MODELLING OF RADIONUCLIDE MIGRATION THROUGH THE GEOSPHERE WITH RADIAL BASIS FUNCTION METHOD AND GEOSTATISTICS

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ABSTRACT

The modelling of radionuclide transport through the geosphere is necessary in the safety assessment of repositories for radioactive waste. A number of key geosphere processes need to be considered when predicting the movement of radionuclides through the geosphere. The most important input data are obtained from field measurements, which are not available for all regions of interest. For example, the hydraulic conductivity, as input parameter, varies from place to place. In such cases geostatistical science offers a variety of spatial estimation procedures. To assess the long term safety of a radioactive waste disposal system, mathematical models are used to describe the complicated groundwater flow, chemistry and potential radionuclide migration through geological formations. The numerical solution of partial differential equations (PDEs) has usually been obtained by finite difference methods (FDM), finite element methods (FEM), or finite volume methods (FVM). Kansa introduced the concept of solving PDEs using radial basis functions (RBFs) for hyperbolic, parabolic and elliptic PDEs. The aim of this study was to present a relatively new approach to the modelling of radionuclide migration through the geosphere using radial basis functions methods and to determine the average and sample variance of radionuclide concentration with regard to spatial variability of hydraulic conductivity modelled by a geostatistical approach. We will also explore residual errors and their influence on optimal shape parameters.

Key Words: radionuclide migration, porous media, partial differential equation, radial basis function, numerical solution, Kansa method, geostatistics.

I. INTRODUCTION

Waste disposal has become a key issue in these environmentally conscious times (Chapman and McKinley, 1989). The objective of geological disposal of radioactive waste is to remove it from man's environment and ensure that any releases remain within accepted limits. Extensive research and

development in the field of management and disposal of radioactive waste is conducted in many countries. To improve the understanding of various strategies for radionuclide transport modelling, an international cooperation project was set up with the participation of a number of organisations active in waste management research. Within the project INTRACOIN (1986), a comparison has been made between different computational codes describing transport of radionuclides in geologic media. In Slovenia, two disposal concepts/siting options are currently being considered for a facility: a surface vault disposal facility; and an underground (tunnel) disposal facility (ARAO, 1999).

The modelling of radionuclide transport through the geosphere is necessary in the safety assessment

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of repositories for radioactive waste. Confidence in a model may be gained from its ability to fit dynamic laboratory and field experiments, which can differ in scale from a few centimetres to tens of metres. Assessment of the release and the transport of long-lived radioactive nuclides from the repository to the biological environment is an important part of the safety analysis of repository concepts. In this assessment, mathematical models describing the mechanisms involved in nuclide transport from the repository to the biosphere are essential tools.

When modelling flow and contaminant transport in the geosphere, it is important to consider both internal processes (e.g. advection, dispersion, retardation) within the geosphere, and external processes associated with the near-field and the biosphere. For example, near-field processes can influence water flow and chemistry in the geosphere surrounding the disposal facility, whilst biosphere processes such as flooding, erosion, weathering, recharge and environmental change all can have an impact on the geosphere (SKI and NEA, 1991).

The general reliability and accuracy of transport modelling depend predominantly on input data such as hydraulic conductivity, water velocity on the boundary, radioactive inventory, and hydrodynamic dispersion. The output data are concentration, pressure, etc. The most important input data are obtained from field measurement, which are not available for all regions of interest. For example, hydraulic conductivity as an input parameter varies from place to place. In such cases geostatistical science offers a variety of spatial estimation procedures (Deutsch and Journel, 1998).

The numerical solution of partial differential equations has been usually obtained by either finite difference methods (FDM), finite element methods (FEM), finite volume methods (FVM), or boundary elements methods (BEM) (Power and Barrac, 2002). These methods require a mesh to support the localized approximations. The construction of a mesh in two or more dimensions is a nontrivial problem. Usually, in practice, only low-order approximations are employed resulting in a continuous approximation of the function across the mesh but not its partial derivatives. The discontinuity of the approximation of the derivative can adversely effect the stability of the solution. While higher-order schemes are necessary for more accurate approximations of the spatial derivatives, they usually involve additional computational cost (Power and Barrac, 2002).

