

# $^{137}\text{Cs}$ : PARAMETRIC ANALYSIS OF THE DARCY VELOCITY INFLUENCE ON THE CONTAMINANT CONCENTRATION AT RECEPTOR WELL

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## ABSTRACT

Safety analysis of nuclear installations involves the study of scenarios related to the release of radionuclides in the environment and their subsequent transport to a critical group of population. If transport in air and water is solved by the knowledge of few parameters easily measurable, groundwater transport is more complicated for the difficulty in measuring or finding the parameters involved in bibliography. At the current state of the art, groundwater calculations are usually performed by software platforms such as FRAMES 1.6. Correct utilization of FRAMES 1.6 as regards groundwater involves the knowledge of parameters such as soil absorption coefficients, bulk density, soil class and Darcy velocity. The latter is one of the most important parameter for simulation. It depends on permeability, dynamic viscosity and porosity of the soil. This work presents a parametric analysis on the influence of Darcy velocity for a case of radioactivity release in groundwater. The study is intended to give the safety analyst an instrument to device figures, for instance minimum–maximum approximations, on radionuclide transport in groundwater.

*Keywords:* aquifer, Darcy velocity, groundwater, radioactive contaminant, safety analysis, vadose zone.

## 1 INTRODUCTION

In safety analysis of nuclear installations, software platform such as FRAMES 1.6 is used by analysts to simulate transport and fate of radioactive contaminants in environment. Concentration in water and air, effective dose and risk are calculated using this platform. Groundwater transport and fate of radioactive contaminant are influenced by various environmental parameters, such as soil bulk density, soil class, soil porosity, Darcy velocity; by contaminant parameters, such as half life and water solubility; and by partition coefficient [1, 2].

In this paper, influence of Darcy velocity on groundwater MEPAS (a FRAMES 1.6 module) models has been studied. FRAMES 1.6 is used by government agencies, regulators and nuclear companies [3–11]. MEPAS modules have been created by a cooperation between EPA, DOE and Battelle Institute [12]. The groundwater model accounts for the major mechanism of constituent mobility, persistence, advection and hydrodynamic dispersion. Advection is described by constant, unidirectional flow in the vertical direction of the vadose zone and in the longitudinal direction of the aquifer [12, 13]. The velocity of this flow is described by Darcy velocity. Birdsell *et al.* [14] underline the importance of water flux in groundwater simulations. In this paper,  $^{137}\text{Cs}$  [15] release has been simulated and a parametric study has been performed to study how Darcy velocity influences concentration at receptor wells and the time at which maximum concentration is reached. The purpose of this paper is to go more in depth in knowledge of MEPAS model and to develop an instrument to evaluate, during plant design and management, the influence of Darcy velocity on  $^{137}\text{Cs}$  propagation and, consequently, to establish the accuracy of Darcy velocity evaluation for the assessment, that involves spending of time, human and economic resources.

## 2 THE METHOD

Calculations have been carried out by FRAMES 1.6, an object oriented software platform, developed by DOE, EPA and Battelle Institute [12], capable to integrate different codes both for radioprotection and for transport and fate calculations. In particular, having referred the study to the common geological situation of a vadose zone which separates the land surface from the saturated zone, the modules 'MEPAS Vadose' and 'MEPAS Aquifer' [42] have been utilized.

### 2.1 Transport phenomena in the unsaturated zone

Calculations are carried out by means of 'MEPAS Vadose' [16] module, which calculates radionuclides (or any other contaminants, such as chemicals) transport and fate in the unsaturated zone by means of the monodimensional advective–dispersive equation under the following hypotheses:

- at the beginning of transport process vadose zone is considered free from all kinds of contaminants;
- vadose zone is considered homogeneous and isotropic;
- water flow in the vadose zone is uniformly distributed in geometric space and stationary in time;
- vadose zone thickness is finite and constant;
- contaminants adsorption by the soil is described by means of the parameter  $K_d$  [1, 2] only;
- only longitudinal dispersivity is considered for each contaminant.

### 2.2 Transport phenomena in the saturated zone

'MEPAS Aquifer' [16] calculates radionuclides (or any other chemical constituents) transport in the saturated zone by means of the tridimensional advective–dispersive equation [16] under the following hypotheses:

- at the beginning of transport process, saturated zone is considered free from all kinds of contaminants;
- saturated zone is considered homogeneous and isotropic;
- water flow in the saturated zone is spatially and uniformly distributed;
- thickness of saturated zone is finite and constant while its width is boundless;
- in the time domain, the water flow of saturated zone is in steady-state conditions: whichever transitory effects, such as thickness variations due to piezometric wells are not taken into account;
- adsorption of contaminants by the soil is described by means of the parameter  $K_d$  [1, 2] only.

