Coupling of unsaturated zone and saturated zone in radionuclide transport simulations

Coupling of Unsaturated Zone and Saturated Zone in Radionuclide Transport Simulations

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Abstract

In the management of radionuclide release in the environment, the unsaturated zone could be a natural barrier to delay or to stop the radionuclide migration through the environment and to protect the groundwater from radiological risks. Thus, a suitable scientific evaluation of any radionuclide transport problems related to groundwater may to take into account the processes affecting flow through the unsaturated zone. In this work, an approach that involves the interactions between unsaturated zone and saturated zone both from hydrogeological and radionuclide transport point of view is proposed. This approach was tested developing a case study on an Italian nuclear site. The behavior of unsaturated zone as protective barrier for the groundwater was highlighted and identified as a fundamental aspect in the development of environmental analysis concerning the radionuclide transport into the environment. Promising results were found to improve the design of a radiological monitoring network.

Keywords: unsaturated zone; saturated zone; radionuclide transport; software tool coupling; release of radionuclide; radiological monitoring

1. Introduction

In the nuclear field, Safety Assessment involves all safety activities in order to manage a nuclear facility from the siting to the post-closure phase (International Atomic Energy Agency (IAEA), 2004; IAEA, 2009). In particular, as far as the radioactive waste management is concerned, the isolation of radioactive waste from the external environment has to be performed and investigated in order to avoid radiological risks for population and environment. Each nation involved in peaceful use of nuclear energy has a radioactive waste management strategy to safeguard population and environment (e.g. Sanders and Sanders, 2016). The modelling of radionuclide migration into the environment is one of the activities that allows to predict the dynamics of the radiological risk due to a nuclear facility operation, and its quantitative and qualitative impact on human beings and environment. The modelling can also be a supporting tool to plan the monitoring network, that is able to detect radionuclides in case of accidental release from a nuclear facility in order to make mitigation actions and/or to restore previous conditions.

Numerous studies on the environmental and radiological impact due to nuclear facilities can be found in the recent literature. Yim and Simonson (2000) reviewed performance assessment models for low level radioactive waste disposal facilities. Three different categories are identified. The first one concerns the near-field models, e.g. degradation of the facility, waste contained, and seepage of radionuclides into the surrounding subsoil. The second category involves the radionuclide migration from the subsoil to potential human exposure sites. Finally, the last category tracks the uptake, exposure, and dose equivalent due to transported radionuclides. Seher et al. (2016) investigated flow and transport processes in generic landfills that only contain nuclear decommissioning waste, through two different software SiWaProDSS (Ingenieurgesellschaft and Dresden, 2007), and SPRING (Delta h, 2010). Skuratovič el al. (2016) investigated unsaturated zone at two radioactive waste disposal sites in Lithuania. In particular, they collected data and determined distributions of tritium and stable isotope ratio of oxygen and hydrogen in precipitation, unsaturated zone and groundwater. They underlined the importance of unsaturated zone as the first natural barrier to limit the spread of contaminants.

In this context, our study focuses on the groundwater as natural resource, which should provide water for human purposes, taking into account the possible effect of radionuclide migration in unsaturated zone and groundwater. In fact, from the hydrogeological point of view, the subsoil profile can be characterized by two zones, with different hydrogeological features: unsaturated zone, that is the part above the water table level, and saturated zone, which is the part below the water table level. In the management of radionuclide release in the surface environment, the unsaturated zone could be a natural barrier to delay or to stop the radionuclide migration through the environment and to protect the groundwater from contamination risk; moreover, understanding the contaminant dynamics in the unsaturated zone allows to protect the biodiversity and the agricultural use of this zone. Thus, a suitable scientific evaluation of any pollutant or radionuclide transport towards groundwater has to take into account the processes affecting flow through the unsaturated zone. Despite groundwater research has highlighted the importance of unsaturated zone, its representation has been often simplified during modelling due to the complexity, the computational demand and the lack of data necessary to characterize hydraulic properties (Keese et al., 2005). Another relevant challenge regards the need to integrate the modeling between the two hydrogeological systems. Different software have been used to model unsaturated zone and saturated zone, separately. The software simulating both unsaturated and saturated zone often oversimplify the modelling of the unsaturated zone. For this reason, in this study a literature research was carried out in order to identify which simulation tools are widely accepted and used by scientific community to model unsaturated zone and saturated zone, respectively. Thus, suitable software to perform separate studies was applied and tested on the unsaturated zone (Testoni et al., 2014), through HYDRUS 1D (Šimůnek et al., 2013), and on groundwater (Testoni et al., 2015; 2016), through MODFLOW (Harbaugh A., 2005) and MT3DMS (Zheng and Wang, 1999). Then, these software were coupled in order to represent and model both zones as unique system, considering the effect of unsaturated zone on the transport in groundwater. This choice is confirmed by MODFLOW literature research. A promising approach to integrate the modelling between unsaturated zone and saturated zone has been suggested, to fill the gap in the integration of simulation codes for unsaturated and saturated systems, for safety assessment studies of nuclear facilities. The Variability Saturated Flow (VSF) process (Thoms et al., 2006), the Unsaturated Zone Flow (UZF1) package (Niswonger et al., 2006) and the HYDRUS package (Seo et al., 2007) are unsaturated zone modelling packages that can be coupled with MODFLOW, from the hydrogeological point of view. The performance comparison of these different tools was investigated by Twarakavi et al. (2008a), identifying the HYDRUS package for MODFLOW as the more promising ones. A coupled model simulates the effects of near-surface hydrological processes on groundwater flow by linking a groundwater model with a selected unsaturated zone model in space and time. This coupling makes it possible to estimate both qualitative and quantitative relationships between the two hydrogeological systems. In this work, a modelling approach that involves the interactions between the unsaturated zone and the saturated zone, both from hydrogeological (HYDRUS 1D-MODFLOW) and radionuclide transport (HYDRUS 1D-MT3DMS) point of view, is proposed. In addition to previous studies, the radionuclide transport was investigated taking into account unsaturated zone. This approach was tested developing a case study on an Italian nuclear site.

