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Radionuclides Transport Phenomena in Vadose Zone

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***Abstract*—**Radioactive waste management is fundamental to safeguard population and environment by radiological risks. Environmental assessment of a site, where nuclear activities are located, allows understanding the hydro geological system and the radionuclides transport in groundwater and subsoil. Use of dedicated software is the basis of transport phenomena investigation and for dynamic scenarios prediction; this permits to understand the evolution of accidental contamination events, but at the same time the potentiality of the software itself can be verified. The aim of this paper is to perform a numerical analysis by means of HYDRUS 1D code, so as to evaluate radionuclides transport in a nuclear site in Piedmont region (Italy). In particular, the behavior in vadose zone was investigated. An iterative assessment process was performed for risk assessment of radioactive contamination. The analysis therein developed considers the following aspects: i) hydro geological site characterization; ii) individuation of the main intrinsic and external site factors influencing water flow and radionuclides transport phenomena; iii) software potential for radionuclides leakage simulation purposes.

***Keywords*—**HYDRUS 1D, radionuclides transport phenomena, site characterization.

I. INTRODUCTION

NUCLEAR activities produce radioactive waste, which must be managed in suitable way. Disposal of radioactive waste is the final step in its management. Containment and isolation are the pillar concepts of radioactive waste management. Engineers' challenge is to design effective and redundant barriers to avoid the interactions between radioactive waste and environment. Despite engineering barriers can be very effective as radionuclides containment, it is important to study the possible dynamics of pollutants in environmental matrices, which constitute the environment surrounding a nuclear site. Interactions must be investigated in order to assess accidental radionuclides leakage, but also to evaluate a proper environmental monitoring network. In this context, radiation protection evaluation could be performed as well, in order to assess the radiological risks for population and environment. For this reason, safety analysis is essential; the safety objective is to protect people and the environment from harmful effects of ionizing radiation [1]. The radiological risks could involve all environmental matrices: surface water, groundwater, soil, air, etc. All of them are possible pathways for radionuclides accidentally released by the nuclear facilities. A soil profile is divided in two main zones by water table: the vadose zone (or unsaturated zone) and the saturated zone.

In this paper, the vadose zone was investigated. Radionuclides propagation in the subsoil and groundwater is subjected to different phenomena: hydro-geological, chemical-physical, and biological phenomena [2]. As far as radionuclides transport in environmental matrices is concerned, hydrogeology deals with all aspects related to movement of water in the subsoil due to advection, molecular diffusion, and dispersion. The chemical-physical phenomena consider the interaction between radionuclide and subsoil solid matrix, or among radionuclides/contaminants, and these phenomena also include the change of concentration due to radionuclides' chemical-physical evolution. Adsorption, volatilization, precipitation, radioactive decay, etc., are part of this category. The biological phenomena involve degradation phenomena and transformation of contaminants through biotic agents (bacteria, microbes, etc.); these processes were not considered in this work.

Environmental assessment can involve site and environmental matrices characterization, as well as prevision analysis by means of computer code. Foresight activity is essential in order to better manage the possible loss and radionuclides transport in the environment. The water flow and solute transport phenomena can be investigated with the aid of specific softwares, which allow obtaining pressure head profile and radionuclides concentration in soil and water. Some specialized open source softwares are: HYDRUS 1D [3], MODFLOW [4], PHREEQC [5]. HYDRUS 1D focuses on simulation of water, heat, and solute movement in one-dimensional variably saturated (vadose) media. MODFLOW is a computer code that numerically solves the three-dimensional groundwater flow equation for a porous medium by using finite-difference method. This software can be coupled with MT3DMS [6], that allows to introduce transport phenomena. The coupled HYDRUS - MODFLOW has been also developed to enhance the interaction between vadose zone and groundwater [7]. PHREEQC performs in detail speciation, batch-reaction, one-dimensional transport, and inverse geochemical calculations.

