters turning over. An ecosystem has P and R connected by circulation; often there is overlooked a tremendous physical energy of waves, current or streams in getting the nutrients released from photosynthesis over to respiration and the products of respiration back to plants. When circulation stops the whole thing slows down and the animals and plants are no longer adapted. They die and there is developed a reduced or abbreviated system that is an ecosystem which may not be related to the one in which you are interested.

BELTER: Have you actually done any of this for strontium or cesium or any other of the isotopes? I take it you have looked at zinc and phosphorus.

ODUM: We've got an actual electrical circuit scaled only for carbon. We have made a calculation for zinc and phosphorus from real data. We have some data for strontium but it is just not complete enough for the systems we have metabolic carbon data, also. We should do these things.

BELTER: This is just a calculation on zinc and phosphorus?

ODUM: Figure 7 has numbers for carbon and zinc from analysis and has flux rates for the carbon. There were a series of zinc tracer experiments done also, but the analyses of the curves are not yet complete.

NELSON: How much of that zinc is actually available?

ODUM: It's very hard to tell.

NELSON: Usually when you add a tracer you add so few atoms of your radioactive one that unless you put in a lot it is hard to detect by chemical methods.

ODUM: That's what one would imagine, but there was a rapid steady-state flux but with a tiny concentration in the water.

KAUFMAN: How do you accommodate the fact that when the storage capacitance is large, transfer takes place out of storage over a considerable period of time and during this time your transfer rates, or your current, is going to vary? Now, how do you accommodate for these non-steady state, non-climax considerations in your transport element of this model?

ODUM: Well, we talked about two things. One was the behavior of the carrier element in postulating that the concentration in each compartment controls the rate through an analog of Ohm's Law. The other was the catch-up of a tracer to steady-state model with capacitancies scaled according to the system. If you change the time constant of the carrier system you are not in climax.

KAUFMAN: Let's say that remains constant but for seasonal reasons. In variations of solar input you do have changes in your transport?

ODUM: Right; now where does the energy source come into this?

Well, in this particular system it's in the photosynthetic process which raises the voltage, raises the concentration and the energy value of a carbon cycle and other cycles coupled with it. In the carbon cycle everything is down hill from there on. That would not be true in a system where it is dominated by clays moving through a stream with not much biota. What's your energy source there? It's a physical transport mechanism and I think you will have to add a separate voltage source for the transport energy. In other words, put your voltage where the energy is. When we start to make circuits for the elements which are mostly piggyback riders then this may be power inputs in several places.

KAUFMAN: But if this varies, and you know what it is, you could simply introduce it in your model.

ODUM: I hope so. If you have a situation where you want the concentration to affect rate without losing your storage you can put in an operational amplifier to make the voltage not changed due to drainage. This may be necessary.

SPOFFORD: What do you do in the case where you don't have linear relationships between the parameters in your model?

ODUM: I might ask what about ten other sciences would we do if we couldn't assume linearity?

SPOFFORD: Aren't non-linear resistors available for the simulation of non-linear relationships?

ODUM: They are.

SPOFFORD: I know that one of the universities in the Boston area has a very specific analog computer for the solution of hydraulic networks. This employs tubes whose resistance increases with the 1.85 power of the current.

ODUM: It seems like you could work a system out linearly first, see where it falls down and then change it. This kind of thinking does give you another hypothesis to keep testing. In sanitary engineering you've known for a long, long time that the sewage would decay logarithmically. You could plot a semi-log and get a rate constant out of it. It's characteristic for sewage as a universal medium all over the world for a given temperature. The question that has been raised for the microbiologist is what about the death curve, shall we say, for the organic biomass that is not in sewage. I might also add that Jenny and Olson proved that soil did this also. So, that where you get a mixture of many species, "micros" mostly mixed in with the clay and all consumed, you apparently get this. What happens, for example, with a bunch of tubificid worms or clams? What is their decay rate? The circuit method now permits us to combine population ecology which up to now has been with a digital method. Replacing plots of death curves and life tables of numbers of individuals, we may be able to improve that science by replacing it with a biomass decay curve and life table of the biomass.

SPOFFORD: Wuhrmann, in Zurich, Switzerland, has studied the biological decay rates of various substrates individually. It appears to him that not only do the various substrates have different decay rates but that these decay rates are constant with time. He, therefore, postulates that the biological decay of sewage, which is usually assumed as a first order reaction, is actually a series of linear relationships superimposed on one another.

ODUM: We asked if that was the case. Byers has a number of runs in his microcosm. When you turn the lights off the respiration, and this has the higher plants, animals, rotifers and things, does seem to follow this curve during the night. We have had to give up in our curve analysis; we no longer assume respiration constants. That was the first approximation. Now we compute diffusion separately and then read the respiration. This is a very worthwhile subject.

SERATA: It seems to me that capacitance should also change with a change in the peak.

ODUM: We faced that. If you've got a steady state, that means your biomass is a constant then your capacitance is a constant because you have so much clay and so many animals and you put so much zinc into it before it levels off, and so on. The question is what do you do when you are growing "critters." You add a little photosynthesis and then the cell divides and you've

got 2 of them and then 4 and then 8 and then 16 not only have you got more circuits but you have a lower resistance in such a problem because you are adding capacitance for every new animal and you have a new capacitance for zinc. The solution was a very exciting thing. In any trophic level, there is storage, and conductance, and respiration-conductance to ground. Photosynthetic fuel comes into the trophic first as stored organic matter then as reproduction. So we run a feed back (we are still doing this manually I might add) to linearly lower the resistance in proportion to the condenser voltage. Behold, you get the logistic equation for population growth so that you can derive the basic ideas of population ecology from this circuit based on food concentrations without ever mentioning a birth rate. So, this is an exciting, separate subject. In what I presented today, I stayed off of this by sticking to steady-state.

KAUFMAN: This comes closest to what I was wondering about.

ODUM: The only time of consideration we mentioned a while ago was the special case of studying the flux of a small tracer to an existing carrier where the tracer amount was too small to affect the concentration and where the carrier was in steady state. In that event your capacitances are fixed and your conductors are fixed and that actually is the problem you are faced with. You are adding tracers that really are not changing the concentration gradients. In ecology we are interested in this other problem and we need a little more electrical engineering to give us the electrical feed back for this. I don't believe you need it for solving the kind of problem we had at this conference.

NELSON: Something disturbs me about your considering the energy available to organisms due to water current which you mentioned several times. It looks like to me that if you have current you have certain types of organisms and they are particularly adapted to this situation and if you don't have current you have other organisms so how can you quantify this current factor as far as the feeding behavior or organisms is concerned?

ODUM: If you have had enough evolution, natural selection and migration of species (and in microcosm we make sure it gets in by just seeding them) you have present those which have become specialized to increase their metabolic rate-settings in exchange for having the oxygen, CO_2 , and food brought to them. They are adapted to make maximum profit by it. If you did not bring it to them they may die because they have given up their alternative ability; they have got to have that concentration gradient provided for them. Does that answer the question?

NELSON: The thing that worries me is that maybe they give up energy in one aspect and use it in another. If you can consider this from the standpoint of adding energy to your system, this is all tied up in the metabolism of the "beast" itself and the transfer of energy of flowing water to the biological system.

ODUM: To give you another example, this is very much like the agricultural situation in the U.S. I couldn't give you proof but we say we are growing potatoes and corn on light. Yet figure up the energy that has been supplemented by man in tractors, weeding chemicals, and in breeding resistant varieties. The supplements permit agricultural plant varieties to drop out some of its normal wild functions, with energy drain, such disease resistances and weed resistances and specialize in a way that would eliminate them immediately without supplements. By the agricultural industry focusing all that energy in specializing, we are supplying energy through fossil energy. When you eat a potato I suspect it might be 90% fossil energy and 10% sun's energy. We are kidding ourselves about progress in agriculture because the minute we stop subsidizing it with tractors, with agriculture stations, with fertilizers, with weeding agents and all the other things we basically pay for by the fuel machines, from civilization as a whole, we will no longer have this yield; the same goes for these organisms.

NELSON: Is this a good analogy? Take China where coolies carry the rice; you are still driving on the same productive capacity. We have a higher standard of living because we can draw on these fossil fuels and divert our energy in other directions.

ODUM: Let's assume the animal on the rock has increased his metabolism rather than expending that energy on diffusion shell gradients which it must pay for. If you ride a tractor aren't you taking energy into your activity so that whatever you do you are no longer completely on your metabolic energy?

NELSON: That is true. But, we have a long period of adaptive evolution in this whole thing.

ODUM: You have got to have a circuit or there is no evolution; with no information from the past there is no evolution.

SPOFFORD: Could you give me some idea why you chose an analog computer over a digital, when it appears that you have a severe restriction; namely, you have to depend on the development of the proper electrical device in order to simulate the interrelationships of the system variates?

ODUM: I might add that Neil Armstrong, who is a graduate student on our program is a computer programmer over on the CDC 1604. One of his ultimate aims is to program the digital model of this once we understand it. This is our thinking device here. You were asking about what happens in reproduction. Well you could not decide the approach with a pack of cards. The circuit helped us convert biological thinking consistent with structure into theory without going through the math until after the answer was found.

STRONTIUM UPTAKE BY ALGAE

by JOHN H. AUSTIN

I. INTRODUCTION

OBJECTIVES OF STUDY

With the ever increasing use of radionuclides in industry, medicine, and power production, the radionuclide load on the environment will be continually increasing. The assimilative capacity of the aquatic environment will play a major role in determining the limits for the release of radionuclides to the environment.

The general objective of this study was to relate the assimilative capacity of natural waters to the environmental factors influencing the uptake of radionuclides by aquatic organisms at various trophic levels. By a systematic study of the passage of radionuclides from one trophic level to the next, it should be possible to ascertain the potential hazard to man.

MPC LIMITATIONS

Maximum permissible concentrations (MPC) of radionuclides in air and water have been adopted by the International Commission on Radiological Protection (1) and the United States National Committee on Radiation Protection (2). These standards are based on certain premises which must not be overlooked in the evaluation of low level waste disposal to the environment. The allowable body burden is based on the amount of the radionuclide in question which is deposited in the entire body and produces the maximum permissible dose rate to the particular body organ of interest (1). The MPC's for water are based on the intake by the standard man during 50 years of occupational exposure. Usually several organs are considered in order to determine the critical organ. Both soluble and insoluble materials are investigated. Values are given for a 40-hour work week and continuous exposure throughout the week. The MPC values for water may be applied to food, but consideration must be given to the amount of food so contaminated and the period of time over which the food was consumed.

Under many circumstances these MPC limitations have been interpreted to mean that surface waters could receive radionuclides up to the MPC level

for the particular radionuclide, or some fraction of it if the general population were the critical population. Thus, the average man would not receive excessive radiation over his lifetime if the average amount of water was taken in each day. The concentration of radionuclides at various steps in aquatic food chains is not often considered. A water at or below the MPC value for a particular radionuclide may be of no concern as far as drinking water is involved, but could possibly pose serious problems due to concentration processes which take place in the hydrosphere.

FOOD CHAIN CRITERIA

The setting of permissible daily discharge levels for radioactivity in England has on several occasions been controlled by the uptake by aquatic organisms. At Windscale (3) detailed investigations showed that the critical population was composed of the consumers of the seaweed Porphyra. The discharge was controlled so that the concentration of the activity by Porphyra would not cause the allowable critical organ concentration in man to be exceeded. At Winfrith the critical population was the lobster fishermen and their families and the critical organism the lobster. Thus, the discharge was based on such factors as the dose received by the fishermen in handling fishing gear, and the consumption of lobsters by the fishermen and their families.

In fresh water systems the concentration of nuclides by aquatic forms or by irrigated plants could be the controlling factor in setting discharge activity concentrations from nuclear installations. Under some circumstances large activity concentrations might be allowed in cases where the concentrating organisms are outside a food chain leading to man. However, this must be verified by suitable studies.

SCOPE OF INVESTIGATION

Many elements are worthy of consideration in a study of the passage of radionuclides through aquatic food chains to man; however, strontium-90, of major concern because of its bone-seeking properties and its presence in fallout, was chosen for use in this investigation. Studies have been under way for some time on the uptake of strontium-90 by higher forms of life, but little has been done in way of a systematic appraisal of the environmental factors affecting strontium uptake at lower trophic levels. This investigation was undertaken to study some of these factors as they affect strontium uptake by a member of the first trophic level, the unicellular alga Chlorella pyrenoidosa, and by gaining an understanding of the mechanisms involved in the accumulation and uptake to propose a relationship relating the strontium activity of the cells to environmental parameters. In particular, the main objective of the investigation was to ascertain the significance of calcium and magnesium on the mechanism and the extent of strontium uptake. The work was carried out on continuous algal cultures so that constant temperature, light, flow rates, pH, ionic composition of medium, and biomass could be maintained.

II. REVIEW OF PREVIOUS WORK

The role of calcium in the metabolism of Chlorella pyrenoidosa is not well defined, although a number of investigators have studied the calcium requirement. Scott (4) reported that calcium was not an element essential for the growth of Chlorella pyrenoidosa, but that it was taken up. Also, magnesium was required for the synthesis of chlorophyll, and an increase in the calcium to magnesium ratio in the medium gave an increase in the calcium uptake by the cell. On the other hand, Walker (5) showed that calcium and/or strontium was required by Chlorella pyrenoidosa. He reported that strontium could replace calcium in most, if not all, of its functions in the cell. Knauss and Porter (6) have shown similarities in strontium and calcium uptake by Chlorella pyrenoidosa.

Lowman (7) suggested that the uptake and retention of radioactive materials by marine plankton are determined by the residence time of the organisms in the contaminated water, the chemical form of the various contaminants, isotopic dilution, planktonic requirement for specific elements, and the ability of divalent cations and multivalent anions to complex with biological surfaces. Corcoran and Kimball (8) reported that strontium uptake by open-sea phytoplankton was an adsorption process, since the accumulation of strontium in heat-killed cells was equal to that in living cells.

Krumholz and Foster (9) indicate that the principal mode of accumulation of radiomaterials by fish is through the food chain. Williams and Pickering (10) have shown the importance of food chain transmission in bluegill fingerlings.

In summarizing the literature, it appears that strontium is sorbed on the cells and that the extent of the sorption is dependent in some way on the ionic composition of the medium. The nature and extent of this sorption is important, since food chain transmission of strontium has been shown to be the major mechanism of uptake under certain circumstances.

III. LABORATORY INVESTIGATION

PARAMETERS UNDER CONSIDERATION

The work of Scott (4) and others has revealed the interactions of various divalent ions such as magnesium, calcium, and strontium with cells of Chlorella pyrenoidosa. The main objective of this investigation was to ascertain the significance of calcium and magnesium on the mechanism and extent of strontium uptake by Chlorella pyrenoidosa in order to estimate the potential hazard of strontium being transmitted to man via the aquatic food chain. The temperature, light intensity, flow rate, and biomass were held constant while various

combinations of calcium and magnesium concentrations were used in the medium.

EQUIPMENT

The apparatus, shown in Figures 1 and 2, consisted essentially of four culture units, which were operated in parallel. Each unit was independent of the others in regard to rate of feed. The units could be fed from the same or different feed reservoirs. They were aerated from the same gas source. Figure 1 shows diagrammatically the continuous flow culture apparatus for one culture unit. The culture tube, component 2 in the figure, held approximately one liter of algae suspension. Recirculating water, 9, from a refrigerating-heating unit, 10, flowed through a cooling tube, 8, to maintain a constant temperature, $\pm 0.5^{\circ}\text{C}$.

Each unit was fed from two feed reservoirs, 18 and 19, identical in all respects except that one reservoir contained strontium-90. A Sigma metering pump, 16, was used to pump liquid into the culture units. An automatic timer, 17, controlled the operation of the pump (see Figure 1).

The cultures were aerated continuously with a mixture of air and carbon dioxide from two cylinders of compressed gas, 26 and 27.

Using sterile technique, algae from a stock culture on a refrigerated agar slant was transferred to one liter of sterile algae medium. The bottle was incubated at $23 \pm 1^{\circ}\text{C}$ in the light of a daylight fluorescent lamp for about 10 days. Then the culture was transferred, using sterile technique, to the continuous culture tube, 2.

The air- CO_2 mixture was bubbled through the culture tube without the continuous addition of new medium to the system for several days. When the cell population approached the desired level, the pumping system was put into operation and the continuous flow study was begun. The biomass of algae was allowed to come to an equilibrium value and then the feed was changed to the reservoir containing the strontium. This feed was continued until the strontium reached an equilibrium value in the culture and this level was maintained for several days. Then the feed was switched back to the strontium-free reservoir and the elution of strontium from the culture was followed. A residence time of about three days was used in these studies. The algae titer ranged between 10^7 and 10^8 cells/ml.

From the literature it appeared that strontium was sorbed by the algae from the liquid phase in a relatively exchangeable manner and little, if any, strontium was fixed in the cell. From this it was possible to propose that the rate of exchange of strontium for other ions on the cell was rapid and controlled by the rate at which exchangeable ions were presented to the cell surface and not by a rate-limiting metabolic process. In other words, for a given ionic composition of medium, the hydraulic flow rate determines the rate of exchange. Furthermore, it appeared that the algal cells could be compared to

1. CONTINUOUS CULTURE
2. CULTURE TUBE
3. CULTURE OVERFLOW
4. EFFLUENT COLLECTION FLASK
5. ALTERNATE OPENING FOR EFFLUENT COLLECTION
6. ALCOHOL-FILLED TEST TUBE
7. FLUORESCENT LIGHT
8. COOLING TUBE
9. RECIRCULATED WATER
10. REFRIGERATING HEATING UNIT
11. CULTURE DRAIN
12. EMERGENCY COLLECTION BEAKER
13. GAS-INFLUENT INJECTION
14. OPENING FOR INFLUENT SAMPLING
15. BACKFLOW CHECK VALVE
16. METERING PUMP
17. PUMP CYCLE TIMER
18. STERILE ALGAE MEDIUM
19. STERILE RADIOACTIVE ALGAE MEDIUM
20. OPENING FOR RADIONUCLIDE ADDITION TO MEDIUM
21. INFLUENT SIPHON
22. AIR OPENING
23. COTTON FILTER
24. ROTAMETER
25. PRESSURE REGULATOR
26. CARBON DIOXIDE SUPPLY
27. COMPRESSED AIR SUPPLY
28. FLOW REGULATOR

FIGURE 1. AUTOMATIC CONTINUOUS FLOW ALGAL CULTURE APPARATUS

FIGURE 2. AUTOMATIC CONTINUOUS ALGAL CULTURE SYSTEM — FRONT VIEW

FIGURE 3. AUTOMATIC CONTINUOUS ALGAL CULTURE SYSTEM — VIEW OF RESERVOIRS AND METERING PUMPS

FIGURE 4. VARIATION OF STRONTIUM-90 ACTIVITY IN ALGAE WITH TIME FOR DIFFERENT GROWTH CONDITIONS

FIGURE 5. COMPARISON OF STRONTIUM-90 SORPTION BY ALGAE GROWN IN MEDIA OF DIFFERENT CALCIUM AND MAGNESIUM CONCENTRATIONS

ion exchange resins and the selectivity coefficients and distribution coefficients of ion exchange theory would be useful tools in estimating equilibrium activity concentrations of the cell. The following equation can be used:

From this the selectivity coefficient $K_{\text{Ca}}^{\text{Sr}}$ would be:

$$K^{\text{Sr}} = \left(\frac{q}{C} \right)_{\text{Sr}} \left(\frac{C}{q} \right)_{\text{Ca}}$$

where

q_{Sr} = concentration of Sr in algae, cpm/mg

C_{Sr} = concentration of Sr in liquid phase, cpm/ml

q_{Ca} = concentration of Ca in algae, mg/mg

C_{Ca} = concentration of Ca in liquid phase, mg/ml

$$\left(k_d \right)_{\text{Sr}} = \left(\frac{q}{C} \right)_{\text{Sr}}$$

$$\left(k_d \right)_{\text{Ca}} = \left(\frac{q}{C} \right)_{\text{Ca}}$$

The results of the experimental investigations substantiated the hypothesis of the readily exchangeable nature of the strontium sorbed by Chlorella pyrenoidosa and demonstrated the necessity of considering the major divalent ionic species; for example, calcium and magnesium, in the estimation of strontium sorption by algae.

The highest rate of exchange for the continuous culture used in these experiments was that hydraulic flow rate which just prevented the cells from being washed out of the system faster than they were replaced by new growth. In a system where the cells were held in place, such as on a membrane filter, exchange rates with half times of less than one minute were obtained (cf. Table I).

The significance and applicability of the findings of this investigation can be demonstrated by comparing the conditions under which the experiments were carried out to those in nature. Although temperature, light intensity, flow rate, composition of media, etc., were carefully controlled, the continuous flow cultures do approximate conditions in a flowing body of water. It has been found from laboratory investigations (11) that a water containing greater than 0.05 mg/l magnesium will support a growth of Chlorella pyrenoidosa. As the

TABLE I
REMOVAL OF ACTIVITY FROM ALGAL CELLS
DEPOSITED ON MEMBRANE FILTER

Rinse Water Calcium Concentration mg/l	Contact Time sec	Percentage Activity Removed
0	15	30.8
4.9	25	49.6
19.6	27	58.5

TABLE II
COMPARISON OF CALCIUM AND MAGNESIUM CONCENTRATIONS
IN SEVERAL SURFACE WATERS

Cation	Concentration in River, mg/l				
	Mississippi Holden, Louisiana ^a	Columbia Rufus, Oregon ^b	Sacramento Sacramento, California ^b	Colorado Austin, Texas ^a	Colorado Yuma, Arizona ^b
Magnesium	0.8	6.4	7	12	41
Calcium	1.9	25	14	49	114

^aUSGS Water Supply Paper No. 1522 (15).

^bUSGS Water Supply Paper No. 1523 (16).

magnesium level increases to approximately 50 mg/l the growth increases. Beyond 50 mg/l there is no increase in growth. Of course, the required trace elements and nutrients must also be present at commensurate levels. Most natural surface waters fall in this range and thus would support *Chlorella* growths. Calcium is not required for the growth of *Chlorella pyrenoidosa*, but most natural surface waters do contain some calcium. Table II compares the calcium and magnesium concentrations of five surface waters in various parts of the country.

It is seen that these surface waters are all within the range to support *Chlorella* growths, assuming that they have the other nutrients required, such as potassium, phosphorus, nitrogen, etc. If the magnesium and calcium concentrations shown in Table II are compared with those of Table III, an estimate of the $(k_d)_{Sr}$ value may be obtained. From this comparison, one would expect the maximum sorption of strontium in a water similar to that of the Mississippi River and the least sorption in a water similar to that of the Colorado River at Yuma, Arizona. In all these circumstances, it must be remembered that not only are the algae involved in the assimilative capacity of the environment, but also other aquatic forms and both organic and inorganic sediments. Thus, given the same activity concentration, the amount of radioactivity sorbed by algal cells in nature would be expected to be lower than that sorbed by algal cells in the pure cultures of the laboratory.

The cells demonstrated a slight preference for calcium over strontium in that the mass action constant K_{Ca}^{Sr} , ranged from 0.53 to 0.62 in these studies. The discrimination against strontium found in these studies is similar to results found by other investigators. Table IV indicates a three-fold difference in discrimination factors (D.F.'s). Various methods have been used in determining the available strontium in the precursor, and part of the variation in the D.F.'s may be due to this.

Also, strontium and calcium are used in different ways in the various acceptors listed; and the availability of the various forms of strontium may be different for algae, higher plants, and bone, so that one would not expect the same discrimination factor in all systems.

This study has shown that the continuous flow apparatus is a useful tool in maintaining steady state populations of *Chlorella pyrenoidosa*. However, as would be expected, the maintenance of a constant biomass becomes progressively more difficult as environmental conditions deviate from optimum growth conditions. The membrane filter technique for measuring the biomass of algae in the system facilitates control of the system and also provides a simple and expedient technique for relating strontium uptake to biomass of the organisms. This technique shows promise of being a useful tool in the determination of the nature of the processes occurring at the cell surface, such as the sorption process, the antagonistic effect of competing ions, etc. Furthermore, this method should be useful in determining the exchange capacities of natural

TABLE III
MAGNESIUM-CALCIUM-STRONTIUM EXCHANGE
ON CHLORELLA PYRENOIDOSA

Influent Concentration			$(k_d)_{Sr}$	$(k_d)_{Ca}$	K_{Ca}^{Sr}
Mg mg/l	Sr c/ml	Ca mg/l	ml/mg	ml/mg	
243	3.7×10^{-4}	0.00	50.5	--	--
		1.32	49.8	--	--
		4.73	33.7	--	--
		18.5	38.8	67.5	0.58
		35.5	33.5	54.2	0.62
49	3.6×10^{-4}	0.00	5,000	--	--
		0.94	3,083	--	--
		4.88	861	1,500	0.58
		19.6	250	480	0.53

TABLE IV
DISCRIMINATION FACTORS

Acceptor	Precursor	Discrimination	
		Factor	Reference
Corn	Soil	0.58	(12)
Oats	Soil	0.60	(12)
Cabbage	Soil	0.78	(12)
Soybean	Soil	0.79	(12)
Rat Bone	Non-milk diet	0.27	(13)
Rat Bone	Milk diet	0.57	(13)
Rat Bone	Milk and corn diet	0.45	(13)
Man Bone	Normal mixed diet	0.5	(14)
Man Bone	Normal mixed diet	0.25	(14)

sediments where limited quantities of the material are available.

The following conclusions can be drawn from the results of this study:

1) The equilibrium concentration of the strontium activity on the cells is related to the ionic composition of the medium. It is dependent not only on the concentration of the chemically similar element calcium, but also on the concentration of the divalent ion magnesium.

2) The use of such relationships as "observed ratio," "distribution function," or selectivity coefficient are only valid if the effect of other ions (such as magnesium in the case of strontium and calcium) is considered. The quantities $(k_d)_{Sr}$ and $(k_d)_{Ca}$ together with the other divalent ion concentrations are required in order to predict the extent to which strontium may be sorbed by Chlorella pyrenoidosa.

3) The sorption process for strontium is rapid and is not limited by a metabolic process.

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DISCUSSION

COWSER: Is there a difference in the effect of calcium-strontium compared to magnesium-strontium or did calcium and magnesium affect the K_d in a similar fashion?

AUSTIN: It is not possible to show the effect of calcium on strontium uptake in the absence of magnesium since magnesium is required for growth. It is needed for chlorophyll formation. At a high magnesium concentration (243 mg/l) the calcium had little effect on strontium uptake. Note that the values of $(K_d)_{Sr}$ in Table III range from 50.5 with no calcium to 33.5 with 35.5 mg/l of calcium. However at a magnesium concentration of 49 mg/l the calcium had a much greater effect on the strontium uptake. Again from Table III, with no calcium the $(K_d)_{Sr}$ was 5000 and at a calcium concentration of 19.6 mg/l, $(K_d)_{Sr}$ dropped to 250.

THOMAS: Do you have any other mathematical apparatus for formulating absorption and other mechanisms?

AUSTIN: We are still working on a mathematical model for the sorption mechanism.

THOMAS: Did you do your work originally with Dr. Eugene Harris?

AUSTIN: I was with Dr. Woodward at the USPHS Sanitary Engineering Center in Cincinnati.

ASPECTS OF BIOLOGICAL TRANSPORT OF PHOSPHORUS - 32 IN STREAMS

by FRANK HOOPER

Mr. Belter has dictated that this is a think session on his problem not on ours. Since I am living in the close shadow of Mr. Belter, as Cleopatra said to Mark Anthony, "I am not prone to argue." I have taken a study that is oriented toward quite a different purpose, namely just learning things about a stream "eco system" and what I have tried to do here is to pick out a few things that might be interesting to this engineering group. The area that we are working in is the biological transport and cycling of phosphorus in streams. We have been dosing (spiking) a stream with millicurie amounts of P^{32} and then watching what happens to this pulse of phosphorus in the stream system.

Figure 1 shows our experimental stream near Cheboygan, Michigan. The stream is in the northern part of the lower peninsula. We have been working on a section about 2 1/2 miles long.

You will see that some of the data refers to specific sections; we will talk about what has happened in these sections A, B, C, D and E. The spike was put in a 50 gallon drum and diluted with water. We released 23 millicuries over thirty minutes. This was about half the MPC level.

The procedure is to follow this pulse of phosphorus through the food chain. We first look at the initial distribution in the stream. We get this initial distribution by repeated sampling. The uptake can be calculated for various sections. One can determine the uptake per unit distance, or the percentage-uptake of what is available. One can actually go to the trouble of calculating how far downstream the molecules of P^{32} go before being taken up by the stream biota. The amount of phosphorus we are talking about here is small that we are not anywhere near the saturation point. The figures in Table I represent the average distance that the phosphorus moved before being cycled into the system. Where uptake is rapid it was 450 yards where uptake was very poor it was 4,000 to 5,000 yards. What you see in this tabulation are figures for 1958, 1959, and 1960. These figures then represent a capacity that the stream has to take up or to metabolize the phosphorus. The interesting thing about this is the spectacular differences in one part of the stream to the next. From Section A to Section B there is a tremendous difference. This reflects something about the environment. Look at 1959; the capacity to cycle phosphorus is quite uniform up and down the stream. In 1960 we again had a great difference from section to section. The total amount of material that left the area is very small.