The aim of this paper is the assessment of radionuclide transport in the subsoil and groundwater, taking into account the unsaturated zone. In general, this zone is oversimplified or neglected in the studies of radionuclide transport for Safety Assessment purposes. We study the radionuclide transport in a real system constituted by unsaturated zone and saturated zone, by means of HYDRUS 1D and MODFLOW/MT3DMS software, respectively. This approach can be a useful tool which can support safety assessment studies of current or future nuclear activities.

2. Radionuclide transport: travel time and retardation factor

One of the most conservative evaluation of radionuclide migration through the subsoil is to assume that the solid matrix has no ability to slow radionuclide movement. Consequently, the radionuclides would travel in the direction and at the same rate of water. Due to the geochemical features of the soil minerals and to the chemical properties of the elements, this assumption is appropriate for certain radionuclides such as tritium and technetium, but is too conservative for other ones, such as americium and cesium.

The water and solute flow in unsaturated zone is characterized by three main parameters: water content, pressure head and hydraulic conductivity. The law that characterizes the dynamic of water and solute in unsaturated zone is the Richards' equation. Richards' equation describes the water flow in porous media, under assumption that air phase plays an insignificant role in the liquid flow process (Šimůnek et al., 2013). This zone is characterized by a water flow mainly in vertical direction. Equation 1 describes the Richards' law in a one-dimensional system:

$$\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial z}\left[K\left(\frac{\partial h}{\partial z} + \cos \alpha\right)\right] - S \tag{1}$$

where:

h is the pressure head [m];

θ is the volumetric water content [$m^3 \cdot m^{-3}$];

t is time [d];

z is spatial coordinate [m];

S is the sink term, the volume of water removed by the soil through plant uptake per unit time [d^{-1}];

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 [$m \cdot d^{-1}$] given by:

$$K(h, z) = K_s(x) \cdot K_r(h, z) \tag{2}$$

where K_s is the saturated hydraulic conductivity [$m \cdot d^{-1}$] e K_r the relative hydraulic conductivity [-].

Model of van Genuchten (Šimůnek et al., 2013) was assumed to describe the hydraulic properties of the unsaturated zone:

$$\theta(h) = \begin{cases} \theta_r + \dfrac{\theta_s - \theta_r}{[1 + |\alpha_{air} h|^n]^m} & h < 0 \\ \theta_s & h \geq 0 \end{cases} \tag{3}$$

$$K(h) = K_s S_e^l \left[1 - \left(1 - S_e^{1/m}\right)^m\right]^2 \tag{4}$$

Where:

θ_r and θ_s are the residual and saturated water content, respectively [-];

α_{air} is the inverse of the air-entry value [1/m];

n is a pore size distribution index, and $n > 1$ [-];

m is an empirical parameter defined as $1\text{-}1/n$, [-];

l was estimated equal 0.5 for many soils [-];

K_s is the saturated hydraulic conductivity [$m \cdot d^{-1}$];

S_e is the effective water content, equal to $(\theta\text{-}\theta_r)/(\theta_s\text{-}\theta_r)$ [-];

The assessment of the travel time of radionuclides in the unsaturated zone is complicated due to: a wide set of physical, chemical and biological processes that affect the flow; the variable size and the geometry of the unsaturated zone in natural conditions that often is not well known; the water flow that is a highly non-linear process and depends on the saturation degree of water (Mattern and Vanclooster, 2010). The estimation of the travel time through the unsaturated zone has received far less attention in comparison to the travel time assessments in groundwater, due to these complexities.

As far as the travel time in saturated zone is concerned, the analysis is performed starting from the Darcy's law:

$$\mathrm{q} = -\mathrm{K}\frac{d\phi}{ds} = -Ki \tag{5}$$

where:

q represents the Darcy's velocity [m·d^{-1}]. It is equal the volumetric flux divided the through area;

K is the hydraulic conductivity [m·d^{-1}];

$d\phi$ is the hydraulic head difference between two assessment points [m];

ds is the distance between the two points [m];

i is the hydraulic gradient [-].

In order to describe the contamination retardation, the processes such as absorption on minerals, matrix diffusion into internal pores and precipitation have to be considered. Many contaminants react strongly with natural and engineered adsorbents and are found to migrate from 10^2 to 10^6 times more slowly than the water (Kaplan et al., 1995). The relationship between contaminants dissolved in aqueous phase and the contaminants sorbed on solid phase can be described both for the unsaturated zone and for the saturated zone by three equations (Table 1) (Šimůnek et al., 2013): linear, Freundlich, Langmuir. In the linear equation, the parameter that directly measures the partitioning of a contaminant between solid and aqueous phases is the distribution coefficient, K_d [m^3·kg^{-1}]. In Freundlich equation, K_f is the Freundlich constant [m^3·kg^{-1}]^a, and *a* is the Freundlich exponent [-]; they are empirical coefficients. In Langmuir equation, K_l is Langmuir constant [m^3·kg^{-1}], and $\bar{S}$ is the total concentration of sorption sites available [kg·kg^{-1}]; K_l is an empirical coefficient.

