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The Spread of a Dye Stream in an Isotropic Granular Medium

GEOLOGICAL SURVEY PROFESSIONAL PAPER 411-G

*Prepared in cooperation with the
U.S. Atomic Energy Commission,
Division of Isotope Development and
Division of Reactor Development*



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By AKIO OGATA

FLUID MOVEMENT IN EARTH MATERIALS

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UNITED STATES GOVERNMENT PRINTING OFFICE, WASHINGTON : 1964

UNITED STATES DEPARTMENT OF THE INTERIOR

STEWART L. UDALL, *Secretary*

GEOLOGICAL SURVEY

Thomas B. Nolan, *Director*

The U.S. Geological Survey Library catalog cards for this publication appear after page G11.

For sale by the Superintendent of Documents, U.S. Government Printing Office
Washington, D.C. 20402 - Price 20 cents (paper cover)

PREFACE

The ground-water hydrologist seeks to identify and use the physical parameters controlling ground-water flow. Scientists are seldom required to define a more heterogeneous or complex region than that involved in an underground flow system. In this respect, the hydrologist's problem is different from that of, say, the electrical engineer or the physicist, who normally analyzes regions constructed with homogeneous characteristics and definable geometries. The heterogeneity of the flow region involved in ground-water studies is so great that it is difficult to find physical characteristics adequate to describe the region. In effect, then, the region must be described by more complex parameters than are generally used in other scientific fields.

Two such parameters are particularly significant. The first is the factor of heterogeneity, which somehow must be identified. But the nature of the heterogeneous region can hardly be described through reference to the individual geometric discontinuities. Such a description would require an endless compendium of individual descriptions, a device so obviously impractical that it renders the region not amenable to description by measurement of any of the characteristics visible or accessible from the surface of the region. The second factor embraces the flow relationships at any given point and these relations often reflect an angular discrepancy between potential gradient and direction of flow. Thus tensor description is required because any scalar factor is inadequate.

The measurement of the two factors—heterogeneity and flow relations—has been one of the principal difficulties in accomplishing an engineering analysis of ground-water flow. Laboratory experiments and study have illuminated some of the basic principles involved in the measurement of these factors. The effects of heterogeneity have been studied by hydraulic experiments on two types of laboratory models, one type constructed of heterogeneous materials and the other of homogeneous materials. A comparison of the experimental results for flow through the two model types shows that for heterogeneous porous material a dissolved constituent is dispersed widely; conversely, for material that is simply homogeneous a dissolved constituent is not dispersed in the moving fluid. This paper discusses results of experiments on the latter type material. The paper was prepared and should be read as a companion to USGS Professional Paper 411-F, by H. E. Skibitzke, in which experiments on heterogeneous materials are discussed.

Because the effects of heterogeneity cannot be expressed quantitatively, experimental procedures must entail the use of a more suitable concept. The observation of the flow is generally accomplished by following a tracer element in its progress downgradient. However, as tracer elements flow downstream they are dispersed to a degree depending on the conditions met in the porous medium—assuming the flow and fluid properties are unchanged. If it were possible to delineate a given series of streamlines, as they would be defined by Darcy's law, the need for describing the flow might be circumvented provided some measure of heterogeneity could be established. Generally, however, establishment of such measure is impossible because of physical limitations in experimental techniques.

Because it is physically impossible to trace out any given streamline, the description of the dispersive mechanism may be of importance. Dispersion in porous media has been studied to some extent, and a general description of the spreading of the flowlines can be predicted for isotropic media. This paper describes, for such a medium, an experiment on dispersion transverse to the direction of flow. Good correlation was obtained between analytical and experimental results. The distribution of concentration about the centerline thread of flow was almost symmetrical.

The tracer elements illustrate the history of events as the fluid progresses downstream. This seems to imply that if the dispersive effects can be separated the flow system can be

completely defined within the region. Although the tracer elements show the historical progress of each fluid element and even if it were possible to describe geometrically any flowline, the process of integrating this description throughout a region is extremely formidable—more likely impossible; hence, there is the need to establish a new outlook in describing the flow system, as discussed in some detail in Professional Paper 411-F.

Further research is required to determine the utility of a macroscopic concept of the flow system to replace the prevailing microscopic concept. The use of the microscopic analysis has led to the conclusion that the Navier-Stokes equation is valid; however, the integration of the point function throughout the region becomes impossible. In other words, each streamline needs to be described as a separate entity. Further, the microscopic concept has led to a statistical analysis of flow employing an ordered porous body and a random-flow system, which is in direct contradiction to the conclusion that the Navier-Stokes equation is valid. In essence, then, the microscopic analysis has produced a method of describing a point, whereas the description of a large-scale aquifer is the goal of the hydrologist.

The implications of studies already accomplished are that further experiments should be tried in larger regions and that a few small-scale supporting laboratory experiments should be added. Because in many of the tests made to date only dissolved constituents were observed, some different and more suitable type of tracer must be used to reveal the components of the dispersion process. Radiotracers have proven especially well suited for this purpose in most laboratory experiments.

The analysis of dispersion processes will necessarily require the use of some tracer techniques, simply because dispersion is the product of the motion of the dissolved component. In a macroscopic sense, of the order involved in ground-water field studies, the dispersion of contaminant does not depend upon any measurable hydraulic factor. Therefore, part of the experimental technique must involve addition of dissolved components to the fluid. Because the dispersion is also dependent upon the location and geometry of the injection site, no naturally occurring tracer element will suffice. Furthermore, once a tracer is decided upon, it will, in general, need to be a radiotracer because the problem of remote sensing is involved in ground-water studies. It seems necessary, then, that techniques be developed for extending the laboratory use of radiotracers into the field.

It is important at this point to note also that the use of tracers to determine hydraulic parameters is at present a useless endeavour. This has been discussed by Skibitzke (1958) in an earlier paper.

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FLUID MOVEMENT IN EARTH MATERIALS

THE SPREAD OF A DYE STREAM IN AN ISOTROPIC GRANULAR MEDIUM

By AKIO OGATA

ABSTRACT

The mechanics of dispersion transverse to the direction of fluid flow was studied both analytically and experimentally. A discussion of the overall mechanism is presented and the analytical expressions obtained are compared with experimental data. Correlation between experimental and theoretical results for the spread of a tracer is fair. However, from the data available an initial approximation is made for the dispersion constant in the direction transverse to the direction of flow.

INTRODUCTION

When a region saturated by a given fluid (say, fresh water) is invaded by another fluid (say, a tracer or salt water), a process of mixing occurs if the two fluids are miscible. Of special import is the mixing of salt and fresh water along coastal regions (Cooper, 1959). Additional occurrences of economic importance are the injection of low-level fission products into ground-water systems and the secondary recovery methods utilized by oil companies. For recovery the fluids can be immiscible and a zone of mixing is not created.

In the other two processes, because the carrier is usually water, there is creation and movement of a zone called "the zone of diffusion." This is a region in which there exists a gradation from the concentration of the original fluid to that of the invading fluid. Recent investigations have been concerned about the zone of diffusion because the final disposition of the contaminant in the transient state is dependent on the creation and movement of this zone. The purposes of the investigations reported herein are to describe the mechanisms involved and to predict quantitatively the final disposition of the contaminant with the passage of time.

