International Journal of Engineering Research and Technology.
ISSN 0974-3154 Volume 6, Number 1 (2013), pp.

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Review of Mathematical Models For Transport of Radionuclides in Surface Waters

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Abstract

The environmental behaviour of radionuclides and stable elements present a challenging problem for their regular monitoring in the atmosphere and hydrosphere. These behaviours can be well understood by developing a suitable model to assess the concentration profile in different environmental compartments. More over every aquatic ecosystem is unique, and yet it is impossible to study each system in the detail necessary for case-by-case assessment of ecological threats. In this situation, quantitative mathematical models are essential to predict, to guide assessment and to direct interventions. In this paper a comprehensive review of mathematical models available for predicting radionuclide dispersion in aquatic environment (surface waters) is presented. The major types of models covered by this paper are Analytical, Box type and Numerical models available globally for the aforesaid purpose.

Keywords: Radionuclides, Mathematical model, Types of Mathematical model, Overview

1. Introduction

Predictive models are fundamental tools for forecasting the consequences of different remedial actions. The term "predictive model" is used to indicate models in which one or a few important y-variables are predicted from a few x variables that can be obtained easily from standard maps or monitoring programme. That is the goal, but

that goal is often very difficult to achieve. It is as much a challenge to develop practical models as it is to create comprehensive validated ecosystem models that predict the time-dependent interactions of many y-variables from many x-variables.
Practically useful models must satisfy some categorical features that make them reliable tools for environmental management:

- They must be characterized by a relevant and simple structure, i.e., involve the smallest possible number of driving variables;
- The values of the necessary driving variables should be easy to access and/or to measure;
- The models must be validated for a variety of circumstances showing a wide range of environmental characteristics.

In broad terms, the variables used in environmental models may be divided into two categories:

1) Variables for which site-specific data must be available, such as lake volume, mean depth, water discharge, amount of suspended particulate matter in water, etc.;
2) Variables for which generic (= general) values are used because of the lack of site-specific data, e.g., the sedimentation rate and/or rates for internal loading (like diffusion and advection rates).

The variables belonging to the first category are often called "site-specific variables", "environmental variables" or "lake-specific variables". They can generally be measured relatively easily and their experimental uncertainty should not significantly affect the overall uncertainty of the model predictions of the target variable(s). The second category, the "model variables" or "model constants", are often difficult to access for each specific system, such as the transfer rates from the sediment to the water, the deposition velocity of X from water to sediments, the migration rate from catchment to lake, etc. The model variables may contribute significantly to the model uncertainty unless they have been validated and reliable sub-models for their generic values established from critical tests.

In the aquatic environment, detailed models for transport, generally based on numerical solutions of the advection ± dispersion equations, are available, and modern computing power means are applied to a range of problems concerning radionuclide transport. In this case, however, the detailed input data that would be required are generally not available. In simple terms, the aquatic environment (particularly seas) is generally less accessible than the atmospheric environment.
In addition, significant problems exist in representing radionuclide transport on sediments; for some problems this can be the key issue in determining the impacts of radionuclides entering the aquatic environment.

In a strict sense, there is no such thing as a general ecosystem model, which works equally well for all ecosystems (of a give type) and at all scales because all models need to be tested against reliable, independent empirical data. The data used in such validations must of necessity belong to a restricted domain. At any modelling scale, the complexities of natural ecosystems always exceed the complexity and size of any model. Simplifications are always needed, and this entails problems. The

ultimate obstacle in achieving predictive power and general validity for a model is to find the most appropriate simplifications, and/or omit small and irrelevant processes related to the given target variables to be predicted ([1]-[2]Monte, 1997a,b; [3]-[4]Monte, 1996a,b; [5]-[6]Monte et al., 1995a,b; [7]Hakanson and Peters, 1995).

