RADIONUCLIDES TRANSFER IN THE ADJACENT AREA OF THE NATIONAL RADIOACTIVE WASTE REPOSITORY, BAITA BIHOR

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Abstract: A simulation of the ^{226}Ra radionuclides transfer through a conceptual model of double porosity media was run for the National Radioactive Waste Repository site at Baita Bihor. The radionuclides transfer simulation was carried out using the Tang Analytical Solution. Starting from the mass transport equation, an explicit solution of the concentration variation through the fracture and through the porous media, and respectively the penetration depth of the radionuclides in the matrix was obtained.

Key words: Tang analytical solution, radionuclides transfer simulation, radioactive decay, concentration distributions.

1. Introduction

Baita Bihor National Radioactive Waste Repository (NRWR) is located in Bihor Mountains, in a former uranium mine (Galleries 50, and 53), 2 km E from the mining village of Baita Plai, Bihor County.

The scope of this paper is the simulation of the ^{226}Ra radionuclides transfer through the fracture – porous matrix system, estimating the exit (realized) concentration, and the depth of penetration of radionuclides in the matrix.

2. Geological conditions

Baita Bihor Repository is located within an area with a complex geological and structural setting, which is the result of multiple tectonic episodes, magmatic intrusions, and hydrothermal activity.

The main geological and structural characteristics of Baita Bihor Repository area are as follows (*Watson S. P. et al., 2006)*:

- The area has been the subject of multiple episodes of overthrusting of the older rocks over the younger ones.
- The area is characterized by a faulting system, striking NNW-SSE, and minor reverse faults.
- In the vicinity of the repository, the magmatic activity is associated with a faulting process. Diabase, and diorite dykes were found within the meta-sediments. The extensive mineralization in this area it is largely associated with hydrothermal alteration resulting from the magmatic activity.
- The repository lies within meta-sandstone, mica schist, and crystalline meta-carbonate rocks (marble and skarn). The meta-carbonate produced a karst drainage system in the area to the NW of the repository.
- The valleys downstream of Baita Sat/Nucet are covered with Neogene and Quaternary alluvial deposits, typical of areas downstream of an active orogen.

3. Hydrological conditions

Baita Bihor NRWR is located in the upper part of the Crisul Negru hydrological basin.

The main tributary of the Crisul Negru River, in Baita Bihor NRWR area, is Crisul Baita, whose main tributary is Valea Baita River.

The repository is located between Mic Creek (with permanent flow in the lower part of its course) to SE and Lupului Creek (with intermittent flow). A second creek (Stancii Creek) drains the central part of the Baita open pit mine.

4. Hydrogeological conditions

According to the Management Plan of Crisurile Hydrographic Area, 2009 – 2015, National Administration Apele Romane, Crisuri Water Catchment, Baita Bihor NRWR area belongs to Vascau ROCR05 groundwater body, Codru – Moma Mountains.

Vascau ROCR05 groundwater body, Codru – Moma Mountains, is characterized by a karst – fissural aquifer, quartered in Triassic limestone and dolomite *(National Administration Apele Romane, Crisuri Water Catchment, 2009)*.

The hydrogeology of the area is dominated by the presence of the hydro – karst system, with divergent feeding. The NRWR area is included in Crisul Baita karst system, which is located in the upper hydrological basin of the valley with the same name. As a result of intense tectonic processes that occurred in the area, the morphology

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of the terrain is rough (*Oraseanu I. et al., 1991, Oraseanu I., 1999).*

The outcrops of carbonate deposits have no permanent surface flows of water. The small streams flowing on the slopes that are not limestone, infiltrate in the karst system when they reach over the carbonate surface (*Oraseanu I. et al., 1991, Oraseanu I., 1999).*

Lithological characteristics

The Baita Bihor NRWR galleries are dough in Permian strata of Arieseni Nappe (*Watson S. P. et al.*, *2006*). The uppermost rocks intercepted by the repository galleries, are metamorphosed sandstones and phyllites which were penetrated by magmatic intrusions, preponderant diabase dykes. In the repository area, the metamorphosed sandstones are, in general, layered granular rocks with intrinsic porosity and lower permeability.

