Using Pedotransfer Function (PTF) analysis in the assessment of the parameters of potential radionuclide transport through the vadose zone of PRAW “Novi Han” – an initial stage of a model case study

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Abstract. The Repository for Radioactive Waste in Novi Han, Lozen Mountain (Bulgaria), dates from the early 1960s. In the present study, the complex geoenvironmental setting of the repository site was analysed from the viewpoint of assessment of potential radionuclide migration from the repository to the geosphere. Thus, components of the mass transport field were elaborated as a part of a conceptual model. In connection with this, a detailed characterization of the subsurface, especially of the vadose zone around the repository, was performed. The fractions of sand, silt and clay based on the grain-size distribution curves of samples from the different hydrogeological units gathered on the site of RAW-Novu Han were implemented in the ROSETTA program, and the respective hydraulic parameters were determined. As a result, the entire vadose zone was hydraulically determined.

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INTRODUCTION

The Repository for Radioactive Waste (RAW) in Novi Han, Lozen Mountain (Bulgaria), further noted as RAW-Novu Han, dates from the early 1960. Its purpose is the storage of unconditioned waste from the medicine, military sources, and scientific research activities in desolate site relatively close to the capital region. The official exploration of the repository started in 1964 with few m³ and up until 2010 the gross volume of the waste went up to around 670 m³ (Nuclear Regulatory Agency, 2011). In addition, according to the Bulgarian Strategy for Spent Fuel and Radioactive Waste Management SSFRAWM, the RAW-Novu Han will be subject to closure after the New National RAW Repository

starts operations to accept the respective waste there (Council of Ministers, 2011).

According to international documents, e.g., those of the International Atomic Energy Agency IAEA, such facilities should present a multibarrier approach (“defense in depth”) in order to ensure the safety of operations during and after the exploitation period (IEAE, 2006).

The choice of the RAW-Novu Han site was made in the early 1960s on the basis of expert evaluation of several sites. Being a “Radon” type facility, RAW-Novu Han’s engineered barriers represent specific units constructed from reinforced concrete and additional barriers such as steel layers, bitumen layers and brick walls (Mateeva, 2001). Although the geosphere around the ground site should be considered

as a main barrier against unanticipated radionuclide migration in the future decades (Stefanova, 2003), up until the 1990s there were not any in-depth geological, engineering geological and hydrogeological investigations concerning the aspects of transport of radionuclides from the viewpoint of a geoenvironmental impact of the repository site (Evstatiev and Kozhoukharov, 2000).

The first survey focused on the geosphere properties around the RAW-Novi Han was performed in 1992 – a complex assessment of the geological and geotechnical conditions of the repository site (Evstatiev *et al.*, 1994; Evstatiev and Kozhoukharov, 2000), followed by a detailed investigation between 2000 and 2002 of the geological, engineering geological and hydrogeological conditions on and along the repository site (Kozhoukharov *et al.*, 2002; Vassilev and Kozhoukharov, 2004). The last two studies were undertaken according to the documents of the IAEA, *e.g.*, IAEA 1994 and IAEA 1999 (both documents are superseded by IAEA, 2014, and IAEA, 2011, respectively), and the national regulations and legislation (Kozhoukharov *et al.*, 2002).

Based on the results of the last investigation, there is a preliminary research of four radionuclides fate and transport in the saturated part of the profile based on laboratory determination of their distribution coefficients (Galabov *et al.*, 2003). A second research work was based on “Interaction Matrix of System of Processes and Phenomena” modelling (Mateeva and Antonov, 2004). The methodology of the latter relies on the general description of natural conditions by applying such values of parameters which will lead to conservative results (IAEA, 2004). In a more recent study, the characteristics of the geosphere were analyzed from the viewpoint of the long-term stability of the repository (Antonov and Varbanov, 2013).

In summary, there are studies concerning some aspects of the natural conditions of the site, but there is no research work integrating the sources, the vadose and phreatic zones with their specific characteristics.