In this paper an Italian nuclear site was investigated to perform the radionuclides transport by means of HYDRUS-1D code. The source terms analyzed are Am-241, Co-60, Cs-137, measured in soil sample [8]. An analysis on the main factors that influence radionuclides transport phenomena was developed for investigation of simulations accuracy.

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II. WATER FLOW AND RADIONUCLIDES TRANSPORT

The water flow in porous media, in vadose zone, under assumption that air phase plays a negligible role in the liquid flow process, is described by Richards equation [3]:

$$\frac{\partial \vartheta}{\partial t} = \frac{\partial}{\partial x}\left[K\left(\frac{\partial h}{\partial x} + \cos\alpha\right)\right] - S \qquad (1)$$

where h is the pressure head; ϑ is the volumetric water content; t is time; x is spatial coordinate; S is the sink term; the volume of water removed by the soil through plant uptake per unit time α is the angle between the flow direction and the vertical axis (i.e. $\alpha = 0°$ for vertical flow, 90° for horizontal flow, $0° < \alpha < 90°$ for inclined flow); K is the unsaturated hydraulic conductivity function given by:

$$K(h,x) = K_s(x) \cdot K_r(h,x) \qquad (2)$$

where K_s is the saturated hydraulic conductivity e K_r the relative hydraulic conductivity.

Water flow in saturated porous media is described by Darcy's law [2], in which flow rate Q_x in x direction through section A perpendicular to x is given by:

$$Q_x = -\frac{kA}{\eta}\frac{\partial P}{\partial x} \qquad (3)$$

where k is the intrinsic permeability of the porous media; η is the dynamic viscosity of the fluid; P is the pressure.

Transport phenomenon is described by the mass transport differential equation [3]. The mass transport differential equation for non-conservative contaminants (meaning that chemical-physical and/or biological phenomena are not negligible) is as follows:

$$D_x\frac{\partial^2 C}{\partial x^2} + D_y\frac{\partial^2 C}{\partial y^2} + D_z\frac{\partial^2 C}{\partial z^2} - v_e\frac{\partial C}{\partial x} - \lambda C = R\frac{\partial C}{\partial t} \qquad (4)$$

or, in general form:

$$div(DgradC - Cv_e) - \lambda C = R\frac{\partial C}{\partial t} \qquad (5)$$

where $-div(Cv_e)$ is the advection term; $div(DgradC)$ is the hydrodynamic dispersion term; D is the coefficients tensor of the hydrodynamic dispersion; C is the concentration of radionuclide in the water; R is the retardation factor; v_e is the flow velocity; λ is the constant decay. One important coefficient is the retardation factor, described by the following equation:

$$R = 1 + \rho/\beta_e \cdot K_d \qquad (6)$$

where ρ is the soil density; β_e is the effective porosity and K_d is the distribution coefficient.

The water flow in vadose zone and radionuclides transport equations are implemented by means of HYDRUS 1D code. The software solves these equations numerically, applying the Galerkin linear finite elements scheme [9], [10]. The hydraulic model of the soil was implemented using soils-hydraulic functions of van Genuchten [11]. The main 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 (or bubble pressure) α; the pore size distribution index n; the pore connectivity parameter l. These coefficients are strictly dependent on the soil layer (sand, sandy loam, etc.). HYDRUS 1D includes several soil catalogs: one performed by Carsel and Parrish [12] for the van Genuchten model, another performed by Rawls et al. [13] for the Brooks-Corey model (soils-hydraulic functions used for coarse grain size layers) [14] and Rosetta DLL (Dynamically Linked Library) [15]. Rosetta implements pedotransfer functions, which predict van Genuchten coefficients in a hierarchical manner from soil textural class information, the soil textural distribution, bulk density.

