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Tracer use for the protection of water resources in nuclear sites

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Abstract

Natural and artificial tracers can be used for nuclear sites management. Tracer techniques coupled with modelling and environmental monitoring activities are an effective tool to characterize and to foresee the radionuclide dynamic in the environment. In particular, the safeguard of water resources for human purposes must be guaranteed. In this work, the transport of tracers H-3 and I-129 in groundwater and subsoil was evaluated by means of Hydrus 1D and AMBER codes. Information on how radionuclides migrate in the environment were obtained and preliminary hypothesis on how to design the environmental monitoring network of the investigated site were deduced.

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Keywords: tracers; groundwater; transport modelling; Hydrus 1D; AMBER; environmental monitoring network; water resources protection

1. Introduction

In the nuclear field, the radioactive waste management includes different activities, which involve handling, pre-treatment, treatment, conditioning, transport, storage and disposal of radioactive waste [1]. A suitable radioactive waste management allows to protect the environment, in particular the water resources. Water is one of the main transport pathways of radionuclides and also an essential resource for the population. Studies to characterize the site and to forecast how polluting phenomena evolve, also supporting Safety Assessment of the nuclear activity, are needed for the design of monitoring networks and/or for mitigation procedures. Many tools and techniques are used

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to characterize the transport of source term into environmental matrices (e.g. groundwater, surface water, subsoil, etc.); tracer tests and their modelling are effective techniques to gain data from the environment, in particular on groundwater systems. Tracer techniques are a multipurpose method to investigate and characterize subsurface, evaluating transport velocity, porosity, dispersivity, preferential flow pathways, structural anisotropy, etc. [2]. In the nuclear field, the tracer application allows to design the environmental monitoring network, to monitor power plants in operation, to characterize future disposal sites, to design effective containment and isolation barriers (engineered barrier), to evaluate the accidental migration of radionuclides, to design proper actions for mitigation of polluting phenomena. The modelling of tracer migration allows to estimate where and when radionuclides can be found in an environmental matrix in accidental situations even though in not measurable concentration levels. In this work, the modelling of H-3 and I-129 migration in groundwater was carried out, using data collected in a specific site. This site was investigated in the past because it was considered a possible site where locate the national near-surface repository for Low Level Radioactive Waste [3]. Thus, data collected to describe it allow to perform realistic transport studies for the aim of this work. Tritium and iodine 129 were chosen for their high mobility, because in ordinary and accidental situations these tracers are the first which leak from the nuclear facility and which can be detected in the environment. In this way, it is possible to design and to perform possible mitigation actions to minimize the migration of low mobility radionuclides (e.g. Cs-137, Sr-90, …). The modelling of the tracer transport was undertaken by means of Hydrus 1D [4] to investigate the vadose zone and to analyze the tracer behavior near the source (near-field). Then, the system "repository-geosphere" was schematized by means of the AMBER code [5] to evaluate the tracer transport in depth (far-field). The analysis performed investigate a short time scale (years), in order to realize immediate mitigation action in case of radionuclide leakage from the nuclear facility and to protect the water resources.

2. Modelling of the tracer transport

The tracer migration in environmental matrices can be modelled to foresee their movement towards possible targets. In the radioactive waste management, this evaluation defines the relationships between the repository and the environment, the design of the engineered barriers and the planning of the radiological monitoring network.

As far as the geological framework is concerned, the soil stratigraphy which characterizes the investigated site is shown in Table 1, its water table level fluctuates between 8 m and 11 m under the ground. This stratigraphy is featured by a gravel layer between 25.5 m and 32 m in soil depth. In this work, a well to extract water for human purposes was hypothesized in the gravel layer (target). The leakage of radionuclides could be a risk from the radiological point of view, if radionuclides reach this layer. For this reason, the use of tracers associated with the modelling is one tool to perform safety studies and to assess the impact of a radioactive waste repository on the environment.

Table 1. Soil stratigraphy.