Metasomatic, and albitisation processes are frequent, which caused alterations on foliation and joints.

Structural details

The repository area is bounded toward NW by Lupului Fault System (LFS), and toward SE by the Central Fault System (CFS), with dip angles of 50 - 80°, in both systems. Along LFS the tectonic contact between diabase, to NW, and meta-sandstone, at SE, is visible. At the surface, CFS separates the meta-sandstone outcrops from the black phyllites (*Watson S. P. et al., 2006)*.

The rocks of the repository area are highly fractured, with minor displacement. The discontinuities in the rocks have been classified by *Watson S. P. et al., 2006* in three groups:

- **S0** – the original bedding planes, which can play a major role in the movement of the groundwater;
- **S1** – metamorphic foliation, which is almost coincident with the bedding;
- **S2 –** the main faulting/fracturing system.

Most of the fissures and fractures from repository galleries belong to the group (**S2**) which according with the fracture prevalent direction was divided into four systems as follows:

<u>F1 System</u> – associated with LFS;

<u>F2 System</u> – oriented NE - SW which is less important;

<u>F3 System</u> – This system has a low frequency, but has an important hydrogeological significance. In the repository area this system is represented by a fault oriented NNW – SSE, which is transverse to the galleries.

<u>F4 System</u> – oriented NW – SE represents the main tectonisation direction from the repository galleries. This system (together with its supporting micro faults and fissures) occurs in almost all repository galleries.

Location of the regional and local faulting systems from Baita Bihor NRWR area is presented in the Figure 1.

Watson S. P. et al., 2006 revealed that the main directions of regional faulting and fracturing system observed at the surface are similar with the one identified underground, and influence the water infiltration-drainage process.

The aquifer system of the repository area can be compared with a flow with double porosity system, composed by a micro-fractured matrix with lower permeability cut by faults higher transitive, and the fractured areas.

Based on these observations a generic model composed of a fault which intersects the repository, and is in direct contact with the porous matrix (micro-fractured) was considered.

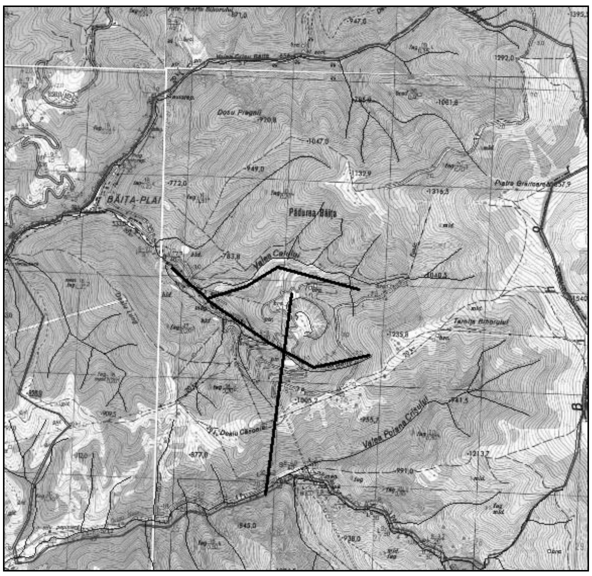

Fig. 1 **Location of the regional and local faulting systems from Baita Bihor NRWR area, Ministry of Defense, Military Topographic Directorate**

5. Tang Analytical Solution

Tang Analytical Solution (TAS) *(Tang et al., 1981)* is based on the assumption of one infinite fracture, with plan-parallel sides, which is in direct contact with the porous matrix.

The TAS model is based on the following geometric properties and hydraulic processes, taking place in the fracture – porous matrix system (Figure 2):

- The modeled area is in the saturated zone of the vertical fracture – porous matrix system.
- At the surface, near to the fracture, there is a radionuclides source with a constant concentration.
- The groundwater flow rate, through the fracture, is constant.
- The width of the fracture is much smaller than its length.

The permeability of the porous matrix is low, and the transport of radionuclides in the matrix is mainly by molecular diffusion. The transport from the fracture into the porous matrix occurs through diffusion, because of high concentration gradients between the fracture and the matrix. In the case of a radionuclide, the tracer concentration is influenced also by the radioactive decay process.

