Uptake and Transport of Radionuclides by Stream Sediments

GEOLOGICAL SURVEY PROFESSIONAL PAPER 433-A

Prepared in cooperation with the
U.S. Atomic Energy Commission

U.S. DEPARTMENT OF THE INTERIOR
March 3, 1849

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By W. W. SAYRE, H. P. GUY, *and* A. R. CHAMBERLAIN

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By W. W. Sayre, H. P. Guy, and A. R. Chamberlain

ABSTRACT

The factors involved and the present status of research concerning disposal of radioactive wastes in natural streams are reviewed with respect to (1) waste characteristics, (2) dispersion theory, (3) sorption by sediments, and (4) sediment transport. Some criteria for identifying potentially hazardous streams and areas where knowledge is deficient are discussed.

Evidence indicates that sorption of radiocontaminants by stream sediments is the rule rather than the exception. The extent of sorption is dependent upon a large number of variables: the nature of the sediment and the substance being sorbed; the available surface area; the concentration and types of solute in solution; time; and, to a lesser extent, temperature.

The transport of sorbed radioactive components by fluvial sediments depends on factors such as the types and amounts of sediment supplied by the drainage area, channel and flow characteristics, the time distribution of streamflow, and channel controls.

Pool and riffle-type streams, characterized by wide variations in discharge and containing large amounts of fine-grained sediments, appear to be potentially the most dangerous type of stream with respect to the development of hazards arising from radiocontaminated sediments.

Present knowledge is not adequate for the purpose of predicting the ultimate distribution of radioactive wastes in streams, particularly as affected by fluvial sediments. Considerable research and study will by required in all the subject areas of this report before this situation can be improved materially.

Certain deficiencies in knowledge and theory stand out—for example, there is a dearth of information in the literature on the quantities and physicochemical properties of low-level liquid wastes.

Current theoretical, laboratory, and field knowledge of the mechanics of turbulent diffusion involving instances of a confined channel, net uptake by sediment, and net uptake by biota is entirely inadequate.

The effect of some of the variables associated with sorption phenomena is not well understood. This is particularly so in complex sorption systems which include competing solutes, mixed sediments, and chemical additives such as detergents and electrolytes.

Despite notable advances, some of the factors involved in sediment-transport phenomena remain unresolved, and much of the associated theory is still in a stage of partial development.

INTRODUCTION

Radioactive wastes present a unique disposal problem (Warde and McVay, 1955) because unlike other industrial wastes they cannot be disposed of by burning, evaporation, or filtering, nor can they be rendered biologically harmless by chemical treatment. Radioactivity is not detectable by any of man's five senses. In general, radioactive pollutants on a weight basis are many thousand times (Thomas, Terrill and Gilmore, 1959)—and in some cases more than a million times (Jensen, Eldridge, Everts and Clare, 1957)—as dangerous to the health of man as are nonradioactive pollutants.

Since the early days of nuclear energy development, the U.S. Geological Survey has cooperated with the Atomic Energy Commission on certain technical problems relating to management of radioactive wastes. Considerations arising from the geology and hydrology of waste-disposal sites have been an important area of cooperation. This study was made cooperatively by the Geological Survey and the Atomic Energy Commission with staff assistance from Colorado State University. The purpose of the study was to determine the factors involved and the present status of research concerning disposal of radioactive wastes in natural streams.

DISPOSAL OF RADIOACTIVE WASTES IN STREAMS

Approximately 95 percent of the short-lived low level liquid radioactive wastes are discharged into surface streams (Joseph, 1955). The radioactivity level of these wastes is low, but the total volume is large. At the Hanford installation, for example, an average of more than 20 million gpd of radioactive wastes are discharged. Since 1944, more than 2 million curies of radioactive materials have been released into the environment at Hanford (U.S. Atomic Energy Commission, 1957).

Radioactivity surveys on the Columbia (Robeck, Henderson and Palange, 1954) and Tennessee (Garner and Kochtitzky, 1956) River systems have indicated that contamination of these waterways has not yet approached the danger level. There is real cause for concern in the future, however, when it is considered that the nation is just on the threshold of the nuclear age. Present estimates (U.S. Atomic Energy Commission, 1957) predict that the nuclear power plant capacity in the United States will increase by a factor of about fivefold every 5 years in the decades preceding 1980. Considered economically, disposal of wastes by discharge into surface streams is the most attractive method. It is only natural that the growing nuclear industry will use streams as a disposal medium to the maximum extent permissible.

Considering the ever-intensifying domestic, industrial and agricultural use of the nation's water resources together with the expanding nuclear industry, and the probable use of streams for waste disposal, it becomes apparent that the chances for harmful exposure to radiation resulting from accident or error are rapidly multiplied.

RADIOACTIVITY BUDGET IN STREAMS

In order that the chance for error or miscalculation be minimized, it is essential that the working of the dispersal mechanisms of streams with respect to radioactive wastes be thoroughly understood. In a simplified qualitative form, the problem for a particular radionuclide may be illustrated in terms of a radioactivity budget.

$$R+r_n=[r_w+r_s+r_o]e^{\lambda t}+r_d \tag{1-1}$$

in which

R is the radioactivity in curies of waste discharged into the stream at time $t=0$,

r_n is the background radioactivity in the stream due to natural causes and fallout at time $t=0$,

r_w is the radioactivity carried in solution by the water at time t,

r_s is the radioactivity sorbed by stream sediments at time t,

r_o is the radioactivity taken up by living organisms and entering the ecological cycle, at time t,

r_d is the radioactivity discharged from the watershed—that is,

$$\int_0^t r(t)e^{\lambda t}dt,$$

where $r(t)$ is the radioactivity discharged from the watershed as a function of time, and

$e^{\lambda t}$ is a radioactive decay factor dependent upon time t and the radioactive-decay constant λ which depends on the nuclide.

The radioactivity budget illustrates the complexity of the problem by suggesting the variety of physical, chemical, and biological laws which are operating simultaneously on the system. The problem is further complicated by the fact that the radioactivity in the stream is transported at different rates which vary according to both the nature of the transporting media, and the stream conditions. In addition, exchange of radioactivity occurs among the various transporting media. The extent of exchange depends on complex chemical relationships involving the characteristics of the radioactive waste effluent, quality of the stream water, particle size, concentration and mineralogic content of the stream sediments, and the nature of the aquatic biota.

At present, the quantities of radioactivity released into stream waters are governed by the maximum permissible concentrations (MPC), usually with a safety factor of about 10, of radioisotopes in drinking water recommended by the National Committee on Radiation Protection (U.S. National Bureau of Standards 1953, 1955, 1959). The MPC value for each radionuclide is based on its particular radiological and chemical properties with respect to their biological hazard potential to man. The MPC values do not take into account the various mechanisms in the stream system which are at work taking up and concentrating radioactivity. In some cases the concentration of radioactivity in organisms and on sediment particles exceeds the concentration of radioactivity in the water by several orders of magnitude (Robeck, Henderson and Palange, 1954).

Present data and knowledge are entirely inadequate for evaluating the radioactivity budget in any given stream system. The deficiency is even greater with respect to use of the budget concept as a means of predicting the behavior of radioactive wastes in streams. Knowledge is particularly lacking with respect to the uptake processes and the initial dispersion of radioactivity in streams by turbulent diffusion.

CHARACTERISTICS OF RADIOACTIVE WASTES WHICH ARE DISCHARGED INTO STREAMS

Perhaps the outstanding general characteristic of low-level liquid wastes is the extremely wide variation in their chemical and physical properties (Blomeke, 1958). It is safe to say that as the size and complexity of the nuclear industries increase, the wastes will increase both in quantity and diversity.

Low-level wastes are arbitrarily defined as having a radiation intensity at the emitting surface of less than 50 milliroentgens per hour as opposed to high-level wastes which have an intensity of more than 2 roentgens per hour (Joseph, 1955). The activity of low-level wastes may be as much as 10^4 times the long-term exposure limit considered permissible for humans (Gorman, 1955).

Although the specific activity of the low-level wastes is relatively low in contrast to high-level wastes, considerable total amounts of radioactivity are involved owing to the large volumes of waste effluents. At the Hanford plant, which is the largest producer of low-level wastes, approximately 640 million gallons of waste effluent containing about 50,000 curies of radioactive materials is discharged into the environment each month (U.S. Atomic Energy Commission, 1957). On this basis the average specific activity of this waste is on the order of 0.02 microcuries per ml (μc/ml).

SOURCES OF WASTES

Low-level, liquid wastes of the types which are released into streams originate from many kinds of sources. By far the greatest source of radioactive wastes are the chemical-processing plants where irradiated reactor fuels are processed to reclaim the unburned nuclear fuel (Blomeke, 1958). The low-level wastes resulting from these operations arise from second- and third-cycle operations, fuel dejacketing, chemical and analytical laboratories, evaporator condensates, and cell- and equipment-decontamination wastes. Another large source is the water used in reactor cooling systems. Other sources result from the medical and industrial use of radioisotopes, various laboratory, laundry and decontamination operations, and the processing of natural radioactive ores.

Some of the low-level wastes are derived from intermediate and high-level wastes which have undergone chemical treatment for removal of some of the more dangerous radionuclides. Others have been retained in storage for varying periods of time to permit reduction of activity by natural radioactive decay.

With respect to the more hazardous radionuclides, it is probable that in the future more problems will arise from the combined operations of many small enterprises, than from the larger enterprises which are able to afford elaborate treatment facilities. Accidental releases of the hazardous radionuclides are always a possibility.

NATURE OF WASTES

It is impossible to formulate an adequate description of the nature of low-level liquid wastes in a few general statements because of the wide variety in the composition of the different wastes. As with other industrial wastes, the characteristics of atomic wastes may vary not only from one installation to another but also to some extent with respect to time at any given site. The nature of radioactive wastes may be classified, however, with respect to criteria such as the properties and types of radionuclides, chemical characteristics, and significant physical characteristics.

Obviously the factor which sets atomic wastes apart from other wastes is radioactivity. A stream has no self-purification capacity for radioactive wastes in the sense that it does for ordinary organic pollutants (Tsivoglou, Harward and Ingram, 1957). Organic pollutants undergo biochemical oxidation resulting from bacterial attack in a stream, by which process the stream rids itself of the pollutant in a short time. No comparable process occurs for radioactive pollutants.

There is no known chemical treatment by which radioactive wastes can be rendered biologically harmless. Even though the physical and chemical form of the waste may undergo change the radioactivity remains unaffected and is reduced only by natural radioactive decay which occurs at a definite rate, different for each radionuclide.

The apparent concentration of radionuclides in the water may be reduced owing to uptake by stream sediments and biota. However, this does not represent a reduction in the total amount of radioactivity in the stream environment, but rather a transfer from one phase of the stream environment to another.

PROPERTIES AND TYPES OF RADIONUCLIDES

The significant properties of radionuclides with respect to waste-disposal problems are (1) nuclear, (2) biochemical, and (3) chemical.

The important nuclear properties are the rate of radioactive decay, the type of radiation, and the energy of radiation. Wastes containing radionuclides characterized by a slow rate of decay—a long half life—if continuously discharged into the environment in significant amounts would build up the concentration of radioactivity in the environment over a long period of time before reaching a condition of equilibrium with respect to activity level. The concentration of radioactivity of radionuclides having a rapid decay rate, however, would reach a level of steady equilibrium rather quickly. Curves illustrating this phenomenon are shown by Rodger (1954). Furthermore, radioactivity emanating from long-lived nuclides would contaminate the environment for a long period if a large quantity were released accidentally, whereas, the radioactivity would decay rapidly where short-lived nuclides were involved. After a time interval of 10 half lives, radioactivity decays to about 0.1 percent of the activity level at the beginning of the time interval. The half life of radionuclides ranges from fractions of a second to more than a million years.

The relative hazards resulting from ingestion of radionuclides aside from the quantity and energy level of radioactivity depend primarily on biochemical considerations. Some of these biochemical considerations are initial body retention, fraction going from blood to critical body tissue, radiosensitivity of tissue, size and essentiality of critical organ, biological half life, and specific ionization and attenuation of energy in body tissue (U.S. National Bureau of Standards, 1953).

From a biologist's standpoint, other factors being equal, radionuclides with an intermediate decay rate, say a half life of 5 to 50 years, are the most hazardous when ingested on a weight basis (U.S. National Bureau of Standards, 1953). Long lived nuclides must be present in relatively larger quantities in order to give off as much radiation as shorter lived radionuclides during a given time interval, say a person's normal remaining life span. At the other extreme, radionuclides with very short half lives normally are less hazardous unless exposure is maintained by continued uptake, since the activity of such radionuclides when deposited in the body soon decays to an insignificant level.

When taken internally, alpha emitters, owing to their high, specific ionization are considered to be about 20 times as damaging as either beta or gamma emitters (U.S. National Bureau of Standards, 1953). However, gamma rays, owing to their great penetrating power, are considered to be the most hazardous for external radiation of the body, excepting the artificially induced neutron emitters.

The maximum permissible concentrations for various radionuclides as recommended by the U.S. National Bureau of Standards (1953, 1955, 1959) have been computed on the basis of relative hazard from nuclear and biochemical considerations. It is significant to note that as more information on the adverse biological effects of radiation has become available, the tendency has been to revise downward, in many cases rather sharply, the MPC's for most radionuclides. The MPC for many of the radionuclides found in radioactive waste solutions are listed in table 1. It is recommended (U.S. National Bureau of Standards, 1959) that the MPC of unidentified radionuclides in water not exceed 10^{-7} *μc/ml.*

The chemical properties of radioactive isotopes are essentially identical to those of the corresponding nonradioactive isotopes having the same atomic number. With respect to waste disposal in streams, the chemical properties are important in so far as they influence sorption by sediments, and uptake by aquatic biota. For example, studies on the Columbia River (Robeck, Henderson and Palange, 1954) revealed concentrations of P^{32} in the river plankton which were as much as 10,000 times the concentration in the water.

Fortunately most of the radionuclides which have been identified in the low-level liquid wastes are of the less hazardous variety. In table 1 are listed the nuclides and some of their properties that have been identified in the Columbia River (Robeck and others, 1954), (Conley, 1954), (Foster and Rostenbach, 1954), and in the cooling water of the Low Intensity Test Reactor (LITR) at the Oak Ridge National Laboratory (Moeller, 1957). These should be fairly typical of the radionuclides found in reactor-cooling water.

CHEMICAL AND PHYSICAL CHARACTERISTICS

The importance of the chemical and physical characteristics of waste effluents is in their effect on the reactions occurring between the radioactive materials and the stream environment. One example is how the chemical constituents of the waste effluent might affect the uptake of radioactivity by stream sediments and biota. Physical characteristics of the waste effluent such as specific gravity, viscosity, surface tension and temperature are important in so far as they affect dispersion by turbulent diffusion of the wastes throughout the flow cross section. These are areas which apparently have received very little systematic study; however, some of the known characteristics of low-level liquid wastes can be listed, and possible areas of importance suggested.

In general, reactor-cooling waters contain significant quantities of corrosion products; used solvents contain kerosenes, tributyl phosphates, I_2, and Ru; and solvent washes contain NaOH and Na_2CO_3 (Joseph, 1958). Condensates usually consist of very nearly pure water. The pH range of low-level liquid wastes is usually between 4 and 9.

At Hanford the reactor-cooling water discharged into the river consists of treated Columbia River water with an approximate dissolved-solids content of 100 ppm before treatment (Barker, 1959, personal communication).

At Knolls Atomic Power Laboratory, Schenectady, N.Y., radioactive laboratory wastes and "hot" laundry wastes are combined and discharged into the Mohawk River. The properties of these wastes vary considerably, and may be acidic at one time and basic at another. It has been observed at KAPL, however, that differences in temperature between the waste and the river water control the mixing far more than do the differences in composition (Barker, 1959, personal communication).

At the Dresden Nuclear Power Station near Chicago, it is proposed that several types of low-level wastes be discharged into the river. These include high-purity water (1 ppm), medium-purity water (100 ppm), laboratory-drain wastes and other corrosive waters, and laundry wastes, all of which will be mixed together in varying proportions before discharge (Falk, 1958).

Laundry and laboratory wastes are apt to contain significant quantities of detergents and water-softening agents which may be important with regard to uptake characteristics.

Where radionuclides in the waste effluent are accompanied by isotopic carriers, their behavior in the stream environment may vary considerably from that of the corresponding carrier-free nuclides.

