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Sensitivity Analysis of Geological Parameters Influencing a Solute Transport from a Deep Repository of Spent Nuclear Fuel

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Abstract: When evaluating Nuclear Waste DGR Safety, it is necessary to confirm its safety in a long run and above all its safety towards the biosphere which is more precisely that the biosphere will not be in any hazard caused by radioactive substances. With the aid of geologists, a model of a hypothetical area was elaborated and described with the use of geological and hydrogeological parameters. The volume of isotopes released out of the massif at the borderline of the near/far field from the DGR was determined. The paper results showed that ground water flow and transport of substances within the area were first to be determined. The Flow123D SW was used for the determination. The resulting outcome represents a determination of transported substances concentration depending on time. The disadvantage of the model is the fact that all the input parameters were set deterministically. The problem is solved by using the sensitivity analysis (changing the input parameters) or using the Monte Carlo Method. The major results are: calculations of the radionuclide concentrations in the elements depending on time and determination of parameters that have the biggest impact on the sensitivity of the whole model.

Key words: Deep repository waste, safety model, Flow123D, sensitivity analysis.

1. Introduction

In the Czech Republic, there are two nuclear power plants: Dukovany and Temelin (with six blocks) which produce highly-active nuclear waste. The waste has to be handled in a long run well to guarantee that it will not be misused anytime and in the same time, to minimize risks for biotopes. Preparation-work and the future realization of such a Safe Nuclear Waste (Deep Geological Repository—DGR have been carried by the DGR Authority of the Czech Republic (Sprava Radioactive Waste—SURAO) [1-3].

The basic concept of the nuclear waste depository is described by Vokal, A. et al. [2, 4-7]—"The Reference Project of a Nuclear Waste DGR in a Hypothetical Locality and Its Actualization". The idea is that nuclear waste will be deposited in granite massive underground in the depth of about 500 m (in the Czech Republic). Now, suitable localities (that have been selected) are being checked and passed through for a detailed geological and hydrogeological research. The own building of such a DGR is forecasted for the year 2040. The hot issue of nuclear waste DGR represents of course a complex problem—combined with THMC (thermo-hydro-mechanical and chemical) processes.

Calculations on ground water flow and on dissolved substances transport are based on a hypothetical locality in the Melechov Massive. The locality is not determined for the building of the deep geological repository, but a detailed geological and hydrological research had been carried there in the past. To calculate ground water flow and substances transport, the software (SW) Flow123D was used [8-11]. The main advantage of the software Flow123D is that it processes data on flow and transport parameters on the

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3 dimensional areas which is divided into 2 dimensional and 3 dimensional elements. The resulting output of a task on the flow is the determination of velocity and a direction of the flow and the determination of volume of a substance coming over the walls of the elements. Analogously, the resulting output of a task on the substance transport is the determination of a dissolved substance concentration (volume activity) depending on time in individual elements.

The paper deals with an analysis of critical radioactive substances transport out of a nuclear waste DGR. A 3D model of the area was designed and it is described with 37,000 elements (where the 2D elements represent fissure and the 3D elements represent the massive). The task on flow and substances transport was calculated with the SW Flow123D. The geological and hydro-geological parameters at the input corresponded with the geological and hydrogeological research which had been carried out there in the Melechov Massive in the past. The Melechov Model contains the following parameters:

- hydraulic conductivity of rocks and fissures;
- porosity of rocks and fissures zones;
- fissures opening.

The substances transport is described with a real model containing the following:

- Time-variable radioactive substances release out of the near/far field border. Results on quantity of the released radioactive substances were detected using the software GoldSim. All of the critical isotopes according to Karlsson, S. and Bergström, U. [2, 16] were covered;
- Radioactive degradation of substances [16];
- Sorption parameters K_d and diffusion coefficients D_e of transported radionuclides [17-19].

The model did not consider the maximal dissolution of a substance in the water as the quantity of released radioactive substances is about 2 $g{\cdot}yr^{-1}$ year maximally. The output of the task is the determination of the substance concentration (1E-9 $g{\cdot}m^{-3}$) re-calculated in volume activity ($Bq{\cdot}m^{-3}$) in individual elements—based on time. A special stress is put on the ground elements where radioactive substances penetrate into biotopes. To calculate the "received" dose (μSv), there was a scenario based on a presumption that a human drank water from the ground (of the area) in the quantity of 1 m^3 a year.