A fairly new approach for solving PDEs is through radial basis functions. Kansa (1990a; 1990b) introduced the concept of solving PDEs using radial basis functions for hyperbolic, parabolic and elliptic PDEs. A key feature of the RBF method is that does not require a grid. The only geometric properties that are used in the RBF approximation are the pair wise distances between points. Distances are easy to compute in any number of spatial dimensions, so working in higher dimensions does not increase the difficulty.

The numerical methods are developed both with regard to efficiency and ability to solve a wider variety of problems. A high efficiency is necessary to be able to solve physically complicated problems in two or three dimensions. The most common present methods often suffer the drawback that they require fine discriminations to solve predominantly advective problems. In the conclusions of the INTRACOIN project it was reported that there are two complementary lines of development in the field of radionuclide transport modelling. The first is towards more sophisticated and detailed models for deterministic analyses and the second towards simpler models for probabilistic analyses.

The aim of this study was to focus on a simpler model and present a relatively new approach to modelling of radionuclide migration through the geosphere using a radial basic functions method (RBFs) and to determine the average and sample variance of radionuclide concentration with regard to spatial variability of hydraulic conductivity modelled by a geostatistical approach. We will also explore residual errors and their influence on optimal shape parameters.

II. GEOSTATISTICS

The term geostatistics is employed here as a generic term, meaning the application of the theory of random fields in the earth sciences (Kitanidis and VoMvoris, 1983). The parameters are distributed in space and can thus be called regionalized variables. The parameters of a given geologic formation can conveniently be represented as realisations of random variables which form random fields.

Stochastic simulation is a widely accepted tool in various areas of geostatistics. The goal of stochastic simulation is to reproduce geological texture in a set of equiprobable simulated realizations. Simulations are termed globally accurate through the reproduction of one-, two-, or multiple-point statistics representative of the area under study. In mathematical terms, the most convenient method for simulation is sequential Gaussian simulation (Deutsch and Journel, 1998) because all successive conditional distributions from which simulated values are drawn are Gaussian with parameters determined by the solution of a simple kriging system.

Sequential Gaussian simulation procedure:

1. First, use a sequential Gaussian simulation to

transform the data into a normal distribution.

- 2. Then perform variogram modelling on the data. Select one grid node at random, then krige the value at that location. This will also give us the kriged variance.
- 3. Then draw a random number from a normal distribution that has a variance equivalent to the kriged variance and a mean equivalent to the kriged value. This number will be the simulated number for that grid node.
- 4. Select another grid node at random and repeat. For the kriging, include all the previously simulated nodes to preserve the spatial variability as modelled in the variogram.
- 5. When all nodes have been simulated, back transform to the original distribution. This gives us first realization using a different random number sequence to generate multiple realizations of the map.

Kriging (named after D. G. Krige, a South African mining engineer and pioneer in the application of statistical techniques to mine evaluation) is a collection of generalized liner regression techniques for minimizing an estimation variance defined from a prior model for a covariance (semivariogram) (Deutsch and Journel, 1998). Since the semivariogram is a function of distance, the weights change according to the geographic arrangement of the samples. Kriging can be used to make contour maps, but unlike conventional contouring algorithms, it has certain statistically optimal properties.

III. A RADIAL BASIS FUNCTION METHOD

Radial basis functions method for interpolation, as high accuracy approximations, are not appropriate only for functions or values, but also for their derivatives. The method is useful for scattered data or irregular grids, and can easily be extended to high-dimensional problems (Zhong, 1999). The RBFs method will be shown to provide an alternative choice with respect to FDM or FEM, which require a mesh to support the localized approximations.