The transport along the fracture is manly advective and dispersive, being much faster than in the matrix.

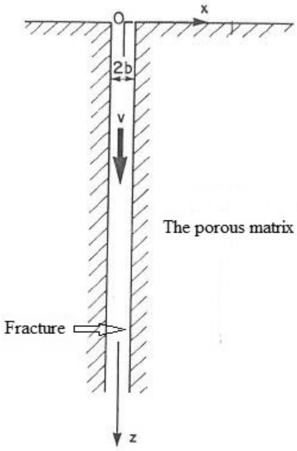

Fig. 2 **Fracture – porous matrix system, after Sudicky et al., 1982**

6. The Conceptual Model

In NRWR case, the considered conceptual model was a conservative one, in that the degree of matrix saturation is very high, the capillary pressure being under penetration pressure. Thus, the "buffer" character of the unsaturated zone is reduced to the minimum.

The simulation transport of radionuclides in the area DNDR was run through FEFLOW software package *(Diersch H. J.G., 2005)* which analyses flow and transport by finite element method.

The simulation was run in conditions of a plan - vertical problem, the studied domain consist of a major fracture, extending to the limit of the saturated zone, which is in direct contact with the porous matrix. The domain extension was 30m horizontally, and 70m vertically.

The ^{226}Ra radionuclides transfer was simulated considering the main transport mechanisms, with the values of the hydrogeological parameters for porous matrix and for the fracture, uniformly distributed throughout the entire domain *(Paraschivoiu D.L., et al., 2014)*.

Porous matrix:

- Matrix effective porosity 15 %;
- Diffusion coefficient 10^{-9} m^2/s;
- Longitudinal dispersivity 250 m;
- Transversal dispersivity 10 m;
- Matrix conductivity $72x10^{-8}$ m/s.

Fracture:

- Fracture opening 1 mm;
- Longitudinal dispersivity 20 m;
- Transversal dispersivity 0 m;
- Lateral surface of the fracture $5x10^{-4}$ m^2;
- Fracture conductivity 0,817 m/s.

The flow boundary condition was given as an imposed flux at the end of the fracture with the mean value of the infiltration flux of W = 0,100 mm/day *(*Danchiv A. et al.*, 2007)*, as resulted from water balance performed for the catchment area of the repository.

The radionuclides transport was simulated imposing to the fracture the source term provided by SNC Pitesti as an imposed concentration.

The major fault was modeled with discrete elements, vertical plans, which penetrate the rock layers throughout their thickness.

To observe the evolution of the concentration of radionuclides during transfer through the fracture - porous matrix system, four observation points located along the fault were considered, and restitution curves were plotted for each point:

- Observation point 1: at gallery depth (the source);
- Observation point 2: at 20m deep;
- Observation point 3: at 50m deep;
- Observation point 4: at 75m deep (at the unsaturated medium boundary).

Two situations were analyzed, to find the percentage with which mechanical transfer mechanisms and the radioactive decay influence the ^{226}Ra isotopes concentration within the rock:

- The ^{226}Ra isotopes transfer in the double porosity media taking into account the decay process;

- The transfer of one fictive-ideal tracer, having its source identical with the ^{226}Ra isotope, ignoring the decay process through the double porosity media.

The ^{226}Ra radionuclides transfer was simulated using variable time steps. The results of the simulation were presented at time intervals equal with 0.5, 1.0 and 1.5 half-lives (T½). Thus, the ^{226}Ra radionuclides transfer was presented at 800, 1.600 and 3.200 years from the release from the source. Finally a spatial distribution of the concentration was obtained, and the mass flow balance at different time intervals.

7. The ^{226}Ra isotopes transfer

The half-life and the decay constant were taken into account for the simulation of the ^{226}Ra isotopes: the half-life T½ = 1.600 years (the ^{226}Ra half-life is much higher than the residence time into the rock). It results a decay constant $\lambda = \ln 2 / T_{1/2} = 1.36 \times 10^{-11}$ s^{-1}.