The main data characterizing water flow and solute transport in HYDRUS 1D code are shown in Tables I and II, respectively. Water flow phenomena are described by soil profile (soil layer or soil textural class information); meteorological data as boundary conditions that include precipitation and evapotranspiration data (i.e. maximum and minimum air temperature, humidity, wind speed, crop data, etc.); water content or pressure head as initial conditions. Contaminants transport involves solute transport parameters (i.e. soil bulk density, longitudinal dispersion, mobile water content, etc.); solutes properties such as distribution coefficient, radiological decay constant or biodegradation decay constant, etc.; concentration flux or solute concentration as boundary conditions and solute concentration or radioactivity as initial condition.

TABLE I
MAIN DATA CHARACTERIZING WATER FLOW PHENOMENA IN HYDRUS 1D

Water Flow Data	Parameters
Soil Profile	Stratigraphy, textural class
	Water Table
Boundary Conditions	Meteorological data (evapotranspiration and precipitation data)
Initial Conditions	Water Content
	Pressure Head

TABLE II
MAIN DATA CHARACTERIZING CONTAMINANTS TRANSPORT IN HYDRUS 1D

Contaminants Transport Data	Parameters
Solute Transport Parameters	Bulk density
	Longitudinal dispersion
	Mobile water content
	Molecular diffusion coeff. in free water
	Molecular diffusion coeff. in soil air
Solutes Properties	Distribution coefficient
	Decay constant
	Biodegradation rate, evaporation rate, etc.
Boundary Conditions	Concentration flux
	Concentration, radioactivity
Initial Conditions	Concentration, radioactivity

In this context, through data analysis used for water flow and transport phenomena in HYDRUS 1D code, it is possible to identify the main parameters that influence the radionuclides transport in vadose zone. Two main categories have been identified: intrinsic factors of the investigated site and external factors. The first one includes the soil stratigraphy or the soil textural class, i.e. knowledge of sand, clay and silt composition percentage. This information is needed in order to obtain the parameters characterizing the soils-hydraulic functions of van Genuchten. The knowledge of the soil layers or the soil textural class is used for soil parameters definition, affecting simulation results. Another intrinsic factor is the distribution coefficient that depends both on soil layer and radionuclide. The distribution coefficient characterizes the adsorption phenomena involving contaminants and subsoil. Other parameters included in this category are: soil bulk density and longitudinal dispersion coefficient; they depend on the soil layers composition. Concerning the external factors, meteorological data (evapotranspiration and precipitation) condition the system response. In particular, precipitations interact with radionuclide transport in vadose zone: rainfall intensity and evapotranspiration phenomena influence movement of contaminants.

The impact of these different factors has been validated through the radionuclides transport analysis in a nuclear site, developing a model by means of HYDRUS 1D.

III. SITE BACKGROUND

The considered nuclear site is located in North Italy - Piedmont region. Several nuclear activities are situated in this area, thus radio-geological data are available. The transport analysis was focused on vadose zone. The main geological data concern the stratigraphy and the soil textural class. In Table III, the soil stratigraphy and the corresponding sample points in textural triangle (Fig. 1) are shown. The vadose zone is characterized from six layers with different soil compositions (Fig. 1).

Am-241, Co-60, and Cs-137 have been measured during the biannual radioactivity monitoring performed in the nuclear site. In Table IV, radioactive levels of Am-241, Co-60 and Cs-137 are reported [8].

TABLE III
SOIL STRATIGRAPHY AND CORRESPONDING SAMPLE POINTS IN TEXTURAL TRIANGLE

Points in textural triangle	Soil depth (cm)	Type of soil
1	0-25	Loamy sand
2	25-50	Sandy loam
3	50-75	Sandy loam
4	75-100	Loamy sand
5	100-125	Sand
6	125-150	Sand