### 2.3 Mathematical models: advective–dispersive equations

The advective–dispersive equation [16] for solute movement through a porous medium with a constant, steady-state flow velocity forms the basis of all groundwater solution algorithms. The algorithms are developed for the limiting case of unidirectional advective transport with three-dimensional dispersion in a homogeneous aquifer.

Let  $n$  and  $n_e$  represent total and effective porosities, respectively; then  $n - n_e$  is the remaining void fraction devoted to immobile pore water. A mass balance on the differential volume  $dV = dx \, dy \, dz$  gives the expression:

$$\begin{aligned} n_e \frac{\partial C}{\partial t} + (n - n_e) \frac{\partial G}{\partial t} + \rho_s (1 - n) \frac{\partial P}{\partial t} + n_e u \frac{\partial C}{\partial x} = n_e \left[ D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2} + D_z \frac{\partial^2 C}{\partial z^2} \right] + \\ + (n - n_e) \left[ D'_x \frac{\partial^2 G}{\partial x^2} + D'_y \frac{\partial^2 G}{\partial y^2} + D'_z \frac{\partial^2 G}{\partial z^2} \right] - n_e \lambda C - (1 - n) \rho_s \lambda P - (n - n_e) \lambda G \end{aligned} \quad (1)$$

where  $n$  = total porosity (dimensionless);  $n_e$  = effective porosity (dimensionless);  $C$  = dissolved concentration in the mobile liquid phase (g/mL or Ci/mL);  $\rho_s$  = soil particle density (g/cm<sup>3</sup>);  $t$  = time (s);  $G$  = dissolved concentration in the immobile liquid phase (g/mL or Ci/mL);  $P$  = particulate concentration on the solid phase (g/g or Ci/g);  $u$  = the x-component groundwater velocity (also referred to as pore-water, seepage, or average linear velocity) (cm/s);  $D_x, D_y, D_z$  = the dispersion coefficients for the mobile liquid phase in the x-, y-, and z-directions, respectively (cm<sup>2</sup>/s);  $\Lambda$  = decay constant.

The previous equation can be streamlined with two simplifying assumptions. First, there is no diffusion of contaminant into the immobile liquid phase. Field studies indicate that including this phenomenon produces inappropriate analysis, as the retardation factor can never approach unity when the total porosity does not equal to the effective porosity. Second, the contaminant sorption process can be described by a constant ( $K_d$ ) representing the ratio between the contaminant adsorbed to the soil matrix ( $P$ ) and the contaminant dissolved in solution ( $C$ ). Using these assumptions, previous equation can be rewritten as

$$\frac{\partial C}{\partial t} + \frac{u}{R_{f1}} \frac{\partial C}{\partial x} = \frac{D_x}{R_{f1}} \frac{\partial^2 C}{\partial x^2} + \frac{D_y}{R_{f1}} \frac{\partial^2 C}{\partial y^2} + \frac{D_z}{R_{f1}} \frac{\partial^2 C}{\partial z^2} - \lambda C \quad (2)$$

in which

$$R_{f1} = 1 + \frac{\beta_d}{n_e} K_d \quad (3)$$

and

$$D = \alpha u + D_{mol} \quad (4)$$

where  $R_{f1}$  = retardation factor (dimensionless);  $\beta_d$  = bulk density (g/cm<sup>3</sup>);  $K_d$  = equilibrium (partition or distribution) coefficient (ml/g);  $\alpha$  = dispersivity in the x-, y-, or z-direction (cm);  $D_{mol}$  = molecular diffusion.

### 3 ADOPTED SCENARIO FOR RADIOACTIVE RELEASE

A radioactive flow is released through a  $0.5 \times 0.5$  m<sup>2</sup> crack in a tank of a nuclear installation. Radioactivity release is  $7.5 \times 10^6$  Bq of <sup>137</sup>Cs diluted in 3 m<sup>3</sup> of water (2.5 Bq/ml) per year, the time length of the release is 4 years. The contaminated solution enters the vadose zone (30 m thick) and moves vertically towards the interface with the saturated zone (Fig. 1). It is assumed to deal with sand soil. Once in the saturated zone, which is 10 m thick, the flow moves in the three directions according to eqn (2) [16] assuming a plume of Gaussian concentrations.