EXPERIMENTAL AREA
T. 33 N., R. 3 W.
Cheboygan County, Michigan

① → Sampling Station

Figure 1

TABLE I

UPTAKE OF RADIOPHOSPHORUS BY SECTIONS OF THE WEST BRANCH OF THE STURGEON RIVER, 1958-1960. TWENTY-THREE MILLICURIES OF P^{32} ADDED EACH YEAR

Section (Stations) (flow cfs)	Length (yards)	1958				1959				1960			
		Uptake		Loss		Uptake		Loss		Uptake		Loss	
		Total	Per-	Half	Average	Total	Per-	Half	Average	Total	Per-	Half	Average
		(Micro-	cent-	dis-	dis-	(Micro-	cent-	dis-	dis-	(Micro-	cent-	dis-	dis-
		uries)	age ₁ /	tance	tance ₅ /	uries)	age ₁ /	tance	tance ₅ /	uries)	age ₁ /	tance	tance ₅ /
				D	D _x			D	D _x			D	D _x
				1/2	x			1/2	x			1/2	x
				(yards)	(yards)			(yards)	(yards)			(yards)	(yards)
A (2-5) (38.73)	550	16,189	70.4	312	450	6,382	27.8	1,155	1,667	10,996	47.3	586	847
B (5-8) (43.48)	450	625	9.2	3,122	4,504	4,084	24.5	1,114	1,608	479	4.0	7,804	11,236
C (8-12) (47.53)	1,550 ₂ /	1,225	24.7	3,093	5,525	9,080	72.4	832	1,201	6,021	52.2	1,453	2,096
D (12-14) (49.72)	600 ₃ /	2,634	53.1	636	917	962	27.9	1,260	1,818	982	17.8	2,081	3,003
E (14-16) (52.5)	2,100 ₄ /	1,803	77.5	1,084	1,565	...	...	...	...	3,085	68.2	1,828	1,267

₁/ Percentage of isotope entering a section that was taken up by the section.

₂/ 1,250 yards in 1958.

₃/ 700 yards in 1958.

₄/ 2,300 yards in 1958.

₅/ Average distance a P^{32} atom travels before loss.

About 95% was taken out in the 2 1/2 miles.

So much for distribution in the stream. All we know at this point is that there was a certain quantity of phosphorus deposited in various sections. Now where is this phosphorus when deposited? What is the state of the material? The major fraction is lost to the periphyton of the stream. We measure this by putting out artificial substrates which grow a crop of periphyton. We also make measurements of the periphyton attached to rocks. Measurements are also made of the uptake by aquatic plants. Our calculations indicate that by far the largest part of the phosphorus goes into the periphyton part of the community. Sediments pick up very little if any. As a matter of fact, there are very few organic sediments in the stream. Probably better than 90% goes into the periphyton attached to gravel and rocks. Some of it goes into filter feeders; however, the filter feeders have to get it from bacteria and other things that remove it from the water. However we find it very soon in the filter feeders.

Now what is the change in distribution with time? This is the next thing that we will study. Table II shows that the activity soon disappears in the water. Within an hour after spike has passed the phosphorus activity of the water cannot be measured. The only way to measure any residual recycled material is by using a biological method. This is done by exposing a substrate containing periphyton to the water and remove enough activity to be counted. It can be seen (Table II) that for a long long time after the spike has been released there is a small quantity of activity passing downstream and that this has varied tremendously in different years. This is activity left that has been recycled back into the water. This is a very sensitive way of studying the amount of residual activity coming downstream. Water activity is low but it continues to move downstream in this fashion.

The next question is can we see the movement of this pulse of activity through the food chain as a whole? Let us look at Figure 2 to get some information on this. Here is a caddis, two fish (brown trout and Cottus), a Mayfly, a fishfly, and filter feeders that live on the rock. One thing that we noticed right away is that these organisms have an activity curve which is characteristic of the species. Some of these curves are rather nicely reproduced. Question here is what do these peaks mean? Do they have any relationship to food activities? The peaks mean two things: they first represent the biological turnover, which is a function of the size of the animal. They have something to do with food habit. The invertebrate predators are late in showing maxima. Trout and a fish are also quite late. Therefore you can see that food habits have something to do with the shape of these activity curves. The maxima reflect the position of these organisms in the whole food web.

Figure 3 will show how we can integrate this all into a meaningful picture. Time in minutes has been plotted on a logarithmic scale to show the distribution of activity. First is the water activity maxima. Next you can see the periphyton is at a maximum, followed by filter feeders, browsers, detritus feeders and fish. The figure represents in a general way the flow of this pulse of phosphorus

TABLE II

LEVEL OF RE-CYCLED P^{32} AS MEASURED BY THE ACTIVITY DENSITY
OF PERIPHYTON EXPOSED TO THIS SOURCE OF ACTIVITY

Exposure in stream (hours after spike release)	Activity density (corrected counts/min/g)						
	1958			1959		1960	
	Station			Station		Station	
	3	8	12	3	8	8	12
1-4	...	...	...	60	80	...	...
1-24	...	...	...	95	200	510	840
1-96	...	...	...	180	200	...	...
1-168	...	...	...	100	250	...	...
1-288	...	...	...	...	...	610	100
120-288	...	310	616	...	...	...	...
288-456	284	447	321	...	...	...	...
288-576	...	...	...	...	...	50	60
456-624	268	318	357	...	...	...	...

TABLE III

PERCENTAGE OF WATER ACTIVITY IN SOLUBLE, PARTICULATE AND
ADSORBED FORMS FOLLOWING ADDITION OF SPIKES OF P^{32} IN 1959
AND 1960. SOLUBLE FRACTION IS THAT PASSING AN HA MILLIPORE
FILTER. ADSORBED FRACTION IS THAT RECOVERED BY WASHING
FILTER WITH 0.1

N. H Cl.

	Station 3		Station 5	Station 8		Station 14
	1959	1960	1960	1959	1960	1960
Soluble	12	65	48	47	81	67
Particulate	70	13	32	47	0	18
Adsorbed	18	22	20	6	19	15

STATION 8

Figure 2

TIME OF P³² ACTIVITY

Figure 3

FIGURE 4

TOTAL WATER ACTIVITY PASSING
SAMPLING STATIONS FOLLOWING
ADDITION OF P^{32} SPIKE

through the food chain. The figure shows that after about 40 days if we wish to know where this activity is located we should look for it in fish.

In Table III, we are concerned with the effects water conditions themselves might have upon cycling of this material. We see that from year to year this differed considerably. In 1959, at Station 3 near the place we introduced the material, a small part of it was in a soluble form with about 70% in the particulate form. The reverse was true in 1960; about 65% was in a soluble form and about 13% particulate. Obviously this is likely to have quite a difference on the way this material cycles through the food chain. We have two more years of data not shown on the figure. The elaborate fractionation I mentioned earlier was done in 1962.

We became interested in the effect of the size and state of material upon its cycling through the food chain and here this is what we found. The two years 1958 and 1960, (versus 1959), go together; they are very similar. Periphyton activity was very high (taking all the stations together) and filter feeders were also high. In 1959 periphyton was moderate - or low. Activity was low in fish in 1958 and 1960, and high in 1959. Detritus feeders were very low in 1958 and 1960 and high in 1959. Apparently differences in the state of the material accounted for some of these differences in the cycling through the food chain. In the years where we had a lot of activity in particulate form we had very high activities in the fish and in the years where we had most of it in the soluble form we found a much larger percentage in periphyton and a lower percentage in fish.

In the next experiment (1961) we introduced the material in a particulate form (as bacteria). We grew the bacteria so they picked up and held the phosphorus intercellularly. It was not absorbed but was very tightly biochemically fixed. What we saw in 1961 was not exactly comparable to either of these two years. We found very little activity in the food system. Most of it went right on down the stream (about 50%). Most of the activity that remained in the stream was in the filter feeders. Some of it dropped out as a sediment and was regenerated; however, this was a very, very small amount.

These rather simple-minded experiments tend to emphasize one thing, that is, that differences in the form of the phosphorus may direct the material into different food webs. Hence one must be a little cautious in statements about the path materials are cycled. Such subtle things as the state of the material make a big difference. Figure 4 shows a summary of all 4 years.

DISCUSSION

BELTER: What were some of the factors that caused the differences in the year 1959 over 1960?

HOOPER: This is speculative because we don't have the best information that we wanted but the thing that we think is happening is the fact that there had been a rain which flushed a lot of organic material out of bogs and the surrounding area. We first believed this to be bacteria. There was much more particulate matter in the water. Now (after our 1961 experiment) I don't believe that it was necessarily bacteria.

KRUMHOLZ: Did you have any isopods or amphipods or anything like that in the creek?

HOOPER: Yes, we had *Grammarus* Sp. at one station and we have some data on this species; however, we did not have it at all stations so it was not one of the organisms we studied intensively. We only worked on species present at all stations during the whole period. If an insect dies or flies away then you can not get an activity curve.

KRUMHOLZ: Where would you put *Grammarus* Sp. in the data that you have there on the board?

HOOPER: It's activity curve indicates that it is very close to a plant feeder, at least it is somewhere between a plant feeder and a detritus feeder, according to its peak activity. In judging these things you always have to consider the size of the animal. If you looked at the thing and it is pretty big then obviously the turnover time is going to be pretty long. You have to look at all organisms with that in mind.

THOMAS: Your half-distances from one reached to the next can change by order of magnitude. Can you tell us something about differences in the characteristics of the hydraulic radius of these reaches?

HOOPER: Actually these areas are quite similar. The kind of things that we suspect are shaded versus open area. We do know that one of those areas is an area which is very highly shaded. We were, in effect, mapping pools of phosphorus in the stream. So it would make a big difference in uptake

if one area had more phosphorus than in others.

THOMAS: It depends on the slope of the curve doesn't it?

HOOPER: It is independent of concentration. The physical stream factors are fairly uniform, I would say. Biological factors and the physical factors such as light and these kind of things give us our trouble.

KAUFMAN: If you were to repeat that first experiment the following night, what would you expect the differences to be in terms of the uptake and releases by the smaller organisms?

HOOPER: From judging how much of this activity is actually inside the cell versus how much is absorbed we wash this off and better than half of it actually gets inside the cell. So some of this loss is an absorptive loss. I think it would be a big difference, actually.

KAUFMAN: I wonder if you couldn't use P^{32} turnover in order to measure the organism activity as Goldman at Davis is using Carbon-14 to measure the productivity?

HOOPER: I wish I knew if phosphorus was equated with energy. If I knew that every bit of this phosphorus was tied up with a certain package of energy this would be lovely. We could study energy as well as phosphorus. However, at this time I am a little bit conservative.

NELSON: Suppose in the 1959 situation, the rain had carried in some available phosphorus from the water shed. Did you make some stable phosphorus measurements here?

HOOPER: Yes, that's the thing. The stable phosphorus in this stream does not vary. It is almost a uniform day in and day out, this is all ground water and we can't get any variation. As a matter of fact it is so uniform that it is disgusting. This is the reason we went to isotopes. It was obvious that something was happening and we could not study the system by measuring stable phosphorus.

KRUMHOLZ: Maybe this is a constant that you can use to determine everything else by.

HOOPER: A night versus day run would be a good experiment. I agree that uptake would be proportional concentration. Whether curves are linear or not is a good question.

NELSON: Does your activity always go straight downstream?

HOOPER: No, under certain conditions it might go only 500 feet before you lose all your activity. The bigger the phosphorus pool in the stream the

more P^{32} is going to exchange with the stable phosphorus.

NELSON: Yes, but suppose you start with a continuous discharge of phosphorus.

HOOPER: If you did that you would still be exchanging with the stable phosphorus and it wouldn't go anywhere. The more stable phosphorus you have the more quickly you get rid of it.

KAUFMAN: I would like to ask Dr. Thomas a related question here. You have the results of a transient P^{32} release on this stream showing the build-up and decay of various elements. If a continuous supply of P^{32} were released would you expect a build-up in these organisms? Would it be the integral of the individual transients?

THOMAS: The build-up would be determined primarily by the total mass rather than by characteristics of the transients.

HOOPER: There are several other experiments we would like to run; however, it is quite a bit of trouble to get all the data that we actually need. Working with the food chain as a whole becomes very laborious. We can do some of these things very simply if they do not require all of the biological sampling.

KAUFMAN: Aren't your sampling errors enormous and is this probably one of your problems?

HOOPER: In the case of the bugs you must pool a lot of specimens to get a reliable sample. A hot bug here and bug there will make a large variation. You would have to have organisms that are going to stay in the stream the whole period and are not going to fly away. Also, they must not reproduce too fast. If you pick up something that wasn't in the stream when the spike was introduced, it's no good. If you get some that drifts down from upstream and therefore was not there when the spike passed this causes some difficulty.

FRIEND: What were you counting here?

HOOPER: We counted beta. We used a coincidence counter last year which was real nice. Unfortunately when testing was resumed our background went up and we didn't gain much precision even with our new \$9,000 counter.

SERATA: How many spikes were introduced each year?

HOOPER: One spike per year. This is all we could work out.

SERATA: Was this experiment repeated about the same time?

HOOPER: Yes, we tried to get everything uniform from year to year.

SERATA:
this stream?

What consideration was given to the hydrological conditions of

HOOPER: This spring-fed stream has a very stable flow; it doesn't vary.
Our stage was almost uniform from year to year. It varied from 37 cfs upstream
to 60 cfs at the sampling station farthest downstream.

BIOLOGICAL TRANSFER OF NATURAL AND FALLOUT RADIONUCLIDES IN STREAMS^{1/}

by LOUIS A. KRUMHOLZ^{2/}

I am just going to talk about what we are doing in a small stream which is about 38 miles southwest of the University of Louisville. We are working with no kind of radioactivity, but with all kinds of radioactivity - the natural radioactivity in the soils and the fallout that has accumulated and passed through the system in past years. We selected this stream, called Doe Run, because it was a small stream that could be very easily handled. It is about 9 1/2 miles long and flows directly into the Ohio River and has only one large tributary called Blue Spring that is about a mile long. The entire drainage basin is pretty well demarcated by the surrounding hills. Doe Run rises as a torrent spring with a flow ranging from 20 to 600 cfs depending upon a local rain storms and showers. The modal flow probably is around 20 to 25 cfs. The stream is about 30 feet wide over its entire length; however, there is only one thing wrong with our selection of a stream. We should have selected one that was only a half-mile long, then we think then we could have gotten some really good data. The principal difficulty is that the stream is 9 miles too long. See Figure 1.

We have spent many, many hours collecting samples of biota, and have carried out water chemistry tests an average of twice a month since October 1959. We have selected five stations on the stream, one at the source and others at various locations downstream depending upon the area, i. e. smooth sand bottom, high riffle area, or in a place where there are intermittent and almost continuous small cataracts from 6 inches to 2 feet in height. A whole series of these falls extends perhaps for a quarter of a mile. We have identified some 140 kinds of plants and animals in the area, and have attempted to follow the passage of the radioactive materials from the fallout and the natural radioactivity as they pass through this whole series of ecosystems, if we are going to use the same terms as Dr. Odum used this morning because in his

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² Contribution No. 63 (New Series) from the Department of Biology, University of Louisville, Louisville 8, Kentucky.

FIGURE 1

MAP OF DOE RUN, MEADE COUNTY, KENTUCKY, SHOWING TRIBUTARIES, STREAM MILES, COLLECTING STATIONS, AND OTHER FEATURES. THE BROKEN LINES INDICATE

INTERMITTENT STREAMS, THE SOLID LINES, PERMANENT STREAMS. INSERT A SHOWS THE LOCATION OF MEADE COUNTY. INSERT B SHOWS LOCATIONS OF WEATHER STATIONS AND GEOGRAPHIC RELATIONS OF STUDY AREA WITH LOUISVILLE

opinion virtually any little group of plants or animal is an entire ecosystem in itself. Each of the stations selected probably is considered an ecosystem in itself.

As Dr. Hooper mentioned, in order to get a good sample of an animal, so that you can analyze the degree of activity, you have to collect a great many individuals. This means that you collect a sample of the various kinds of organisms and then pick it over for a couple of hours in order to obtain perhaps 2/3 of a gram or a full gram of organisms to use for analyses. Some of these organisms were analyzed through the courtesy of the Taft Sanitary Engineering Center of the U. S. P. H. S. at Cincinnati. They were analyzed on a 256-channel gamma spectrometer and there were good measurable amounts of uranium-238 and thorium-232. There were also good measurable amounts of cesium-137, zirconium-95, ruthenium-106, total rare earths as indicated by cerium-144, and so on. Because of the tremendous cost of having analysis made and because of the almost impossible situation of getting enough animals to analyze we settled on analyses for strontium-90 and cesium-137 by radiochemistry.

Among the first things we found out was that we do not know what a atrophic level is, and I am quite certain that the level cannot be defined as clearly as it is in the books. The reason I say this is that the only one that can be rigorously defined is the primary producer. That is the organism that can use the energy from the sun and by the process of photosynthesis can produce a carbohydrate, or protein, or some other organic compound. This is the one that makes the food on which the entire ecosystem depends.

We found that allochthonous detritus was much more important than we had anticipated and that a very great amount of the nutrient materials was contributed by the leaves from the surrounding trees that had fallen into the stream. Such leaves fall into the stream, some of them sink, and others float downstream and pile up into rafts in the bends of the stream, they rot, they sink to the bottom, and the bacteria attack them and eventually they are changed into a completely different chemical form than when they entered the stream. Such material probably is one of the principle sources of nutrients in the stream and probably amounts to as much as 40 to 60% of the total nutrient materials.

Now, let me give you some idea of the magnitude of the radioactivity in some selected organisms. There are algae, mosses, rooted aquatic plants and a great many different kinds of invertebrate animals which range in size from microscopic up to the largest crayfishes. In addition, there are many species of fishes, some of which are closely related to the Cottus that Dr. Hooper mentioned. Radioactivity is accumulated by the various organisms throughout the length of the stream without any pattern of accumulation when the total aspect of the stream is considered.

Doe Run starts as a spring and the first half-mile is very interesting. There is a flat, level sand bottom without much on the floor except fine sand and a good bit of algae and moss. The moss is Fissidens the algae is mostly

Vaucheria and there are millions of snails of the genus Goniobasis. These are essentially the basic organisms there except the animals living in the Fissidens and the algae, the most abundant one is an endemic species of isopod of the genus Asellus. One of the interesting things is the growth form of Fissidens. It grows like the shingles on a roof except there is a water space in between the leaves. There are micro-currents within the moss beds, and we found, on the basis of micro-sampling, that there is about one or two orders of magnitude of difference in the oxygen content at the surface of the bed compared with that deep within the bed. The same applies for carbon dioxide, and other chemicals. This presents a whole area of different micro-climates and Asellus, the largest specimen we have seen is about 6 millimeters long, live in and around this moss bed. The largest sample contained about 14,400 per square foot. These little animals seem to live solely on the periphyton. They do not use the plants, they use the diatoms that live on the plants. So, here we have the plants supporting the diatoms and the diatoms supporting the Asellus. There are other animals that eat the Asellus and so on through the food web to the fishes.

We selected the food web leading to Cottus, and here are some figures on the amounts of the radioactivity. The water, and these are data from 1960-1961, before the Russian tests began: about 10 $\mu\text{c}/\text{l}$ of water total radioactivity and this gradually lessened from 1959 until August 1961 and then it increased following the Russian tests and at times there has been as much as 30 to 40 $\mu\text{c}/\text{l}$ total radioactivity. A good bit of this radioactivity comes in following sudden rain storms when there is a high silt burden in the stream.

The plants, that is the Fissidens and Vaucheria, will accumulate radioactivity, and these again are 1960 data, to somewhere between 10 to 25 $\mu\text{c}/\text{gram}$. Remember that the water was 10 $\mu\text{c}/\text{l}$ per liter this is 10 to 25 $\mu\text{c}/\text{gram}$ or about 1,000 times as much. Then the isopods were accumulating radionuclides from the diatoms living on the Fissidens up to about 5 $\mu\text{c}/\text{gram}$, and the amphipods of the genus Gammarus about 4 to 5 $\mu\text{c}/\text{gram}$. There were two crayfishes living in the stream, an omnivorous one in the upper reaches that will eat almost anything, and one farther downstream that is herbivorous which accumulates about 5 $\mu\text{c}/\text{gram}$ body tissue opposed to the omnivorous one with only 1 $\mu\text{c}/\text{gram}$ of body tissue in the whole organism.

Another thing I was talking about just a moment ago - this series made up of Fissidens, Vaucheria, Goniobasis and Asellus - below this region there is a whole series of pure stands of aquatic plants. First is a pure stand of Potamogeton and this grades off very abruptly into a pure stand of Nitella. Below Dam 1 there is an almost pure stand of Myriophyllum. A pure stand of one plant is replaced by a pure stand of another within 50 feet and there is no visible difference in the bottom of the stream and it is very difficult to see the difference on the basis of sand-particle size or anything like that. Asellus lives in almost all of these plants; however, they are most abundant in the Fissidens. Further downstream there are a great many other plants and animals. The two most abundant plants to the person who might walk up to the stream are the two European importations the watercress and the forget-me-not. These two plants concentrate large

amounts of radioactivity, but little is known about the physiology of watercress and the forget-me-not. In 1959 and 1960, they were carrying about 20 to 25 μc /gram of tissue, wet weight.

Following the beginning of the Russian tests, and the continuing American and Russian testing that has gone on intermittently since September 1961, there has been somewhere around a ten- to fifty-fold increase in the amounts of radioactivity accumulated by these organisms. These samples have been analyzed for Sr^{90} and Cs^{137} . In 1960 and 1961, any where from about 10 to 30 or 40% of the radioactivity in these organisms was fallout materials as indicated by strontium and cesium. In 1961, for which we have not yet received a radiochemical analyses on strontium and cesium, we must assume that the amount of natural radioactivity is the same and that the increase is attributable to increased fallout materials. This means that where we were having anywhere from 10 to 40% of the total radioactivity in the organism it is now anywhere from 50 to 60 or maybe even 80 to 90% of the total radioactivity. One of the reasons for this explanation is that so far no one has yet demonstrated that the transuranic elements enter into the normal metabolic processes of organisms; they are not used as nutrient materials in any way. Any occurrence of them in a plant or animal I think is pretty much incidental. I think the principal thing we have found out is that all the organisms in Doe Run have the data, but I just have not had time to work it up. We are accumulating and retaining certain amounts of fallout radionuclides. Such accumulations are for specific nuclides for each organism and the tenure of radionuclides in the stream biota depends largely on the biology of the organisms concerned.

DISCUSSION

KAUFMAN: Do you have any idea as to how the radioactivity will vary in the water as compared to the length of the stream?

KRUMHOLZ: No.

KAUFMAN: Would you expect the strontium and cesium to be the same at the head of that stream as the tail end?

KRUMHOLZ: We have no reason to think otherwise.

I should mention that this is a karsted area. Within two miles of the source of Doe Run there are more than a hundred sink holes. It is part of the Indiana, Kentucky, Tennessee, St. Louis Limestone of Mississippian origin that Mamouth Cave lies in, caves in southern Indian, caves in Kentucky; this is a big cave area that extends from Bloomington, Indiana down into probably the area east of Nashville, Tennessee.

KAUFMAN: Do you have a large number of water analyses that have been carried out for these isotopes?

KRUMHOLZ: I think about 50. I think the average of 10 μmc that I gave you was on about 50 samples. We have been sampling these various areas for water, plants, and so on, every two weeks since 1959 to get these data and we are finding them difficult to analyze.

KAUFMAN: This would be interesting from the standpoint of ground water contamination.

KRUMHOLZ: In this area, if we look at the source of Doe Run and it flows north, with weather stations at Irvington, Ft. Knox, Elizabethtown, and Louisville, there is just no correlation between the rainfall and weather at any one of these stations, with the volume of flow at the source.

In the source area there are sink holes. These sink holes obviously lead into the aquifer which feeds the source of Doe Run. Native residents of the area have told us that when we get a rainfall at Big Springs, the source at Doe Run will rise 13 hours later, and this really happens. When you

get a heavy shower at Big Springs between 12-1/2 and 13 hours later the water starts to rise in the source of Doe Run. Where this water goes in the meantime I don't have the foggiest notion. The source of Doe Run, a cave, collapsed since we started to work. Fortunately, none of us were in it. As a matter of fact we wouldn't go in the thing because it looked too rotten. The cave collapsed in March or April 1960, I believe. Now this water coming out through these interstices. Following a heavy rain one time Dr. Minckley and I were cruising the stream to see what we could find. There had been a 2-inch rainfall the night before and a 4-inch rainfall the afternoon before, local showers in a very small area, covering maybe a 1/2 mile in diameter. While we were walking down the side of the stream, we could hear water roaring but we didn't know where it was coming from. As we rounded a small hill we came upon an area we had categorized as a dry sinkhole, but water was boiling up through this hole to a height of about 3 feet. Of course, Doe Run was running very high because the Rushing River was full. We have never before or since seen water come out of this hole and my guess is that this underground pattern through here is almost constantly changing, and at times the aquifers feeding Doe Run are just a series of caves and when they get filled up the water comes out the most convenient opening. There is another hole near the source that in high water will contribute to the flow at the source. If we had it to do over again we would not select this stream for study.

THOMAS: You remarked, did you not, that the stream was too long and if you had it to do over again you would concentrate your efforts in one-half mile?

KRUMHOLZ: No. I would pick a stream only one-half mile long.

THOMAS: Am I correct in saying the reason that you would do this would be to get correlation between your samples.

KRUMHOLZ: No. There are just too many things going on in 9 miles of stream where there is a limited group like we have to work on. We believe now that we probably could handle a stream one-half to three-quarters of a mile long.

THOMAS: You have gained valuable information. The lack of correlations for the 9 mile stream is in itself important knowledge.

KRUMHOLZ: Yes, we have had some help on that. Some developers from Louisville were interested in that area and bought some land just above the flood plain at the Ohio River. There is a chemical factory owned by Olin Mathieson that operates right near the source of Doe Run. These developers impounded the lower end of Doe Run, and there is now a lake about 4 miles long in this narrow valley. They built this lake to supply fresh water to Olin Mathieson. In order to seal the caves in the area they put in several million yards of concrete in the floor of that valley. It is a beautiful location but the nature of the aquifers have certainly been changed with all that concrete grouted into the floor of the valley just above where they built the dam.

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RELATIONSHIPS BETWEEN THE CONCENTRATION OF RADIONUCLIDES IN COLUMBIA RIVER WATER AND FISH

by RICHARD F. FOSTER

The few brief remarks which I have are by no means in the form of a prepared presentation. I did bring a few slides. This is not strictly new information by any means. I have been somewhat guided by Dr. Gloyna's agenda and I have tried to pick a few things that might fit into this ecological section. There have been several other presentations here which indicate that a bit of information on the broader programmatic aspects of what goes on at Hanford might be in order.

As a broad frame of reference, which we can later narrow down into one of my favorite subjects, namely fish, there are two basic approaches which one can take in making studies on streams or aquatic forms. We have seen samples of both of these today. One of these involves the radiological aspects. Such things as potential exposure to humans, or hazard potentials, if you will. We can also include in the radiological aspects some of the potential effect on aquatic organisms and communities. Quite often our incentive to initiate studies is motivated by these radiological aspects. There is, however, quite a different incentive which involves radioactive materials. This concerns studies which are of a more basic or fundamental nature, and involve mineral metabolism, productivity, community structure, the cycling of minerals through streams, and so forth. Obviously, these are not mutually exclusive by any stretch of the imagination and they often lead nearly to the same kind of specific studies somewhere along the line. One may start out with the radiological aspects. Once the cream has been skimmed off the surface, however, you begin to look under the surface and at ways and means of control. Then you get into fundamental kinds of studies.

If one is working in a laboratory or on a small stream a single tracer is usually introduced. You know what you had put in so it's rather simple to look for this isotope, and often it is simple to count. As Dr. Nielsen pointed out yesterday, the Columbia River is tagged with some 100 or more different kinds of radionuclides. Throughout the entire scope of studies in which we are engaged it is important that we separate the isotopes by radiochemical techniques or spectrometric techniques so that we know each individual material which is present and its relation to the whole. This obviously involves quite elaborate and complex analytical problems which one does not have when dealing with simple laboratory tests. I think it goes without saying that virtually all of the work which has gone on at Hanford has been oriented toward the radiological

aspects. We do put out a considerable amount of radioactive material both in liquid effluents and in gaseous effluents. Some of our problems are associated with the Columbia River, but not all of them.

Our organizational structure tends to be more along disciplinary lines rather than to have an organizational component which deals exclusively with all kinds of Columbia River problems. For this reason, we have at Hanford, at the present time, some three or four different organizational groups that are involved in the work on the Columbia River in some way. We also have at Hanford the mathematical capability for assistance in complex mathematical models. This is our Applied Mathematics group, which is of great assistance in some areas such as transfer mechanisms.

We also have some work which relates to the Columbia River being carried out by the major component which operates the reactors -- the Irradiation Processing Department. These people are interested in two aspects: One, they use Columbia River water as a coolant; therefore, they are very interested in the quantity and quality of the water which is available to the reactors. Secondly, they are concerned with what materials are discharged from the reactors back into the Columbia River.