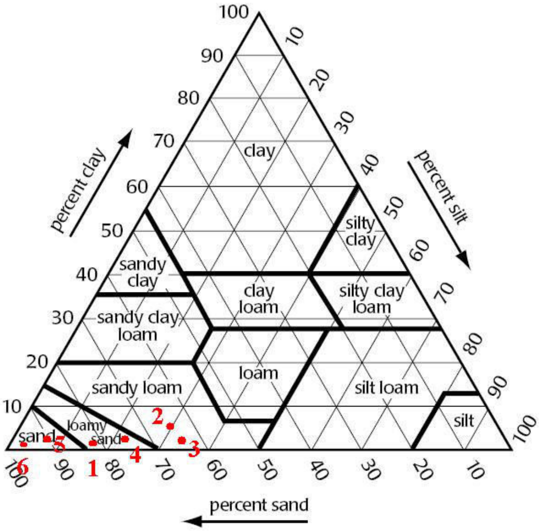

Fig. 1 Soil textural triangle with corresponding sample points for the investigated site

TABLE IV
RADIOACTIVE ACTIVITY OF AM-241, CO-60 AND CS-137

Date of measure	Description	Am-241 (Bq/kg)	Co-60 (Bq/kg)	Cs-137 (Bq/kg)
27/03/2013	Depth 0.80m	4.75 ± 1.43	0.30	898 ± 91

IV. RESULTS AND DISCUSSION

A detailed model of the investigated nuclear site was built. A van Genuchten model for porous media was considered. The aim of the analysis was to evaluate the effects of the intrinsic and external factors on the studied site, and the possible radionuclides migration up to the water table (with fluctuation of 2-5m) [16]. As far as the hydraulic aspect is concerned, a calibration model was developed. For a specific site, the development of a calibration model means to operate on parameters, influencing the model, so that the outputs reflect more accurately the data measured on-site. In this case, the pressure head data as a function of time and depth were provided for period from June to July, 2000 and are shown in Table V. The calibration model allows transient simulation of pressure head as a function of soil depth thus permitting the comparison with measured data. The pressure head data of June 7th were used as the initial condition and it was described by means of a fourth-order polynomial equation.

The calibration model was used as starting point for the development of the radionuclides' (focusing on Am-241, Co-60, and Cs-137) transport model. In this case, no information about measured radioactivity with time and at different soil depth were gathered. The only data available, shown in Table IV, were imposed as initial condition. Thus, the prevision model was implemented.

TABLE V
PRESSURE HEAD DATA (cm_{H2O}) MEASURED AT DIFFERENT DEPTH AND IN DIFFERENT DAY

Depth (cm)	07/06/00	08/06/00	22/06/00	28/06/00	05/07/00
-20	-210	-253	-231	-386	-367
-40	-205	-219	-190	-271	-313
-60	-199	-206	-183	-236	-282
-80	-195	-198	-164	-210	-236
-100	-193	-192	-168	-178	-199
-120	-196	-195	-169	-191	-208

A. Water Flow Model

The calibration model was developed in consecutive steps. First of all, the analysis was focused on the impact of intrinsic factors. The response of the model considering two different soil catalogs was investigated. As first approach, a simple model based on the soil catalog by Carsel and Parrish was analyzed, just considering the layer composition (loamy sand, sandy loam, sand). The second step consisted of more refined definition of the soil composition, by means of the Rosetta soil catalog. It provides precise data based on sand, silt and clay composition percentage. The parameters characterizing van Genuchten functions changed appreciably. The improvement in results of pressure head calculations justifies a detailed characterization of the analyzed soil. Afterwards, the impacts of external factors were analyzed to further refine the calibration model. External phenomena were introduced: the evapotranspiration, the precipitation and the hysteresis phenomena. They appreciably influenced the results. Evapotranspiration is influenced by: maximum and minimum air temperature, humidity, wind speed, sunshine, crop data (root depth, crop type, etc.). This information was provided by on-site weather station, as well as the precipitation data. As far as the hysteresis is concerned, it was not possible to get reliable information. For this reason the default parameters setting were not modified. In Fig. 2, the result of this calibration procedure, in term of comparison between measured pressure head (hollow markers) and simulated pressure head (filled markers) is shown. The rather good calibration level can be appreciated.

