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Vrankar, L., Turk, G. in Runovc, F. 2004. Modelling of radionuclide migration through the geosphere with radial basis function method and geostatistics. *Journal of the Chinese Institute of Engineers* 27, 4: 455–462.  
DOI: 10.1080/02533839.2004.9670895.

## Modelling of Radionuclide Migration through 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 measurement, which are not available for all regions of interest. For example, the hydraulic conductivity as input parameter varies from place to place. In such case geostatistical science offers a variety of spatial estimation procedures. To assess a long term safety of 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 been usually obtained by either 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 modelling of radionuclide migration through geosphere using radial basis functions method (RBFs) and to determine the average and sample variance of radionuclide concentration with regard to spatial variability of hydraulic conductivity modelled by geostatistical approach. We will also explore the residual errors and their influence on optimal shape parameter.*

**Keywords** - *Radionuclide migration, Porous media, Partial differential equation, Radial basis function, Numerical solution, Kansa method, Geostatistics.*

## 1. INTRODUCTION

Waste disposal has become a key issue in these environmentally conscious time [1]. 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 the waste management research. Within the project INTRACOIN [2], a comparison has been made between different computational codes describing transport of radionuclides in geologic media. In Slovenia, the two disposal concepts/siting options are currently being considered for the facility: a surface vault disposal facility; and an underground (tunnel) disposal facility [3].

The modelling of radionuclide transport through the geosphere is necessary in the safety assessment 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 the nuclide transport from the repository to the biosphere are essential tools.

When modelling flow and contaminant transport in 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, environmental change all can have an impact on the geosphere [15].

The general reliability and accuracy of transport modelling depend predominantly on input data like hydraulic conductivity, water velocity, radioactive inventory, hydrodynamic dispersion, etc. 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, the hydraulic conductivity as input parameter varies from place to place. In such case geostatistical science offers a variety of spatial estimation procedures [8].

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), and boundary elements methods (BEM) [11]. 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 [11].

A fairly new approach for solving PDEs is through radial basis functions (RBFs). Kansa [4], [5] 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 space 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 INTRACOIN project was told that there are two complementary lines of development in 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 to simpler model and present a relatively new approach to modelling of radionuclide migration through geosphere using 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 geostatistical approach. We will also explore the residual errors and their influence on optimal shape parameter.

## 2. GEOSTATISTICS

The term geostatistics is employed here as a generic term, meaning the application of the theory of random fields in the earth sciences [9]. 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 for the area under study. In mathematical terms, the most convenient method for simulation is sequential Gaussian simulation [8] 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 transform the data into a normal distribution. 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. 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. 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. 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 linear regression techniques for minimizing an estimation variance defined from a prior model for a covariance (semivariogram) [8]. 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.

### 3. A RADIAL BASIS FUNCTION METHOD

Radial basis functions method for interpolation, as a high accurate approximation are not appropriate only for the functions or values, but also for their derivatives. The method is available for scattered data or irregular grid, and can easily be extended to high-dimensional problems [12]. 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 [4], [5] 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 to solve various PDEs.

A radial basis function [12] is a function  $\phi_j(x) = \phi(\|x - x_j\|)$ , which depends only on distance between  $x \in \mathbf{R}^d$  and a fixed point  $x_j \in \mathbf{R}^d$ . Here,  $\phi$  is continuous and bounded on any bounded sub-domain  $\Omega \subseteq \mathbf{R}^d$ .

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,
$\phi(r) = e^{-\alpha r^2},$	Gaussian,
$\phi(r) = (r^2 + c^2)^{\frac{1}{2}},$	multiquadric,
$\phi(r) = (r^2 + c^2)^{-\frac{1}{2}},$	inverse multiquadric,

In our case we used multiquadric (MQ) and inverse multiquadric. MQ method was first introduced by Hardy [13]. The parameter  $c > 0$  is a positive shape parameter controlling the fitting of a smoothing surface to the data.

## 4. 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 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 the 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 advection-dispersion equation [10]. 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 the 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.

### 4.1 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. Anisotropic porous media and incompressible fluid were assumed in this analysis. The equation has the following form [10]:

$$K_x \frac{\partial^2 p}{\partial x^2} + K_y \frac{\partial^2 p}{\partial y^2} = 0, \quad (1)$$

where  $p$  is the pressure of the fluid and  $K_x$  and  $K_y$  are the components of hydraulic conductivity tensor. The corresponding boundary condition is

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

where  $s_x$  in  $s_y$  are the components of the unit vector normal to the boundary.