My group, the Environmental Studies and Evaluations Operation, is attached to the Radiation Protection component. Our job principally involves the measurement of environmental contamination, and the interpretation of this in terms of its potential effect on exposure to people. The Columbia River is one of the major areas which we study. This particular group also provides a central point where all information which relates to environmental exposure can be consolidated. Between the several groups, the communication system is quite short, quite informal, and I think quite effective.

Historically, we began studying the Columbia River many years ago. We have been interested in the radioactive effluent from the reactors going into the Columbia River because of its potential effect on aquatic life as well as on human exposure. Some of the first things we tried to do in relation to aquatic life was to find out what kinds of organisms in the Columbia River were most radioactive, which radionuclides tended to concentrate in the organisms and which ones did not. We also tried to determine the importance of direct uptake by the organisms versus that which might be taken up by foods. We were a bit naive in the early days and thought perhaps that all radioactive materials behaved in a similar manner. We started measuring gross beta activity and soon found that in fish this was principally due to radio-phosphorus. As some of the radiochemists, including Dr. Nielsen, led the way to more sophisticated radioanalysis we began to see a number of other nuclides in the organisms, which had been there all along but which we had not previously recognized.

In doing these early studies we recognized that it was not going to be possible to measure routinely the activity in all kinds of organisms throughout the entire Columbia River for the full 52 weeks of each year. So, we tried to mold

the program in such a way that certain aspects of this could be studies in breadth and others in depth. We attempted to cover all the major groups of organisms in the near stretch of the Columbia River and we selected certain key species that would allow us to follow the seasonal variations. In some species we followed the activity completely down the Columbia River. Some samples were taken in the ocean nearby. This early work gave us a good fix on the overall scope of the problem. The seasonal variations which we found in the organisms proved to be particularly interesting since we found that they did not follow the variations in the water very closely.

The reactor effluent water which is discharged into the Columbia River contains some hundred different radionuclides. On a routine surveillance basis we measure the more dominant nuclides, particularly those that have some significance in terms of potential radiation exposure to people. The first place downstream from the reactors that we measure the amounts of material in the Columbia River water is at Hanford. Figure 1 shows the relative abundance of radionuclides at Hanford Ferry. The total is about $6 \times 10^{-5} \mu\text{c/cc}$. Because of the length of time the river takes to reach Pasco, Washington, and since there is depletion in the river, as pointed out by Dr. Nielsen, the total activity is diminished considerably at this point -- (about $1.5 \times 10^{-5} \mu\text{c/cc}$). The shorter lived materials have tended to decay out. You can see that chromium-51 becomes dominant. As we go another 100 miles or so down the Columbia River to Vancouver, which is close to Portland, the total activity has diminished to about $2.4 \times 10^{-6} \mu\text{c/cc}$ and the chromium-51 has become more important. This situation represents only one point in time. This would indicate that it is a static situation, but this definitely is not the case. All of the concentrations are continually changing and the sizes of the diagrams are also changing.

If we look at this in terms of concentration of radioactive materials in river water at Pasco we can see that seasonally these things change markedly. This is shown in Figure 2. The change can be attributed to the difference in flow of water and it can also be attributed to variations in the rate of formation of the different nuclides in the reactors which is not the same throughout the year. We are interested in making more material balances in the river down to Vancouver in order to get a better concept of the total situation. Our routine sampling throughout the river system is on a frequency of weekly to biweekly and this information is quite useful for material balances, provided we have the radiochemical capability of measuring the small amounts that are present. The program has gradually developed as improvements have been made in the radiochemical capability of measuring the isotopes in the presence of all of the other materials. We look forward in the future of being able to estimate the inventory of nuclides in bottom sediments from routine analyses. We recognize from the problems that we had earlier in the game that a rather larger amount of sophisticated radiochemical analysis is required to accomplish such studies.

As a part of our routine surveillance program we have a continuous fish sampling program. In order to make some sense out of trends it is necessary to confine a pictorial presentation to one kind of fish taken from one section of

RELATIVE ABUNDANCE OF RADIONUCLIDES IN COLUMBIA RIVER WATER

Figure 1

CONCENTRATIONS OF SEVERAL RADIONUCLIDES IN COLUMBIA RIVER WATER AT PASCO, WASHINGTON

Figure 2

the river. Figure 3 shows the total beta, contributed almost entirely by phosphorus³² in the flesh of whitefish caught between Hanford Ferry and Richland. You will note there are major seasonal fluctuations. It appears that the peak occurs in October and November. If one wishes to follow long range trends we feel it is perhaps more useful to take the top of the peaks for successive years and compare these rather than trying to come up with some sort of an overall average number for the year.

The seasonal changes can be attributed to several things including changes in the concentration of radio-phosphorus in the water, and changes in the metabolic or feeding rate of the fish. The phosphorus comes principally from the food which the fish eat and additional variability results from changes in the life cycle or community structure of the food organisms.

Figure 4 shows the radionuclide composition of whitefish flesh and Columbia River water in the fall of 1961. You will notice that the phosphorus³² and zinc-65 are the most predominant nuclides. We have also identified sodium-24, cobalt-60, and potassium in the flesh. The background activity comes from potassium-40. From the relative size of the two diagrams (concentrations in the flesh versus that of water) represents a gross concentration. The small wedge representing the P³² in water should be compared to the in the whitefish flesh.

Figure 5 shows the rate of uptake and retention of phosphorus³² by trout tissue. This test was run several years ago. Phosphorus was fed to the fish continuously and this figure shows that they reached an equilibrium in 3 or 4 weeks. The point that I wish to show here is that the fish require some substantial number of days, even with P³², to reach an equilibrium. Once the feeding of the P³² has stopped, as one would predict, the level in the fish begins to drop off. There are two reasons why it would drop off. One, is due to the radioactive decay of the isotope and the other is that biological turnover can eliminate the isotope. The elimination curves for the bone and the muscle have the same slope as the radioactive decay of P³². The one for the liver is slightly more rapid.

Figure 6 indicates the concentration of P³² in the flesh of whitefish caught between Hanford Ferry and Richland.

Each point on Figure 6 represents a fish sample and the position of the lines represents a collection of fish at a particular location. Ordinarily, we try to get our collectors to pick up about 10 fish for a sample. Sometimes fishing is not good enough to produce this many. We feel it is necessary to sample at frequent intervals in order to follow the seasonal trends.

One would like to know what produces the seasonal variations and whether some control might be effective, such as the rate at which phosphorus is released to the river. Therefore, I would like to relate the level in the fish to the output of P³² from the reactors so that one might perhaps be able to predict

P³² IN WHITEFISH - RINGOLD TO RICHLAND

MAXIMUM AVERAGE CONCENTRATION

SEASONAL VARIATION

CLASSIFICATION _____
 CLASSIFIED BY _____
 DATE BY _____ DATE _____

Figure 3

RADIONUCLIDE COMPOSITION OF WHITEFISH FLESH AND COLUMBIA RIVER WATER. FALL, 1962

WHITEFISH FLESH
($1 \times 10^{-3} \mu\text{c/g}$)

WATER
($1.5 \times 10^{-5} \mu\text{c/cc}$)

Figure 4

UPTAKE AND RETENTION OF P³² BY TROUT TISSUES

Figure 5

CONCENTRATION OF P^{32} IN THE FLESH OF WHITEFISH CAUGHT BETWEEN HANFORD
FERRY AND RICHLAND

Figure 6

what the level in the fish might be with different release rates from the reactors. We recognize from the information that already has been presented that uptake by the fish involves a sluggish system. It requires a considerable time to get up to equilibrium level. So, there is not a static, stable system in terms of transfer coefficients or energy flows. We really have here a dynamic system. It occurred to me that we might pick any point on the curve and say that at that particular point in time a fish had radio-phosphorus present. If that fish, or a group of fish, did not feed or gain any new radio-phosphorus at all, then the level of radio-phosphorus in the fish would decline. From what we saw from the lab experiments it would decline at the rate of the radiological half-life of P^{32} . However, if after a week's time we know that the level did not drop off; or it might actually increase, this would indicate then that during the particular week the fish gained P^{32} -- at least enough needed to make up for the radio-active decay. Assuming that the average line drawn through the broad scatter was valid, we divided the data into weekly intervals. The incremental addition of P^{32} which the fish must have gained during that week in order to keep a fish on the curve was computed.

Figure 7 then shows the gain in phosphorus per unit time. In effect, it says from January through May that the new phosphorus being added to the fish is being added at a very slow rate. At several points it is essentially zero which means that the fish just did not get any new P^{32} during those weeks. From June through November, the rate at which the fish was incorporating the new P^{32} rose to slightly over 300 μmc of P^{32} per gram of fish per week. After November the rate declined but it did not come down quite as low as it had the previous January and February.

This particular curve has a feature one can strip out and that is the amount of P^{32} which is present in the water. We can take this curve and normalize it to what it would have been if the concentration of P^{32} in Columbia River water had been uniform throughout the year. We did that and came up with a slightly modified curve. Figure 8 shows the rate at which the fish would be gaining new P^{32} if the Columbia River had a uniform concentration of P^{32} throughout the year. The rate is very low from January through May, rises again during the summer and early fall, and then drops off to a low rate in the winter. The curve does not tell us why this happens. We then must begin to postulate what other factors are involved. From laboratory experiments we know pretty much that it has to be food. We have postulated in the past that it is pretty well associated with temperature which governs metabolic rate and feeding rate of fish, so I have plotted, on Figure 8, the water temperature curve. It appears from the temperature scale that above 12°C the rate of intake or deposition of P^{32} increased, and after the temperature dropped below 12°C the rate dropped off. However, it would appear that the rate of deposition is dependent upon a big "blackbox," an ecological box. Temperature is obviously a factor, but I don't think it is the only one. Also on Figure 8 you will note flow, but this particular year has had the flow factor stripped out of it. Flow can be an important feature here in changing the habitat area of the fish. Stream ecologists will recognize that there is bottom scouring during the spring freshet. There will be elimination

Figure 7

RATE OF DEPOSITION OF P^{32} RELATED TO RIVER STAGE AND TEMPERATURE

Figure 8

of a reasonably good feeding area until the flow is stabilized and a new food base is built up.

Figure 9 shows the specific activity of P^{32} in Columbia River organisms. This figure was made from information obtained several years ago. We know approximately the concentration of P^{31} in the Columbia River and we know the concentration of the radio-isotope; therefore, we derived this activity as micro-curies of P^{32} per gram of P^{31} . This is the label we put on P^{31} of the system. In the water it comes out to about 25; on net plankton also contains about 25. The numbers are about what you would expect if you had a rapid exchange between the systems. As we move up the food chain, the ecological cycle, the specific activity begins to drop down. When this was taken (in the fall) the activity in the fish was about 5. About three half lives have elapsed from the time isotope was "fresh" in the water and became "aged" in the fish. The phosphorus in the fish has a mean age of about six weeks.

Figure 10 shows the concentration of zinc-65 in the flesh of the whitefish. The same fish that were shown in Figure 6 are shown here for the zinc-65 concentration in the flesh. The total range of seasonal variation is not as great here as in P^{32} , as one would expect, because zinc-65 is longer in half-life. The range between the fish in the same sample is also not quite as great for this longer-life isotope. I would like to be able to run the same kind of analysis of increments for zinc as I did for the phosphorus. I cannot do it yet because we do not have the laboratory work finished which shows the rate at which zinc-65 is lost in the fish after equilibrium has been reached. One would expect the biological half-life would be a more dominant factor in diminishing the activity of zinc-65 than in the case of P^{32} .

Our sampling program also takes oysters from Willapa Bay, a salt water body outside of the Columbia River. Samples are received about every two weeks. We have established an arrangement with a good oyster grower who collects samples for us from the same beds. Figure 11 shows that the zinc-65 concentration in the oysters throughout the year has remained remarkably stable. We can also see the variation in the radio-phosphorus in these same oysters. The phosphorus level is not as stable as the zinc-65. It is interesting to me that both in 1961 and 1962, after the first of July, there was a decline in the phosphorus³². I would not speculate at all on this because the problem here is so complex. It involves the circulation of the Columbia River water into the sea, the fraction of this that gets into Willapa Bay, and the feeding habits of the oysters.

We have recently run experiments on the chronic feeding of strontium and yttrium-90 to trout. This is continuous feeding at three different levels. Those of you who are familiar with the levels of strontium will recognize that the high level here is a substantial amount. The fish weighed about 200 grams, as shown in Figure 12, and so some of these fish were getting 100 μ c or more of strontium each day. The object of the test was to find the burden of strontium in the fish that actually caused the damage. I show this to indicate that this kind of work

SPECIFIC ACTIVITY OF COLUMBIA RIVER ORGANISMS

NOTE: THESE VALUES WERE ESTIMATED FROM MATERIAL COLLECTED AT DIFFERENT TIMES AND ARE SUBJECT TO REVISION.

Figure 9

CONCENTRATION OF Zn^{65} IN THE FLESH OF WHITEFISH

Figure 10

CONCENTRATION OF RADIONUCLIDES IN OYSTERS FROM WILLAPA BAY

Figure 11

Effect of Chronic Feeding of $\text{Sr}^{90}\text{-Y}^{90}$ on Growth of Trout

Figure 12

Effect of Chronic Feeding of $\text{Sr}^{90}\text{-Y}^{90}$ on Mortality of Trout

Figure 13

is going on so that we can measure contamination levels in Columbia River fish and have something to compare them with. I hasten to point out that strontium-90 is not one of the things we have to contend with in the Columbia River, any more than in the other rivers of the country except for the Clinch. Figure 13 shows the effect of chronic feeding of strontium-90 and yttrium-90 on mortality of trout.

DISCUSSION

KRUMHOLZ: Which trout was that in Figure 6?

FOSTER: Rainbow trout.

KAUFMAN: How is the fish fed the P^{32} ?

FOSTER: It was in capsule form. The isotope was spiked in the fish's food in a known quantity, and the quantity fed was related to mass of the fish.

KAUFMAN: Was the fish hand fed?

FOSTER: Yes, hand fed.

KAUFMAN: Was the water then removed from around the fish so that it wasn't picking up any from this route or source relative to the water.

FOSTER: These are in hatchery troughs supplied with Columbia River water. This Columbia River water did have in it some small fraction of effluent from up-river reactors. The Laboratory was located downstream from the reactors so the amount of effluent present in the water in the troughs in which these fish were living is quite characteristic of the water to which the fish in the Columbia River are exposed. However, the water as it comes into the troughs does not have enough food materials to make any difference as far as the fish is concerned. At zero time for the experiment the fish had been in the river water for a number of weeks. Actually, before this particular experiment was started the Sr^{90} level was down below the base level in Figure 6. The level began going up as soon as we began feeding.

KAUFMAN: In Figure 6 was the data corrected for decay?

FOSTER: No, it is not corrected for decay. The rate at which the fish lost the P^{32} was almost exclusively radioactive decay, not biological elimination.

THOMAS: If you used both the physical half life and the biological half and if you put these two together (as in the formulation for MPC) do you then agree that the decay would be a check?

FOSTER: These data points represent the amount of phosphorus that remains in the fish for given times after the feeding was stopped. This is not a theoretical curve, it is an experimental curve.

KAUFMAN: If you corrected this for decay it would be the answer to Dr. Thomas' question.

FRIEND: What is happening in April and May in Figure 7?

FOSTER: This small hump in the curve seems to be real. If it had only been in one collection of fish for this particular time I would not have been inclined to draw the curve through the point. However, it does show up in other places in other kinds of fish and it is explainable on the basis that at about this time of the year the Columbia River water begins to warm up a bit so that the fish begin to feed. The concentration of P^{32} starts up until we get the spring flood. Then the river flow greatly dilutes the effluent water and lowers the concentration.

PARKER: What is the migration range of these fish in Figure 7?

FOSTER: We don't know how far these whitefish migrate; however we do know it is a number of miles. We have not tagged any fish but there are two indications of this: one, the dams in the Columbia River have fishways and routinely a count of the number of fish that go over the dams is made. Whitefish are one of the species that are counted, and there are substantial numbers that go up and down river over the dams. A dam is located upstream from the Hanford Reservation. Just below Priest Rapids Dam lies a favorite fishing spot for whitefish. As a matter of fact, this is one of the places where we sample for whitefish. The closest reactor is somewhere around five to seven miles downstream from here. The samples of whitefish that we get here contain some fish which have P^{32} . The numbers of fish which contain the P^{32} that we have obtained in this location varies throughout the year. It rises during the fall, October and November, and drops again and there is another little rise in the spring. This actually represents the fish from below the Hanford reactors that have moved up.

THOMAS: Is there a good biological reason why one fish should vary so much from one stretch to another?

FOSTER: Obviously there must be. From these factors we can postulate two things and both of them are related to food. One of these is that a "cold" fish may be feeding for a few days on food organisms that have lower amounts of phosphorus in them. Another possibility is that they may have been in a slightly different part of the river, perhaps up river from reactors, where the food organisms are in a less contaminated section of the river. You can superimpose on top of these variations, some individual variations between individuals that one might find in a laboratory experiment in which he knew that the fish were getting the same amount of phosphorus each day.

KENNEDY: What about the distribution of the radioactive elements in the fish? Would that tell you whether they had been upstream and then had come back down?

FOSTER: We actually tried this for one year by comparing the activity in the scales to that in the flesh, in the bone, in the liver, etc. We thought perhaps, that a fish collected at Priest Rapids might have most of the activity in the bone and not so much in the muscle. This method didn't work very well. This result might have been predicted from the laboratory experiment where P³² was fed to the fish. You notice that the flesh and the scale and the bone all went down at about the same rate. It was only the liver which had a slightly steeper curve or more rapid loss of phosphorus than the other tissues.

KENNEDY: What about the sodium versus the phosphorus?

FOSTER: Sodium is gained or lost so rapidly that it reflects only the location of the fish for a day or two prior to sampling.

KRUMHOLZ: At the Oak Ridge National Laboratory we had much the same experience, where we would take three fish out of the same net at the same time and we would have a 200 to 300% range among comparable tissues of the fish.

FOSTER: This is a problem that is built into the system - one which also influences estimates of the potential of exposure to people and complicates the situation. I don't know how you are going to resolve this except by realizing that you are going to have to sample a lot of fish.

KAUFMAN: Do you find the variation great downstream as you see here or closer to the site?

FOSTER: Yes, its great all over.

SERATA: I have a question related to Dr. Thomas' question. I imagine the metabolic rate and also the feeding habit of the fish would be quite different in the different ages. I wonder if you found any effect on the age of the fish?

FOSTER: We do know that the smaller fish tend to have a higher level P³² than the old fish. Beyond this I can't answer the question. It is a good assumption and we have assumed that the change with the age of the fish is due to the change in metabolic rates or feeding habits, I don't know for certain.

SERATA: Do you have samples for the various sizes of fish?

FOSTER: They are of various sizes but most all are in the "legal size" range. There are no very small ones. Most are all 8 inches or bigger.

BERNARD: But that wouldn't be the reason for the orders of magnitude difference in P³² level, would it?

FOSTER: No, this does not account for the wide range in the concentration between specimens of the same collection.

KAUFMAN: Did you say that curve of P^{32} uptake is probably proportional to the rate of fish growth throughout the year?

HOOPER: This is the same question that I was going to ask. I don't know about white fish but if you take a trout you probably have a maximum growth increment from about June to July then you have a fall off. Fall off occurs about the end of July is true but that extra additional growth in October and November is what bothers me.

FOSTER: You could relate this to temperature, again.

KAUFMAN: Even simpler; the P^{32} rate must be equal to the P^{31} rate, the P^{31} concentration in the fish is constant so you have a relationship with P^{32} as growth, haven't you?

HOOPER: But the fish doesn't follow that, Q. E. D.

FOSTER: I don't know enough about the growth rate of the whitefish in the river to make a x-y plot of these things; intuitively you would say the curves should look quite the same.

HOOPER: This second peak could represent any batch.

FOSTER: I think this is the key to the situation. The food which the fish is eating also has a dynamic change in the P^{32} concentration. This is the big "black box" which is left out of the situation because we don't know that much about it at the moment. What I had hoped would happen by stripping out the factor of concentration in the water and throwing in the temperature factor was that these two things would be so dominant that the food "box" would be less important. However, the data show that this is not the case.

KAUFMAN: May I ask you two questions. Would you agree that the P^{32} that you put in the river labels the P^{31} that is already there?

FOSTER: Yes.

KAUFMAN: Would you agree that there is a constant phosphorus content in the flesh of the fish?

FOSTER: Yes.

KAUFMAN: If we assume that the stable and radioactive phosphorus are present in constant concentrations and that the fish contain a constant percentage phosphorus, then, the rate of loss of phosphorus, either stable or radioactive must be proportional to the rate of fish growth. There are, of course, many

complicating considerations, but the underlying principle is a very simple one.

NELSON: When do the whitefish spawn?

FOSTER: They spawn in November, usually.

NELSON: A lot of pre-spawning organisms build up higher concentrations of phosphorus. I wonder if this in any way be related to your situation?

FOSTER: I think that spawning may enter into the total physiological response which could change the rate at which the "new" phosphorus was being taken up by the fish rather than any major shift of phosphorus from one site into sex products.

NELSON: When you go back to the equilibrium equation of yours and avarian growth, then the food consumption has to go up. If the fish doubled their food consumption, then they would also double their phosphorus equilibrium concentration.

KRUMHOLZ: He is talking about flesh, he is not talking about fish.

NELSON: Yes, but still he is going to have to increase his food consumption and the amount of phosphorus will be shifted around; this is hardly the black box. All the phosphorus is not going to be shunted into the ovary; you would expect some of it to get into other tissues.

FOSTER: You can postulate several things. All the new phosphorus being put in is not being converted into egg production or any one thing.

KRUMHOLZ: Do you know whether the same thing happens in Rainbows?

FOSTER: No, I don't know.

FRIEND: The experiments shown on Figures 13 and 14 must have become a radiation hazard.

FOSTER: This required a substantial amount of radiation control, i. e., short time limits; the fish were handled with lead gloves, these fish had to be dissected under water for shielding.

KRUMHOLZ: What were your criteria for growth, gain in weight?

FOSTER: This was one of the parameters. We also recorded mortality. The object of showing this figure was that at the two low levels we essentially saw no effect. The high level not only stopped the growth, but it also killed them.

KRUMHOLZ: Did you try anything on that to see if you were getting strontium

rickets on the basis of that much stable strontium?

FOSTER: Yes, we did follow up on some of these but we saw no bone tumors of this sort in connection with it.

KAUFMAN: That is a fair slug of strontium.

FOSTER: Yes, it points out that if the strontium concentration in the fish is kept to a level suitable for human food, you are going to keep the level orders of magnitude below that which caused injury to the fish in this experiment.

BELTER: You might indicate how many pounds of Willapa Bay oysters you would have to eat for those concentrations of 1961-62.

FOSTER: One pound of oysters per week would lead to a whole body dose of about 7 millirem per year. If you use a limit of 500 millirems per year as a maximum permissible dose, 1,800 pounds would just about give it to you. You could probably eat around 35 pounds per week or at least in this order of magnitude.

KAUFMAN: I think Odum's model would work very beautifully on your P³² and it looks like to me this would be a really good test on something like this.

BELTER: This would be a very good idea.

FOSTER: From this particular data I think you can use three basic compartments as far as the fish is concerned. One, the concentration of the water, the other being the temperature, and the third being the intake of food. In the model I think you could very nicely strip out what this concentration from food would have to be. We can from this, I believe, compute the grams of food that the fish would have had to eat each week, if we could identify the dominant food item of the fish. This is a more sophisticated treatment and I hope some of our biology friends can help solve it.

BELTER: We still have not begun to cover the subject of physical characteristics or parameters which are involved in the transport of radionuclides in water. I believe we have ample time left today in order to at least get started on this subject. We have several presentations scheduled on the hydro-dynamics of streams and Mr. Calvin Patterson from the University of Texas will discuss the general subject of diffusion first.

DISPERSION MEASUREMENTS

by CALVIN C. PATTERSON

This talk will be a description of dispersion tests that have been made here over the past 18 months. Before going into the details of the actual tests conducted a few words about the importance of dispersion in the transport of nuclides are in order.

First, dispersion determines the location and concentration at the point of maximum concentration for nuclides that remain in solution. The time-concentration curve of nuclides at points in the stream and particularly along the stream bottom is determined by the dispersion mechanism. Colloidal and floating microbial populations that have sorbed nuclides from the solution will also be dispersed by the same mechanism. The dispersion phenomenon is part of the mixing process that provides the intimate contact between nuclides and sorption media required for uptake.

An individual faced with the problem of judging the mixing characteristics of a stream has very little to go on at the present state of knowledge. The theory appears to be on firm footing. Solutions exist for various types of boundary conditions and turbulent fields. However, such solutions have constants or a constant in them, which is called by various parties the diffusion, dispersion, or mixing coefficient. These coefficients have been determined in laboratory scale experiments; however, very little data have been obtained in prototype streams.

To my knowledge, Parker's work in the Mohawk River, Thomas' work in the Chicago Ship Canal, Shapiro's work in the Ohio River, and Frederick's work in the Clinch River represent the sum total of diffusion data taken in upland streams. We feel there is a need for additional data from streams of different characteristics than those previously mentioned. We also feel that there is a need for additional data from streams of different characteristics than those previously mentioned. We also feel that there is a need to attempt to correlate laboratory and prototype data.

A section of the Colorado River about 20 miles southeast of Austin was chosen as a test site. It is one of the longest-straightest sections of the river in this area. See Figure 1. It has a fairly uniform depth as shown in Figure 2. Near the upper end the bottom is composed of sand and cobblestones. It changes to pure sand about 1/3 of the way down. The sand layer in this area is at least 3 feet deep. There are no ripples on the sand. Dunes appear 30 to 40+ feet long

DOWNSTREAM VIEW

UPSTREAM VIEW

COLORADO RIVER

Figure 1

Figure 2

with 0.5 ft. heights. What appears to be a pool is in reality a meander within the main channel that the stream assumes at low flow. Figure 3 is a cross-section, taken downstream of where the meander crosses the channel centerline. At this section the meander is near the right bank. The channel develops another deep spot along the left bank downstream of this section. This causes a slight lateral drift from right to left in the main channel.

Another stream chosen as a test site is the Pierce Canal. This canal is the main irrigation canal on the Pierce Ranch, at El Campo, Texas. This canal is near uniform in cross section, 33 feet wide at the bottom, with a depth of 3.5 feet, and side slopes one on one. Figure 4 shows upstream and downstream views of the canal. There is a very slight bend in the channel where the tree overhangs. Depth is maintained in the channel by means of a check shown on the slide.

Having selected sites for testing, the next problem was to collect the data. This required tagging a volume of river water with a suitable tracer, and determining the concentration of the tracer in the river as the mixing progresses. Fluorometric techniques were chosen using fluorescent dyes and a Turner fluorometer. The method was chosen because the dyes create no health hazard, the cost of tagging a large volume of water is small, and the detection is sensitive. Concentrations on the order of 0.2 parts per billion and 1 ppB are detectable for Rhodamine B and fluorescein, respectively.

Originally, sample collection was to be made in two ways. Stationary samplers were constructed such that they could be placed at known locations in the river and collect water samples at predetermined times. A portable continuous flow sampler was built so that it could be put in a boat. The boat was located at known points and river water was pumped through the fluorometer. A concentration-time curve can be obtained in this way from the fluorometer which is equipped with a recorder.

A stationary sampler is shown in Figure 5. It consists of a series of three weights which are released at five minute intervals by withdrawing a supporting pin. The pin is driven by a battery operated motor which is actuated by a clock rigged as a timer through the alarm system. As the weights are released, they draw a 20 ml. sample into a syringe. The whole system is supported on a 1 1/4 inch pipe.

The samples are taken instantaneously. This in itself is not good design, because the average concentration at a point is what one is interested in; and even under steady injection the tracer concentration at any point in the turbulent field is very much time dependent. Furthermore, operating troubles were encountered with this design. The support pipe must be placed in a nearly vertical position or the weights do not fall. It required about an hour per sampler to make a field set up. We tried leaving the support pipes in overnight to speed things up for the next day's test; however, floating weeds would collect on the support pipes and bend them about 20° out of vertical.

$Q = 3200 \text{ cfs}$

Figure 3

CROSS SECTION NUMBER ONE COLORADO RIVER

UPSTREAM VIEW

DOWNSTREAM VIEW

CHECK AT
END OF CHANNEL

PIERCE CANAL

Figure 4

STATIONARY SAMPLER

**MODIFIED STATIONARY
SAMPLER**

Figure 5

The samplers were redesigned so that the syringe was opened directly by the motor. These samples are superior to the others in view of the fact that they operate on an incline, they are light, and they withdraw a sample over a 5 minute interval.

The continuous flow apparatus is shown in Figure 6. It consists of a generator set, centrifugal pump, flowmeters, and fluorometer. Water is pumped at 5 gpm from the river. The valves are set so that 0.3 gpm is shunted through the fluorometer. The concentration of the tracer in the river water is read out of the fluorometer on a recorder. The performance of this apparatus is good. It was originally intended to use it for longitudinal dispersion only, but it will be used for transverse dispersion also.