Such information on the chemical and physical characteristics of low-level liquid wastes as is available should be systematically assembled and analyzed. Supplemental information as required to fill in the

TABLE 1.—*Radionuclides in reactor-cooling water*

Half life: s, seconds; m, minutes; h, hours; d, days; y, years
Mode of decay: *K*, *K*-electron capture; IT, isomeric transition
MPC: Maximum permissible concentration, based on hazard resulting from exposure continued over a long period of time. (From U.S. National Bureau of Standards, 1959)
Mode of formation: FP, fission product

[Data from Kinsman, 1957; Moeller, 1957; Oak Ridge Nat. Lab., 1960]

Location	Radionuclide	Half life	Mode of decay	MPC in water (μc/ml)	Primary mode of formation
Columbia River	As^{76}	26.8h	β^-, γ	2×10^{-4}	Induced
	Ba^{140}	12.8d	β^-, γ	2×10^{-4}	FP
	Co^{60}	5.3y	β^-, γ	5×10^{-4}	Induced
	Cr^{51}	27.8d	K, γ	2×10^{-2}	Induced
	Cu^{64}	12.8h	$K, \beta^-, \beta^+, \gamma$	2×10^{-3}	Induced
	Mn^{56}	2.6h	β^-, γ	10^{-3}	Induced
	Na^{24}	15.0h	β^-, γ	3×10^{-4}	Induced
	P^{32}	14.3d	β^-	2×10^{-4}	Induced
	Si^{31}	2.6h	β^-, γ	2×10^{-3}	Induced
	Sr^{89}	53.0d	β^-	10^{-4}	FP
	Sr^{90}	28.0y	β^-	10^{-6}	FP
Oak Ridge LITR	Ba^{140}	12.8d	β^-, γ	2×10^{-4}	FP
	Br^{80}	18.0m	$K, \beta^-, \beta^+, \gamma$	---	Induced
	Br^{80m}	4.6h	IT, γ	---	Induced
	Br^{82}	35.9h	β^-, γ	4×10^{-4}	Induced
	Ce^{141}	32.5d	β^-, γ	9×10^{-4}	FP
	Ce^{143}	33.0h	β^-, γ	4×10^{-4}	FP
	Co^{60}	5.3y	β^-, γ	5×10^{-4}	Induced
	Cr^{51}	27.8d	$K, \gamma,$	2×10^{-2}	Induced
	Cu^{64}	12.8h	$K, \beta^-, \beta^+, \gamma$	2×10^{-3}	Induced
	F^{18}	1.9h	β^+	5×10^{-3}	Induced
	Fe^{59}	45.1d	β^-, γ	6×10^{-4}	Induced
	I^{131}	8.1d	β^-, γ	2×10^{-5}	FP
	I^{132}	2.3h	β^-, γ	6×10^{-4}	FP
	I^{133}	21.0h	β^-, γ	7×10^{-5}	FP
	I^{135}	6.7h	β^-, γ	2×10^{-4}	FP
	La^{140}	40.0h	β^-, γ	2×10^{-4}	FP
	Mn^{56}	2.6h	β^-, γ	10^{-3}	Induced
	Mo^{99}	2.8d	β^-, γ	4×10^{-4}	Induced
	Na^{24}	15.0h	β^-, γ	3×10^{-4}	Induced
	Np^{239}	2.3d	β^-, γ	10^{-3}	Induced
	Pa^{233}	27.4d	β^-, γ	10^{-3}	Induced
	Sb^{122}	2.8d	$\beta^-, K, \beta^+, \gamma$	3×10^{-4}	Induced
	Sr^{91}	9.7h	β^-, γ	5×10^{-4}	FP
	Sr^{92}	2.7h	β^-, γ	6×10^{-4}	FP
	Tc^{99m}	6.0h	IT, γ	6×10^{-2}	Induced
	Te^{132}	3.2d	β^-, γ	2×10^{-4}	FP
	Xe^{131m1}	4.8×10^{-10}s	IT	---	FP
	$\mathrm{Xe}^{133}+\mathrm{Cs}^{133m}$	5.3d	β^-, γ	---	FP
	$\mathrm{Xe}^{135}+\mathrm{Cs}^{135m}$	9.2h	β^-, γ	---	FP
	Y^{91m}	50.0m	IT	3×10^{-2}	FP
	Zn^{65}	245.0d	K, β^+, γ	10^{-3}	Induced

gaps should be added, and the combined results published.

TURBULENT-DIFFUSION THEORY FOR OPEN CHANNELS

Waste, contaminants, or tagged materials when introduced into an ambient flow field—a natural stream, for example—may be (1) fixed in place, (2) dispersed by molecular diffusion, (3) dispersed by diffusion of a submerged jet, (4) dispersed by the process of turbulent diffusion, or (5) subjected to two or more of these processes simultaneously—for example, the simultaneous occurrence of molecular and turbulent diffusion is common. In nearly every field situation, however, turbulence is by far the most important mechanism causing dispersion in streams.

Considering the whole problem of waste dispersal, however, dispersion of waste solutions is not the only effect produced by turbulent exchange. In sediment-carrying streams, for example, sediment particles are distributed by turbulence. This is a very important consideration when the nature of the contaminant is such that it can be sorbed and subsequently released from stream sediments.

In view of the roles of turbulent diffusion with respect to both waste dispersion and the distribution of contaminated sediments, turbulent diffusion assumes great importance in studies concerned with use of natural streams as a disposal medium for radioactive materials.

Application of turbulent-diffusion theory to problems of waste disposal in alluvial streams, canals, and rivers appears to be slight. While hundreds of papers on the turbulent diffusion of material into a fluid of homogeneous properties are in the literature, the number referring to the roles of diffusion and fluvial sediments in waste disposal is small. It is gratifying to note, however, that considerable work is being initiated in this field of study.

In this section the field conditions which must be represented by the boundary conditions in an analytical treatment will be discussed and classified. Next, a summary of diffusion theory will be given, in which specific reference will be made whenever the work reviewed was in regard to open-channel or closed-conduit flow-diffusion problems. The effect of fluvial sediments on the concentration of radionuclides in the stream will also be introduced. And lastly, some experiments reported in the literature will be summarized.

Molecular diffusion is neglected herein, though in many field problems it is not correct to neglect it. The relative roles of molecular and turbulent diffusion is discussed in an excellent review of turbulent diffusion by Batchelor and Townsend (1956). An incompressible fluid is assumed.

CLASSIFICATION OF FIELD, BOUNDARY, AND INITIAL CONDITIONS

There are very few major field problems such as radioactive-waste disposal in alluvial streams where large sums of money cannot be saved by considering the problem on paper before going into the field. Before doing this paper work, however, it is necessary to classify the problems to be resolved. This section will present a typical classification of field conditions which could be directed toward aiding theoretical analysis of turbulent diffusion in alluvial channels.

For theoretical purposes the field environment—which controls the boundary conditions—assuming an alluvial channel, could be classified in order of increasing complexity as follows—adding radioactive decay only at the end.

FLOW SYSTEM CONTAINING NEITHER SEDIMENT NOR BIOTA

This environment may be subdivided, as follows:

1. Channel of infinite width and constant slope with steady flow.
2. Channel of fixed finite width and constant slope with steady flow.
3. Channel of infinite width and constant slope with unsteady flow.
4. Channel of infinite width and variable slope with unsteady flow.
5. Finite width of uniform channel with unsteady flow.
6. Finite width of nonuniform channel with unsteady flow.
7. Finite width of uniform channel containing curves with steady flow.
8. Finite width of uniform channel containing curves with unsteady flow.
9. Finite width of nonuniform channel containing curves with steady flow.
10. Finite width of nonuniform channel containing curves with unsteady flow.

FLOW SYSTEM CONTAINING SEDIMENT BUT NOT BIOTA

The environment for this situation becomes even more complex than that described in the previous paragraph. Considering the theoretical approach that will probably first be applied to this condition—the continuity equation for mass—it seems that the presence of sediment can best be handled by assigning to the sediment field a spatially distributed sink strength for radioactivity. This will undoubtedly have to take into account the effect of sediment size and associated chemical activity. This sink must be added into the continuity equation; the description of the channel and flow environment remains the same as in the section above. Since the sediment will not always permanently retain the sorbed radioactivity, perhaps the artifice of a secondary source, effective sink strength or multicomponent continuity equations will have to be employed.

Apparently practically none of the existing diffusion theories have been applied to the above field condition. However, the basic continuity equation for conservation of mass for "k" phases has been formulated (de Groot, 1952). The interaction case such as exists with the condition of sediment being present is not clearly formulated in the diffusion theories.

FLOW SYSTEM CONTAINING BOTH SEDIMENT AND BIOTA

The problem of the first and second situation discussed is again compounded. To date, apparently neither the basic multiphase kinematic or dynamical equations involved have been published; nor have really appropriate boundary conditions been developed to include sediment and biota. However, the situation of a channel containing both sediment and biota is the most common one in nature.

Parker (1958) is one of the few to attempt an analytical expression for concentration including sediment and biota. He multiplied the expression for the concentration distribution derived from so-called Fickian theory for a simple geometry by e^{-Nt} where e is the base of Naperian logarithms, N is a "river-uptake coefficient" and t is the time. This procedure leaves much to be desired.

It seems reasonable to consider both the sediment and biota in the flow system as sinks for the radioactivity, to be added to the basic continuity equation. These sinks would have to be spatially distributed and, of course, have appropriate nuclear-decay multipliers incorporated. The biological sinks, and secondary sources such as dying zooplankton, would have to be idealized. Also, in using sinks and sources to represent the influence of biota, account would have to be taken, at least if short time periods are being considered, of the cyclic behavior both in time and location of such sources and sinks—due to diurnal feeding and rhythmic vertical migration of the biotic life.

RADIOACTIVE DECAY

Decay of the radioactive constituents in the waste can be accounted for in the theory for any of the above conditions by introducing the appropriate time-dependent exponential decay expressions.

Initial conditions for analytical purposes are mostly associated with the character of the source of contaminant. However, the relative mass density of the contaminant and the ambient flow field is also important—density currents may completely change the origin of the source in the analytical coordinate system employed as far as applying turbulent diffusion theory is concerned.

Common sources of radioactivity to be associated in the theory with the above environmental factors may be classified, in general, as follows:

1. Zero relative mass density of contaminant compared to ambient fluid.
2. Finite nonzero relative mass density.

- a. Plane source normal to the direction of the mean flow; instantaneous or of finite time.
- b. Source at or above the stream bed:
  1. Instantaneous point or volume source.
  2. Line source across or along the direction of flow; finite or infinite

The above situations are involved in setting initial conditions in an analytical study. Perhaps the most common is the continuous (or infinite) line source from a point, such as a pipeline outlet, in the direction of flow. A constant source strength, in time, is usually assumed. Also, in general, any relative mass density is usually due only to temperature differences of the two fluids. It seems that researchers have assumed, based on some evidence, that the viscosities and other physical properties of the waste effluents are nearly identical to those of pure water.

In considering the turbulent-diffusion problem with respect to the type of source, it becomes evident that in many situations consideration must be given to the turbulent diffusion of submerged jets. In this report, however, this problem has been considered as secondary since the scale of diffusion in the vicinity of the source is small compared to turbulent diffusion from the turbulence generated by channel-boundary shear and wind shear. Where density currents of waste products are suspected, the use of submerged-jet theory (Rouse, 1950) must be given attention if study of turbulent diffusion immediately adjacent to the source is desired. Jet theory also becomes important if the waste is discharged into the stream at velocities much different from the stream velocity. However, under other circumstances it may be desirable to use density-current flows to keep the waste concentrated in a small portion of the open channel.

TURBULENT-DIFFUSION THEORIES

Nearly all diffusion theories with which the hydraulics engineer is acquainted fall into two classes: the Fickian and the statistical. The Fickian theories are sometimes considered the "old" theories while the statistical are the "new" theories. A few theories, which have heretofore had only limited application in hydraulics, will also be mentioned.

The equation commonly used as a basis for analysis of diffusion is the Fickian equation extended to turbulent diffusion

$$\frac{\partial C}{\partial t}+\tilde{v}\cdot\nabla C=K\nabla^2 C \qquad (3\text{–}1)$$

where C is "concentration", t is time, $\tilde{v}$ is the velocity vector, K is the diffusion "constant" and ∇^2 is the Laplacian operator. The $\tilde{v}\cdot\nabla C$ term accounts for convection transfer. Seldom are the limitations of this equation mentioned, or the character of K, in applying it to hydraulic problems. A derivation adopted from Pai (1957) points out some of the background of this equation.

If $C=C(x,y,z,t)$ is the concentration at a point in space then $\int_v C dr=$total mass of subject material in v. By conservation of mass

$$\frac{\partial C}{\partial t}+\nabla\cdot(C\tilde{v})=0. \qquad (3\text{–}2)$$

Substituting in $\tilde{v}=\tilde{v}'+\bar{\tilde{v}}$ and $C=C'+\overline{C}$ with the customary definitions and applying Reynold's averaging rules,

$$\frac{\partial \overline{C}}{\partial t}+\frac{\partial}{\partial x}(\overline{C}\overline{u})+\frac{\partial}{\partial y}(\overline{C}\bar{\tilde{v}})+\frac{\partial}{\partial z}(\overline{C}\overline{w})=\frac{\partial}{\partial x}(-\overline{C'u'})$$
$$+\frac{\partial}{\partial y}(-\overline{C'\tilde{v}'})+\frac{\partial}{\partial z}(-\overline{C'w'}). \qquad (3\text{–}3)$$

Applying Boussinesq's exchange coefficients,

$$-\overline{C'u'}=v_{xx}\frac{\partial\overline{C}}{\partial x}+v_{xy}\frac{\partial\overline{C}}{\partial y}+v_{xz}\frac{\partial\overline{C}}{\partial z}$$

$$-\overline{C'v'}=v_{yx}\frac{\partial\overline{C}}{\partial x}+v_{yy}\ldots\frac{\partial\overline{C}}{\partial y}+v_{yz}\frac{\partial\overline{C}}{\partial z}$$

$$-\overline{C'w'}=v_{zx}\frac{\partial\overline{C}}{\partial x}+v_{zy}\ldots\frac{\partial\overline{C}}{\partial y}+v_{zz}\frac{\partial\overline{C}}{\partial z}. \qquad (3\text{–}4)$$

It is now evident that the nine v_{ij}'s define the diffusion tensor which in general is a function of the spatial coordinates.

It is frequently assumed that the x,y,z axes are the principal axes of the diffusion tensor. Here $v_{ij}=0$ for $i\neq j$. Now dropping the averaging bars, letting $K_i=v_{ij}$, where $i=j$, and assuming $\nabla\cdot\tilde{v}=0$,

$$\frac{\partial C}{\partial t}+\tilde{v}\cdot\nabla C=\sum_{i=1}^{3}\frac{\partial}{\partial xi}\left(K_i\frac{\partial C}{\partial x_i}\right). \qquad (3\text{–}5)$$

Assuming further that K_i is a constant such that $K_1=K_2=K_3$ (homogeneity) then

$$\frac{\partial C}{\partial t}+\tilde{v}\cdot\nabla C=K\nabla^2 C. \qquad (3\text{–}1)$$

Some of the available solutions of this equation for special cases, largely drawn from atmospheric diffusion, will be presented later.

Statistical turbulent-diffusion theories in Lagrangian form and their applications are well summarized (Frenkiel 1953). In general, only an infinite or semi-infinite domain has been considered. One of the most important points made by Frenkiel is that the Fickian theories are applicable only for large dispersion times such that

$$t>>L_h$$

where L_h is the Lagrangian time scale of turbulence such that

$$L_h=\int_0^\infty R_h(\alpha)d\alpha$$

in which R_h is the Lagrangian velocity auto-correlation coefficient defined by

$$R_h(t)=\frac{\overline{u'_A(t)u'A(t+h)}}{\sqrt{\overline{[u'_A(t)]^2}}\sqrt{\overline{[u'_A(t+h)]^2}}} \qquad (3\text{–}6)$$

The fundamental equation of statistical turbulent diffusion in the Lagrangian system in an isotropic and homogeneous turbulence field is

$$\overline{y^2}=2\overline{v'^2}\int_0^t(t-\alpha)R_h(\alpha)d\alpha=2\overline{v'^2}\int_0^t d\alpha_2\int_0^{\alpha_2}R_h(\alpha_1)d\alpha_1 \qquad (3\text{–}7)$$

where $\overline{y^2}$ is the variance of y or a measure of the diffusion in the y direction as a function of time. When $t>>L_h$ it follows that

$$\overline{y^2}\approx 2\overline{v'^2}L_h t$$

and when $t<<L_h$

$$\overline{y^2}\approx\left[1-\frac{1}{6}\frac{t^2}{\lambda_h^2}\right]\overline{v'^2}t^2 \qquad (3\text{–}8)$$

where

$$\frac{1}{\lambda_h^2}=-\frac{1}{2}\frac{d^2R_h(0)}{dh^2}.$$

The λ_h is called the Lagrangian microscale of turbulence. Also, if $t<<\lambda_h$,

$$\overline{y^2}\approx\overline{v'^2}t^2. \qquad (3\text{–}9)$$

Statistical turbulent-diffusion theories in Eulerian form have been worked on by several authors (Batchelor, 1951), (Taylor, 1921), (Goldstein, 1951), (Michelson, 1954). However, no application has been made to open-channel flow problems of waste disposal. These researchers have used or extended continuous stochastic processes or random-walk (discontinuous-movement) theories.