The Laplace equation was solved by RBF and direct collocation [6]. We add an additional set of nodes (outside of the domain) adjacent to the boundary and, correspondingly, add an additional

set of collocation equations.

The approximate solution can be expressed as :

$$p(x, y) = \sum_{j=1}^{N+N_b} c_j \varphi_j(x, y) \quad (3)$$

where  $c_j, j = 1, \dots, N + N_b$  are the unknown coefficients to be determined. By substituting (3) into (1) and (2), we have:

$$\sum_{j=1}^{N+N_b} \left( K_{x_i} \frac{\partial^2 \varphi_j}{\partial x^2} + K_{y_i} \frac{\partial^2 \varphi_j}{\partial y^2} \right) c_j = 0, \quad i = 1, 2, \dots, N_i + N_b, \quad (4)$$

$$\sum_{j=1}^{N+N_b} \left( \frac{\partial \varphi_j(x_i, y_i)}{\partial x} s_x + \frac{\partial \varphi_j(x_i, y_i)}{\partial y} s_y \right) c_j = g(x_i, y_i), \quad i = 1, \dots, N_b, \quad (5)$$

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.

The pressure gradient is evaluated by:

$$\frac{\partial p_i}{\partial x} = \sum_{j=1}^{N+N_b} c_j^n \frac{\partial \varphi_j(x_i, y_i)}{\partial x}, \quad (6)$$

$$\frac{\partial p_i}{\partial y} = \sum_{j=1}^{N+N_b} c_j^n \frac{\partial \varphi_j(x_i, y_i)}{\partial y}. \quad (7)$$

For the calculation of velocity in principal directions we use Darcy's law [10]:

$$v_{x_i} = - \left( \frac{K_{x_i}}{n \rho g} \right) \frac{\partial p_i}{\partial x}, \quad (8)$$

$$v_{y_i} = - \left( \frac{K_{y_i}}{n \rho g} \right) \left( \frac{\partial p_i}{\partial y} + \rho g \right). \quad (9)$$

where  $\rho$  is the density of the fluid,  $n$  is porosity, and  $g$  gravitational acceleration.

## 4.2 ADVECTION-DISPERSION EQUATION

In the next step, the velocities obtained from 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 [10]:

$$\begin{aligned}
 R \frac{\partial u}{\partial t} &= \left( \frac{D_x}{\omega_e} \frac{\partial^2 u}{\partial x^2} + \frac{D_y}{\omega_e} \frac{\partial^2 u}{\partial y^2} \right) - v_{x_i} \frac{\partial u}{\partial x} - R\lambda u, & (x, y) \in \Omega, \quad 0 \leq t \leq T, \\
 u|_{(x,y) \in \partial\Omega} &= g(x, y, t), & 0 \leq t \leq T \\
 u|_{t=0} &= h(x, y), & (x, y) \in \Omega,
 \end{aligned} \tag{10}$$

where  $x$  is the groundwater flow axis,  $y$  is the transverse axis,  $u$  is the concentration of contaminant in the groundwater [ $\text{Bqm}^{-3}$ ],  $D_x$  and  $D_y$  are the components of dispersion tensor [ $\text{m}^2\text{y}^{-1}$ ] in saturated zone,  $\omega_e$  is the effective porosity of the saturated zone[-],  $v_{x_i}$  is Darcy velocity [ $\text{my}^{-1}$ ] at interior points,  $R$  is the retardation factor in saturated zone [-] and  $\lambda$  is the radioactive decay constant [ $\text{y}^{-1}$ ].

For the parabolic problem, we consider the implicit scheme:

$$R \frac{u^{n+1} - u^n}{\delta t} = \left( \frac{D_x}{\omega_e} \frac{\partial^2 u^{n+1}}{\partial x^2} + \frac{D_y}{\omega_e} \frac{\partial^2 u^{n+1}}{\partial y^2} \right) - v_{x_i} \frac{\partial u^{n+1}}{\partial x} - R\lambda u^{n+1}, \tag{11}$$

where  $\delta t$  is the time step,  $u^n$  and  $u^{n+1}$  are the contaminant concentration at the time  $t_n$  and  $t_{n+1}$ .