B. Radionuclides Transport Model

After the development of calibration model, the radionuclides transport phenomena were carried out. A prevision model was implemented, considering the soil catalog Rosetta [15] assumption formulated for the water flow study. The numerical evaluation requires the definition of the radionuclide source, Am-241, Co-60, and Cs-137. A uniform 80cm contamination layer around a leaking source was considered as initial condition.

Firstly, the intrinsic factors, related to radionuclides transport, were investigated: distribution coefficient K_d, soil bulk density ρ, longitudinal dispersion coefficient D_L and radioactive half-life $T_{1/2}$.

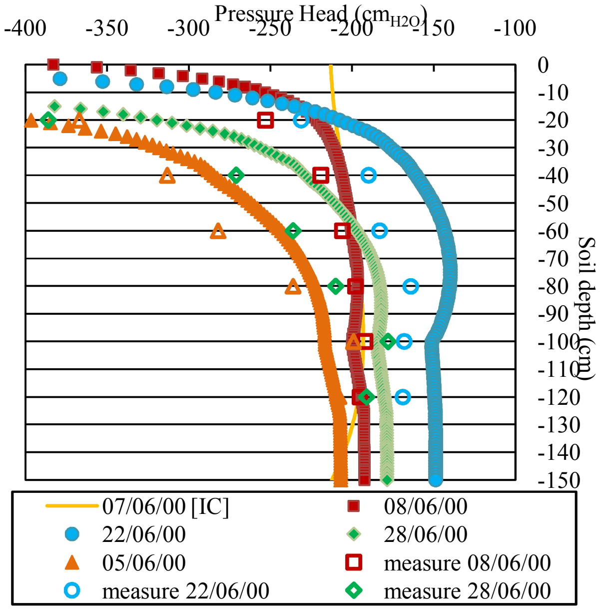

Fig. 2 Comparison between measured pressure head (hollow markers) and simulated pressure head (filled markers)

The distribution coefficient depends on the soil layer and radionuclide; its value was not gathered for the analyzed site, thus literature values [17]-[20] were used. The soil bulk density and the longitudinal dispersion coefficients depend on the soil layers; the measured values of these parameters are not available. For this reason, a literature research on sandy soil features led to individuate a suitable value of $1.6g/cm^3$ for bulk density and 15cm for longitudinal dispersion [2]. The radioactive half-life is an intrinsic property of the radionuclides. In Table VI, the value of distribution coefficient and the radioactive half-life for Am-241, Co-60, and Cs-137, are reported.

TABLE VI
DISTRIBUTION COEFFICIENT AND RADIOACTIVE HALF-LIFE FOR AM-241, CO-60 AND CS-137

Radionuclide	K_d (cm^3/g)	$T_{1/2}$ (years)
Am-241	8.2	432.2
Co-60	10	5.27
Cs-137	10	30.17

External factors were analyzed, in particular the effect of precipitation phenomena on the concentration profile with respect to the soil depth. This study was performed in a transient period of 30 days. The reference simulation condition is based on meteorological data provided by on-site weather station; then some critical conditions were hypothesized, considering different rainfall intensities. Two particular analyzed conditions were: a) a rainfall of 1.5cm/d for 10 days, b) a rainfall of 12cm/d for 3 days. Each environmental analysis must consider the climate change, which induces always more intense precipitations concentrated in a short time period. This consideration justifies the investigation case of a rainfall of 12cm/d for 3 days. Am-241, Co-60, and Cs-137 concentrations in water up to the soil depth of 110cm for the previous two cases, are shown in Figs. 3-5, respectively. Intense rainfall carries radionuclides deeper in the soil profile. The second case result is more critical than the first one. The impact of the intrinsic factor, distribution coefficient, is highlighted by an approximately uniform 80cm contamination layer. The behavior of the concentration in the first 20cm of soil depth is due to the evapotranspiration and rain accumulation phenomena.