2. Development of Model

The main solution methods available for transport modelling involve the use of single or multiple uniformly mixed compartments, or numerical or semi-analytical solutions of the relevant transport equations. In some situations purely empirical methods can be used, where measurements provide relationships between quantities of interest, and no attempt is made to model the processes involved. Semi-analytical methods express the solutions to the equations in a closed form but may require, for example, numerical integrations to be undertaken to evaluate the quantities of interest. These methods lie somewhere between fully analytical and fully numerical solutions to the equations.

2.1: Analytical Method

A main approach for pollutant transport modelling is to rely on the fundamental advection–dispersion equation (ADE), a partial differential equation (PDE) describing the advective–diffusive mass transport in surface waters (Socolofsky and Jirka, 2005[8]; Wallis and Manson, 2004[9]; Fischer et al.,1979[10]; Guymer, 1998[11]). The ADE is widely employed to build models for pollutant transport, and it has proved to be suitable for a large number of cases (Marsili-Libelli and Giusti, 2008[12]; Rowinski et al., 2007[13]; Boxall and Guymer, 2007[14]; Jirka and Weitbrecht, 2005[15]; Pujol and Sanchez-Cabeza, (2000)[16]. Analytical solutions of the ADE have been proposed by several workers, e.g. Van Genuchten and Alves (1982)[17], and have been used in water quality modelling for many years, e.g. Runkel and Bencala (1995)[18], Zoppou and Knight (1997)[19], Socolofsky and Jirka (2005) and Dunnivant and Anders (2005)[20].

In the research presented by E.-C. Ani et al.[21] used ADE for the development of mathematical models for an instantaneous pollutant release. The model uses an analytical solution of the ADE, and is implemented in Matlab. The analytical model was developed by using the solution of the ADE for an instantaneous injection of a mass of conservative tracer (Jirka andWeitbrecht,2005[15]; Guymer,1998)[11], as given by:

$$c(x,t) = \frac{M}{A\sqrt{4\pi t D_x}} \exp\left(-\frac{(x-(x_0+V_x t))^2}{4D_x t}\right) \quad \text{----------(1)}$$

Where M is the mass of released tracer [mg]; A is the cross-sectional wetted area of the channel [m^2], and x_0 is the location of the source [m].

Many studies on different numerical models have been published. The majority concern the numerical discretisation of the ADE using finite difference, finite element or finite volume methods (e.g. Morton and Sobey, 1993[22]). Most

recently, Rubio et al. (2008)[23] developed a numerical code to simulate advective–diffusive–reactive solute transport in rivers. Other papers describe software packages, e.g. OTIS (Runkel, 1998)[24].

2.2 Single-compartment models

Simple single-compartment models are widely used for modelling the fate of radionuclides entering the marine environment close to the coast; Activity is partitioned between the dissolved and suspended sediment phases depending on an equilibrium sorption coefficient, and losses of activity from the system are due to the removal of water from the compartment, scavenging to bottom sediments, and radioactive decay.

Some models extend the simple one-compartment approach to incorporate explicitly one or more separate compartments for bottom sediments. For example, Simmonds et al. (1995)[25] employed three sediment compartments. In addition to the transfer of radionuclides from the water column to bottom sediments by sedimentation, transfer to and from sediments by diffusion and bioturbation is also included.

2.3. Semi-analytical models

Semi-analytical models lie somewhere between the simple single-compartment models and more complex numerical models. Examples of such models are those described by Maul (1986a, b)[26-27]. A key feature of the local dispersion of contaminants in the sea is the movement of the tide parallel to the coast. Over a tidal cycle, the total excursion may be 20 km or more, so that a contaminant released into the sea may travel many kilometres along the coast before being brought back past the release point. Thus, a more natural way to represent the dispersion is by movement with the tide along the coast. Instead of the usual fixed co-ordinate system, the advection ± diffusion equation is solved in a frame of reference which moves with the tide as it travels up and down the coast. Discharges made at different times in the tidal cycle will have different dispersion characteristics, and these can be represented directly in the moving coordinate system. Mathematically, the resulting solution to the dispersion problem in the moving co-ordinate system is transformed back into the fixed co-ordinate system to give estimates of average concentrations as a function of both the position along the coast and distance offshore.

