POLLUTANT TRANSPORT IN NATURAL STREAMS

by

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A paper accepted for presentation at the Topical Meeting on Computational Methods in Nuclear Engineering in Charleston, S. C., on April 15-17, 1975, and sponsored by the American Nuclear Society.

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A mathematical model has been developed to estimate the downstream effect of pollutant releases to tributary streams and rivers. The one-dimensional dispersion model was employed along with a dead zone model to describe stream transport behavior. Options are provided for sorption/desorption, ion exchange, and particle deposition in the river. The model equations are solved numerically by the LODIPS computer code. The solution method was verified by application to actual and simulated releases of radionuclides and other chemical pollutants.

Work performed under USERDA Contract No. AT(07-2)-1.
INTRODUCTION

An accurate prediction of pollutant movement in a stream or river is necessary to assess the downstream effect of normal and accidental releases of radionuclides or process chemicals from a nuclear or chemical facility. This problem is becoming increasingly important due to the growth of the nuclear industry and the development and construction of commercial nuclear fuel reprocessing facilities. The modeling of pollutant transport is complicated by the complex hydraulic nature of natural streams and rivers. Chemical and physical reactions that can occur within the stream or river tend to further complicate the problem. These reactions include:

1. sorption by bed sediments and aquatic plants,
2. desorption into the flowing water,
3. ion exchange between the pollutant and some substance in the stream, and
4. particle deposition by biomass removal.

White and Gloyna (1) have described the transport of nuclides in a model river system according to the one-dimensional dispersion model, in which sorption and desorption by aquatic plants and the streambed sediments are included as sink and source effects. However, one-dimensional dispersed flow does not adequately describe the complex hydraulic mixing phenomena in most natural streams. This shortcoming of the model when applied to a real system is caused by the nonuniformity of flow of a typical stream or river cross section. Hays (2) has better approximated this behavior by dividing a stream or river cross section into two distinct zones: a main channel and a stagnant, or dead, zone. In the main channel, the dispersed flow model is applied; however, only lateral mass transfer is considered in the dead zone. Although Hays' description approximates the hydraulic behavior more accurately, a complete river model must include all chemical and physical reactions that can occur in the river.

In this study, a stream-river model was developed to treat advective and dispersive transfers along the longitudinal axis of a stream or river according to the one-dimensional dispersion model. Chemical and physical processes occurring within the stream are included in the model as source/sink effects. These processes include particle deposition, ion exchange, and sorption/desorption. Also, the dead zone model is included to better approximate stream flow conditions. A JOSHUA module LODIPS, has been written to solve the model equations under a variety of release conditions. The model is also combined with
a parameter identification algorithm to estimate stream parameters from controlled-release experiments.

THEORY

Radionuclides or other pollutants in a moving fluid stream are transported by the combined effects of turbulent diffusion and differential mass transfer. These combined effects, referred to as dispersion, can be represented in one dimension:

\[
\frac{\partial C(x,t)}{\partial t} = \frac{\partial}{\partial x} \left( D(x) \frac{\partial C(x,t)}{\partial x} \right) - \frac{\partial}{\partial x} \left( U(x) \frac{\partial C(x,t)}{\partial x} \right) + \sum_{j=1}^{J} S_j \quad (1)
\]

Boundary Conditions:
1. \( C(0,t) = r(t) \)
2. \( C(x_m,t) = C_{\infty} \)

or

\[
\left. \frac{\partial C(x,t)}{\partial x} \right|_{x = x_m} = 0
\]

Initial Condition: \( C(x,0) = 0 \)

where:

- \( C(x,t) \) = concentration of pollutant in stream or river at a point \( x \) and time \( t \)
- \( D(x) \) = dispersion coefficient at a point \( x \)
- \( U(x) \) = longitudinal transport velocity at a point \( x \)
- \( S_j \) = uptake or release of particles by a dead zone (\( j = 1 \)) or interactive phase (\( j > 1 \)) within the stream
- \( J \) = total number of source/sink effects
- \( r(t) \) = analytic function describing form of release
- \( x_m \) = point far downstream
- \( C_{\infty} \) = specified concentration

The terms in Equation (1) represent, respectively:

(I) the temporal change in concentration

(II) turbulent diffusion
(III) longitudinal convective mass transfer

(IV) source/sink effects within the stream.

The fourth term is necessary to approximate the departure of the stream flow behavior from the one-dimensional bulk dispersion model description. This departure is due to the presence of dead zones in the stream. This also represents stream-pollutant interaction through the processes of ion exchange, sorption/desorption, and particle deposition. The temporal change in an interactive phase or dead zone concentration is represented by:

\[
\frac{\partial W_j(x,t)}{\partial t} = f_j[C(x,t), W_j(x,t)]
\]  

(2)

where:

\[ W_j(x,t) = \text{concentration of pollutant in the dead zone (} j = 1 \text{) or interactive phase (} j > 1 \text{)} \]

\[ f_j[C(x,t), W_j(x,t)] = \text{a function describing the dynamic characteristics of a dead zone (} j = 1 \text{) or interactive phase (} j > 1 \text{)} \]