The tracer injection apparatus consists of a tank of compressed nitrogen which is used to force the tracer solution out of a bottle through a hose into the river. 9 to 18 liters of dye solution can be introduced into the river in 10 seconds. This is assumed to be an instantaneous point source.

The test consists of introducing the tracer at a preselected point in the river, moving downstream to preselected points with the boat in the center of the tracer plume, and collecting the data read out of the fluorometer as the plume passes the boat. The boat is then moved to the next location and the process is repeated. If transverse data is required the plume can be criss-crossed with the boat.

Some typical data of longitudinal tests are presented in Figures 7 and 8. The first is taken in the Colorado. The plotted points are representative points taken from the recorder. The solid line on this figure was used to compute the dispersion coefficient. The next slide shows the Pierce Ranch data. In computing the coefficient, these curves are assumed to represent a concentration distance curve made at the instant the maximum concentration reaches the sampling point. Time on the recorder output was converted to distance using the mean velocity at the sampling point. The standard deviation of the curve was computed and the longitudinal dispersion coefficient was calculated using the formula:

$$D_1 = \frac{\sigma^2}{2T}$$

There is some interest here in whether or not a microbial system disperses in the same manner as a liquid system. A test was made recently in the Hydraulics lab at The University of Texas, which was designed to help answer this question. An algal bacterial population was grown in an aquarium and introduced into a one foot wide flume. Rhodamine B was used separately but under identical flow conditions to determine liquid dispersion.

The tracers were placed in the flume as continuous point sources. Vertical dispersion only was determined at a point 7 feet downstream of the release point.

CONTINUOUS FLOW APPARATUS

Figure 6

Microbial counts were made in a Levy counting chamber. Liquid dispersion was determined by fluorometry.

The results of this one preliminary experiment indicate that there is indeed a difference in the dispersion of the two systems. However, we feel that more experiments are necessary to make any strong conclusions in this regard. This line of experimentation will be continued.

This has been a brief look at our successes and failures in dispersion measurements. The job is not finished. A few additional longitudinal dispersion measurements are planned. A major effort in the Colorado will be given to transverse tests beginning this March. Flow in the Colorado is controlled by a series of dams beginning in Austin. Water is being conserved at present and the flow in the Colorado is too low to conduct the tests. Irrigation releases will begin in March.

Construction will begin this week on a 200 foot long flume at the University's Balcones Research Center. Among several experiments planned for this flume, we intend to hydraulically model the Pierce Canal and Colorado River and compare prototype and model dispersions.

In concluding this talk, I would like to say again that this type of data is important. Beyond measuring a parameter that is directly involved in transport equations, additional benefits may be obtained. What one really measures here is turbulence. It has already been shown that longitudinal dispersion correlates with oxygen reaeration. It has long been accepted that suspended sediment is related to the vertical diffusivity in a stream. I feel confident that the future will show many other items of more direct interest in nuclide transport will exhibit correlation with the turbulent mechanism of a stream.

DISCUSSION

FRIEND: What kind of tracer was used?

PATTERSON: We used fluorescent dyes, Rhodamine B and Fluorescein.

BELTER: How does this canal differ from the Chicago canal?

PATTERSON: I am not too well acquainted with the Chicago canal. I have only read Thomas' 1957 Progress Report and not his final report.

BELTER: That is, with reference to the uniform cross-section etc. ?

PATTERSON: The difference is in scale, certainly the depth, width, and probably the velocity.

BELTER: I just wondered if you had reviewed the Chicago work and how this differs?

YOTSUKURA: The Chicago canal is about 160 feet wide and 26 feet deep. It has very uniform cross-sections.

PATTERSON: It is excavated from rock isn't it?

YOTSUKURA: Yes, the sidewalls are; there may have been some surface irregularities.

In the first dispersion examination you found that the dispersion coefficient decreased in downstream direction. Is this because of the channel contracting?

PATTERSON: No, the channel wasn't contracting. I believe I had some dye going by that was below the sensitivity of the fluorometer. The trouble is you are not picking it up outside of the curve that you use to compute with. The reduction in dispersion at the second sampling point in Figure 7, I think, was a problem not having the boat positioned on the center line of the plume.

THOMAS: You know the Taylor Theory. Did you see whether your values compared with those are predicted by this theory?

PATTERSON: No, I haven't checked that.

THOMAS: Parker and I found something like an order of magnitude of the ratio between observed and predicted in the Mohawk investigation where there was a higher degree of irregularity in the channel.

PARKER: Wasn't it one or two orders of magnitude difference in natural channels?

FREDERICK: I have made some similar analyses on various straight reaches up to 13,000 feet long and on very crooked natural streams of about the same cross-sectional area, but of somewhat different configuration. For the straight reaches, the dispersion coefficient derived from field observations differed from the dispersion coefficient computed from Taylor's formula by a factor of only 2 to 4. In the natural streams, however, the difference between "observed" and "Taylor" dispersion coefficients was as much as a hundred.

BELTER: Is this on the Clinch River?

FREDERICK: This was on the Clinch River in Tennessee, the Coachella Canal in California, the Clinch River in Virginia and several other streams.

SAYRE: I am wondering that since you made your introduction from a point source and that since it took the slug on the order of minutes to go by the sampling station, how the concentration distribution of the dye across the canal might have varied with time?

PATTERSON: The centerline concentration is affected by transverse diffusion in proportion to $\frac{1}{\sqrt{Dyt}}$; however, this will not affect the variance of the curve.

SAYRE: I would think that the leading end of the slug coming by would be more concentrated at the center of the channel and the trailing end may have been more subjected to the lateral diffusion.

FREDERICK: I would say right off-hand that at the first station you didn't have anywhere near complete lateral dispersion or lateral mixing.

PATTERSON: Lateral dispersion did not cover more than 80% of the channel at the last sampling station, except for the Pierce Canal where lateral mixing was complete at both sampling stations.

FOSTER: Later this year we are planning a project to map the effluents from the Hanford Reactors in some detail using dye. Although our work here is not specifically for the testing of mathematical models, it will provide a good chance to try some of these out in a practical case. Carl Bennett in the Applied Mathematics group is quite interested in the mathematical model applications and he can help in this area. He is quite interested in knowing how some of these mathematical models may or may not prove out in a large size river.

REMARKS

by W. G. BELTER

As we mentioned yesterday, we hope to continue on this morning, possibly through the lunch hour period in order to insure that we will be able to complete the program. Some of you have indicated that you would like to visit the facilities at the University of Texas and Dr. Gloyna has agreed to provide escorts. Some of you also have indicated an interest in visiting the University of Texas Port Aransas Marine Institute. Dr. Odum and Dr. Gloyna have indicated that they will help provide transportation, leaving possibly sometime this afternoon.

As I look at the schedule for this morning, I note there is a place for the Summary at the end. I think in view of the many different disciplines involved in this meeting, that the way we would like to conduct this part of the conference, is to take the three or four sections that we have covered - the sections on chemical and mineral characteristics, mineralogy, sediment uptake, transport, and the last part on hydrodynamics - and have each of the participants in those groups think in terms of the questions that were posed at the beginning of the meeting. That is, 1) what we know now in these various areas, 2) what areas are we relatively weak in now or putting it another way what emphasis should our future studies take, and thirdly, is there anything we have done so far that does have application to other stream environments. At the end of the meeting I would like to throw it open for discussion and general comments by the various participants, to give us some idea of what you have obtained from this meeting and to also indicate, from what you have heard in the past two days, the direction you think our program should be going for the next several years.

So, without any further discussion on that point, I think we will get started this morning and hear from Dr. Yotsukura of the U. S. Geological Survey who will talk to us on turbulent dispersion of miscible materials in open channels.

TURBULENT DISPERSION OF MISCIBLE MATERIALS IN OPEN CHANNELS

by NOBUHIRO YOTSUKURA

INTRODUCTION

When the U. S. Geological Survey started its turbulent dispersion study in 1958, very little information was available on dispersion in natural streams. Of the published papers, the most helpful were the theoretical works by G. I. Taylor (1954) and R. E. Glover (1955), and the measurement of dispersion in the Mohawk River reported by F. L. Parker (1955), H. A. Thomas, Jr. (1956), and E. S. Simpson, W. A. Beetem and F. H. Ruggles (1958). Consequently, the Survey's tests in various rivers were rather preliminary in scope and were made to help develop a better understanding of the significance of dispersion coefficients and to establish the pattern of dispersion (R. G. Godfrey and B. J. Frederick, in press).

Concurrently other investigators produced a number of papers reporting dispersion measurements in natural streams. Although the theoretical development of equations describing open channel dispersion had not been very rapid, significant contributions were made by several investigators, namely, I. E. Thomas (1958), J. W. Elder (1959) and G. T. Orlob (1959). At the beginning of 1962, however, results of tests in natural streams clearly demonstrated that the existing theories were unsatisfactory and that a lot more needed to be known about the fundamentals of open channel dispersion. Thus, for the past year and a half, efforts have been devoted to basic theoretical work and laboratory tests of dispersion. A summary of this work which is limited to longitudinal dispersion following an instantaneous release of miscible material is presented in this paper. It is recognized that laboratory study of dispersion in steady uniform flow is significant, however, many factors need to be considered before applying the results of model studies to natural streams; some are: 1) the effects of cross-sectional variation (non-uniformity) and temporary storage in the river, 2) the effects of stream alignment (bends), 3) the effects of branch flow and flood waves (unsteadiness,) and 4) the effects of stratified flow due to temperature and density gradients.

REVIEW OF EXISTING THEORIES

Fundamental theory of turbulent diffusion was developed by G. I. Taylor in 1921. His equation for one-dimensional diffusion in an isotropic turbulence is

$$\sigma_x^2 = 2 \overline{u'^2} L_t t - 2 \overline{u'^2} \int_0^t \xi R(\xi) d\xi, \quad (1)$$

where σ_x^2 is the variance of diffused distance, $\overline{u'^2}$ the variance of turbulent velocity, L_t the time scale of turbulence, t the time coordinate and $R(\xi)$ is the Lagrangian correlation function. Because the distribution of diffused material should be Gaussian under such a condition,

$$C = \frac{G}{\sqrt{2\pi\sigma_x^2}} \exp \frac{-(x-\bar{x})^2}{2\sigma_x^2}, \quad (2)$$

where C is the concentration at distance x , $\bar{x}$ the mean position of diffused material (center of concentration cloud) and G is an arbitrary constant. These basic formulae can easily be adapted to the longitudinal dispersion in an open channel when the flow is steady and uniform. For the flow with mean velocity U , equation (2) is transformed into

$$C = \frac{M}{A \sqrt{2\pi\sigma_x^2}} \exp \frac{-(x-Ut)^2}{2\sigma_x^2}, \quad (3)$$

where M is the total amount of dispersed material and A is the cross sectional area of the flow. It is also seen that if the dispersion coefficient K is defined by

$$K = \frac{1}{2} \frac{d\sigma_x^2}{dt}, \quad (4)$$

then, from equation (1), K is equal to the product of intensity and scale of turbulence. For very large t ,

$$K \approx \frac{\sigma_x^2}{2t},$$

and equation (3) becomes identical with the Fickian distribution equation,

$$C = \frac{M}{A\sqrt{4\pi Kt}} \exp \frac{-(x-Ut)^2}{4Kt} \quad (5)$$

An associated theoretical problem is to predict the magnitude of K without going into the tedious measurements of $\overline{u'^2}$ and L_t . This work was undertaken by Taylor in 1954. Using the Reynolds analogy and the resulting dispersion equation, Taylor estimated that in a straight circular pipe,

$$\frac{K}{aU_*} = 10.1, \quad (6)$$

where a is the radius of the pipe and U_* is the mean shear velocity. Subsequently I. E. Thomas and Elder applied Taylor's method to a wide open channel and obtained, respectively,

$$\frac{K}{DU_*} = \left(\frac{U}{U_*}\right)^3 \Phi(n) + \left(\frac{U_*}{U}\right) \Psi(n), \quad (7)$$

and

$$\frac{K}{DU_*} = 5.93, \quad (8)$$

where D is the depth, $\Phi_{(n)}$ and $\Psi_{(n)}$ are constants depending on the Reynolds number of the flow. G. T. Orlob examined the lateral dispersion at the water surface in a channel by means of floating particles and could determine the pertinent scale of turbulence. Applying Kolmogoroff's similarity principle to the phenomenon, he concluded on the basis of experimental data that the lateral dispersion coefficient is

$$K_L = 0.0136 E^{1/3} L_x^{4/3}, \quad (9)$$

where E is the rate of energy dissipation and L_x is the Lagrangian length scale of turbulence.

THEORETICAL WORK

The Survey's work consisted of solving the dispersion equation by numerical method. The basic dispersion equation which is similar to that of Elder and I. E. Thomas is

$$\frac{\partial C}{\partial t} + u \frac{\partial C}{\partial x} = \frac{\partial}{\partial y} \left(k \frac{\partial C}{\partial y} \right), \quad (10)$$

where u is the velocity at depth y and k is the lateral diffusion coefficient. All variables are reduced to nondimensional quantities. Logarithmic velocity distribution was assumed in the y direction. In brief, the results of the computations show better agreement with experimental observations than the existing theories. For a flow with a high Reynolds number, the Longitudinal coefficient of dispersion, K , is about $14 DU_*$ and the skew coefficient is about unity at $x = 80D$. It was observed that K became almost constant at about $x = 80D$, and the center of the concentration cloud travelled with the mean velocity. However the maximum concentration decreased with $1/t$ rather than $1/\sqrt{t}$ and the skew, which was a maximum at $x = 20D$, decreased linearly with increasing x , showing that the dispersion at this point is not of a purely diffusive type. Presently computations are being extended to include points farther downstream to obtain the dispersion configuration which can be compared directly with experimental data. Also tests are planned at various Reynolds numbers to determine their effects on K .

LABORATORY WORK

DESCRIPTION OF THE FLUME TESTS

A rectangular flume at the National Bureau of Standards, Washington, D. C., about 150 feet long, 4 feet wide, and with side walls about 15 inches high is used in the laboratory studies. The flume has a recirculating system with a sump containing about two million gallons of water. Tests were run in an 18-inch wide sub-section. Three kinds of roughness were employed; 1) 0.4 mm sand glued on all boundaries, 2) 1/2-inch square butt laid across the bottom width with longitudinal spacing of 1 foot, and 3) 1-inch square butt with spacing of 5 feet. Miscible material was a mixture of hydrochloric acid and ethyl alcohol with specific gravity of unity. The conductivity probe was a pair of electrodes having circular surfaces of 5/32-inch diameter. The signals from the probe were fed into a galvanometer and the needle deflection was impressed continuously on a chart. Three probes were placed in downstream direction with a spacing of 36 feet. About 80 runs have been completed with various combinations of depth (2 inches to 8 inches), velocity, slope (0-0.01) and roughness. The data were processed on a Balgol 220 digital computer which computed the true concentrations from conductivity data as shown on the charts. The computer also determined the mean, the standard deviation, the skew and the total area of the concentration-time curve. Various quantities are defined in Figure 1.

ANALYSIS OF DATA: I

When the concentration of miscible material is observed with respect to time at a fixed point, x is fixed and equation (5) becomes

$$C(t) = \frac{M}{A\sqrt{4\pi Kt}} \exp \frac{-(X-Ut)^2}{4Kt} \quad (11)$$

By introducing two parameters

$$T = \frac{X}{U} \quad \text{and} \quad J = \frac{X U}{K}, \quad (12)$$

Fig.1 Definition sketch of concentration vs time curve observed at a fixed point

the quantities pertinent to the distribution function are rigorously derived as follows:

$$\int_0^{\infty} C(t) dt = \frac{M}{UA} \quad , \quad (13)$$

$$C_{\max}(t) = \frac{M}{AUT} \sqrt{\frac{J}{4\pi}} \left(\frac{J}{\sqrt{1+J^2}-1} \right)^{\frac{1}{2}} \exp \frac{-(J-\sqrt{1+J^2}+1)}{4\sqrt{J^2-1}} \quad ; \quad (14)$$

$$t_{\max} = \frac{\sqrt{1+J^2}-1}{J} \cdot T \quad ; \quad (15)$$

$$\bar{t} = \frac{J+2}{J} \cdot T \quad ; \quad (16)$$

$$\sigma_t^2 = \frac{2J+8}{J^2} \cdot T^2 \quad ; \quad \text{and} \quad (17)$$

$$\sqrt{\beta}_{1t} = \frac{\sqrt{2}(3J+16)}{(J+4)^{3/2}} \quad . \quad (18)$$

In order to check the flume data with the Fickian solution (5), equations (14), (17), and (18) were plotted in non-dimensional form. See Figures 2, 3, and 4. The solid lines in these graphs are expected Fickian curves. Using the dispersion coefficient determined by equation (4), some of the experimental data are superimposed on the curves. It is seen that, for smooth and sand roughened boundaries, the standard deviation σ_t/T converges to Fickian value at $J = 700$.

The maximum concentration, $\frac{C_{\max} AUT}{M}$, also converges to Fickian but with considerable scatter. As for the skew $\sqrt{\beta}_{1t}$ it is greatly in excess of Fickian value even at $J = 1000$, though it is converging gradually to the curve. The data from tests using butt roughness are not plotted in these figures, but they show much better agreement with the Fickian curves mainly because the turbulence created by the butts tends to be more uniform, and the pattern of dispersion approaches the conditions of isotropic diffusion. The standing waves usually associated with these roughness flows had no effect on the dispersion. From the data analysis, it is concluded that the Fickian distribution (5) is an asymptotic solution and not all properties of the Fickian solutions are satisfied in a flume test of reasonable dispersion time. By trying various methods of determining K , it is concluded that the best is to plot experimental σ_t^2 against t

Fig.2 Relationship between C_{max} and J

Fig. 3 Relationship between δ_t^J and J

Fig.4 Relationship between skew coefficient and J

and to read the slope. If the concentration curve is available only at a point, the determination should be done either by using equation (17) or by plotting log

$C \sqrt{t}$ against $\frac{(X-Ut)^2}{4t}$ and by reading the average slope. All other methods which are described in the literature tend to produce large errors unless the data used are from very long dispersion time, or from tests using an extremely rough boundary.

ANALYSIS OF DATA: II

As to the magnitude of K , it is convenient to use the nondimensional dispersion coefficient as proposed in the theoretical discussions. It is noticed that an extension of Taylor's result to an open channel, by substituting the hydraulic radius R for the pipe radius a , results in $\frac{K}{RU_*} = 20.2$. Flume data yielded

an average value of $\frac{K}{RU_*}$ of about 15 in a two-dimensional flow with sand

roughness. With data from all tests included, $\frac{K}{RU_*}$ ranged from 2 to 17, but

an attempt to correlate the magnitude of this nondimensional coefficient with hydraulic parameters was not successful. However, the correlation of K with the depth D and the rate of energy dissipation E had a fair degree of success. This correlation is in line with Orlob's concept even though the two flow situations are considerably different. Thus far, K has been observed to be proportional to $E^{1/3}$ but no proper measure of scale has been determined. Using the depth as a tentative scale, the proposed correlation was

$$K \propto D^m E^{1/3}, \quad (19)$$

where m varied from $4/3$ to $1/3$. As the tests are being extended to the 4-foot wide section of the flume, it is hoped that a better measure of scale will be obtained in consideration of depth, width, and roughness. As a quick reference, the magnitude of K in natural streams ranges from 1 to 1000 square feet per second whereas it ranges from 0.001 to 1 in experimental flumes. It is noted that the total range is much narrower than that in ocean dispersion because of the limiting effect of boundaries on the turbulence scales in open channel flow.

FUTURE PLANS

Because our ultimate goal is the prediction of dispersion pattern in natural streams, work is planned on the following items next year:

- 1) One dimensional theory for nonuniform flow by the use of equation

$$\frac{\partial(AC)}{\partial t} + \frac{\partial}{\partial x}(AUC) = \frac{\partial}{\partial x} \left(AK \frac{\partial C}{\partial x} \right) \quad (20)$$

- 2) Analog simulation of dispersion in a three dimensional channel
- 3) Correlation of existing stream data on the basis of equation (19) or its developed form, and
- 4) Tests of dispersion in a few streams where the uniform steady condition is realized.

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DISCUSSION

SAYRE: How did you introduce the material in the flow?

YOTSUKURA: We introduced the material by surface dumping. I was quite concerned about this but we could not find any other easy way of introducing the material. The device was a simple trough across the channel which dumped the material directly in.

SAYRE: We used the same method and I think for depths which are not large it appears to be quite good.

YOTSUKURA: We have not done too much work on checking which method of injection is good.

PARKER: You will remember, Dr. Yotsukura, that when we ran our tests in your flume we did put some dye in to see what distance the salts extended and it wasn't too bad.

YOTSUKURA: Of the measuring stations located every 36 feet, the last station is 108 feet downstream and the effect of initial injection disappears at this station. Therefore, we feel we can get a fairly good idea of what is going on.

KAUFMAN: Why do you feel that you will never get the Fickian solution in flume tests unless you introduce a very rough bottom?

YOTSUKURA: In a smooth or sand-roughened channel with reasonable flow depth and slope, it takes exceptionally long distances, say, several hundred feet, to obtain Fickian distribution. A very rough boundary introduces violent eddies which predominates the convective effects in controlling the dispersion. Thus, we get Fickian distribution in a much shorter distance. I think that the turbulent structure is more homogeneous and isotropic than in the case of smooth channels.

THOMAS: The trouble is the theory was derived for smooth channels.

YOTSUKURA: Dr. Parker made this study of dispersion by introducing waves. What dispersion did you get Dr. Parker?

PARKER: We are still analyzing the results.

YOTSUKURA: From these Figures 2, 3 and 4, we have made several conclusions. One is that the standard deviation is the most stable quantity which satisfies the so-called Fickian solution, whereas all other properties such as skew, maximum concentration and so forth, are deviating from the Fickian solution. Therefore, when we compute the dispersion coefficient under the assumption of Fickian solution it is best to use the standard deviation.

FREDERICK: Have you computed the area under some of these curves to determine what percentage of the injected material you are actually observing?

YOTSUKURA: No. I made an attempt to check the area under the curve but I found this kind of trial is meaningless. As long as the area is somewhat consistent, say a fluctuation of 10% or so from one station to another, it is all right. If you have a very large rate of increase of the area as you go downstream then this is a clear indication that you are having some density problems. The areas are very hard things to check unless you have a perfectly uniform concentration over the cross-section.

FREDERICK: It seems to me that your mathematical computations are a waste of time if you do not have complete distribution.

YOTSUKURA: I don't think so. It is not necessary to have a uniform distribution over the cross-section. From our theoretical and experimental work, we know that the dispersion coefficient, in which we are interested, becomes constant much earlier than the uniform distribution obtains.

FREDERICK: It is necessary to make these mathematical computations. I will grant that you don't always have it, but if you don't have it you are not going to be able to make your mathematical model.

SAYRE: I think that if you have just a fair approximation of a uniform concentration throughout the cross-section it wouldn't make much difference in the diffusion coefficient. If the coefficient is decidedly non-uniform it would probably make a considerable difference.

FREDERICK: You should find out what sort of a distribution you have.

YOTSUKURA: What I mean is that if you have the area underneath the curve increasing or decreasing with a great variation then you should be very cautious. But, as long as it stays within reasonable range then this is some sort of an indication that there is uniform distribution.

PARKER: No, that is not necessarily so. If you have a density current then you have a certain percentage of it in the area that you are measuring with a much larger slug that is going along undetected near the bottom.

SAYRE: What you are saying is that if you come out within 10% of the calculated area that you should come out with, then this is an indication that the

distribution is pretty good.

FREDERICK: The curve we are discussing is actually concentration versus time. If you integrate it over the total period of time you should get the total amount of activity. Have you done that?

YOTSUKURA: In the early tests I checked this and got some kind of a uniformity and since then I have not done any more work in this area. Our main interest is in the dispersion coefficient. When you integrate the tail it comprises such a small portion of the area that it may not be important.

KAUFMAN: In a natural stream you do not have a sharp bottom boundary. An interchange between the body of water in the stream and the groundwater must occur. It is the transfer rate between the groundwater and the surface water that causes the tail. If you persevere a complete recovery of the tracer will be obtained, providing the stream is not an effluent one.

SAYRE: Tails have also been observed in concrete-lined channels.

YOTSUKURA: I would like to check if the long tail is an instrumental problem. We will have to do this in a model flume and a natural stream. If it is not the instrumental problem, there are a number of reasons that might cause it.

FREDERICK: I think the same thing has been observed when there is not opportunity for groundwater flow, that is, in concrete-lined channels.

SAYRE: In addition to Dr. Kaufman's comment about natural streams, in natural streams with irregular banks, a considerable amount of separation often occurs near the boundary where the water near the edges of the channel is essentially circulating but not flowing. In such cases you might also logically expect to find a long tail.

YOTSUKURA: Yes, that is what we should expect from the butt roughness used in our study, but we did not find any long tails by using a conductivity probe. I confess I simply do not know.

TRANSPORT OF RADIONUCLIDES IN FRESH WATER: DISPERSAL OF BED SEDIMENTS

by W. W. SAYRE and D. W. HUBBELL

INTRODUCTION

When radionuclides become attached to sediment particles, sediment transport in streams and rivers assumes importance as one of the phenomena which control the distribution of radionuclides in the environment.

Bed-material sediments are nearly always transported at a rate which is much slower (often several orders of magnitude) than the velocity of flow. Therefore, conditions which are favorable for uptake of radionuclides by bed-material particles may result in an objectionable buildup of radioactivity in the stream bed even though the concentration of radionuclides in the stream water at the time of release is far below the maximum permissible concentration. Fine sediments (clays and silts) on the other hand are generally transported in suspension at a rate more nearly equal to the velocity of flow. However, under circumstances such as impoundment or conditions which are conducive to flocculation, fine sediments may be deposited on the bed and their transport greatly retarded.

Fine sediments tend to have the greatest sorption capacities, however, as was pointed out in Dr. Kennedy's presentation, sand particles coated with clay and certain kinds of rock fragments in the sand size range may also have relatively high sorption capacities. Depending on the specific conditions, a hazard could be associated with almost any kind of sediment. Therefore, an evaluation of hazards arising from the uptake of radionuclides by fluvial sediments should ideally include a study of the transport of all types of sediment.

The study of sediment transport can be approached in either of two ways. In the first approach, attention is focused on a given cross section of the stream, and on the description of the motion, the concentrations and the characteristics of sediment particles as they pass the cross section. By analogy with classical mechanics, this is called an Eulerian description. The observer stands at the cross section and watches the particles go by. In the second approach attention is focused on a particular particle or group of particles and its (their) motion is described over a period of time. Using the same analogy, this is called a Lagrangian description. The observer moves downstream with the particles and watches their behavior as they go.

In general, information derived from one of these descriptions cannot be directly applied to the other system and vice versa. The study of the dispersal of contaminated sediment in a natural stream environment ties in naturally with the Lagrangian method of description. However, the great bulk of theories and measurement techniques associated with current and past sediment transport research is geared to the Eulerian system. Therefore, much of this knowledge is not directly applicable to the problem at hand and new theories and measurement techniques must be developed.

DISPERSION OF BED MATERIAL

Theory - H. A. Einstein (1937) was one of the first to establish a theoretical foundation for describing the motion of bed-material particles in a Lagrangian frame of reference. Unfortunately, very little has been done in the intervening years to follow up the promising beginning made by Einstein. This has no doubt been largely due to the experimental difficulties associated with tracing the motions of a given particle or group of particles. Indeed until the advent of radioactive tracer techniques, the experimental verification of the theory, except on a very limited laboratory scale, was virtually impossible.

In an alluvial channel, the part of the total sediment discharge that is composed of particles having sizes the same as those of particles commonly found in the bed material is called the bed-material discharge. The motion of bed-material particles over a period of time consists of a series of discrete steps. A particle may roll along the bed or be entrained temporarily in the flow by a turbulent eddy, then come to rest on the bed where it will remain, usually becoming covered by other particles, until it is re-exposed and takes another step. Thus, the motion of a particle can be described by the length of the steps that it takes and the duration of the rest periods between steps, provided, of course, that the aggregate resting time is very large in comparison to the aggregate time spent in motion.

The step lengths and rest periods that a particle takes are random phenomena that depend on factors such as flow and turbulence characteristics, sediment characteristics, and bed form. Due to the randomness, the step lengths and the durations of the rest periods are variable. Consequently, if a group of particles are released simultaneously from a source that extends across the width of the channel, the particles will not move downstream in a body, but will become dispersed along the length of the channel. In addition to being distributed longitudinally, the particles will be dispersed throughout some depth into the bed; the depth depends on the characteristics of the bed forms.

Because the transport and dispersion of bed-material particles is essentially a random process it is logical to use probability theory to derive a function for describing the dispersion process. H. A. Einstein (1937) considered bed-load transport from the viewpoint of probability theory. Beginning with

essentially the same assumptions, Sayre and Hubbell (1963) arrived at distribution functions which are identical to those obtained by Einstein. However, the two derivations are considerably different.

In order to acquaint the reader with the nature of the distributions, the derivation as presented by Sayre and Hubbell (1963) is reviewed.