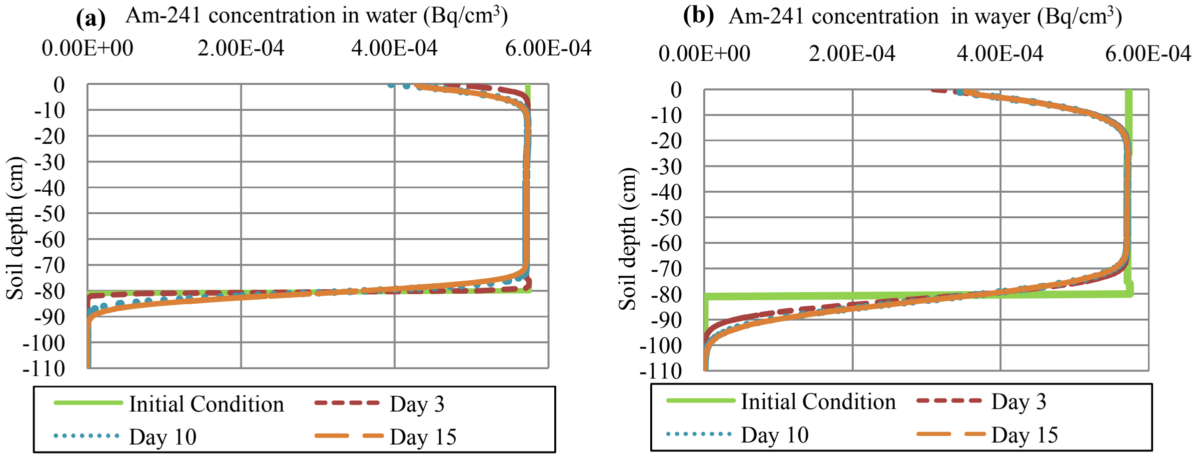

Fig. 3 Am-241 water concentrations respect to soil depth: (a) in case of 1.5 cm/d of rainfall for 10 days and (b) in case of 12 cm/d for 3 days

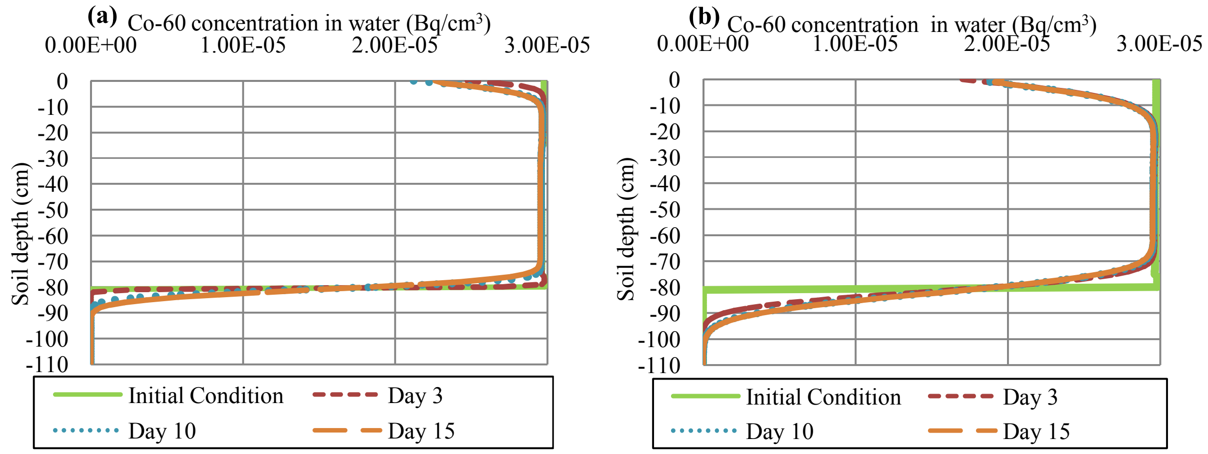

Fig. 4 Co-60 water concentrations respect to soil depth: (a) in case of 1.5 cm/d of rainfall for 10 days and (b) in case of 12 cm/d for 3 days.