The aim of the study is to evaluate the parameters of both the vadose and saturated zones as a part of conceptual model and thus to provide detailed characterization of the subsurface around the repository.

As a further study, the potential radionuclide transport through the vadose zone will be studied based on the conceptual model, using numerical modeling with two different scenarios: with existing radioactive waste storage facilities and without such an inventory.

GEOENVIRONMENTAL SETTING

Geosphere

Geological and engineering geological setting

The RAW-Novi Han is located in the Lozen Mountain, where the geological structures of the area consist of Lower Paleozoic, Upper Paleozoic, Lower Triassic, Neogene and Quaternary deposits, and several magmatic bodies of granitoids and lamprophyre dykes (Fig. 1) (Vassilev and Kozhoukharov, 2004). Due to the purpose of the study, the engineering geological setting of the site area will be presented in details. According to the performed surveys (Kozhoukharov *et al.*, 2002; Antonov and Varbanov, 2013), four engineering geological zones (EGZ) are identified, distinguished both in plan-view and in cross-section of the site (Figs 2, 3).

Hydrogeological setting

Regarding the hydrogeological conditions of both the ground site of RAW-Novi Han and the area along it, two zones subject to different hydraulic characteristics are presented. A relatively thick unsaturated zone exists in the upper part of the ground site profile incorporating the whole anthropogenic zone and a part of the highly weathered zone. Downwards, in the weathered part of the phyllite massive, an aquifer is developed that is recharged only by precipitation. The flow shows a strict direction to the north (Fig. 4). The mean annual infiltration recharge for the area is assessed as 29.3 mm, the total groundwater recharge – 0.7 l/s (for area of 0.75 km²), hydraulic gradient – 0.095, and the average aquifer transmissivity – 0.3 m²/d (Kozhoukharov *et al.*, 2002).

Definition of the mass transport model

Based on the hydrogeological setting of the repository site, as well as the position of the storage facilities and engineered barriers, a convective-dispersive model of the water flow, which accounts for the retardation properties of the engineered layers and the geosphere, should be developed. Such models have been applied in similar studies in Bulgaria (Stoyanov, 2009, 2012; Antonov, 2017) and abroad (*e.g.*, Robinson and Bussod, 2000; Mallants *et al.*, 2001; Šimůnek *et al.*, 2006; Mallants *et al.*, 2011; Merk, 2012; Piqué *et al.*, 2013).

Regarding the aim of the whole investigation, as a next step, the model should be implemented in two scenarios: with stored wastes (with some arbitrary values for radionuclide concentration) and

Fig. 1. Geological map of the RAW Repository Novi Han, Lozen Mountain (after Vassilev and Kozhoukharov, 2004, with amendments): 1 – Deposits formed by human activity; 2 – Popov Dyal Formation (Late Miocene–Early Pliocene); 3 – Unit of the argillaceous sandstones and clays of the Chukurovo coal layers (Miocene–Helvetian); 4 – Upper Cretaceous dykes of syenite-granite porphyry; 5 – Triassic sediments; 6 – Upper Carboniferous and Permian sediments; 7 – Phyllite schists and phyllite unit (Ordovician); 8 – Diabase–phyllitoid complex (Late Riphean–Vendian); 9 – Precambrian metamorphic rocks of the Arda Group; 10 – Bedding; 11 – Fault; 12 – Reverse fault; 13 – RAW Repository Novi Han.

with removed waste but contaminated subsurface soil layer(s).

Elements of the Mass Transport Field – conceptual model

In order to achieve its goals, *i.e.*, to perform a reliable assessment of the radionuclide migration, the model should consider the following elements:

- Engineered barriers. According to the international safety requirements, the engineered barriers

are subject to destruction after 100 years of usage of the waste facility. Due to the operation period of RAW-Novı Han (since 1964), this period is halved. On one hand, the barrier is still in operation and, on the other, appearance of some cracks and fissures in the concrete parts is possible. For the purpose of the study, the model takes into account an arbitrary value of the radioactive concentration into the pore water as an outflow from the bottom of the facilities.