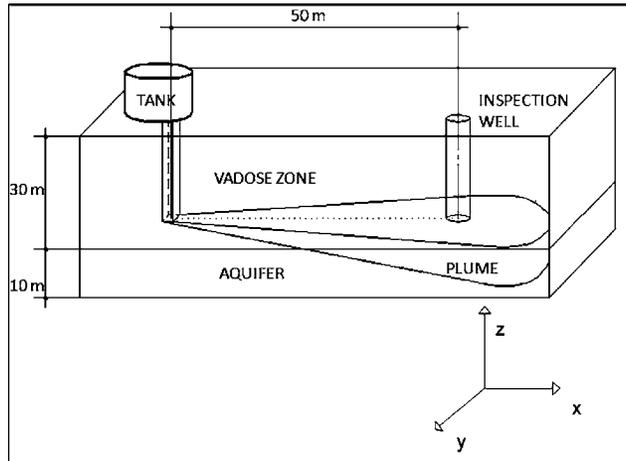


Figure 1: Environmental scenario in which  $^{137}\text{Cs}$  release happens.

Table 1: Representative sand soil characteristics.

Property	Value	Unit
Percent of sand	92	%
Percent of silt	5	%
Percent of clay	3	%
Saturated hydraulic conductivity	6.6E-03	cm/s
Representative root-zone depth	73	cm
Porosity	38	%
Dry bulk density	1.64	g/cm <sup>3</sup>
Field capacity	9	%
Soil-type coefficient	4.05	–

Note: Based on Davis and DeWiest, 1966 [17]; Freeze and Cherry, 1979 [18]; Harr, 1962 [19]; Lane and Nyhan, 1984 [20]; Linsley, Kohler, and Paulus, 1975 [21]; Mills *et al.*, [22]; MEPAS On-line Handbook [23].

At 50 m from the emission point, at phreatic surface,  $^{137}\text{Cs}$  concentration is measured.

The most important characteristics of the soil and of the contaminants are reported in Tables 1 and 2.

The adsorption coefficient has been assumed equal to 25.5 ml/g, which is the mean of the values reported in FRAMES 1.6 database. As regards dispersivity parameters, a value of 3 m has been assumed for longitudinal dispersivity in the unsaturated zone, while for the saturated zone longitudinal dispersivity has been assumed equal to 5 m, transverse dispersivity 1.65 m and vertical dispersivity for well equal to 0.0125 m. For  $^{137}\text{Cs}$  solubility in water has been considered a very high value.

Table 2:  $^{137}\text{Cs}$  properties.

Property	Value	Unit
Molecular weight	137	g/mole
Decay half life in groundwater	11,000	days

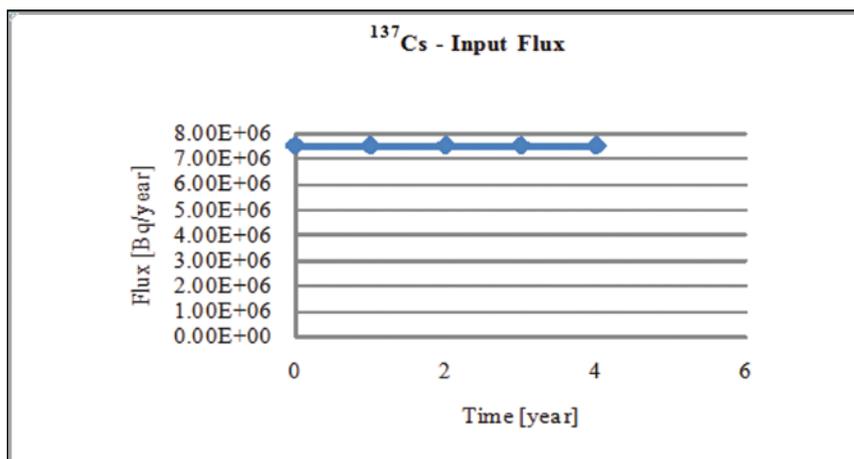


Figure 2: Input flow as calculated by FRAMES 1.6.

#### 4 RESULTS AND DISCUSSION

Two sets of simulations have been carried out, one for the calculation of the maximum concentration and the other one for the time of maximum concentration. Every set has considered eight values of Darcy velocity, ranging from 10 to 110 cm/day.

In every run, FRAMES calculates the input flow as reported in Fig. 2 and produces an output like that of Fig. 3 (Fig. 3 is the case with Darcy velocity  $V_d = 10$  cm/day, the other calculation with different values of  $V_d$  has produced similar outputs).