The calculated concentration field depends critically on the hydrographical parameters employed, particularly the residual tide (the velocity parallel to the coast averaged over the tidal cycle). In reality, residual tides are never constant, being particularly affected by meteorological conditions. Even though this model is relatively simple compared with detailed numerical models, a large amount of site-specific data is needed to undertake a full validation of the model; such data will generally not be readily available. This approach to dispersion modelling avoids some of the known shortcomings of single-compartment models. Models of this type can be used to investigate the variability of water phase concentrations, and the sensitivity of these estimates to assumptions about hydrographic parameters.

An example of a 1-D semi-empirical model is that developed by Schaefer (1975)[28]. It is assumed that activity in the river water reduces as activity is transported downstream according to:

$$Q_w = Q_0 \exp[-kx] \quad \text{and} \quad k = k' + \frac{\lambda_r}{u_w} \qquad \text{----------(2)}$$

where Q_w is the activity passing a given point in unit time (Bq /l), u_w is the river water speed, and for a steady source, Q_0 is the source strength (Bq /1). The empirical factor k represents an effective loss to river sediments. Once the plume is well mixed across the river, the total concentration in the river water will be given by:

$$C = \frac{Q_w}{A u_w} \qquad \text{----------(3)}$$

where A is the cross-sectional area of the river. This activity will be partitioned between that in the water phase and that associated with suspended sediments. It is readily shown that at equilibrium, if the river bed sediments are moving at a speed us, the activity per unit length of river bed, Ms (Bq/m.1) is given by:

$$M_s = \frac{Q_0 k'}{(k u_s - \lambda_r)} [\exp(-\lambda_r x/u_s) - \exp(-kx)] \qquad \text{----------(4)}$$

This quantity has a maximum value at:

$$x_m = \frac{1n \frac{k u_s}{\lambda_r}}{(k - \frac{\lambda_r}{u_s})} \qquad \text{----------(5)}$$

The model can be used with a number of different sections, with river characteristics varying between sections and the output from one section providing the input to the next.

2.4. Multiple-compartment models

A number of multiple-compartment models have been developed to address various issues associated with radionuclides in the marine environment, ranging from regional scale models concerned with discharges into the sea from coastal nuclear facilities, to larger scale models concerned with the disposal of radioactive waste to the seabed. Regional scale models generally assume uniform vertical mixing, so that a 2-D array of compartments is employed, whereas models concerned with radionuclide inputs from the seabed need a full 3-D array of compartments. Depending on the application, radionuclide transport in bottom sediments may be modelled very simply (as an effective sink for losses from the water column) or in some detail (where the profile of radionuclides through the sediment column is important).

Long-term advective exchanges between water compartments occur due to slow moving residual currents. These can be represented straightforwardly, and multi-compartment models can be used to solve advection ± diffusion type problems, but the approach is limited to a single type of discretisation (upwind differencing). Different models require different choices for the distance over which the concentration gradient is calculated; this may, for example, be taken as the distance between the centers of the grids. When a significant fraction of the activity in a water compartment is associated with suspended sediment, this approach implies an inconsistency. If the suspended sediment loads are different in the two compartments, there is an implied net flux of sediment from one compartment to the other according to the direction of advective flow. The best way to represent sediment transfers between compartments in multi-compartment models is still open to debate. An example of a multi-compartment model is given by Simmonds et al. (1995), who used a set of regional compartments to represent dispersion in European waters and to calculate collective doses from discharges into those waters. This model derives from work undertaken on the MARINA project (CEC, 1990)[29]. The exchange rates between water compartments have been tuned using observed concentrations of ^{137}Cs, assumed to be conservative (i.e. not significantly sorbed onto sediments).