Table 1 Summary of aqueous-solid phases relationships and obtained retardation factors for unsaturated and saturated zone.

	Aqueous-solid phases relationship	Retardation factor
Linear	$\bar{C} = K_d C$ (6)	Unsaturated zone $R = 1 + \frac{\rho}{\theta} K_d$ (9)
		Saturated zone $R = 1 + \frac{\rho}{p} K_d$ (10)
Freundlich	$\bar{C} = K_f C^a$ (7)	Unsaturated zone $R = 1 + \frac{\rho}{\theta} a K_f C^{a-1}$ (11)
		Saturated zone $R = 1 + \frac{\rho}{p} a K_f C^{a-1}$ (12)
Langmuir	$\bar{C} = \frac{K_l \bar{S} C}{1+K_l C}$ (8)	Unsaturated zone $R = 1 + \frac{\rho}{\theta}\left[\frac{K_l \bar{S}}{(1+K_l C)^2}\right]$ (13)

		Saturated zone $R = 1 + \frac{\rho}{p}\left[\frac{K_l\bar{S}}{(1+K_lC)^2}\right]$ (14)

R = the retardation factor [-]; ρ_b = the bulk density of the unsaturated zone or saturated zone [kg/m^3]; K_d = the distribution coefficient [m^3/kg]; ϑ = the volumetric water content of the porous medium [-]; p = the porosity [-]. The use of the water content instead of porosity is correct when the retardation factor is referred to the unsaturated flow.

The used software codes are able to implement all three equations for the mutual dynamic of contaminants dissolved in aqueous phase and the contaminants sorbed on solid phase, introducing the parameters described in Table 1. In our study, we implemented a linear behavior as validated by studies performed in the area (Iezzi et al., 2009).

3. Method

Twarakavi et al. (2008a and 2008b) studied the computational efficiency of the coupled HYDRUS–MODFLOW tool from the hydrogeological point of view. HYDRUS 1D (Šimůnek et al., 2013) focuses on simulation of water, heat, and solute movement in one-dimensional variably saturated 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 that allows to introduce transport phenomena.

The coupling of a one-dimensional model and a three-dimensional model is acceptable due to the physic of the unsaturated zone and saturated zone, respectively. The unsaturated zone is characterized by a water flow mainly in vertical direction, thus the schematization of the unsaturated zone in one-dimensional system is representative of the real system. Instead, the saturated zone must be implemented as a three-dimensional system. This zone is characterized by a water flow in horizontal and vertical direction. To implement this coupling, unsaturated zone must be subdivided in specific domains, featured by the same soil and hydrogeological properties; as a consequence, each domain is characterized by a specific stratigraphy profile of the unsaturated zone. Through the implementation of the specific mathematical models for unsaturated zone and saturated zone coupling, each unsaturated domain exchanges data, such as water table level and flow fluxes, with groundwater domain, which is modelled as a three dimensional system. In Figure 1, a simplified scheme of the coupling is shown (Twarakavi et al., 2008a).

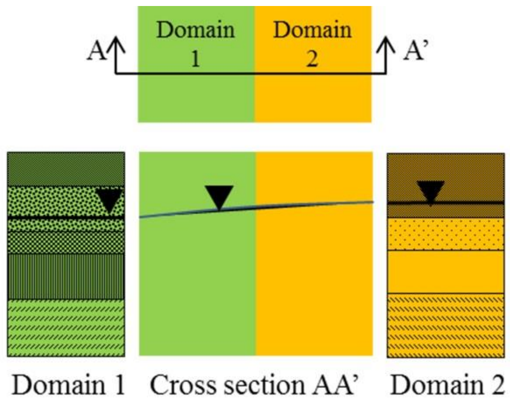

Figure 1 Simplified scheme of the coupling of unsaturated zone and saturated zone.

The proposed methodological approach was developed through the following steps.

First, the coupled HYDRUS-MODFLOW tool is enhanced by simulating unsaturated zone and saturated zone flows at their own, often different, time steps. This is needed because a proper treatment of the Richards' equation requires smaller time steps than those usually used in MODFLOW simulations. The two zones interact, exchanging data about the groundwater recharge and the groundwater level, only at the end of each MODFLOW time step, during which HYDRUS may perform multiple time steps to simulate unsaturated zone flow. MODFLOW receives the recharge flux from HYDRUS and calculates a new water table depth for the next time step. A new water table depth is calculated and assigned as the pressure head bottom boundary condition in the HYDRUS package for the next MODFLOW time step (Twarakavi et al., 2008a; Seo et al., 2007). In this way, the water table levels and the exchanged fluxes between unsaturated zone and groundwater are identified. During this step, information such as the topography and the stratigraphy of the area, initial hydraulic heads, meteorological conditions, presence of surface water, must be introduced to represent the hydrogeological behavior of the area as real as possible.

Then, the radionuclide transport is implemented in HYDRUS 1D and the radionuclide concentration at the interface between unsaturated zone and groundwater is identified. In this step, the initial concentration of the involved radionuclide, the localization of the source term, the interaction between radionuclide and solid matrix, represented by the distribution coefficients, must be implemented as initial and boundary conditions.

Finally, the data obtained by the hydrogeological coupling (water table level and exchanged fluxes) and by the radionuclide transport in the unsaturated zone (radionuclide concentration at the interface between unsaturated and saturated zone) have to be used for a second calculation, to implement an environmental system model that makes it possible to study the radionuclide migration into the groundwater (MODFLOW-MT3DMS).