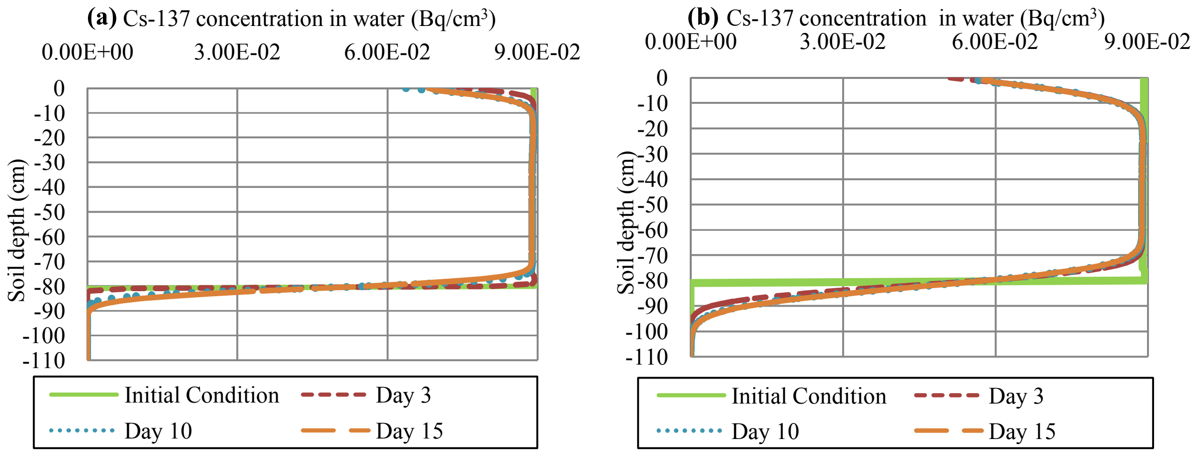

Fig. 5 Cs-137 water concentrations respect to soil depth: (a) in case of 1.5 cm/d of rainfall for 10 days and (b) in case of 12 cm/d for 3 days.

The radionuclides concentration in soil can be evaluated by the outputs of the simulations. In Fig. 6, the Co-60 concentration in soil, in case of a real condition, is shown.

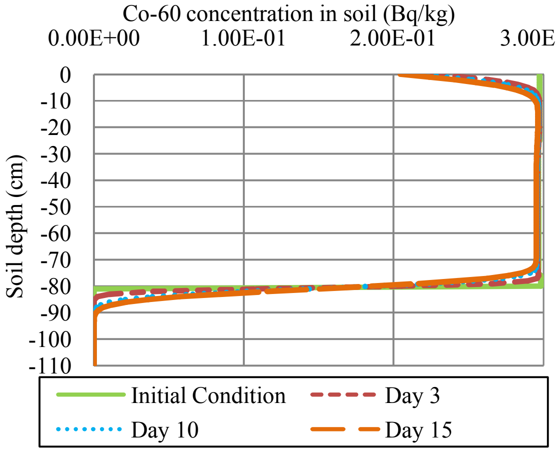

Fig. 6 Co-60 concentration in soil for different time

Other simulations were performed in order to analyze the long-term behavior. A transient simulation was run on a period of 200 days. In this case the effect of evapotranspiration is negligible. Results show that the concentration in the first 80cm of soil depth is constant due to the interaction between soil and radionuclides, established by the distribution coefficient. The reduction of the concentration is due to the radioactive decay. In Fig. 7, Co-60 concentration in water profile is shown. The concentration reduction of this radionuclide is noticeable as Co-60 has a radioactive half-life equal to 5.27 y, which is less than the half-life of Am-241 and Cs-137.