Assume a straight, uniform alluvial channel in which the water and bed-material discharges per unit width are uniform and constant. Now, consider a group of contaminated particles which have identical transport characteristics. Let these particles be released simultaneously, at time $t = 0$ and station $x = 0$, from a uniformly-distributed plane source that extends laterally across the channel and vertically from the surface of the bed to the lower limit of the zone of particle movement. Also, let the particles move and disperse downstream in such a way that there is no tendency to develop a vertical concentration gradient of contaminated particles within the bed. Furthermore, assume that the number of contaminated particles is small enough so that the bed-material discharge is virtually unchanged by the release of the particles, yet is large enough to approximate an infinite population.

If the foregoing conditions are satisfied the stream channel can be characterized as a homogeneous probability field in space and time. If, as it is, reasonable to assume, the length of any particular step and the duration of any particular rest period of any given particle are independent of x and t and of each other, the step lengths and rest periods can each be represented as a set of independent, identically-distributed random variables. Also, the total distance from the origin travelled by a particle in n steps can be represented as the sum of n independent, identically distributed random variables.

Let it be assumed that the step lengths are exponentially distributed with mean step length $1/k_1$ and that the durations of the rest periods are exponentially distributed with mean duration $1/k_2$. With these assumptions it can be shown (Parzen, 1962) that the total distance, x , travelled by a particle in n steps is gamma distributed, and that the number of waiting periods, i , in the time interval $(0, t)$ is Poisson distributed. Expressed mathematically,

$$f(x|n) = k_1 e^{-k_1 x} \frac{(k_1 x)^{n-1}}{\Gamma(n)} \quad 0 < x < \infty \quad (1)$$

and

$$p_t(i) = e^{-k_2 t} \frac{(k_2 t)^i}{i!} \quad i = 1, 2, \dots \quad (2)$$

Equation 1 is the probability density function of the gamma distribution. When equation 1 is multiplied by dx , it expresses the probability that a contaminated particle will be located in the length increment $(x, x + dx)$, which is to say that the particle will have been transported the exact distance $(0, x)$, given that it has taken exactly n steps. Equation 2 is the probability mass function of the Poisson distribution and it expresses the probability that during the time interval $(0, t)$ a particle will have completed exactly i rest periods.

As a particle moves downstream in a sequence of discrete steps, each rest period is followed by a step which, in turn, is followed by another rest period, etc. Therefore, assuming that the particle is initially at rest and begins its first rest period at $t = 0$, during any given rest period the particle must have undergone the same number of complete rest periods as steps. Hence, a particle which has taken n steps also will have undergone n rest periods.

From a practical standpoint, it is not possible to keep track of the number of steps (or rest periods) that a particle undergoes; consequently, equations 1 and 2 as they stand are of little practical use. However, by using the concept of conditional probability, equations 1 and 2, in effect, can be combined to form the density function $f_t(x)$ from which the probability of the location of the contaminated particle in any length increment, $(x, x + dx)$, at time t , can be obtained. To this end it is convenient to express equation 1 in the form of a conditional probability distribution function as follows:

$$F(x|n) = \int_0^x f(x'|n) dx' = P[x' \leq x | i = n] \quad (1a)$$

in which

P denotes probability,
 x' is a dummy variable of integration.

Applying the definition of conditional probability,

$$F_t(x|n) = P_t[x' \leq x | i = n] = \frac{P_t[x' \leq x, i = n]}{P_t[i = n]} = \frac{P_t[x' \leq x, i = n]}{p_t(n)} \quad 0 < t < \infty$$

When n is given, knowledge of t adds no information, hence

$$F_t(x|n) = F(x|n)$$

and

$$P_t[x' \leq x, i = n] = F(x|n) p_t(n) \quad (3)$$

The marginal distribution function in x corresponding to the joint probability function in equation 3 is

$$\begin{aligned} F_t(x) &= \sum_{n=0}^{\infty} P_t[x' \leq x, i = n] = \sum_{n=0}^{\infty} F(x|n) p_t(n) \\ &= \sum_{n=1}^{\infty} F(x|n) p_t(n) + F(x|0) p_t(0) \end{aligned} \quad (4)$$

Differentiation of equation 4 with respect to x yields the density function for x at time t

$$f_t(x) = \frac{\partial}{\partial x} [F_t(x)] = \sum_{n=1}^{\infty} f(x|n) p_t(n) + p_t(0) \frac{\partial}{\partial x} [F(x|0)]$$

Performing the substitutions indicated by equations 1 and 2, and noting that

$$\begin{aligned} p_t(0) &= e^{-k_2 t} \\ F(x|0) &= 1 \quad \text{when } x = 0 \\ &= 0 \quad \text{elsewhere} \end{aligned}$$

the density function for x at time t can be written

$$f_t(x) = k_1 e^{-(k_1 x + k_2 t)} \sum_{n=1}^{\infty} \frac{(k_1 x)^{n-1}}{\Gamma(n)} \frac{(k_2 t)^n}{n!} \quad (5)$$

which transforms readily into

$$f_t(x) = k_1 e^{-(k_1 x + k_2 t)} \sqrt{\frac{k_2 t}{k_1 x}} I_1(2 \sqrt{k_1 x k_2 t}) \quad (5a)$$

where $I_1(2 \sqrt{k_1 x k_2 t})$ is a modified Bessel function of the first kind of order one. Equations 5 and 5a were first derived by H. A. Einstein (1937).

If the displacement of a contaminated particle from the origin at time t is considered as a stochastic process, it can be shown that equations 5 and 5a represent the density function corresponding to a compound Poisson process with exponentially distributed incremental step lengths (Parzen, 1962).

It should be noted that the integral of the density function in equations 5 and 5a over the entire range of x is

$$\int_0^{\infty} f_t(x) dx = 1 - e^{-k_2 t} = 1 - p_t(0)$$

where $p_t(0)$ is the probability that the particle has not yet moved from the origin at time t . Thus the density function $f_t(x)$ applies only to particles which have taken at least one initial step.

If, as assumed initially, a very large number of contaminated particles that are initially distributed uniformly in a plane extending throughout the zone of movement at $x = 0$ all begin their first rest period at $t = 0$, the density function, $f_t(x)$, as defined by equations 5 and 5a can be used to estimate the concentration, $\phi(x, t)$, of contaminated particles in the bed at any distance, x , downstream from the source at time t . In fact, $\phi(x, t)$ will be directly proportional to $f_t(x)$ if the following conditions are satisfied: (1) the average depth to which the contaminated particles are distributed remains constant downstream from the source; (2) the concentration of contaminated particles in the bed does not, on the average, vary with the depth to which they are distributed; (3) at any given instant, no significant fraction of the contaminated particles are in suspension. Given these conditions, if the concentration is defined as the weight of contaminated particles per unit volume of bed material, the concentration is

$$\phi(x, t) = \frac{W}{Bd} f_t(x)$$

in which

- W is the total weight of contaminated particles released into the channel
 B is the width of the channel
 d is the average depth beneath the bed surface to which the contaminated particles are distributed.

Thus, the concentration-distribution function at time t for contaminated particles which have moved from the origin where they began their first rest period at $t = 0$ is

$$\phi(x, t) = \frac{W k_1}{Bd} e^{-(k_1 x + k_2 t)} \sum_{n=1}^{\infty} \frac{(k_1 x)^{n-1}}{\Gamma(n)} \frac{(k_2 t)^n}{n!} \quad (6)$$

or

$$\phi(x, t) = \frac{W k_1}{Bd} e^{-(k_1 x + k_2 t)} \sqrt{\frac{k_2 t}{k_1 x}} I_1(2 \sqrt{k_1 x k_2 t}) \quad (6a)$$

The displacement of the center of concentration of the contaminated particles at any time, t , is given by

$$\bar{x} = \int_0^{\infty} x f_t(x) dx = \frac{k_2 t}{k_1}$$

hence the mean rate of movement of the contaminated particles is

$$\frac{\bar{x}}{t} = \frac{k_2}{k_1}$$

If a contaminated particle is introduced into the channel at $x = 0$ in such a way that it takes its first step at time $t = 0$, then during any given rest period it will have completed one less rest period than steps. In this case, equation 4 becomes

$$F_t^*(x) = \sum_{n=1}^{\infty} F(x|n) p_t(n-1) \quad (8)$$

where the asterisk denotes the initial condition of beginning with motion. Likewise equations 5 and 5a become

$$f_t^*(x) = k_1 e^{-(k_1 x + k_2 t)} \sum_{n=1}^{\infty} \frac{(k_1 x)^{n-1}}{\Gamma(n)} \frac{(k_2 t)^{n-1}}{(n-1)!} \quad (9)$$

and

$$f_t^*(x) = k_1 e^{-(k_1 x + k_2 t)} I_0(2\sqrt{k_1 x k_2 t}) \quad (9a)$$

where $I_0(2\sqrt{k_1 x k_2 t})$ is a modified Bessel function of the first kind of order zero. As were equations 5 and 5a, equations 9 and 9a were first obtained by Einstein (1937).

In a manner analogous to that which was used to obtain the concentration-distribution function, (x, t) , which specifies the concentration of contaminated particles in the bed at any distance x and any time t , a discharge-distribution function, (t, x) , which specifies the discharge of contaminated particles past a cross section at any distance x downstream from the source at any time t , can be obtained from the density function $f_x(t)$. A particle which begins its first rest period at the origin at $t = 0$ will, as it passes a point a distance x downstream from the origin, have completed one less step than rest periods. Thus, the probability density $f_x(t)$ which relates to the probability that the particle will pass the point x during the time interval $(t, t + dt)$ corresponds to $f_t^*(x)$ except that the variables x and t and the coefficients k_1 and k_2 are interchanged so that

$$f_x(t) = k_2 e^{-(k_1 x + k_2 t)} \sum_{n=1}^{\infty} \frac{(k_2 t)^{n-1}}{\Gamma(n)} \frac{(k_1 x)^{n-1}}{(n-1)!} \quad (10)$$

or

$$f_x(t) = k_2 e^{-(k_1 x + k_2 t)} I_0(2\sqrt{k_1 x k_2 t}) \quad (10a) \quad a)$$

The corresponding discharge-distribution function in weight of contaminated particles per unit time per unit of cross-sectional area is

$$\psi(t, x) = \frac{W k_2}{Bd} e^{-(k_1 x + k_2 t)} \sum_{n=1}^{\infty} \frac{(k_2 t)^{n-1}}{\Gamma(n)} \frac{(k_1 x)^{n-1}}{(n-1)!} \quad (11) \quad //$$

or

$$\psi(t, x) = \frac{W k_2}{Bd} e^{-(k_1 x + k_2 t)} I_0(2\sqrt{k_1 x k_2 t}) \quad (11a) \quad // a)$$

For any given set of streamflow and sediment conditions the coefficients k_1 and k_2 will have the same values for both distribution functions (equations 6 and 11).² The functional relationship between k_1 and k_2 and sediment and hydraulic parameters is not yet known, hence the values of k_1 and k_2 for a particular set of conditions can at present be determined only by experiment.

Experiment - In November 1960, the dispersion of bed-material particles was investigated experimentally in the North Loup River, near Purdum, Nebraska, by means of radioactive tracer techniques. The reach selected for the experiment is 1800 ft long, approximately 50 ft wide, and fairly straight in alignment. The bed material in the North Loup River is composed of sand having a median fall diameter of 0.29 mm. At the time of the experiment the water discharge was about 260 cfs, the water-surface slope was about 0.00083, the mean depth was about 2.5 ft, and the bed configuration consisted of large dunes which averaged about 1.3 ft in height and 12 ft in length.

In the experiment 40 lbs of sand particles labeled with 40 millicuries of Iridium-192 were deposited along a line extending across the channel bed at the upstream end of the test reach on November 3, 1960. This initial condition approximated an instantaneous, uniformly distributed plane source. The subsequent dispersion of these tracer particles was observed by making periodic surveys along the stream bed with a sled-mounted scintillation detector (Figure 1) which was pulled downstream at a controlled rate by a boat as shown in Figure 2. Core samples were also obtained to determine the distribution of tracer particles with respect to depth beneath the bed surface. In addition, the distribution of tracer particles across the channel was checked at selected cross sections, and was found to be reasonably uniform.

The observed counting rates were converted to concentrations (weight of tracer particles per unit volume of bed material) on the basis of detailed laboratory calibrations which were carried out under simulated experimental conditions. Several typical curves which illustrate the distribution of tracer particles along the channel at various times following the dosing are shown on Figures 3 and 4. The secondary peak near the source shown on Figure 4 is of interest because it represents a group of tracer particles which became buried beyond the range of the detection equipment in a deep trough near the source

Figure 1

Figure 2

Figure 3 Longitudinal distribution of labelled particles along the right side of the channel, Nov 3-8, 1960

Figure 4 Longitudinal distribution of labelled particles along the center of the channel

shortly after dosing and did not become re-exposed until nearly a week later. This illustrates that under some conditions, slugs of contaminated particles can remain buried for considerable periods of time during which no dispersion will take place.

The core sample data indicated that the tracer particles were distributed to an average depth of about 1.5 ft below the surface of the bed. These data also indicated that within this zone there was no pronounced tendency for the concentration of tracer particles to either decrease or increase with depth beneath the bed surface.

The parameters k_1 and k_2 in the theoretical concentration distribution function (equation 6) were determined by comparing the rate of movement of the mode along the channel and the rate of attenuation of the peak concentration with respect to time for the observed longitudinal concentration-distribution curves with the corresponding characteristics indicated by the theoretical concentration distribution function. These calculations resulted in a mean step length ($1/k_1$) of 36 ft, a mean rest period ($1/k_2$) of 12 hours and a mean rate of particle movement (k_2/k_1) of 3 ft per hour. Several theoretical concentration distribution curves which were calculated from equation 6 using the above values of k_1 and k_2 are compared with observed concentration distribution curves in Figures 5 and 6.

An idea of the range of applicability of the theoretical dispersion model can be obtained by comparing certain features of this experiment to the corresponding features of experiments reported by Einstein (1937). Einstein used the same model to describe the dispersion of 30 mm diameter gravel particles which were being transported at mean rates as high as 1300 ft per hour. Hence, the two sets of experiments bracket a hundredfold range of particle sizes and a several hundredfold range of transport rates.

A considerably more detailed description of the design, conduct and results of the North Loup River experiment is given in a publication by Sayre and Hubbell (1963).

Application to real situations - The theory and experiment discussed in the preceding sections are based on the idealized concept of an instantaneous source of contaminated particles which are distributed uniformly over a plane which extends laterally across the width of the stream and vertically from the surface of the bed to the lower limit of the zone of particle movement. Although such idealized source conditions exist rarely if ever in nature, the concentration distribution function corresponding to the instantaneous plane source (equation 6) is of fundamental practical importance. This is because concentration-distribution functions corresponding to any known source distributions in space and time can be obtained from the instantaneous plane source function by the principle of mathematical superposition. The same principle applies to the discharge-distribution function (equation 11) also.

Some examples of concentration-distribution functions corresponding to source distributions where the source strength varies with longitudinal distance and/or time are listed below. In these examples, uniform concentration distributions in the lateral and vertical directions are assumed.

1. Instantaneous longitudinally-distributed source of variable strength, $S(\xi)$:

$$\phi(x, t) = \frac{1}{Bd} \int_0^L S(\xi) f_1(x-\xi, t) d\xi \quad (12)$$

where

ξ is the distance downstream from $x = 0$ along which the source is distributed. ($0 \leq \xi \leq L$)

$f_1(x-\xi, t)$ is the instantaneous plane source density function, $f_t(x)$ (equation 5), with x replaced by $x-\xi$.

2. Continuous plane source at $x = 0$, discharging at a variable rate, $S(\tau)$:

$$\phi(x, t) = \frac{1}{Bd} \int_0^{t_1} S(\tau) f_1(x, t-\tau) d\tau \quad (13)$$

where

τ is the time after $t=0$ during which the source is discharging. ($0 \leq \tau \leq t_1$)

$f_1(x, t-\tau)$ is the instantaneous plane source density function with t replaced by $t-\tau$.

3. Continuous, longitudinally-distributed source of variable strength and discharging at a variable rate, $S(\xi, \tau)$:

$$\phi(x, t) = \frac{1}{Bd} \int_0^{t_1} \int_0^L S(\xi, \tau) f_1(x-\xi, t-\tau) d\xi d\tau \quad (14)$$

where

$f_1(x-\xi, t-\tau)$ is the instantaneous plane source density function
with x replaced by $x-\xi$, and t replaced by $t-\tau$.

By integrations such as these an infinite variety of real situations can be approximated. In addition to longitudinal concentration-distribution curves, the instantaneous plane source density functions can be used to construct concentration-time curves, longitudinal discharge-distribution and discharge-time curves, concentration and discharge growth and decay curves, etc. The theoretical distribution functions also provide a means for making estimates such as the time of travel and reduction of maximum concentration of a slug of contaminated bed-material as it travels from one location to another location further downstream, the duration of contamination in excess of some specified level at a particular site, and the level of and length of time required to reach an equilibrium concentration of contaminated particles which are being released from a continuous source.

Another idealized concept which was employed in the theory is the implication that a stream can be represented as a homogeneous probability field in space and time. Such an assumption will not be justified in many cases. However, using available knowledge of stream characteristics and flow patterns it should be possible to break most streams up into reaches in which the assumption of homogeneity is an acceptable approximation for limited periods of time. This would be analogous to the procedure commonly used in calculating back-water curves in or routing floods through non-uniform channels.

In the theory and experiment reported herein, the effects of exchange of radionuclides among various phases of the transporting media has not been considered. It was assumed that once a particle became contaminated by radionuclides it would remain so. Using the mathematical device of secondary sources and sinks it should be possible to allow for the effects of the exchange of radionuclides, for example between sediment and solution or biota, in the theoretical distribution functions.

In the immediate future there is a need to determine experimentally the values of the coefficients k_1 and k_2 over a wide range of sediment and hydraulic conditions both in the laboratory and in the field, and to obtain an understanding of how they vary with these factors. This will permit bridging the gap between the Eulerian and Lagrangian descriptions of particle motion so that the mass of Eulerian sediment transport data which has been accumulated over the years can be used for the purpose of predicting dispersion.

Another need is to reduce the data which is being obtained on the release of radionuclides into streams and the subsequent uptake of radionuclides by sediment particles to the form of equations which describe the "source strength" of contaminated particles as a function of space and time, so that this information can be incorporated into the dispersion equations. Quantitative information on

expected rates of exchange of radionuclides among the various phases of the transporting medium (water, sediment, and biota) is needed for the same reason.

DISPERSION OF FINE SEDIMENTS

In contrast to the coarser bed sediments such as sands and gravels the individual particles of which move in discrete steps at an overall rate which is much slower than the velocity of flow, the very fine sediments such as clays in a dispersed state tend to be maintained in continuous suspension in the flow and to be dispersed in a manner more nearly like the miscible materials as was just described by Dr. Yotsukura.

We have just begun an experimental program on the transport of fine sediment the purpose of which is to ascertain and evaluate the differences between the dispersion of suspended fine particles, and the dispersion of a miscible liquid in the same flow field. We have conducted one set of experiments in a concrete-lined canal in which we compared the dispersion of a dispersed Wyoming Bentonite to the dispersion of Rhodamine-B dye.

The experiments were conducted in the Horsetooth Feeder Canal near Fort Collins, Colorado, in a 1600-ft straight reach with a 7-ft bottom width and 1-1/4 to 1 side slopes. At the time of the experiments the water discharge was 24 cfs, the depth of flow was 1.7 ft and the water-surface slope was 0.00012.

The dye, in solution form, and the sediment, in the form of a well mixed slurry, were introduced into the flow from a trough that extended across the channel and was positioned a few inches above the water surface. This method of introduction was a good approximation of an instantaneous plane source. The concentrations of dye and sediment were measured at several locations downstream from the source as the slug passed the measuring point. The dye concentrations were measured by a continuously-recording fluorometer. The sediment concentrations were determined by collecting one-quart samples in rapid sequence and analyzing the samples in the laboratory by conventional weighing and drying procedures.

The results of the dye dispersion experiments are shown in Figure 7 and those of the fine sediment dispersion experiments in Figure 8. In Figure 9 dye and sediment concentration-time curves obtained 1400 ft downstream from the source are compared. The comparison indicates that for this particular set of experimental conditions the dispersion of bentonite is not significantly different from that of the dye. The agreement is not as good for the sampling stations closer to the source, however, this is attributed to differences in the response characteristics of the two sampling methods which are, of course, most apparent when the rate of change of concentration with respect to time is rapid. This conclusion is borne out by the fact that the sediment and dye curves tend to converge

Figure 7

Figure 8

Figure 9

to one another with increasing distance from the source, whereas they would diverge if there were any essential differences in the dispersion processes.

CONCLUDING REMARKS

We have made a start in studying the role of fluvial sediment in transporting and dispersing radionuclides which are released into streams as radioactive wastes and subsequently sorbed on sediment particles. We have considered the limiting cases, i. e., the dispersion and transport of sand-size particles which move as bed-material load, and the dispersion and transport of extremely fine clay particles which move in a continuously suspended state. In between there remains an entire spectrum of sediment types and conditions which is unexplored territory as far as dispersion research is concerned.

We advocate that research associated with the physical, biological and chemical aspects of river systems receiving radioactive wastes be oriented, whenever feasible, toward providing data that can be interpreted in terms of probability laws and/or stochastic processes. The success of this method of treatment in deriving a theoretical model for the dispersion of contaminated bed material particles, coupled with the success of the statistical theories of turbulent diffusion in providing a model for the dispersion of miscible and extremely fine particulate contaminants, lead one to speculate that similar analyses might also prove fruitful in examining other phases of the system. Speculating further, it appears reasonable to suppose that a mathematical model could be set up which would treat the response of an entire river system as a multiple component stochastic process composed of jointly distributed random variables.

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FIGURE TITLES

1. Sled and scintillation detector. Note the parallelogram linkage that allows the detector freedom of vertical motion and the wooden dish-shaped piece that supports the detector on the bed.
2. Arrangement of boat and sled for the longitudinal traverses.
3. Longitudinal distribution of labelled particles along the right side of channel, November 3-8, 1960.
4. Longitudinal distribution of labelled particles along the center of the channel, November 9 and 11, 1960.
5. Comparison of theoretical and measured longitudinal concentration distribution curves, Runs 3R and 4R.
6. Comparison of theoretical and measured longitudinal concentration distribution curves, Runs 5R and 6R.
7. Concentration-time curves illustrating longitudinal dispersion of Rhodamine-B dye from an instantaneous plane source in Horsetooth Feeder Canal near Fort Collins, Colorado.
8. Concentration-time curves illustrating longitudinal dispersion of suspended Wyoming bentonite from an instantaneous plane source in Horsetooth Feeder Canal near Fort Collins, Colorado.
9. Comparison of Rhodamine-B and Wyoming bentonite dispersion curves at 1400 ft from the source.

DISCUSSION

THOMAS: Do you allow the particle to go back upstream in your theory?

SAYRE: No, we do not allow the particle to go back upstream.

PARKER: In the sampling of the Wyoming bentonite, in case of the fine sediments, how did you determine the concentration?

SAYRE: We pumped samples out of the canal into quart bottles, dried the samples out and weighed them.

THOMAS: These curves downstream at 1400 feet for the dissolved tracer look like they fit your theory better than they do Taylor's theory. They appear to accord with your formulation despite the fact that your theory was derived for bedload movement.

SAYRE: Yes, they seem to.

FREDERICK: Did you compute your theory and plot it on Figure 9?

SAYRE: No. This is an empirical curve, it is not a theoretical curve.

FREDERICK: Why didn't you?

SAYRE: We have not really analyzed this data yet. We have had a problem particularly in the dye curves that in computing the variances by the method of moments we did not know exactly where to cut the tails off. It makes quite a difference.

THOMAS: You know there are other models for turbulence beside Taylor's.

SAYRE: Yes, I know this. I just wanted to show these as a comparison between the dispersion of dye and the dispersion of fine suspended sediments.

YOTSUKURA: In the sigma test, using the betonite, do you get the same long tail?

SAYRE: No, not as much. The reason for this may have been that our

sampling methods were not as accurate and we got down to a scattering about the background which consisted of the dissolved solids in the water. The individual samples scattered about so that we could not really pinpoint where the curve came down to background.

I might also mention here that in earlier runs the fit of the two was not as good, but I think this can be attributed to the differences in the sampling techniques. As the distance from the source increased the dye and sediment curves converged to one another. If there were any important differences between the dispersion mechanisms they would tend to diverge rather than converge.

KAUFMAN: Suppose you had reduced the velocity progressively in this channel?

SAYRE: I don't think that this would have made any difference in the degree of dispersion of the sediment in comparison to that of the dye. I'm not making any claims here that this would be universally true and that there would never be any difference in the dispersion of the two. My conclusion applies only to flows with a Reynolds number as high or higher than in our experiment and to very fine sediments in a dispersed state.

KAUFMAN: These particles are fine enough to behave as molecules.

SAYRE: Yes, that is probably correct.

PARKER: Did you intergrate under the curves to see if you got the same amount of solids?

SAYRE: Yes we did; we got fairly consistent results. However, there was some scattering.

PARKER: I was wondering why the first ones did not appear to agree. Upstream you had some larger particles so they did not settle out?

SAYRE: No. These were all fine particles so they did not settle out. We did not lose area under the curve. So, I do not think this was affecting us particularly.

A LEAST COST EVALUATION OF LOW LEVEL LIQUID ATOMIC WASTE TREATMENT-SIMPLIFIED DETERMINISTIC MODEL

by W. O. SPOFFORD, JR. and H. A. THOMAS, JR.

I INTRODUCTION

The following example illustrates the use of a simplified economic decision making process as applied to a water quality problem. The flows of water and the flux of pollutant are deterministic and constant in contrast to a more realistic stochastic system.¹ These vectors were made deterministic in order that the problem would lend itself more readily to an analytical solution. The problem was made as simple as possible so that the theoretical principles involved could be elucidated.

Only one decision making "knob" or decision variable is exhibited in this example, for any more would only have made the mathematics of an analytical solution more complicated without concomitant gain in exhibiting the principle of the economic analysis. For most complex multivariate water resource systems, however, a large number of possible decision "knobs" exist.

This is a specific problem in systems analysis in which two investments are made most economically to provide a given required output, that being a maximum permissible concentration of a given pollutant.

The general problem is discussed in the next section whereas the objective function, cost function for the treatment plants, mathematical development, and a numerical example follow in subsequent sections.

¹ deterministic process: a degenerate case of a stochastic process in which the error in forecasting the future is zero; hence in such a process a knowledge of the past is sufficient to predict the future perfectly.

Stochastic: as an adjective, connotes the presence of a random variable. A stochastic process is one in which at least one of the variables in the system is a random variable or has an element of randomness. Opposed to a deterministic system. The word stochastic derives from a Greek word applying to a forecast of the future course of events with the intention of predicting the truth.

II GENERAL PROBLEM

It is assumed that the flows of water Q'_R , Q_R , Q_1 and Q_2 and the flux of pollutant X_0 and Y_1 shown on Figure 1 are deterministic, constant and given for this example. The system is in a steady state condition with reference to both the pollutant and hydrology and only one isotope without any radioactive daughters is considered. For this simple case, one use point is assumed as shown on Figure 1. The river enters the system in three separable and beneficial ways; namely it (a) provides for dilution, (b) provides for decay and hold-up, and (c) may improve treatability such as providing a free coagulant aid. It is assumed that treatment will be provided at both the waste discharge point, (1), and the water intake point (2).² These are known as the economic input vectors to the system. A storage reservoir in the stream midway between the outlet and the intake is considered as a third possible treatment mechanism in a subsequent section. In this section this alternative is not considered. Another technological possibility - an alternative source of water supply - is not considered in this model. Also the possibility of carrier addition is not considered.

The flux of pollutant enters the system at treatment plant 1, as the quantity X_0 and passes through the waste treatment plant to the effluent, X_1 . The degree of treatment at the waste treatment plant 1, must be balanced with the degree of treatment at the water treatment plant 2 in order to maximize economic efficiency. These involve an economic balance subject to a constraint; the constraint being the maximum permissible flux of pollutant in the drinking water at the use point. The isotope reaches the stream with the effluent, Q_1 , of the waste treatment plant and mixes with the quantity of water flowing in the stream, Q'_R . In the stream, the waste is diluted, the isotope is held up³ and also decays.

A portion, Q_2 , of this stream water, Q_R , containing a flux of pollutant, Y_0 , is taken in at point 2 for use. After this water passes through the water treatment plant, the remaining flux of pollutant is designated as Y_1 . Y_1 is taken as a constraint that must be met by employing either or both of the treatment plants, 1 and 2.

The object of this example is to minimize the total cost of the project, C_T , subject to the constraint $Y_1 = K$, by varying the flux parameter, X_1 . Stated differently, the goal is to minimize the objective function subject to the constraint. The objective function is given in the next section.

² Design alternatives include a zero scale of development for each plant.

³ Part of the waste moves directly with the wash load of the stream and part is routed to benthal deposits where its rate of passage down-stream is halted or retarded.