In this way, it has been possible to consider the effect of the unsaturated zone both from the hydrogeological and radionuclide transport point of view. In Figure 2, the flow chart of the described approach is shown.

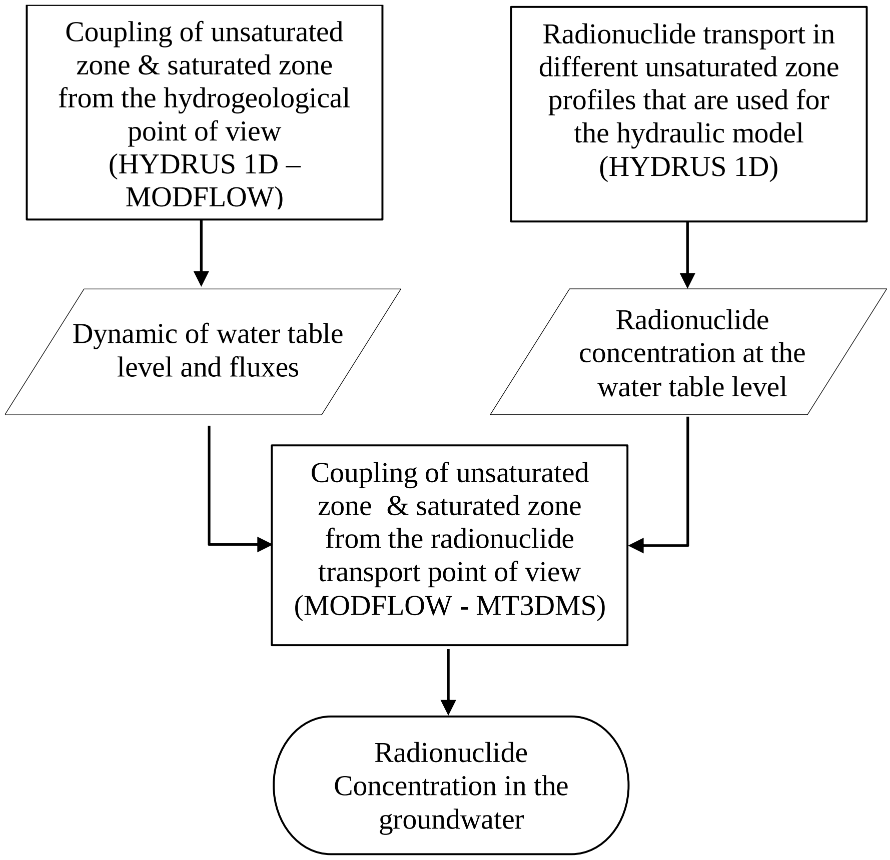

Figure 2 Flow chart of the proposed method.

4. Site Description

The proposed method to model the coupling of unsaturated zone and saturated zone both from the hydrogeological and radionuclide transport point of view was applied and tested on an Italian nuclear site. This site is located in the North Italy at Saluggia (VC). It hosts several nuclear facilities such as a fuel reprocessing plant, in decommissioning, temporary radioactive waste disposal, waste ponds to collect liquid effluent, a nuclear research reactor that now is used as temporary repository, etc. Figure 3 shows the investigated area and the location of piezometers. Piezometers were used to measure the water table level by ENEA (Italian National Agency for New Technologies, Energy and Sustainable Economic Development).

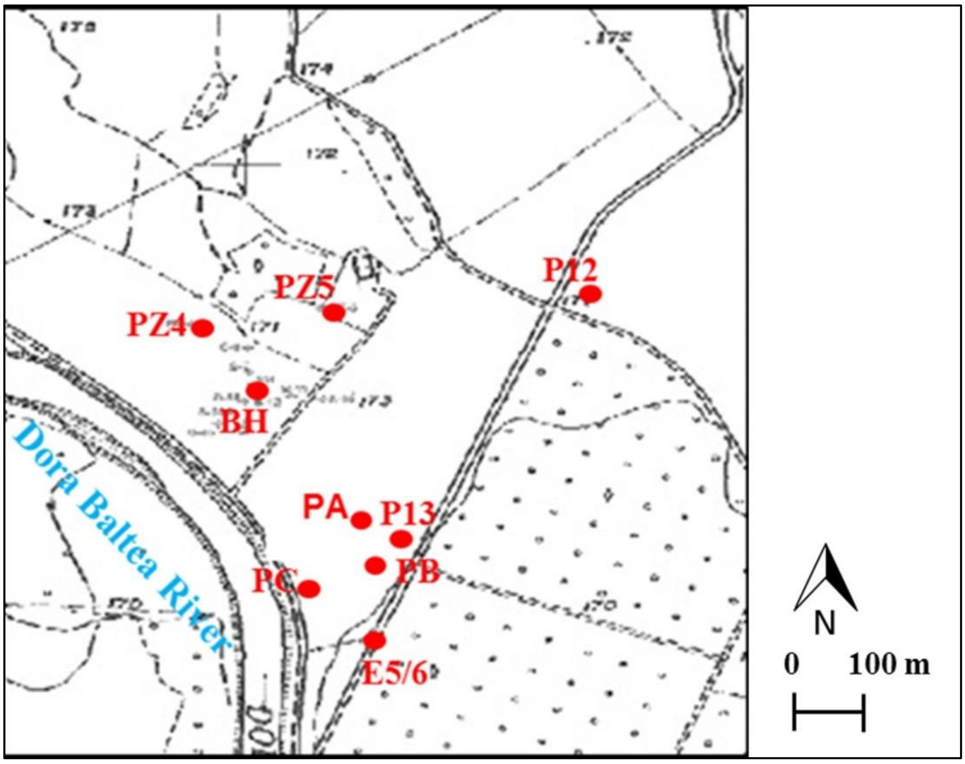

Figure 3 Italian nuclear site in the North Italy and the localization of piezometers of the site.