The dead zone model (1) assumes that the stream is made up of two distinct zones: a main channel in which transport is described by Equation (1), and a dead zone. This concept is depicted in Figure 1. The concentration of pollutant in a given dead zone cross section is assumed to be uniform; thus,

\[
f_1 = \frac{MP}{A_d} [C(x,t) - W_1(x,t)]
\]  

(3)

where:

\[ M = \text{mass transfer coefficient} \]

\[ P = \text{wetted perimeter between the dead zone and stream} \]

\[ A_d = \text{cross sectional area of the dead zone} \]

Similarly:

\[
S_1 = \frac{MP}{A_s} [W_1(x,t) - C(x,t)]
\]  

(4)
FIGURE 1. Dead Zone Representation
where:

\[ A_s = \text{cross sectional area of the stream} \]

For the interactive phases, the transfer of particles is:

\[ f_j = T_{pj} \{ g_j \{ C(x,t) \} - W_j(x,t) \} \]  \hspace{1cm} (5)

where:

\[ g_j[C(x,t)] = \text{the transfer function relating the concentration of particles in water to the equilibrium level in phase } j \]

\[ T_{pj} = \text{the transfer coefficient for phase } j \]

\[ W_j = \text{the concentration of particles in the } j^{th} \text{ phase} \]

For the processes under consideration, the equilibrium concentration of radionuclides or other pollutants has the form (2):

\[ g_j[C(x,t)] = k_j C_j(x,t) \]  \hspace{1cm} (6)

where \( k_j \) and \( n_j \) are constants pertaining to a particular radionuclide or pollutant and the \( j^{th} \) interactive phase. For most radionuclides \( n = 1 \); thus,

\[ f_j = T_{pj} [k_j C(x,t) - W_j(x,t)] \]  \hspace{1cm} (7)

Likewise:

\[ S_j = T_{sj} [W_j(x,t) - k_j C(x,t)] \]  \hspace{1cm} (8)

where:

\[ T_{sj} = \text{transfer coefficient between stream and } j^{th} \text{ phase} \]

For convenience of notation, \( f_j \) and \( S_j \) are defined in general as:

\[ f_j = T_{pj} [C(x,t) - \bar{W}_j(x,t)] \]  \hspace{1cm} (9)

\[ S_j = T_{sj} [\bar{W}_j(x,t) - C(x,t)] \]  \hspace{1cm} (10)
where:
\[
\overline{W}_j = \begin{cases} 
  W_j(x,t) & j = 1 \text{ (dead zone)} \\
  W_j(x,t)/k_j & j > 1 \text{ (interactive phase)}
\end{cases}
\]

For the dead zone:
\[
T_{pl} = \frac{MP}{A_d}
\]

and
\[
T_{sl} = \frac{MP}{A_s}
\]

**METHOD OF SOLUTION**

The method of solution of Equations (1) and (2) involves a full finite difference approximation of the space and time domains. The continuous time domain is divided into intervals \( \Delta t_1, \Delta t_2, \ldots, \Delta t_i, \ldots \) by defining discrete times \( t_0, t_1, \ldots, t_i, t_{i+1}, \ldots \) where \( t_0 = 0 \). A fully implicit (backward) difference approximation to Equation (1) is:

\[
C^{i+1}(x) - C^i(x) = h \left[ \frac{3}{3x} D(x) \frac{3}{3x} C^{i+1}(x) - \frac{3}{3x} U(x) C^{i+1}(x) \right] + h \sum_{j=1}^{J} S_i^{i+1} \quad \text{(11)}
\]

where:
\[
h = t_{i+1} - t_i
\]

Similarly, Equation (2) is:
\[
W_j^{i+1}(x) - W_j^i(x) = f_j[C^{i+1}(x), W_j^{i+1}(x)] \quad \text{(12)}
\]

Substitution of Equations (9) and (10) into Equations (11) and (12) yields, with some manipulation, the following:
Spatial difference equations are derived from Equation (13) by first considering the stream to be made up of homogeneous segments, called reaches, which are characterized by a particular set of transport and interactive properties. Each reach is further subdivided into finite increments with a mesh point centered in each increment. This geometric representation is illustrated in Figure 2. The concentration is assumed to be segmentally linear between mesh interfaces and mesh points. At the mesh interfaces, continuity of flux and concentration conditions are applied.

The resulting difference equations can be written in matrix form as:

\[
C^{i+1}(x) - C^i(x) = h \left[ \frac{\partial}{\partial x} D(x) \frac{\partial}{\partial x} C^{i+1}(x) - \frac{\partial}{\partial x} U(x) C^{i+1}(x) \right]
- \sum_{j=1}^{J} \frac{T_{sj}}{1 + h/T_{pj}} C^{i+1}(x)
+ h \sum_{j=1}^{J} \frac{T_{sj}}{1 + h/T_{pj}} W^i_j
\]

\[
W^{i+1}_j = \frac{1}{1 + h/T_{pj}} \left[ h T_{pj} C^{i+1}(x) + W^i_j(x) \right]
\]

Spatial difference equations are derived from Equation (13) by first considering the stream to be made up of homogeneous segments, called reaches, which are characterized by a particular set of transport and interactive properties. Each reach is further subdivided into finite increments with a mesh point centered in each increment. This geometric representation is illustrated in Figure 2. The concentration is assumed to be segmentally linear between mesh interfaces and mesh points. At the mesh interfaces, continuity of flux and concentration conditions are applied.