Type of layer	Soil Depth [m]
Silty Clay	0-3
Sandy Loam	3-3.4
Silty Clay	3.4-8
Clayey Silt	8-10.5
Gravel	10.5-15.5
Silty Clay	15.5-25.5
Gravel	25.5-32

High mobility tracers, H-3 and I-129, were chosen to carry out the transport modelling. Their high mobility is fundamental in accidental situations because they are the first radionuclides that can be detected through the monitoring network, but a low quantity could be stored in the repository, thus the on-site detection is not always easy (i.e. radionuclides are difficult to be measured). The concentration of tritium (half-life 12.33 y) and iodine 129 (half-life $1.5 \cdot 10^7$ y) present in the Italian radiological inventory [6] is $3.36 \cdot 10^3$ GBq and $2.80 \cdot 10^2$ GBq, respectively.

The study was developed analyzing the transport (advection, dispersion, diffusion) in the vadose zone and in the entire soil profile during the post-closure phase of the near-surface repository life.

HYDRUS 1D focuses on simulation of water, heat, and solute movement in one-dimensional variably saturated (vadose) media. The model was developed focusing on the first 5 m of the vadose zone. Hydrus 1D numerically solves the Richards equation for variably-saturated water flow and advection-dispersion type equations for heat and solute transport:

$$\frac{\partial \vartheta}{\partial t} = \frac{\partial}{\partial x}\left[K\left(\frac{\partial h}{\partial x} + \cos\alpha\right)\right] - S$$

where: *h* represents the pressure head [m], ϑ is the water content [-], *t* is the time, *x* is the spatial coordinate [m], S is the sink term [d^{-1}], α represents the angle between the flow direction and the vertical axis, *K* represent the non-saturated hydraulic conductivity [$m \cdot d^{-1}$].

The software solves the Galerkin linear finite elements scheme. The hydraulic model of the soil was implemented using soils-hydraulic functions of van Genuchten [4]. The parameters characterizing the van Genuchten expression are: the residual and saturated water content, θ_r and θ_s respectively; the saturated hydraulic conductivity K_s; the inverse of the air-entry value α; the pore size distribution index n; the pore connectivity parameter l. In the developed simulations, water flow and contaminant transport models were implemented. The Hydrus 1D application was tested and validated analyzing a specific soil profile in [7].

After the investigation of the vadose zone, the system "repository-geosphere" was developed by means of the code AMBER. This software evaluates the radionuclide transport through different compartments, which represent the environmental matrices, and calculates concentrations and radioactive doses. It is used mainly to implement the transport model in the geosphere and biosphere. The concentration in the compartment is considered uniform and the transport between compartments is simulated assuming suitable transfer rates for advection, diffusion and dispersion among compartments. Radionuclides can be considered in solid, liquid or gaseous phase. The variation of the pollutant *N* in the compartment *i*, is expressed by the equation:

$$\frac{dN_i}{dt} = \left(\sum_{j\neq i} \lambda_{ji} N_j + \lambda_M M_i + S_i(t)\right) - \left(\sum_{j\neq i} \lambda_{ij} N_i + \lambda_N N_i\right)$$

where: *i* and *j* represent the source compartment and the target compartment respectively, *N* and *M* are the amount of pollutant in the compartment, *M* is the first radionuclide of the radioactive decay chain [mol], *S(t)* is the external source of the pollutant in [mol/y], λ_M and λ_N are the radioactive decay constants of M and N in [y^{-1}], λ_{ji} and λ_{ij} are the transfer rates between the compartments in [y^{-1}].

The concentrations obtained in simulations must be compared with the Minimum Detectable Activity (MDA). The MDA represents the sensitivity of the measurement and gives also an immediate idea about the importance of the data collected from measurements and of radiological consequences. The MDA to detect H-3 and I-129 is equal to 0.2 Bq/l using standard instrumentations. Another aspect which must be considered is the radiological impact. It is represented by the annual dose to the population, which must be lower than 1 mSv/y. In this study the main radiological risk is due to the ingestion of water, thus it is possible to estimate the dose considering the effective dose coefficients for ingested particulates, assuming a mean ingested water by a man of 0.73 m^3/y. The effective dose coefficient is equal to $1.8 \cdot 10^{-11}$ Sv/Bq for tritium and $1.1 \cdot 10^{-7}$ Sv/Bq for iodine 129 [8].

3. Results

3.1. Modelling of the tracer transport in the vadose zone

Firstly the hydraulic behavior in the vadose zone was investigated. Parameters used in van Genuchten model, obtained by the soil catalog Rosetta [9], are shown in Table 2.