Since Kansa (1990a; 1990b) successfully modified the radial basis functions for solving PDEs of elliptic, parabolic, and hyperbolic types, more and more computational tests showed that this method is feasible for solving various PDEs.

A radial basis function (Zhong, 1999) is a function $\phi_j(x) = \phi(||x-x_j||)$, which depends only on distance between $x \in \mathbb{R}^d$ and a fixed point $x_j \in \mathbb{R}^d$. Here, ϕ is continuous and bounded on any bounded sub-domain $\Omega \subseteq \mathbb{R}^d$.

The commonly used radial basis functions are:

 $\phi(r)=r$, linear, $\phi(r)=r^3$, cubic,

 $\phi(r)=r^2\log r$, thin-plate spline,

 $\begin{array}{ll} \phi(r) = e^{-\alpha r^2}, & \text{Gaussian,} \\ \phi(r) = (r^2 + c^2)^{1/2}, & \text{multiquadric,} \\ \phi(r) = (r^2 + c^2)^{-1/2}, & \text{inverse multiquadric,} \end{array}$

In our case we used multiquadric (MQ) and inverse multiquadric. The MQ method was first introduced by Hardy (Hardy, 1971). The parameter c>0 is a positive shape parameter controlling the fitting of a smoothing surface to the data.

IV. MODELLING OF THE RADIONUCLIDE MIGRATION

The central issue in modelling is, on the one hand, consistency between conceptual and mathematical models, and, on the other hand, between conceptual models and scenarios. A conceptual model is a qualitative description of the functioning of the system in a form which is amenable to mathematical representation. It should make explicit all the assumptions and interpretations which are necessary to bridge the gap between the real system and mathematical equations. Each scenario is a set of features, processes and events which has to be considered together to assess the impact of disposal in the future. It is convenient to distinguish between process and model structure identification. The number of processes that may affect flow and transport is very large. Model structure identification refers to the definition of parameter variability, boundary conditions, etc. The most important processes affecting the movement of water and solutes underground are advection, dispersion, and sorption.

The movement of solutes is mostly simulated with an advection-dispersion equation (Bear and Verruijt, 1987). According to this equation, mass transport is controlled by two mechanisms: advection and dispersion. Advection accounts for the movement of the solute, linked to the fluid, with the average water velocity. Average water velocity can be assessed through Darcy's law. Dispersion accounts for mixing caused by diffusion and by random flow from the mean stream. The dispersive component is evaluated by assuming the dispersive mass flux to be proportional to the concentration gradient, similar to Fick's law of molecular diffusion. Without fundamental modifications, the advection-dispersion equation can treat other processes such as sorption, radioactive decay, chemical reactions, sink sources, ion exchange and matrix diffusion.

1. Laplace Equation

The first step of radionuclide transport modelling is to solve the Laplace equation to obtain the Darcy velocity. In this case the Neumann and Dirichlet boundary conditions will be defined along the boundary. Homogeneous and anisotropic porous media and incompressible fluid were assumed in this analysis. The equation has the following form (Bear and Verruijt, 1987):

$$K_x \frac{\partial^2 p}{\partial x^2} + K_y \frac{\partial^2 p}{\partial y^2} = 0 \tag{1}$$

where p is the pressure of the fluid and K_x and K_y are the components of hydraulic conductivity tensor.

The corresponding boundary conditions are:

$$\frac{\partial p}{\partial x}s_x + \frac{\partial p}{\partial y}s_y = g_1(x, y) \tag{2}$$

or

$$p = g_2(x, y) \tag{3}$$

where s_x and s_y are the components of the unit vector normal to the boundary. The Laplace equation was solved by RBF and direct collocation (Fedoseyev *et al.*, 2002). We add an additional set of nodes (outside of the domain) adjacent to the boundary and, correspondingly, add an additional set of collocation equations. We assume the collocation points are arranged in such a way that the first N_I points are in Ω , whereas the last N_B points are on $\partial \Omega$.