The dilution of the contaminant may be attributed to three major processes—mechanical or hydrodynamic dispersion, adsorption, and dispersion due to heterogeneous aquifer conditions. Other minor mechanisms, such as ionic diffusion or discontinuous pore channels, have a tendency to cause spreading. However, the magnitude of spread due to these minor mechanisms is small in comparison to that due to the other processes except when the flow rate of the ground water is of the

same order as the mean flux due to diffusion. The rate of movement and the path traversed by this zone of dispersion are dependent on the existing flow condition and the method of injection. Thus, assuming that other things are equal—such as density of fluid and external pressures—and that Darcy's law is valid, the major mechanisms of dilution are classified as follows:

1. Mechanical or hydrodynamic dispersion—due to microscopic velocity variation controlled by viscosity and pore-channel diameters; principle mechanism in homogeneous and isotropic media.
2. Adsorption—due to mass transfer from liquid to solid phase controlled by chemical properties of contaminant and solid phase.
3. Heterogeneity—due to regional or macroscopic changes in the properties of the granular media.

Most papers on dispersion in a porous medium published to date have dealt exclusively with "mechanical dispersion" in the direction of flow, which is of primary importance in the analysis of reactor columns. However, in ground-water systems, adsorption and variation in the permeability of the media may be the major causes of the apparent dispersion. Adsorption or ion-exchange processes may cause significant dilution in a short distance owing to the presence of a large surface area available for the transfer of mass. Heterogeneity on the other hand relates to the finite changes in the properties of the aquifer. The occurrences of the discontinuities cause additional spreading that is due to refraction of the streamlines.

There is a different type of problem in regions near the seacoast. Although the problems involved are not the same, the mechanism of mixing is similar. Studies have been initiated (Kohout, 1960; Henry, 1959) to analyze the mixing in response to the tidal oscillation in the ocean and the natural velocity with which the ground water flows through the system. These effects, however, have not been separated and the importance of each process cannot yet be evaluated. Investigation is as yet in its initial stage, and a great deal of field data

is being accumulated, especially in Florida. However, the distribution of velocity within the two fluids and the effect of the change in densities along their interface remain to be determined either by in-place measurements or by model studies.

The discussion that follows is confined to dispersion of miscible fluids without density variations. It should be useful in determining the final disposition of radioactive waste in the event of spillage or leakage. The accumulated knowledge of the dispersion process in the ground-water system is directly applicable provided the geologic conditions can be described.

In essence, the final disposition of any contaminant is dependent on the path traversed. In ordinary circumstances the liquid waste first flows through the unsaturated zone overlying the ground-water system. Most of the contaminants are adsorbed in this region owing to the existing exchange potential of the soil. Because the adsorption depends on the types of materials present, all the contaminants may be fixed to the surface of the soil particles. A few of the fission products, however, may enter the saturated zone in which further dilution may occur.

In the saturated zone there are two possible means of reducing the concentration of the contaminated fluid. These are dilution by spreading, caused by mechanical dispersion, and dilution by actual removal of the contaminant from the liquid phase by adsorption. The fate of the contaminant in this region is thus dependent on the rate of flow, the geologic conditions, and the types of materials present.

Most of the reported studies of the dispersion phenomena are based on a gross analysis in which is assumed a diffusion process described by a relationship similar to Fick's law. The magnitude of the constant of proportionality, or the dispersion coefficient, is determined experimentally. These investigations of longitudinal dispersion (for example, de Josselin de Jong, 1958; Orlob and Radhakrishna, 1958) indicate that the magnitude of the dispersion coefficient D is proportional to the magnitude of the velocity u —that is, $D=mu$. The constant m is dependent on the size and distribution of aquifer material and also on the degree of saturation; thus it varies according to the individual situations considered. This relationship has been obtained from studies of unconsolidated-sand models composed of fairly uniform material. Thus, for application of the results to the existing field condition, or to consolidated materials, correction factors must be introduced. In regions where materials are extremely heterogeneous, it may be possible to increase the constant m to describe the dispersion or spread of contaminant.

The mechanism that controls the lateral spread

has not yet been extensively correlated experimentally. de Josselin de Jong (1958) and Saffman (1960) concluded from the study of statistical models that the lateral spread is also dependent on the average longitudinal velocity. However, in extremely slow flow, ionic diffusion becomes important (Beran, 1957). Laboratory study has been reported by Simpson (1962), and some laboratory investigations of lateral dispersion have been reported in chemical engineering journals. The investigations reported in these journals deal with flow rates much higher than those found in ground-water reservoirs. In most studies, good correlation is obtained between the lateral-dispersion coefficient and the velocity in the longitudinal direction.

For both lateral and longitudinal dispersion in isotropic media, the spread is due primarily to microscopic controls or variations. But, in nonhomogeneous or consolidated materials, macroscopic variations may have a marked effect. When these variations constitute changes in geologic conditions, the term "heterogeneity" applies. Heterogeneous conditions exert a large influence on the breaking up and the spread of a stream of contaminant progressing through the saturated zone. There is little doubt that these macroscopic discontinuities exist in many aquifer systems and depend on environmental conditions at the time the aquifer materials were deposited. Of course, the objective is still to obtain quantitative measures of the spread of a contaminant. Because the prevailing environmental conditions at the time of deposition are not known, it would be virtually impossible to describe quantitatively the individual discontinuities or changes in a large region. But reason suggests that, as for dispersion in homogeneous media, all macroscopic variations may be treated separately by using an averaging process. By this means it may be possible to describe the spread or mixing due to heterogeneity, assuming that the constant m is dependent on the size and distribution of isolated discontinuities.

The mechanism of adsorption or ion exchange, which is of primary importance in predicting the fate of the contaminant, is usually not mentioned in studies of dispersion. Adsorption cannot be classified as a dispersion phenomenon; it is primarily a surface chemical process due to the existence of an exchange potential between the dissolved contaminant and the surface of the media through which the fluid flows. This exchange capacity is primarily dependent on the solid material and the type of contaminant present (Kaufman and Orlob, 1956). The dilution of the liquid phase is realized because of the transfer of dissolved solid from the liquid state to the solid state. Although the contaminant may not be permanently fixed, the rate of exchange from liquid to solid is

usually large enough that considerable dilution is realized. In this way adsorption may be the most effective method of reducing contamination hazards. Adsorption is discussed further in the next section.

Analysis of dispersion with linear adsorption isotherm superimposed on diffusion has been attempted. However, major simplification is usually necessary to carry out analytical details. In laboratory experiments utilizing continuously injected organic dye tracers, a considerable travel delay due to adsorption or ion exchange is usually noted, but ordinarily no quantitative description is attempted. Investigations of Houghton and Marshall (1947) are an example of this attempt; however, the fluid was gas and the simplification utilized cannot be applied to liquid flows.

The lack of available data on transverse dispersion and effects of heterogeneity prompted the Phoenix research office of the U.S. Geological Survey to construct models for laboratory analysis of dispersion. Owing to time limitations and the urgency of other projects, laboratory-model studies of heterogeneity were intended to furnish qualitative rather than quantitative description of the process (Skibitzke and Robinson, 1963). Both mathematical and laboratory studies were made to analyze transverse diffusion. A value of the transverse-diffusion coefficient has been obtained; however, inasmuch as only one test run was made, this value must be substantiated further.

The laboratory experiment and the analysis of the experimental block was carried out by H. T. Chapman and J. M. Cahill of the Phoenix research office. Their diligence and ingenuity in overcoming many difficult technical problems resulted in the successful conclusion of the experimental phase.

ADSORPTION

Although adsorption is not a dispersion process, a short discussion is presented because the experiment reported requires knowledge of concentration relationships between the liquid and the solid phases. Generally, if the concentration of the tracer used in the experiment is low, a linear relationship can be assumed provided an equilibrium condition is reached. This relationship follows from the assumption that the Langmuir exchange isotherm is valid. All the isotherms reported are semiempirical in nature. Although there are many published reports of transfer from solutions to solid surfaces, the exchange isotherms utilized are valid only for the systems reported, inasmuch as the theoretical expressions are obtained only after a series of simplifications to construct a mathematical model.