FIG. 1 PLAN OF RIVER SYSTEM SHOWING WATER & WASTE TREATMENT PLANTS

III OBJECTIVE FUNCTION

In systems analysis, the term "objective function" commonly refers to a function in which the dependent variables are to be maximized or minimized. The dependent variable may be velocity, time, distance, weight, cost, net benefits, economic growth, income redistribution, etc. Usually the objective function contains constraints of a physical or economic nature.

For this particular problem, the objective function can be formulated as a cost function⁴; namely,

$$C_T = C_1 + C_2 + \dots + C_n \quad (1)$$

$$= \sum_{i=1}^n C_i \quad (2)$$

where C_T is to be minimized and n is the number of economic input vectors. The cost at treatment plant 1 is C_1 and is a function of the volume of the effluent, the initial concentration of pollutant, the final concentration of pollutant, parameters characteristic of the treatment process and economic parameters such as discount rate and economic time horizon. Expressed mathematically,

$$C_1 = f(x_1^1, x_2^2, x_1^3, x_1^4, \dots, x_1^r) \quad (3)$$

or in general $C_i = f(x_i^1, x_i^2, \dots, x_i^r) \quad (4)$

where r equals the number of parameters necessary to describe the cost function of a given treatment unit.

If the objective function is not constrained, a necessary but not a sufficient condition for a maxima or a minima is that the first derivatives with respect to all the independent variables must be equal to zero.

$$\frac{\partial C_T}{\partial C_1} = 0, \quad \frac{\partial C_T}{\partial C_2} = 0, \quad \dots \quad \frac{\partial C_T}{\partial C_n} = 0 \quad (5)$$

⁴ Cost, C_i , is a present value cost of the initial construction plus the operating and maintenance costs over the economic time horizon.

$$C_1 = f(x_1^1, x_1^2, \dots, x_1^r), C_2 = f(x_2^1, x_2^2, \dots, x_2^r), \dots, C_n = f(x_n^1, x_n^2, \dots, x_n^r)$$

$$\frac{\partial C_T}{\partial x_1^1} = \frac{\partial f_c}{\partial c_1} \cdot \frac{\partial c_1}{\partial x_1^1} = 0, \quad \frac{\partial C_T}{\partial x_1^2} = \frac{\partial f_c}{\partial c_1} \cdot \frac{\partial c_1}{\partial x_1^2} = 0, \dots, \frac{\partial C_T}{\partial x_1^r} = \frac{\partial f_c}{\partial c_1} \cdot \frac{\partial c_1}{\partial x_1^r} = 0$$

$$\frac{\partial c_1}{\partial x_2} = \frac{\partial f_c}{\partial c_2} \cdot \frac{\partial c_2}{\partial x_2} = 0, \dots$$

• • • • •

$$\frac{\partial C_T}{\partial x_n} = \frac{\partial f_c}{\partial c_n} \cdot \frac{\partial c_n}{\partial x_n} = 0 \dots \dots \dots \frac{\partial C_T}{\partial x_n} = \frac{\partial f_c}{\partial c_n} \cdot \frac{\partial c_n}{\partial x_n} = 0$$

When the objective function is constrained, the method of Lagrange multipliers⁵ may be used. For the sake of simplicity, assume $C_T = f(C_1, C_2)$ and $C_1 = f_1(x_1)$ and $C_2 = f_2(x_2)$. The total cost, C_T , can now be represented as follows:

$$C_T = f(x_1, x_2) \quad (6)$$

Assume the variables x_1 and x_2 are constrained by the relationship, $\phi(x_1, x_2) = 0$. Using the method of Lagrange multipliers the following equations obtain:

$$\frac{\partial C_T}{\partial x_1} + \lambda' \frac{\partial \theta}{\partial x_1} = 0 \quad (7)$$

$$\frac{\partial C_T}{\partial x_2} + \lambda' \frac{\partial \theta}{\partial x_2} = 0 \quad (8)$$

$$\phi(x_1, x_2) = 0 \quad (9)$$

Equations 7, 8 and 9 contain 3 unknowns x_1, x_2 and λ' which can be solved for readily. Here, λ' is known as the undetermined multiplier.

⁵ I. S. Sokolnikoff and R. M. Redheffer, Mathematics of Physics and Modern Engineering, McGraw Hill Book Co., Inc., NY, 1958, Chap. 3, p. 250.

In order to continue with the problem as stated in Section II, one must now determine the form of the cost function for the treatment units and the necessary parameters to describe it. In the following section the cost function for the treatment units will be developed.

IV COST FUNCTION FOR TREATMENT UNITS

The cost functions of the treatment units to provide various degrees of treatment was presented in Section III as a function of several independent variables; namely,

$$c_i = f(x_i^1, x_i^2, x_i^3, \dots, x_i^r) \quad (4)$$

Thomas⁶ has postulated that for several types of conventional treatment processes, the degree of removal of a pollutant is a function of the influent flux of the waste, the discharge rate of the waste water, and the economic investment⁷ made to treat the waste; or expressed mathematically,

$$1 - p = \left[\frac{1}{1 + a \left(\frac{x_0}{Q} \right)^m \left(\frac{C}{Q} \right)} \right] \quad (10)$$

where p = proportion removed

x_0 = influent flux of pollutant

Q = discharge rate of waste water

C = total cost of unit⁷

a = parameter of the particular treatment unit

m = parameter of the particular treatment unit

Figure 2 shows a plot of proportion of waste remaining in the effluent ($1-p$) vs. cost per unit of waste water $\left(\frac{C}{Q}\right)$. The two curves A and B represent a low influent concentration and a high influent concentration of waste, respectively.

⁶ H. A. Thomas, Jr., Unpublished notes, Harvard University, 1963

⁷ "economic investment" implies initial cost of construction plus the present value cost of operation, maintenance, chemicals, etc.

FIG. 2 PROPORTION OF WASTE REMAINING IN EFFLUENT vs COST PER UNIT OF WASTE WATER TREATED

FIG. 3 COST OF TREATMENT PLANT 1 vs FLUX OF POLLUTANT IN EFFLUENT FOR GIVEN FLUX OF POLLUTANT IN INFLUENT

FIG. 4 COST OF TREATMENT PLANT 2 vs FLUX OF POLLUTANT IN INFLUENT FOR GIVEN FLUX OF POLLUTANT IN EFFLUENT

If p equals the proportion of waste removed, then the expression $(1-p)X_0 = X_1$ obtains for treatment plant 1, from Figure 1.

$$(1-p) = \frac{X_1}{X_0}$$

where X_1 is the flux of pollutant remaining in the effluent. Equation 10 for treatment plant 1 now takes the form:

$$\frac{X_1}{X_0} = \left[\frac{1}{1 + a_1 \left(\frac{X_0}{Q_1} \right)^{m_1} \left(\frac{C_1}{Q_1} \right)} \right] \quad (11)$$

$$a_1 \left(\frac{X_0}{Q_1} \right)^{m_1} \left(\frac{C_1}{Q_1} \right) = \frac{X_0 - X_1}{X_1} \quad (12)$$

It follows from equation (12) that:

$$C_1 = \left(\frac{Q_1}{a_1} \right) \left(\frac{Q_1}{X_0} \right)^{m_1} \left(\frac{X_0 - X_1}{X_1} \right) \quad (13)$$

Figure 3 shows a typical plot obtained from equation 13.

Similarly, the cost function for treatment plant 2 is:

$$C_2 = \left(\frac{Q_2}{a_2} \right) \left(\frac{Q_2}{Y_0} \right)^{m_2} \left(\frac{Y_0 - Y_1}{Y_1} \right) \quad (14)$$

where Y_1 is constrained, and Y_0 is the flux of pollutant in the water intake. Figure 4 shows a typical plot obtained from equation (14).

The effluent flux of pollutant from treatment plant 1, X_1 , is to be considered as a system parameter to be adjusted to maximize economic efficiency.

In the river, the pollutant, X , decays according to the logarithmic law so that the flux of pollutant at any time, t , is equal to:

$$X_t = e^{-\lambda t} X_1 \quad (15)$$

If the time of travel of water from the effluent of the waste treatment plant 1 to the intake at the water treatment plant 2 is T units of time, the intake flux of pollutant, Y_0 , equals;

$$Y_0 = \left[\frac{Q_2}{Q_R} \right] e^{-\lambda T} X_1 \quad (16)$$

where Q_2 = rate of influent to water treatment plant

Q_R = rate of flow in river

λ = parameter reflecting the self-purification of the river including hold-up, biological uptake and decay

T = time of travel from waste treatment outlet to the use point inlet

The quantity $\frac{Q_2}{Q_R} e^{-\lambda T}$ can be thought of as a constant for this problem and for convenience is denoted as b . Therefore, $Y_0 = b X_1$ (16a)

The total cost of the project, C_T , is the sum of the individual unit costs and is equal to:

$$C_T = C_1 + C_2 \quad (2)$$

$$C_T = \left(\frac{Q_1}{a_1} \right) \left(\frac{Q_1}{X_0} \right)^{m_1} \left(\frac{X_0 - X_1}{X_1} \right) + \left(\frac{Q_2}{a_2} \right) \left(\frac{Q_2}{Y_0} \right)^{m_2} \left(\frac{Y_0 - Y_1}{Y_1} \right) \quad (17)$$

V MATHEMATICAL DEVELOPMENT

In this section, the equation for the optimal flux of pollutant in the effluent of the waste treatment plant will be developed.

The total cost function, equation (2),

$$C_T = C_1 (X_1) + C_2 (Y_0) \quad (2)$$

is subject to the constraint

$$\emptyset = bX_1 - Y_0 = 0 \quad (16 a)$$

The Lagrange function for this is,

$$L = C_1(X_1) + C_2(Y_0) + \lambda'(bX_1 - Y_0) \quad (19)$$

$$\frac{\partial L}{\partial X_1} = \frac{\partial C_1}{\partial X_1} + \lambda' b = 0 \quad (20)$$

$$\frac{\partial L}{\partial Y_0} = \frac{\partial C_2}{\partial Y_0} - \lambda' = 0 \quad (21)$$

$$\frac{\partial L}{\partial \lambda} = bX_1 - Y_0 = 0 \quad (22)$$

From equations (20) and (21),

$$-\frac{\partial C_1}{\partial X_1} = b \frac{\partial C_2}{\partial Y_0} \quad (23)$$

$$\begin{aligned} \frac{\partial C_1}{\partial X_1} &= \left(\frac{Q_1}{a_1}\right) \left(\frac{Q_1}{X_0}\right)^{m_1} (X_0)^{-1} (X_1)^{-2} \\ &= -\left(\frac{Q_1}{a_1}\right) \left(\frac{Q_1}{X_0}\right)^{m_1} \left(\frac{X_0}{X_1^2}\right) \end{aligned} \quad (24)$$

$$\begin{aligned} \frac{\partial C_2}{\partial Y_0} &= \left(\frac{Q_2}{a_2}\right) (Q_2)^{m_2} (Y_0)^{-1-m_2} \left(\frac{Y_0}{Y_1} - 1\right) + \left(\frac{Q_2}{a_2}\right) (Q_2)^{m_2} (Y_0)^{-m_2} \left(\frac{1}{Y_1}\right) \\ &= \left(\frac{Q_2}{a_2}\right) \left(\frac{Q_2}{Y_1}\right)^{m_2} [(1-m_2)(Y_0)^{-m_2} - m_2(Y_1)(Y_0)^{-1-m_2}] \end{aligned} \quad (25)$$

$$-\frac{\partial C_1}{\partial X_1} = b \frac{\partial C_2}{\partial Y_0} \quad (23)$$

$$\left(\frac{Q_1}{a_1}\right) \left(\frac{Q_1}{X_0}\right)^{m_1} \left(\frac{X_0}{X_1}\right) = b \left(\frac{Q_2}{a_2}\right) \left(\frac{Q_2}{Y_1}\right)^{m_2} [(1-m_2)(Y_0)^{-m_2} - m_2(Y_1)(Y_0)^{-1-m_2}]$$

$$Y_0 = bX_1 \quad (16a)$$

$$\left[\frac{Q_1^{1+m_1} X_0^{1-m_1} Y_1 a_2 b^{m_2}}{Q_2^{1+m_2} a_1} \right] = (1-m_2) b (X_1)^{2-m_2} + m_2 (Y_1) (X_1)^{1-m_2} \quad (26)$$

From equation 26, the flux of pollutant in the effluent of the waste treatment plant, X_1 , can be solved for, thereby giving the degree of treatment in the waste treatment plant, 1, and the water treatment plant, 2, necessary to maximize economic efficiency.

VI NUMERICAL EXAMPLE AND RESULTS

Two numerical examples will be given in this section; namely, (1) an example that does not consider the possibility of a storage reservoir between the waste outlet and the use inlet and (2) an example that considers a reservoir of a given fixed size.

The assumptions in both of these problems are as follows:

$$a_1 = 0.833$$

$$a_2 = 10.5$$

$$m_1 = m_2 = 0.5$$

$$X_0 = 3750 \text{ curies/day}$$

$$Y_1 = k = .375 \text{ curies/day}$$

$$Q_1 = 1 \text{ mgd} = 3750 \text{ meters}^3/\text{day}$$

$$Q_2 = 1 \text{ mgd} = 3750 \text{ meters}^3/\text{day}$$

Example 1 - No Storage Reservoir $b = 0.01$

From equation (26),

$$\frac{(3750)^{1.5} (3750)^{0.5} (10.5)(0.01)^{0.5} (.375)}{(3750)^{1.5} (.833)} = (0.5)(0.01)X_1^{1.5} + (.5)(.375)X_1^{0.5}$$

$$28.9 = 0.005 X_1^{1.5} + 0.1875 X_1^{0.5}$$

$$X_1 = 300$$

$$Y_0 = b X_1 = 0.01 (300) = 3$$

From equation (13), the cost of the waste treatment plant is

$$C_1 = \left(\frac{3750}{0.833} \right) \left(\frac{3750}{3750} \right)^{0.5} \left(\frac{3750-300}{300} \right) = \$51,700$$

and from equation (14), the cost of the water treatment plant is

$$C_2 = \left(\frac{3750}{10.5} \right) \left(\frac{3750}{3} \right)^{0.5} \left(\frac{3-0.375}{0.375} \right) = \$88,400$$

The total cost is therefore,

$$C_T = C_1 + C_2 = \$51,700 + \$88,400 = \$140,100$$

Example 2 - With Storage Reservoir

$$b = 0.001$$

If a storage reservoir is employed, the mean time of travel, T , from the discharge point to the intake point will be increased. From equations (16) it can be shown that as T increases, the quantity $b = \left[\frac{Q_2}{Q_R} \right] e^{-\lambda T}$ decreases. The

introduction of a reservoir in this example was done only to demonstrate the effects of a reservoir on the total cost of treatment plants 1 and 2. It is assumed that in this example $b = 0.001$ as compared to $b = 0.01$ in example 1.

From equation (26),

$$\frac{(3750)^{1.5} (3750)^{0.5} (10.5) (0.001)^{0.5} (.375)}{(3750)^{1.5} (0.833)} = (0.5)(0.001)X_1^{1.5} + (0.5)(.375)X_1^{0.5}$$

$$9.19 = (0.0005)X_1^{1.5} + (0.1875)X_1^{0.5}$$

$$X_1 = 470$$

$$Y_0 = bX_1 = (0.001)(470) = 0.470$$

From equation (13), the cost of the waste treatment plant is

$$C_1 = \left(\frac{3750}{0.833} \right) \left(\frac{3750}{3750} \right)^{0.5} \left(\frac{3750-470}{470} \right) = \$31,400$$

and from equation (14), the cost of the water treatment plant is

$$C_2 = \left(\frac{3750}{10.5} \right) \left(\frac{3750}{.470} \right)^{0.5} \left(\frac{.470-.375}{.375} \right) = \$80,800$$

The total cost is therefore,

$$C_T = C_1 + C_2 = \$31,400 + \$80,800 = \$112,200$$

The difference in total cost of treatment plants 1 and 2 between example 1 and 2 is

$$\$140,100 - \$112,200 = \$27,900$$

indicating that for water quality control the dam has a benefit of \$27,900. In this example, it is assumed that the dam was built to a given fixed size ($b = 0.001$) for purposes other than just for water quality benefits.

Table I summarizes the costs of the individual treatment plants for example 1 and 2.

TABLE I
SUMMARY OF COSTS OF NUMERICAL EXAMPLES

Example	Release of Activity to Stream curies/day	Waste Treatment Cost C_1 , \$	Water Treatment Cost C_2 , \$	Total Cost C_T \$
1 No Dam	300	51,700	88,400	140,100
2 With Dam	470	31,400	80,800	112,200
Benefit for Dam = Difference				27,900

VII ERROR ANALYSIS AND PROBLEM IDENTIFICATION

Systems analysis is usually thought of as a problem solving tool. However, more properly it should be regarded equally or even more useful as a problem identification tool. By synthesizing the best available data in a theoretical construct, even though this information may be incomplete, and by carrying forward the mathematical simulation together with an error analysis and the calculation of confidence limits, it is possible to show quantitatively the effect upon practical conclusions of inadequacy of different types of input data and the cost of having to base decisions on poor rather than good data. The simulation results yield a rational basis for the evaluation of the relative importance of variables and classes of input data obtained from field and laboratory observations. This constitutes problem identification and provides a valuable frame of reference for planning and ranking of future-data-collection projects.

In this section, the effects of the various system parameters on the total cost of the project will be demonstrated in order to show that if more research is justified on the project, where it should be invested first.

In this simple problem, the total cost, C_T , is given as a function of several independent system parameters; namely

$$C_T = \phi (Q_1, Q_2, a_1, a_2, m_1, m_2, X_0, X_1, Y_1, b) \quad (27)$$

Any change in the total cost, dC_T , is given mathematically as

$$dC_T = \frac{\partial \phi}{\partial Q_1} dQ_1 + \frac{\partial \phi}{\partial Q_2} dQ_2 + \frac{\partial \phi}{\partial a_1} da_1 + \dots + \frac{\partial \phi}{\partial b} db \quad (28)$$

The quantities $\frac{\partial \phi}{\partial Q_1}$, $\frac{\partial \phi}{\partial Q_2}$, $\dots$, $\frac{\partial \phi}{\partial b}$ can be thought of as the set of constants, $\alpha_1, \alpha_2, \dots, \alpha_{10}$ so that equation (28) now takes the form

$$dC_T = \alpha_1 dQ_1 + \alpha_2 dQ_2 + \alpha_3 da_1 + \dots + \alpha_{10} db \quad (29)$$

The total differentials of each parameter, $dQ_1, dQ_2, \dots, db$ can be regarded as measures of the randomness of the data available for each parameter; the greater the spread of data about the respective mean values, the greater the magnitude of these differentials.

$$\text{Let } dC_T = (C_T - \mu_c), dQ_1 = (Q_1 - \mu_{Q_1}), \dots db = (b - \mu_b) \quad (30)$$

where $\mu_c, \mu_{Q_1}, \dots, \mu_b$ equal the population means of the individual parameters. If equation (30) is substituted into equation (29), the following equation results:

$$(C_T - \mu_c) \approx \alpha_1(Q_1 - \mu_{Q_1}) + \alpha_2(Q_2 - \mu_{Q_2}) + \alpha_3(a_1 - \mu_{a_1}) + \dots + \alpha_{10}(b - \mu_b) \quad (31)$$

By squaring both sides of equation (31) and taking the expected values,⁸ term by term, equation (32) results

$$\begin{aligned} E\{(C_T - \mu_c)^2\} &\approx E\{\alpha_1^2(Q_1 - \mu_{Q_1})^2\} + E\{2\alpha_1\alpha_2(Q_1 - \mu_{Q_1})(Q_2 - \mu_{Q_2})\} \\ &\quad + E\{\alpha_2^2(Q_2 - \mu_{Q_2})^2\} + E\{\alpha_3^2(a_1 - \mu_{a_1})^2\} + E\{2\alpha_1\alpha_3(Q_1 - \mu_{Q_1})(a_1 - \mu_{a_1})\} \\ &\quad + E\{2\alpha_2\alpha_3(Q_2 - \mu_{Q_2})(a_1 - \mu_{a_1})\} + \dots + E\{\alpha_{10}^2(b - \mu_b)^2\} \end{aligned} \quad (32)$$

Given⁽⁸⁾ that the expected values $E\{(x - \mu)\} = 0$ and $E\{(x - \mu)^2\} = \text{var } x = \sigma_x^2$ and that the parameters are statistically independent, equation (32) takes the form

$$\begin{aligned} \sigma_{C_T}^2 &\approx \alpha_1^2 E\{(Q_1 - \mu_{Q_1})^2\} + 2\alpha_1\alpha_2 E\{(Q_1 - \mu_{Q_1})\} E\{(Q_2 - \mu_{Q_2})\} \\ &\quad + \alpha_2^2 E\{(Q_2 - \mu_{Q_2})^2\} + \alpha_3^2 E\{(a_1 - \mu_{a_1})^2\} + 2\alpha_1\alpha_3 E\{(Q_1 - \mu_{Q_1})\} \times \\ &\quad E\{(a_1 - \mu_{a_1})\} + \dots + \alpha_{10}^2 E\{(b - \mu_b)^2\} \\ &\approx \alpha_1^2 \sigma_{Q_1}^2 + \alpha_2^2 \sigma_{Q_2}^2 + \alpha_3^2 \sigma_{a_1}^2 + \dots + \alpha_{10}^2 \sigma_b^2 \end{aligned} \quad (33)$$

⁸ D. A. S. Fraser, Statistics: An Introduction, John Wiley & Sons, Inc., New York, 1960.

By substituting the values of $\alpha_1, \alpha_2, \alpha_3, \dots, \alpha_{10}$ back into equation (33) the equation of the variance of the total cost is:(9)

$$\sigma_{C_T}^2 \approx \left(\frac{\partial \phi}{\partial Q_1} \right)^2 \sigma_{Q_1}^2 + \left(\frac{\partial \phi}{\partial Q_2} \right)^2 \sigma_{Q_2}^2 + \left(\frac{\partial \phi}{\partial a_1} \right)^2 \sigma_{a_1}^2 + \dots + \left(\frac{\partial \phi}{\partial b} \right)^2 \sigma_b^2 \quad (34)$$

From equation (34), it is observed that each error term has two factors; a variance in the data, σ_x^2 , and a weighting factor $\left(\frac{\partial \phi}{\partial x} \right)^2$. Equation (34) indicates

which parameters and to what extent these parameters affect the variance of the total cost $\sigma_{C_T}^2$. It is clear from this equation, that in general, the largest uncertainty pertaining to the estimated cost is associated with those parameters for which the products of the weighting factor and the variance in the data are the greatest.

For this simple example, the weighting factors are derived as follows:
Given that

$$c_1 = \frac{Q_1}{a_1} \left(\frac{Q_1}{X_0} \right)^{m_1} \left(\frac{X_0 - X_1}{X_1} \right) = \frac{Q_1^{(1+m_1)}}{a_1} \left(\frac{X_0^{(1-m_1)}}{X_1} - \frac{1}{X_0^{m_1}} \right) \quad (13)$$

$$c_2 = \frac{Q_2}{a_2} \left(\frac{Q_2}{bX_1} \right)^{m_2} \left(\frac{bX_1 - Y_1}{Y_1} \right) = \frac{Q_2^{(1+m_2)}}{a_2} \left(\frac{b^{(1-m_2)} X_1^{(1-m_2)}}{Y_1} - \frac{1}{b^{m_2} X_1^{m_2}} \right) \quad (14)$$

the following partial derivatives obtain:

9

A. Hald, Statistical Theory with Engineering Applications, John Wiley & Sons, Inc., New York, 1952, Chap. 5, p. 117.

$$\frac{\partial \theta}{\partial Q_1} = \frac{(1+m_1)}{a_1} \left(\frac{Q_1}{X_0} \right)^{m_1} \left(\frac{X_0 - X_1}{X_1} \right) \quad (35)$$

$$\frac{\partial \theta}{\partial Q_2} = \frac{(1+m_2)}{a_2} \left(\frac{Q_2}{bX_1} \right)^{m_2} \left(\frac{bX_1 - Y_1}{Y_1} \right) \quad (36)$$

$$\frac{\partial \theta}{\partial a_1} = - \frac{1}{a_1^2} \left[\left(\frac{Q_1^{(m_1+1)}}{X_0^{m_1}} \right) \left(\frac{X_0 - X_1}{X_1} \right) \right] \quad (37)$$

$$\frac{\partial \theta}{\partial a_2} = - \frac{1}{a_2^2} \left[\left(\frac{Q_2^{(m_2+1)}}{(bX_1)^{m_2}} \right) \left(\frac{bX_1 - Y_1}{Y_1} \right) \right] \quad (38)$$

$$\frac{\partial \theta}{\partial m_1} = \left(\frac{Q_1}{a_1} \right) \left(\frac{Q_1}{X_0} \right)^{m_1} \left(\frac{X_0 - X_1}{X_1} \right) \log_e \left(\frac{Q_1}{X_0} \right) \quad (39)$$

$$\frac{\partial \theta}{\partial m_2} = \left(\frac{Q_2}{a_2} \right) \left(\frac{Q_2}{bX_1} \right)^{m_2} \left(\frac{bX_1 - Y_1}{Y_1} \right) \log_e \left(\frac{Q_2}{bX_1} \right) \quad (40)$$

$$\frac{\partial \theta}{\partial X_0} = \left(\frac{Q_1}{a_1} \right) \left[\left(\frac{Q_1}{X_0} \right)^{m_1} \frac{1}{X_1} - m_1 \left(\frac{Q_1^{m_1}}{X_0^{(m_1+1)}} \right) \left(\frac{X_0 - X_1}{X_1} \right) \right] \quad (41)$$

$$\begin{aligned} \frac{\partial \theta}{\partial X_1} = & - \left(\frac{Q_1}{a_1} \right) \left(\frac{Q_1}{X_0} \right)^{m_1} \left(\frac{X_0}{X_1^2} \right) + \left(\frac{Q_2}{a_2} \right) \times \\ & \left[\left(\frac{Q_2}{bX_1} \right)^{m_2} \left(\frac{b}{Y_1} \right) - m_2 \left(\frac{Q_2^{m_2}}{b} \right) \left(\frac{1}{X_1^{(m_2+1)}} \right) \left(\frac{bX_1 - Y_1}{Y_1} \right) \right] \end{aligned} \quad (42)$$

$$\frac{\partial \theta}{\partial y_1} = - \left(\frac{Q_2}{a_2} \right) \left(\frac{Q_2}{bX_1} \right)^{m_2} \left(\frac{bX_1}{Y_1^2} \right) \quad (43)$$

$$\frac{\partial \theta}{\partial b} = \left(\frac{Q_2}{a_2} \right) \left[\left(\frac{Q_2}{bX_1} \right)^{m_2} \left(\frac{X_1}{Y_1} \right) - m_2 \left(\frac{Q_2}{X_1} \right)^{m_2} \left(\frac{1}{b^{(m_2+1)}} \right) \left(\frac{bX_1 - Y_1}{Y_1} \right) \right] \quad (44)$$

For numerical values of the parameters given in Section VI, the numerical values of the partial derivatives of equations (35) through (44) are as follows:

$$\alpha_1 = \frac{\partial \theta}{\partial Q_1} = \frac{(1+0.5)}{0.833} \left(\frac{3750}{3750} \right)^{0.5} \left(\frac{3750-300}{300} \right) = 20.7$$

$$\alpha_2 = \frac{\partial \theta}{\partial Q_2} = \frac{(1+0.5)}{10.5} \left(\frac{3750}{3} \right)^{0.5} \left(\frac{3-0.375}{0.375} \right) = 35.2$$

$$\alpha_3 = \frac{\partial \theta}{\partial a_1} = - \frac{1}{(0.833)^2} \left[\left(\frac{(3750)^{1.5}}{(3750)^{0.5}} \right) \left(\frac{3750-300}{300} \right) \right] = - 62,600$$

$$\alpha_4 = \frac{\partial \theta}{\partial a_2} = - \frac{1}{(10.5)^2} \left[\left(\frac{(3750)^{1.5}}{(3)^{0.5}} \right) \left(\frac{3-0.375}{0.375} \right) \right] = - 8,450$$

$$\alpha_5 = \frac{\partial \theta}{\partial m_1} = \left(\frac{3750}{0.833} \right) \left(\frac{3750}{3750} \right)^{0.5} \left(\frac{3750-300}{300} \right) \log_e \left(\frac{3750}{3750} \right) = 0$$

$$\alpha_6 = \frac{\partial \theta}{\partial m_2} = \left(\frac{3750}{10.5} \right) \left(\frac{3750}{3} \right)^{0.5} \left(\frac{3-0.375}{0.375} \right) \log_e \left(\frac{3750}{3} \right) = 634,000$$

$$\alpha_7 = \frac{\partial \theta}{\partial x_0} = \left(\frac{3750}{0.833} \right) \left[\left(\frac{3750}{3750} \right)^{0.5} \frac{1}{300} - (0.5) \left(\frac{(3750)^{0.5}}{(3750)^{1.5}} \right) \times \left(\frac{(3750-300)}{300} \right) \right] = 8.14$$

$$\alpha_8 = \frac{\partial \theta}{\partial x_1} = - \left(\frac{3750}{0.833} \right) \left(\frac{3750}{3750} \right)^{0.5} \left(\frac{3750}{(300)^2} \right) + \left(\frac{3750}{10.5} \right) \left[\left(\frac{3750}{3} \right)^{0.5} \left(\frac{0.01}{0.375} \right) - (0.5) \left(\frac{3750}{0.01} \right)^{0.5} \left(\frac{1}{(300)^{1.5}} \right) \left(\frac{3-0.375}{0.375} \right) \right] = 0$$

$$\alpha_9 = \frac{\partial \theta}{\partial Y_1} = - \left(\frac{3750}{0.833} \right) \left(\frac{3750}{3} \right)^{0.5} \left(\frac{3}{(0.375)^2} \right) = - 272,000$$

$$\alpha_{10} = \frac{\partial \theta}{\partial b} = \left(\frac{3750}{10.5} \right) \left[\left(\frac{3750}{3} \right)^{0.5} \left(\frac{300}{0.375} \right) - (0.5) \left(\frac{3750}{300} \right)^{0.5} \left(\frac{1}{(0.01)^{1.5}} \right) \left(\frac{3-0.375}{0.375} \right) \right] = 5,760,000$$

Table II shows the various weighting factors, $(\alpha_i)^2$ and error terms $[(\alpha_i)^2 \sigma_i^2]$ for the analysis of the variance of the total cost of example 1 of Section VI. The relative importance of the various error terms is demonstrated in column (7) where each parameter is ranked in order of relative importance. From this column, it can be seen that the parameter which affects the total cost of the project most is m_2 . This parameter is relatively six times as important as the next most important parameter, Q_2 . The remaining parameters, ranked in order of effect on the variance of total cost, are, Y_1 , a_1 , b , and so on.