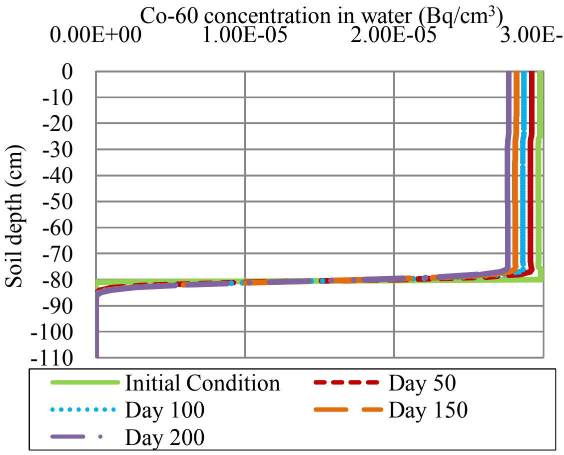

Fig. 7 Co-60 water concentration for several time in a transient simulation of 200 days

Analyzing the several prevision scenarios simulated, it was observed that radionuclides contamination does not reach the groundwater. In particular, the maximum concentration values correspond to initial condition imposed. The effect of intense rainfall causes deeper migration of the radionuclides, but the radioactivity of the three investigated radionuclides is negligible from the radiological risk point of view.

The transport dynamic studies must be coupled with on-site measurement, in order to establish the accuracy order with which to carry out the simulations. The device used for measuring and its efficiency must be known. For the detection of Co-60 and Cs-137, the gamma spectrometry with high-purity germanium detector p-type or n-type is used. The determination of α-emitter activity allows identifying total activity due to α-emitter; complex radiochemical methods allow separating a single radionuclide and identifying α activity due to Am-241. The measurement sensitivity is specified by the key parameter, the Minimum Detectable Activity (MDA), representing the smallest radioisotope quantity detectable with 95% confidence. The measurement sensitivities achievable for α-emitter (Am-241 included, unless it is not separated), Co-60, and Cs-137 for some standard instrumentations used for this purpose are shown in Table VII. For the analysis performed in this paper, these devices allow to detect the radioactivity in the first 80cm of soil depth.

TABLE VII
MINIMUM DETECTABLE ACTIVITY (ORDER OF MAGNITUDE) FOR ALPHA-TOT, CO-60 AND CS-137

Parameter	α-tot	Co-60	Cs-137
MDA (Bq/l)	0.1	0.002	0.005

V. CONCLUSION

The radioactive waste management and, in general, the accidental presence of artificial radionuclides in the environment need specific surveys. In this paper, radionuclide transport phenomena in the vadose zone have been evaluated, focusing on the radionuclides concentration profile in water and soil. These aspects can be implemented by means of proper software. Thus, prevision simulations can be run and possible mitigation actions can be planned. These studies may be important to properly design environmental monitoring network, in order to place appropriate monitoring stations in radiological risky areas.

A numerical analysis by means of HYDRUS 1D code, so as to evaluate radionuclides transport dynamics in a nuclear site in Piedmont region (Italy), has been carried out, focusing on the main impact factors affecting the model response. The key concepts in the analysis of the transport dynamics of radionuclides, obtained by the vadose zone simulations, are: i) a detailed site characterization from the geological point of view; ii) the adequate measurement of intrinsic parameters, such as distribution coefficient, soil bulk density, etc.; iii) the impact of the external factors, such as evapotranspiration and precipitation phenomena. The main factors, which influence the radionuclides transport, are the external factors, in particular the rainfall.

These analyses allow also evaluating the time and soil depth at which a radionuclide concentration reaches the MDA value. The radiological impact in the nearby of the leaking source is limited and negligible outside of the nuclear site.

Software potentials for radionuclides leakage simulation purposes must be investigated and examined in depth. A sensitivity analysis of the parameters, influencing the model response, is fundamental to appropriately manage the models. These aspects allow deep understanding of the proper way to develop a computational model for a real site and, as a consequence, tweak the codes according to the problems that must be studied.

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