A sensitivity analysis of the model was designed with a parameter change. The analysis is for finding parameters that have the most impact on the concentration change of the selected elements —mainly the ground ones. The following parameters were changed: hydraulic connectivity of rocks and fissures, rocks porosity, opening of fissures and—for each of the transported substances—sorption K_d. The changes describe changed hydrogeological conditions of the researched area in a long run. The output of the sensitivity analysis is a table where all of the stated parameters are shown, those which have the most impacts on the increased concentration of substances on the ground elements.

The paper extends case studies issued before with the following:

- time-variable feature of released isotopes;
- radioactive degradation of substances;
- implementation of sorption and diffusion coefficients;
- calculation for more substances;
- results of re-calculation of received dose (μSv) for the selected scenario.

2. Materials and Method

The designed model of the DGR is situated in the locality of the Melechov Massive which located about 150 km to the Southeast from Prague.

In order to perform the desired analysis, a mathematical mesh was formed describing the massif of Melechov. The mesh covers the area of about 10,000 m × 10,000 m × 1,300 m. The model mesh was created basing on geologic data [14, 15]. The suggested model contains a few vertical and horizontal rift fissures and 40 types of materials display different physical characteristics like hydraulic conductivity.

For the Melechov Massive, the geometry was prepared first. Then, there generated a calculation mesh of the system. Calculation of simulating task was used software Flow123D. The mesh contains in total 7,174 nodes and 37,069 elements. There are 2,140 ground elements where a transport of radiation into the biosphere can happen. The challenge was to specify (make more accurate) the distribution function of concentrations in the elements of the area.

A geological research had been carried in the area in the past. The results of the research helped to set types of rocks, the location of fissures and to determine their hydraulic conductivity. Later, the following was discovered:

- its geological structure—porous massif or fractural zones of the rock;
- types of rocks—granit of melechov type, granit of kout type, granit of Lipnice type and gneiss.

Then, there were considered different physical features of the environment according to the depth analysis of the area for the depths of 75, 150, 400, 600, 800 and more than 800 m. Input details of hydraulic conductivity for a particular type of a rock and fissures are stated in Table 1. To set the boundary condition, three basic parts of boundaries were stated within the simulated area. The boundary condition and hydraulic conductivity in the individual parts within the simulated massif were calibrated to:

- the piezometric height in the upper layer of the model area at the level of underground water;
- one percent of rain level deeper than 800 m under the ground.

Sorption parameters K_d and diffusion coefficients D_e of substances were taken from a reference project. The values of those parameters are stated in Table 2.

Another input into the model was substance quantity released from the near to the far field in time. A conservative case was considered—that was the

Table 1 Hydraulic conductivity of particular type of rock and rifts (mm·year^{-1}).

Type of rock				
Depth (m)	Melechov type	Kout type	Lipnice type	Gneiss
75	18,000	5,400	1,800	360
150	2,880	860	288	57.6
400	288	43	28.8	5.76
600	28.8	8.6	2.88	0.36
800	5.4	1.53	0.54	0.072
> 800	1.8	0.324	0.12	0.012
Vertical rifts				
Depth (m)		Without specification of rock		
75		14,400		
150		7,200		
400		7,200		
600		3,600		
800		1,800		
> 800		720		
Horizontal rifts				
Type of rock		Hydraulic conductivity		
Melechov type		72,000		
Kout type		36,000		
Lipnice type		36,000		
Gneiss		18,000		

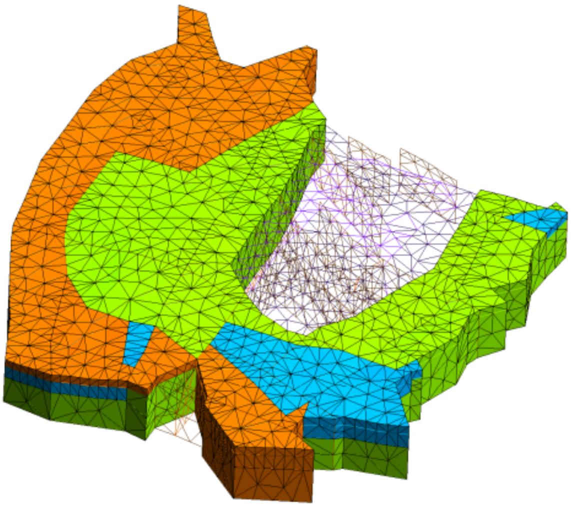


Fig. 1 Visualization of prepared geometry in the software GMSH (three dimensional finite element mesh generator).