The approximate solution is expressed as :

$$u(x, y, t_{n+1}) = \sum_{j=1}^N c_j^{n+1} \varphi_j(x, y) \tag{12}$$

where  $c_j^{n+1}$ ,  $j = 1, \dots, N$  are the unknown coefficients to be determined.  $\varphi_j(x, y)$  is the Hardy's multiquadrics function [7]:

$$\varphi_j(x, y) = \sqrt{(x - x_j)^2 + (y - y_j)^2 + c^2} \tag{13}$$

where  $c$  is shape parameter.

By substituting (12) into (11), we have:

$$\sum_{j=1}^N \left( R \frac{\varphi_j}{\delta t} - \frac{D_x}{\omega_e} \frac{\partial^2 \varphi_j}{\partial x^2} - \frac{D_y}{\omega_e} \frac{\partial^2 \varphi_j}{\partial y^2} + v_{x_i} \frac{\partial \varphi_j}{\partial x} + R\lambda \varphi_j \right) c_j^{n+1} = R \frac{u^n(x_i, y_i)}{\delta t}, \tag{14}$$



where  $i = 1, 2, \dots, N_i$ . By substituting (12) into the second of equations (10) we obtain

$$\sum_{j=1}^N \varphi_j(x_i, y_i) c_j^{n+1} = g(x_i, y_i, t_{n+1}), \quad i = N_i + 1, N, \quad (15)$$

where:

$$\frac{\partial \varphi_j}{\partial x} = \frac{(x_i - x_j)}{\varphi_j(x_i, y_i)}, \quad (16)$$

$$\frac{\partial \varphi_j}{\partial y} = \frac{(y_i - y_j)}{\varphi_j(x_i, y_i)}, \quad (17)$$

$$\frac{\partial^2 \varphi_j}{\partial x^2} = \left[ 1 - \frac{(x_i - x_j)^2}{\varphi_j^2(x_i, y_i)} \right] \frac{1}{\varphi_j(x_i, y_i)}, \quad (18)$$

$$\frac{\partial^2 \varphi_j}{\partial y^2} = \left[ 1 - \frac{(y_i - y_j)^2}{\varphi_j^2(x_i, y_i)} \right] \frac{1}{\varphi_j(x_i, y_i)}, \quad (19)$$

from which we can solve the  $N \times N$  linear system of ((14)-(15)) for the unknown  $c_j^{n+1}, j = 1, \dots, N$ . 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 (12) can give us the approximate solution at any point in the domain  $\Omega$

## 5. NUMERICAL EXAMPLE

The simulation was implemented for rectangular area which was 600 m long and 300 m high. The source was Thorium ( $Th - 230$ ) with activity  $1 \cdot 10^6 Bq$  and half-life of 77000 years. The source was located on 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 location of the radioactive source is presented with symbol  $\diamond$ .

The components of dispersion tensor are approximated by  $D_x = a_L v$  and  $D_y = a_T v$ . Longitudinal dispersivity,  $a_L$  is 500 m and transversal dispersivity,  $a_T$  is 2 m,  $v$  is Darcy's velocity. Porosity is 0.25 whereas hydraulic conductivity was generated in different points with geostatistics [8] 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 [ $\frac{m}{y}$ ]). Positive variance contribution or sill is size 1.0 and nugget effect size 0.0 as variogram parameter are chosen. Simple kriging is chosen as a type of kriging.

In the second case the 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 [ $\frac{m}{y}$ ]).

Positive variance contribution or sill size 0.7 and nugget effect size 0.3 as variogram parameters are chosen. Ordinary kriging is chosen as a type of kriging.

Distribution of hydraulic conductivity and velocities based on 8-point data set are shown on fig. 1, distribution of hydraulic conductivity and velocities based on 16-point data set are shown on fig. 2. Distribution of average of contaminant concentrations (8 points) and standard deviation of contaminant concentrations (8 points) are shown on fig. 3 and fig. 5. Distribution of average of contaminant concentrations (16 points) and standard deviation of contaminant concentrations (16 points) are shown on fig. 4 and fig. 6.

