MODELLING THE RADIOACTIVE GROUND POLLUTION BY TRANSURANIUM ELEMENTS USING CELLULAR AUTOMATA

K. Breitenecker[1], A. Brandl[2], H. Böck[1], M. Villa[1]

[1] Atomic Institute of the Austrian Universities
1020 Vienna, Stadionallee 2, Austria
[2]Nuclear Engineering Seibersdorf GmbH
2444 Seibersdorf, Austria
Katharina.Breitenecker@ati.ac.at (K. Breitenecker)

Abstract

In the course of decommissioning the former ASTRA Research Reactor, the Seibersdorf site has to be surveyed for possible contamination by radioactive materials, including transuranium elements. The site survey encompasses measurements of both, the site's area and soil. In the case of remediation measures, the exact boundaries of the contamination and the activity concentration have to be determined. To limit costs due to systematic sampling and time consuming laboratory analyses, a mathematical model that describes the migration of transuranium elements and that includes the local topography of the area where deposition has occured, was established. The concept is to find a mathematical function that determines the contamination by modelling the pathways of Transuranium elements. The model approach chosen is cellular automata (CA). For this purpose, a hypothetical acitivity of transuranium elements is released on the ground in the center of a simulated area. Under the assumption that migration of these elements only takes place by diffusion, advection and sorption, these equations are modeled in the CA-model by a simple discretization for the existing problem. The radioactive decay of the transuranium elements of concern has to be considered in this model, just like the different migrational behaviour of these elements (e.g. sorption). The CA-model was compared to measurment and up to now the simulated migrational behaviour appears to describe the migration of transuranium elements rather well.

Keywords: Cellular Automata, Migration in Soils, Radioactive Pollution, Transuranics

Presenting Author's Biography

Katharina Breitenecker studied Technical Physics at the Vienna University of Technology. She did her Master's at the Atomic Institute of the Austrian Universities, currently she is working as research and teaching assistant at the Atomic Institute of the Austrian Universities, where she is also enrolled as a PhD graduate student. Besides, she works part-time at Nuclear Engineering Seibersdorf GmbH and contributes in operational radiation protection. Her major scientific interrests are nuclear environmental analytics and nuclear forensics.

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1 General

The International Atomic Energy Agency (IAEA) has launched an environmental remediation project to deal with the problems of radioactive contamination worldwide. Several documents, including documents on the characterization of contaminated sites have been prepared [1], [2], [3].

A critical element in any such project is the characterization of the contamination and of the various environmental compartments in which radioactive contamination are found, which is necessary in order choose the proper remediation techniques. Careful delineation of the contamination has to ensure that the entire radioactive material is treated, commensurate with the IAEA recomondations.

Understanding the processes of migration and dispersion is particulary important when natural attenuation is to be relied upon as a mechanism for remediation and dose prevention.

Although contamination may originally be dispersed, secondary processes may concentrate, fractionate or otherwise redistribute the radionuclides. Conversely, erosion may lead to the dispersion of radionuclides, particulary in a mining environment. Techniques for remediating sites with well defined contaminated areas containing high concentrations of radionuclides are well established. On the other hand, remediation of dispersed contamination is a challenge, when considering factors such as expected dose, removal of radionuclides at minimal disturbance of the environment.

In order to decrease costs and expenditure of time, a mathematical model that describes the migration of transuranium elements and includes the local topography of the area where deposition has occured, has been established. The concept is to find a mathematical formula which specifies the activity concentration on a larger area while only limited sampling is performed.

2 Radioactive Release in the Environment

It is frequently not realized that the fabric of our planet is radioactive and that the earth has been continuously bombarded with particles from extraterrestrial sources. These particles induce radioactive nuclides in the surrounding atmosphere. Thus, all life from its very beginnings has been subject to a pervasive stream of ionizing radiation, emanating from the environment in which it exists and from the materials from which it is constructed.