The approximate solution can be expressed as:

$$p(x, y) = \sum_{j=1}^{N_I + 2N_B} \alpha_j \varphi_j(x, y)$$
 (4)

where α_j , j=1, ..., N_I+2N_B are the unknown coefficients to be determined. By substituting (4) into (1), (2) or (3), we have:

$$\sum_{j=1}^{N_I+2N_B} \left(K_x \frac{\partial^2 \varphi_j}{\partial x^2} + K_y \frac{\partial^2 \varphi_j}{\partial y^2} \right) \bigg|_{x_i, y_i} \alpha_j = 0$$

$$i=1, 2, \dots, N_I + N_B$$
(5)

$$\sum_{j=1}^{N_I+2N_B} \left(\frac{\partial \varphi_j(x_i, y_i)}{\partial x} s_x + \frac{\partial \varphi_j(x_i, y_i)}{\partial y} s_y \right) \alpha_j = g_1(x_i, y_i)$$

$$i=N_I+N_B+1, \dots, N_I+2N_B \tag{6}$$

or

$$\sum_{j=1}^{N_I + 2N_B} \varphi_j(x_i, y_i) \alpha_j = g_2(x_i, y_i)$$

$$i = N_I + N_B + 1, \dots, N_I + 2N_B$$
(7)

The pressure gradient is evaluated by:

$$\frac{\partial p}{\partial x} = \sum_{i=1}^{N_I + 2N_B} \alpha_i \frac{\partial \varphi_i(x, y)}{\partial x}$$
 (8)

$$\frac{\partial p}{\partial y} = \sum_{j=1}^{N_I + 2N_B} \alpha_j \frac{\partial \varphi_j(x, y)}{\partial y}$$
 (9)

For the calculation of velocity in principal directions we use Darcy's law (Bear and Verruijt, 1987):

$$v_x = -\frac{K_x}{\omega \rho g} \frac{\partial p}{\partial x} \tag{10}$$

$$v_y = -\left(\frac{K_y}{\omega \rho g}\right)\left(\frac{\partial p}{\partial x} + \rho g\right) \tag{11}$$

where ρ is the density of the fluid, ω is porosity, and g gravitational acceleration.

2. Advection-Dispersion Equation

The velocities obtained from the Laplace equation are used in the advection-dispersion equation. The advection-dispersion equation for transport through the saturated porous media zone with retardation and decay is (Bear and Verruijt, 1987):

$$R\frac{\partial u}{\partial t} = \left(\frac{D_x}{\omega} \frac{\partial^2 u}{\partial x^2} + \frac{D_y}{\omega} \frac{\partial^2 u}{\partial y^2}\right) - v_x \frac{\partial u}{\partial x} - R\lambda u$$

$$(x, y) \in \Omega, \ 0 \le t \le T$$

$$u|_{(x, y) \in \partial \Omega} = g(x, y, t) \qquad 0 \le t \le T$$

$$u|_{t=0} = h(x, y), \qquad (x, y) \in \Omega \qquad (12)$$

where x is the groundwater flow axis, y is the transverse axis, u is the concentration of contaminant in the groundwater [Bqm⁻³], D_x and D_y are the components of the dispersion tensor $[m^2y^{-1}]$ in the saturated zone, ω is porosity of the saturated zone [-], v_x is Darcy velocity $[my^{-1}]$ at interior points, R is the retardation factor in the saturated zone [-] and λ is the radioactive decay constant $[y^{-1}]$. In these cases [y] means years.

For the parabolic problem, we consider the implicit scheme:

$$R\frac{u^{n+1} - u^n}{\delta t} = \left(\frac{D_x}{\omega} \frac{\partial^2 u^{n+1}}{\partial x^2} + \frac{D_y}{\omega} \frac{\partial^2 u^{n+1}}{\partial y^2}\right)$$
$$-v_x \frac{\partial u^{n+1}}{\partial x} - R\lambda u^{n+1} \tag{13}$$

where δt is the time step and u^n and u^{n+1} are the contaminant concentrations at the time t_n and t_{n+1} .