The distribution of the average value of contaminant concentration after 100 000 years is given. These values were obtained after repeating 100 simulations. Distribution of contaminant concentration after 100 000 years at 8 points and 16 points data set of hydraulic conductivity are shown on fig. 7 and fig. 8

## 6. OPTIMAL SHAPE PARAMETER

In our problem we used multiquadric (MQ) and inverse multiquadric RBFs. MQ's performance depends on the choice of a user-specified parameter  $c$ , which is often referred 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 [4]. In practice, the optimal value of the shape parameter can be determined by numerical experiments. The optimal shape parameter depends on the properties of numerical solution, number and locations of the collocations points. So, It appears always a question how to find the optimal shape parameter for arbitrary real problem given by geometry and hydrological parameters of continuum.

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

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

From the fig. 1 and fig. 2 we can see that the conductivity of porous media has influence on magnitude of velocities of the fluid. Different standard deviations of contaminant concentrations are due to different input data sets of the conductivity and geostatistics.

## **7. CONCLUSION**

This work presents modelling of radionuclide migration through geosphere using radial basis 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 pressure of the fluid  $p$  obtained from Laplace differential equation. In the second steps the advection- dispersion equation was solved to find a concentration of the contaminant. In this case the method of evaluation was verified by comparing results with the one obtained from finite difference method (Fig. 10 and 11). Both methods give very similar results.

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

A good parameter assessment was obtained from graphic presentations. So, 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.

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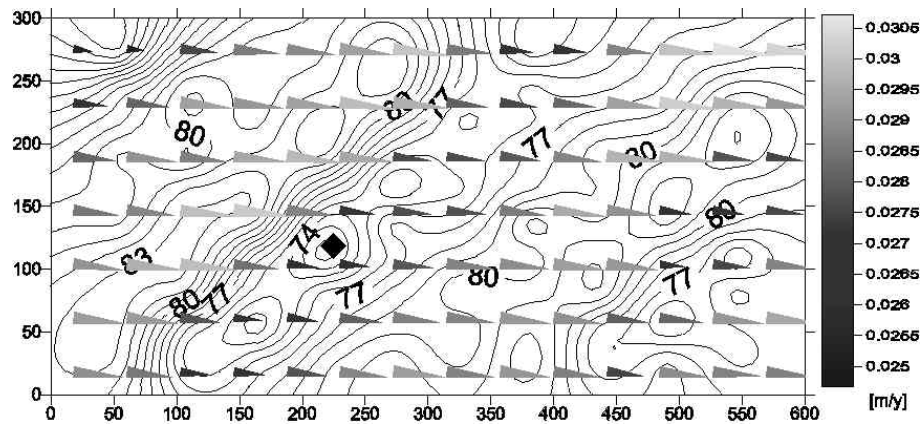


Fig. 1: Conductivity and velocities based on 8-point data set, shape parameter: 4.5

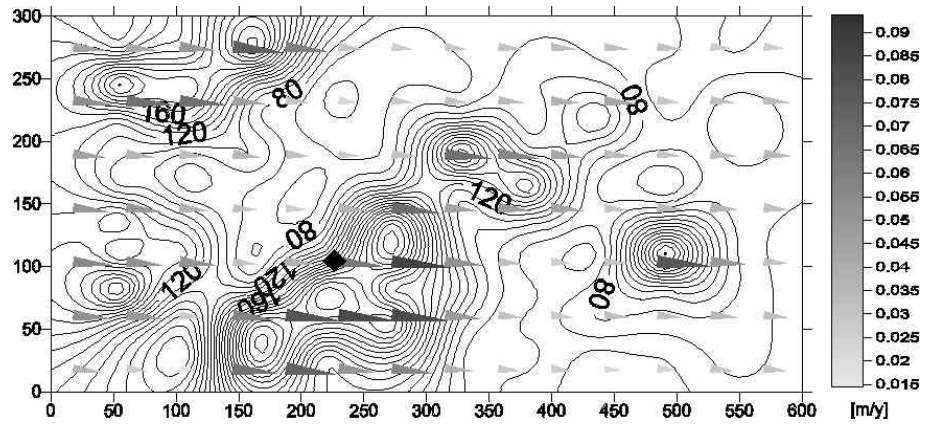


Fig. 2: Conductivity and velocities based on 16-point data set, shape parameter: 4.9