The ^{226}Ra isotope transfer

For each observation point, restitution curves were made, the main characteristics beeing summarized in the Table 1. The T_{max} and C_{max} represent the time period in which the maximum concentration value is reached for each observation point, respectively the maximum concentration registered for that point.

Table 1 **The maximum concentrations of ^{226}Ra radionuclides in the observation points**

Obs. point	Depth of obs. point (m)	T_{max} (years)	C_{max} (Bq/m^3)
1	source	50	7.49E+08
2	20	1.550,9	2.01E+05
3	50	4.918,8	2.32E+03
4	75	9.941,6	1.23E+02

The Ra radionuclides penetrate the rocks-mass with significant concentration of 7.49E+08 Bq/m^3. The penetration process is accelerated at the upper part of the rock-mass, and at the beginning of the transfer process when the concentration gradient between the fault and the rock-mass it is the highest. It was observed that the penetration area of radionuclides increases with the increasing time after release. After 1.551 years reaching 20m deep, at 4.919 years after release reaching 50m deep, and after 9.9 years reaching a depth of 75m.

Within 50 years from the launch of isotopes at the source, the maximum value of the ^{226}Ra isotope concentration is reached, 7.49E+08 Bq/m^3. After 1.551 years from the release, the concentration decreases by three orders of magnitude in the observation point 2 (located at 20m deep), to the value of 2.01E+05 Bq/m^3. In observation point 3, (located at 50m deep) after 4.919 years from the release, the concentrations decreases with an additional two orders of magnitude, to the value of 2.32E+03 Bq/m^3. As expected, the concentration decreases with increasing depth. In the observation point 4, the concentration reaches the value of 1.23E+02 Bq/m^3, after 9.941,6 years from the release time.

In brief, the ^{226}Ra isotopes concentration decreases by six orders of magnitudes in 9.9 years, at 75m depth from the source.

The transfer of an ideal-tracer, from the same source as the ^{226}Ra isotope, ignoring the decay process

For each observation point, restitution curves were made, the main characteristics beeing summarized in the Table 2. The time period in which the maximum concentration value is reached for an observation point, is noted with T_{max}. The registered maximum concentration is noted with C_{max} .

Table 2 **The maximum concentrations of an ideal-tracer for each observation point (without radioactive decay)**

Obs. point	Depth of obs. point (m)	T_{max} (years)	C_{max} (Bq/m^3)
1	source	50	7.49E+08
2	20	2.926,0	8.07E+05
3	50	43.370,5	2.32E+05
4	75	100.316,7	1.94E+05

In observation point 2 (located at 20m depth), after 2.926,0 years, the concentration decreases by three orders of magnitude, to the value of 8.07E+05 Bq/m^3. In observation point 3 (located at 50m depth), after 43.370,5 years from the release, the concentration decreases to the value of 2.32E+05 Bq/m^3. A concentration of 1.94E+05 Bq/m^3 was reached after 100.316,7 years from the release, in observation point 4 (located at 75m depth).

Ignoring the decay process, the ideal-tracer concentration, decreases by only three orders of magnitude in 100.316,7 years, before it reaches 75m depth.

Unlike the previous version, after reaching the maximum value, the ideal-tracer concentration decreases very slowly, as the mass of the tracer is dispersed over a long time period. This aspect characterizes the predominant diffusive processes. An important mass of radionuclides diffused into the matrix because of the high concentration gradient between the fault and the porous matrix. The transfer is very slow due to the low value of diffusion coefficient of the rock. In the upper part of the domain, where the concentration gradient is maximum, the diffusion intensity in the rock mass is highest. As radionuclides enter into the matrix, the gradient decreases and the transfer process is slowed down. This explains the large difference between the concentration at the source and the concentration registered in the observation point 2, respectively the small differences between the maximum concentrations registered in the observation points 2, 3, and 4.

The trend of the maximum concentration for all four observation points, with respect to depth can be observed in Figures 3 and 4.