The resulting difference equations can be written in matrix form as:

\[
\mathbf{A} \cdot \mathbf{C} = \mathbf{Q}
\]

where:

\( \mathbf{A} = \) a tridiagonal matrix containing the contribution of all advective, dispersive, dead zone, and interactive terms

\( \mathbf{C} = \) column vector of average pollutant concentrations over each mesh interval

\( \mathbf{Q} = \) column vector of source terms; includes releases from dead zones and interactive phases, and contributions from the release and from the previous time step

The set of coupled-algebraic equations represented by Equation (15) is solved in LODIPS at each time step using the method of forward elimination-backward substitution (2,4). Equation (14) is used to update the dead zone and interactive phase concentrations. Release profiles can be simulated either by a time-varying function or by tabular data.
FIGURE 2. Stream Geometrical Representation
MODEL PARAMETER ESTIMATION

Experimental methods for estimating stream model parameters, such as the dispersion coefficient, are not very successful, because it is difficult to duplicate field conditions in the laboratory. This is especially true of the dispersion coefficient (1), which is very sensitive to the form of the velocity profile in the stream.

An alternative approach to direct measurement of model parameters is the system identification (model matching) technique (5,6,7,8). The objective of this technique is the minimization of the deviation between the model predicted response for the system and the experimentally measured response by varying the model parameters. The set of parameters that gives the best agreement between model and experiment is considered to be the true physical parameters of the system under study (9).

To implement this method, an error function $E(x)$ is defined:

$$E(x) = \sum_{\ell=1}^{N} \left\{ \left[ C_{E\ell}(x) - C_{\ell}(x) \right] W_{\ell}(x) \right\}^2$$

(16)

where:

$C_{E\ell}(x) =$ experimentally measured concentration at point $x$

$C_{\ell}(x) =$ concentration as predicted by the model at point $x$

$W_{\ell}(x) =$ weighting function

$\ell =$ the measurement time index

$N =$ total number of measurement times

The function $E(x)$ is, of course, non-linear and cannot be minimized according to the normal least squares procedure. A gradient search technique known as the conjugate gradient method (7,9) is employed for this minimization. The gradients required for the search are approximated by solving the model equations repetitively while varying, in turn, each of the variable parameters by a small amount. This procedure is repeated, until the function $E(x)$ is less than a specified convergence value.
RESULTS

The transport prediction and parameter estimation methods have been applied successfully in the analysis of many controlled release experiments as well as some accidental release situations. The utility of the parameter estimation method is illustrated by the following example:

Data from a USGS (10) dye study of the Savannah River were analyzed to estimate the dispersion coefficient and mean travel time. The dye study was conducted by injecting dye at an upstream location and measuring the downstream concentrations at two stations located approximately 6.4 miles apart. The time-concentration profile at each station is shown in Figure 3. Also shown in Figure 3 is the matched conservative transport model response (i.e., no chemical or physical reactions are considered). The dispersion coefficient and mean travel time derived from the non-linear least-squares fitting are, respectively, 241 ft$^2$/sec and 349 minutes.

The differences in the shapes of the curves could probably be resolved with a dead zone model if appropriate transfer coefficients were available. The parameter estimation method is concurrently being extended to include dead zone modeling as well as estimation of transfer coefficients for non-conservative constituents.

A study of an accidental release of water containing low levels of tritium as HTO from the heavy water separations facility at the Savannah River Plant (SRP) illustrates the utility of the LODIPS module. The aqueous stream flowed from a drainage ditch to Beaver Dam Creek and then to the Savannah River, as shown in Figure 4. The time-concentration profiles measured at the Highway 301 and Purysburg sampling stations are shown in Figure 5. Two LODIPS predictions of the release behavior are presented. The first was calculated using the dispersion coefficient derived from the dye study. The agreement of the predicted and measured travel times is excellent as is evident from Figure 5; however, the difference in the shape of the observed and predicted response indicates a much stronger dispersion mechanism over this segment of the river (approximately 25 miles below the dye study reach). The other curve is the LODIPS predicted response using the dispersions coefficient derived by applying the parameter estimation method to the tritium release data. The dispersion coefficient derived from this analysis was approximately ten times larger than that for the dye study. The large difference in the values
FIGURE 3. USGS Dye Study Results
of the dispersion coefficients derived from the two studies indicates a need for further studies of tributary streams and river systems through controlled release experiments to develop appropriate data for modeling both conservative and nonconservative water soluble pollutants.

CONCLUSIONS

The LODIPS module provides an accurate method for prediction of the consequences of normal and accidental pollutant releases to tributary streams and rivers. The module is very versatile since both conservative and non-conservative pollutants can be modeled. Also, the parameter estimation method incorporated in the LODIPS module provides an effective means for stream parameter identification.
FIGURE 4. Sampling Locations on the Savannah River
FIGURE 5. Results of a Tritium Release Study
REFERENCES