container damage in the time of 20,000 years. A concrete plug is considered to be around the container with a bentonite package preventing immediate contamination of ground water. The calculation of the quantity of released substances was done through the simulation SW GoldSim. Fig. 2 shows a graph describing critical isotopes release in time. Calculations of different variants of container damage were done in the analysis. Those scenarios were less conservative and that is why they are not mentioned here.

3. Results and Discussion

3.1 Basic Model Results

A model analysis, which was performed using the SW Flow123D helped to determine the state of concentration (a volume activity) in time for all the elements used in the calculated mesh. Within the area at the beginning of the simulation, there is supposed to

Table 2 Sorption parameters K_d and diffusion coefficients D_e and the maximal dissolution of substances.

Substances	K_d ($m^3 \cdot kg^{-1}$)	D_e ($m^2 \cdot s^{-1}$)	Dissolution ($mol \cdot m^{-3}$)
C14	0.0005	5.00 E-15	Unlimited
Cl36	0	8.00 E-15	Unlimited
Ni59	0.01	2.80 E-14	
Se79	0.0005	4.00 E-14	2.59 E-6
Mo93	0.0005	1.00 E-15	1.00 E-03
I129	0	8.00 E-15	Unlimited
Cs135	0.01	9.00 E-13	Unlimited
Ra226	0.01	3.70 E-14	1.2 E-1

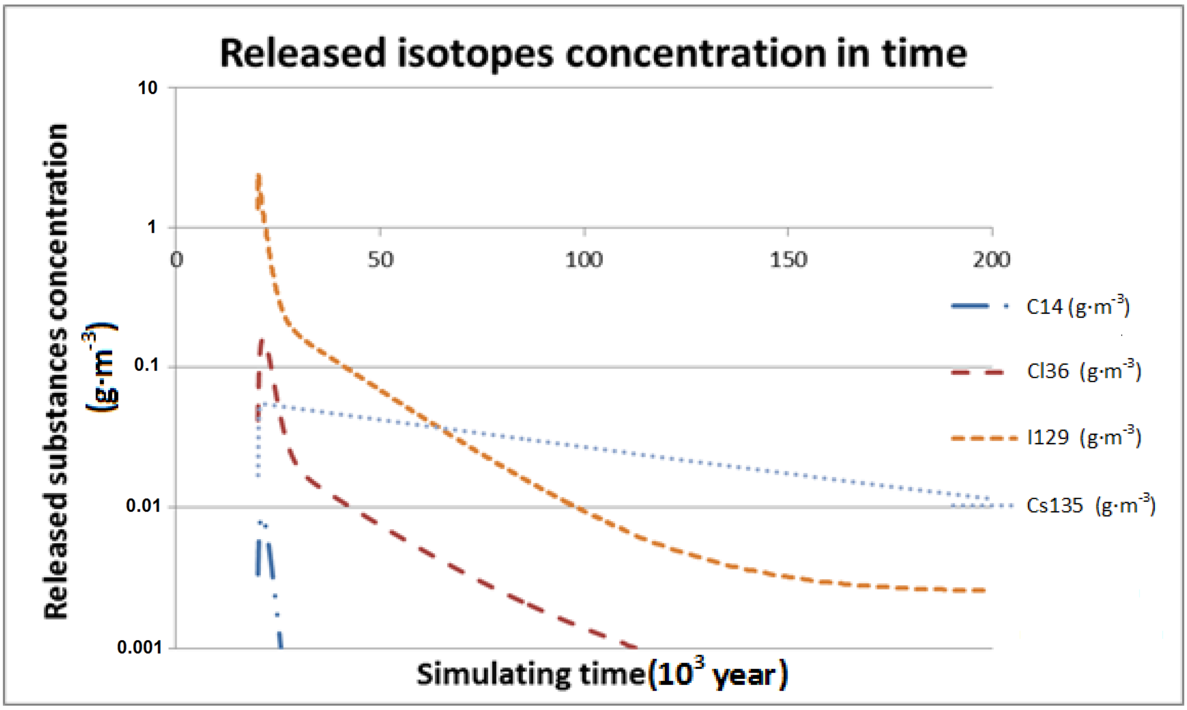


Fig. 2 Quantity of released isotopes in time.

be the zero concentration of isotopes. The DGR in the calculated mesh was modeled in the way that an element was withdrawn and a non-zero concentration on the walls of the withdrawn element was entered. Thanks to the element withdrawal, it is possible to model variable-in-time quantity of released radioactive substances.