The continuous stochastic process approaches consist of finding the equation for the probability $P(\tilde{r},t)$ that the point defined by the vector $\tilde{r}$ lies in the volume of fluid containing the contaminant. Assuming only that the turbulent flow field is homogeneous and the mean flow zero,

$$\frac{\partial}{\partial t}[P(\tilde{r},t)]=v_{ij}\frac{\partial^2 P(\tilde{r},t)}{\partial x_i\partial x_j} \qquad (3\text{–}10)$$

from which it can be shown that v_{ij} is a tensor of second order. It can be shown that for very small $(t-t_0)$

$$v_{ij}\approx(t-t_0)U_i(t_0)U_j(t) \qquad (3\text{–}11)$$

and for very large $(t-t_0)$ that $v_{ij}\rightarrow$constant. Thus v_{ij} is initially zero, then increases linearly with time and finally tends to a constant value—thus, demonstrating that for large times v_{ij} can be replaced by K=constant.

The random-walk or discontinuous-movement method of treating turbulent diffusion, an application of the theory of random functions, was first extended by Taylor (1921) in considering diffusion by continuous movements. This resulted in the same equation as that given immediately above. Goldstein (1951) extended Taylor's analysis upon quite rational grounds. Goldstein's equation has the form of the telegraph equation of theoretical physics. Several diffusion cases can be derived from his basic-difference equation. For isotropic diffusion in one dimension—the motions are continuous and the displacements take place at finite velocities—the basic diffusion equation is

$$\frac{\partial^2 C(y,t)}{\partial t^2}+\frac{1}{A}\frac{\partial C(y,t)}{\partial t}=v^2\frac{\partial^2 C(y,t)}{\partial y^2} \qquad (3\text{–}12)$$

where v satisfies $y=vt$ and A is a constant. For anisotropic diffusion, the equation becomes

$$\frac{\partial^2 C}{\partial t^2}+\left(\frac{1}{A}+\frac{1}{B}\right)\frac{\partial C}{\partial t}+\frac{v}{B}\frac{\partial C}{\partial y}-v^2\frac{\partial^2 C}{\partial y^2}=0. \qquad (3\text{–}13)$$

The first form above of the telegraph equation corresponds to the case of no leakage from the cable. The second case includes leakage. These equations are derived on the assumption of partial correlations between the directions of motions in any two consecutive time increments dt. Also, these equations are of the hyperbolic type—the solutions are somewhat different from the parabolic equations of the Fickian theory.

The above two equations, which appear on theoretical grounds to more truly represent the diffusion phenomena, at least in their derivation, have been only incompletely applied to the diffusion problem. Michelson (1954) has investigated some points in the mathematics. It is significant to note that the parabolic form of the turbulent-diffusion equation can be derived from the same difference equation by assuming zero correlation between consecutive motions and that the particles move with infinite velocity. Harleman (1959) has pointed out that, close to the source, the solution of these equations differs materially from the Fickian theory solutions.

In the category of miscellaneous theories, there is Richardson's (1926) distance-neighbor theory, which has some similarity to Kolmogoroff's subsequent work. In Richardson's theory the basic equation for each range of separation L, for the distribution of a quantity P is

$$\frac{\partial P}{\partial t}=\frac{\partial}{\partial L}\left[F(L)\,\frac{\partial P}{\partial L}\right] \qquad (3\text{–}14)$$

where

$$F(L)\approx \sigma L^{4/3}$$

in which σ is a diffusion parameter. Comparison to the Fickian theory quickly shows the dependency of the diffusion on the scale of the phenomena being investigated. In a confined channel the scale must, of course, reach a maximum.

In the development of a theory to explain the increase of the diffusion coefficient K in the Fickian theory with distance from the source in a semi-infinite domain, Lettau (1951) introduced a term for the advection due to boundary shear. He replaces the usual K by $K_{(\text{apparent})}$ where

$$K_{(\text{apparent})}=K+K\,\frac{\partial u}{\partial z}\left(\frac{x-x_0}{u}\right)$$

where $(x-x_0)$ is the distance from the source.

SOME THEORETICAL SOLUTIONS NOW AVAILABLE

One of the best ways to evaluate the present level of theoretical knowledge of turbulent diffusion in alluvial open channels is to summarize some of the theoretical solutions now available. The subsequent tabulation will make clear that not even the simplest of conditions involving sources and sinks, such as used to represent sediment and biota, has been resolved for open channels. Also, only very simple conditions in confined channels have been treated—most works discuss semi-infinite domains with a unidirectional flow. However, consideration has been given by various authors to vertical velocity profiles, elevated sources and thermal stability (in the atmosphere). Crank (1956) includes some solutions for conditions where the diffusion coefficient is dependent on concentration.

For Fickian theories:

1. Instantaneous point source at the origin in an infinite isotropic homogeneous turbulence field.

   If a quantity Q of material is released at a point source at $t=0$ from an origin moving with the mean flow, the concentration given by Roberts (1923) is

$$C(\bar{r},t)=\frac{Q}{(4\pi Kt)^{3/2}}\,e^{-\left(\frac{r^2}{4Kt}\right)}. \qquad (3\text{–}15)$$

2. Instantaneous point source at the origin in an infinite anisotropic homogeneous turbulence field.

   Releasing the isotropy condition above, Roberts (1923) gives

$$C(x,y,z,t)=\frac{Q}{(4\pi t)^{3/2}\sqrt{K_xK_yK_z}}\,e^{-\frac{1}{4t}\left(\frac{x^2}{K_x}+\frac{y^2}{K_y}+\frac{z^2}{K_z}\right)}. \qquad (3\text{–}16)$$

3. Continuous infinite line source along the bed, normal to the stream velocity.

   Roberts (1923), under the conditions of a velocity profile such that

$$u=\bar{u}_1\left(\frac{z}{z_1}\right)^m;\ m\geq 0;\ K_z=K_1\left(\frac{z}{z_1}\right)^{1-m};\ \frac{\partial C}{\partial t}=0$$

gives the concentration as

$$C(x,z)=\frac{Q}{\bar{u}_1\Gamma(s)}\left[\frac{\bar{u}_1}{(m-n+2)^2K_1x}\right]^s e^{-\left[\frac{\bar{u}_1 z^{(m-n+2)}}{(m-n+2)^2K_1x}\right]}, \qquad (3\text{–}17)$$

where $S=\dfrac{(m+1)}{(m-n+2)}$, $n=1-m$, and Γ is the gamma function.

4. Same as case 3 immediately preceding, but with approximate logarithmic velocity profile. See Calder (1949).
5. Continuous point source, infinite isotropic domain.

   Roberts (1923), when Q is the emission rate, $r^2=x^2+y^2+z^2$ and $\overline{U}$ is the mean velocity in the x-direction, gives

$$C(x,y,z)=\frac{Q}{4\pi Kr}\, e^{\frac{-\overline{U}}{4Kr}(y^2+z^2)}. \tag{3-18}$$

6. Continuous point source, infinite anisotropic domain. Roberts (1923), when Q is the emission rate, $r^2=x^2+y^2+z^2$ and $\overline{U}$ is the mean velocity in the x-direction, gives

$$C(x,y,z)=\frac{Q}{4\pi r\sqrt{K_yK_z}}\, e^{-\frac{\overline{U}}{4r}\left[\frac{y^2}{K_y}+\frac{z^2}{K_z}\right]}. \tag{3-19}$$

7. Continuous infinite cross-stream line source in bed of isotropic homogeneous flow field.

Roberts (1923), where Q is again the emission rate, states

$$C(x,z)=\frac{Q}{2(\pi\overline{U}\sqrt{x^2+z^2})^{1/2}}\, e^{-\frac{\overline{U}z^2}{4K\sqrt{x^2+z^2}}}. \tag{3-20}$$

Solutions for continuous infinite elevated cross-stream sources in isotropic and anisotropic conditions may be found in Sutton (1953). Kellogg (1952) has considered spherical volume sources. Yih (1952) solved for several cases with m and n independent the two-dimensional equation

$$z^m\left(\frac{\partial c}{\partial x}\right)=D\frac{\partial}{\partial z}\left(z^n\frac{\partial c}{\partial z}\right). \tag{3-21}$$

Many other special conditions are considered in the literature.—See, for example, the references in Meteorological Abstracts (1953).

For the statistical theories, Frenkiel (1953) gives:

1. Instantaneous point source at the origin in an infinite isotropic homogeneous region, where Q is the quantity of material released,

$$C(x,y,z,t)=\frac{Q}{(4\pi\overline{y^2})^{3/2}}\, e^{\frac{-r^2}{2\overline{y^2}}}. \tag{3-22}$$

Gifford (1955) has considered relatively arbitrary volume sources related to this.

2. Instantaneous point source at the origin in an infinite anisotropic homogeneous region, where Q is the total quantity of material introduced,

$$C(x,y,z,t)=\frac{Q}{(2\pi)^{3/2}(\overline{x^2}+\overline{y^2}+\overline{z^2})^{1/2}}\, e^{-\frac{1}{2}\left[\frac{x^2}{\overline{x^2}}+\frac{y^2}{\overline{y^2}}+\frac{z^2}{\overline{z^2}}\right]}. \tag{3-23}$$

3. Continuous point source in an infinite isotropic homogeneous domain with origin at the source

$$\left.\begin{matrix}\text{Relative mean}\\ \text{concentration}\end{matrix}\right\}=\frac{1}{(2\pi)^{3/2}\overline{U}}\int_0^\infty \frac{1}{(\overline{y^2})^{3/2}}\, e^{-\left[\frac{(x-\overline{U}t)^2+y^2+z^2}{2\overline{y^2}}\right]}dz. \tag{3-24}$$

(Solutions are also given for short and long distances from the source.)

4. Infinite-line sources normal to mean velocity. Frenkiel also gives solutions for this situation. They are too lengthy to present here.

EXPERIMENTS ASSOCIATED WITH THEORY

Very few experiments directly applicable to turbulent diffusion of material from sources in alluvial open channels are available. Those of Taylor (1954), Orlob (1958), Parker (1958), and Harleman and others (1959) seem the most applicable. There are, however, some excellent papers in the literature on the use of radioisotopes for beach- and harbor-drift studies. Also, many papers are available on experiments in atmospheric and oceanographic diffusion, normally including only a plane horizontal boundary and no confining walls or hills. Few of the papers consider, analytically or experimentally, the effect of sediment or biology on waste dispersion.

Taylor (1954) solved analytically for the diffusion coefficient K for flow in a uniform straight circular pipe in the turbulent-flow regime. His result is

$$K=10.1r_0U\frac{\sqrt{\tau_0/\rho}}{U}.$$

His analysis is based on the "universal velocity-distribution" equation of mixing-length theory, where r_0 is the pipe radius, U the mean velocity, $\sqrt{\tau_0/\rho}$ the shear velocity. He then compared this to experiments in straight and curved pipelines. It was demonstrated that the theory compared well with experiments for a straight pipeline, but not for a curved pipeline.

The expression for K may be transformed so as to apply to the open-channel situation. The result is

$$K=7.15RU\sqrt{f}=20.2R\sqrt{\tau_0/\rho}, \tag{3-25}$$

where R is the hydraulic radius, U the mean stream velocity and f the Darcy-Weisbach coefficient of channel friction. Taylor does not give the required magnitude of the dispersion time before the equation has reasonable validity.

Orlob (1958) conducted an excellent piece of hydraulic research on turbulent diffusion. He investigated the turbulent diffusion of floating polyethylene particles in the homogeneous turbulence field at the surface of a uniform steady open-channel flow in a flume. He demonstrated that the Fickian diffusion coefficient increased rapidly close to the source and gradually approached a constant value as distance from the source increased. Some indications of a preliminary nature are given for turbulence intensity and scale as a function of Reynolds number, channel slope, and other parameters. These are pioneering experiments in obtaining turbulence measurements in open channels.

Parker (1958) reported on several field experiments on pipelines, reservoirs and open channels. These experiments do not adequately elucidate turbulent diffusion in open channels. Field experiments by other scientists have also done little to date to explain diffusion in open channels. Theory and experiment frequently give concentrations differing by 100 percent even when no sediment or biota is present.

Glover (1956, 1957, unpublished data) reported on two laboratory experiments and one field experiment. These data will be of value in future work. Glover also reported some theoretical work; largely similar to the work of earlier investigators. He did, however, attempt to consider the effect of vertical boundaries, confining a stream, perhaps the only author other than Taylor (1954) to do so. Unlike Taylor, however, Glover did not consider in an explicit manner the influence of velocity gradients. Glover considered the diffusion coefficient K to be a constant.

Harleman and others (1959), have conducted both an extensive review of the literature and some very fundamental laboratory research. Primary emphasis was on the problem of waste disposal in estuaries and other large bodies of water. They concluded from one-dimensional tests in a turbulence flume and from review of the literature, that for waste disposal problems having large dispersion times, the Fickian theory is as satisfactory as the statistical theories. They also made a suggestion for translation of coordinates in order to account for the effect of near-source deviations of the diffusion coefficient K from a constant value.

SORPTION OF RADIOCONTAMINANTS BY STREAM SEDIMENTS

One aspect of the problem of radiocontaminants in streams which has received relatively little systematic attention is concerned with the role of fluvial sediment in dispersing, transporting, and concentrating radioactive substances. Indications are that sorption reactions are the rule rather than the exception when extremely dilute solutions, such as low-level liquid wastes, come into contact with solid surfaces (Carritt and Goodgal, 1953). The concentration of radioactive substances on the surface of sediment particles may be many thousands of times greater than the concentration in the surrounding solution.

When significant amounts of radioactivity are sorbed by sediment particles, knowledge of the sediment supply and the sediment-transport characteristics of the stream clearly becomes an important factor in assessing the rate of dispersal and the ultimate distribution of radioactivity in the stream environment.

Sorption of radionuclides can be either an asset or a liability. Under controlled conditions, sorption of radionuclides by sediment can effectively reduce the concentration of dissolved radionuclides in the waste water. In a natural stream environment, however, uncontrolled sorption of radioactive substances by sediments must be considered as a potential hazard. Radioactivity could build up over long periods of time in sections of a stream where the rate of accumulation of radioactivity induced by sediment deposition exceeds the rate of natural radioactive decay. Such deposits might occur along the convex banks of meandering rivers, or in reaches of a stream where flow velocities are reduced. Some of these contaminated deposits would be periodically flushed out at high water, and then be redeposited farther downstream as the floodflow receded.

The key to the sediment aspect of the problem of low-level liquid-waste disposal is sorption. To what extent does sorption occur? What forms of sorption are involved? What factors control the extent of sorption, and what are the relationships among these factors? For the most part only qualitative answers can be given to these questions at the present time. Many of these are at best tentative and incomplete. Following is a brief summary of: (1) current sorption theories, (2) some of the relevant literature, and (3) factors determining the extent of sorption.

SORPTION THEORIES

Sorption may be described as the process of taking up and holding either by adsorption or absorption. In solutions, adsorption refers to the uptake and concentration of a solute at the surface of a sorbent, whereas absorption implies a generally uniform penetration of the solute into the sorbent. Sorption reactions involving stream sediments and radioactive solutes generally belong in the adsorption category. However, in practice it is often impossible to separate the effects of adsorption from those of absorption. Consequently, the nonspecific term "sorption" will be used in the remainder of this section.