In addition, the last century has seen an additional anthropogenic contribution to this background radioactivity (i.e. medical diagnostic, clinical uses of radiation, operation of nuclear power plants, testing of nuclear weapons). These latter sources make only a minor contribution to the overall background radioactivity, but tend to figure disproportionately in the public imagination. Nuclear accidents, such as Three Mile Island and Chernobyl, tend to reinforce this assumption, even though they contributed only a relatively small fraction to the overall dose received by the general population away from the accidents spots [4]. Thus, the different radioactive sources shall be introduced in the following sections.

2.1 Cosmic Radiation

Cosmic radiation has two main sources, i.e. galactic and solar. Both types of radiation consist mainly of protons. Galactic cosmic rays are produced in stellar flares, supernovae explosions, etc. Solar cosmic rays are produced in the Sun, where particles are continuously emitted.

On entering the atmosphere, those particles may interact with nuclei of atmospheric gases (nitrogen, oxygen and argon) in a variety of nuclear reactions. The products include, amongst neutrons, muons, etc. radioisotopes, such as ^{3}H (tritium) and ^{7}Be (beryllium). Exposure to cosmic rays increases with altitude and hence, air travel increases exposure to cosmic radiation [4].

2.2 Terrestrial Sources of Radiation

The work of Pierre and Marie Curie already showed that radioactive material also exists naturally in the environment. This group of radionuclides is responsible for most of the radiation exposure of the vast majority of individuals. The principal radionuclides are ^{40}K (potassium-40, half life $1,28 \cdot 10^9$ years), ^{232}Th (thorium-232, half life $1,41 \cdot 10^{10}$ years) and ^{238}U (uranium-232, half life $4,47 \cdot 10^8$ years). The thorium and uranium radionuclides are the starting points for decay chains of several radionuclides. Members of these decay chains may be important, either as contributors to human exposure or for other radioactive properties. A radioactive nuclide may deceay to another radioactive nuclide, which in turn again decays further again, until a stable isotope is reached [4].

2.3 Anthropogenic Radionuclides

In the 1920's, phycisists realised that energy could be released by splitting or fissioning uranium nuclei. Fisson is induced by the absorption of a neutron by a nucleus and it was the first nuclear process to be developed. It is also the basis for current research and power reactors.

Fission products (^{137}Cs, ^{144}Ba, ^{89}Kr,...) are radioactive and the released neutrons can induce more fission processes. This leads to the possibility of a self sustaining series of nuclear fissions.

Also the atomic bombs used on Japan in World War II were fission bombs, a ^{235}U device was exploded over Hiroshima, followed by a ^{239}Pu device, a transuranium element, over Nagasaki [4].

2.3.1 Transuranium Elements

Most of transuranium elements were first discovered in extensive and expensive experiments. Only two transuranium elements, einsteinium and fermium, were discovered by coincidence. They were discovered in environmental samples taken after the first thermonuclear explosions at the Bikini - Atoll, November

1952.

Also, transuranium elements are created by the absorption of neutrons, see figure 1 as an example. A nucleus captures and neutron and this new neutron-rich nucleus transforms via β-decay to the next higher element, as shown in the creation of ^{239}Pu. Neutron sources are i.e. nuclear reactors. Within a reactor, transuranium elements, ^{237}Np, $^{239-242}$Pu, ^{241,243}Am and ^{244}Cm can be produced in a weighable amount. The next higher elements are often refered as the transcurium elements. During processing of spent nuclear fuels, these elements can be seperated chemically.

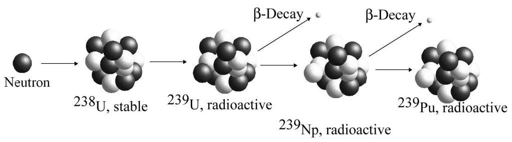

Fig. 1 Creation of transuranium elements by neutron capture

Some transuranium elements can be used for nuclear weapons or as device for nuclide batteries, which are used in satellites [5].

If transuranium elements are released in the environment, they may spread and decay, see figure 2, until a stable product is created.