The approximate solution is expressed as:

$$u(x, y, t_{n+1}) = \sum_{i=1}^{N} \alpha_j^{n+1} \varphi_j(x, y)$$
 (14)

where α_j^{n+1} , $j=1, \dots, N$ are the unknown coefficients to be determined. $\varphi_j(x, y)$ is Hardy's multiquadrics function (Hardy, 1971):

$$\varphi_i(x, y) = \sqrt{(x - x_i)^2 + (y - y_i)^2 + c^2}$$
 (15)

where c is the shape parameter.

By substituting (14) into (12), we have:

$$\sum_{j=1}^{N} \left(R \frac{\varphi_{j}}{\delta t} - \frac{D_{x}}{\omega} \frac{\partial^{2} \varphi_{j}}{\partial x^{2}} - \frac{D_{y}}{\omega} \frac{\partial^{2} \varphi_{j}}{\partial y^{2}} + v_{x} \frac{\partial \varphi_{j}}{\partial x} + R \lambda \varphi_{j} \right) \bigg|_{x_{i}, y_{i}}$$

$$\cdot \alpha_{j}^{n+1} = R \frac{u^{n}(x_{i}, y_{i})}{\delta t},$$

$$i=1, 2, \dots, N_I$$
 (16)

$$\sum_{j=1}^{N} \varphi_{j}(x_{i}, y_{i}) \alpha_{j}^{n+1} = g(x_{i}, y_{i}, t_{n+1})$$

$$i = N_{I} + 1, N$$
(17)

where

$$\frac{\partial \varphi_j}{\partial x} = \frac{(x_i - x_j)}{\varphi_i(x_i, y_i)} \tag{18}$$

$$\frac{\partial \varphi_j}{\partial y} = \frac{(y_i - y_j)}{\varphi_j(x_i, y_i)} \tag{19}$$

$$\frac{\partial^2 \varphi_j}{\partial x^2} = \left[1 - \frac{(x_i - x_j)^2}{\varphi_i^2(x_i, y_i)}\right] \frac{1}{\varphi_i(x_i, y_i)}$$
(20)

$$\frac{\partial^2 \varphi_j}{\partial y^2} = \left[1 - \frac{(y_i - y_j)^2}{\varphi_i^2(x_i, y_i)}\right] \frac{1}{\varphi_i(x_i, y_i)}$$
(21)

from which we can solve the $N \times N$ linear system of (16)-(17) for the unknown coefficients α_j^{n+1} , $j=1, \dots, N$. Let $N=N_I+N_B$ be the number of collocation points, N_I is the number of interior points and N_B is the number of boundary points. Then (14) can give us the approximate solution at any point in the domain Ω .

V. NUMERICAL EXAMPLE

The simulation was implemented for a rectangular area 600 m long and 300 m wide. The source (initial condition) was Thorium (Th-230) with activity $1\cdot10^6$ Bq and half-life of 77000 years. The source was located on the left side of the area. The groundwater flow field is presented for a steady-state condition. Except for the inflow (left side) and outflow (right side), all boundaries have no-flow condition $\frac{\partial p}{\partial s} = 0$ (s taken normal to the boundary). The inflow rate was 1 m/y. At the outflow side, time-constant pressures at the boundaries were set. The

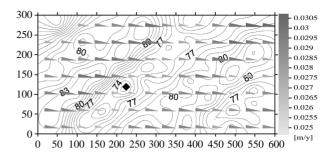


Fig. 1 Conductivity and velocities based on 8-point data set, shape parameter: 4.5

location of the radioactive source is presented with symbol \diamondsuit .