In the case of Fig. 3, a maximum concentration of about  $1.4 \times 10^{-4}$  Bq is reached after 50 years from emission.

##### 4.1 First set of simulations – maximum concentration value

The first set of simulations has been carried out considering eight values of Darcy velocity, ranging from 10 to 110 cm/day. Results are reported in Table 3 and are given also in terms of Max Concentration %, which represents the ratio of the calculated concentration with the original concentration of the flow at the point of emission.

Figure 4 shows that for small values of Darcy velocity concentrations grow up. This happens because when velocity grows up the influence of radioactive decay becomes less important. After Darcy velocity about 50 cm/day the effects of advection and dispersion predominate and concentrations decrease. For a Darcy velocity of 50 cm/day the concentration at the well is about 1/100 of the concentration at the point of release.

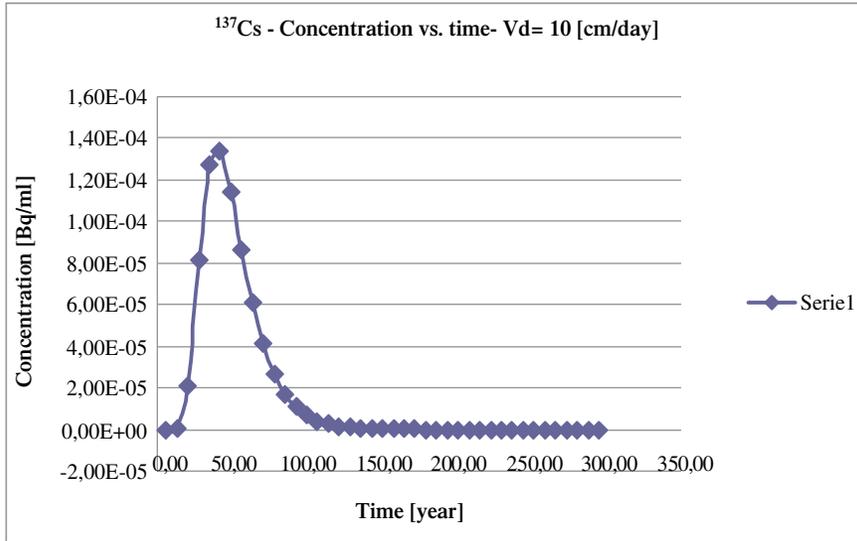


Figure 3: <sup>137</sup>Cs concentration in the well versus time.

Table 3: Maximum concentration of <sup>137</sup>Cs versus Darcy velocity. Max concentration % represents the percentage ratio with the original concentration of the flow in the point of emission.

Darcy velocity [cm/day]	Max concentration [Bq/ml]	Max concentration [%]
10	1.33E-04	5.34E-03
20	2.05E-04	8.20E-03
30	2.39E-04	9.54E-03
40	2.57E-04	1.03E-02
50	2.68E-04	1.07E-02
75	2.68E-04	1.07E-02
100	2.56E-04	1.02E-02
110	2.53E-04	1.01E-02

4.2 Second set of simulation – maximum concentration time

This set of simulations calculated the maximum concentration time versus Darcy velocity. Also in this case eight values of Darcy velocity, ranging from 10 to 110 cm/day were considered. For this second set of simulations, every run, for a given value of Darcy velocity, outputs a curve like the concentration curve of Fig. 3. Table 4 and Fig. 5 are obtained by considering the maximum of the concentration curve for every run.

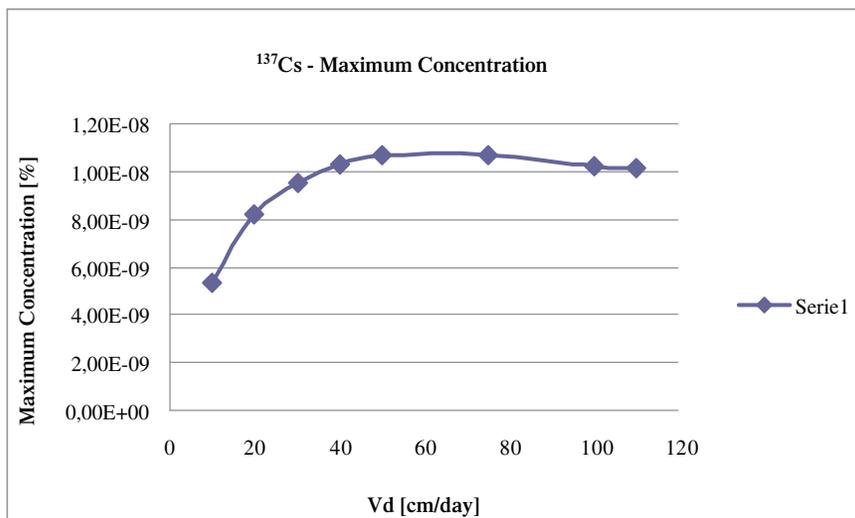


Figure 4: Ratio  $\times 100$  of the  $^{137}\text{Cs}$  concentrations in the well and at the emission point versus Darcy velocity.