- Vadose zone. The vadose zone coincides with the embankment (anthropogenic zone) and with the

Fig. 2. Engineering geological map of the RAW Repository Novi Han site (after Antonov and Varbanov, 2013): 1 – Anthropogenic embankment – first engineering geological zone EGZ (EGZ 1); 2 – Highly weathered rock-core (EGZ 2); 3 – Rock-core, phyllites weathered in different degree (EGZ 3); 4 – Fissured phyllites (argillites), in association with quartz dykes and granitoids (EGZ 4); 5 – Fault distinguished by geological mapping; 6 – Fault distinguished by geophysical surveys; 7 – Fault supposed by geological mapping; 8 – Geological boundary; 9 – Profile line; 10 – Prospecting motor borehole.

Fig. 3. Cross-section through the RAW Repository Novi Han (after Antonov and Varbanov, 2013, with amendments): 1 – Anthropogenic embankment – first engineering geological zone EGZ (EGZ 1); 2 – Highly weathered rock-core (EGZ 2); 3 – Rock-core, phyllites weathered in different degree (EGZ 3); 4 – Fissured phyllites (argillites), in association with quartz dykes and granitoids (EGZ 4); 5 – Fault; 6 – Fault distinguished by geophysical surveys; 7 – Groundwater level; 8 – Motor borehole.

upper part of the weathered zone (between 7 m and 13.5 m in total).

- Phreatic zone. The phreatic zone underlies the vadose zone and it is formed in the fractured parts of the Ordovician phyllites.

METHODOLOGY OF THE RESEARCH

The constructed model will be implemented in programs numerically solving the partial differential equations for the governing convection-dispersion equations of the same-named model. Hence, the computer code HYDRUS-1D (Šimůnek *et al.*, 2008) will be used for the vadose zone transport modeling and, respectively, the codes ModFlow (Hughes *et al.*, 2017; Langevin *et al.*, 2017) and MT3D-MS (Bedekar *et al.*, 2016) will be used for the saturated zone transport modeling (Stoyanov, 2019).

Characterization of the vadose zone and its hydraulic parameters

The computer program HYDRUS-1D numerically solves (using the method of final elements) the advection-dispersion equation on the basis of the solution of the partial differential equation of Richards, which describes the water flow in a variably saturated porous medium (Šimůnek *et al.*, 2008). Resolving the latter governing equation requires

the knowledge of two nonlinear functions, namely the soil water retention curve $\theta(h)$ and the hydraulic conductivity function $K(h)$, and the theoretical fundamentals are given in brief.

The convection process in variably saturated porous medium is described by the partial differential equation of Richards. For one-dimensional vertical flow, this equation is:

$$\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial z} \left[K(h) \left(\frac{\partial h}{\partial z} + 1 \right) \right], \quad (1)$$

where θ is volumetric soil water content ($\text{m}^3 \text{m}^{-3}$); t is time (s); z is vertical coordinate (reference level for h reading) (m); K is hydraulic conductivity (m s^{-1}); and h is soil water pressure head (m).

Equation (1) is usually solved by the usage of the two above-mentioned nonlinear functions, $\theta(h)$ and $K(h)$. One of the most used and sustained models, empirically describing these functions and implemented in the HYDRUS-1D computer code, was developed by van Genuchten (1980) based on the statistical pore size distribution of Mualem (1976), and is defined as follows:

$$\theta(h) = \begin{cases} \theta_r + \frac{\theta_s - \theta_r}{\left(1 + (\alpha |h|)^n\right)^m}, & h < 0 \\ \theta_s, & h \geq 0 \end{cases} \quad (2)$$

and

$$K(h) = \begin{cases} K_s K_r(h) & h < 0 \\ K_s & h \geq 0 \end{cases} \quad (3)$$