Precipitation and evapotranspiration phenomena were included in the model. The atmospheric conditions were assumed as upper boundary condition, thus the rainfall and the evapotranspiration were considered. The variable pressure head condition, which allows to define the water table level, was assumed as lower boundary condition. The initial condition was imposed on the water content, assuming it equal to 0.1 [3].

Table 2. Soil parameters for the van Genuchten model.

Soil Type	Θ_r [-]	Θ_s [-]	α [cm^{-1}]	n [-]	K_s [cm/d]
Silty Clay	0.07	0.36	0.5	1.09	0.0048
Sandy Loam	0.065	0.41	7.5	1.89	1.061

Two simulations were performed, one over a period of one month, another one over a period of one year. In Fig. 1, the water content and the water flux over a period of 30 days and at specific soil depths are shown. The analysis of the profile at different depths allows to understand the advective behavior. In Fig. 1a, the water content at the different soil depths is almost constant over the entire simulation period. It is higher at the surface of the soil profile due to precipitations, which have influence on the first centimeters of the soil profile, where the layer is less permeable. The water content decreases with the depth, because the precipitation influence decreases and due to the evapotranspiration. The lowest water content is present at -3 m of the soil depth, where a more permeable layer (sandy loam layer) is located. This layer confined between two silty clay layers is partially isolated. The advective exchanges are very small. In Fig. 1b, the water flux shows negative values, where the flux enters in the system, and positive values, where the flux exits from the system. At the surface, the water flux is influenced from the precipitations, therefore the profile has zero value or negative value. The water flux at different depths is of the order of 10^{-18} m^3/m^2d.

The simulation developed over a period of 365 days allows to deduce comparable considerations respect the simulation over 30 days, both for water content and water flux. Some simulations which consider high rainfall conditions, such as precipitation of 100 mm or 200 mm in a day, were implemented to consider extraordinary meteorological events. These simulations did not identify evident variations in the water behavior except in the first centimeters of the soil profile. The major amount of the meteoric water flows for evapotranspiration and surface runoff.

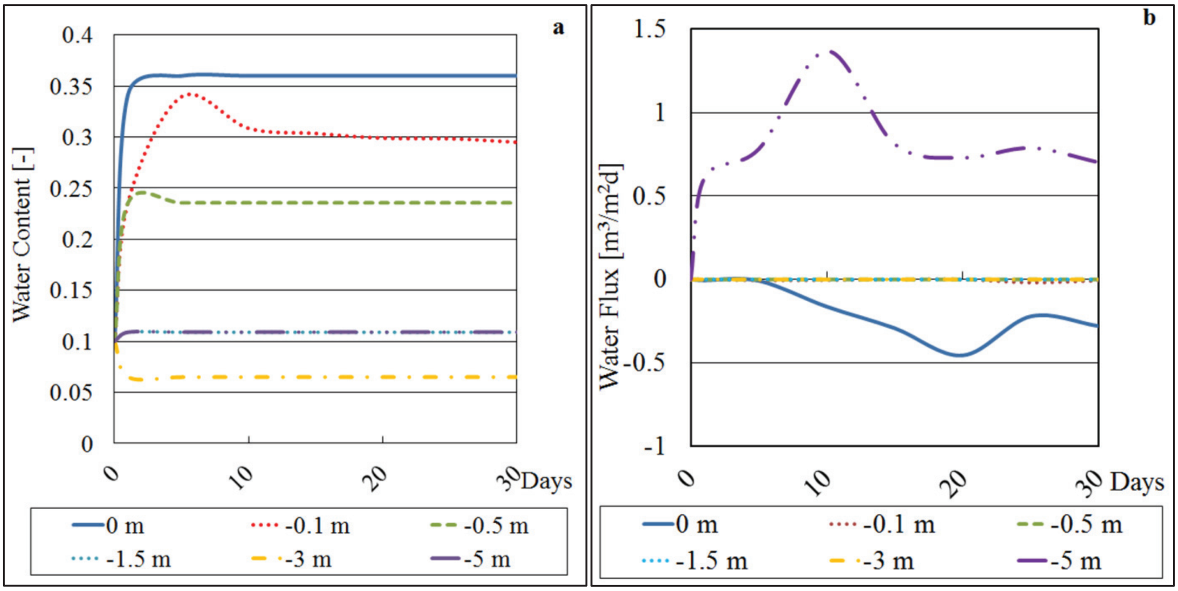

Fig. 1. Water content (a) and water flux (b) profile at different soil depths over a simulation period of 30 days.