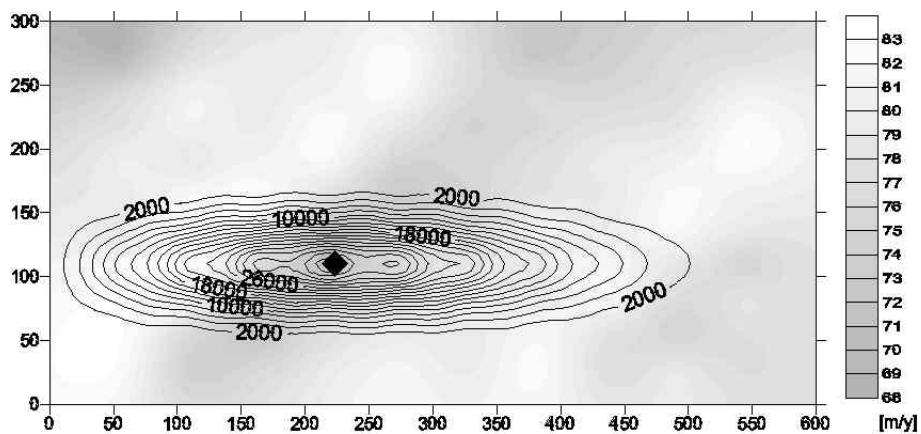


Fig. 3: Average of concentrations (8 points), shape parameter: 140



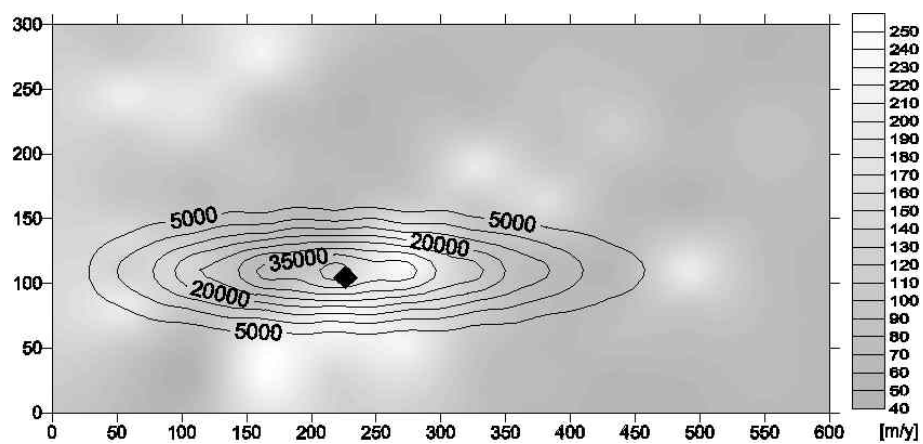


Fig. 4: Average of concentrations (16 points), shape parameter: 120

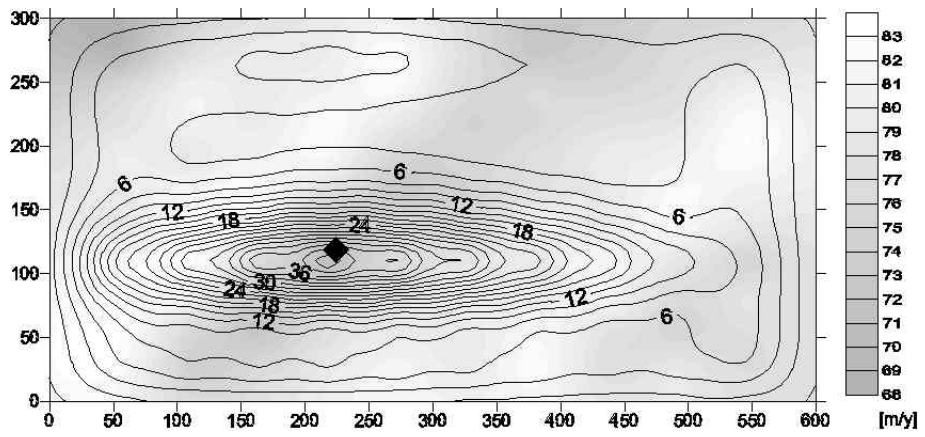


Fig. 5: Standard deviation of concentrations (8 points), shape parameter: 140

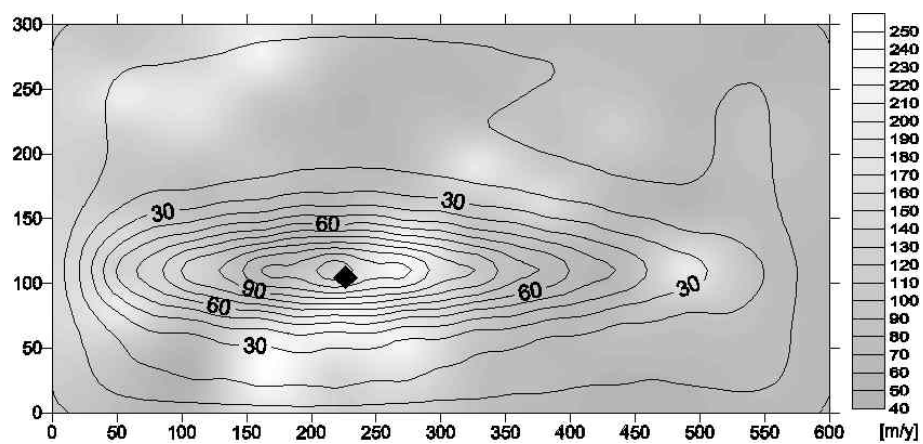