O.M.Zhukova (2002)[30] et al. suggested a model of transfer of radionuclides in a river system, which relies on the principle of the chamber model, for the case of hydraulically stationary and chemically equilibrium conditions of interaction of radionuclides in the systems "water−suspensions" and "water−bottom sediments." The model is based on analytical solutions of a system of equations for different conditions of ingress of radioactive contaminants into the river system: in the inlet cross section; with fallout of radiactive aerosols on the water surface; with ingress of radionuclides with surface flow from a contaminated water catchment. The model is verified using the data of radiation monitoring carried out on an experimental water catchment of the Iput river.

This case, a quasi-stationary model of migration of radionuclides under chemically equilibrium conditions of their interaction in the systems "water−suspensions" and "water−bottom sediments" with the following assumptions:

(1) The flow rate of the water in control volumes of the river is constant;
(2) The characteristic time of run-off from the water catchment is greatly in excess of the time of turnover of the water in the river;
(3) Sorption equilibrium between the water and the suspension and the water and the bottom sediments sets in instantaneously for exchange forms of the radionuclides;
(4) Equilibrium between turbidity and sedimentation takes a very short time;
(5) The flow rate of the bottom sediments in control volumes of the river is constant;
(6) The river channel is not deformed.

The assumptions made allow one to pass from the system of differential equations to a system which comprises algebraic balance equations describing the hydrological block and the block of transfer of suspended and entrained sediments under

hydraulically stationary conditions, and the differential equation of change in the concentration of radionuclides in the chambers with

$$\frac{dV_iC_i}{dt} = C_{w.enti}Q_{w.enti} + C_{r.enti}Q_{w.enti}S_{m.enti} + C_{b.enti}Q_{b.enti}\,\rho_{bi} +$$

$$+ C_{w.tri}Q_{w.tri} + C_{r.tri}Q_{w.tri}S_{m.tri} + C_{b.tri}Q_{b.tri}\,\rho_{bi} +$$

$$+ C_{w.ci}Q_{w.ci} + C_{r.ci}Q_{w.ci}S_{m.ci} + C_{w.gri}Q_{w.gri} -$$

$$- C_{ri}Q_{wi}S_{mi} - C_{bi}Q_{bi}\,\rho_{bi} - C_{wi}Q_{wi} - \lambda_d V_i C_i\,, \qquad \text{---------(6)}$$

$$C_{ri} = C_{wi}\,\rho_{bi}K_{d.ri}\,, \qquad \text{--------(7)}$$

$$C_{bi} = C_{wi}K_{d.bi}\,, \qquad \text{--------(8)}$$

$$C_i = C_{wi}R_i\,, \qquad \text{--------(9)}$$

$$R_i = R_{ri} + R_{bi}\,, \qquad \text{-------(10)}$$

$$R_{ri} = 1 + S_{mi}\,\rho_{bi}K_{d.ri}\,, \qquad \text{-------(11)}$$

$$R_{bi} = \frac{V_{bi}}{V_{wi}}(1-\Delta_i)\,K_{d.bi}\,\rho_{bi}\,. \qquad \text{--------(12)}$$

With certain initial conditions and specific conditions of ingress of radionuclides into the chambers, the differential equation (6) is solved analytically.
In this stage of investigation, we consider three cases of ingress of radioactive contamination into the river system.
I. Ingress of Radionuclides in the Inlet Cross Section of the River during a Limited or an Unlimited Period of Time. With the initial condition
$t = 0$, $C_{wi}(0) = 0$, $Cri(0) = 0$, $C_{bi}(0) = 0$ ----------(13)
and the condition of ingress of radionuclides

$$C_{ent1} = C_{0ent1}\exp(-\lambda_d t) \qquad \text{---------- (14)}$$

they obtained the analytical solution of Eq. (6)

$$C_{wi}(\mathrm{I}) = C_{0ent1}\exp(-\lambda_d t)\,A_0\prod_{j=1}^{i}\lambda_{ent1}\left[B_i - \sum_{j=1}^{i-1}B_{ij}\exp(-\lambda_j t) - B_{jj}\exp(-\lambda_i t)\right]\delta_1 \qquad \text{----(15)}$$