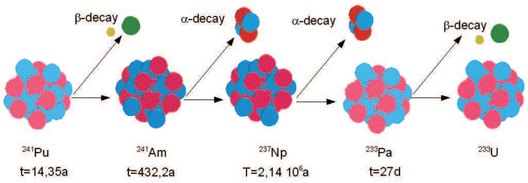

Fig. 2 Decay of ^{241}Pu to ^{233}U

3 Basic Principles for the Migration Model

The central processes for radionuclide migration are radioactive decay, matrix diffusion, the distribution of groundwater flow, and sorption [6]. These provide key terms for the transport equations of the migration model. Others, like colloid-facilitated transport, influence of microbes, gas mediated transport, precipitation and dissolution and off-diagonal Onsager processes are of secondary significance and can, if necessary, be carefully taken into account by adding them to one of the four main processes.

Although the afore mentioned main processes primarily cause the retention and transport of radionuclides in studies of deep geological respositories, they are also applicable - with some constraints - to other geological environments [6].

Transuranium elements might be released to the environment i.e. by fallout after nuclear weapons testing, or by a damaged satellite. Actinides amongst other radionuclides may be spread in the air by wind until they adhere to plants and crops. When it rains, those radioelements are washed towards the grassroots, and deposition in the soil occurs, see figure 3. Now, further migration can take place via two different processes, which are basic key terms in the transport equation, namely diffusion in the soil, and advection with rain water, see figure 3.

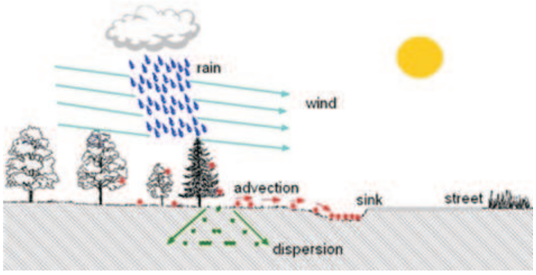

Fig. 3 The different ways of migration in the soil

3.1 Radioactive Decay

The first central process for radionuclide migration is the **radioactive decay**. The rate of decay of a radionuclide is described by its activity A, that is, by the number of atoms that decay per unit time [7]. The probability of radioactive decay is represented by the decay constant λ, and depends on the half life of the radioculide concerned.

$$A(t) = A_0 \cdot e^{-\lambda \cdot t} \qquad (1)$$

Depending on the type of radionuclides which were released (some Am or Pu isotope), the activity concentration increases or decreases with time, depending on the radioactive equilibrium between mother and daughter nuclides. If the released radionuclide decays to a stable product the activity concentration will decrease. But if the daughter-nuclide is also radioactive, it will also decay further. Due to the fact that the most common uranium isotopes ^{238}U and ^{235}U have half lives of billions of years, all decay chains in this model have their break-up at the respective uranium isotope.

3.2 Diffusion

Diffusion is a physical equilibrium process, in which molecules, atoms or ions move from sites containing high concentration to sites containing low concentration, caused by Brownian motion. The amount of material, that diffuses through a fictitious unit surface is proportional to the concentration gradient. The underlying three-dimensional equation is also known as *Fick's Law*, and the diffusion constant D can be derived experimentally, where D_W means the diffusion constant in water [8].

$$\frac{\partial c}{\partial t} = D_W \cdot \left(\frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial y^2} + \frac{\partial^2 c}{\partial z^2}\right) = D_W \cdot \nabla^2 c \qquad (2)$$

Migration due to ordinary diffusion in water is a comparably negligible process, because the migration probability for transuranium elements by diffusion is very little, and the migration rate is usually much lower than the velocity of water, i.e. rainwater advection [6], [9].

But there are not only diffusion processes in a body of standing water, diffusion also occurs in **soil matrix**, which is considered the second central process for migration models in the geosphere. This process is quite similar to the process of ordinary diffusion, except that it occurs in the porous space and its possible fillings. The difference between ordinary diffusion and matrix diffusion results in the different system specifications [9], [10].