The annual groundwater dynamic was studied in detail in (Testoni et al., 2016). As far as the hydrogeological aspect is concerned, the elevation of top soil is in the range of 170-175 m above sea level. The site is located in the flood of the Dora Baltea River. The hydrogeological system is made up of two main aquifers: a shallow aquifer and a deep aquifer. The shallow aquifer is located in sandy gravel and gravelly sand layers, with a mean thickness of 45 m. It is characterized by a free water table, whose mean depth from the ground surface is about 3-5 m. Its flow direction is North-South. A study, that was performed in the past by ENEA for proper environmental analysis, has collected environmental samples that were used for the characterization of the unsaturated zone. Several stratigraphies were identified; they are mainly made up of loamy sand, sandy loam, gravel, sand, silty gravel, silt, silty loam. The performed analysis was focused on the shallow aquifer, due to the results obtained in (Testoni et al., 2015). In fact, it was deduced that the detailed study of the shallow aquifer has proved to be sufficient to protect the deep aquifer from the radiological contamination point of view. In particular, developed scenarios showed a concentration negligible, from the radiological risk point of view, at the interface between shallow aquifer and deep aquifer. In Table 2, the stratigraphies considered in this work are reported.

Table 2 Stratigraphies that characterize the investigated site

Altitude above sea level [m a.s.l.]	Depth below the ground [m]	Type of soil
Stratigraphy 1		
175.00-174.00	0.00-1.00	Sand
174.00-171.00	1.00-4.00	Silty Gravel
171.00-170.50	4.00-4.50	Sand
170.50-145.00	4.50-30.00	Sandy Gravel
145.00-130.50	30.00-44.50	Fine Gravelly Sand
130.50-130.00	44.50-45.00	Clay
Stratigraphy 2		
170.80-170.10	0.00-0.70	Loamy Sand
170.10-167.80	0.70-3.00	Sandy Loam
167.80-166.40	3.00-4.40	Silty Gravel
166.40-140.80	4.40-30.00	Sandy Gravel
140.80-126.30	30.00-44.50	Fine Gravelly Sand
126.30-125.80	44.50-45.00	Clay
Stratigraphy 3		
170.00-169.70	0.00-0.30	Silt
169.70-167.80	0.30-2.20	Sandy Loam
167.80-165.70	2.20-4.30	Silty Gravel
165.70-140.00	4.30-30.00	Sandy Gravel
140.00-125.50	30.00-44.50	Fine Gravelly Sand
125.50-125.00	44.50-45.00	Clay
Stratigraphy 4		
171.50-170.50	0.00-1.00	Loamy Sand
170.50-167.20	1.00-4.30	Gravel
167.20-141.50	4.30-30.00	Sandy Gravel
141.50-127.00	30.00-44.50	Fine Gravelly Sand
127.00-126.50	44.50-45.00	Clay
Stratigraphy 5		
173.00-172.00	0.00-1.00	Sand
172.00-171.70	1.00-1.30	Gravel
171.70-170.90	1.30-2.10	Sandy Loam
170.90-168.90	2.10-4.10	Silt Loam
168.90-143.00	4.10-30.00	Sandy Gravel
143.00-128.50	30.00-44.50	Fine Gravelly Sand
128.50-128.00	44.50-45.00	Clay
Stratigraphy 6		
171.50-171.30	0.00-0.20	Loamy Sand
171.30-170.50	0.20-1.00	Sandy Loam
170.50-167.50	1.00-4.00	Gravel
167.00-166.45	4.00-4.55	Sand
166.45-141.50	4.55-30.00	Sandy Gravel
141.50-127.00	30.00-44.50	Fine Gravelly Sand
127.00-126.50	44.50-45.00	Clay
Stratigraphy 7		
170.00-169.50	0.00-0.50	Loamy Sand
169.50-168.40	0.50-1.60	Sandy Loam
168.40-167.50	1.60-2.50	Sand
167.50-165.55	2.50-4.45	Sandy Gravel
165.55-140.00	4.45-30.00	Sandy Gravel
140.00-125.50	30.00-44.50	Fine Gravelly Sand
125.50-125.00	44.50-45.00	Clay
Stratigraphy 8		
171.00-170.50	0.00-0.50	Loamy Sand
170.50-166.50	0.50-4.50	Sand
166.50-141.00	4.50-30.00	Sandy Gravel
141.00-126.50	30.00-44.50	Fine Gravelly Sand
126.50-126.00	44.50-45.00	Clay

As far as the radionuclide concentration is concerned, the analysis of databases of the Regional Agency for the Protection of the Environment of Piedmont (ARPA Piemonte) (ARPA web page) has highlighted an almost constant Cs-137 concentration in the samples collected in correspondence of the well E5/6 in the period 2009 – 2015. Cs-137 concentration is in the range of 0.0266 – 0.0739 Bq/l.