First, consider the exchange isotherms for equilibrium conditions. The isotherms presented are based on

the assumption that only a monomolecular film is formed around the solid particles. This fact, in essence, tends to limit the use of the isotherms presented unless extremely low concentrations are used in the experiments.

Adsorption of particles from gaseous mixtures onto a solid surface depends primarily on temperature and pressure (Adamson, 1960). In transfer of mass from a solution, concentration of the solution replaces the pressure. The isotherms proposed by Langmuir and Freundlich are those most widely used, because of their simplicity. Freundlich's isotherm, at first thought to be an empirical relationship, is written

$$F = aC^{1/n}$$

where

F = amount of adsorbate per unit amount of solid,

a and n = constants, and

C = concentration of liquid phase.

Langmuir's isotherm, on the other hand, can be written

$$F = abC/(1 + bC)$$

where

a = a constant related to the specific surface area of the solid, and

b = a constant related to the heat of adsorption.

Roughly speaking, the constants appearing in the Freundlich isotherm may be interpreted to represent the same specific area and heat of adsorption. For both the Freundlich and Langmuir isotherms, a curve of F versus C shows a convexity towards the C axis. However, there is one major difference: the Langmuir isotherm reaches a saturation value of a as C increases and bC becomes much greater than unity, whereas the Freundlich isotherm does not reach an upper limit. Furthermore, when the concentration is small, or $bC \ll 1$, the Langmuir isotherm gives a straight-line relationship between the liquid concentration and the solid concentration, though the Freundlich isotherm does not show this effect and the straight-line approximation is obtained only when $n \rightarrow 1$. In this circumstance, the isotherm is a straight line regardless of concentration.

Investigations of adsorption on solids from solutions indicate that porous adsorbents usually obey Langmuir's equation because the pore volume limits the amount adsorbed (Condon and Odishaw, 1958). However, this evidence does not prove that Langmuir's expression is superior to Freundlich's expression. On

the contrary, many laboratory data show better correlation with the Freundlich isotherm, but when concentrations are low Langmuir's isotherm seems better suited for a large number of adsorption systems.

Later researchers have reported a sigmoid isotherm (fig. 1), which is indicative of a multilayered formation for transfer from the vapor phase to the solid phase (Adamson, 1960). Although very little is known as to whether multilayers actually form, there seems to be a general acceptance of this possibility. The theoretical analysis involves the use of Langmuir's isotherm for n layers. The expression was developed by Brunauer, Emmett, and Teyerson and is generally known as the BET equation (Condon and Odishaw (1958) or Adamson (1960)).

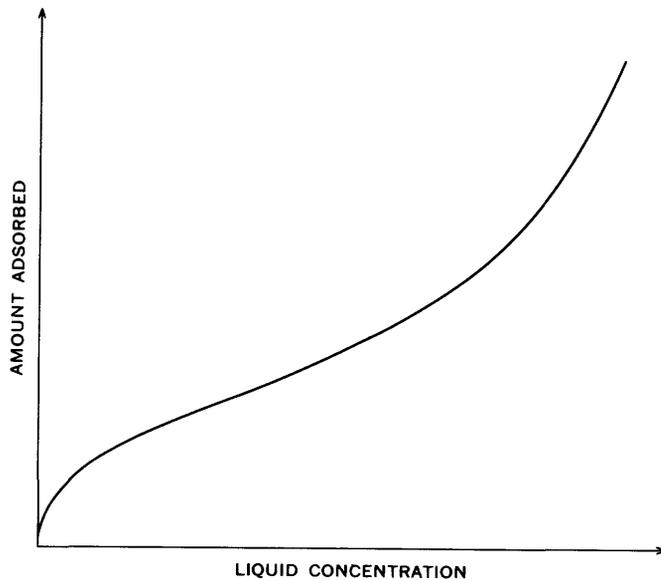


FIGURE 1. Graph of the isotherm characteristic of multilayer adsorption.

In utilizing the equilibrium isotherms discussed previously, it is assumed that there is instantaneous transfer of mass. The assumption of instantaneous adsorption does simplify to a great extent the analysis of the combined effects of adsorption and diffusion. However, for a fluid flowing through a stationary porous bed, this assumption may not be valid unless the time elapsed is large. In nonequilibrium conditions the expression that must be considered is

$$\frac{\partial F}{\partial t} = f(C),$$

assuming that rate of transfer is a function of the liquid concentration only. Generally the function $f(C)$ is not described but an assumed relationship, usually linear, is superposed on the diffusion equation.

Two types of nonlinear expressions for $f(C)$ were discussed by de Vault (1943). Generally, $f(C)$ is assumed to be linear; a typical relationship (for example, that given by Merriam and others (1952)) has the form

$$\frac{\partial F}{\partial t} = a(bC - F).$$

Here again it must be assumed that the exchange process is instantaneous. Thus, whether equilibrium is reached or not, the assumed relationship of F or $\partial F/\partial t$ to C is similar to Langmuir's or Freundlich's isotherm.

MATHEMATICAL ANALYSIS OF TRANSVERSE DISPERSION WITH RADIAL SYMMETRY

In the mathematical development of dispersion in granular media, various simplifications are made so that the problem becomes tractable. The first simplification is the assumption that a piston-type flow exists, that is, that every element of fluid is flowing at a velocity equal to its average velocity. The second assumption is that the apparent dispersion due to microscopic variation of velocity in the individual pore channels is described by a relationship similar to Fick's law of diffusion. The component of total mass transported in the x direction (say) for isotropic media is thus given by the relationship

$$-q_x = D_x \frac{\partial C}{\partial x} - uC \quad (1)$$

where

- q = the total mass flux across any given plane,
- D_x = the constant of proportionality depending only on direction x ,
- C = tracer concentration in fluid,
- x = directional component, and
- u = average fluid velocity in the x direction.

If no chemical reaction occurs, the law of conservation of mass gives the differential equation

$$\frac{\partial J_x}{\partial x} + \frac{\partial J_y}{\partial y} + \frac{\partial J_z}{\partial z} = \frac{DC}{Dt}$$

where

$$\frac{D}{Dt} = \frac{\partial}{\partial t} + u \frac{\partial}{\partial x} + v \frac{\partial}{\partial y} + w \frac{\partial}{\partial z} \quad (2)$$

- J_i = mass flux due to dispersion only in the direction i , and
- u, v, w = constant average velocity components in the $x, y,$ and z directions, respectively.

In anisotropic media where the dispersion coefficient D depends on the direction, D takes on the properties

of a tensor. Hence, the components of dispersion at any given point are described by

$$\left. \begin{aligned} -J_x &= D_{11} \frac{\partial C}{\partial x} + D_{12} \frac{\partial C}{\partial y} + D_{13} \frac{\partial C}{\partial z} \\ -J_y &= D_{21} \frac{\partial C}{\partial x} + D_{22} \frac{\partial C}{\partial y} + D_{23} \frac{\partial C}{\partial z} \\ -J_z &= D_{31} \frac{\partial C}{\partial x} + D_{32} \frac{\partial C}{\partial y} + D_{33} \frac{\partial C}{\partial z} \end{aligned} \right\} \quad (3)$$

In other words, the mass flux in any given direction is not only dependent on the concentration gradient in the same direction but also on the gradient in the other two directions. Owing to mathematical difficulties and the lack of experimental data available for anisotropic conditions, it is assumed that the principle direction of dispersion is along the axis of the coordinate system chosen.