II. Fallout of Radionuclides on the Water Surface of the Entire River or Separate sections of It.

It is assumed that the radionuclides that entered interact with the elements of the river medium instantly. With the initial condition

$t = 0$, $C_{0i}(0) = a_{0i} / H_{mi}$ ----------*(16)*

they obtained the analytical solution of Eq. (6) in the following form:

$$C_{wi}(\text{II}) = \exp(-\lambda_d t)\left\{\sum_{j=1}^{i-1} D_{ij}\exp(-\lambda_j t) + \left(C_{0i}A_i - \sum_{j=1}^{i-1} D_{ij}\right)\exp(-\lambda_i t)\right\}. \quad \text{---------(17)}$$

III. Ingress of Radionuclides into the River System with Surface Flow from the Contaminated Water Catchment (or Separate Sections of It).

To determine the amount of radioactive contamination entering the river system from the surface of the water catchment, we write the equation of balance of the radionuclides on the *i*th section of the water catchment:

$$F_c h_c \frac{dC_s}{dt} = -C_{w.s}Q_{w.c} - C_{r.s}Q_{w.c}S_{m.c} - C_{w.s}U\alpha F_c - \lambda_d F_c h_c C_s, \quad \text{----------(18)}$$

$$C_s = C_{w.s}\theta_s R_s, \quad \text{----------(19)}$$

$$R_s = \left[1 + K_{d.s}\frac{\rho_s(1-\mu_s)}{\theta_s}\right], \quad \text{----------(20)}$$

$$C_{r.s} = C_{w.s}K_{d.s}\rho_s. \quad \text{----------(21)}$$

Integrating Eq. (18), we obtain the solution for the *i*th section of the water catchment:

$$C_{w.si} - \frac{C_{s0i}}{\theta_{si}R_{si}}\exp(-(\lambda_{wi} + \lambda_d)t). \quad \text{----------(22)}$$

With the initial condition (13) and the condition of ingress of radionuclides with surface flow from the water catchment(22), we found the solution of Eq. (6) for the i^{th} chamber in the following form:

$$C_{wi}(\text{III}) = \exp(-\lambda_d t)\left[w_i\exp(-\lambda_{wi}t) + \sum_{j-1}^{i-1} w_i\exp(-\lambda_{wi}t) - \sum_{j-1}^{i-1} E_{ij}\exp(-\lambda_j t) - E_{ii}\exp(-\lambda_i t)\right], \quad \text{-------(23)}$$

$$w_i = \frac{C_{s0i}}{\theta_{si}R_{si}}\frac{\lambda_{ci}}{\lambda_i - \lambda_{wi}}. \quad \text{-------(24)}$$

Above equations are the main part of the mathematical model describing migration of radionuclides in the river system.

Another famous application of Multi Compartment Model is shown by Luigi Monte et al., who showed that the most important hydrological processes occurring in

a lacustrine system that influences the behaviour of radionuclides in the lake is obviously the outflow of water from the outlet. Such a process is, indeed, responsible of the removal of radionuclides from the water body. The process is usually modelled according to the following formula:

$$\Phi_r = \Phi C_w \quad \text{----------(25)}$$

where Φ_r is the flux of radionuclide (Bq s^{-1}) removed by the outlet, Φ is the outlet flux (m^3 s^{-1}) and *C*w is the radionuclide concentration in the lake water (Bq m^{-3}). The other processes of radionuclide migration involve the complex interaction of dissolved radionuclide in water with suspended particles and bottom sediments. ^{137}Cs-ECOPRAQ, ^{90}Sr-ECOPRAQ, ^{90}Sr-MOIRA, ^{137}Cs-MARTE and LAKECO are typical first-order models. They comprise three or four compartments corresponding to the radionuclide in the lake water and in two or three layers of bottom sediment. The structures of the above models do not show substantial differences. The models developed in the frame of the AQUASCOPE project are substantially similar to the other box models. AQUASCOPE models are indeed based on the evaluation of the response of water contamination to a single pulse deposition input of radionuclide.