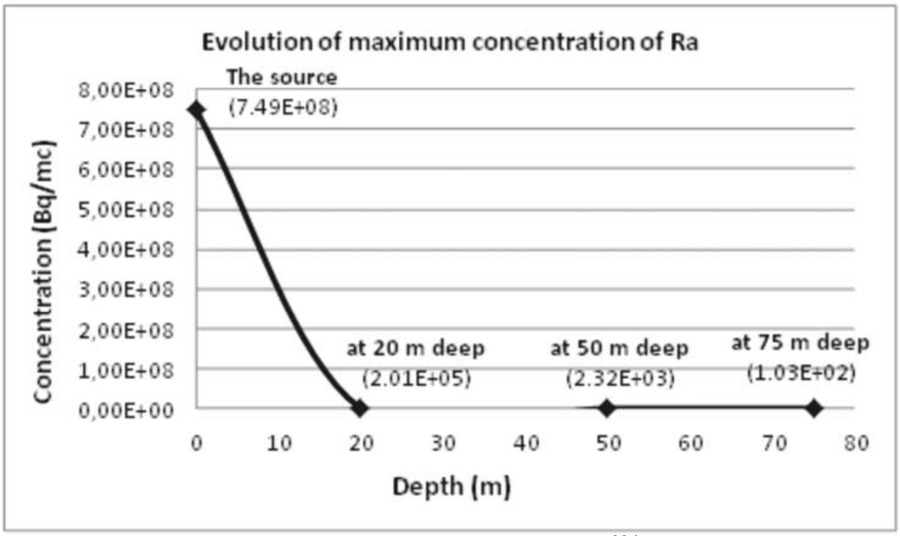

Fig. 3 **The evolution of the maximum concentration of ^{226}Ra in relation to the depth in that 4 observation points**

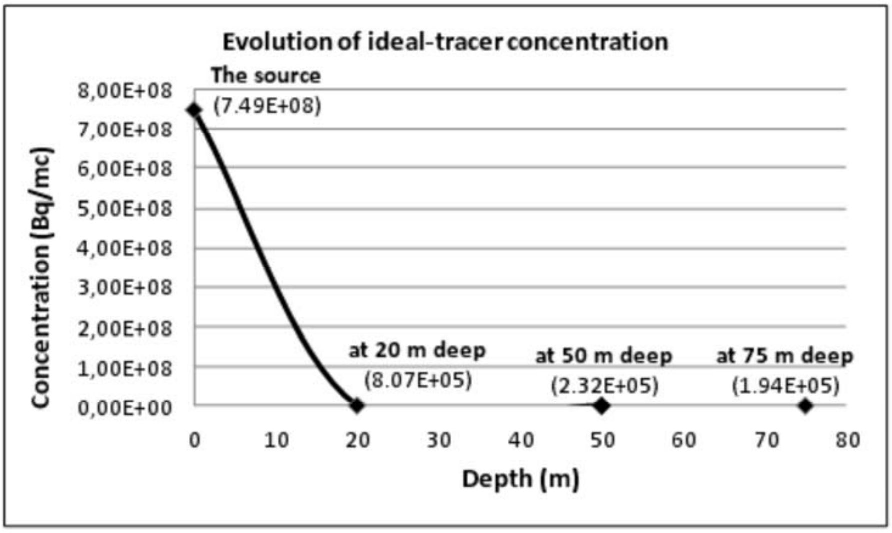

Fig. 4 **The evolution of the ideal-tracer concentration in relation to the depth in that 4 observation points (without radioactive decay)**

From Figures 3 and 4 it can be noted a large percentage of radioactive decay contribution. In the observation point 4, the maximum concentration is with three orders of magnitude lower when considering radioactive decay, even if the rock thickness at this site is relatively small, and the half-life is 1.600 years.

Even in these conservative circumstances, of the model, the role of the buffer zone is important, the radionuclides exiting the buffer zone at a value of about 0.16×10^{-4} Bq/m^3 of the initial concentration registered at the source.

As previously noted, the radioactive decay of ^{226}Ra leads to a decrease of the isotope transit time through the porous matrix, and thus to a concentration decrease by several orders of magnitude. The radioactive decay process is much more evident in observation points 3 and 4, located at greater depths.

For the observation point 3, the maximum concentration registered of 226Ra, was 2.32E+03 Bq/m^3, which is with two orders of magnitude smaller than in the case of the ideal-tracer (2.32E+05 Bq/m^3).