According to McBain (1950), sorption comprises a number of phenomena, the most commonly recognized of which is the accumulation of one or more substances at an interface. Combination of the sorbed substance with a solid at an interface may involve strong chemical-valence bonds—chemisorption—or weaker physical interaction through van der Waals forces—physical sorption—or both. The uptake of radioactivity by stream sediments may involve either chemisorption or physical sorption; frequently it involves both types, whence analysis of the problem becomes considerably more complicated.

Sorption may occur wherever there is a surface or an interface. Sorption may occur at any surface; the most important, and most complex surfaces, however, are the interfaces between solutions and solids, such as the surfaces of colloidal particles in a liquid medium. The substances sorbed at an interface may be products of reaction or hydrolysis, unaltered molecules, or particular ions. Sorption may be complicated by the orientation of molecules at the interface, by diffusion into a solid, and by the surface mobility of sorbed molecules.

According to Daniels (1948), there must be a layer of atoms and molecules at the extreme surface of a solid which have some valence forces or other attractive forces that are not so fully utilized as those associated with the atoms and molecules in the interior which are completely surrounded by other molecules. The residual forces on the surface of sediment particles in solution are used up, and the free energy reduced, by means of sorption reactions. The extent of sorption may vary widely. It is dependent on the specific nature of the sorbent solid and the substance being sorbed, the available surface area, the concentration and types of solute in solution, the time, and the temperature.

Surface-tension phenomena are known to be intimately related to sorption reactions, but the mechanics of tensions at solid-liquid interfaces are not very well understood. It is known though that when the surface tension of a solvent remains constant as the concentration of solute increases, no sorption occurs (McBain, 1950). This behavior is typical over a certain range of solute concentration in solutions containing soap or a detergent. Neither the critical range of solute concentration nor the requisite concentration of soap or detergent is defined.

Sorption isotherm.—The Freundlich equation, which is derived from the empirical classical isotherm, states that

$$\frac{x}{m}=kc^n \qquad (4\text{–}1)$$

when

x is the weight of material sorbed by m grams of sorbent,
c is the concentration of solution in equilibrium with the solid,
k is an empirical constant, and
n is an empirical constant having a value between about 0.5 and 0.1.

The values of the empirical constants k and n are determined by experiment for each solute, adsorbent and temperature. Temperature, however, according to McBain (1950), does not greatly affect sorption from solutions.

The classical isotherm applies with fair accuracy to the various types of sorption from solution over a very wide range of concentrations. The form of the isotherm indicates correctly that appreciable sorption occurs even at extremely low concentrations, especially at low values of n. It has been shown that c is a true equilibrium concentration which is reached by both sorption and desorption for a particular value of x/m.

The classical isotherm holds true only for definite total times of exposure. Owing to the inaccessibility of fine pores of solids, the competition between solvent and solute, and the diffusion into the solid, sorption reactions may require long periods of time, even up to years, to reach completion. McBain (1950) gives Burt's empirical formula

$$\frac{x}{m}=kt^n \qquad (4\text{–}2)$$

where t is now time and k and n again are empirical constants, for the time effect. For very prolonged sorption this expression is modified to the form

$$\log\frac{\sigma}{\sigma-s}=kt^n \qquad (4\text{–}3)$$

where σ is the saturation value and s is the value of x/m at any given time.

Competition between solute and solvent for the surface of a solid is an important factor when solution concentrations are high. However, in extremely dilute solutions, such as most low-level radioactive waste liquids, sorption of the solvent has only a negligible effect upon sorption of the solute.

In mixed solutions which contain more than one solute the competition is more complicated, and the attainment of equilibrium is slower. Each solute lessens the sorption of others; however, it has been demonstrated that in time the solutes which sorb strongly largely displace the more weakly sorbed solutes.

Physical sorption.—Physical sorption is characterized by small heats of adsorption, by reversibility, and by rapid attainment of equilibrium (Glasstone, 1940). Physical sorption is nonspecific in character and occurs on chemically inactive surfaces (Taylor and Taylor, 1942). This type of sorption is attributed to the effect of van der Waals forces which are thought to result from molecular attraction due to dispersion forces. The classical isotherm is applicable to systems in which physical sorption predominates.

Sorption of solutes on the surface of sand and silt particles in streams is primarily physical. Although physical sorption also occurs with colloidal clay particles, it is secondary to other types of sorption, particularly ion exchange.

Owing to the reversibility and rapid attainment of equilibrium which are typical of physical sorption, the concentration of solutes on the surface of sand and silt particles would tend to be quite sensitive to the concentration of solutes in the water. The sensitivity would be less when diffusion of the solute into the solid is a significant factor.

Ion exchange.—Ion exchange, which is a type of chemisorption, may be described as the process by which ions sorbed at the surface of a solid are replaced by other ions of like sign. In a disperse system, such as that represented by a suspension of sediment particles in water, ion exchange occurs when ions in solution first displace and then replace ions which are sorbed on the surface of the particles.

In soil chemistry, ion-exchange phenomena are generally associated with the fine-grained soils and in particular with the expanding-lattice montmorillonite clays. Ion exchange occurs to some extent with larger grained sediments; however, exchange capacity diminishes rapidly as particle size increases. Ion-exchange capacity depends both on the specific surface and the chemical and mineralogical characteristics of the sediments involved.

Particles in a colloidal suspension carry an electric charge. Most clay particles carry a negative charge and in a colloidal system behave essentially as anions (Baver, 1956). Consequently the negatively-charged clay particles sorb available positively-charged cations from solution. This phenomenon is known as ionic adsorption. The magnitude of the electric charge on the clay particles is known as the zeta potential. High zeta potentials are associated with high ionic-adsorption capacities and stable suspensions. Conversely potentials of small magnitude are associated with flocculation and subsequent settling of clay particles.

In the cation-exchange process, those ions which are sorbed strongly tend to displace those which are not as strongly sorbed. The energy with which an adsorbed cation is held to a particular type of clay particle, in an aqueous environment, is determined by considerations relating to hydration and valence (Baver, 1956). For isovalent cations the bonding energy varies inversely with the effective radius of the hydrated ion, and hence directly with the atomic weight. The relative bonding energies of some common exchangeable cations may be expressed in terms of the Hofmeister or lyotropic ion series

$$Cs^+ > Rb^+ > K^+ > Na^+ > Li^+$$

for monovalent cations, and

$$Ba^{++} > Sr^{++} > Ca^{++} > Mg^{++}$$

for divalent cations.

Bonding energy tends to increase with valence, although there is a slight amount of overlap at the beginning of the monovalent series and the end of the divalent series.

Considering the facts that: (1) Sr^{90} and Cs^{137} are the most abundant of the long-lived fission products, and (2) Ca, Mg, K, Na are among the most abundant exchangeable cations in clay-mineral soils, the relative positions of the ions in the Hofmeister series is of especially great practical significance where waste solutions contain fission products.

An exception to the rule is the banding energy associated with the hydrogen ion, which exceeds that of the divalent cations. This apparent contradiction is generally attributed to the explanation that the adsorbed hydrogen is actually in the form of a hydronium ion $(H_3O)+$.

Jenny and Reitemeier (1935), who contributed a great deal toward establishing the foregoing relationships, also demonstrated that ionic exchange increases approximately as the sixth power of the zeta potential.

Thus the close relationship among the various phenomena of cation exchange, energy of adsorption, zeta potential and flocculation seem to be fairly well established.

The chemical and mineralogical nature of clays as well as the sorbed cations are instrumental in determining the surface behavior of clay particles. Important properties of some clay minerals are listed in table 2.

LITERATURE REVIEW—SORPTION OF RADIONUCLIDES

Radioactive materials are essentially identical with their stable counterparts in ordinary chemical and physical behavior. Although sorption is influenced to a slight extent by the difference between isotopic weights of radioisotopes and corresponding stable isotopes, this effect is negligible when the atomic number of the sorbed ion is greater than 10. In fact the influence is so small that radioactive tracers are commonly used in quantitative investigations of sorption reactions. Few studies have been undertaken, however, with a definite view toward obtaining a better understanding of the role of fluvial sediments in concentrating radioactivity by sorption. Studies by Carritt and Goodgal (1953) and Barker (1958) are among the few which are directly applicable to the problem.

Carritt and Goodgal studied the uptake of various radioactive components from solution by suspensions of Roanoke River and Chesapeake Bay sediments, bentonite, fuller's earth, and powdered Pyrex glass. Each of the solutions studied contained one of the following dissolved radioactive substances: Phosphate, iodide, sulfate, strontium, copper, and iron. Phosphate, iodide, and sulfate occur as anions in waste solutions.

TABLE 2.—*Important properties of some clay minerals*

[After R. E. Danielson, 1956, written communication]

Group and lattice type	Name	Typical empirical formula	Cation-exchange capacity (meg/100q)	Approximate size range (μ)
Kaolinite group (1:1 lattice)	Kaolinite	$Al_2Si_2O_5(OH)_4$	3–15	0. 5–2
	Halloysite	$Al_2Si_2O_5(OH)_4nH_2O$	40–50	. 04–. 2
Montmorillonite group (2:1 expanding lattice).	Montmorillonite	$(Al_{2.67}Mg_{.33})Si_4O_{10}(OH)_2Na_{.33}\ nH_2O$	80–125	. 01–. 1
	Beidellite	$Al_{2.16}(Al_{.88}Si_{3.14})O_{10}(OH)_2Na_{.33}\ nH_2O$	60–90	. 05–. 5
	Nontronite	$Fe_2(Al_{.33}Si_{3.67})O_{10}(OH)_2Na_{.33}\ nH_2O$	60–70	. 01–. 1
	Saponite	$Mg_3(Al_{.33}Si_{3.67})O_{10}(OH)_2Na_{.33}\ nH_2O$	20–30	. 01–. 1
Illite or hydrated mica group (2:1 nonexpanding lattice).	Illite	$(Al_{1.76}Mg_{.36})(Al_{.60}Si_{3.40})O_{10}(OH)_2K_{.60}$	20–40	
	Pyrophyllite	$Al_4Si_8O_{20}(OH)_4$		

Strontium, iron, and copper usually occur as cations in solutions. Additional variables which were either controlled or measured in the experiments were pH (hydrogen-ion concentration), temperature, contact time, concentration of suspended solids and reacting dissolved solids, and ionic strength.

The methods employed in these experiments were simple. Basically they consisted of assembling measured amounts of the various ingredients, placing them in a stoppered flask, keeping the contents in suspension by means of a shaking machine, and withdrawing samples for analysis at periodic time intervals. The solids in the samples were centrifuged from suspension and analyzed for radioactivity. Analysis of the supernatant solution for radioactivity provided an independent check.

The results of the experiments by Carritt and Goodgal indicate that, depending upon the specific conditions involved, any of or all the variables they investigated may have an important influence on the rate and extent of sorption. Most of their data plotted according to the classical empirical isotherm, and the extent of sorption was definitely shown to increase with contact time. Not much can be added to the above in the way of general conclusions. The data are too few and the variables too many to permit an adequate evaluation of the effects of pH, temperature, ionic strength, and concentration of suspended solids which would cover all the combinations of sediment types and radioactive components.

Some of the more specific conclusions reached by Carritt and Goodgal (1953) are as follows:

1. The uptake of phosphorus by Roanoke River solids—bentonite and fuller's earth—was shown to be maximum in the pH range (4–7) in which the $H_2PO_4^-$ ion predominates;
2. It was demonstrated that the mechanism by which phosphorus was taken up involved both a rapid adsorption process and a slow diffusion process;
3. Strontium, copper, and ferric iron were found to precipitate from solution as basic salts under appropriate conditions. Added solids acted as a "sweeper" aiding the settling of finely divided precipitate;
4. Very little iodide was sorbed by any of the suspended solids.
5. The sorption of sulfate sulfur by Roanoke River sediment was found to be maximum in the pH range below 4, which favors formation of the HSO_4^- ion.

In general, the percentage of radionuclides sorbed by sediments with respect to the initial quantity of radionuclides in solution ranged between 10 percent and 90 percent in Carritt and Goodgal's experiments. The initial concentrations of radioactivity in these experiments were several orders of magnitude in excess of the recommended maximum permissible concentrations. In natural streams where radioactivity concentrations might be on the same order of magnitude as the MPC, a greater percent of sorption would normally be expected under otherwise comparable conditions.

Calculations based on one of the isotherm tests in which the sediment concentration was 629 ppm of Roanoke River solids, and the initial solution concentration was 11.5 microgram-atoms of phosphorus per liter, showed that approximately 10 percent of the phosphorus was sorbed in 1 minute at which time the concentration of phosphorus on the surface of the sediment particles was estimated to be more than 10,000 times that in the surrounding solution.

Barker (1958) conducted sets of experiments similar to those of Carritt and Goodgal in which sorption due to ion exchange of cesium and strontium by kaolinite, illite, halloysite and two types of montmorillonite was investigated. Cation-exchange capacity determinations of these clays through the use of trace amounts of Sr^{90} and Cs^{137} compared well with exchange-capacity determinations by other methods. The effect of various concentrations of sodium chloride and calcium chloride, which are common electrolytes in natural waters, on the sorption of carrier-free strontium and cesium by illite, halloysite and montmorillonite 21 (bentonite) also was investigated. Sorption of cesium and strontium by these three clay minerals from solutions containing added amounts of strontium nitrate and cesium chloride carriers was studied in addition. Sediment concentrations and contact times were maintained constant at 4,000 ppm and 17 hr in these experiments.

Barker found that the percentage of carrier-free cesium sorbed by clay is decreased as the concentration of calcium or sodium in solution is increased. Results for the calcium and sodium solutions were very nearly the same. The concentration of calcium was found to have a much greater effect on the sorption of strontium than the concentration of sodium. The fact the strontium sorption from the calcium solution was considerably less is attributed to the marked chemical resemblance between strontium and calcium, the ions of which compete for exchange positions on a nearly equal basis. Barker concluded that as a first approximation the sorption of small quantities of cesium by clays from most natural waters would depend only upon the total concentration of cations in the water, but that predictions concerning the sorption of strontium would necessitate the additional consideration of the sodium-calcium ratio in the water.

Comparison between the results of the carrier-free and the carrier-added experiments indicated that both the concentration of radioactive substance and the concentration of sodium and calcium affect the amount of radioactivity sorbed. In general, however, changes in the total ionic concentration of the solution were found to have a greater effect than changes in the cesium or strontium concentration within the range to be expected in most natural waters.

In summary it might be said that Carritt and Goodgal's more comprehensive work, involving a relatively large number of variables, points the way toward many areas which require further attention. However, Barker's more intensive work, though smaller in scope, has resulted in more definitive answers. Both approaches are needed at this time.

FACTORS DETERMINING EXTENT OF SORPTION

From the foregoing discussion it is clearly evident that characteristics of the sediment and the waste effluent, and the quality of stream water are all intimately related to sorption phenomena involving radioactive waste components and stream sediments. Considering the relatively meager amount of available knowledge and data on the subject it would be presumptuous to draw conclusions pertaining to the relative importance of the many factors involved. Factors which are known to be significant and their estimated qualitative effects are listed in the following paragraphs.

SEDIMENT CHARACTERISTICS

Probably the most important single sediment characteristic affecting the extent of sorption is particle size. Particle size is important in determining both the extent and type of sorption. Sorption capacity of stream sediments is roughly proportional to the available surface area. From figure 1 it is seen that for a given weight or concentration of spherical sediment particles, the total surface area of 1-micron clay would be 1,000 times as great as that for 1-millimeter sand. The fact that the chemical activity of soil particles tends to decrease as the particle size increases is instrumental in making ion exchange the normally dominant mechanism in sorption by clays, and physical sorption the principal mechanism in sorption by sands and silts.

The chemical and mineralogic nature of sediments, particularly of clays, is also important in determining the extent of sorption. The cation-exchange capacity of the clay and the position of the adsorbed cations in the Hofmeister ion series with respect to that of the radioactive cations in solution are significant factors in this regard.

Apparently very little information which relates various types of sediments to their capacities for absorption by diffusion of a solute is available.

Concentration of sediment in a stream is important insofar as it is related to the total surface area available for sorption reactions.

CHARACTERISTICS OF WASTE EFFLUENT AND STREAM WATER

Waste effluent and stream water are considered together because it is the mixture of the two which determines the characteristics of the solution in the sorption system.

Concentration of radioactive waste effluent in stream water is of course an important factor in determining the amount of radioactivity which is sorbed by stream sediments. The classical or Freundlich isotherm equation indicates that for a particular solution-sediment system, the amount sorbed depends largely on the concentration in the solution of the material being sorbed. Any one of a number of chemical characteristics of the waste effluent-stream solution may, however, radically alter the complexion of the entire sediment-solution sorption system.