The radionuclide concentration in water at instant t following a single pulse deposition event at instant t is

$$C_w(t) = DG(t-\tau) \quad \text{----------(26)}$$

where $G(t-\tau)$ is the response to a deposition pulse of 1 Bq m^{-2} and D (Bq m^{-2}) is the radionuclide deposition per square meter. The radionuclide concentration $C_w(t)$ for deposition processes depending on time ($D(t)$ = radionuclide deposition rate Bq m^{-2} s^{-1}) is:

$$C_w(t) = \int_0^t D(\tau)G(t-\tau)d\tau. \quad \text{----------(27)}$$

It is well known that any linear model such as ^{137}Cs-ECOPRAQ, ^{90}Sr-ECOPRAQ, ^{90}Sr-MOIRA, ^{137}Cs-MARTE and LAKECO is characterised by a function $G(t-\tau)$ that allows one to evaluate the radionuclide concentration in water (or in any other target variable) by Eq. (3). From now on we will call $G(t-\tau)$ the Green Function (GF) of the model. It is instructive to start our analysis by considering ^{90}Sr behaviour in the water-sediment sub-system of a lake. We compare, for instance, the model ^{90}Sr-AQUASCOPE with the ^{90}Sr-MOIRA model.

The ^{90}Sr-MOIRA model for predicting the migration of radionuclide from water to sediments is composed of two active boxes (Fig. 1):

1. radionuclide dissolved in water (Water, C_w, Bq m^{-3});
2. radionuclide deposited in sediment (Bottom sediment, D_s, Bq m^{-2});

and a ‘passive box’ (Deep sediment) representing the radionuclide subject to nonreversible removal processes from the active deposit. The equations controlling the radionuclide migration processes are the following:

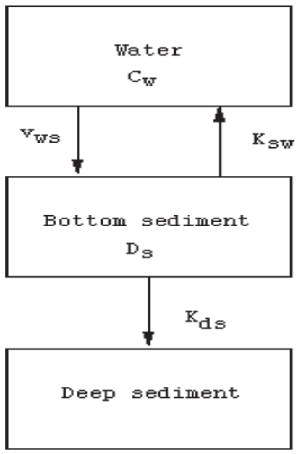

Fig. 1. Structure of the ^{90}Sr-MOIRA box sub-model for predicting ^{90}Sr migration from water to sediments (internal process)

$$\frac{dC_w}{dt} = -\frac{v_{ws}}{h}C_w + \frac{K_{sw}}{h}D_s - \lambda_r C_w \qquad \text{----------(28)}$$

$$\frac{dD_s}{dt} = v_{ws}C_w - (K_{sw} + K_{ds})D_s - \lambda_r D_s \qquad \text{---------- (29)}$$

where h is the average depth (m) of the lake, v_{ws} is the migration velocity (m s^{-1}) of radionuclide to the bottom sediment, K_{sw} is the migration rate (s^{-1}) from bottom sediment to water, K_{ds} is the removal rate (s^{-1}) of radionuclide from the bottom sediment, λ_r is the radioactive decay constant (s^{-1}) and t is the time (s). The solution of the previous equation system, following a deposition pulse D (Bq m^{-2}) at time 0 (GF of the MOIRA model), is

$$C_w(t) = \frac{D}{h}\frac{\left(\lambda_2 - \frac{v_{ws}}{h}\right)}{(\lambda_2 - \lambda_1)}e^{-(\lambda_1+\lambda_r)t} + \frac{D}{h}\frac{\left(\frac{v_{ws}}{h} - \lambda_1\right)}{(\lambda_2 - \lambda_1)}e^{-(\lambda_2+\lambda_r)t} \qquad \text{------- (30)}$$

where λ_1 and λ_2 are as follows:

$$\lambda_{1,2} = -\frac{-\left(\frac{v_{ws}}{h} + K_{sw} + K_{ds}\right) \pm \sqrt{\left(\frac{v_{ws}}{h} + K_{sw} + K_{ds}\right)^2 - 4\frac{v_{ws}K_{ds}}{h}}}{2}. \qquad \text{----------(31)}$$