This analysis ranks each parameter in importance relative to the other parameters and therefore indicates a priority on future data collection projects. It does not imply, however, how much of an economic investment should be made on one parameter relative to the investments made on the others. The final decision as to the allocation of investment in additional research and development would have to be based not only upon the foregoing considerations (weighting factor and variance in data) but also upon the relative cost of obtaining improved precision for each parameter. This is discussed in Section VIII.

TABLE II
WEIGHTING FACTORS & ERROR TERMS FOR THE VARIANCE OF TOTAL COST

Parameter	α_1	Weighting Factors $[\alpha_i]^2$	σ_i	$\text{Var}^{(10)}_{\sigma_i}$	Error Terms $(\text{Var}) (\alpha_i)^2$	Rank
(1)	(2)	(3)	(4)	(5)	(6)	(7)
Q_1	20.7	4.28×10^2	375	1.4×10^5	6.0×10^7	5
Q_2	35.2	1.24×10^3	375	1.4×10^5	1.735×10^8	2
a_1	-62,600	3.92×10^9	0.0833	6.92×10^{-3}	2.71×10^7	7
a_2	-8,450	7.12×10^7	1.05	1.1	7.83×10^7	4
m_1	0	0	0.05	2.5×10^{-3}	0	9
m_2	634,000	4.0×10^{11}	0.05	2.5×10^{-3}	1.0×10^9	1
X_0	8.14	6.6×10^1	375	1.4×10^5	9.25×10^6	8
X_1	0	0	30	9×10^2	0	10
Y_1	-272,000	7.4×10^{10}	0.0375	1.4×10^{-3}	1.04×10^8	3
b	5,760,000	3.32×10^{13}	0.001	1×10^{-6}	3.32×10^7	6

¹⁰ the variance is estimated from the mean for demonstration purposes assuming a coefficient of variation of 0.10 ($C_V = \frac{\sigma}{M} = 0.10$)

$$\sigma_{C_T}^2 = \sum_{i=1}^{10} (\alpha_i)^2 \sigma_i^2 = 14.85 \times 10^8$$

$$\sigma_{C_T} = (14.85 \times 10^8)^{1/2} = 3.85 \times 10^4 = \$38,500$$

VIII DECISION PROCESS FOR REFINEMENT OF TOTAL COST ESTIMATE⁽¹¹⁾

As mentioned in the previous section, the final decision as to apportionment of investment in future data collecting projects would have to be based not only on considerations discussed in the previous section (weighting factor and variance in data) but also upon the relative cost of obtaining improved precision for each parameter.

Initial Investment

Assume that by investing u_i dollars in additional research and development of parameter, i , the reduction in the standard deviation of that parameter, $\Delta\sigma_i$ will be

$$\Delta\sigma_i = \sigma_i(0) - \sigma_i(u_i) \quad (45)$$

From equation (34)

$$\sigma_{C_T}(u_{Q_1}, \dots, u_b) = \left\{ \left[\frac{\partial \theta}{\partial Q_1} \right]^2 \sigma_{Q_1}^2(u_{Q_1}) + \left[\frac{\partial \theta}{\partial Q_2} \right]^2 \sigma_{Q_2}^2(u_{Q_2}) + \dots + \left[\frac{\partial \theta}{\partial b} \right]^2 \sigma_b^2(u_b) \right\}^{1/2} \quad (46)$$

By squaring both sides and differentiating with respect to u_i , equation (46) becomes

$$\sigma_{C_T} d\sigma_{C_T} = \left[\left(\frac{\partial \theta}{\partial Q_1} \right)^2 \sigma_{Q_1} \frac{\partial \sigma_{Q_1}}{\partial u_{Q_1}} du_{Q_1} + \left(\frac{\partial \theta}{\partial Q_2} \right)^2 \sigma_{Q_2} \frac{\partial \sigma_{Q_2}}{\partial u_{Q_2}} du_{Q_2} + \dots + \left(\frac{\partial \theta}{\partial b} \right)^2 \sigma_b \frac{\partial \sigma_b}{\partial u_b} du_b \right] \quad (47)$$

¹¹ Harold A. Thomas, Jr., Decision Process for Refinement of Cost and Benefit Estimates in Preliminary Screening and Exploration Studies of River Basin Development Plans, unpublished memorandum, Harvard Water Resources Seminar, December 19, 1962.

By dividing both sides of equation (47) by σ_{C_T} and letting the terms

$$\left(\frac{\partial \theta}{\partial Q_1} \right)^2 \frac{1}{\sigma_{C_T}}, \left(\frac{\partial \theta}{\partial Q_2} \right)^2 \frac{1}{\sigma_{C_T}}, \dots \left(\frac{\partial \theta}{\partial b} \right)^2 \frac{1}{\sigma_{C_T}}$$

equal $\beta_1, \beta_2, \dots, \beta_{10}$, equation (48) obtains

$$d\sigma_{C_T} = \left[\beta_1 \sigma_{Q_1} \frac{\partial \sigma_{Q_1}}{\partial u_{Q_1}} du_{Q_1} + \beta_2 \sigma_{Q_2} \frac{\partial \sigma_{Q_2}}{\partial u_{Q_2}} du_{Q_2} + \dots + \beta_{10} \sigma_b \frac{\partial \sigma_b}{\partial u_b} du_b \right] \quad (48)$$

Hopefully, the standard deviation of the individual parameters decreases as the dollar investment is increased. The general shape of this function is shown on Figure 5.

The values of $\beta_1, \beta_2, \dots, \beta_{10}$ are all positive values and can be calculated from Table II, Section VII. As shown on Figure 5, the values of

$\frac{\partial \sigma_{Q_1}}{\partial u_{Q_1}}, \frac{\partial \sigma_{Q_2}}{\partial u_{Q_2}}, \dots, \frac{\partial \sigma_b}{\partial u_b}$ are all negative. Also the values of $\sigma_{Q_1}, \sigma_{Q_2}, \dots$

σ_b are positive and are shown on Table II, column (4) for an initial value of σ_i corresponding to a zero additional investment [$\sigma_i(0)$]. Therefore, it can be seen from equation (48) that the change in the standard deviation of total cost, $d\sigma_{C_T}$, is necessarily a negative quantity indicating that with some investment,

$du_{Q_1}, du_{Q_2}, \dots, du_b$, the magnitude of the standard deviation of total cost, σ_{C_T} , will decrease.

The objective of this type of analysis is to attain the greatest reduction in σ_{C_T} , ($\max \{-d\sigma_{C_T}\}$), for a given amount of money, u_i . From equation (48) the most attractive investment will be made on that parameter having the minimum value (maximum absolute value) of the coefficient of du_i .

In other words,

FIG. 5 CURVE SHOWING REDUCTION IN STANDARD DEVIATION, σ_i WITH INCREASE IN INVESTMENT, u_i

$$\text{Min} \left\{ \left(\frac{\partial \emptyset}{\partial Q_1} \right)^2 \frac{\sigma_{Q_1}}{\sigma_{C_T}} \frac{\partial \sigma_{Q_1}}{\partial u_{Q_1}}, \left(\frac{\partial \emptyset}{\partial Q_2} \right)^2 \frac{\sigma_{Q_2}}{\sigma_{C_T}} \frac{\partial \sigma_{Q_2}}{\partial u_{Q_2}}, \dots, \left(\frac{\partial \emptyset}{\partial b} \right)^2 \frac{\sigma_b}{\sigma_{C_T}} \frac{\partial \sigma_b}{\partial u_b} \right\} \quad (49)$$

or

$$\text{Max} \left\{ \left| \left(\frac{\partial \emptyset}{\partial Q_1} \right)^2 \frac{\sigma_{Q_1}}{\sigma_{C_T}} \frac{\partial \sigma_{Q_1}}{\partial u_{Q_1}} \right|, \dots, \left| \left(\frac{\partial \emptyset}{\partial b} \right)^2 \frac{\sigma_b}{\sigma_{C_T}} \frac{\partial \sigma_b}{\partial u_b} \right| \right\} \quad (50)$$

In ranking parameters in the optimal order for additional research toward refining the total cost of a project, a parameter will have a high priority for further study if (a) it has a large weighting factor, $\left(\frac{\partial \emptyset}{\partial X_i} \right)^2$; (b) it has a large

error, σ_i ; and (c) if the cost of reducing the error, $\frac{\partial u_i}{\partial \sigma_i}$, is small.

Table III shows the various weighting factors, α_i^2 , errors, σ_i , and relative costs of decreasing the errors, $\frac{\partial \sigma_i}{\partial u_i}$, for the various parameters.

The ranking of parameters in optimal order of addition investment is demonstrated in column (7). From this column, it can be seen that the most profitable investment for the reduction of the error in the total cost of the project, σ_{C_T} , is on the parameter Y_1 . Note that this is different from the parameter

ranked first in the previous section indicating that the cost of refining date is an important factor in the over-all analysis. Here, the cost of reducing the error, $\frac{\partial u_i}{\partial \sigma_i}$, was assumed for each parameter in order that the method may be eluci-

dated. This ranking, however, implies only where the initial investment should be made, for after the initial investment is made, the priority terms,

$\left[\left(\frac{\partial \emptyset}{\partial X_i} \right)^2 \frac{\sigma_i}{\sigma_{C_T}} \frac{\partial \sigma_i}{\partial u_i} \right]$, should be recalculated as shown on Table III and a

new ranking of parameters established. The next section will discuss the proper allocation of funds to each parameter under a budget constraint of U dollars.

TABLE III
PRIORITY TERMS FOR OPTIMAL RANKING OF PARAMETERS
FOR ADDITIONAL RESEARCH

Para- meter	Weighting Factor $[\alpha_i]^2$	σ_i	(12) $\frac{\sigma_i}{\sigma_{C_T}}$	(13) $\left(\frac{\partial \sigma_i}{\partial u_i}\right)$	Priority Term $[\alpha_i]^2 \frac{\sigma_i}{\sigma_{C_T}} \frac{\partial \sigma_i}{\partial u_i}$	Rank
(1)	(2)	(3)	(4)	(5)	(6)	(7)
Q_1	4.28×10^2	3.75×10^2	9.75×10^{-3}	3.75×10^1	1.57×10^2	4
Q_2	1.24×10^3	3.75×10^2	9.75×10^{-3}	3.75×10^1	4.53×10^2	2
a_1	3.92×10^9	8.33×10^{-2}	2.16×10^{-6}	8.33×10^{-4}	7.05	8
a_2	7.12×10^7	1.05	2.73×10^{-5}	1.05×10^{-2}	2.04×10^1	6
m_1	0	5.0×10^{-2}	1.3×10^{-6}	5.0×10^{-4}	0	9
m_2	4.0×10^{11}	5.0×10^{-2}	1.3×10^{-6}	5.0×10^{-4}	2.6×10^2	3
X_0	6.6×10^1	3.75×10^2	9.75×10^{-3}	3.75×10	2.41	5
X_1	0	3.0×10	7.8×10^{-4}	3.0×10^{-1}	0	10
Y_1	7.4×10^{10}	3.75×10^{-2}	9.75×10^{-7}	3.75×10^{-2}	2.7×10^3	1
b	3.32×10^{13}	1.0×10^{-3}	2.6×10^{-8}	1.0×10^{-5}	8.63	7

¹² The value of σ_{C_T} is taken from Table II

¹³ The relative cost of decreasing the errors in each parameter, $\left(\frac{\partial \sigma_i}{\partial u_i}\right)$, is assumed here for demonstration purposes.

Allocation of Funds to Each Parameter Under a Budget Constraint

The previous section, Initial Investment, provided a means for optimally ranking the various parameters for further initial investment. However, nothing was said about the optimal allocation of funds on research on the various parameters to maximize the reduction in error of total cost of the project, σ_{CT} , under a budget constraint of some fixed amount, U . This can be determined by Lagrangian analysis.

From equation (34) and the constraint,

$$U = \sum_{i=1}^n u_i = u_{Q_1} + u_{Q_2} + \dots + u_b \quad (51)$$

the Lagrangian function obtains as equation (52)

$$L = \sqrt{\left[\frac{\partial \sigma}{\partial Q_1}\right]^2 \sigma_{Q_1}^2 (u_{Q_1}) + \dots + \left[\frac{\partial \sigma}{\partial b}\right]^2 \sigma_b^2 (u_b) + \lambda(U - \sum u_i)} \quad (52)$$

$$\left. \begin{aligned} \frac{\partial L}{\partial u_{Q_1}} &= \sigma_{CT} \left[\frac{\partial \sigma}{\partial Q_1}\right]^2 \sigma_{Q_1} \frac{\partial \sigma_{Q_1}}{\partial u_{Q_1}} - \lambda = 0 \\ \frac{\partial L}{\partial u_{Q_2}} &= \sigma_{CT} \left[\frac{\partial \sigma}{\partial Q_2}\right]^2 \sigma_{Q_2} \frac{\partial \sigma_{Q_2}}{\partial u_{Q_2}} - \lambda = 0 \\ &\vdots \\ \frac{\partial L}{\partial u_b} &= \sigma_{CT} \left[\frac{\partial \sigma}{\partial b}\right]^2 \sigma_b \frac{\partial \sigma_b}{\partial u_b} - \lambda = 0 \end{aligned} \right\} \quad (53)$$

Solution of equations (53) give the following first order conditions for optimal allocation of funds

$$\left[\frac{\partial \theta}{\partial Q_1} \right]^2 \sigma_{Q_1} \frac{\partial \sigma_{Q_1}}{\partial u_{Q_1}} = \left[\frac{\partial \theta}{\partial Q_2} \right]^2 \sigma_{Q_2} \frac{\partial \sigma_{Q_2}}{\partial u_{Q_2}} = \dots = \left[\frac{\partial \theta}{\partial b} \right]^2 \sigma_b \frac{\partial \sigma_b}{\partial u_b} \quad (54)$$

These, together with the constraint of equation (51), fix the optimal allocation.

This analysis is shown graphically on Figure 6. The values of u_i on the upper diagram determine the optimum allocation of funds.

IX SUMMARY AND CONCLUSIONS

The simplified economic decision making model exemplified in this paper illustrates some of the important features of systems analysis such as (1) optimal economic design; (2) problem identification, which quantitatively ranks the various system parameters in order of relative importance on total cost of the project; (3) error analysis, which demonstrates the effect of errors in the estimate of the various system parameters on the precision of the estimate of total cost; and (4) optimal allocation of funds for future data collection projects on the various system parameters.

Generally, water resource systems are much more complex than the one presented here and consist of (1) many use points rather than the one; (2) numerous waste discharge points rather than one; (3) stochastic inputs for hydrology and waste effluents rather than the simplified deterministic inputs, (4) complex combinations of wastes rather than single isotopes, etc. Complex problems such as these employ such methods as finite calculus, queueing theory, probability theory, finite Markov chains, etc. in conjunction with mathematical models⁽¹⁴⁾ and computer simulation⁽¹⁵⁾ as tools for their solution. These so-called modern techniques are only extensions of the conventional mathematical techniques elucidated in this paper. However, the underlying approach is identical for both, the former being more sophisticated than the latter.

¹⁴ mathematical model: a formalized symbolic expression or description of a casual situation which has engendered observed data.

¹⁵ simulation method: a mathematical system that is analogous to a physical system under study and which may be manipulated to produce output data closely similar to observed data. The variables of interest in the former correspond to physical variables such as flow of water or heat. Also there is a more or less complete correspondence of elements in the two systems (for example, the flow of a stream may be reproduced as a stochastic process).

FIG. 6 GRAPHICAL DETERMINATION OF OPTIMAL ALLOCATION OF FUNDS UNDER A BUDGET CONSTRAINT

For an optimal economic design of this simplified deterministic system, both the total cost function, equation (17) developed in Section IV, and equation (26) of Section V, which gives the optimal flux of waste in the effluent of the waste treatment plant, are required as demonstrated with numerical examples in Section VI.

$$\frac{Q_1^{1+m_1} X_0^{1-m_1} Y_1 a_2^{m_2}}{Q_2^{1+m_2} a_1} = (1-m_2) b (X_1)^{2-m_2} + m_2 (Y_1) (X_1)^{1-m_2} \quad (26)$$

$$C_T = \left[\frac{Q_1}{a_1} \right] \left[\frac{Q_1}{X_0} \right]^{m_1} \left[\frac{X_0 - X_1}{X_1} \right] + \left[\frac{Q_2}{a_2} \right] \left[\frac{Q_2}{b X_1} \right]^{m_2} \left[\frac{b X_1 - Y_1}{Y_1} \right] \quad (17)$$

For a given set of deterministic values for the various system parameters, equation (26) can be solved for the optimal flux of waste in the effluent of the waste treatment plant, X_1 . This optimal value of X_1 is substituted into equation (17) in order to determine the minimum total cost of the project, C_T .

Inherent in the measurements of the magnitude of the individual system parameters are errors. Assuming that these errors are in precision rather than of bias, equation (34) obtains.

$$\sigma_{C_T}^2 = \left(\frac{\partial \phi}{\partial Q_1} \right)^2 \sigma_{Q_1}^2 + \left(\frac{\partial \phi}{\partial Q_2} \right)^2 \sigma_{Q_2}^2 + \dots + \left(\frac{\partial \phi}{\partial b} \right)^2 \sigma_b^2 \quad (34)$$

Imprecision derives from insufficient basic data. Additional data collection will therefore improve the precision of the various system parameters and consequently the estimate of total cost. Equation (34) shows that the variance in total cost of the project is equal to the sum of products of a weighting factor and the variance in the data for each system parameter. This equation points out clearly which parameters affect the variance of total cost most and by how much relatively to one another thereby indicating the "weak links" in the analysis. This constitutes problem identification, implying a priority ranking of parameters to be investigated in future data collection projects.

The final decision as to the apportionment of investment in future data collection projects would have to be based, however, not only on considerations of weighting factor and errors in the data of each parameter but also on the estimated relative cost of obtaining improved precision for these parameters.

Equation (48) shows the relationship between the weighting factor, $\left(\frac{\partial \phi}{\partial x_i} \right)^2$,

the error in the data, σ_i , and the relative cost of obtaining improved precision,

$\left(\frac{\partial u_i}{\partial \sigma_i}\right)$ for each parameter.

$$d\sigma_{C_T} = \sum_{i=1}^{10} \left(\frac{\partial \theta}{\partial x_i} \right)^2 \frac{\sigma_i}{\sigma_{C_T}} \frac{\partial \sigma_i}{\partial u_i} du_i \quad (48)$$

Clearly, the most profitable investment will be made on that parameter with the greatest absolute value of the coefficient of du_i , or in other words,

$$\text{Max} \left\{ \left| \left(\frac{\partial \theta}{\partial x_i} \right)^2 \frac{\sigma_i}{\sigma_{C_T}} \frac{\partial \sigma_i}{\partial u_i} \right| \right\} \quad (50)$$

The optimal allocation of funds for further research under a budget constraint will obtain when these coefficients are equal as shown by equation (54). The final solution of equation (54) is, of course, subject to the budget constraint, equation (51).

$$U = \sum_{i=1}^n u_i \quad (51)$$

$$\left(\frac{\partial \theta}{\partial Q_1} \right)^2 \sigma_{Q_1} \frac{\partial \sigma_{Q_1}}{\partial u_{Q_1}} = \left(\frac{\partial \theta}{\partial Q_2} \right)^2 \sigma_{Q_2} \frac{\partial \sigma_{Q_2}}{\partial u_{Q_2}} = \dots = \left(\frac{\partial \theta}{\partial b} \right)^2 \sigma_b \frac{\partial \sigma_b}{\partial u_b} \quad (54)$$

In Sections VII and VIII of this paper, the relative precision of the data of the various parameters was discussed but nothing was said about the bias of the values of the various parameters. The parameter subject most to bias is probably the estimate of the future quantity of water for municipal use, Q_2 . Figure 7 shows a curve of total cost of project, C_T , vs. quantity of water for municipal use, Q_2 , both for unconstrained values of X_1 and for a value of X_1 constrained at 200 curies per day. If the actual quantity of water, Q_2 , in say 1975 is not equal to 3750 meters³/day as designed but say 2000, the approximate unnecessary cost of the water and waste treatment plants would be \$140,000 - 74,400 = \$65,700. See Table IV. This figure demonstrates clearly the cost of overestimating the future population and the saving incurred with more conservative (unbiased) estimates of future water demands.

FIG. 7 VARIATION IN TOTAL COST WITH CHANGE IN Y_1 & Q_2 FOR BOTH X_1 UNCONSTRAINED AND CONSTRAINED AT 200

TABLE IV
VARIATION IN TOTAL COST WITH CHANGE IN Y_1 & Q_2 ;
(X_1 NOT CONSTRAINED)

Parameter	Value of Parameter	Flux of Waste in Effluent of Plant 1, X_1	Cost 1	Cost 2	Total Cost of Project, C_T
(1)	(2)	(3)	(4)	(5)	(6)
Y_1	. 200 curies/day	185	\$86, 800	\$132, 500	\$219, 300
	. 300	255	61, 600	103, 000	164, 600
	. 375	300	51, 700	88, 400	140, 100
	. 500	370	41, 100	73, 500	114, 600
	. 750	485	30, 300	53, 100	83, 400
	1. 20	670	20, 700	38, 800	59, 500
Q_2	10, 000 meters ³ /day	98	\$168, 000	\$155, 000	\$323, 000
	5, 000	220	72, 200	109, 000	181, 200
	3, 750	300	51, 700	88, 400	140, 100
	3, 000	385	39, 300	73, 500	112, 800
	2, 000	565	25, 400	49, 000	74, 400

TABLE V
VARIATION IN TOTAL COST WITH CHANGE IN Y_1 & Q_2 ;
(X_1 CONSTRAINED AT 200)

Parameter	Value of Parameter	Flux of Waste in Effluent of Plant 1, Y_1	Cost 1	Cost 2	Total Cost of Project, C_T
(1)	(2)	(3)	(4)	(5)	(6)
Y_1	. 200	185	\$86, 800	\$132, 500	\$219, 300
	. 300	200	80, 000	87, 500	167, 500
	. 375	200	80, 000	67, 000	147, 000
	. 500	200	80, 000	46, 300	126, 300
	. 750	200	80, 000	25, 800	105, 800
	1. 20	200	80, 000	10, 300	90, 300
Q_2	10, 000	98	\$168, 000	\$155, 000	\$323, 000
	5, 000	200	80, 000	103, 800	183, 800
	3, 750	200	80, 000	67, 100	147, 100
	3, 000	200	80, 000	47, 900	127, 900
	2, 000	200	80, 000	26, 000	106, 000

The value of the constraint, Y_1 , set by public health authorities, has a significant effect on the total cost of the project, as shown on Figure 7 for values of Y_1 from 0.2 to 1.2 curies/day. Setting these constraints at a lower tolerance level than economically justified can be quite costly. For example, the cost of changing the constraint under the given conditions from 0.375 curies/day to 0.2 curies/day amounts to $\$219,300 - \$140,100 = \$79,200$ or a 56.4 percent increase in the original cost.

For many years, decisions have been made as to water quality criteria and standards; for example, in the United States, drinking water should have an average coliform concentration not greater than about one organism per 100 milliliters. This type of criteria and many others like it represent socio-economic decisions and reflect in each case the inherent tension between human desires and human capabilities.⁽¹⁶⁾ "The setting of any quality criterion or standard relating to health and well being inevitably entails making an implicit estimate of a cost:benefit ratio based on whatever data or other factors are available for judgment. In situations where the costs are known within reasonable limits, the establishment of a standard amounts to assigning a certain utility to human life, health or well being."⁽¹⁶⁾

Now if the benefits, measured in terms of utility, derived from this water quality constraint are less than the benefits derived from an equal investment in medical research, highways, schools, or other sectors of the public health budget, these people are constrained to drink over-pure water. If in this problem, the tolerance level of the constraint is increased from 0.375 to 0.750 curies/day the saving would be $\$140,100 - \$83,400 = \$56,700$. If this saving would give a greater return if invested in another sector of the public health budget pertaining to radiological health, it may be economically justifiable to relax the existing constraint to 0.750 curies/day.

Figure 7 also shows the effect of a constraint on the flux of pollutant, X_1 , discharged to the stream with the effluent of the waste treatment plant. In this example, the flux is constrained at 200 curies/day. This restriction becomes more costly as the value of the constraint, Y_1 , is relaxed. Are the benefits derived by this constraint ($X_1 = 200$) greater than the benefits that might be derived from an equal investment into another sector of the radiological health budget?

ACKNOWLEDGMENT

This mathematical model has been drawn from a report, "Operations Research in Liquid Radioactive Waste Disposal" (June 1963) by the Harvard Water Resources Group based on research (Contract 44-720-7140) sponsored by the Environmental and Sanitary Engineering Branch, Division of Reactor Development, U. S. Atomic Energy Commission.

¹⁶ Harold A. Thomas, Jr., The Animal Farm: A Mathematical Model for the Discussion of Social Standards for Control of the Environment, The Quarterly Journal of Economics, Vol. LXXVII, Harvard University, February 1963.

LIST OF SYMBOLS

HYDROLOGIC PARAMETERS

Q'_R = Flow in River above Waste Treatment Outfall - meters³/day

Q_R = Flow in River between Waste Treatment Outfall and Water Treatment Intake

Q_1 = Quantity of Waste Discharge at Waste Treatment Plant - meters³/day

Q_2 = Quantity of Water for Municipal Use - meters³/day

WASTE PARAMETERS

X_O = Flux of Waste before Waste Treatment - curies/day

X_1 = Flux of Waste after Waste Treatment - curies/day

Y_O = Flux of Waste in Water Treatment Influent - curies/day

Y_1 = Flux of Waste in Water Treatment Effluent - curies/day

TREATMENT PARAMETERS

a_1 = Parameter of the Waste Treatment Plant

a_2 = Parameter of the Water Treatment Plant

m_1 = Parameter of the Waste Treatment Plant

m_2 = Parameter of the Water Treatment Plant

b = $\left[\frac{Q_2}{Q_R} \right] e^{-\lambda T}$ = Parameter of the River

λ = Parameter Including Effects of Hold-up and Decay

T = Mean time of Travel of Water from Waste Treatment Discharge to Water Treatment Intake

K = Maximum Permissible Flux of Waste in Municipal Water Supply

COST PARAMETERS

C_1 = Present Value Cost of Constructing and Operating Waste Treatment Plant

C_2 = Present Value Cost of Constructing and Operating Water Treatment Plant

C_T = $C_1 + C_2$ = Present Value Total Cost of Constructing and Operating Plants 1 and 2

DISCUSSION

BELTER: Do you think from what you have heard in the past two days, that based on the data presented by both Hanford and Oak Ridge, they could go through some kind of analysis similar to this?

THOMAS: Yes, I am sure of it. As a matter of fact, as soon as the parameters are identified.

BERNARD: Will we have to provide a better background information than we have now?

SPOFFORD: The answer to that question is difficult at this time. One of the great innovations of systems analysis lies in its ability to identify which and by just how much each of the input parameters affect the desired objective, thus ranking the parameters in relative importance. If we find, for example, that one of the input parameters only affects the objective slightly over wide ranges of that parameter, this would indicate that it would not pay to invest much time or money into collecting data for this parameter. In other words, there is necessarily an economical balance between the cost of getting more data, and the benefits, in terms of the objective, derived from receiving this new data.

COWSER: Is this in terms of quality of water at a particular point within the system?

SPOFFORD: Yes, at any point that you would want.

COWSER: Presumably, one would have to extend beyond that point as a simple model. Simply consider consuming water for drinking purposes. This may be about one of a half dozen avenues of exposure. I assume now that one can expand on this model to include all evidence, and one might reverse it in the sense that you can define the problem in terms of dose rather than costs.