For modelling purposes, the simulated time was 200,000 years with a damage of all the containers containing spent nuclear fuel set on 20,000 years. The time (20,000 years) represented quite a pessimistic forecast, more optimistic forecasts were also modeled (those ones also considered quite a different character of containers damage—the Weibull Distribution).

The input parameters of the model are stated in Chapter 2 (the definition of the area geometry, hydraulic conductivity and porosity of rocks and fissures zones, fissures openings, substances sorption parameters, diffusion coefficient and released volumes of substances in time). These parameters are analyzed (in the view of the model sensitivity and supposing the same calculated mesh but different input parameters—those of hydraulic conductivity or porosity) in the Chapter 4.

Progress (state) of volume activities of ground water (Bq·m^{-3}) in time was found for all the elements in the calculated mesh within the task. In total, 2,000 elements with non-zero concentration were recorded (from the total amount of 37,000) and from those elements, the following ones were analyzed in a more detailed way:

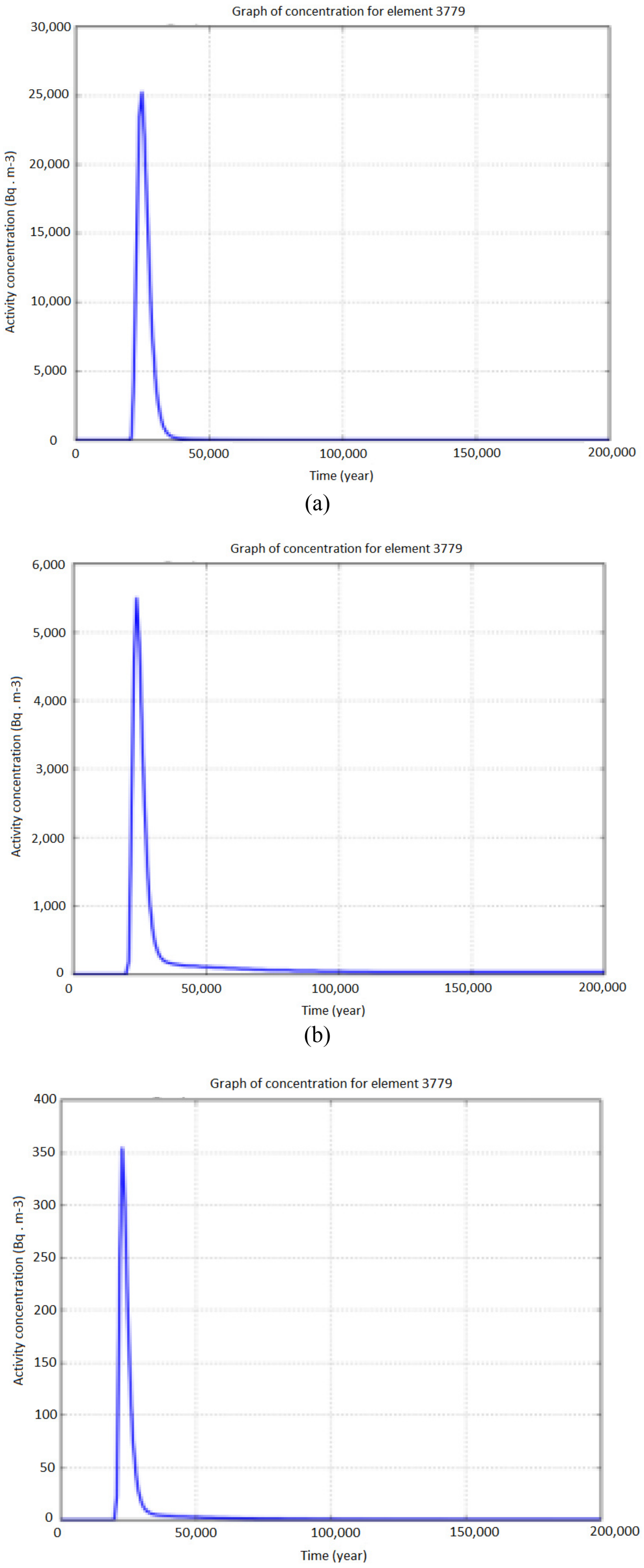
- Elements that were found on the surface of the area where isotopes penetrated biotopes (out of the geosphere). There were 2,140 surface elements within the area. In the paper, results for one element are presented. The element with the most volume activity of isotopes. Results of other elements are stated [2, 3, 13];
- Elements that formed the main transport line out of the DGR onto the surface of the area of the maximal recorded volume activity. The main transport line is formed with 10 elements;
- Elements that were found near the surface of the area (forming a layer at more than 400 m above the sea level), which could turn into surface elements (due to denudation) in future.