^{90}Sr-AQUASCOPE predicts ^{90}Sr concentration in the water of so-called “closed” lakes by the following equation for a pulse deposition input at initial time t = 0.

$$C(t) = Ae^{-(K+\lambda_r)t} + D\eta e^{-(g+\lambda_r)t} \qquad \text{----------(32)}$$

where K, g and h are empirical parameters

The model LAKECO makes use of the following equations for predicting the rate constants of the migration processes:

a) Migration from water to top sediment layer:

$$k_{ws} = \left(\frac{D_m}{d_{s1}h} + \frac{\varepsilon R_w \phi_1}{h} + \frac{R_w \rho_1 K_{d1}(1 \quad \phi_1)}{h} + \frac{\sigma K_{dw}}{h}\right)\frac{1}{1 + K_{dw}L}. \quad \text{----------(33)}$$

The four terms on the right side of the previous equation are:

- The rate constant of diffusion from water to sediment pore water;
- The rate constant for radionuclide transfer from surface water to pore water in sediment due to physical mixing and bioturbation;
- The rate constant for radionuclide transfer from the water column to top sediment layer due to physical mixing and bioturbation;

b) Migration from top sediment to water column:

$$k_{sw} = \left(\frac{D_m}{d_{s1}^2\phi_1} + \varepsilon\frac{R_w}{d_{s1}}\right)\frac{\phi_1}{\phi_1 + K_{d1}\rho_1(1-\phi_1)} + \frac{R_w}{d_{s1}}\frac{K_{d1}\rho_1(1-\phi_1)}{\phi_1 + K_{d1}\rho_1(1-\phi_1)}. \quad \text{----------(34)}$$

The terms on the right side are:

- The rate constant for the radionuclide diffusion from sediment to water;
- The rate constant for radionuclide transfer from sediment porewater to water column

due to physical mixing and bioturbation;

- The rate constant for radionuclide transfer from top sediment layer to the water column due to physical mixing and bioturbation.

c) Migration from the top sediment layer to the deep sediment layer:

$$k_{s1s2} = \frac{D_m}{d_{s1}d_{s2}\phi_1}\frac{\phi_1}{\phi_1 + K_{d1}\rho_1(1-\phi_1)} + \frac{\sigma}{\rho_1(1-\phi_1)d_{s1}}\frac{K_{d1}\rho_1(1-\phi_1)}{\phi_1 + K_{d1}\rho_1(1-\phi_1)}. \quad \text{----------(35)}$$

The terms on the right side are:

- The rate constant for diffusion from the top to the deep sediment layer;
- The rate constant for the migration from the top to the deep sediment layer due to burial mechanisms.

d) Migration from the deep to the top sediment layer:

$$k_{s2s1} = \frac{D_m}{d_{s2}^2\phi_2}\frac{\phi_2}{\phi_2 + K_{d2}\rho_2(1-\phi_2)}. \quad \text{----------(36)}$$

e) Finally, radionuclide burial from the deep sediment layer:

$$k_{s2->} = \frac{\sigma}{\rho_2(1-\phi_2)d_{s2}}\frac{K_{d2}\rho_2(1-\phi_2)}{\phi_2 + K_{d2}\rho_2(1-\phi_2)}. \quad \text{----------(37)}$$

The five radionuclide fluxes of LAKECO model are controlled by the five aggregated rate constants k_{ws}, k_{sw}, k_{s1s2}, k_{s2s1} and k_{s2}. The model structure corresponds to a three components exponential the radionuclide concentration in water following a pulse event of contamination. The model calculates the values of the rate constants by 14 primary parameters most of which are related to fundamental processes like the molecular diffusion of the radionuclide through water and the interaction of the radionuclide in dissolved form with sediment particles. In principle the reliability of

the model is strictly dependent on the accuracy of the values of these primary parameters. LAKECO is “fundamental process specific” as it relates the behaviour of radionuclides in the environment to specific physical and chemical fundamental processes. LAKECO is a so called “reductionistic model”. In other words, it includes, at least in principle, as many relevant details as reasonably possible by modelling them according to primary laws from fundamental disciplines such as physics and chemistry.