After the analysis of the hydraulic behavior, the transport phenomena were introduced. A complete contamination of the vadose zone was assumed as initial condition. The initial contamination was imposed equal to the amount of H-3 and for I-129 in the inventory to represent an accidental situation in which a total release happens, even if the probability of this event is extremely low. Parameters which were introduced to describe the transport are: soil density, longitudinal dispersivity, distribution coefficient, radioactive half-life. A density of 2500 kg/m^3 and of 1600 kg/m^3 was assumed for the silty clay layer and for the sandy loam, respectively [3]. H-3 does not interact with the soil, thus its distribution coefficient is zero [10]; I-129 has a distribution coefficient of 0.0002 m^3/kg [11]. The longitudinal dispersivity was assumed equal to 0.5 m. In Fig. 2, the concentration in water of H-3 and I-129 are shown. From a qualitative point of view, both profile show a constant concentration except in the sandy loam layer (more permeable layer), enclosed between two silty clay layers. Quantitatively, the activity of these tracers in the inventory is very low respect to other radionuclides such as caesium 137, but not negligible from the radiological point of view. The constant behavior of the tracer concentrations allows to deduce that mitigation actions can be performed to avoid the leakage of low mobility radionuclides (e.g. Cs-137, Sr-90, Am-241, etc.). Indeed, radionuclides with low mobility are often more dangerous from the radiological point of view than radionuclides with high mobility. The simulated results highlight that the vadose zone protects the groundwater. The hypothesis of the initial condition equal to the inventory activity shows concentration results higher than the MDA and also the ingestion dose due only to these two tracers is higher than law limit of seven order of magnitude.

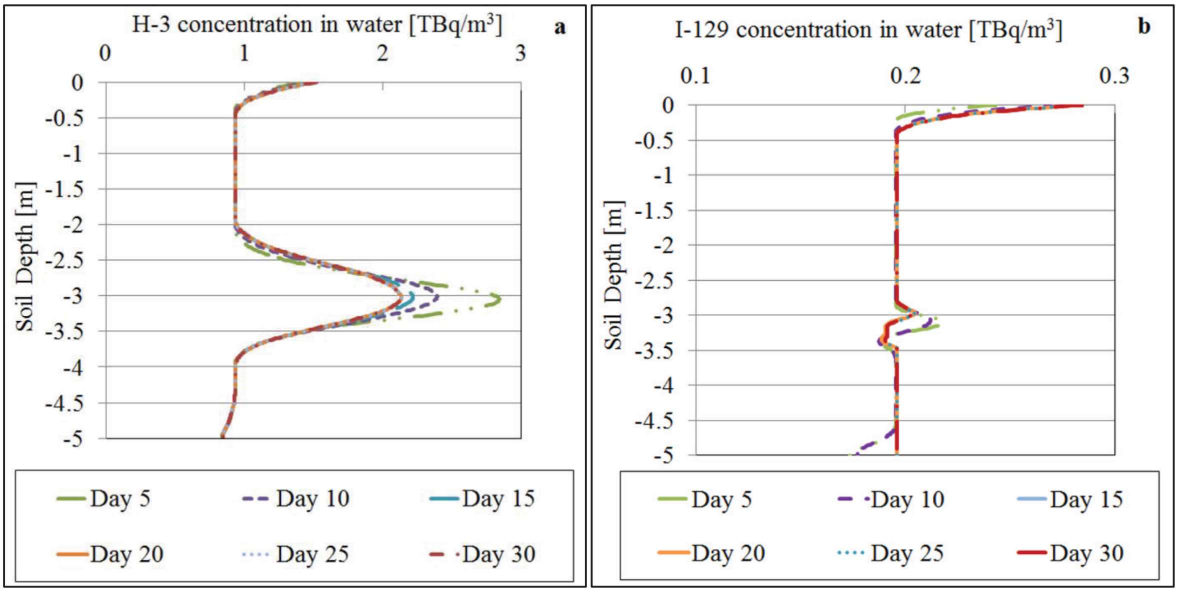

Fig. 2. Concentration in water of H-3 (a) and I-129 (b) in soil depth over a simulation period of 30 days.