as

$$K_r = S_e^l \left[1 - \left(1 - S_e^{1/m} \right)^m \right]^2, \quad (4)$$

where θ_r is residual water content ($\text{m}^3 \text{m}^{-3}$); θ_s is saturated water content ($\text{m}^3 \text{m}^{-3}$); α (m^{-1}), n (–), and m ($m = 1 - 1/n$) are empirical constants; h is soil water pressure head (m); l is empirical constant, assumed as equal to 0.5; K_r is relative hydraulic conductivity (–); K_s is saturated hydraulic conductivity (m s^{-1}); and S_e is saturation degree (–) given with:

$$S_e = \frac{\theta - \theta_r}{\theta_s - \theta_r}. \quad (5)$$

According to the van Genuchten model, if the five hydraulic characteristic parameters θ_r , θ_s , α , n and K_s are known, the two functions $\theta(h)$ and $K(h)$ could be described (van Genuchten, 1980).

Fig. 4. Hydrogeological map of the RAW Repository Novi Han site (after Kozhoukharov *et al.*, 2002, with amendments): 1 – Zone with prevailing groundwater in porous medium; 2 – Zone with prevailing groundwater in fractured medium; 3 – Groundwater table elevation; 4 – Groundwater flow line; 5 – Watershed; 6 – Piezometers – respectively, number and water level in meters; 7 – Spring (elevation in meters); 8 – Border line of the RAW Repository Novi Han.

The values of these parameters for a relevant soil can be determined directly by field and/or laboratory hydraulic tests or estimated indirectly through prediction from more easily measured data based upon quasi-empirical models. Such models are known as pedotransfer functions (PTFs) because they translate basic soil data into hydraulic properties (Schaap and Leij, 1998; Schaap *et al.*, 1998).

Such PTF algorithm based on particle-size distribution (PSD) analysis is used in the computer code ROSETTA (Schaap *et al.*, 2001) incorporated in HYDRUS-1D. By using ROSETTA code, the retention function and the hydraulic conductivity function, *i.e.*, their respective hydraulic parameters for each individual layer of the site/model, were determined.

RESULTS AND DISCUSSION

On the base of all gathered geological, engineering geological, borehole, and geophysical data from the ground site profile of RAW–Novi Han, four hydrogeological units with a different rank have been established. The domains of these units approximately coincide with the spatial ranges of the engineering geological zones described in Fig. 2 and Fig. 3.

The particles' percentage values (fractions) of sand, silt and clay based on the grain-size distribution curves (not presented in the study) of twenty-four samples in total of the different hydrogeological units gathered by three motor-boreholes MB1,

MB2 and MB3 (Fig. 2) on the site of RAW–Novi Han were used. In case of the existence of fractions greater than 2 mm, their percentage values were added to the sand one. The latter approximation does not change the tendencies of the hydraulic characteristics, especially about the hydraulic conductivity ones, due to the similarity between the physical characteristics of the gravel- and sand-sized grains in a soil-rock complex. Then, each sample's fraction data were implemented in the ROSETTA program, and the respective hydraulic parameters were determined (Tables 1–3).

In summary, one set of hydraulic parameters was determined for the embankment, four sets of parameters were determined for the weathered rock (eluvium) all above the groundwater table, fourteen sets of hydraulic parameters were received for the phyllites zone all in the phreatic zone and, finally, five sets of parameters for the association of granitoids, phyllites and quartz dykes were determined in both the vadose and phreatic zones. Analysis of the received set of data shows that the hydraulic parameters' values of each specific unit in a specific place (borehole) are not dispersive, *i.e.*, those of the weathered rock (Table 1), those of the association of granitoids and phyllites (Table 2), those of the phyllites (Table 2), etc. There is one exception for this tendency concerning the phyllites (Table 3 and partly Table 1). In contrast, the parameters' values for one and the same unit vary in spatial direction when comparing the hydraulic characteristics of MB1, MB2 and MB3, respectively. Thus, the spe-