But the total of all diffusive processes can be summed by the so called impedance factor in a quantitative way. The impedance factor is described as a proportional factor between the effective diffusion constants, which are operational in the soil, and the diffusion coefficient, that operates in free water [10]:

$$D = \gamma \cdot D_W \tag{3}$$

where γ is the impedance factor and D is the combined diffusion constant for all diffusion processes. So, in this migration model for transuranium elements, matrix diffusion, one of the central processes, and ordinary diffusion in standing water, which is almost negligible, are combined in the global diffusion term D.

The diffusive spread is a rather slow process and proceeds only in the area surrounding of the point of release. Due to the fact that diffusion occurs in the complete soil matrix [6], [9], it is treated as a three dimensional process.

3.3 Groundwater Advection

The dominant transport mechanism for solutes in fractured rock is groundwater advection, the third central process for migration models. Advection occurs due to the interconnected network of soil, that provides flowpaths for groundwater and therefore enables the transport of radionuclides [6].

In this migration model, advection is defined as the movement of dissolved particles in the flow of the surrounding water and describes the pollutant dispersal, which moves with the velocity of rainwater through the interconnected network [10].

Radionuclides can migrate with rainwater in various chemical forms and interact with other matter present in water. Transport routes for advective transport by rainwater are formed by fractures which appear in dry soils and, during rainfall, become filled with rainwater. Adhered radionuclides can be transported along a natural incline towards some natural or artificial barrier (i.e. ditch).

The advective term in the model is described in the transport equation by the convective flow, which is caused by material flow with a certain velocitiy $\vec{u}$ and can be defined as:

$$\frac{\partial c}{\partial t} = -\vec{u}\,\nabla c(\vec{r}, t) \tag{4}$$

where $\vec{u}$ is the velocity of the rainwater that flows along the incline to some barrier, and the term $-\nabla c(\vec{r}, t)$ the so called convective term [8].

Advection by rainwater is the dominant migration process for transuranium elements, but it occurs only during rainfall, an important fact, which has also to be considered in the migration model. As advection just takes place at the interface between soil and air, it is treated as a 2-dimensional process.

Most of the sorbing radionuclides exhibit very slow movement in the geosphere environment, with typical estimated travel times from respository depths to the surface environment exceeding half-lives of many radionuclides [6].

Up to now, the migration process has to be described by the diffusion equation and the advective term for migration by rainwater:

$$\frac{\partial c(\vec{r}, t)}{\partial t} = D \cdot \nabla^2 c(\vec{r}, t) - \vec{u}(\vec{r}, t) \nabla c(\vec{r}, t) \tag{5}$$

The chemical mechanisms for radionuclide retention are **sorption** and precipitation. In areas, where radionuclide concentrations can generally be expected to be very low, sorption is the dominant process and precipitation can usually be neglected [6].

3.4 Sorption

Sorption, the fourth central process in the migration model, refers to the action of either absorption or adsorption and means the physical or chemical binding of dissolved particles to the surface soil matrix, fracture surfaces, fracture fillings and pore surfaces of the rock matrix. These retention processes occur at low concentrations at the water-solid interface, by which a solute is bound or released by sites on the surface of minerals and organic matter [6], [10].

In the transport equation, sorption illustrates a sink term and corresponds to the dissolved amount of substance that moves from the solution to the sorbed phase per unit time. Considering sorption processes, the transport equation is extended by a sorption term S:

$$\frac{\partial c(\vec{r}, t)}{\partial t} = D \cdot \nabla^2 c(\vec{r}, t) - \vec{u}(\vec{r}, t) \nabla c(\vec{r}, t) - \underbrace{S}_{\frac{\rho}{\Theta}\frac{\partial c_{sorb}}{\partial t}} \tag{6}$$

where S represents a term that consists of the sorption rate $\frac{\partial c_{sorb}}{\partial t}$, and a more or less constant factor, which describes the situation of the soil: ρ is the closeness of grain and Θ the humidity ratio.