For the observation point 4, the maximum concentration registered, when ^{226}Ra was considered had a value of 1.23E+02 Bq/m^3, which is three orders of magnitude smaller compared with the simulation of the ideal tracer (1.94E+05 Bq/m^3).

Spatial distribution of ^{226}Ra concentrations

To highlight the relationship fault-porous matrix, the spatial distribution of ^{226}Ra concentrations were represented at different time intervals (800, 1.600, and 3.200 years, respectively 0.5, 1.0, and 2.0 half-lives). These time intervals were chosen because the half-life of ^{226}Ra is much longer than the residence time in the rock. The spatial distribution of ^{226}Ra is presented in the Figures 5, 6, and 7.

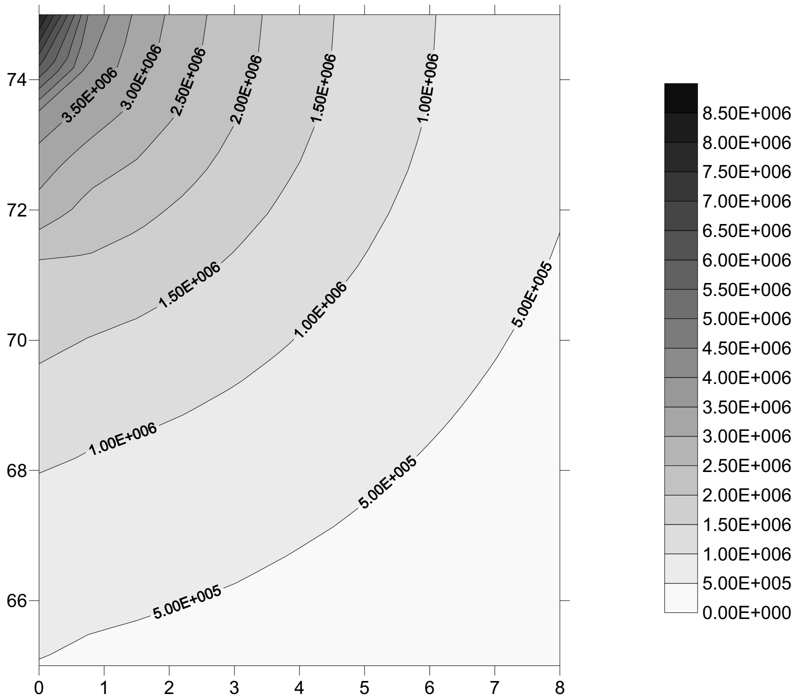

Fig. 5 **The distribution of ^{226}Ra isotope concentration after 800 years (Bq/m^3), Paraschivoiu D.L., et al., 2014**

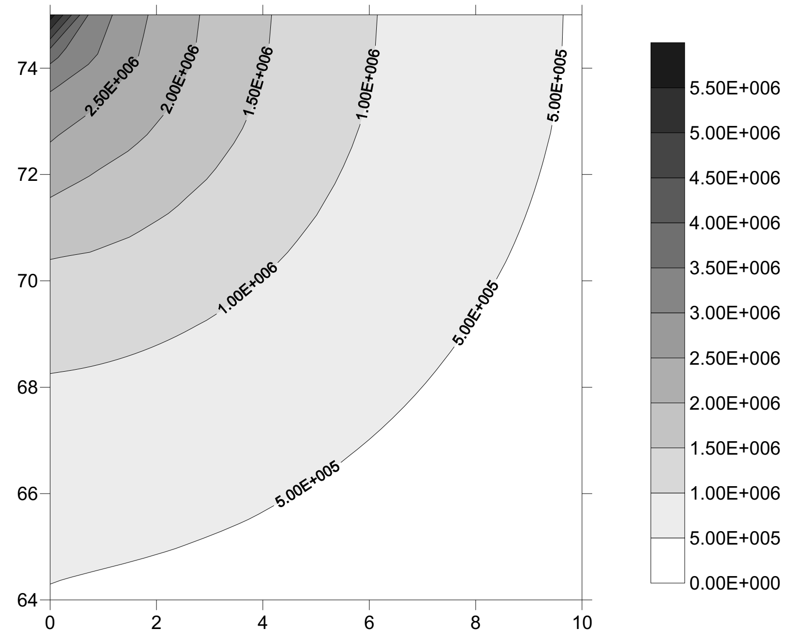

Fig. 6 **The distribution of ^{226}Ra isotope concentration after 1.600 years (Bq/m^3), Paraschivoiu D.L., et al., 2014**