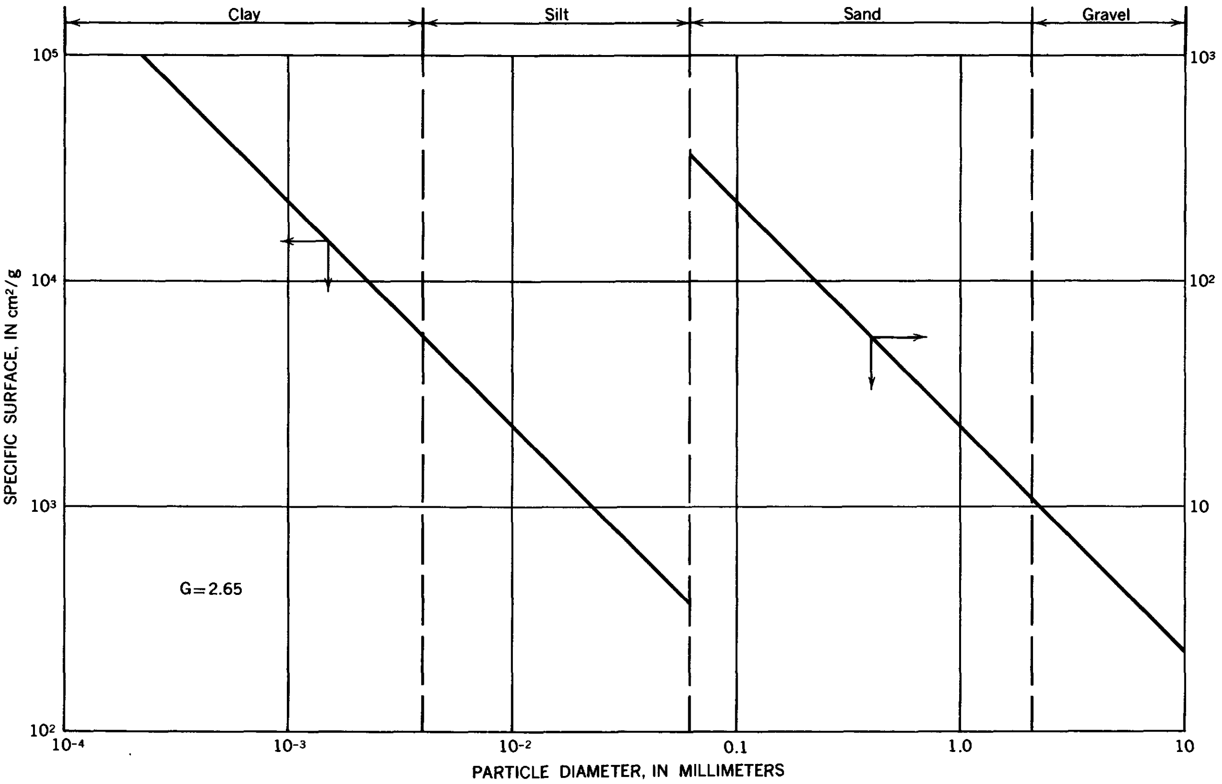


FIGURE 1.—Variation of specific surface of spherical sediment particles (sp gr 2.65) with particle diameter.

The composition of the radionuclide-bearing substance is important in this regard. For example, systems containing appreciable quantities of sediments with exchangeable cations would show a marked preference for sorbing cations over anions, regardless of whether the cations and anions are radioactive or stable.

In systems involving ion exchange the position in the Hofmeister ion series of the radioactive cation is also a significant factor. As indicated on p. 13, exchange occurs according to the order of the cations in the series; cations toward the beginning of the series tend to displace those which follow. Also, polyvalent cations tend to displace divalent cations, which in turn tend to displace monovalent cations.

The presence in the solution of competing nonradioactive substances may greatly reduce the uptake of radioactivity by sediments. In streams, competing nonradioactive substances might originate from other types of industrial wastes or from natural sources.

It has been demonstrated that the extent of sorption may be greatly influenced by the effect of the pH of the solution on the ionic forms in solution. Changes in the pH of the solution lead to the formation of different ions which may be either more or less readily sorbed than before. The critical pH range depends on the various components of the system. Considerable further investigation is required in this area.

Another area requiring investigation is the effect of detergents in solution on sorption. It is well known that detergents reduce the surface tension of water, and it is thought that sorption is reduced by the presence of detergents in solution. However, the relationship between the concentration of detergent and the effect on sorption is largely unknown.

Under appropriate conditions it is known that certain radioactive ions will combine as salts with other materials and precipitate from solution. Although uptake of radioactivity by sediments will be reduced by the extent to which the concentration of radioactivity in solution is decreased by precipitation reactions, the radioactive precipitates may themselves become a source of hazard. Detergents in solution tend to inhibit precipitation.

Concentrations of flocculating agents in solution sufficient to induce flocculation and subsequent deposition of clay particles constitute another source of potential hazard. The addition of neutral salts will in general reduce the zeta potential or negative charge of clay particles, thereby inducing flocculation. The flocculating power of a salt increases with the sorbability of its cation component. Flocculating agents in natural streams may arise from such sources as industrial wastes and the intrusion of brackish tidal waters.

The physical characteristics of the waste effluent such as density and viscosity affect uptake by sediments insofar as they affect the distribution of the radioactive components over the stream cross section by turbulent diffusion. Sorption from solution is to some extent affected by the temperature of the solution, but temperature effects over the normal range of stream temperatures generally seem to be secondary to other considerations.

It is apparent from the preceding discussion that the factors influencing the uptake of radioactivity by sediments are many, varied, and often not very well understood. In natural streams the sorption systems are further complicated by the fact that several factors will normally be operating simultaneously, thereby obscuring cause and effect relationships. A great deal of additional research and information is a prerequisite to adequate criteria for predicting the extent to which sorption of radiocontaminants by sediments may occur in natural streams.

SEDIMENT TRANSPORT AS AFFECTED BY DRAINAGE AREA AND STREAM CHARACTERISTICS

Einstein (1950) has classified the total sediment load or sediment transport of a stream into fine-material load and bed-material load. The fine-material load consists of particles finer than most of the material found in the stream bed, whereas particles composing the bed-material load are found abundantly in the bed.

The particles of fine-material load are moved by small fluid forces and, therefore, are in nearly continuous suspension by natural streams. The quantity of such material in transport by a stream then depends on the rate at which these fine particles become available as affected by the environment or characteristics of the drainage area, and not on the ability of the flow to transport them. On the other hand, sediment of the coarser sizes found in large quantities in the stream bed of alluvial channels will be transported at a rate depending on the relative capacity of the stream for moving these sizes.

Experience of the Geological Survey in measuring sediment transport during the past 2 decades substantiates these general theories. Most streams readily transport silt and clay fractions (as much as 0.062 mm in diameter) as wash load. The quantity of these fines moved by the stream at a given time is very nearly equal to that released by the environmental factors within the drainage basin. The quantity of the various coarser sizes (sand and gravel) in transport is closely related to water discharge—if the supply is not proportional to the water discharge, then the stream will aggrade or degrade its bed, thus maintaining its load-capacity relationship.

The sediment load, or transport, of a stream then involves variables from two categories, as follows:

1. Environmental factors associated with the supply of sediment from the drainage area and included within the broad terms of climate, physical character of the watershed, and land use.
2. Stream channel and flow factors associated with the quantity of flow, channel geometry, particle size of sediment in transport, and channel control.

As expected, the difficult and broad subject of the environmental factors has been investigated in only small and usually unrelated segments—unrelated because studied in different geographical areas and with different objectives. Examples would include the data from small reservoir surveys collected by various agencies of the Department of Agriculture and the Department of the Interior and by some State agencies. Limited interpretation of these data is underway. (See Glymph, 1954; Maner, 1958; Langbein and Schumm, 1958; and Stall and Bartelli, 1959.) The area of investigation is so large that progress seems negligible.

Stream channel and flow characteristics of streams, on the other hand, have been or are in a more advanced state of investigation. The magnitude and pattern of streamflow have been intensively studied on a routine basis. In the process of obtaining these data, much information about channel geometry is being obtained. Channel roughness and certain hydraulic characteristics as may affect sediment transport are being actively investigated with recirculating flumes. Progress in this area has been sufficient for design and initiation of a sound course of study that should yield substantial progress in the near future.

DRAINAGE-AREA CHARACTERISTICS

It has been noted that the transport of fine sediment in a stream is largely controlled by factors of supply in its drainage basin. Some of these factors include:

1. Nature, amount, and intensity of precipitation.
2. Condition and density of the channel system.
3. Orientation, degree, and length of slope of the watershed area.
4. Soil type.
5. Land use.

These factors can be allied logically to either or both of two forces: those that resist or those that advance the rate of erosion and transport. For example, precipitation of low intensity occurring at ideal intervals may advance the growth of vegetation and thereby advance the resisting force. If precipitation is very intense after a drought, or occurs over an area without cover, it is likely to cause a large amount of erosion. Because of the interrelationship of these factors, and the large variance associated with each, it is difficult to define erosion and transport from a small area.

Many workers in the field of erosion and soil loss have correlated considerable data, but they find large areas within the factors that cannot be defined by present data. In one recent investigation, Wischmeier and others (1958) showed that an index consisting of the product of rainfall energy and maximum 30-minute intensity of the storm is the outstanding variable for correlation with soil-loss data from 5 locations in the Midwest each having 131 to 207 observations.

One of the most perplexing problems in handling soil-loss data is that of accounting for deposition within the drainage basin. The sediment yield of a large basin is usually less than the sum of its subbasins because of deposition and channel aggradation during transport from the small areas. The factor "condition and density of the channel system" is probably the most important index of this phenomenon. Data assembled by Glymph (1951) shows the relation of net sediment yield to size of drainage area. For a drainage area of 5 square miles, the yield ranges from 400 to 4,000 tons per square mile, for 500 square miles, 100 to 2,000 tons, and for 50,000 square miles, 60 to 1,200 tons.

Because of the complicated interrelations among the environmental factors affecting sediment yield, and because they have not been studied sufficiently, a detailed discussion of each factor will not be attempted in this treatise. Some of these factors have been considered indirectly in an analysis of the trend of sediment yield of the Brandywine Creek at Wilmington, Del. (Guy, 1957). In this study the parameters of total runoff, rainfall intensity, and season of the year evaluate to some degree the factors considered in this section.

GENERAL CONSIDERATIONS OF SEDIMENT TRANSPORT

Streamflow turbulence results in the continuous motion of fluid vertically across a horizontal plane within the stream and transports not only fluid and momentum but also suspended sediment. This condition alone would tend to produce a uniform distribution of sediment in a given vertical section of streamflow; however, the constant settling of the particles due to gravitational force counteracts this process. Under equilibrium conditions, the upward components of turbulence travel toward a region of lower concentration, and the downward components toward a region of higher concentration. Thus, the rate of increase in suspended-sediment concentration toward the bottom depends on the degree of turbulence in the stream and the settling velocity of the particles.

Sediment of extremely small size (colloidal) may be suspended by Brownian movement or electrolytic charges. Particles of this kind are, therefore, suspended independently of turbulence and theoretically are transported by a velocity equivalent to the suspending fluid.

It is logical that all particles in a colloidal suspension will maintain uniform vertical as well as horizontal dispersion through a reach of stream, if they are dispersed uniformly at the entrance of the reach. Observations show further that natural turbulence in most streams is sufficient to suspend clay and silt (diameters less than 0.062 mm) uniformly for all practical purposes. Sand, however, is suspended less easily and therefore its concentration in a vertical section of a stream must increase from top to bottom in accordance with the operation of the turbulence-suspension theory. Figure 2 (U.S. Inter-Agency com-

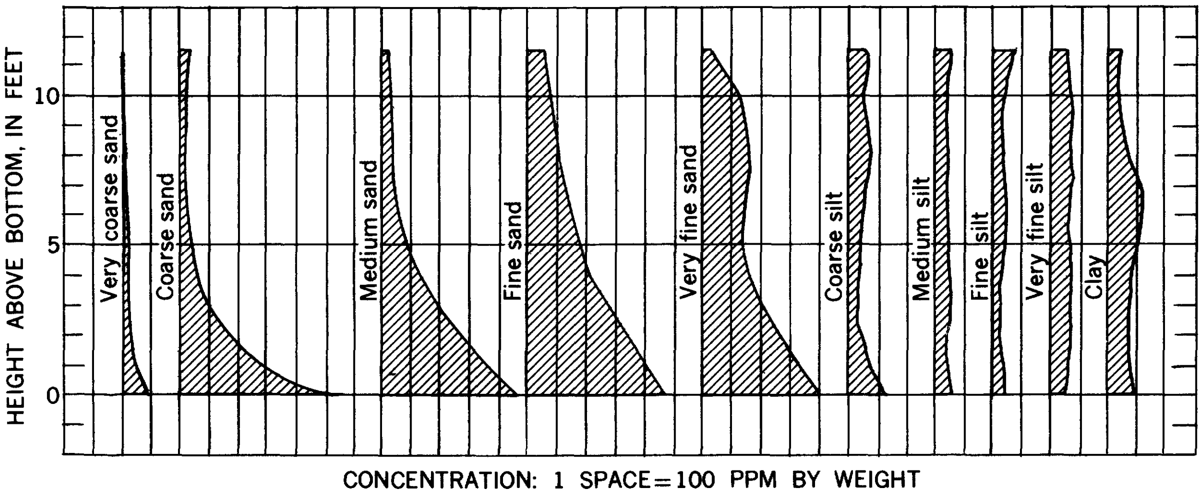


FIGURE 2.—Distribution of sediment at a vertical in the Missouri River at Kansas City, Mo. From U.S. Inter-Agency Committee (1948).

mittee, 1948) illustrates the vertical concentration of several size classes from clay to very coarse sand, thus contrasting the nearly uniform concentration from top to bottom of the clay and silt with the increasing concentration of sand from top to bottom.

The concentration distribution of sand in suspension by a stream then is a function of the size available for transport, the roughness of the stream bed, and the velocity characteristics of the stream. Hubbell and others (1956) show that the suspended-sediment concentration at different verticals in the cross section of a sand-carrying stream may vary widely and that sand transport is associated with hydraulic characteristics of the stream that fluctuate widely in both time and space for a given stream.

Streams having a relatively uniform rate of water discharge are expected to transport sediment at a relatively uniform rate, particularly the coarser fractions that depend on the transport capacity of the stream. Streams having a widely varying rate of water discharge are expected to exhibit very large variations in sediment discharge because in addition to the increased transport capacity with increased water discharge, rainfall causes a change in supply of fine sediment. This contrast is largely a function of climate and its related factors such as vegetation. Table 3 contrasts the monthly water and sediment discharge of the Scantic River at Broad Brook, Conn., with the Pecos River near Artesia, N. Mex., for the 1954 water year.

TABLE 3.—*Comparative monthly water and sediment discharges for streams in areas with differing climate*

1954 water year	Discharge of Scantic River at Broad Brook, Conn. (area 98 square miles)		Discharge of Pecos River near Artesia, N. Mex. (area 15,300 square miles)	
	Water (cfs-days)	Sediment (tons)	Water (cfs-days)	Sediment (tons)
October	1,860	56	701	321
November	2,560	60	1,710	550
December	5,400	331	1,810	642
January	3,490	91	1,610	330
February	3,800	136	1,200	244
March	5,850	222	1,220	168
April	5,850	540	7,900	70,100
May	6,540	322	9,660	130,400
June	2,830	88	500	216
July	1,610	33	24	6
August	1,780	69	10,000	198,700
September	5,120	512	1,040	927
Total	46,700	2,460	37,400	402,500
Average per square mile	477	25.1	2.44	26.3

GEOMETRICAL CHARACTERISTICS OF STREAMS

Relation of channel geometry to sediment transport.—A natural stream in equilibrium condition has a geometrical pattern of slope and cross section to fit the water discharge and the size distribution and rate of transport of sediment. Leopold and Maddock (1953, p. 51) state that

> . . . even in the youthful topography of the headlands where streams are gradually downcutting and are, therefore, not graded, the stream-channel characteristics and slope tend to be adjusted to the discharge and sediment load. Even the ungraded parts of a channel system exhibit a neat pattern characterized by a tendency toward a regular increase of width and depth downstream with increasing discharge and with a concomitant decrease in slope.

and

> Load and discharge are characteristics of the hydrology, geology, and physiognomy of the drainage basin. They are furnished to the trunk stream channels and are essentially independent of the trunk channel system.