Sorption is usually considered as the reversible attachment of dissolved species to surfaces. In a strongly simplified concept where the partitioning of a solute between a solution and a solid phase is usually described by the linear equilibrium constant K_d. Thus, under equilibrium conditions, and according to the linear sorption model by Henry, the term $\frac{\partial c_{sorb}}{\partial t}$ will be replaced by a term $K_d \frac{\partial c(\vec{r}, t)}{\partial t}$, where K_d is derived experimentally [10]. Transforming equation (6) leads to:

$$\begin{aligned}\frac{\partial c(\vec{r},t)}{\partial t} &= D\cdot\nabla^2 c(\vec{r},t) - \\ &- \vec{u}(\vec{r},t)\nabla c(\vec{r},t) - \frac{\rho}{\Theta}\cdot K_d\cdot\frac{\partial c(\vec{r},t)}{\partial t}\end{aligned}$$

$$\begin{aligned}\frac{\partial c(\vec{r},t)}{\partial t}\cdot(1+\frac{\rho}{\Theta}\cdot K_d) &= D\cdot\nabla^2 c(\vec{r},t) - \\ &- \vec{u}(\vec{r},t)\nabla c(\vec{r},t)\end{aligned}$$

$$\begin{aligned}\frac{\partial c(\vec{r},t)}{\partial t} &= \frac{D}{R}\cdot\nabla^2 c(\vec{r},t) - \\ &- \frac{\vec{u}(\vec{r},t)}{R}\nabla c(\vec{r},t)\end{aligned} \tag{7}$$

The derivation shows how the factor K_d can be replaced by some experimentally determined retardation factor R for retarded advection and retarded diffusion [10]. In the migration model, the retardation factor R is different for different processes (advection, diffusion, adsorption to natural or artificial barriers), and will be termed as R_i, because the K_d-factor is different in soil, water and asphalt.

$$R_i = 1 + \frac{\rho}{\Theta}\cdot K_{d,i} \tag{8}$$

If the advective flow is concentrated in narrow paths, it is said to flow in *channels*. However, narrow channels provide very small areas for interactions between flowing water and soil sediment, and due to their slow flow velocity, travel time is limited and the probability of retention is very small. So, in the migration model, diffusion is the only process where sorption can occur.

Some elements, like iodine, are characterised by relatively weak sorption, whereas others, such as almost all actinides, typically sorb very strongly and are virtually immobile. Actinide chains generally exhibit a high sorption coefficient [6], [9].

In addition to the sorptive sink term a second sink term has to be introduced, which corresponds to adsorption of radionuclides at natural or artificial barriers. This term corresponds to the amount of substance that is dismantled per unit time, where σ represents the dismantling coefficient and c the dissolved concentration of radionuclides [10].

$$\frac{\partial c(\vec{r},t)}{\partial t} = D\cdot\nabla^2 c(\vec{r},t) - \vec{u}(\vec{r},t)\nabla c(\vec{r},t) - \sigma\cdot c(\vec{r},t) \tag{9}$$

Due to the fact that the dismantling coefficient σ represents also some kind of retardation factor, it will also be entered as an R_i in the migration model.

$$R_i = \sigma\cdot c(\vec{r},t) \tag{10}$$

Considering all main processes for radionuclide migration in the environment, the transport equation, a partial differental equation (PDE), will be displayed as follows:

$$\begin{aligned}\frac{\partial c(\vec{r},t)}{\partial t} &= (\frac{D}{R_i}\cdot\nabla^2 c(\vec{r},t) - \frac{\vec{u}(\vec{r},t)}{R_i}\nabla c(\vec{r},t) \\ &- R_i\cdot c(\vec{r},t))\cdot e^{-\lambda t}\end{aligned} \tag{11}$$

4 Modelling Approach

According to the IAEA guidlines, an area should be characterised by in-situ methodologies and systematic sampling, if necessary. Therefore, a special sampling grid for each area has to be designed [1], [2], [3]. Hence, to simulate this sampling process also in the migration model, the modelling approach of cellular automata was chosen.

A cellular automata is a discrete model, which consists of a regular grid of cells, each one in a finite number of states. The grid itself can be in any finite number of dimensions. Time is also discrete, and the state of a certain cell at time t is a function of the states of a finite number of cells (called its neighbourhood) at time $t-1$. These neighbours are a selection of cells relative to the specified cell and do not change. Every cell has the same rule for updating, based on the values in this neighbourhood [11].