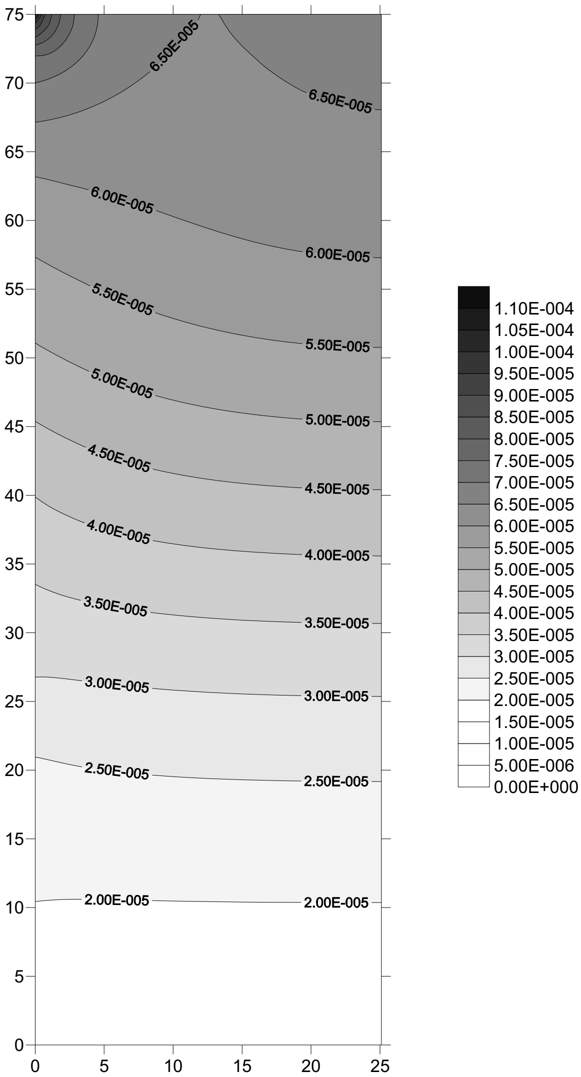

Fig. 7 **The distribution of ^{226}Ra isotope concentration after 3.200 years (Bq/m^3), Paraschivoiu D.L., et al., 2014**

Following the analysis of these simulations it can be observed that:

- After 800 years the radionuclides reach a depth of only 10m, with a concentration of 5.00E+005 Bq/m^3.
- After a half-live (1600 years), the ^{226}Ra isotope concentration in the upper part of the repository, up to 11 meters depth, is comparable with the concentration in the lower parts of the rock layers, at a depth greater than 11 meters, for the 800 years simulation.
- After 3200 years, two half-lives, the radionuclides concentration becomes negligible at a depth of 65 m.

8. Conclusions

The ^{226}Ra radionuclide transfer is manly advective, through the fracture system and diffusive through the porous matrix.

Diffusion transport mechanisms from the fracture into the porous matrix, represents an effective attenuation mechanism that can remove the contaminant mass, thus delaying the radionuclide penetration depth in the adjacent area of the repository. This is caused by both the radioactive decay and the diffusion of radionuclides from the fault into the micro-fissural matrix. The matrix acts as a radionuclide storage system because of the diffusion process. Advection transport through the porous matrix is negligible due to high permeability differences between the fracture and the matrix.

For an infiltration flux with an average of 0.1 mm/day (the multi-annual average value), ^{226}Ra isotopes leave the repository area with a value of approximately 6 orders of magnitude lower compared with the values at the source. After two half-lives, 3200 years, from the release time, the ^{226}Ra radionuclides concentrations become negligible at 65 m depth.

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