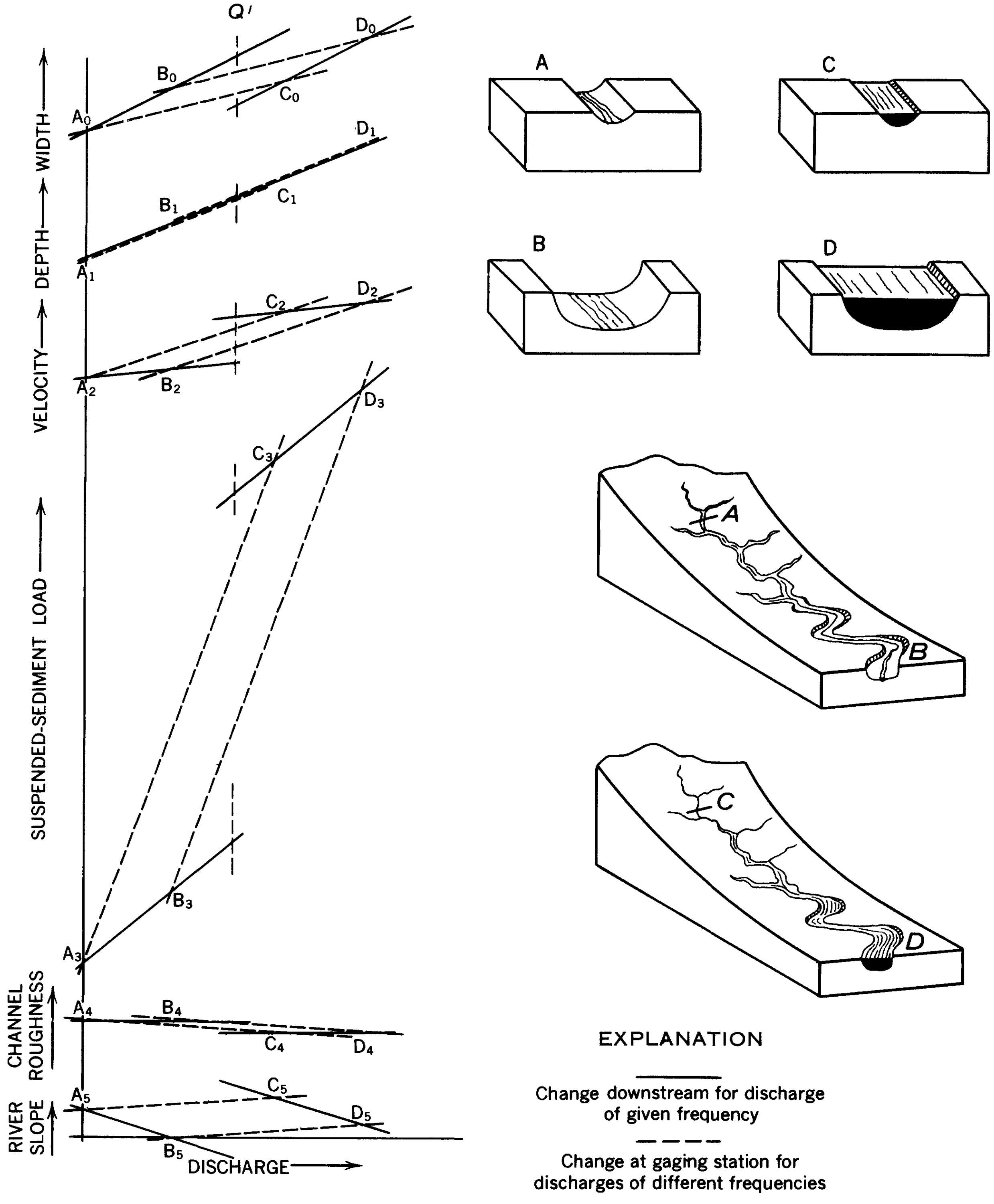
In somewhat more specific terms, figure 3, a reproduction of figure 19 from Leopold and Maddock (1953, p. 27) shows how slope, roughness, sediment load, velocity, depth, and width vary with discharge at a station and downstream. Sections A and C represent headwater conditions of low and high flow, respectively. In considering the differences at the headwater section with increase of flow, note the small increase in slope, the small decrease in roughness, the *large* increase of sediment load, the moderate increase of velocity, depth, and width. With respect to differences in sections A and B, where B is downstream and at the same flow frequency as A, river slope decreases moderately, roughness decreases slightly, sediment load increases moderately—probably because the water discharge increases—, velocity increases slightly, and depth and width increase moderately. The sediment load has the greatest contrast when considering both the increase in flow and a downstream section.

Leopold and Maddock (1953) also state that stream channel geometry varies in response to sediment transport as follows:

> At constant discharge, an increased velocity at constant width is associated with an increase of both suspended load and bed load in transport. At constant velocity and discharge, an increase in width is associated with a decrease of suspended load and an increase in bed load in transport.

Boundary roughness.—Resistance to streamflow in a given reach of a stream is the sum of the roughness created by the size of particles on the stream bed, the size of ripples and (or) dunes made up of these particles on the bed, and the bank roughness. The relative importance of any one of these may depend on the particle size of sediment in transport, the stream velocity, the shape of the cross section and the sediment load in transport. For example, the important roughness factor in a wide shallow stream having a bed of medium sized sand and with a mean velocity of 4 feet per second is the dune-and-ripple roughness.

In addition to the particle roughness, a given stream may flow successively and(or) simultaneously in several



Note: All scales are logarithmic

FIGURE 3. Average hydraulic geometry of river channels expressed by relations of width, depth, velocity, suspended-sediment load, roughness, and slope to discharge at a station and downstream. From Leopold and Maddock (1953).

"regimes" of flow. This is most clearly demonstrated by gradually increasing the velocity in a laboratory flume having a deep bed of sand which has been planed smooth. The flow regimes, with increasing velocity, are ripple, dune, plane, and antidune. The sediment in transport increases in this order. With regard to roughness, the plane-bed regime has less resistance to flow than does the dune regime.

In a natural stream the shift from a dune to a plane regime results in an increase in velocity and, consequently, a decrease in gage height even though the quantity of flow has increased. The regime of stream flow affects the depth as well as the rate of bed movement. For the ripple regime, the depth of disturbance is not as severe as the dune and antidune regimes. Simons and Richardson (1959) found lenses of fines in the bed during the ripple regime which are not present for the other regimes when these fines are carried as suspended load. They also found that under the dune regime the fine material does not settle and coat the bed, but there is a constant exchange between the fine material in suspension and that in the bed. Thus, when "clear" water returned, and if the stream remained in the dune regime, the fine material would be removed from the bed.

The banks of a stream may have little effect on the total resistance to flow if the stream is wide and shallow. A deep narrow section, on the other hand, may receive a large part of the total resistance from the bank. The bank roughness may result from the shape and size of the particles of sediment in the deposits or residual material, from vegetation, or from man-made devices.

The roughness condition of the bank affects the nature and quantity of deposition along the bank during the recession of any storm runoff. Sand-bed streams have been observed that had mud deposits too deep and soft to wade across after the recession of storm runoff (less than bankfull stage) containing a large concentration of fine suspended sediment. These deposits were at a straight reach of the channel where one would not expect bank "building" and, therefore, it is assumed that this deposit would be eroded away during the rising stage of the next runoff. This kind of deposit is, then, only of a semipermanent nature. The roughness of a braided stream would have an increase in bank roughness proportional to the percentage increase of banks encountered for a representative number of cross sections in a reach.

Alinement of channels.—Streams flowing in alluvium generally exhibit rather poor alinement—ranging from an occasional bend for some streams to a high degree of meandering for others. Such bends affect the secondary circulation and hence the location and depth of scour and deposition in the stream bed. The cross section of a river with a straight channel and a relatively flat bed is approximately trapezoidal in shape; whereas, a section at a bend is roughly triangular in shape with the deep part near the outside bank. Carey and Keller (1957) in a study of bends found that the deposit of sand on the inside bank of a bend was mostly that scoured from the outside bank immediately upstream. Thus the sand tends to travel across the thalweg in the reach of reversing curvature. The coarser sands are obviously deposited at the lower elevations and toward the head of the bar during such deposition. This phenomenon of cut and fill is most active at bankfull stage. At less than bankfull stage, velocities will be lower and incapable of depositing the sand at a reasonable height on the inside bend; and at more, the flow down the valley will tend to create currents not parallel to the bend of the stream channel and probably reduce the cut-and-fill velocity. Fine sediment from many sources may be deposited on the upper part of the inside bank during bankfull or higher stages. Occasional lenses of fines (silt and clay) in the fill bank (inside of bend) have been observed which logically are deposited by receding flood waters, then bound in place by rank vegetation before burial by sand during the next flood.

Experiments with hydraulic models at the Waterways Experiment Station (Lipscomb, 1952) show that the size of bends becomes smaller with either a decrease in flood discharges, the slope, or the angle of entrance to the bend; and, that the more erodible the banks, the wider and shallower will be the crossings to transport the greater load.

Both the straight or meandering types of stream on a sand bed have a relatively uniform gradient. However, if the stream channel contains considerable quantities of gravel and possibly rocks, the gradient is likely to be irregular and the channel will consist of alternate pools and riffles. This pool-and-riffle type of stream is logically susceptible to the collection of deposits of fine sediment during relatively long periods of low flow followed by rather sudden removal with increasing velocity during the early part of increased flow in the channel. The deposition of these fine sediments during low flow would be relatively high in concentration of radioactivity because of the small sediment transport of the stream under these conditions.

PARTICLE-SIZE GRADATION

Effect of chemical quality of water.—Concise theory on the nature of flocculation for suspended sediment in natural streamflow is lacking. General experience has been obtained by noting the difference between apparent and ultimate size gradations when a given sediment is analyzed in settling mediums of both native stream water and distilled water with a dispersant. As mentioned previously, streamflow at a given reach has various degrees of turbulence ranging from little at the bank and other "dead water" areas to compound secondary circulation at midchannel. The turbulent motion inhibits flocculation because it, in effect, tends to prevent the drifting of particles toward each other that would ordinarily occur in still water. Turbulence is also a factor in disrupting floccules, if formed in quiet water.

It may be stated, then, that flocculation of sediment probably varies as the chemical nature of the stream, the kind of clay, the concentration of clay, and inversely, with the turbulence of streamflow. Since clay particles are very irregular in shape, it is assumed that mechanical as well as chemical binding is involved. This would be most likely for the clay-to-clay floccule and less likely for the clay-to-silt or clay-to-sand floccule.

Effect on surface area.—Average-size gradation curves for both simultaneous suspended-sediment and bed-material samples are illustrated in figure 4 for three streams. These curves are considered indicative of the typical range of sizes of the particles of sediment transported by alluvial streams. Some streams may be encountered that are more unusual than those illustrated; for example, streams from the Sand Hills of Nebraska may contain suspended sediment of nearly the same particle size as that of the bed, and streams tending toward the pool-and-riffle condition will have a much larger range of bed-material sizes, particularly the coarser sizes.

As indicated in the section of this report "Factors determining extent of sorption", the capacity of sediment for sorption of radioactive waste is related to the surface area of the sediment. On this basis, the particle-size curves of three streams indicate increasing capacity to carry such waste in the order, Middle Loup River at Arcadia, Nebr., Loup River at Fullerton, Nebr., and Mississippi River at St. Louis, Mo. On the basis of these average size-distribution curves, the surface area (computed as loosely packed spheres) of the bed-material sediment is 60, 140, and 100 square-centimeters per cubic centimeter, respectively; and the suspended sediment is 350, 4,530, and 27,400 square centimeters per cubic centimeter, respectively. The sorptive capacity of the suspended sediment is, therefore, many times that of the bed material; and since most of the sediment is moved in the sampled or suspended zone, it is assumed that suspended-sediment transport is the more important consideration. It is further evident that the fines or sediments brought into the stream from sheet erosion are the most important as far as sorptive capacity is concerned. Hence, environmental factors contributing to erosion in the upland areas of the drainage basin may be more critical with respect to radioactive waste disposal than the transport characteristics of the stream channel.

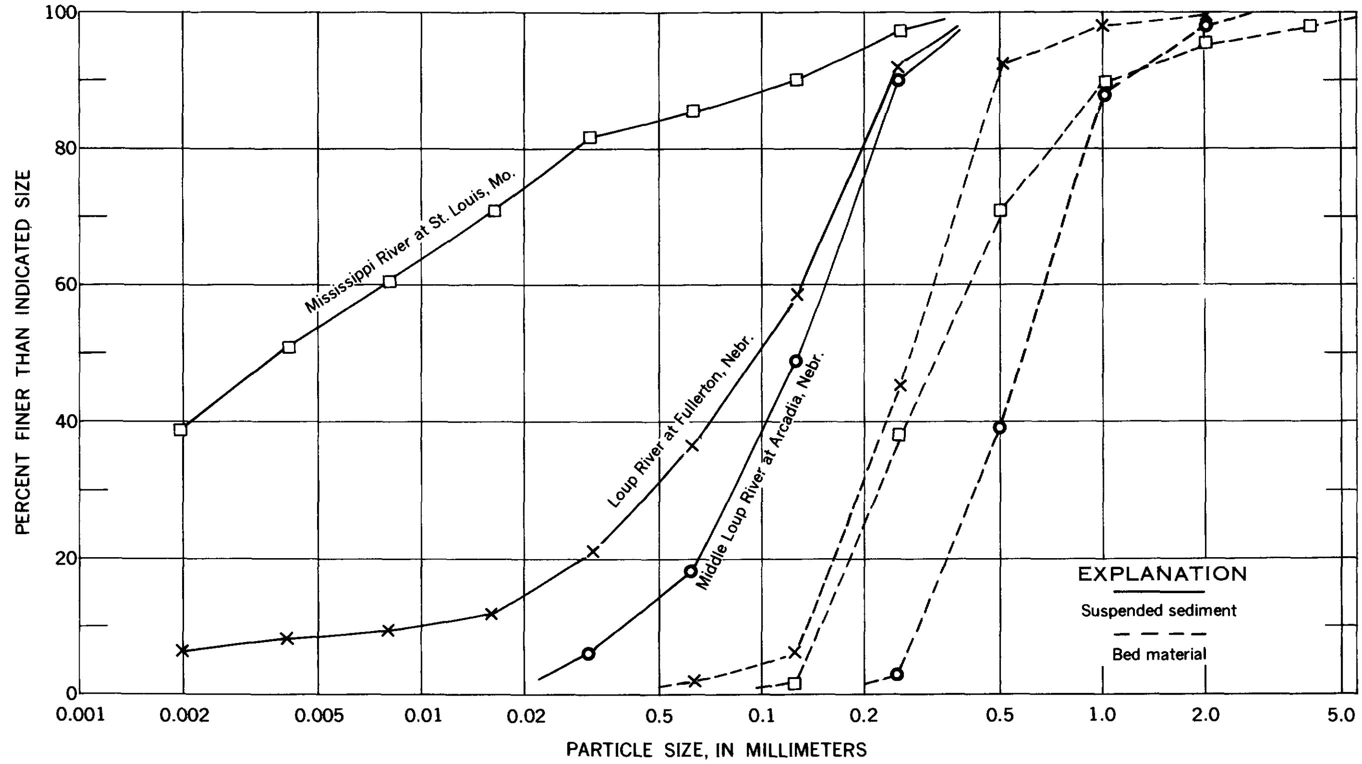


FIGURE 4.—Particle-size gradation of suspended sediment and bed material for three typical alluvial streams.

CHANNEL CONTROLS

Channel controls affect the suspension and deposition of sediment because of their control of the flow of water. A large deep reservoir holding water for weeks or months may trap 100 percent of the inflowing sediment and cause separate deposits of sands and fines, whereas a bank lining would change only the flow characteristics and thereby change the mode of sediment transport. The effectiveness of a given control in altering normal suspension and deposition depends not only on the control itself, but also on the character and concentration of the sediment in transport and on the chemical character of the water.