Table 4: Maximum concentration time versus Darcy velocity.

Vd [cm/day]	Maximum concentration time [years]
10	42.02387
20	24.6874
30	17.72108
40	13.94367
50	11.56856
75	8.265905
100	7.731142
110	7.171494

#### 4.3 Discussion

As shown in Table 3, when Darcy velocity grows from 10 to 110 cm/day, the contaminant concentration remains confined within a variation not greater than 100%. Such a result may suggest to the safety analyst when there is the need of a precise measurement of the Darcy velocity or when a conservative assumption is enough. For instance, having foreseen a particular scenario, and having assumed direct proportionality of constituent concentration versus effective dose to population, if 100% variation of the effective dose to the population remains far below the limit of no radiological significance there is no need to determine Darcy velocity with greater accuracy. From a practical point of view such a result represents

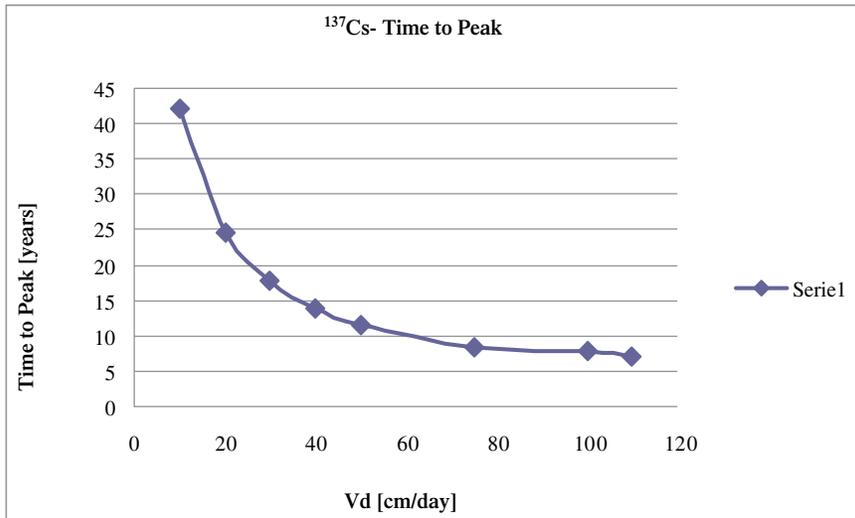


Figure 5: Maximum concentration time versus Darcy velocity.

a great simplification, because correct determination of Darcy velocity is costly and time consuming and, due to possible variations of soil characteristics versus time, it should be repeated periodically during the operative life of the installation.

As regards the maximum concentration time, as shown in Table 4, high values of Darcy velocity produce time delays not smaller than some years; for low values of Darcy velocity time delay is even 40 years. This result could be significant to program environmental measurement, especially if maximum concentration is a little greater than the minimum detectable line of instruments: knowledge of maximum concentration time at a certain distance from the source allows to detect the presence of contaminant and demonstrate the presence of a leak in the containment that otherwise will be unobserved.

## 5 CONCLUSIONS

The present work is intended to show the potentialities offered by the software platform FRAMES 1.6 to the safety analyst and show how Darcy velocity influences calculations of concentration at well (and dose calculation consequently) when MEPAS modules are used. This study could be an instrument for the safety analyst that can help him during the screening process, so he can decide about Darcy velocity accuracy. It is important to emphasize that sometimes performing parametric calculations may overcome practical difficulties, cost and time related to the knowledge environmental parameters. FRAMES 1.6 allows to calculate the concentration at a receptor well even after a periods of time that are not compatible with measures. On the other hand, MEPAS model hypothesis in par. 2.2 shall be considered: in some cases the model cannot be a good representation of real world, because soil structure is complex and soil properties are unhomogeneous and unisotropic. In those cases the model can be used only if the result of simulation is conservative. In every case of study, soil parameter shall be evaluated by a multidisciplinary team, to evaluate soil parameters and approximate in the best way the contaminant migration; an uncertainty analysis on parameters shall be performed [24].

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