The components of the dispersion tensor are approximated by $D_x=a_xv$ and $D_v=a_vv$. Longitudinal dispersivity a_x is 500 m and transversal dispersivity a_v is 2 m, v is Darcy's velocity. Retardation constant R is 800. Porosity ω is between 0.25 and 0.26 whereas hydraulic conductivity was generated at different points with geostatistics (Deutsh and Journel, 1998) based on two different sets of input data. In the first one, hydraulic conductivity at 8 different points is given (values are: 66.00, 71.00, 73.00, 75.00, 76.52, 77.02, 79.74, 83.41 $\left[\frac{m}{v}\right]$). Positive variance contribution or sill size is 1.0 and nugget effect size is 0.0 as variogram parameters are chosen. Simple kriging is chosen as the type of kriging. A spherical model is chosen as a variogram structure. The angles defining the geometric anisotropy are maximum horizontal range 600 m and minimum horizontal range 300 m. It is assumed that the mean in the case of simple kriging is known.

In the second case a data base of 16 different points is used (values are: 66.00, 71.00, 73.00, 75.00, 76.52, 77.02, 79.74, 83.41, 36.00, 21.00, 173.00, 275.00, 96.52, 57.02, 97.74, 63.41 $\left[\frac{m}{y}\right]$). Positive variance contribution or sill size 0.7 and nugget effect size 0.3 as variogram parameters are chosen. Ordinary kriging is chosen as the type of kriging where the constant mean value is replaced by the location-dependent estimate.

Distribution of hydraulic conductivity and velocities based on an 8-point data set are shown in Fig. 1, distribution of hydraulic conductivity and velocities based on a 16-point data set are shown in Fig. 2. We can see that the length of the velocity is greatly dependent on hydraulic conductivity and porosity. In Fig. 1 we cannot see a lot of variability of hydraulic conductivity. One of the reasons could be that there are not many differences between the prescribed values of hydraulic conductivity. In Fig. 2 we can see more variability of hydraulic conductivity.

The distribution of the average value of contaminant concentration after 100,000 years is given. These

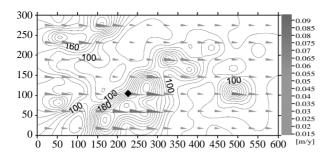


Fig. 2 Conductivity and velocities based on 16-point data set, shape parameter: 4.9

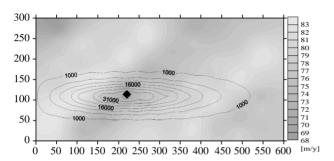


Fig. 3 Average of concentrations (8 points), shape parameter: 140

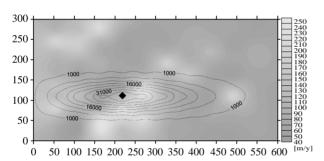


Fig. 4 Average of concentrations (16 points), shape parameter: 120

values were obtained after repeating 100 simulations. Distribution of average of contaminant concentrations (8 points) and standard deviation of contaminant concentrations (8 points) are shown in Fig. 3 and Fig. 5. Distribution of average of contaminant concentrations (16 points) and standard deviation of contaminant concentrations (16 points) are shown in Fig. 4 and Fig. 6. Distribution of contaminant concentration after 100,000 years for one specific simulation at 8 points and 16 points data sets of hydraulic conductivity are shown in Fig. 7 and Fig. 8. Comparison of the results between the average of contaminant concentrations (Fig. 3 and Fig. 4) and concentrations for one particular simulation (Fig. 7 and Fig. 8) shows that the more realizations we have, the more accurate are the results. The scatter of the results is not large, which is also indicated in Fig. 5.