Trap efficiency.—Table 4 shows the estimated efficiency of several kinds of structures for trapping the clay, silt, and sand fractions of sediment. The capacity-inflow ratio for reservoirs, or the acre-feet of capacity per acre-feet of annual flow has been related to trap efficiency of normally ponded reservoirs by Brune (1953). The curve defined by data from more than 40 measurements indicates the following:

Capacity-inflow ratio	*Percent sediment trapped*
0.005	30
0.02	60
0.1	85
0.3	95
1.0	98

TABLE 4.—*Estimated sediment-trap efficiency, in percent, of various types of structures for channel and storage control*

Description and purpose	Clay	Silt	Sand
Large drainage areas 10,000 square miles or more:			
Reservoirs for power and storage (high capacity-inflow ratio)	80–100	100	100
Reservoirs for irrigation and water supply:			
High capacity-inflow ratio	70–100	100	100
Low capacity-inflow ratio	10–100	50–100	100
Flood-control reservoirs	0–50	0–80	20–100
Navigation control	0–50	0–80	20–100
Irrigation diversion	0	0	0–50
Bank or channel control	0	0	0
Soil Conservation Service structures on drainage of 10 square miles or less:			
Detention reservoir with draw-down tubes and sediment storage:			
High capacity-inflow ratio	10–70	50–100	100
Low capacity-inflow ratio	0–30	10–50	80–100
Drop spillways, chutes, and other channel-control structures	0	0	0

Brune also notes that the Tennessee Valley Authority has used the ratio of the period of retention to the mean velocity as an index to trap efficiency. This is similar to the capacity-inflow ratio because both imply a rate of water movement through the reservoir.

Reservoir deposition.—The ideal concept of reservoir deposition assumes that deposition begins at the upstream limit of backwater effect with the deposition of the largest particles in transport. Increasingly finer particles are deposited with progressive decrease in transport capacity of the flow until all material of the size of fine sand or larger is deposited. The silt and clay sizes are transported, in an undefined manner, beyond the sand deposit. Deposition of these fine particles may occur in an unknown sequence along the floor of the reservoir. Density flows may result in the transport of clay throughout the length of the reservoir. This idealistic pattern is complicated by the forward growth of the sand deposits over previously deposited silts, encroachment of one kind of deposit over another owing to fluctuations in pool level, reworking of deposits by waves and erosion, and three-dimensional flow phenomena, such as seasonal turnover of deep waters, owing to temperature gradient (thermocline).

The headwater deposit may be relatively simple as in a long narrow reservoir, or it may be extremely complicated owing to the presence of many side channels, as for the Mississippi River at the Gulf of Mexico.

Figure 5 (Witzigman, 1955) illustrates the relation of suspended-sediment concentration and particle size to cross-sectional area, depth, and mean velocity for the backwater reach of Garrison Reservoir—the narrow or confined type—July 7 to 8, 1954. The concentration decreases with increasing cross-sectional area and depth and decreasing velocity downstream.

Density currents, which are capable of moving fine sediment throughout a reservoir, are caused by differences in temperature, salinity, and more usually, sediment content. The relatively muddy stream entering a clear reservoir collides and partly mixes with the "still" water and then moves along the bottom propelled by the slope of the bed of the impounding reservoir and the pressure of additional muddy water on the delta deposits. Deposits from "dead" density currents will consist of fine clays and may occur in the deepest water near the dam.

Degradation downstream from control.—The Corps of Engineers (1959) in a study of sediment problems in the Arkansas River, notes the following factors for the occurrence of degradation downstream from a control:

1. The stream must have carried a relatively large bed-material load prior to completion of the control, as this is a measure of its ability to transport the existing bed material.
2. The control must provide sufficient detention time of water in the reservoir to trap most of the bedload.
3. If most of the load was transported by large discharges prior to construction, then these large discharges must prevail to some degree after construction.
4. Material in the bed must be of a size ordinarily carried as bedload with the new flow regime.

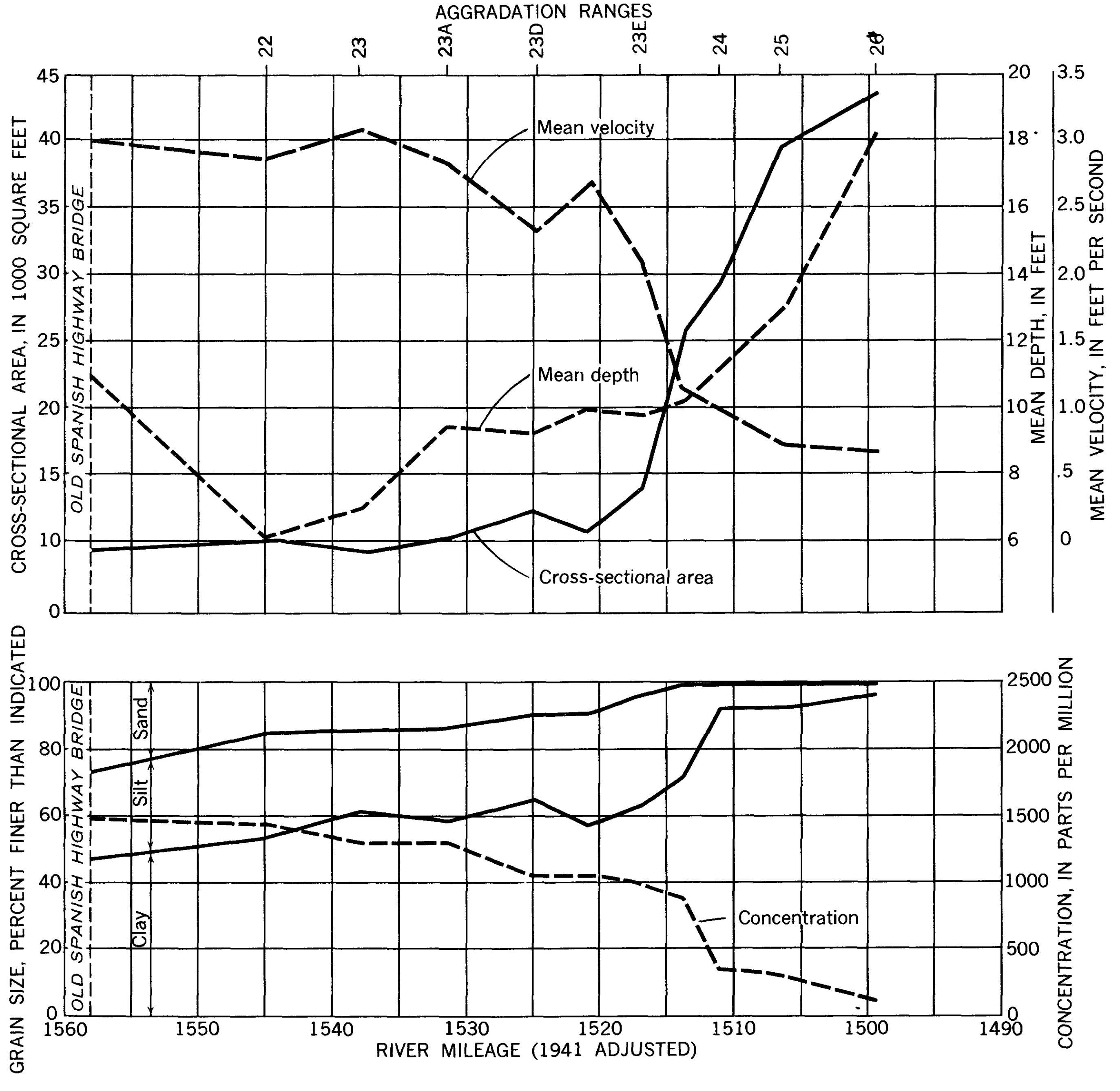


FIGURE 5.—Relation of sedimentation to channel characteristics through backwater reach of Garrison Reservoir, N. Dak., July 7-8, 1954, from Witzigman (1955).

CRITERIA FOR IDENTIFYING POTENTIALLY HAZARDOUS STREAMS

In general the criteria for identifying streams which are potentially hazardous with respect to the deposition of radioactive sediments are not simple and clearcut. More often than not, a hazardous situation will involve the combined action of several factors. Taken individually, none of the factors might cause objectionable conditions; taken collectively, however, the addition or subtraction of a single factor might radically alter the entire picture.

The development of a hazardous situation obviously implies the requirements that there be (1) uptake and concentration of significant quantities of radioactivity by stream sediments, (2) stream geometry and sediment conditions conducive to extensive local deposits of contaminated sediments, and (3) a rate of buildup of radioactivity in deposits which exceeds the rate of natural radioactive decay. In this section the hazard criteria are identified and briefly discussed in terms of these three requirements.

UPTAKE BY SEDIMENTS

With respect to the sediment itself, the total available surface area of the suspended-sediment particles is the most obvious index of hazard potential. Concentration and size distribution are fairly reliable indicators of surface area.

Fortunately the greater sorption capacity of the fine sediments is to some extent offset by the tendency of colloidal materials to remain in suspension except when flocculation occurs. Heavy concentrations of contaminated fine sediments might occur in density currents at the bottom of reservoirs and pools, but there the chances for dangerous exposure to humans would be small.

The chemical and mineralogical nature of the sediments, particularly the fines, are also a factor to be considered in assessing hazard potential. Sediments characterized by high ion-change capacities and high zeta potentials would bear watching. The nature and amount of sorbed nonradioactive components should be considered as an indication of residual sorption capacity for radioactive components.

Concentration of radioactivity in the waste-stream water solution is obviously related to hazard potential. The chemical composition of radioactive components would be important in estimating the extent to which sorption might occur.

The total ionic concentration in solution is likewise an important factor. Normally a low-ionic concentration of nonradioactive substances would mean correspondingly increased sorption of radioactive substances.

The reversibility of sorption reactions should also be considered. Irreversible sorption reactions would in most cases be considered as potentially more hazardous than reversible reactions.

The presence of significant quantities of a flocculating agent in streams containing radiocontaminated clays could be a source of danger.

The degree of dispersion of radioactivity in the stream cross section should be evaluated. For example, concentration of the radioactive wastes in a density current flowing along the bottom of a stream would probably lead to an increased degree of uptake by the more stable bed sediments. Generally this would be considered undesirable.

CONCENTRATION AND LOAD OF SUSPENDED SEDIMENT

The concentration and load of suspended sediment in streams can be considered one of the important criteria for identifying potentially hazardous streams because of the effect on amount of available surface area for sorption. Further, it is important as a factor in the nature and extent of deposition and erosion within the stream system.

The nature of the concentration and (or) load variation with respect to time may be important because it is expected to indicate the capacity or lack of capacity of the stream to transport a given input of waste. For example, a stream with a relatively uniform water discharge and high concentration of fine sediment would probably maintain the waste in suspension on, or with, the fine sediment. On the other hand, a stream with a large range of water discharge and sediment load recieving the same concentration of waste would be expected to carry the waste in "slugs", because the waste not having an abundance of fine suspended sediment for sorption during low flow periods, would probably be sorbed on the particles in the stream bed. Then, during the relatively short periods of storm runoff with high concentrations of sediment, the waste held on the bed may be exchanged to "free" suspended particles and (or) moved downstream considerable distances as bedload.

Discharge-weighted mean concentration of sediment is being mapped for the United States by Rainwater (1959 personal communication) as a part of a Geological Survey project on composition of rivers, and can be used as one means of classifying potentially hazardous streams. The variation of sediment concentration and load with respect to time, however, has not been defined, nor have criteria been established or sufficient data made available for this kind of definition. This can be approximated, however, on the basis of climate, size of drainage basin, and control of the stream flow. The magnitude of time variation in concentration and load would be expected to be greater, for example, on most Kansas streams than on South Carolina streams, and on the uncontrolled upper Colorado than on the controlled lower Missouri River.

CONDITIONS CONDUCIVE TO SEDIMENT DEPOSITION

Conditions conducive to the formation of sediment deposits containing large amounts of radioactive waste are more likely to be found on the inside of a bend on a stream near the source of waste disposal rather than on the banks of a straight stream channel, on a stream exhibiting a considerable range of sediment concentration and water discharge rather than a stream of nearly uniform water discharge and sediment concentration, and on a stream with man-made channel controls rather than on an uncontrolled stream.

Sediment of the coarser fractions, usually deposited on the point bar on the inside of a stream bend, is derived mostly from that being cut from the outside bank of the opposite bend immediately upstream. The finer fractions, usually deposited only at the upper elevations of the bar, or during the recession of the storm runoff event, may be derived from many sources including bank cutting in the immediate channel, or gully and sheet erosion in the upper part of the drainage basin. The point bar at the first bend immediately downstream from a source of waste may then be built of a large mass of contaminated sand. In this discussion it is assumed that the waste is sufficiently mixed with stream flow so that contact with the sediment in transport is relatively uniform.

Streams of widely varying discharge are conducive to bank and flood plain deposits. The most rapid deposition on the inside bank of the meandering channel occurs during the high rates of flow; especially so, for the fines at the top of this deposition bank. Flood plain deposits obviously cannot occur without the generally unusual high rates of flow. Deposits of fines along stream banks other than point bars usually occur during the recession of a storm event for which relatively large concentrations of fines are carried. These may be of only minor significance because they are transported and deposited when stream flow and concentration are considerably greater than average and, therefore, the concentration of waste in the deposit is likely to be low. This again assumes a uniform rate of waste disposal.

Deposits of contaminated sediment may accumulate within the controlled channel when flow and sediment concentration are low for long periods of time; and thus, with a constant rate of waste disposal, the concentration of radioactivity in the water and on the sediment would be high. Similar to channel control, the most critical deposits on the pool-and-riffle type of stream are likely to occur during the long periods of low flow when the fines are deposited in the slack water resulting in an intensive concentration of radionuclides in the pool. With this type of stream, the deposit is usually removed during the rising stage of the next period of storm runoff.

In summary then, the sedimentation criteria for identifying potentially hazardous streams may include:

1. The sediment concentration and water discharge or load. High concentrations of sediment may cause many kinds of deposits but may result in relatively low concentration of radionuclides. Low concentration of sediment may result in some small accumulations of sediment deposits of high waste content.
2. The time variation in sediment concentration and load. The waste, under uniform release, is likely to build up during low flow and then move as a slug for possible deposit on high banks or flood plains during high flow.
3. The nature of channel geometry. A meandering channel is characterized by building of point bars on the inside bends. The pool-and-riffle type of stream accumulates the waste in pool reaches during long periods of low flow. A channel maintaining flow almost continuously in the dune regime moves fine sediment in and out of the bed almost uniformly in time, whereas a channel flowing in the ripple regime most of the time may accumulate fines in the bed as lenses and upon occasional shifting to the dune regime will move these fines from the bed.
4. The nature and extent of water and channel controls. Reservoirs, for example, may trap up to 100 percent of the sediment, whereas other controls may serve only to concentrate the waste only during the low flow.

RATE OF BUILDUP OF RADIOACTIVITY

In order to assess the hazard potential arising from the buildup of radioactivity in sediment deposits, both the decay rates of the radioactive components and the rate of sediment deposit at locations like those discussed in the preceding section would have to be investigated. Also, the frequency at which sediment deposits are flushed out and their subsequent movement and deposition would need to be considered. A combination of conditions involving an intermediate or slow radioactive decay rate, a high deposition rate of contaminated sediments, and infrequent flushing of deposits would usually be the most hazardous.

CLASSIFICATION OF STREAMS IN THE UNITED STATES ACCORDING TO HAZARD POTENTIAL

The hazard potential of radioactive waste in streams may be considered in view of differences in stream flow, in channel geometry, and in the concentration and character of sediment as they may affect deposition of sediment and consequent waste. However, streams are very complex with respect to the magnitude and variation of these features; and, therefore, classification according to hazard potential is difficult and indeed almost remote in view of limitations concerning basic knowledge of the relation of these features.

A review of the sedimentation criteria for identifying potentially hazardous streams for radioactive-waste disposal discussed in the previous section indicates many conflicting aspects for consideration. For example, a stream with relatively large water and sediment discharges may be conducive to extensive deposition and erosion within the channel; but, because of the high rates of water and sediment movement, the concentration of radioactive waste may be very low. Further, a meandering channel results in a nearly continuous building of a deposit as a point bar, but most of this building occurs during high rates of water discharge when the concentration of waste in the water is low.

The sand-bed stream of good alinement and carrying a relatively uniform water discharge and concentration of fine sediment would be the least hazardous kind of stream to transport a given uniform amount of radioactive waste without accumulating extensive and (or) concentrated deposits. This would assume the flow in the channel to be in the dune, plane-bed, or antidune regimes so deposits of the fine sediment would not accumulate in the bed; and, if minor bank deposition occurred, it would be when the flow in the channel and concentration of sediment are relatively high. Such a stream would have relatively minor and uniformly concentrated accumulations of waste in relation to both time and space in the bed and along the banks.