Within the task, volume activity graphs were designed for each of the elements (in time). Another output of the task was maps showing substances concentration at the given sea level. Results obtained (for all the isotopes) were used in radioactive dose calculations. To determine the dose, the scenario of 1 m^3 ground water drunk by a human in a year had been chosen. Based on that scenario, ingestion factors were found and stated for all the critical radionuclides shown in Table 3 [2, 3, 13].

In Figs. 3 and 4, there are volume activity graphs for the critical isotopes for one surface element (the one of the maximal activity) in time. It is quite clear that the released quantity of isotopes and the activity of elements on the surface are strongly dependent on the simulated time, the half-life and the course of its release out of the near/far field borders.

Table 3 The ingestion factor for the critical isotopes, the maximal recorded volume activity on the surface element and the dose calculated basing on the given scenario.

Isotope	Ingestion Factor	Max volume activity	Dose
	(Sv·Bq^{-1})	(Bq·m^{-3})	(Sv·m^{-3})
C14	5.80E-10	30,000	17.4
Cl36	9.30E-10	9,500	8.8
I129	1.10E-07	540	59.4
Cs135	2.00E-09	700	1.4
Ra226	2.80E-07	85	23.8
Sum			110.8

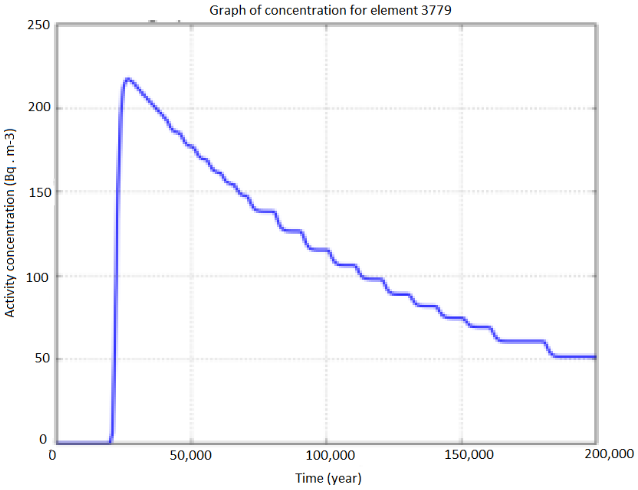


(a)

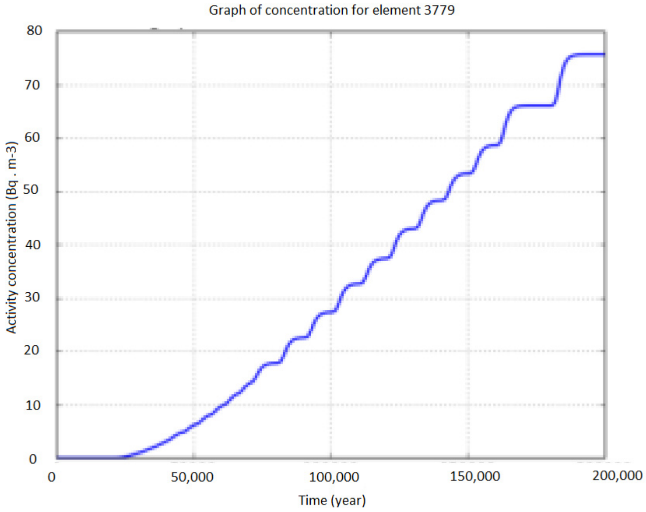
(b)

(c)

Fig. 3 Volume activities of (a) C14, (b) Cl36 and (c) I129 on the surface element in time.