Equation 2 has been expressed in terms of Cartesian coordinates; however, the equation may be written for any system of coordinates. Using appropriate transformations

$$\begin{aligned} x &= r \cos \theta \\ y &= r \sin \theta \\ z &= z, \end{aligned}$$

equation 2 in cylindrical coordinates is

$$\frac{DC}{Dt} = \frac{1}{r} \frac{\partial}{\partial r} \left(r D_r \frac{\partial C}{\partial r} \right) + \frac{1}{r^2} \frac{\partial}{\partial \theta} \left(D_\theta \frac{\partial C}{\partial \theta} \right) + \frac{\partial}{\partial z} \left(D_z \frac{\partial C}{\partial z} \right). \quad (4)$$

The differential equation of diffusion may also be written in spherical coordinates, but it is not as useful as the expression involving Cartesian coordinates for problems of flow through granular media.

Generally in flow through porous media the velocity component can be considered to be unidirectional and the principal axes of diffusion are assumed parallel and transverse to the velocity. Thus for unidirectional flow, equation 2 is reduced to

$$D_i \nabla^2 C = \frac{\partial C}{\partial t} + u \frac{\partial C}{\partial x}; \quad i = x, y, \text{ and } z. \quad (5)$$

Utilizing either the substitution $C = \Gamma \exp \left[\frac{ux}{2D} - \frac{u^2 t}{4D_x} \right]$ or the moving coordinate $z = x - ut$, equation 5 can be reduced to the standard diffusion equation

$$D_i \nabla^2 C = \frac{\partial C}{\partial t}. \quad (6)$$

In the cylindrical coordinate system, where radial symmetry exists, the principal axes of dispersion are

assumed to be in the directions parallel and transverse to the direction of flow. Thus the dispersion equation can be written

$$\frac{\partial C}{\partial t} + u \frac{\partial C}{\partial x} = D_x \frac{\partial^2 C}{\partial x^2} + \frac{D_r}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C}{\partial r} \right). \quad (7)$$

Equation 7 can be reduced to the diffusion equation by employing the moving-coordinate system, or $\eta = x - ut$; hence

$$\frac{\partial C}{\partial t} = D_x \frac{\partial^2 C}{\partial \eta^2} + \frac{D_r}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C}{\partial r} \right). \quad (8)$$

Equation 6 in its one-dimensional form has been solved for various boundary conditions in the investigations of both heat-flow and diffusion processes. Analytical and experimental correlation studies have been carried out by many investigators, as previously stated. These investigations have indicated that the dispersion coefficient is directly proportional to the average velocity of the fluid both transverse and parallel to the direction of flow, but there is no indication that the coefficients are of equal magnitude in both directions.

Industrial interest has prompted extensive study of longitudinal dispersion, in both the laminar range (Reynolds No. < 1) and the so-called turbulent range (Reynolds No. > 1), but few studies have been made either analytically or experimentally of dispersion transverse to the direction of flow. The remainder of this section will therefore be confined to the development of a mathematical description of transverse diffusion.

Analytical difficulties are inherent in equation 7, even for $D_r = D_x$, unless boundary conditions are simple. Thus, to simplify the analysis to a large degree, consider separately the two processes of longitudinal and transverse dispersion. Except in the frontal zone where longitudinal dispersion predominates, the system can be described approximately by

$$D_r \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C}{\partial r} \right) - u \frac{\partial C}{\partial x} = \frac{\partial C}{\partial t}. \quad (9)$$

Von Rosenberg (1956) indicated that the length of the frontal zone, over which the concentration varies about 80 percent, is proportional to the velocity and inversely proportional to the diffusion coefficient. Because the diffusion coefficient is proportional to velocity once the front is established, it remains nearly constant as the stream progresses through the media. Further, if no chemical reaction or adsorption of the contaminant takes place, there is no "holdup" time and the front progresses through the media at a rate equal to the average velocity. The foregoing observations are based on one-dimensional problems; thus, the simplifications may not be wholly justified physically.

To simplify the mathematical model it is assumed that the contribution of the front to the overall lateral spread is small, especially in the zone $x > ut$. In other words, in the mathematical model it is assumed that a piston flow takes place; that is, a sharp front traverses the media with a velocity u , and diffusion occurs normal to the direction of flow. For this simplified model it is apparent that equation 9 holds.

By letting $z = x - ut$ and $\tau = t$, it is possible to simplify equation 9 further: that is,

$$D \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C}{\partial r} \right) = \frac{\partial C}{\partial t}$$

Physically, this equation means that an observer follows a slug of contaminant at a rate equal to the average velocity u as the contaminant progresses through the system. For the unsteady condition this approximation requires that the boundary conditions be

$$\begin{aligned} t=0, \quad C=C_0, \quad r < a \\ t=0, \quad C=0, \quad r > a \\ r=0, \quad \frac{\partial C}{\partial r} = 0. \end{aligned}$$

If the source emits at a constant rate, this problem is the same as diffusion from a semi-infinite cylinder of radius a into the surrounding media. In the steady situation, $\partial C / \partial t = 0$; thus t can be replaced by x/u in the boundary condition specified above.

The solution of the above system has been obtained and discussed extensively (Goldstein, 1932). This solution written in equivalent form is

$$\left. \begin{aligned} \frac{C}{C_0} &= a \int_0^\infty e^{-D\alpha^2 t} J_1(\alpha a) J_0(\alpha r) d\alpha \\ &= \exp \left[-\frac{(\eta-1)^2}{2\eta} \xi \right] \sum_{m=1}^\infty \eta^m e^{-\xi} I_m(\xi) \\ &= \exp \left[-\frac{(\eta-1)^2}{2\eta} \xi \right] \sum_{m=0}^\infty \frac{1}{\eta^m} e^{-\xi} I_m(\xi) \end{aligned} \right\} \quad (10)$$

where $\eta = a/r$, $\xi = ra/2Dt$, and J_1 and J_0 are Bessel functions of the first kind of order 1 and 0, respectively. The integral solution was written in its alternate form to facilitate computation of the general solution. The author showed this solution in graphical form in chapter B of this series (Ogata, 1961). As previously indicated, the solution of the steady-state system is obtained by simply replacing t by $\frac{x}{u}$. Because the experiment that was performed is assumed to have reached steady state, the solution to equation 10 is hereafter written in its steady-state form.

Although equation 10 can be computed, the radial-dispersion coefficient cannot be easily determined from the resulting expression. However, the integral can be integrated for the specific situation of either $r=0$ or $r=a$. Consider first the distribution of concentration along $r=0$. Because $J_0(0)=1$, equation 10 can be written

$$\frac{C}{C_0} = a \int_0^\infty e^{-D\alpha^2 x/u} J_1(\alpha a) d\alpha.$$

Watson (1948, p. 394) gave the value of the above integral as

$$\frac{C}{C_0} = 1 - \exp \left[\left(-\frac{a^2 u}{4Dx} \right) \right]. \quad (11)$$

If the concentration distribution is experimentally determined along $r=0$, the radial component can easily be computed using equation 11.