For the spreading of transuranium elements by diffusion the von-Neumann neighbourhood was chosen, because of their comparatively slow migration velocity, see figure 4. In a von-Neumann neighbourhood cell C has 4 neighbours in a 2-dimensional grid, which are represented by the cardinal directions, thus 6 neighbours in a 3-dimensional grid. Figure 5 shows, how diffusion is spread within the von-Neumann neighbourhood.

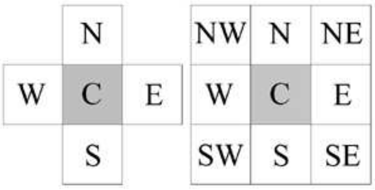

Fig. 4 The von-Neumann neighbourhood on the left, the Moore neighbourhood on the right

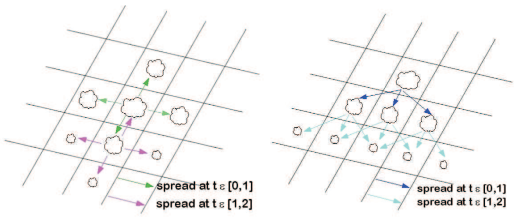

Fig. 5 Diffusion within the von-Neumann neighbourhood on the left, advection within the Moore neighbourhood on the right

For the dispersion by advection, the Moore neighbourhood, see figure 4, was chosen, because migration proceeds fast along the natural incline. That vector component, that has the same direction as the natural incline, drives the strongest force for advection. In a Moore-neighbourhood each cell C has 8 neighbours in a 2-dimensional grid and 26 neighbours in a 3-dimensional grid, see figure 4. Figure 5 illustrates, how advection is spread within the Moore neighbourhood.

The main principle behind this migration model is the conservation of the amount of substance, its physical unit is called mole. Conservation of the amount of substance insists, that not a single mole of transuranics might appear or disappear during migration processes over the time [8]. Figure 6 illustrates how the transport equation is implemented in this migration model, under the assuption of conservation of the amount of substance.

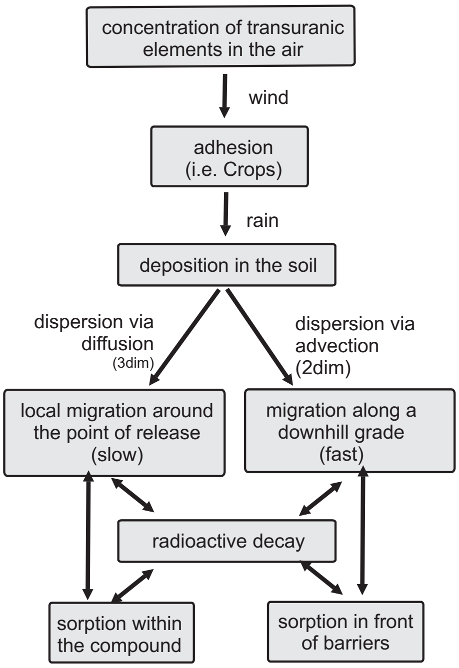

Fig. 6 Schematic Diagram of the Migration Modell

The mole is the base unit that measures an amount of substance and contains Avogadro's number ($6,022 \cdot 10^{23}$) entities, i.e. transuranium particles. Looking at equation (12) for migration the concentration of radionuclides can be replaced by the number of particles N.

$$\begin{aligned} \frac{\partial N(\vec{r},t)}{\partial t} &= (\frac{D}{R_i} \cdot \nabla^2 N(\vec{r},t) - \\ &- \frac{\vec{u}(\vec{r},t)}{R_i} \nabla N(\vec{r},t) - R_i \cdot N(\vec{r},t)) \cdot e^{-\lambda t} \end{aligned} \tag{12}$$

Due to the fact that each radionuclide has its own decay constant, this PDE has to be set up for every radionuclide, that will be considered in this model. The total amount of radionuclides is therefore the sum of all these PDEs.