5. Model setup and application

The performed analysis was run for a simulation period of 365 days. First, the hydrogeological model was implemented coupling unsaturated zone model and saturated zone model through HYDRUS 1D and MODFLOW, respectively. The unsaturated zone was analyzed on the basis of available data: topography, water table level, several stratigraphy collected in the area. Starting from these data, the investigated site was divided in 8 domains that show similar soil and hydrogeological properties (Figure 4). Each domain was represented by a one-dimensional profile, characterized by a specific stratigraphy (Table 2) and that was discretized with a grid spacing from 1 cm to 15 cm. For each unsaturated zone profile, grass cover and evapotranspiration was assumed. The meteorological data were collected by the local meteorological station. The data were provided by ENEA. In Table 3, the meteorological data are reported; they represent the mean annual behavior of precipitation and transpiration, that were introduced in the calculations.

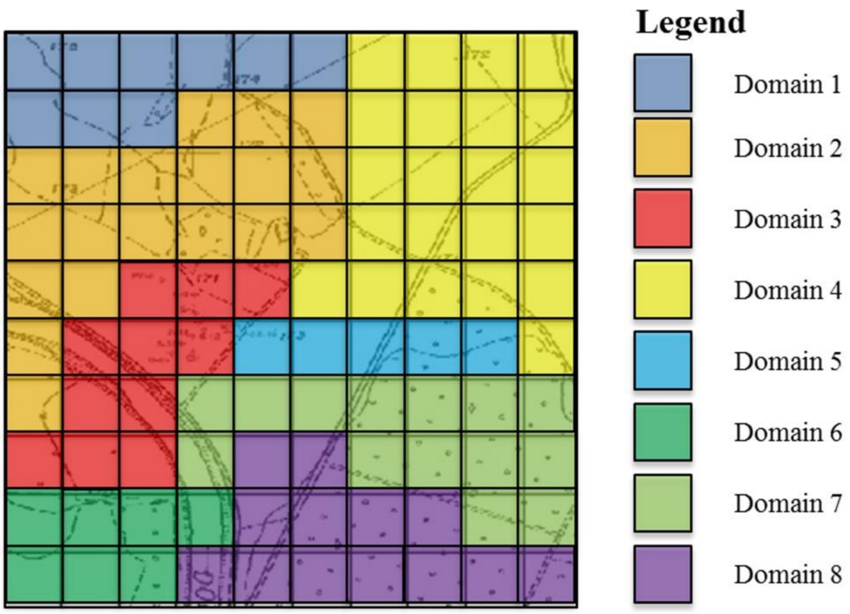

Figure 4 MODFLOW domains used to define HYDRUS soil profiles in the developed problem.

Table 3 Meteorological data collected by the local meteorological station .

Time [d]	Precipitation rate [mm/d]	Potential transpiration [mm/d]
30	0.0002	0.000015
60	0.006225	0.000001
90	0.00286	0.000018
120	0.04684	0.00002
150	0.051	0.000015
180	0.0721	0.00003
210	0.01132	0.000011
240	0.01905	0.000005
270	0.020325	0.000001
300	0.02562	0.000001
330	0.0006	0.000001
360	0.01644	0.000001

The groundwater model was discretized in cells 100 m by 100 m in x-direction and y-direction. The subsoil profile was divided in three main layers: sandy gravel (almost 30 m thick), fine gravelly sand (almost 12 m thick), clayey silt (0.5 m thick). The detailed stratigraphies in Table 2 include also the unsaturated zone profile. The grid spacing was defined by means of an iterative approach, until the convergence of the measured and simulated hydraulic heads was reached.

The unsaturated zone and the groundwater models were integrated and the water table levels and the exchanged water fluxes between unsaturated zone and saturated zone were obtained.

As far as the radionuclide transport is concerned, the radioactivity measurements performed by ARPA (ARPA web page) in 2009-2015 were analyzed. This study identified a source of Cs-137 is in the range of 0.0266-0.0739 Bq/l, in correspondence of well E5/6 for the period. An hypothetical source of Cs-137 equal to 0.05 Bq/l was implemented as punctual source term in the environment in correspondence of the well E5/6, in order to represent a real condition of the investigated area and to compare simulated data with measurement data. Comparing the position of the wells and the subdivision of the unsaturated medium in zones, the identified source term is located in domain 8. Concerning the approach, the transport in the unsaturated zone was considered introducing the interaction of the Cs-137 with the solid matrix by means of the distribution coefficient, the radioactive decay, and the diffusion and dispersion phenomena. In Table 4, the parameters that characterize the layers of the unsaturated zone are shown. The data useful to solve van Genuchten equations (eq_s. 3-4) were obtained through the software database, Rosetta (Schaap et al., 2001), introducing the types of soil. Distribution coefficient, diffusion coefficient and porosity were found in studies performed by Iezzi et al. (2009), and Varalda et al. (2006). In particular, Iezzi et al. determined empirically distribution coefficients, applying analytical procedures based on ASTM D 4319-93. Instead, Varalda et al. made investigations in situ in the Saluggia nuclear site. They studied in detail the hydrogeological system on the basis of collected samples and laboratory analysis. Then, the output concentration obtained by the unsaturated zone was introduced as input data in the saturated zone model. In Table 5, the soil characteristics of the saturated zone are reported. These data allow to solve Darcy's law (eq. 5) and contaminant transport. The introduction of the radionuclide transport needs the refinement of the space discretization in particular near the imposed radioactive source, in order to obtain more accurate concentration estimation. The model grid consists of 17 rows and 12 columns, with a grid spacing of 50-100 m in the x-direction and of 50-100 m in the y-direction. The z-direction consists of three layers, such as those ones of the hydrogeological model.

Table 4 Parameters that characterize the layers of the unsaturated zone (Iezzi et al. (2009), and Varalda et al. (2006)).