3.2. *Modelling of the tracer transport in the soil profile*

The system repository-geosphere was studied by means of the AMBER code. The near-surface repository was assumed with a base under the ground of 5 m and the Italian inventory quantities of H-3 and I-129 were introduced. A leaching rate from the near-surface repository, considering the correct functionality of the repository and the periodical monitoring and maintenance (normal functionality scenario), was assumed over a period of 500 years. In this way, it was possible to evaluate the usefulness of tracers for the identification of small leakage from the repository. The geosphere includes the vadose zone (silty clay and clayey silt layers) and the saturated zone (gravel, silty clay and gravel layers). The aim of these simulations is to evaluate the amount of concentration that reaches the gravel layer (25.5-32 m), where a well for drinking water could be eventually located (target). The clay layers are mainly characterized from the molecular diffusion, but also a small advection phenomenon was included to realize a more realistic model. In the gravel layer, advection was imposed as unique transport phenomenon. The main parameters introduced in the model to describe advection, dispersion and diffusion transfer rates are shown in Table 3.

Table 3. Main transport parameters implemented in the model

	Permeability [m/y]	Hydraulic Gradient [-]	Total Porosity [-]	Effective Porosity [-]	Density [kg/m^3]	Hydrodynamic Dispersivity [m^2/y]	Diffusion Coefficient [m^2/y]
Waste	$1.00 \cdot 10^{0}$	0.005	0.2	0.2	1000	2100	$3.15 \cdot 10^{-5}$
Silty Clay	$3.15 \cdot 10^{-4}$	0.005	0.5	0.08	2500	2100	$3.15 \cdot 10^{-5}$
Clayey Silt	$3.15 \cdot 10^{-3}$	0.005	0.55	0.1	2500	2100	$3.15 \cdot 10^{-5}$
Gravel	$3.15 \cdot 10^{+2}$	0.03	0.35	0.25	1800	2100	$2.21 \cdot 10^{-3}$
Silty Clay	$3.15 \cdot 10^{-4}$	0.005	0.5	0.08	2500	2100	$3.15 \cdot 10^{-5}$
Gravel	$3.15 \cdot 10^{+2}$	0.03	0.35	0.25	1800	2100	$2.21 \cdot 10^{-3}$

In Fig. 3, the concentration profile of H-3 and I-129 is shown over the period of 500 years in the last gravel layer (target). It is possible to deduce that the monitoring of tritium can be performed to identify accidental situations in the first 200 years and the monitoring of iodine 129 can be carried out in the following years to control the polluting phenomenon. This consideration depends on the radioactive half-life of the two tracers. However, in this case the amount of I-129 is of the same order of magnitude of the minimum detectable activity (0.2 Bq/l), but ordinary condition is under investigation. Thus, in accidental situations, the possibility to use these tracers to detect a polluting phenomenon is verified. As far as the radiological impact is concerned, the calculation of ingestion dose due to these tracer is six order of magnitude under the law limit (1 mSv/y).

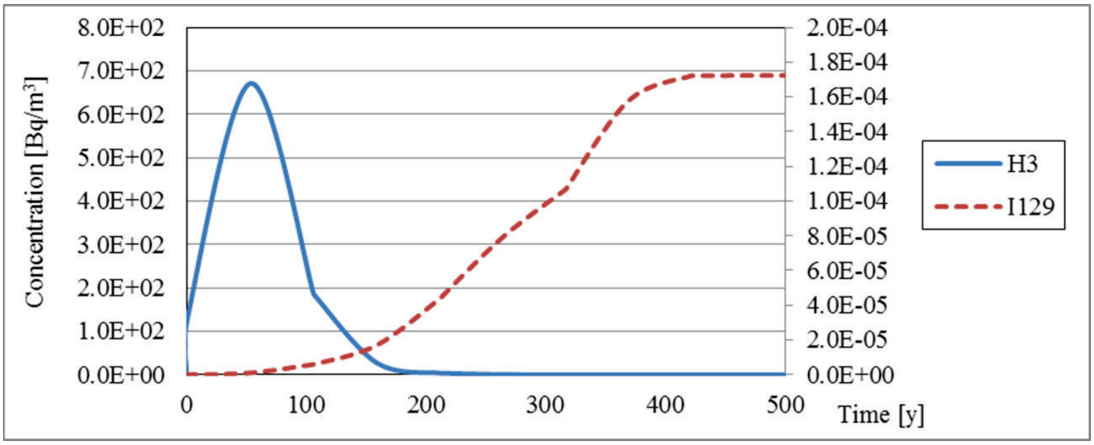

Fig. 3. Concentration profile of H-3 (left label) and of I-129 (right label) in the gravel layer.