(a)



(b)

Fig. 4 Volume activities of (a) Cs135 and (b) Ra226 on the surface element in time.

The crucial impact on the total radioactive dose (after the containers damage) had the I129, which did not absorb with unlimited dissolution and with a too long half-life (15.7 E6 year). Another disadvantage is its immediate release out of the damaged container—resulting in the worst possible situation where all the containers are damaged at once in the given simulated time. Therefore, the total dose is not dependent on the time of the damage of all the containers—only a shift of the maximal activity towards the maximum is shown in the Figs. 3 and 4. Using the Weibull Distribution to describe the containers life-time, it is necessary to consider the value of β parameter. The similar problems are with C14 that has a shorter half-life time and absorbs very little. The both isotopes penetrate into biotopes and into food-chain too easily and this fact increases their potential perilousness.

Another of the elements that has a crucial impact on the total dose is the Ra226, arisen with radioactive degradation of uranium matrix—with a short half-life (1,600 years). The released quantity is increasing with natural degradation of safety barriers and a large volume of U238 concentrated in a small volume.

Table 3 shows the maximal volume activities of isotopes on the surface element of the maximal volume activity in the given simulated time. The total dose received by a human with the given scenario (1 m^3 of ground water is drunk a year) is calculated out of the volume activities. The other isotopes (not mentioned in Table 3) have lower impact on the given scenario—less than 1 μ Sv. The total dose on other elements is lower (as it is shown in the Figs. 3 and 4) [21].

The map of volume activity was designed (Fig. 5) in a similar way. The map shows the surface area with volume activity of isotopes. It is clear that a substance reaches only a small part of the modeled area and that is why it is not necessary to extend the modelled area. Similar maps were designed for cuts of a given sea-levels. The cuts can be used to model denudation of the area in future.

Quality of the model were proved by a model designed using the SW GoldSim [22] which is determined for modelling of stochastic processes and helps to calculate substances transport and simulate changes of input parameters. The basic principal of the SW is in modular settings of defined components on the basic surface and determinations of physical or mathematical relations among particular blocks.

The SW GoldSim does not provide calculations of ground water flow. Knowing the preference line—1D geometry or cut geometry—2D area could be connected using components of physical relations and the resulting concentration of a substance (in the area) could be calculated. "Cell Pathway"—representing a mix chemical reactor and "Pipe Pathway"—representing chemical colony—belong among those basic components.

The transport module (knowing flow among components) is for the transport of more substances calculating: variable-in-time quantity released on the near/far filed border, sorption parameters, radioactive degradation, diffusion coefficient and the maximal dissolution of a substance in ground water.

The task model of flow and transport described at the beginning of the Chapter (using the SW Flow123D) was also analyzed using the SW GoldSim. Geometry of the area which was chosen basing on elements with non-zero concentration of a substance—they represented the input data for the model using the SW GoldSim. The complexity of the conversion was due to the fact that the elements entered using the SW Flow123D are of the tetrahedron shape and that is why the conversion of over-flows between walls in the flow task is so complex. A sample of a part of the model for a few elements near the DGR is shown in Fig. 6. Both tasks reached quality—correspondence in model results.

3.2 Sensitivity Analysis through Parameter Change

Sensitivity analysis through parameter change is based on a comparison of substance concentration between the reference task and the task with changed input parameters [20]. The basic parameters that changed are the following:

- Hydraulic conductivity of fissures and rocks that was changed as follows: 0.1, 0.2, 0.5, 2, 5 and 10 multiple of the former value calculated for every type of rock or fissure. There are 38 types of rocks within the area, which represented 228 tasks in total;