Soil Type	**Θ_r** [-]	**Θ_s** [-]	**α** [m^{-1}]	**n** [-]	**Ks** [m/day]	**ρ_b** [kg/m^3]	**δ** [-]	**D** [m^2/y]	**K_d** [m^3/kg]
Loamy Sand	0.057	0.41	12.4	2.28	3.502	1800	0.25	0.00221	0.03
Sand	0.045	0.43	14.5	2.68	7.128	1800	0.25	0.00221	0.03
Sandy Loam	0.065	0.41	7.5	1.89	1.061	1500	0.25	0.00221	0.03
Gravel	0.03	0.34	20	3	269.57	1500	0.25	0.00221	0.03
Silty Gravel	0.03	0.23	20	2.57	55.296	1500	0.25	0.00221	0.03
Silt	0.034	0.46	1.6	1.37	0.06	1500	0.25	0.00221	0.03
Silt Loam	0.067	0.45	2	1.41	0.108	1500	0.25	0.00221	0.03

Θ_r = the residual water content; Θ_s = the saturated water content; α = the inverse of the air-entry value (or bubble pressure); n = the pore size distribution index; K_s = the saturated hydraulic conductivity; ρ_b = the bulk density; δ = the effective porosity; D = the diffusion coefficient; K_d = the distribution coefficient.

Table 5 Soil characteristics of the saturated zone. (Iezzi et al. (2009), and Varalda et al. (2006)).

Soil Type	**ρ_b** [kg/m^3]	**δ** [-]	**D** [m^2/y]	**K_d** [m^3/kg]
Sandy gravel & Fine gravelly sand	1800	0.25	0.00221	0.03
Clayey silt	2500	0.08	0.0000315	0.08

6. Results

6.1 Coupling of unsaturated zone and saturated zone from the hydrogeological point of view

First, the coupling was investigated from the hydrogeological point of view. The software HYDRUS 1D and MODFLOW were used to investigate the unsaturated zone and saturated zone, respectively. The soil profiles that characterize the unsaturated zone were introduced. These profiles are described from the stratigraphies 1-8 reported in Table 2. In particular, unsaturated zone is represented by layers up to 4.10-4.55 m below the ground. This quote represents the water table level in each domain. The implementation of unsaturated zone through HYDRUS 1D requires the introduction of: layer properties to apply van Genuchten equations (eqs. 3-4, and Table 4); the profile of pressure head along the subsoil depth, which makes it possible to identify the position of water table level; the subsoil depth involved in the transpiration phenomenon; the plant root water uptake parameters obtained by the software database (Wesseling, and Brandyk, 1985); the meteorological data (Table 3). All these information are required for each stratigraphy. The MODFLOW scripts were implemented introducing the hydrogeological framework of the area under investigation. The hydrogeological framework of the saturated zone of the investigated nuclear site was analyzed in detail in Testoni et al. (2015). A qualitative study on the collected data in the site by ENEA and ARPA was performed, identifying the main dominant phenomena that regulate the groundwater flow: regional recharge due to glaciers and snow melting in spring and summer, and river dynamics which drains groundwater in the flood plain most of the year. Then, an hydrogeological model was implemented and calibrated. The simulated dynamics allowed to evaluate travel times in groundwater and compare the obtained estimation with measurements.

For what concern the modelling performed in this paper, only a part of the nuclear site was considered in comparison of the area investigated in (Testoni et al., 2015), due to available unsaturated zone data only in a part of the nuclear site. Topography of the area, hydraulic heads, subsoil stratigraphy, and layer properties (e.g. hydraulic conductivity, bulk density, porosity, etc.) were implemented in the model. One of the main boundary condition is represented by the river. The parameters introduced to describe the river are: flow, elevation, bottom, section, hydraulic conductivity of the streambed. Another boundary condition introduced is the time variant specified head in the north part and in the south part of the area (Figure 3), in order to impose the condition due to the surrounding of the investigated site. In addition, the groundwater model was subdivided in the 8 domains and at each of these domains was associated the corresponding unsaturated profile. This condition allows the exchange of information (water table level and flow fluxes) between unsaturated zone and groundwater. In Figure 5, the comparison between measured and simulated hydraulic head is shown for two monitoring wells, BH and P13, that are located in the domains 8 and 7, respectively. The comparison between measured and simulated hydraulic head in the shallow groundwater shows a good agreement of the results; the relative error between measured and simulated data was estimated lower than 1 % in each well of the investigated site.

The hydrogeological interaction made it possible to identify the dynamic of water table level and the exchanged fluxes between the unsaturated zone and saturated zone.