THOMAS: The answer to your first question is yes. You must have more use points. The answer to your second question is no; we would want to assume that these factors of 10% are ultimate. In our look into conventional practices in hydrology, we found rules of thumb developed in a technology that is now outmoded and in an economy that is bygone. We want to re-evaluate the

practices now. Ultimately, when we get enough systems analyzed we can come up with more rational methods. You can see from the amount that we have mentioned, that the amount to be released to streams is dependent upon what is permitted to pass the control point. The control point indicates what radioactivity goes into people. We must accept the opinions of radiologists and physiologists about health hazards inside the body that result from ingestion of radioactive substances.

KAUFMAN: Wouldn't you have to say that this is more applicable in Hayden's economy than it is today? In other words, in a sense, economy seemed to make more difference in Hayden's time than it does today.

THOMAS: In systems analysis you can build-in social objectives as well as economic objectives and you can incorporate budgetary constraints as well as institutional constraints. Whenever you have uncertainty it ultimately costs you money. Systems analysis pinpoints this and provides the incentive to go after needed research.

GLOYNA: To a certain degree you have to identify these objectives, and in order to do this you have to look at these weighting factors. I wonder if you would expand a little on how you test the weighting factors.

THOMAS: Well, when I wrote the series for the variants of the overall costs on the blackboard I had a large number of terms, some of which were major, some of which were minor. The major ones are the ones that are holding us up at present. These uncertainties or imperfections in design data have a large dollar cost.

GLOYNA: In setting these up you have to arrive at a method of weighting them.

THOMAS: The weighting factors are derived from an error analysis of the technological function.

KAUFMAN: Do you feel that the analog has any advantage in this type of problem?

THOMAS: We do feel that it has some advantages clearly. I believe both digital and analogue computers are useful. In Pakistan we have been working on the design of an enormous well field. The analogue that Mr. Skibitzky of the USGS was running at Phoenix was useful in predicting trends over times of fifty or a hundred years; also it had capacity to cover an enormous area of aquifer. Our digital computer program which simulated as many as one hundred wells at a time could take into account non-linear relations such as obtain between evaporation rate and depth of ground water. Moreover, cost-benefit functions are not linear but this is easily taken care of in the digital computer.

FOSTER: It appears to me that, in the complete model that you have here,

there is one peculiar feature in the economic aspect. This is the fact that you are making the evaluation both on the cost of the waste treatment plant and of the water treatment clean-up for a community. Since most communities have different ownership, sponsorship, and financial arrangements than the atomic energy plant, I am wondering, in practice, if you can ever get to the point where you can say that part of this whole system will depend upon the efficiency of the operation of the community's water plant and, therefore, can realistically take this into account.

THOMAS: You are raising a very important and broad question. I will say that when we first got together with the economists and governmental scientists they told us that governmental processes are not nearly as inflexible as engineers assume them to be. They are in the long run adjusted to meet the needs of technology and developing science. They said that engineers should examine many alternative designs from the standpoint of economic efficiency. If there are constraints in finance, or if subsidies are operating this should be taken into account at all levels from field planners to the Bureau of the Budget in Washington. The examination of many alternatives will show the cost of non-optimal designs that may be promoted for various reasons other than economic efficiency. If the design is unbalanced nobody gets the benefit; if you balance it up you save money doing it more efficiently and with the saving invest in further resource development.

PARKER: That is what we are doing right now subsidizing water treatment and sewage treatment facilities.

THOMAS: That is right. There are a variety of forms of subsidy. The point is that we do not want to get these subsidies tangled up with one another. At all times we should know what the real costs and benefits are. I might say that engineers and planners in Russia have difficulties in this because they do not have a "market guide." When I was over there looking at their system I was at a loss to see how they arrived at real costs and benefits in their investment in social overhead capital.

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SUMMARY DISCUSSION

BELTER: In this session we will not summarize what has been said in each paper. I would like to ask first the reactions of those who gave papers to present their comments and what they believe we should still be doing in stream studies. What areas are we perhaps weak in? What areas should we be emphasizing further? We will now open the meeting to discussion from the six or seven people who spoke the first day and then anyone else who has any reactions or comments on their phase of work.

FRIEND: I will speak from our own experience, of course. It appears to me from the field trips that we have made to the Mohawk River, the Clinch River, the Savannah River, and so forth, we have a pretty good picture of the general distribution of the radionuclides in the environment, those that we have determined. It seems like to go from here that AEC should be sponsoring research in the laboratories, semi-natural, under controlled conditions with some of the mechanisms - the rates and reactions - and that some of these things might be determined in the, let's say, less-complex environment than the natural one. This will allow us to get some measure of the factors that do affect the distribution within the environments. I think that most of the papers here have indicated that such research is going on, but in talking with Mr. Belter I get the impression that the AEC is not interested too much in sponsoring pure research. Maybe I should not say this. They are more interested in determining the situation as it exists in the stream now.

BELTER: That is not exactly true, Dr. Friend. There is a lot of work going on within the program that can be classified as basic or fundamental research and which is not really directed at practical application. In the Division of Reactor Development it is integrated with the engineering development projects. It is not being sponsored, per se, by the Division of Research, but we have work that is going on that you are familiar with, i. e., the work at Oak Ridge, for instance. I make reference here to the work done by Drs. Jacobs and Tamura. Much of it is of a basic or fundamental nature and not directed at practical engineering application. We realize that we will have work of this type continuing, but this cannot be of major emphasis in the entire program.

FRIEND: I do not think you should stop your research at the laboratory level; if something in the field comes up real "hot," I think you should go back and check it.

BERNARD: To what degree does this particular facet have on the problems touched on in Mr. Cowser's analysis? That would influence us regarding the level of effort that our Division would put into this particular facet because of our engineering application approach.

BELTER: Dr. Nielsen, would you care to comment?

NIELSEN: I think that my interest in some of these phases was intensified - particularly the mechanisms whereby the radioisotopes are adsorbed onto the sediments and move on the sediments. Also, specifically, the biota and what role they play and the order of magnitude of that role. What is the effect of iron and other adsorptive surface material and the possibility that Mr. Kennedy has mentioned that they participate in absorption rather than the clay substrata or whatever is underneath the particle? I think that I would want to look at an actual stream for these to get an idea which ones were the most important ones for that particular stream. In other words, the kind of factors that Dr. Thomas can use rather than taking something straight out of the laboratory. Just how large a variation in nature do you see and this gets into the plus or minus on his figure. However, I think that in order to answer some of these questions we will have to cart some of the things back to the laboratory and make the studies there. For instance, if you assume that there are conditions where iron will be released and its radioactive adsorbed material with it and yet you cannot perhaps see that in nature because it does not occur all of the time, it might only occur at the peak flow of the year, or something, then we would have to try to duplicate that in the laboratory.

BERNARD: Could you duplicate that with a mathematical model in the laboratory?

NIELSEN: Well, if the iron can be released by a reducing environment I would have to cause a reducing environment and see if the iron was released and see if the radioisotopes on the particles associated with the iron were released also and in what form they are released. Are they still particulate or do they go into solution?

PARKER: We can help you a little bit on that because we have just been working on that particular problem.

NIELSEN: This is one of the particular values of this type of meeting so that we can find out what other people are doing.

BELTER: Naturally, this is one of the major benefits that we can derive from this type of discussion.

The main thing that we are interested in is how important are each of the factors. It would be nice if all the information was readily available at the present time. With this not being the case, should we be expanding our effort on sediments, or do we know enough about them to essentially close out

that phase of the program? I am sure this is not the case. In the case of dispersion, what is most important? Have we done enough dispersion experiments in field tests? If we knew the relative importance of the many factors and parameters involved it would simplify our program planning.

PARKER: I do not think it is a static figure. It depends upon the area in which you are working. For Oak Ridge I don't think we can say that sediment transport is important. It may be important in one area and may not be important in another. These are the things we should work to find out answers and this is where we must bring some work into the laboratory, make experiments, and then test it in the field.

BELTER: Do we have to do this at each individual site? In other words, where a nuclear power reactor would be constructed - do we have to do all of this at each site?

FRIEND: That is the point. That is why we must do basic research.

PARKER: There is also another point. We are talking about old-style reactors for the pollution loads. This would be from the gas-cooled and the water-cooled reactors with once thru coolant cycles. The newer reactors have recirculation of the coolant and do not have this problem at all. The new sites that you are talking about will be relatively few in number.

BERNARD: Actually then, you only really have the problems at the existing sites.

BELTER: The point that you are making, Dr. Parker, is that this is a problem at some of the sites we have now, either a large laboratory site or some of the others.

BERNARD: At these sites we know what the sediment problem is, if there is one. Therefore, maybe some of the other organizations should do basic studies at the other sites.

PARKER: That is not quite so, Mr. Bernard. The properties change at these sites, too.

BERNARD: Basically, just from a physical and chemical standpoint the sediments are going to move in the same way no matter what you put on them. Physically and chemically the system may change and this can be determined in a laboratory from general physical chemistry.

BELTER: We can see this is an area where there is not general agreement.

Mr. Kennedy do you have anything that you would like to comment on at this point?

KENNEDY: Well, I agree with Dr. Nielsen pretty well. I think we need to get out and work in the field and then, on the basis of what we see there, determine what laboratory work is appropriate and what parameters should be considered. The complexities are much greater in the field and, until you have some feeling for these complexities, I don't see how you can develop an intelligent laboratory experiment. In some of our research group's work, not tied with the AEC work particularly, the approach has been to take a problem that could be handled in the laboratory, get as many answers as possible there, and then go to the field and see if the answers are applicable. Further laboratory work can be based on the field results. Dr. Nielsen mentioned the problem of iron coatings on minerals under reducing environments. There is the additional fact that we have organic material present in sediments under these conditions and it is well known that ferrous iron can travel as an organic complex. This is a complication that you might not consider if you were to think of it just as a reducing environment. So, I really do not see how we are ever going to set up exactly the same conditions in the laboratory as we have in the field. It may get impossibly expensive to do it this way. There has to be a feed-back from the laboratory to the field, and vice versa, and I do not see how you can separate them.

BELTER: You are perhaps indicating that the prime emphasis should be on conduct of field investigations or studies, and then on the basis of what you find out in the field you go back to the laboratory.

KENNEDY: Yes, this is my feeling.

BELTER: This is also the feeling of Dr. Nielsen and Dr. Parker, I believe.

KENNEDY: There is one aspect of this research that we ignored which certainly needs investigation and that is the role of dead organic material. We find there has been a fair amount of study of the living matter, but I have not been able to find much information on the organic composition of the particulate organic matter. What are its attributes as far as being selective for individual elements? Probably part of the organic material is being broken down after burial with the mineral particles. This helps develop the reducing environment.

BELTER: How important is this aspect?

KENNEDY: At this stage, I do not know.

FREDERICK: Yesterday, I thought I understood it said that there was anywhere from 30 to 40% of the radioactivity tied up in both the dead and live organic material. The "live" people here have only accounted for considerably less than 10%, so there must be a lot of it in the dead organic material.

NELSON: There is not that much dead organic material. If this is counted on a unit weight basis it looks high, but if you figure out what the total organic material there is in the environment, I have looked at a large number of samples

from the Clinch River - in fact quite a number of them - and I only looked at the big particles that I could catch on a 1 mm square sieve, and this was about 1% by weight of the total sample. So, in bulk sometimes it looks high, but in weight it is not very much.

KENNEDY: Well, it is my impression that the organic material in ancient sediments averages about 1.5% and that in recent sediments this runs several percent.

NELSON: I will admit that I missed a lot of the fines that went through the sieve. It would take other techniques to pull this stuff out; there is probably as much or more in there.

KENNEDY: There is a possibility here, and only a possibility, that you have dissolved material attaching itself to some of the moving organic matter in such a way that it really is not free to exchange back and forth from the sediment to solution once it has reacted. This may be fairly important.

BERNARD: I think the extent of research done in any one area could possibly be a function of the degree of hazard that could develop in that particular area. If there were no radioactive wastes, then no research. If there is radioactive waste, how big a problem is it, and how big a problem could it become? This could be developed somewhat between Mr. Cowser's work, the end result or ultimate fate of the radioactive wastes, and what Dr. Thomas and Mr. Spofford are doing at Harvard to determine the effects of the many variables of the systems.

HUBBELL: Nothing has been said about the range of values.

BERNARD: Perhaps knowledge of the range is not required.

HUBBELL: To get the variance you do. This may be the very item we are seeking.

THOMAS: Well, this is not just right. Take the case of the Manning formula on a river. Using the Manning formula and field measurements you can make a statistical analysis and obtain not only the best value of "n," but also a standard deviation.

HUBBELL: This may be true, but do you have enough data in the first place to make the statistical analysis.

THOMAS: You only have to have two measurements to get a variance.

HUBBELL: But, your variance is only as good as the input.

THOMAS: That is certainly true but after you have said that, what have you said?

BERNARD: Maybe you don't need any better data.

KAUFMAN: You do not know how much water the canal or the stream needs to carry.

BELTER: You can always go back to engineering judgment.

KAUFMAN: You can always go back to 1880 for the first records on stream flow.

SERATA: Well, I feel that there is a need for finding out the relative importance of the several mechanisms. How much ion exchange is present; how much bio-absorption is in there? I think before we get into extreme detail on ion exchange I think we have to determine the relative significance of the several mechanisms. To do this there must be a very well defined field study probably relating to mathematical models in relation to laboratory models.

BELTER: There is, Dr. Serata, a lot of work going on in ion exchange, adsorption on sediments, a great deal of which is going on in the laboratory right now in addition to the field studies. What we are trying to decide in our own mind is how much of this should be done further, in the next year, two years, or three years.

FRIEND: The things that are important to me, and I think some will agree, is what you must place your emphasis on are those pathways that will lead radioactivity into man. I think if you get the radioactivity going into the bottom sediments, and it stays there, it is a wonderful place for it. It is well shielded.

BELTER: How important is this in the food chain?

SAYRE: Well, suppose it deposits on a flood plain that is used for grazing by cattle?

FRIEND: I said in the bottom sediments of a stream.

SAYRE: But bottom sediments will deposit on a flood plain during the recession of a flood.

FRIEND: No, but I said stay there. They are under about 30 feet of water and this may be a beautiful place for them; let's don't disturb them.

CLANTON: Along this line I think it would be wise to comment that many types of clays will selectively remove many of the radionuclides. However, in the clays with an expanding lattice, the radionuclides are transient and subject to a mass action effect. If the environment changes so that the equilibrium conditions no longer balance, some of the radionuclides will again be released into the environment.

This is distinctly different from the way cesium is bound to the illite structure. In the illite structure the interlayer cations are bound tightly and it takes some rather intensive weathering conditions to release sorbed interlayer cations.

FRIEND: Maybe you would want to take this out every year or so and calcite it. We must use our imagination.

BELTER: You are!

FRIEND: These are the things we are interested in; I am not interested whether clay takes up cesium or not. If it takes it up and keeps it on the bottom then I am very happy.

BELTER: The question is - is there any release and if there is release is it significant from the overall standpoint of getting the radioactivity into the food chain.

FRIEND: I think that using statistics on the human population is wrong too. The population is made up of individuals, not the one in a thousand.

FOSTER: In that regard one of the things that we have glossed over quite lightly here is the basis of exposure estimates. For non-occupational people, averaging over a period of a year, is specified.

COWSER: Let me make another pitch. Dr. Parker alluded to it and that is the transmutation products that are likely to become quite significant in the overall atomic energy program. This is pointed out by the AEC's response to the President's request on where do we go from here? Emphasis is placed on breeder reactors and as a result of that one gets involved in fuel recycling and as a further result some of the heavy metals become important; i. e., uranium, plutonium, and curium. But, thus far we find ourselves confined to talking about cesium, strontium and phosphorus. Apparently there are several groups that are looking at plutonium movement through the environment but there is no little information to work with.

BELTER: Dr. Kaufman, what is your general reaction?

KAUFMAN: Every point of view is represented here. Sediment uptake is probably relatively less important here than the biota problem, the biota problem being nearer to man as it has been pointed out. When we compare the masses of sediment to the masses of organic material it doesn't lead to any conclusions of importance. I think more attention must be given to the approach that Mr. Cowser gave us the first day. We must first identify the radiological problem as it impinges on man and then follow the problem back to its source. Along this path many unanswerable questions will arise and they should serve as the basis of our research. We can all justify our own basic research on an academic basis but within an engineering group such as Mr. Belter represents one has to

do more than that. The basic research can only be justified in terms of the engineering or radiological problem that it helps to solve.

FREDERICK: Who is going to make the decision as what is important and what isn't?

KAUFMAN: I think that the engineer has not identified the problem in a sufficiently detailed manner to allow adequately oriented basic studies to be defined. The work of Cowser and Thomas will be of great help in identifying the health problems and in systemitizing their analysis. Perhaps we do not understand the science well enough to appreciate the limitations of what data are needed. But I think we are making some progress in this line as evidenced particularly by the recent discussion.

BELTER: What is your reaction to this Mr. Reynolds?

REYNOLDS: Well, the studies in the streams and model studies are both useful. Although a model will never be able to duplicate exact stream conditions, it will help in uniting the past work that has been done on dispersion, sediment uptake and biological uptake. It will help in deciding which factors are important in the ultimate disposition of a particular radionuclide in a stream. For instance, cesium is taken up by most fine grained minerals, therefore much of the cesium will become associated with the bed material. If however, lower forms of life feeding on the bed materials take up some of the cesium, the cesium will enter into the food chain. Model studies would be useful in studying such phenomena under controlled conditions.

BELTER: This points out the importance of the ecological transport problem. Maybe we should get into this area now. Dr. Nelson, what has been your reaction to the discussion for the last two and one-half days?

NELSON: Well it is always interesting to hear the engineering approach to this problem and we sometimes get the feeling that engineers work with great big things and the biologists are always grasping for straws and this is probably the reason behind some of the variations. Maybe you could take biological variations and ignore the whole thing such as Dr. Thomas was talking about. Dr. Kaufman pointed out this does get awfully close to man and this is the reason we are doing all of this in the first place.

As far as the work we are doing, right now my whole research philosophy is based on the fact that if we know the distribution of the stable element in the environment we ought to be able to predict where the radioactive species of this element is going to go. One of your interests is whether you can transfer studies from one area to another and I think this could probably be solved fairly easily with a few simple comparative field studies and also laboratory studies. We should know what the response is to a concentration of elements in the environment as far as uptake on organisms is concerned. We ought to know the effect of other ions, the possible effect of competing ions or similar ions,

on uptake like calcium and strontium.

BELTER: Do you think you could do this right now? Is there a possibility we could transfer some of the results?

NELSON: Sure, I think you could make a pretty good estimate and not miss it over a factor of 10. If we would run a few experiments maybe we could come closer.

BELTER: This could be done without having a ecological program at each site?

NELSON: Well, if you wanted to put in a reactor at some place that is going to release strontium⁹⁰ or a processing plant and a little effluent could get out in the river, I think you could simply look at the distribution of stable strontium in the flesh and in the water and if you could predict your releases it would be a fairly simple matter to predict how much strontium⁹⁰ uptake would take place. This is simple and straightforward with a little knowledge of the seasonal variation of the strontium content in the river and perhaps the same on fish flesh. However, we do not have this information now but we will have it shortly on one species of fish. This is a time-consuming problem at the present time.

I think another thing we ought to look for are the indicator species. We can use clams for this purpose and perhaps some of the insects. The sanitary engineers, if I may introduce the tubificid worms, have long used the tubificid worms as indicators of organic pollution. They have an abundant number of indicators that they could use to indicate organic pollution of one kind. I think one thing we ought to remember with radioactive pollution when comparing it with organic pollution is that there is no self-purification of the stream. The only self-purification you get is the physical half-life of the elements you release. So, organisms can adapt to organic matter - enriched organic matter in the streams - but they cannot adapt to radioactivity as such, they see this as the element and take it up as such. The heart of Dr. Odum's talk was community metabolism. You would like to know, and get a "quick and dirty answer" of what is going on in a community and right now we have no studies to test whether we could use phosphorus or zinc or some other element as a tracer for carbon. I think very definitely there should be some studies along these lines. This might allow us to cut out a lot of details of biological sampling. I think the principles involved probably apply to marine environments as well as fresh water environment.

BELTER: As far as biological sampling is concerned, if we took a reasonable number of fish samples and analyzed these, would this be sufficient?

NELSON: Dr. Foster should answer that.

FOSTER: Dr. Nelson has hit the thing on the head - a way in which we can come up with some predictions on the level of contamination which is apt

to exist in fish. Most of the estimates which we make at the present time tend to hit towards a maxima value. I think our experience generally has shown that if you actually get out and measure the concentrations you will find that some hit the high points but generally the range is below this and subject to terrific variability below the maximum. We need to understand better the ecological factors mainly responsible for causing the great variations and these fluctuations. Again, this is about the same kind of thing that Dr. Friend is talking about in terms of the bottom sediments. You can only push the field work so far and then, I think, you make the greatest gains if you follow up with laboratory studies. In this case, what are the environmental factors that change these rates? We have seen, at least for some of the shorter-lived materials in fish, that the concentrations may vary at different times of the year from essentially zero up to many hundred thousand times. I think it is important to know the environmental features which influence these great changes so that we might possibly be able to take advantage of some of these things which influence these great rates. Again, I would encourage the kind of work which gives us more information about changes in rates for the key radionuclides.

We need to talk in terms of two kinds of problems. One is the chronic situation with continuous releases. The other case is the short term "slug" situation and the importance of a short-term "slug" as it goes through the system.

Dr. Nelson also mentioned indicator species. I think I hold a little different view on this. We used to use indicator species. But when we got down to the problem of evaluation, we did not make radiological evaluations on the indicator species. You make evaluations on the fish which somebody is using for food or on some food which is being eaten by man. If you can't measure the material in the fish which the people are eating, then there is not much of a problem. If you can measure it, you might just as well do it with the fish, rather than trying to extrapolate from the indicator species.

I will say one other thing. We do have for man the MPC values and the radiation protection guides. We do not have these things for aquatic populations and we will always be faced with the people who have the opinion, that certain levels of contamination may affect a community of organisms, or individual species, at lower levels than those deemed safe for man. I think we need more information on levels which are permissible, let's say, from the fish's point of view.

BELTER: I would like to get a reaction on the dispersion work and what we should be doing further along this line. What are the general feelings or reactions about this phase of the transport problem?

PATTERSON: We have not gotten together to develop a general reaction. Personally, I feel we have been "shot out of the saddle." If we take Dr. Kaufman's route and start with man and his dosages and go back to the routes through which the nuclides came eventually we get back to the reactor and assuming we find

someone who wants to evaluate the dispersion then I would say we use this approach. We need to continue the uniform flow flume studies that have been worked on by various groups. We need to extend that to bends and transitions always with the view in mind of developing predictor type equations for transferring this kind of data to prototypes. Also, I think we need a survey of the field data that has been taken to date, put in one report possibly and distributed to the groups interested in it.

BELTER: Certainly Dr. Parker has done a great deal of work in this area.

PARKER: I think it depends upon where you are. It should be done on the basis of hazard and accident situations in most cases.

BELTER: How important do you think the continuance of flume studies are at this point?

PARKER: Since we did some ourselves a very short time ago this is a "loaded question." I obviously believe that there is use for flume studies in the right place at the right time.

NELSON: In connection with the dispersion studies I frankly am very glad, from the standpoint of the biological studies that we are doing, the dispersion information is available and if we want to get a sedentary organism from the river bottom we can go downstream only so far and we are reasonably assured that this "beast" has been exposed to a well mixed effluent. This is important as far as improving the quality of our sampling is concerned.

KAUFMAN: It seems to me that when we have completed all of our flume studies we will still have the impossible task of applying the data to the prototype. On the other hand, consider how relatively simple it is to go out and measure the prototype.

GLOYNA: Actually, field studies are not always simple in cost and time. When you try to cover all of the factors that you need to look at, field studies become very involved.

KAUFMAN: My statement was somewhat of an over-simplification. However, if you want to know the dispersion of the stream what is the simplest way of determining it? That is what we are talking about, not all the other factors. Simply go out and measure it.

GLOYNA: But you do not know which stream you are talking about and wouldn't it be nice to have some general information available.

KAUFMAN: The information required to obtain a dispersion constant from a knowledge of stream velocities and dimensions is very considerable. However, the time required to measure such a constant is not great. We do need some

descriptive generalizations relating stream type to dispersion, but not in the detail that would allow us to synthesize the dispersion curve. This would be unwarranted.

FREDERICK: I am inclined to agree with Dr. Kaufman. I would go further and say that even if we did a lot of work in the laboratory and were fortunate enough to get material which had a transfer value to the field, if there was a problem at a certain site and someone predicted the dispersion on the basis of the lab studies, someone else would go out and make a field study to see whether or not the prediction was any good.

BELTER: From what I have seen, that is certainly a true statement.

SAYRE: I have two groups of comments to make; one that is specifically directed toward the dispersion problem, and the other more general. With respect to dispersion, I would agree that dispersion from an instantaneous source is not of great practical importance in itself. The importance of the dispersion investigations lies more in the solutions to practical problems which can be generated from the solution for the idealized instantaneous source case. For example, by integration of the dispersion equations, one can estimate the rate at which concentrations of radionuclides or contaminated particles will build up along certain reaches of a stream, or the rate at which contaminated bed-material particles will move through a given section. One can predict the length of time required to reach equilibrium, and also the rate of response to a changed input, for example. I would attach more importance to these questions than to dispersion itself. However, we can't make predictions such as these until we have first solved the dispersion problem. In the case of a sudden accidental release of radionuclides, however, knowledge of dispersion from an instantaneous source would become, of course, extremely important in its own right.

As for more general comments, and I think this applies to all of us, we need to improve communication between those of us who are actively engaged in researching the physical, chemical and biological aspects of the problem and the people like Dr. Thomas who are setting up the mathematical models. Such communication will help us to provide the kind of information needed for the input into the model, and will also enable the model to be used for directing research to the areas where it is most needed.

BELTER: These are both very good points Mr. Sayre.

Drs. Foster and Nielsen must leave now to return to Hanford. I would like to express our appreciation to them for coming from the west coast. The Hanford contributions are major ones in all areas.

How does the group, as a whole, feel about the significance and importance of Dr. Thomas' mathematical model?

FRIEND: My personal thoughts are that our efforts should be to collect

data that could be used in the setting up of the model whether it be the Clinch River, Mohawk, or any other stream. I do not particularly agree on the criteria; economics in the case of health has no significance. I think it should be based on a health precept rather than economics. I can visualize the point that if strontium⁹⁰ turns out to be the most significant radionuclide in fish in the Clinch River then you may have to do complete treatment. This is the point I am making, not that I am arguing with Dr. Thomas.

BELTER: I would like to express our appreciation to Dr. Gloyna for the wonderful facilities he has provided us with at the University of Texas, the extra-curricula activities, and the general friendliness and fellowship that we have had. The participation has been very informal, like we wanted, the weather has been extremely cooperative and we would like to thank you and your fellow workers. Each of you deserves a sincere vote of thanks from all of us. We appreciate coming to the University of Texas and we would look forward to coming back here and holding further meetings. In the brief time remaining would you care to summarize your work?

GLOYNA: Very briefly, at the University of Texas, we have been working in the field and in the laboratory on several problems. You have heard three of the speakers describe some of our work; others have been working in the laboratory on specific problems involving radioactivity and radioisotope uptake in a waste treatment system. Some of this work has been done in streams, some in lakes, all involving bottom sediments and organic material that is tied up in the bottom sediments. It occurred to us several months ago, particularly the group that is working in the area now, and specifically the work that Patterson is doing, to utilize flume facilities to check our field studies. We are now in the process of constructing such a flume.

At the Balcones Research Center we have several large tanks, old clarifier tanks, which can be used to develop an ecological system typical of our streams. Knowing the type of material we have and knowing the type of bed load material that can be expected we feel that we can model our streams. Perhaps we can even use some of the experience that Dr. Odum has gained in his studies at the Marine Institute. He has been working with small ponds.

We are building a flume that will be 200 ft. long, 3 ft. wide and roughly 2 ft. deep. The existing tanks are about 8 ft. deep and 70 ft. in diameter. The flume can be elevated if it is desirable to change slopes.

We plan steady-state or unsteady-state releases. We will be able to measure the bio-mass and this will help us check some of the field data under more controlled conditions.

BERNARD: Could Dr. Odum utilize this system to check out his analog approach?

ODUM: We could probably make metabolic measurements in there to go

with it; so, perhaps this might be the system we are looking for.

SAYRE: Is the flow recirculated back from the tanks?

GLOYNA: Yes, the contents can be recirculated since we will have the pump capacity. It has been designed so the effluent can be discharged either into the tank supplying the influent or a separate tank.

BELTER: I would like to take this occasion to express my appreciation to all of you for taking time out from busy schedules and coming to the University of Texas for this meeting. I think we have had a very good exchange of viewpoints and information. On behalf of Mr. Bernard and myself, and Dr. Hooper of the Division of Biology and Medicine, we appreciate your participation and look forward to seeing you again soon.

If there is no further business the meeting is adjourned.

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