In Fig. 4, the concentration in depth is shown for different times and at 250 m of distance from the near-surface. The tracers are confined by the clay layers, due to the absorption of the clay materials and to the low advection phenomenon. In the gravel layer, the transport phenomenon is very high. The H-3 concentration decreases quickly with the time due to the low radioactive half-life respect to the I-129 behavior.

The concentration of H-3 and I-129 was also evaluated at different distance from the near-surface repository in the gravel layer, up to 500 m, and at different times as shown in Fig. 5. In Fig. 5a, the tritium concentration at 250 m of distance from the repository results negligible from the radiological point of view, but it is detectable during monitoring activities. As far as I-129 is concerned (Fig. 5b), the concentration increases in time due to the leaching rate from the repository and to the negligible reduction due to the radioactive decay in the considered period. The concentration at 250 m from the repository results negligible from the radiological point of view and it is of the order of magnitude of the minimum detectable activity.

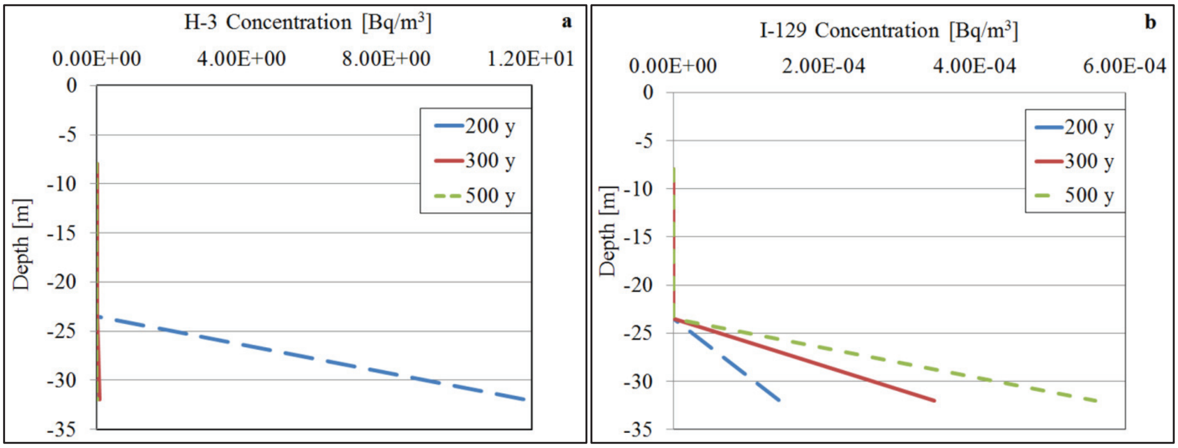

Fig. 4. Concentration in depth of a) H-3, b) I-129 at different times.