However, the radius of a finite source of tracer may be such that within the limits of the experimental model no noticeable change in concentration will occur along $r=0$. For this situation, it would be preferable to determine distribution along a known value of r , say a . When $r=a$, equation 10 can be written

$$\frac{C}{C_0} = a \int_0^\infty e^{-D\alpha^2 t} J_1(\alpha a) J_0(\alpha a) d\alpha.$$

Goldstein (1932) expressed the above integral in terms of the modified Bessel function of the first kind, that is,

$$\frac{C}{C_0} = \frac{1}{2} \left[1 - \exp \left(-\frac{a^2 u}{2Dx} \right) I_0 \left(\frac{a^2 u}{2Dx} \right) \right]. \quad (12)$$

Inasmuch as the function $e^{-x} I_0(x)$ is well tabulated, the term D can be readily computed from any given experimental data. Here again the value of D can be obtained by a curve-fitting process.

Further reflection on the problem of radial dispersion led to the hypothesis that the physical assumptions may not be realistic and that this problem may best be described as diffusion from a moving-disk source in a semi-infinite medium. A second analysis was thus attempted based on the assumption that, owing to radial symmetry, the coefficient of dispersion depended only on the x and r directions. A summary of the results obtained is presented. It should be noted that the complete solution for the unsteady-state system was not computed because of the complexity of the resulting integral. But, for comparison purposes the steady-state system was computed for various values of the ratio D_x/D_r .

In approximating the process of transverse diffusion,

it was assumed that the term $D_x \frac{\partial^2 C}{\partial x^2}$ was negligible in equation 9. It was also assumed that the conditions of the experiment justified the use of the steady-state system. These conditions cannot be checked experimentally although the theoretical solution can be correlated with experimental evidence.

Because radial symmetry exists, equation 7 would describe the process. For the instantaneous point source (Carslaw and Jaeger, 1959, p. 260), it is possible to write the solution of the problem immediately. The expression

$$\frac{C}{C_0} = \frac{u'a'}{2\sqrt{\pi D}} \int_0^t \exp \left\{ -\frac{[x-u'(t-\tau)]^2}{4D(t-\tau)} \right\} \frac{d\tau}{\sqrt{\tau-t}} \times \left\{ \int_0^\infty e^{-D(t-\tau)\lambda^2} J_0(\lambda\rho) J_1(\lambda a') d\lambda \right\} \quad (13)$$

is obtained by integrating the expression for the continuous point source from zero to a , where a is the radius of the disk source. The terms used in equation 13 are

$$\begin{aligned} u' &= u\sqrt{D/D_x} & \rho &= r\sqrt{D/D_r} \\ \chi &= x\sqrt{D/D_x} & a' &= a\sqrt{D/D_r} \end{aligned}$$

Inasmuch as the strength of the source is $u'C_0$, the expression for steady-state conditions is

$$\begin{aligned} \frac{C}{C_0} &= \frac{2}{u'\sqrt{\pi}} \int_0^\infty a' J_0(\lambda\rho) J_1(\lambda a') d\lambda \int_0^\infty \exp \left[-\left(1 + \frac{4D^2}{u'^2} \lambda^2\right) \xi - \frac{\chi^2(u')^2}{16D^2\xi^2} \right] d\xi = \frac{a'}{u'} \int_0^\infty \exp \left[-\frac{\chi u'}{2D} \left(1 + \frac{4D^2}{(u')^2} \lambda^2\right)^{1/2} \right] J_0(\lambda\rho) J_1(\lambda a) \frac{d\lambda}{\left(1 + \frac{4D^2}{(u')^2} \lambda^2\right)^{1/2}} \quad (14) \end{aligned}$$

Equation 14 cannot be integrated further, but comparison with equation 10 shows that when $\frac{4D^2}{(u')^2} \lambda^2 \ll 1$, the expressions are identical.

Consider the concentration distribution along $r=0$. Substituting $\rho=0$ in equation 14 and integrating yields the expression

$$\frac{C}{C_0} = 1 - \exp \left[\frac{ux}{2D_x} \left(1 - \sqrt{1 + \frac{a^2 D_x}{x^2 D_r}}\right) \right] \quad (15)$$

Note that if $\frac{a^2 D_r}{x^2 D_x} \ll 1$ and the approximation $(1+x)^{1/2} \approx (1+x/2)$ is used, the term under the radical in equation 15 can be simplified and equation 15 becomes

$$\frac{C}{C_0} = 1 - e^{-ua^2/4D_x x},$$

which is identical to equation 11. Thus it can be stated that for any given ratio D_x/D_r , the approximate solution is valid for large distances from the source.

The value of the parameter $\frac{a^2 D_x}{x^2 D_r}$ at which the approximation is correct to the nearest thousandth is about 10^{-4} . Figure 2 shows a comparison between equations 11 and 15.

For $r=a$ the solution based on the disk source gives

$$\frac{C}{C_0} = \frac{1}{2} \left\{ 1 - \frac{1}{\pi} \exp \left(\frac{ux}{2D} \right) \int_0^1 \exp \left[\frac{ux}{2D} \sqrt{1 + \frac{4a^2 D_x}{x^2 D_r} \tau} \right] \frac{d\tau}{\sqrt{1-\tau^2}} \right\} \quad (16)$$

Again, as $\frac{a^2 D_x}{x^2 D_r} \tau$ becomes small,

$$\sqrt{1 + \frac{4a^2 D_x}{x^2 D_r} \tau} \approx 1 + \frac{1}{2} \left(\frac{2a}{x} \right)^2 \frac{D_x}{D_r} \tau.$$

Hence

$$\frac{C}{C_0} = \frac{1}{2} \left[1 - \exp \left(\frac{-a^2 u}{2D_r x} \right) I_0 \left(\frac{a^2 u}{2D_r x} \right) \right],$$

which is identical to the approximate solution, or equation 12, for $r=a$.

Above analysis of the disk source thus indicates that if the experimental column is sufficiently long the radial-diffusion coefficient can be computed by equation 11 or 12. In addition, by utilizing equation 15 or 16, the longitudinal-dispersion coefficient can be readily computed provided reliable data are obtained near the source.

Experimental Method

The experiment on diffusion transverse to the direction of flow was conducted with an isotropic, artificially constructed sandstone. Radioactive phosphorous was utilized as a tracer which was introduced into the flow system through a tube three-fourths of an inch in diameter. Except for the diameter of the source and the mode of controlling the quantity of tracer injected, the method utilized was exactly the same as that described by Skibitzke and others (1961). Physical data are as follows:

Length of block.....	43.5 cm
Diameter of source.....	.95 cm
Average fluid velocity.....	2.27×10^{-2} cm per min
Average sand-grain diameter.....	.44 mm
Uniformity coefficient.....	1.92
Total time of experiment.....	383.5 hr

As in the previous experiment, the amount of tracer introduced was determined by the ratio of the cross-sectional areas of the source and the block. By controlling the quantity of flow rather than the head of the source, the problem of measuring extremely small