Fig. 6: Standard deviation of concentrations (16 points), shape parameter: 120

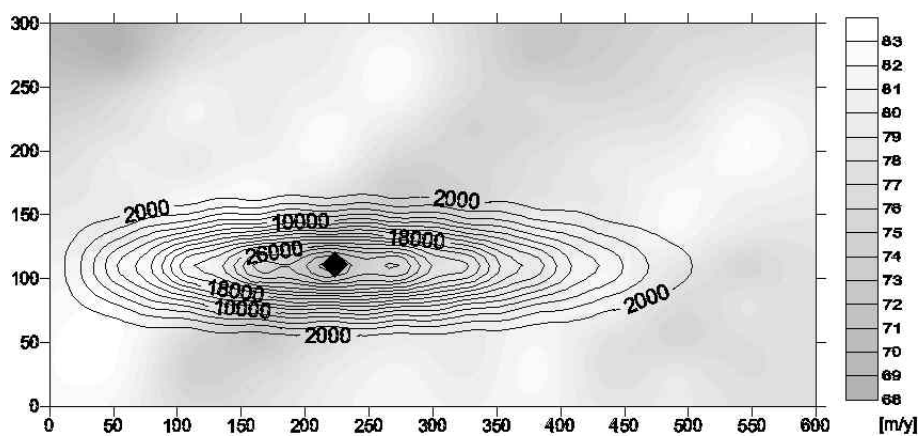


Fig. 7: Concentrations based on 8 points data set for one simulation, shape parameter: 140

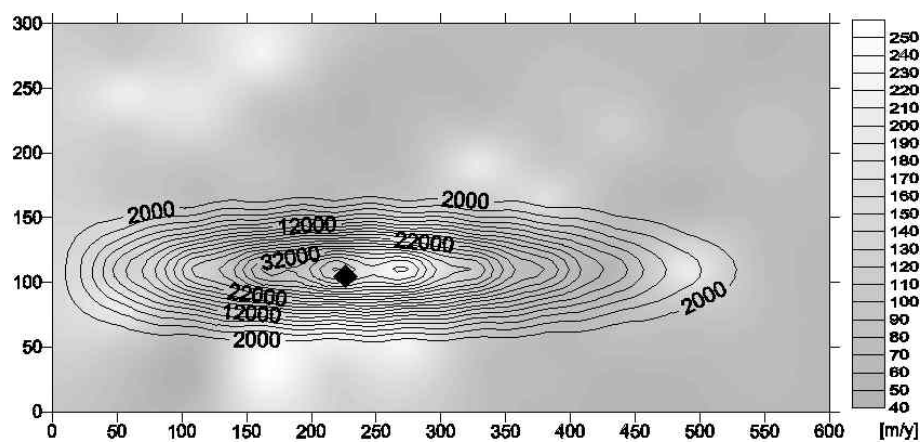


Fig. 8: Concentrations based on 16 points data set for one simulation, shape parameter: 120