On the contrary, MARTE, AQUASCOPE and ECOPRAQ models are based on a holistic approach. They are “environmental process specific”, that is, they relate the behaviour of radionuclides in the environmental systems to relevant *environmental* characteristics and processes. As such they aggregate a great deal of elementary, fundamental processes of physical, chemical, geochemical, biological etc. nature.

2.5. Numerical models

Many numerical models have a hydrodynamic module to calculate water velocities and depths as a function of time through the tidal cycle. Generally, 2-D equations are used, assuming no variations in the vertical plane. There are equations for continuity and for conservation of momentum. The latter depend upon such factors as Coriolis forces, the dispersion of momentum, and wind shear stress at the surface. There are also terms which allow for the input of momentum at the boundaries. CSERAM (Aldridge, 1998)[31] is an example of a recently developed sediment and contaminant transport model. It is based on similar principles to the commercially available MIKE21 (Danish Hydraulics Institute, 1994)[32] code. The hydrodynamic model computes water velocities for the region under consideration, typically at half-hour intervals. These water velocities are used as inputs to the numerical transport model with time steps of 15 min, typically. In addition, a wind wave model has been developed for situations where significant sediment resuspension can take place. For a 2-D depth-averaged form of the model, modern computing power enables long-term simulations (several years) to be undertaken.

Improved computing power now makes such modelling a viable option, provided that suitable data are available. In practice, complex numerical models are very `data hungry', and information on variations in model parameters on the grid distance scales used may not be available. Such models can be seen to be extremely powerful, and one can anticipate the increased use of commercially available packages for environmental modelling, but a proper understanding of the impacts on model calculations of limitations on data availability is needed to avoid inappropriate conclusions.

The numerical model was developed by E.-C. Ani et al.[21] in the Chemical Engineering Module of COMSOL Multiphysics by using the 1D form of the convection and diffusion terms. Starting from this predefined form of the PDE it was possible to develop the model at the desired level of complexity. The source was introduced in COMSOL as an impulse signal, to represent the instantaneous discharge of a known mass of tracer at the beginning of the simulation. A smoothed switch function was used in order to avoid discontinuity problems associated with the

computation of the Dirac signal. The Dirac signal is computed as being a continuous signal during a very short period of time.

3. Summary

Many models are available to calculate the dispersion in surface waters of material originating from routine discharges and accidental releases. The three basic groups of models are the following:

(1) Numerical models transform the basic equations of radionuclide dispersion into finite difference or finite element form. Such models permit most of the relevant physical phenomena to be taken into account in the analysis.

(2) Box type models treat the entire body of water, or sections thereof, as composed of homogeneous compartments. In this type of model, average concentrations are computed for each compartment and transfer constants are set up to relate the variables for one compartment to those in adjacent compartments. Most models dealing with the interactions between radionuclides and sediment are of this type.

(3) Analytical models solve the basic equations describing radionuclide transport with major simplifications made for the geometry of the water body and the dispersion coefficients. This group of models is the one most frequently used in surface hydrological analysis.

In addition, Monte Carlo methods may be used to model water body geometry and to simulate particles.

The selection of a model should be based on the type of discharge (surface or submerged), the type of water body (river, estuary, impoundment, large lake or ocean) and the use being made of the water. The magnitude of the source term under normal operation and potential accident conditions, the required accuracy and the type of water affected should be considered in the selection of the model.

The results from a calculation model should be compared with laboratory data or field data for a specific site. Such validation usually has a limited range of applicability, which should be determined with a full understanding of the model.

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