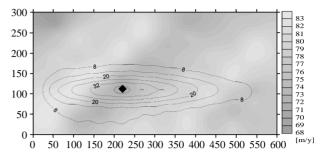


Fig. 5 Standard deviation of concentrations (8 points), shape parameter: 140

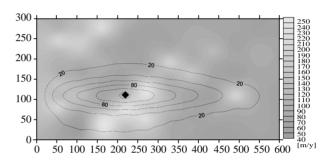


Fig. 6 Standard deviation of concentrations (16 points), shape parameter: 120

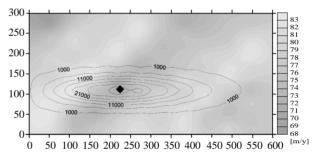


Fig. 7 Concentrations based on 8 points data set for one simulation, shape parameter: 140

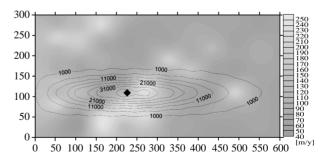


Fig. 8 Concentrations based on 16 points data set for one simulation, shape parameter: 120

VI. OPTIMAL SHAPE PARAMETE

In our problem we used multiquadric (MQ) and inverse multiquadric RBFs. MQ's performance

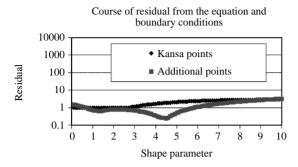


Fig. 9 Residuals

depends on the choice of a user-specified parameter c, which is often referred to as the shape parameter. The shape parameter controls the effective number of collocation points used in the interpolation at any location.

When c is small, the surface fitted to the data contains sharp corners at the collocation points. As c increases, more collocation points are effectively involved in the interpolation and the sharp corners spread out to form a smooth surface. When c is too large and reaches a critical value, the resulting matrix becomes ill-conditioned and the solution is smeared.

In the past, there have been several numerical experiments and empirical formulas that suggest how to choose the optimal value of such parameters, which in general depend on the density of the interpolation centres (Kansa, 1990a). In practice, the optimal value of the shape parameter can be determined by numerical experiments. The optimal shape parameter depends on the properties of the numerical solution, number and locations of the collocation points. Therefore, a question of how to find the optimal shape parameter for an arbitrary real problem given by geometric and hydrological parameters of a continuum always appears as one of the key problem.

In our case, we always try to answer the question of how to find a good optimal shape parameter, which fulfils the equation in the most possible points. Many realizations of the equations were made using different shape parameters at different points. The course of residual errors from the equation in Kansa (basic mesh of the problem) and additional points are shown in Fig. 9. We can see that as the shape parameter gets larger, the residuals get larger. By increasing the shape parameter the residual errors from the PDEs showed in Kansa tend to a minimum value and then grow.

We set the shape parameter at 4.5 and compared results with the test method (the finite difference method). The results were very similar.

VII. CONCLUSION

This work presents modelling of radionuclide migration through the geosphere using a radial basis

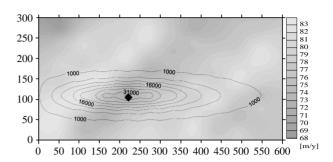


Fig. 10 Concentrations based on 8 points data set for one simulation, finite difference method

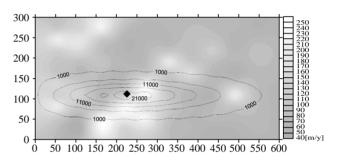


Fig. 11 Concentrations based on 16 points data set for one simulation, finite difference method

functions method and geostatistics.

In the case of radionuclide migration two steps of evaluations were performed. In the first step the velocities in principal directions were determined from the pressure of the fluid p obtained from the Laplace differential equation. In the second step the advection-dispersion equation was solved to find the concentration of the contaminant. In this case the method of evaluation was verified by comparing results with the one obtained from the finite difference method (Fig. 10 and Fig. 11). Both methods give very similar results.

Due to different types of conductivity, variogram input parameters and different types of kriging were necessary to find an appropriate shape parameter which can give us results comparable to the test method.

A good parameter assessment was obtained from graphic presentations. Thus we explore the residual errors from the equation as an error indicator which provides a road map to the optimal selection of the shape parameter value.

In the case of calculating the advection-dispersion equation we can conclude that the Kansa method could be an appropriate alternative to the FDM due to its simpler implementation.

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