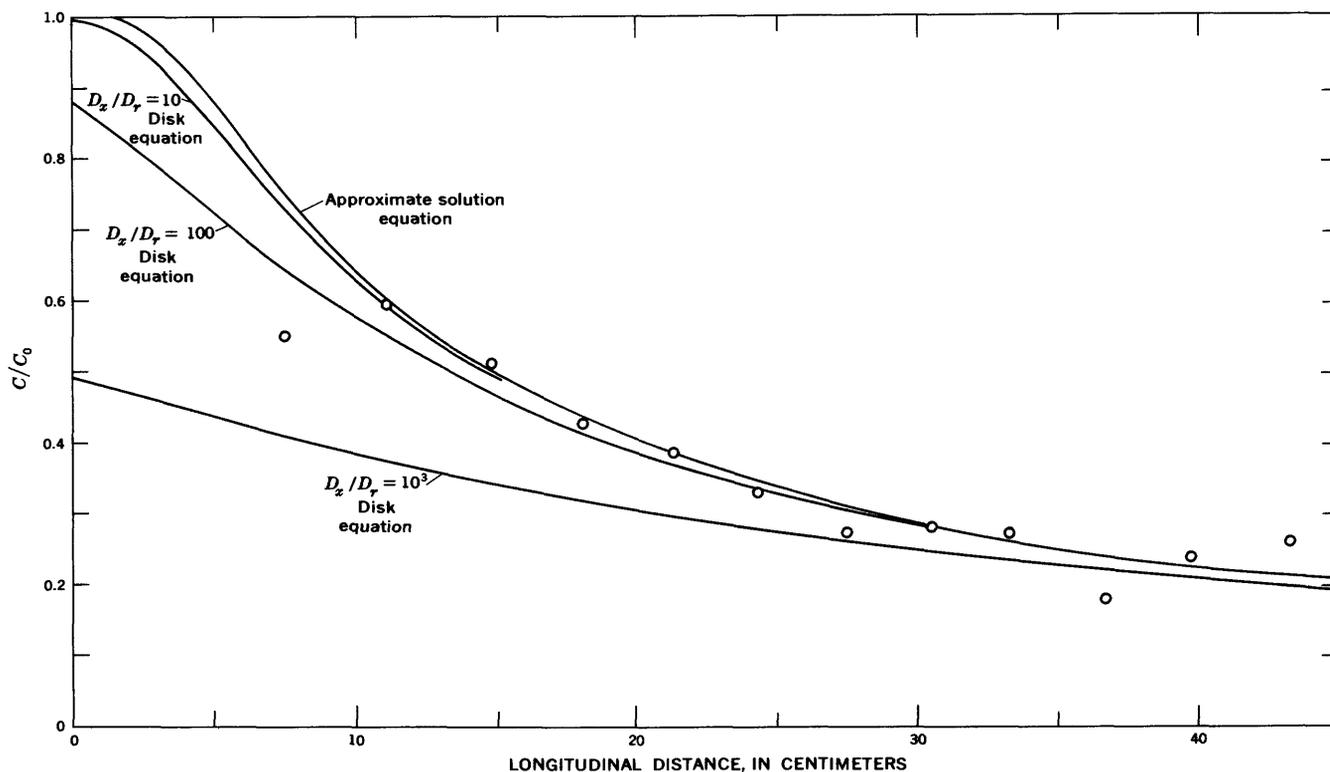


FIGURE 2.—Graph showing comparison between equations 11 and 15 for various ratios of D_x/D_r . Concentration distribution along $r=0$. Curves, theoretical; \circ , experimental.

differences in head was eliminated. The quantity of flow was controlled by dropping glass beads into a storage tank which fed the tracer source. Although this method of quantity control does not satisfy the boundary condition calling for a continuous source, over the period of the experiment (17 days) the condition was averaged; the continuous source was therefore approximated.

After a steady flow of tracer was detected at the outflow end of the block, the experiment was terminated and the block dissected. The cross section of the block was then assayed by means of a needle probe and a photoemulsion method. The data reported were obtained using the needle probe. The emulsion method showed a distinctive pattern of spread; however, no correlation could be obtained between the counts and the density of light transmitted through the film.

ANALYSIS OF EXPERIMENTAL DATA

In analyzing the experimental data, the basic assumption is made that a multilayer of P^{32} was formed by adsorption, except in the lower reaches of the model. Owing to the fairly low concentration readings realized in the lower parts of the block, the relationship of the liquid concentration and the solid concentration was assumed to be almost linear. That is, Langmuir's isotherm was assumed to be valid with $bC \ll 1$. This

basic assumption made possible the comparison of theoretical and experimental values of any given section of the experimental model.

This type of analysis was attempted because of large discrepancies observed along the centerline of the trace filament near the tracer source. The rapid decline of concentration from distances 0 through 10 cm could not be correlated with any of the theoretical results. However, a good fit was obtained for distances greater than 11 cm from the source and the dispersion coefficient D was therefore computed from these data. The computations showed a range from 6.7×10^{-4} to 11.8×10^{-4} cm^2 per min. Similar computations for $r=a$ using equation 12 gave a range in D from 6.0×10^{-4} to 10.3×10^{-4} cm^2 per min. For distances less than 11 cm the value of the coefficient increased approximately tenfold, the largest values being near the source.

Because the concentration of the entering fluid was not measured, the assumption that the adsorption formed a multilayer except in the lower reaches made it necessary that this entering concentration be computed. Inasmuch as the ratios of the concentration $C_{r=a}/C_{r=0}$ for various distances were available for both theoretical and experimental conditions, these ratios were used. Figure 3 shows the theoretical relationship between $C_{r=a}/C_{r=0}$ and $ua^2/4xD_r$. After experimental values were obtained for $C_{r=a}/C_{r=0}$, it was possible to

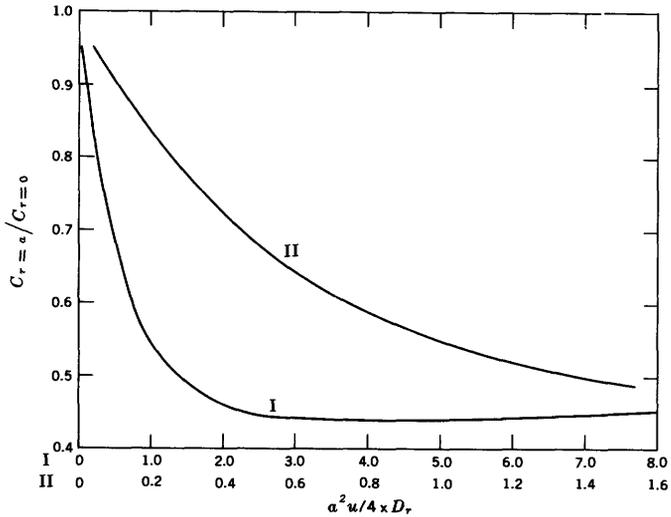


FIGURE 3.—Graph showing theoretical relationship between $C_{r=a}/C_{r=0}$ and $ua^2/4xD_r$.

compute values of D for each given section of the experimental model. The value of D obtained by this method ranged from 3.9×10^{-4} to 6.0×10^{-4} cm² per min, as shown in table 1. The arithmetic average was computed to be 5.0×10^{-4} cm² per min. By utilizing the average value of the dispersion coefficient, an average value of C_0 was computed to be approximately 140 counts per minute.

TABLE 1.—Computation of $ua^2/4xD_r$ from actual ratio $C_{r=a}/C_{r=0}$ for various distances $x > 11.0$ cm
[Average: $q=10.3$ cm; $D_r=4.97 \times 10^{-4}$ cm² per min]

x	$C_{r=0}$	$C_{r=a}$	$C_{r=a}/C_{r=0}$	Theoretical $ua^2/4xD_r$	$q=ua^2/4D_r$	$D_r \times 10^4$
11.1	83.0	48.5	0.584	0.81	9.0	5.68
14.8	72.0	43.0	.597	.76	11.2	4.56
18.1	59.2	39.5	.667	.53	9.6	5.32
21.3	54.0	34.5	.639	.61	13.0	3.93
24.3	46.2	34.5	.747	.35	8.5	6.01
27.6	38.5	28.0	.727	.39	10.8	4.73
30.6	39.3	30.3	.771	.31	9.5	5.38
33.3	37.5	30.0	.800	.26	8.7	5.87
36.7	26.0	19.0	.780	.33	12.1	4.22

RESULTS

By utilizing the values $C_0=140$ cpm and $D=5 \times 10^{-4}$ cm² per min, theoretical and experimental values were compared as shown in figures 4 through 15. Note that for most of the sections the correlation is good. However, in some sections all experimental points fall consistently below the theoretical curve, perhaps primarily because of instrumental errors, though the exact causes are unknown.