One of the most hazardous conditions may occur either on the pool-and-riffle type of stream or a large river controlled with low dams for navigation purposes. For these situations, the pool or pools immediately downstream from the disposal outlet may accumulate extensive and highly contaminated deposits in the bottom of the pools. This results from the relatively low water discharge and very low concentration of sediment, and, consequently, a very high concentration of waste, accumulating for long periods of low water flows. When a relatively large increase in flow occurs after weeks or months of accumulation, most of the contaminated deposit will move downstream and thus become available for bed, bank, or flood-plain deposition.

The range of conditions encountered with regard to (1) the quantity of sediment and water discharge, (2) the variation of sediment and water discharge with respect to time, (3) the mineralogic composition of the sediment, (4) the channel geometry, and (5) the nature and extent of the water and channel controls makes it apparent that each site of radioactive-waste disposal is unique with respect to the effect of sediment in causing hazardous deposits of waste. Most of these criteria vary considerably in a given stream. With regard to channel geometry, for example, Leopold and Wolman (1957) state that straight channels cannot be readily differentiated from meandering and braided patterns by definitive combinations of hydraulic variables, yet the regular spacing and alternation of shallows and deeps is characteristic of all patterns.

Figure 6 illustrates the general magnitude of water-discharge-weighted suspended-sediment concentration for streams east of the Mississippi River as one of the factors in the criteria that may be useful for classification. This map is being extended to include the remainder of the United States, at the time of this writing (1959).

Another factor for which some data are available concerns the particle size or the relative sorptive capacity of the sediment in transport for the different streams. This factor is illustrated in figure 7 where the ordinate is a measure of the difference in bed and suspended-sediment sizes and plotted as a ratio of the median (50 percent) bed size to the median suspended-sediment size; where the abscissa is a measure of sediment fineness and is plotted as the product of the sizes for which both the suspended-sediment and bed sizes contain 15 percent by weight finer. The graph indicates an increasing potential for hazardous deposits in the direction of increasing difference of bed and suspended-sediment size and in the direction of decreasing measure of coarseness.

Many dimensional and dimensionless diagrams may be prepared by different measures resulting from the data on the particle size of suspended and bed sediments. These may include surface area of the particles, the standard deviation of the size gradation—a measure of the range in sizes—and combinations of these with the concentration and load in transport. A logical, but untried, plot would relate the surface area for a unit concentration in suspension to the median size of suspended sediment with the standard deviation of size as the third variable.

CONCLUSIONS

Many of the mechanisms involved in the dispersion, transportation, and concentration of radioactive wastes in natural streams by sediments are complex and not well understood. In this section, the various subject areas of this report are evaluated according to present knowledge. Areas where knowledge is particularly deficient are indicated.

Characteristics of waste materials.—The properties of low-level liquid wastes, which are of interest with regard to potential hazards arising from the contamination of stream sediments, may be classed as (1) nuclear, (2) chemical, and (3) physical characteristics.

Nuclear characteristics of the radionuclides contained in the wastes—such as type and energy level of radiation, and radioactive decay rate—coupled with the biochemical characteristics determine the potential health hazard resulting from a given amount of radioactivity.

The chemical composition of the radioactive waste solution is an instrumental factor in determining the extent to which sorption of radioactive components

FIGURE 6.—Concentration of suspended sediment in streams east of Mississippi River. From written communication, F. H. Rainwater, 1959.

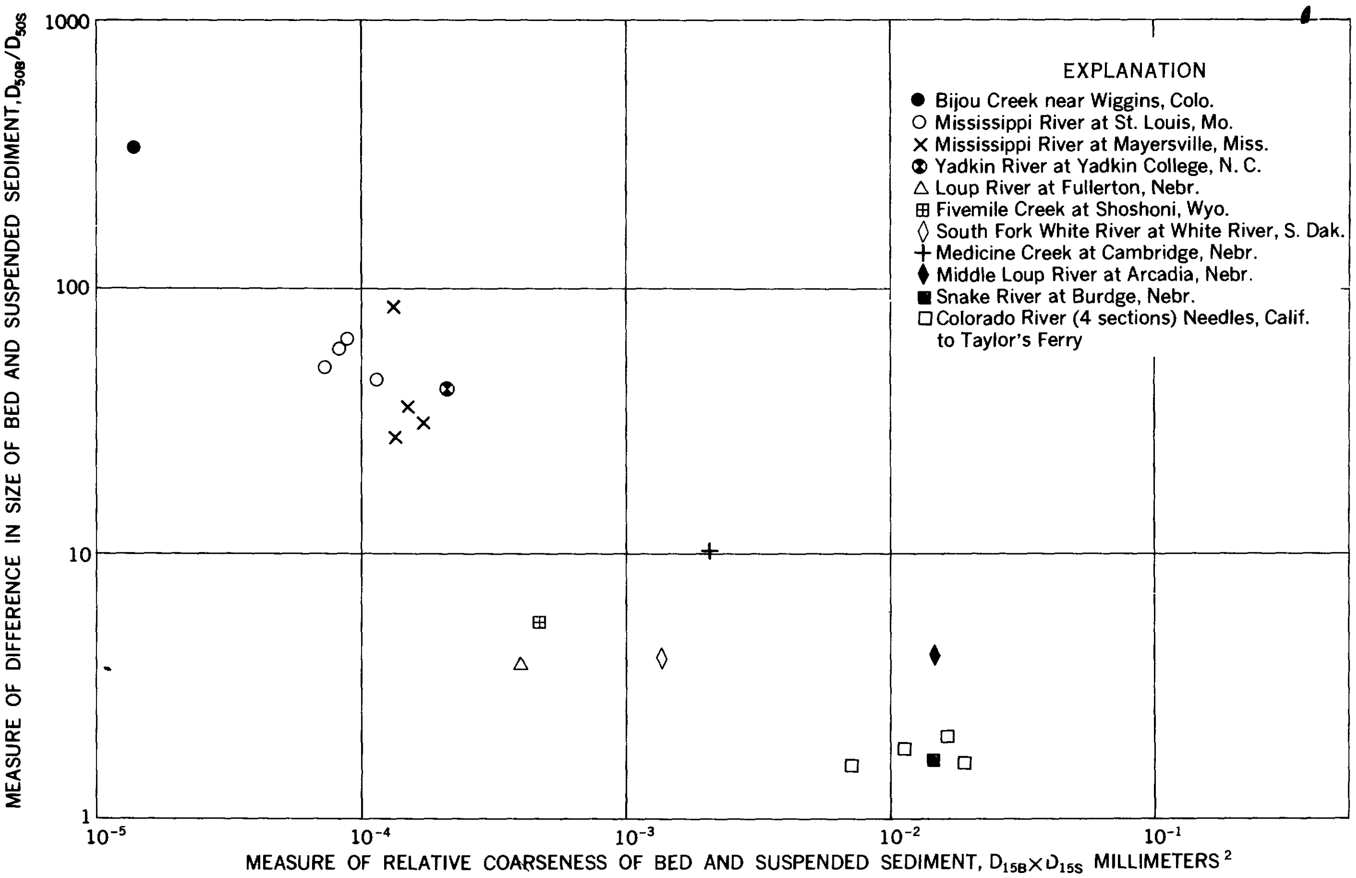


FIGURE 7.—A measure of hazard potential for stream deposition of sediment containing radioactive wastes.

by sediments occurs. Total ionic concentrations, pH values, the presence of organic compounds such as detergents, and reacting dissolved substances are all important in this respect.

Physical characteristics of the waste solution such as viscosity, density, and temperature are important in determining how effectively the radioactivity will be dispersed through the stream section by the process of turbulent diffusion.

There is considerable information in the literature on the nuclear characteristics of low-level radioactive waste solutions, but considerably less information on the chemical composition and physical characteristics. Data on the last two aspects should be systematically assembled and analyzed, and the results should be published.

Turbulent diffusion.—The literature on turbulent diffusion of submerged jets and diffusion by boundary-generated turbulence is very extensive. Nearly all this literature, however, considers an infinite or semi-infinite domain rather than a confined channel. The applicability of the solution for these domains to confined channel problems, whether by statistical or Fickian theory is very much restricted. Literature applying the concept of sediment and aquatic biota as sources and sinks for radioactivity discharged in an alluvial channel is sparse, even when the relatively well-advanced status of the Fickian approach to analysis of turbulent diffusion is considered. Well-planned theoretical investigations and field and laboratory experiments are needed.

Sorption.—Available evidence indicates that sorption of waste materials from solution by stream sediments is the rule rather than the exception. Experiments have demonstrated that sorption of radioactive components from dilute solutions may exceed 90 percent, and that the concentration of radioactivity on the surface of sediment particles may become many thousands of times as great as the concentration in the surrounding liquid.

Ion exchange is apparently the predominant mechanism involved in the sorption of radiocontaminants by stream sediments, although physical sorption may also play an important role particularly where the coarser sediments are concerned. However, owing to the inverse relationship of both specific surface and surface chemical activity to particle size, sorption capacity per unit weight of sediment tends to increase as particle size decreases.

Much literature on sorption phenomena has originated from physical chemistry and allied fields. Ion exchange, in particular, has been subjected to considerable study. Other types of sorption such as physical sorption and, in particular, sorption by diffusion into a solid are less well understood. Experimental techniques are reasonably straightforward and quite well established. In any case a fairly adequate frame of reference for the continued study of sorption phenomena has been developed, at least for the relatively simple systems which involve only a single solvent, solute, and sorbent.

There is, however, a considerable deficiency of knowledge concerning the mechanics of sorption in more complex systems. The effects of pH, total ionic concentration and detergents on sorption reactions require considerable further investigation as does sorption from mixed solutions. The sorption capacities of different types of sediments under various sets of conditions should be studied. The reversibility of sorption reactions is another area requiring attention.

Bed sediments probably sorb less waste than do suspended sediments where the solutes are for the most part uniformly distributed throughout the flow cross section. It appears, however, that little work has been done on this aspect of the sorption problem. It would be advisable to investigate sorption by bed sediments.

The study of sorption phenomena in natural streams will require the collection and compilation of a great deal of additional basic data from streams throughout the nation on the physical and mineralogical properties of sediments, and the presence of competing solutes in the water.

Sediment transport.—Identification of potentially hazardous streams on the basis of fluvial sediment transport is a new field of endeavor. The broad subject of evaluating the drainage area or environmental factors, as they affect the quantity and character of sediment supplied to the streams, has been investigated in only small and usually unrelated segments. The subject of channel and flow characteristics of streams, as they affect the rate of transport and the nature and extent of deposition, is more advanced but still contains many undetermined elements.

The environmental and channel problems cannot be considered separately becuase the accumulation of sorbed radionuclides in the stream bed and in other kinds of deposits will depend on the movement of individual sediment particles, whether they be in suspension or moved intermittently. The nature of particle movement will depend on the flow characteristics of the channel as they affect suspension or other movement. In consideration of gross transport and deposition, the depth to which ripple or dune movement takes place, the interchange and circulation of fine sediment with coarse, and the nature of semipermanent deposits with respect to the frequency and magnitude of runoff events are primary.

GLOSSARY OF TERMS

Absorption	the process of taking up within, implying nearly uniform penetration.
Adsorption	the process of taking up on the surface.
Aggradation	the general raising of the bed of a stream in a given reach.
Anti-dunes	symmetrical sand and water waves which are in phase and moving slowly upstream in groups of two or more, becoming gradually steeper on their downstream face until they break.
Bed load	that part of the solids load of the stream which is moving in almost continuous contact with the streambed, being rolled or pushed along by the force of the moving water.
Clay	crystalline fragments of minerals, essentially hydrous aluminum or magnesium silicates less than about 0.004 millimeters in size.
Concentration	dry weight of material per unit volume or unit weight of solution. Concentration of sediment is often expressed in parts per million (ppm) by weight; of solutes, in parts or equivalents per million; of radioactivity in microcuries per milliliter (μc/ml).
Curie	the quantity of a radioactive substance which undergoes 3.70×10^{10} disintegrations per second.
Degradation	the general lowering of the bed of a stream in a given reach.
Density current	a relatively dense mass of water moving slowly through a body of standing water by virtue of a difference in temperature, concentration of solutes, or concentration of fine sediment.
Desorption	the release of sorbed matter to solution.
Dune	a sand wave of approximately triangular cross section (in a vertical plane in the direction of flow) with a gentle upstream slope and steep downstream slope which travels downstream by movements of the sand up the upstream slope and the deposition of it on the downstream slope.
Exchange capacity	the amount of ions which are chemically sorbed on solids in such a fashion that they can be replaced by other ions. An indication of chemisorption capacity. Exchange capacity is usually expressed in milliequivalents (meq) per 100 g of material at neutrality (pH 7).
Flood plain	the portion of a river valley covered by water in time of flood and built of sediments carried by the stream.

Gravel	inorganic granular material in the size range of 2.00 to 64.0 millimeters.
Half life	one of the fundamental characteristics used to identify a particular radioactive species, the half life of a radioisotope is the time required for one-half the atoms to decay.
Ion exchange	the process by which exchangeable sorbed ions are displaced and subsequently replaced by other ions of like sign.
Sorption isotherm	a curve representing the relationship between sorption from and concentration of solute in solution, for conditions of constant temperature.
Isotope	any of two or more nuclear species having the same atomic number, therefore representing the same element, but having different mass numbers, hence different atomic weights.
Isotropic diffusion	the diffusion process is isotropic in character. Isotropic implies that the character of the diffusion is independent of coordinate direction at any given specified point.
MPC	maximum permissible concentration of radioactivity often expressed as microcuries per milliliter (μc/ml) or micromicrocuries per liter (μμc/l).
Nuclide	any nuclear species characterized by the number of neutrons and protons of which it is composed, and therefore characterized by its atomic number (the number of protons), and its mass number (the number of protons and neutrons).
pH	the negative logarithm of the hydrogen ion concentration in gram atoms per liter. A pH value of 7 indicates neutrality, numbers less than 7 increasing acidity, and numbers greater than 7, increasing alkalinity.
Radioactive decay	a probability process; the rate of decay is proportional to the number of radioactive atoms present at any time.
Radioisotope	a radioactive isotope.
Radionuclide	a radioactive nuclide.
Riffle	a short and shallow reach of stream channel with a slope considerably greater than the stream valley, the section containing rock or a deposit of the more coarse material moved by the stream.
Ripple	small ridges and/or crests, and troughs similar to dunes in shape, but smaller in magnitude, which have rather small width normal to the direction of flow.
Roentgen	a unit of radiation defined as the quantity of X- or gamma-radiation such that the associated corpuscular emission per 0.001293 gm. of air (=1 cc STP) produces, in air, ions carrying 1 electrostatic unit of quantity of electricity of either sign. A roentgen is equivalent to 1.61 x 10^{12} ion pairs per gram of air or the absorption of 83.8 ergs of energy per gram of air. The abbreviation for milliroentgen (10^{-3} roentgen) is mr.
Sand	inorganic granular material in the size range of 0.062 to 2.00 millimeters.
Sediment	fragmental material that originates from weathering of rocks and is transported by, suspended in, or deposited by water.
Sediment concentration	the ratio of dry weight of sediment to total weight of the water-sediment mixture, usually expressed in parts per million (ppm).
Silt	inorganic granular material in the size range of 0.004 to 0.062 millimeters.
Solute	a dissolved substance.
Solvent	a substance capable of, or used in, dissolving something.
Sorbent	a substance that sorbs.
Sorption	the process of taking up either by absorption (within) or adsorption (surface.)
Specific surface	an index of the extent of surface of a disperse system which is expressed as the surface area per unit weight or per unit volume of the dispersed phase.
Suspended sediment or suspended load	sediment moving in suspension in water as a result of turbulent currents or by colloidal suspension.
Trap efficiency	percentage of sediment trapped by a channel or water control structure.
Turbulent diffusion	a term used herein which is also sometimes called turbulent exchange, eddy diffusion or dispersion. The transfer of matter by means of the eddy motion of the fluid in a turbulent flow field.
Wash load	that part of the sediment load of a stream, usually silt and clay, which is composed of particle sizes smaller than those found in appreciable quantities in the shifting portions of the stream bed.
Zeta potential	the magnitude of the electric charge of soil particles in a colloidal suspension, usually expressed in millivolts (mv). More explicitly, the zeta potential is defined as the work required to bring a positive charge from the potential of a free solution to the point where the charge will migrate with the colloid when subjected to an electropotential gradient.

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