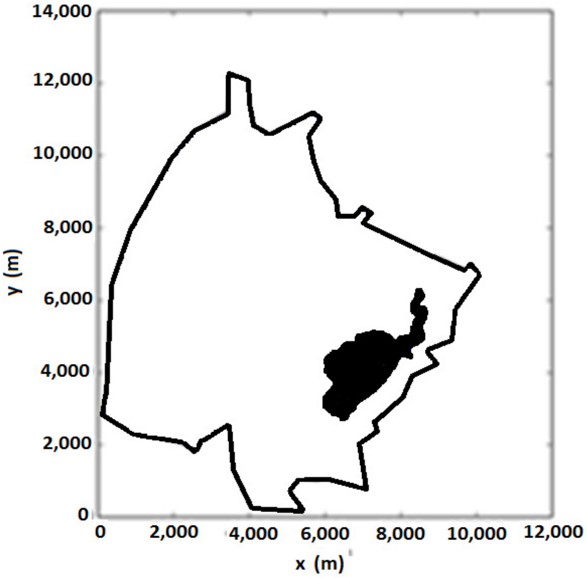


Fig. 5 Map of concentration I129 in the 250 m simulated time of 42,000 years (The black part shows the area with occurrence of concentration).

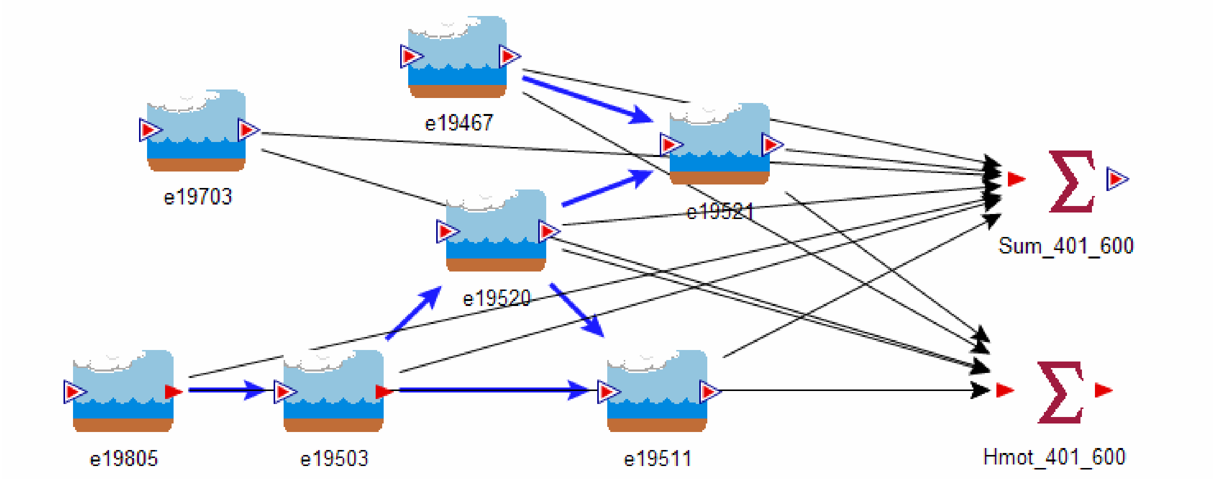


Fig. 6 A sample of a modeled part designed using the SW GoldSim.

- Fissures openings (14 types of openings in total) that were changed in 0.5 and 2 multiple of the former value, which represented 28 tasks in total;
- Porosity that was changed from the former value of 0.5 in 0.25 and 0.75 (mainly fissure zones and the surface area). The former value of 0.05 changed in 0.02 and 0.10 and the former value of 0.01 changed in 0.005 and 0.02, which represented 76 tasks (38 × 2) in total;
- Sorption parameters of substances were changed as follows: 0.1; 0.5; 2 and 10 multiple of the former value (only for absorbing substances), which represented four tasks in total.

Those changes were made for all the isotopes of a higher radioactive dose (more than 1 μSv) in the basic model. They were: C14, Cl36, I129, Cs135 and Ra226. Each of them had to be evaluated individually due to their different half-life, different character of their release out of the near/far field and different absorption properties.