The approximate analysis of the experimental data does seem to indicate that diffusion coefficients transverse to the direction of flow are of the same order of magnitude as ionic diffusion. A spread to approximately three times the width of the source at a dis-

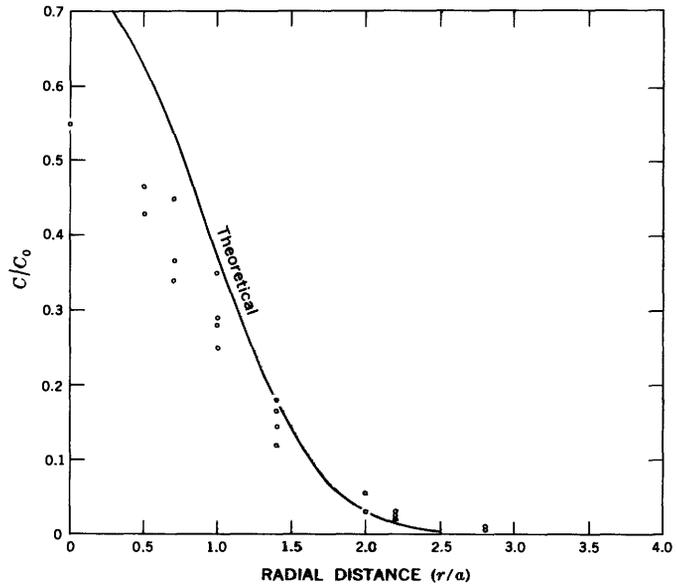


FIGURE 4.—Graph showing concentration distribution along radius r , block 11. $x=7.5$ cm, $a=0.95$ cm. \circ , experimental.

tance of 43 cm seems indicated. Because only one test run was made, these figures should be taken as preliminary in nature.

Future experiments should take into account the nature of the adsorption isotherms. Generally, it is good practice to ascertain the range of concentration for which the linear part of the Langmuir isotherm can be used. Analysis of the data does show that transverse diffusion is small; thus, if future experiments are desired, the described method can be utilized provided the gross transfer of mass between the liquid and the solid phases is determined for the concentration and for the tracer used.

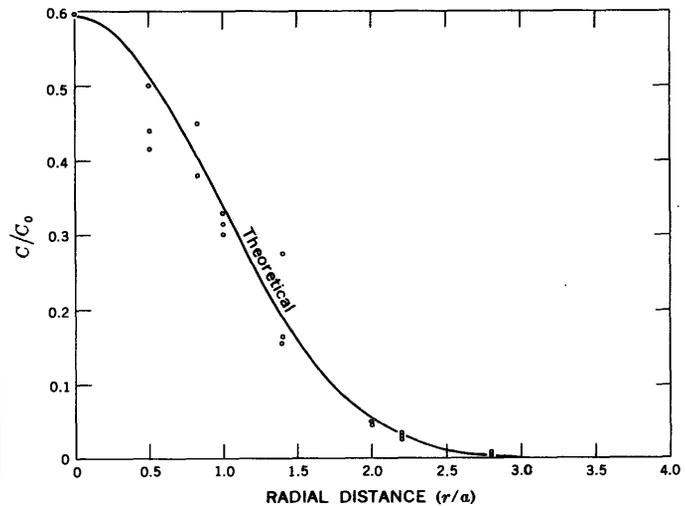


FIGURE 5.—Graph showing concentration distribution along radius r , block 10. $x=11.1$ cm, $a=0.95$ cm. \circ , experimental.

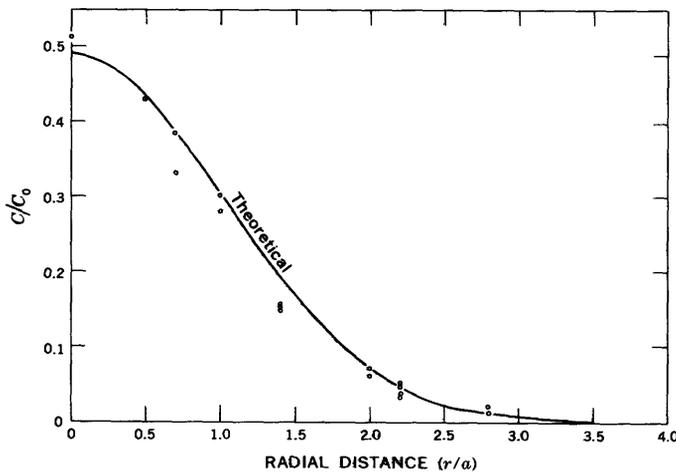


FIGURE 6.—Graph showing concentration distribution along radius r , block 9. $x=14.8$ cm, $a=0.95$ cm. \circ , experimental.

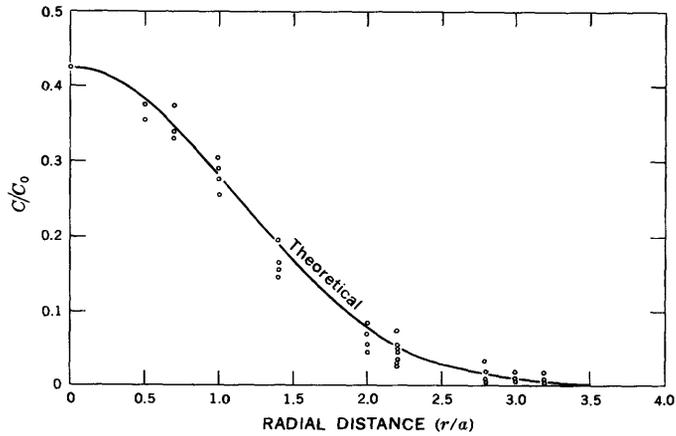


FIGURE 7.—Graph showing concentration distribution along radius r , block 8. $x=18.1$ cm, $a=0.95$ cm. \circ , experimental.

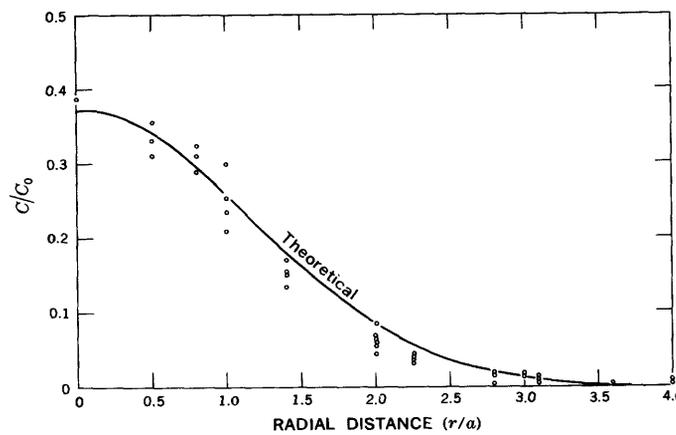


FIGURE 8.—Graph showing concentration distribution along radius r , block 7. $x=21.3$ cm, $a=0.95$ cm. \circ , experimental.