$$\begin{aligned} \frac{\partial N}{\partial t} &= \frac{\partial N_{^{241}Pu}}{\partial t} + \frac{\partial N_{^{240}Pu}}{\partial t} + \\ &+ \frac{\partial N_{^{239}Pu}}{\partial t} + \frac{\partial N_{^{238}Pu}}{\partial t} + \frac{\partial N_{^{241}Am}}{\partial t} \end{aligned} \tag{13}$$

To use these PDEs in a discrete cellular automata model, the underlying transport equations have to be transformed to their discrete form. Doing this, some kind of anti-derivative of the PDEs are created.

$$\begin{aligned} N(x_0, t=1) &= \int dt \frac{D}{R_i} \cdot \nabla^2 N \cdot e^{-\lambda \cdot t} - \\ &- \int dt (\frac{\vec{u}}{R_i} \cdot \nabla N - R_i \cdot N) \cdot e^{-\lambda \cdot t} \end{aligned} \tag{14}$$

The fractions containg the retardation factor R_i are replaced by the constant α_i, which has the dimension of the retardation factor times migration probability times the time. Considering the average of rainy days a year, x, the anti-derivative results in a discrete equation that looks like

$$\begin{aligned} N(t) &= N_{(t-1)}(1 - \alpha_{i\ diff} - \alpha_{i\ adv} \frac{x}{365}) \\ &\cdot\ e^{-\lambda t} + \alpha_{i\ sink} \end{aligned} \tag{15}$$

Considering the different velocities of migration, the different sorption processes and the neighbourhood of the cell considered which contains a finite number of transuranium particles, the discrete migration equation would be as shown in the following as an example for diffusion, where the position vector $\vec{r}$ now is replaced by cell number X_i:

$$\begin{aligned} c(x_0, t=1) &= c(X_0(t_0)) - 6\alpha_i c(X_0(t_0)) + \\ &+ \alpha_i \cdot [c(X_1(t_0)) + c(X_2(t_0)) + \\ &+ c(X_3(t_0)) + c(X_4(t_0)) + \\ &+ c(X_5(t_0)) + c(X_6(t_0))] \end{aligned} \tag{16}$$

Considering also the other processes, advection, sorption and the sink term, the discretised equation will become much longer and more complex, given that the advective term needs 8 directions for its spread.

Now the big advantage of discretising the problem can be seen. If the number of any radionuclide in a certain cell changes due to radioactive decay, the number of decayed particles can be calculated by the decay equation and will be shifted towards the proper nuclide.

For instance, there exist $N\ ^{241}Pu$ nuclides and N_d ^{241}Pu have decayed. But ^{241}Pu can only decay to ^{241}Am [7], so the decayed $N_d\ ^{241}Pu$ are $N_a\ ^{241}Am$ per definition. Thus, the amount of $N_a\ ^{241}Am$ will be added to the amount of $N\ ^{241}Am$ particles in that cell, and the conservation of the amount of substance is ensured.

5 Implementation to MATLAB

A simulated area is divided into a regular grid of cells, each in a state of no transuranium concentration. The size of that area is $49 \cdot 49$ cells and the depth is 15 cells. One time unit is set to $86400s$, which is less than the decay time of most of the radonuclides considered [6].

At time $t - 1$ an activity of i.e. $3,7 \cdot 10^{10}$ $\mathrm{Bq}^{241}Pu$ is released in the middle of the simulated area. During the next time steps, migration takes place. Fig. 7 describes how migration takes place over this area and how the activity concentration of Pu decreases. As this figure only demonstrates the behaviour of ^{241}Pu, the concentrations of the amount of substance can not be taken into account.