The designed tasks of transport with changed parameters were calculated using the SW Flow123D. The resulting concentrations (volume activities ($Bq \cdot m^{-3}$)) were analysed and tasks where the change of an input parameter caused the strongest response, an increase of substance concentration (volume activity) on selected elements were found afterwards. Similar elements to those of the basic task could be included:

- Elements on the surface of the area where a substance penetrates out of the geosphere into biotopes. Results for one element (the one of the biggest volume activity of isotopes) are stated in the paper. Results for the other elements are stated by Chudoba, J. [13];
- Elements forming the main transport line out of the DGR up to the surface of the area with the maximal volume activity. The main transport line is composed of 10 elements;
- Elements at the selected (or higher) sea level. Those elements could describe penetration of a substance out of the geosphere into the biosphere taking into account the rock denudation;
- Elements that are found in a particular part of the modelled area with (supposing) a higher concentration of people;
- One crucial element could be analysed (a surface element of the maximal detected concentration or an element supposed in the area of a higher concentration of people).

The SW Flow123D compares the substance concentration (activity) between the reference task and the task with changed input parameters. A list of tasks of the strongest impact on the substance concentration change in the selected area is in fact the result of the SW analysis. The supposed increase of the evaluation

Table 4 Sensitivity analysis results.

The reference task	C14	Cl36	I129	Cs135	Ra226
Sum of activities (kBq·m^{-3})	16,950	3,400	148	1,850	1,450
Change parameters					
Rock near the DGR, hydraulic conductivity 0.1 x	3.49 x	3.46 x	3.49 x	3.42 x	3.13 x
Rock near the DGR, hydraulic conductivity 0.2 x	2.25 x	2.24 x	2.26 x	2.23 x	2.10 x
Rock near the surface, hydraulic conductivity 2 x	2.07 x	2.08 x	2.08 x	2.08 x	2.08 x
Rock near the surface, hydraulic conductivity 5 x	3.90 x	3.91 x	3.91 x	3.92 x	3.93 x
Rock near the surface, hydraulic conductivity 10 x	4.86 x	4.86 x	4.86 x	4.87 x	4.88 x

of particular tasks differences is the sum of the concentrations found on surface elements in the simulated time (all of the simulated time or times). The surface elements were selected because of radioactive substances penetration out of the geosphere into biotopes.

The increase was "fixed" as it was supposed that: (1) all the simulated time or times were of the same "weight"—the simulated time entry was done in an equidistant way; (2) elements were of the similar volume. That presumption was met and there was no "over-density" of any part of the area reflected on the calculated net. The total simulated time was 200,000 years and the results were recorded into the input files every 1,000 years.

The resulting volume activities on selected elements were compared with the reference task and are stated in Table 4.

It is clear (from the results stated in the Table 4) that the strongest impact on the substance concentration increase in the surface area was caused by:

- Decrease of hydraulic conductivity in the surroundings of the DGR, where the preference is represented by a fissure system and where contamination is not diluted anyway. That is why concentration near the surface is increased (as dilution of concentration happens on the surface);
- Increase of hydraulic conductivity near the surface of the area where transported substances are spread into greater "near the surface" area due to higher hydraulic conductivity.

Decrease of sorption coefficient K_d with substances causes increase concentration in the surface element. According to the results, the sorption parameter has a strong impact on the substances transport in the surroundings of the DGR—also having an impact on the total weight (mass) of substances released out of the near/far field. The weight (mass) of the released substances could have a crucial impact on concentration (volume activity) on surface elements.

4. Conclusions

A complex task describing a substances-transport out of the DGR onto the surface of the area was presented in this paper. The model covered a real area that had passed through a geological and hydrological research in the precedent years. The area was described by a geometry that included the basic geological types of rocks and the location of a fissures-net. The geological "situation" is described with: (1) hydraulic conductivity; (2) rocks porosity; and (3) openings of fissures. The other input parameters were: (1) substance quantity released out of the near/far field border in the surroundings of the DGR and (2) sorption and diffusion coefficients of particular isotopes released out of the geosphere.

A calculation net was designed for the area. Substance flow and transport were calculated with the use of the SW Flow123D. The results of the transport task were verified with the use of the SW GoldSim afterwards.

The radioactive dose was calculated from the obtained substances concentrations (volume activities) of isotopes (in every element of the calculated net).

The selected scenario supposed a human being who drinks 1 m^3 of ground water a year. The maximal dose on some surface elements was 110 $\mu Sv \cdot year^{-1}$.

A sensitivity analysis was done through a parameter change in the model. Parameters of hydraulic conductivity, porosity, opening of fissures and sorption parameters were changed for every critical isotope. Furthermore, the impact of such a change on resulting concentration (volume activity) on surface elements was observed.

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