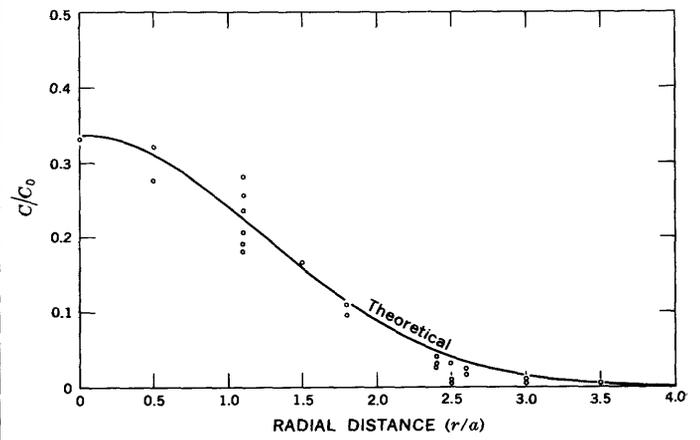


FIGURE 9.—Graph showing concentration distribution along radius r , block 6. $x=24.3$ cm, $a=0.95$ cm. \circ , experimental.

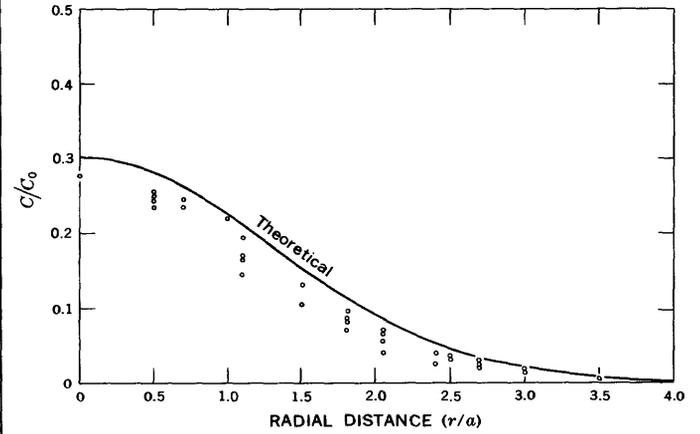


FIGURE 10.—Graph showing concentration distribution along radius r , block 5. $x=27.6$ cm, $a=0.95$ cm. \circ , experimental.

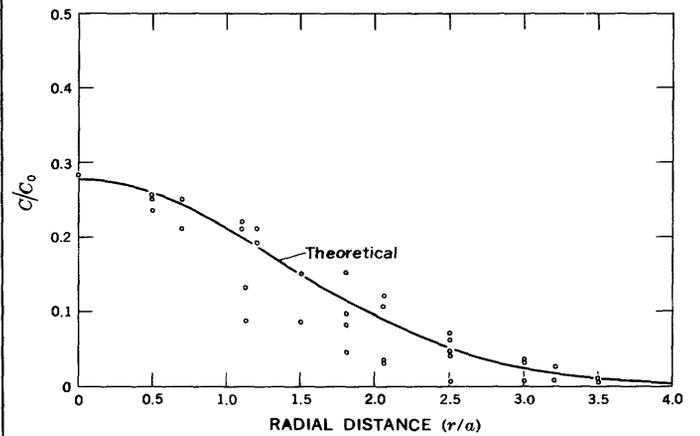


FIGURE 11.—Graph showing concentration distribution along radius r , block 4. $x=30.6$ cm, $a=0.95$ cm. \circ , experimental.

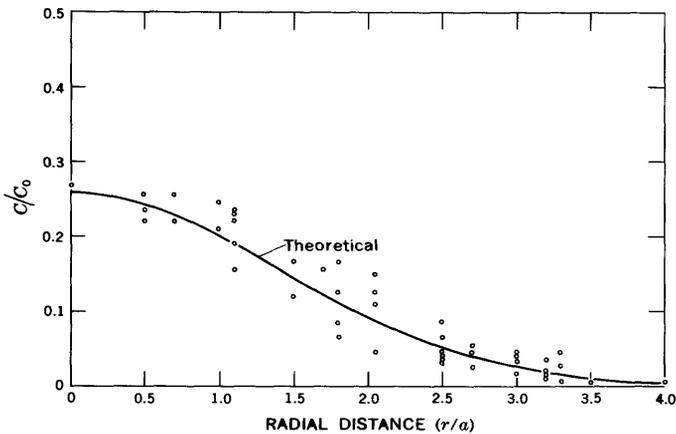


FIGURE 12.—Graph showing concentration distribution along radius r , block 3. $x=33.3$ cm, $a=0.95$ cm. \circ , experimental.

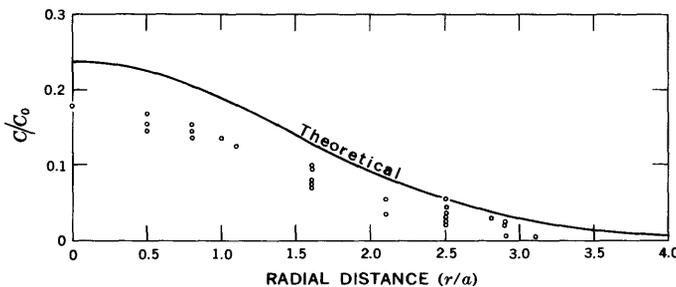


FIGURE 13.—Graph showing concentration distribution along radius r , block 2. $x=36.7$ cm, $a=0.95$ cm. \circ , experimental.

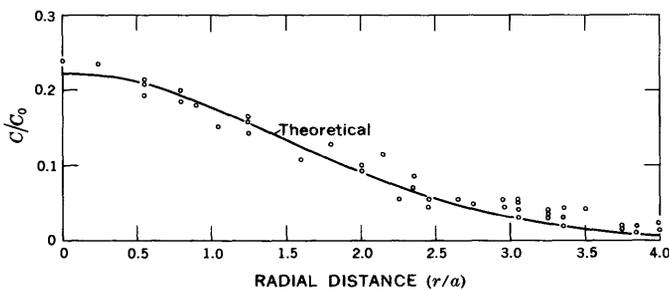


FIGURE 14.—Graph showing concentration distribution along radius r , block 1. $x=39.8$ cm, $a=0.95$ cm. \circ , experimental.

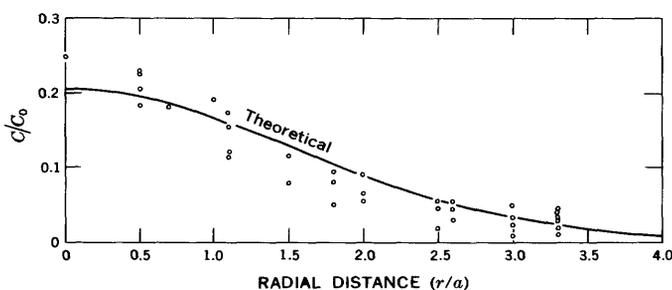


FIGURE 15.—Graph showing concentration distribution along radius r , block 14. $x=43.3$ cm, $a=0.95$ cm. \circ , experimental.

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The U.S. Geological Survey Library has cataloged this publication as follows:

Ogata, Akio, 1927-

The spread of a dye stream in an isotropic granular medium. Washington, U.S. Govt. Print. Off., 1964.

v, 11 p. diags., table. 30 cm. (U.S. Geological Survey. Professional paper 411-G)

Fluid movement in earth materials.

Prepared in cooperation with the U.S. Atomic Energy Commission, Division of Isotope Development and Division of Reactor Development.

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Ogata, Akio, 1927- The spread of a dye stream in an isotropic granular medium. 1964. (Card 2)

1. Dispersion. 2. Water, Underground. 3. Fluids. I. U.S. Atomic Energy Commission. II. Title. (Series)