Table 1
Hydraulic parameters determined by PTF analysis for MB1

MB1 layer	depth (m)	θ_r (–)	θ_s (–)	α (m ⁻¹)	n (–)	K_s (m s ⁻¹)
weathered rock (eluvium)	2.50–3.0	0.0411	0.3847	4.03	1.6809	1.09E-05
weathered rock (eluvium)	6.0–6.50	0.0374	0.3898	4.47	2.0064	1.88E-05
GWT*			6.9 (m)			
phyllites	12.0–12.50	0.0442	0.3823	3.83	1.8355	1.44E-05
phyllites	15.0–15.30	0.043	0.3969	0.98	1.5156	2.98E-06
phyllites	17.0–17.30	0.0393	0.386	4.11	1.6054	9.41E-06
phyllites	19.0–19.30	0.0396	0.3871	3.08	1.4071	4.96E-06
phyllites and quartz dykes	21.0–21.30	0.0417	0.3841	3.86	1.5283	7.5E-06
phyllites, high- ly weathered	22.50–23.0	0.0411	0.3849	4.07	1.7984	1.36E-05
phyllites	26.50–26.80	0.0363	0.3887	3.52	1.4255	6.05E-06
phyllites	29.30–29.50	0.0529	0.3884	2.34	1.3884	2.64E-06
phyllites	29.50–30.0	0.052	0.3919	1.58	1.4364	2.14E-06

*GWT – groundwater table

Table 2
Hydraulic parameters determined by PTF analysis for MB2

MB2 layer	depth (m)	θ_r (–)	θ_s (–)	α (m ⁻¹)	n (–)	K_s (m s ⁻¹)
Weathered rock (eluvium clay)	1.0–1.20	0.0433	0.3837	3.59	1.4463	5.69E-06
association of granitoids and phyllites	7.80–8.0	0.0341	0.3907	3.47	1.4228	6.45E-06
association of granitoids and phyllites	8.0–8.50	0.0325	0.3925	3.67	1.4319	7.09E-06
association of granitoids and phyllites	10.0–10.20	0.0395	0.386	3.77	1.4586	6.30E-06
GWT*			13.50 (m)			
phyllites	14.0–14.50	0.0414	0.3866	2.94	1.4026	4.53E-06
phyllites	22.0–22.50	0.0302	0.3958	4.33	1.4854	8.88E-06
phyllites	27.50–28.0	0.0413	0.3869	2.82	1.4023	4.44E-06

*GWT – groundwater table

Table 3
Hydraulic parameters determined by PTF analysis for MB3

MB3 layer	depth (m)	θ_r (–)	θ_s (–)	α (m ⁻¹)	n (–)	K_s (m s ⁻¹)
embankment	1.0–1.50	0.0426	0.3846	3.49	1.4271	5.29E-06
weathered rocks	3.0–3.50	0.0393	0.386	4.11	1.6054	9.41E-06
GWT*			10.15 (m)			
phyllites	16.50–16.80	0.0281	0.3997	4.37	1.4752	9.52E-06
phyllites	21.0–21.30	0.0417	0.3903	1.73	1.4371	3.51E-06
phyllites	26.0–26.30	0.0378	0.3879	3.23	1.4128	5.46E-05
phyllites	29.70–30.0	0.0413	0.3869	2.82	1.4023	4.44E-06

*GWT – groundwater table

cial diversities should be taken into account for the next step of modeling.

CONCLUSION

Assessment of the potential radionuclide migration is a complex task due to the sophisticated mutual interactions of various elements and processes of a repository facility during time. In this study, an attempt has been made to describe in details the main features of the RAW-Novi Han's building and environment areas, *e.g.*, engineered barriers and vadose and phreatic zones, that have to be integrated in a model case study. The concept of the latter is in congruence with the relevant international documents, especially those of the International Atomic

Energy Agency, and incorporates the governing equations of the water flow in media with variable state of saturation. In addition, as an initial stage of such model case study, after processing with ROSETTA program of samples' grain-size distribution fractions, the hydraulic parameters of the layers of four hydrogeological units, the constituents of the vadose and phreatic zones, are unambiguously determined. As a next step, the model will be implemented in two scenarios: with and without a source, *i.e.*, radioactive waste facilities.

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