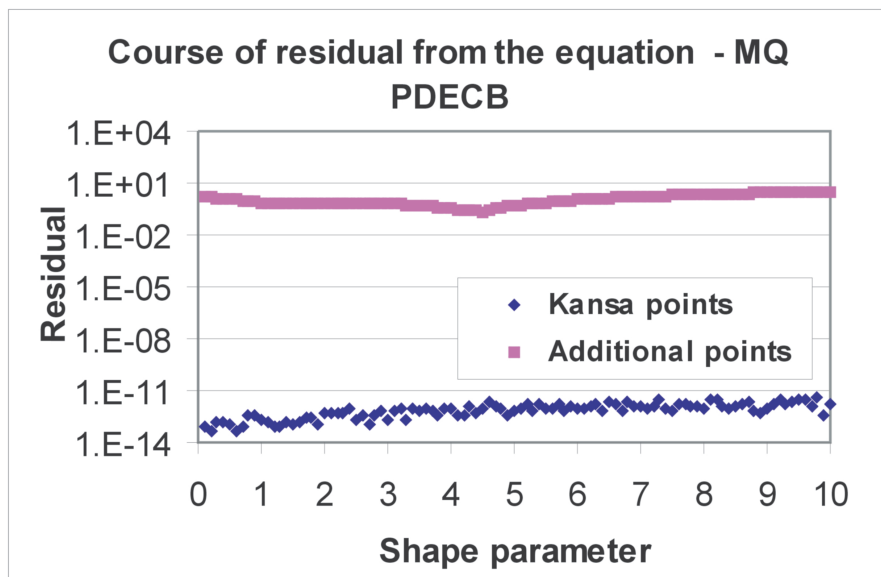


Fig. 9: Residuals

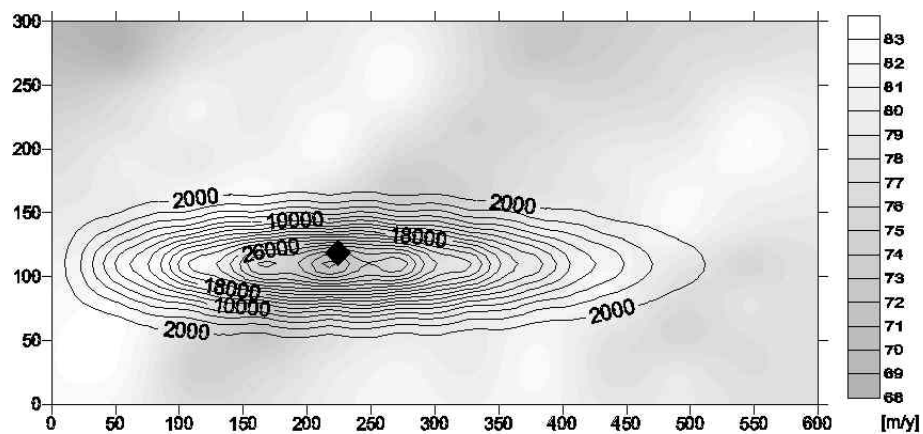


Fig. 10: Concentrations based on 8 points data set for one simulation, finite difference method

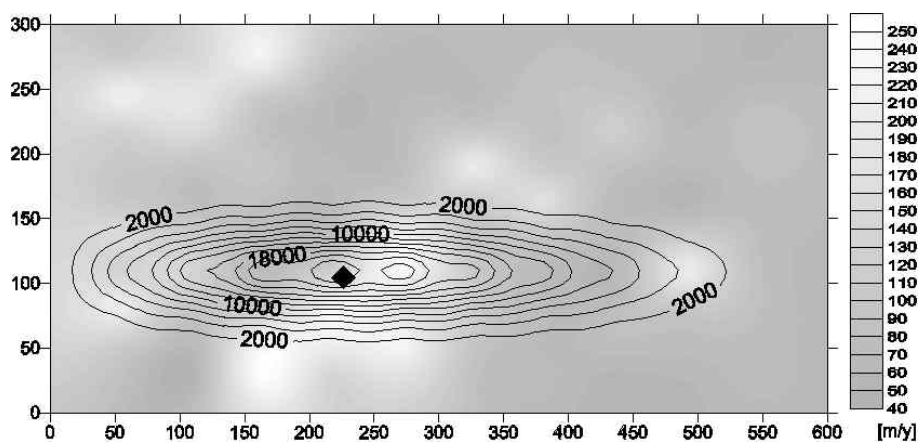


Fig. 11: Concentrations based on 16 points data set for one simulation, finite difference method