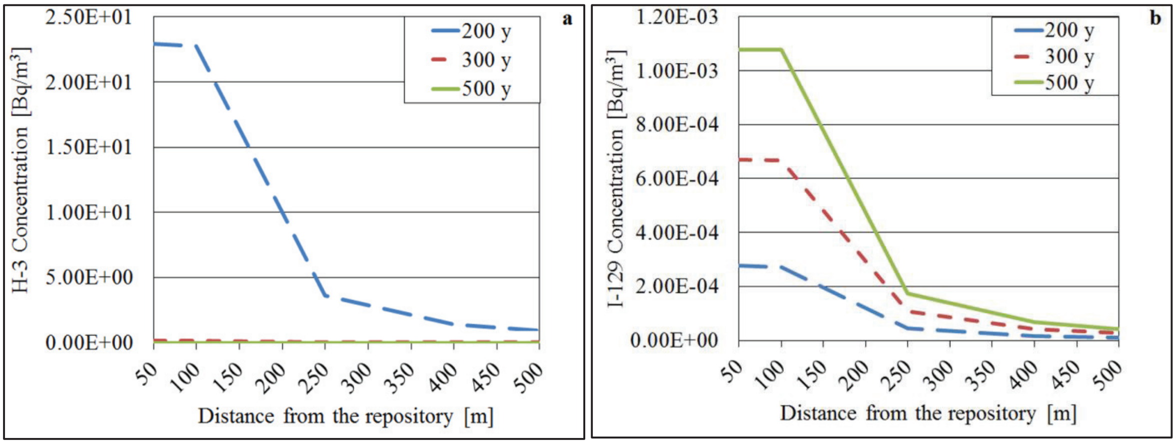

Fig. 5. Concentration at different distances from the repository in the gravel layer for a) H-3 and b) I-129 at different times.

4. Discussion

Preliminary considerations concerning the environmental monitoring network for the analyzed area can be deduced by the performed work. Specific monitoring stations could be hypothesized for the investigated area, considering how environmental monitoring systems were developed in existing near-surface repositories and nuclear facilities [12-14], and the regulatory laws.

As far as the groundwater monitoring is concerned, the concentration values at 250 m from the repository result negligible from the radiological point of view. Monitoring wells in an area surrounding the radioactive waste repository will be suitable to detect possible radionuclides in the environment.

Analyzing the stratigraphy, the presence of gravel layers favors the radionuclide migration. In particular, a sampling system which intercepts the water in the first gravel layer of the soil profile [10.5-15.5 m] may be planned. The detection of radionuclides inside it could highlight an accidental situation and mitigation actions may be undertaken to protect the deeper gravel layer [25.5-32 m], which can be used to extract water for human purposes.

Another consideration regards the mean life of a near-surface repository, which is assumed 300 y by the international scientific nuclear community. This period was identified as the period in which the radioactivity in the repository reaches values comparable with the environmental background. Analyzing the tracer behavior, it is possible to deduce that radionuclides with short radioactive half-life (e.g. tritium), have no a radiological impact from the ingestion dose point of view after 300 y, but the concentration of radionuclides with long half-life (e.g. iodine-129) is not negligible after 300 y. A monitoring over more than 300 y seems necessary for the current inventory.

The analysis performed demonstrates that the concentration of tracers is higher than the MDA, but it is negligible from the radiological impact point of view. In this situation, it is useful to monitor constantly the behavior of the nuclear facility using tritium and iodine 129 as tracers.

5. Conclusion

In the radioactive waste management, tracer techniques and their modelling are an useful tool to characterize the area where a repository could be located and to study the evolution of the system "site-repository". The tracer techniques can be also used to design the environmental monitoring network, to evaluate possible accidental migration of radionuclides and to plan suitable mitigation actions. These techniques can be associated with modelling applications to foresee the dynamic of the tracers in the environment. In this work, the tracer migration in an hypothetical near-surface repository for Low Level Radioactive Waste was evaluated, analysing in detail the transport in the vadose zone and in the entire stratigraphy profile. The tracers H-3 and I-129 were assumed, because of their high mobility. The feature of these tracers allows to identify their presence in the environment and to perform mitigation actions limiting the transport of radionuclides with low mobility. The analysis of the tracer behaviour in the environment by means of suitable software has demonstrated how to obtain preliminary information both on the site characterization and on the groundwater monitoring. In particular, the study of the vadose zone by means of Hydrus 1D highlighted that this zone is such as a barrier which protects the groundwater. On the other hand, the study of the system "site-repository" by means of AMBER allowed to estimate that a well-defined monitoring network on a small spatial scale is able to detect radioactive leakage from the nuclear facility. In addition, the deeper gravel layer, which can be used to collect water for human purposes, resulted vulnerable to the radionuclide migration. Thus, a periodical sampling may be provided to identify possible contamination before it reaches this layer.

In future studies, the couple of software, such as Hydrus 1D and AMBER, could be useful to improve the scheme "repository-geosphere-biosphere", to better define a possible radiological monitoring network in order to obtain a more realistic and detailed representation of the possible radiological impact of a near-surface repository. These studies could increase the knowledge on the site characterization and on the transport of radionuclides in the environment, with the aim to improve the safeguard of the population and of the environment.

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