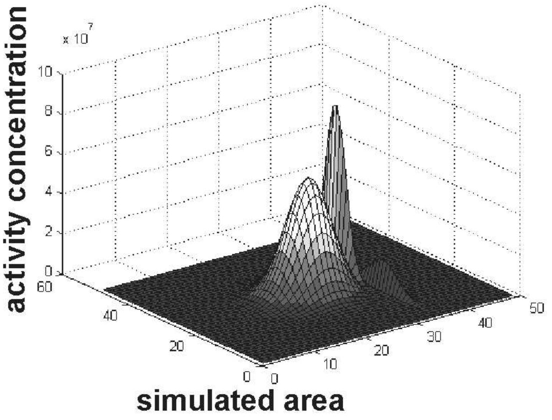

Fig. 7 Migration from the middle of the area to a ditch, where radionuclides become concentrated

6 Experimental

At the Sibersdorf site, there exists an area where historic deposition of transuranium nuclides has occured, revealed by random sampling at the site.

Now, detailed sampling on this area has been performed in order to compare the migration of the model to its natural analogue and to identify the various parameters α_i in the discretised transport equation.

This area is approx. 4 m wide and has a length of about 100 m. The distance from one knot of the sampling grid to the next is about 1 m. On each knot a soil sample with an approximate volume of 450 ml fresh matter is collected and measured by gamma spectrometry for its Am and Cs activity. Up to know, about 50 samples are analysed. The analysis results and also the spread can be seen in fig 8.

Both nuclides, plutonium and americium are quite immobile and will remain together over long periods [9]. But also plutonium and cesium might stay together for decades [12], thus having the americium activity and the cesium activity of a certain sample might provide an indication of the plutonium activity, which can not be measured by gamma spectrometry.

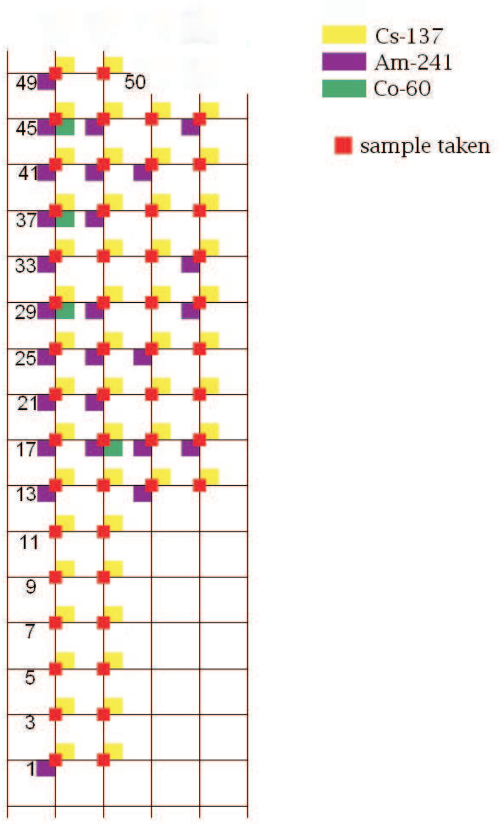

Fig. 8 Spreading of Am and Cs

To identify the parameters α_i for the migration model, the nuclide vector, which contains all the other transuranium isotopes of interest, which cannot be measured by gamma spectrometry, but by LSC or alpha spectrometry shall be established. The isotope ^{241}Am is the only one that can be seen in a gamma spectrum, and will therefore act as the lead nuclide.

The soil samples are dried in a cabinet drier at 450 °C for about eight hours and afterwards grained to a grain size less than 3 μm. 50 g of the sample are weighed in a round bulb and 250 ml of a 8 molar HNO_3 are added. The mixture is boiled for about two days, until the majority of transuranic particles, which are in the soil sample, are dissolved in the acid. The acid-soil sample is filtered and the filtrate will be used to separate transuranium isotopes with the help of ion exchange raisins.

7 Conclusions

Up to know, this migration model is still work in progress, but it appears that it's a reasonable simplification of all the complex and complicated transport mechansims, which describe migrational behaviour of real sites rather well.

Even the modelling approach of using cellular automata and discretising the transport equation, has proved to be sensible, because the model behaves stable, even under rather unusual initial conditions.

Now, systematic sampling and analysis are performed in order to achieve good basic data for identifying the migration model's parameters, α_i.

8 Acknowledgements

The authors thank R. Essbüchl for his assisstance in the determination of americium and cesium in soil samples by gamma spectrometry.

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