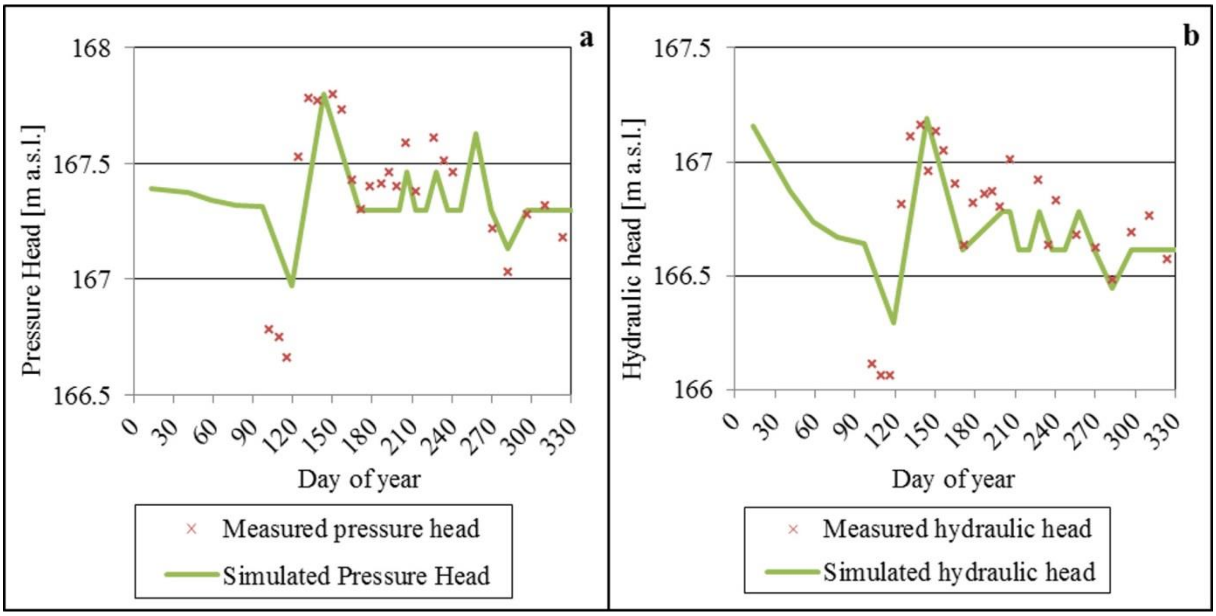

Figure 5 Comparison between measured and simulated hydraulic head in the shallow groundwater, in the monitoring wells BH (a) and P13 (b).

6.2 Radionuclide transport in unsaturated zone

The radionuclide transport in the unsaturated zone was investigated to implement the transport in the unsaturated domain 8 (chapter 5), by means of HYDRUS 1D tool. An initial concentration of Cs-137 was assumed equal to 0.05 Bq/l. The dynamic of water table level obtained by the hydrogeological coupled model was implemented as main boundary condition. Starting from the detected concentration data, different test cases were developed. Three different initial conditions about the radioactive source in the subsoil were hypothesized, to highlight the importance of the coupling of unsaturated zone and saturated zone through suitable software tools:

- the first radioactive source involves the entire unsaturated profile;
- the second one considers a Cs-137 release in the first 4 m of the soil depth;
- the third case implements a radioactive source in the first 2 m of the subsoil depth.

These three conditions allow us to highlight the behavior of the radionuclide transport when the radioactive source is located far or near the water table level. In the domain 8, the initial level of water table was located at 4.5 m below the ground.

For each initial condition two different stratigraphies were assumed: the first one corresponds to the real stratigraphy of the unsaturated profile of the zone 8 (loamy sand [0-0.5 m below the ground] and sand [0.5-4.5 m below the ground] as shown in Table 2); the second one considers a layer of clay instead of the layer of sand, in order to consider a different interaction between radionuclide and solid matrix (distribution coefficient equal to 0.08 m^3/kg (Iezzi et al., 2009)). The latter assumption represents a realistic condition for the surrounding of the investigated site. In fact, the nuclear site is located in an area dominated by the river dynamic, and as a consequence the stratigraphy of the zone cannot be assumed homogeneous on a large scale. Thus, the hypothesized stratigraphy with a clay layer is realistic. The clay layer is featured by a low hydraulic conductivity, therefore it is able to slow down the radionuclide migration. Understanding the radionuclide dynamics in these different scenarios allows to better know and manage the potential radiological risks related to nuclear facilities.

As result, six different scenarios were analyzed:

- case a) real stratigraphy (loamy sand and sand) and source term involves the entire unsaturated profile;
- case b) hypothetical stratigraphy (loamy sand and clay) and source term involves the entire unsaturated profile;
- case c) real stratigraphy (loamy sand and sand) and source term involves the first 4 m of the subsoil depth;
- case d) hypothetical stratigraphy (loamy sand and clay) and source term involves the first 4 m of the subsoil depth;
- case e) real stratigraphy (loamy sand and sand) and source term involves the first 2 m of the subsoil depth;
- case f) hypothetical stratigraphy (loamy sand and clay) and source term involves the first 2 m of the subsoil depth.

In Figure 6, Cs-137 concentration in the unsaturated domain 8 is shown for all test cases. In the Figures 6a, 6c, 6e, the three different initial contamination conditions in the real soil depth stratigraphy are shown, instead in Figures 6b, 6d, 6f, the Cs-137 concentration in the hypothetical stratigraphy loamy sand and clay is shown. The comparison between these different cases shows, as expected, that the hypothesized clay presence slow down the mass transport in depth.

In cases of Figures 6a and 6b, the concentration order of magnitude is the same and variations in time can be observed at the end of the unsaturated zone. In cases of Figures 6c and 6d, a slight variation at the water table level occurs. Instead, in the last two cases (e and f), the concentration at the interface of unsaturated and saturated zone is null. These concentration profiles depend on the initial location of the radioactive source. In particular, in the first two cases (a and b) the radioactive source was hypothesized along the entire unsaturated zone. The third and fourth cases (c and d) have investigated a source distributed for 4 m below the ground and up to 50 cm from the water table level. Instead, the last cases (e and f) do not involve the area near the water table level. The simulation results underline the effect of the clay barrier to radionuclide migration towards groundwater. As we expected, the low hydraulic conductivity, that mainly characterizes the contaminant behavior into clay layer, influences the migration, as well as the interaction between solid matrix and radionuclide, which is represented by the distribution coefficient.

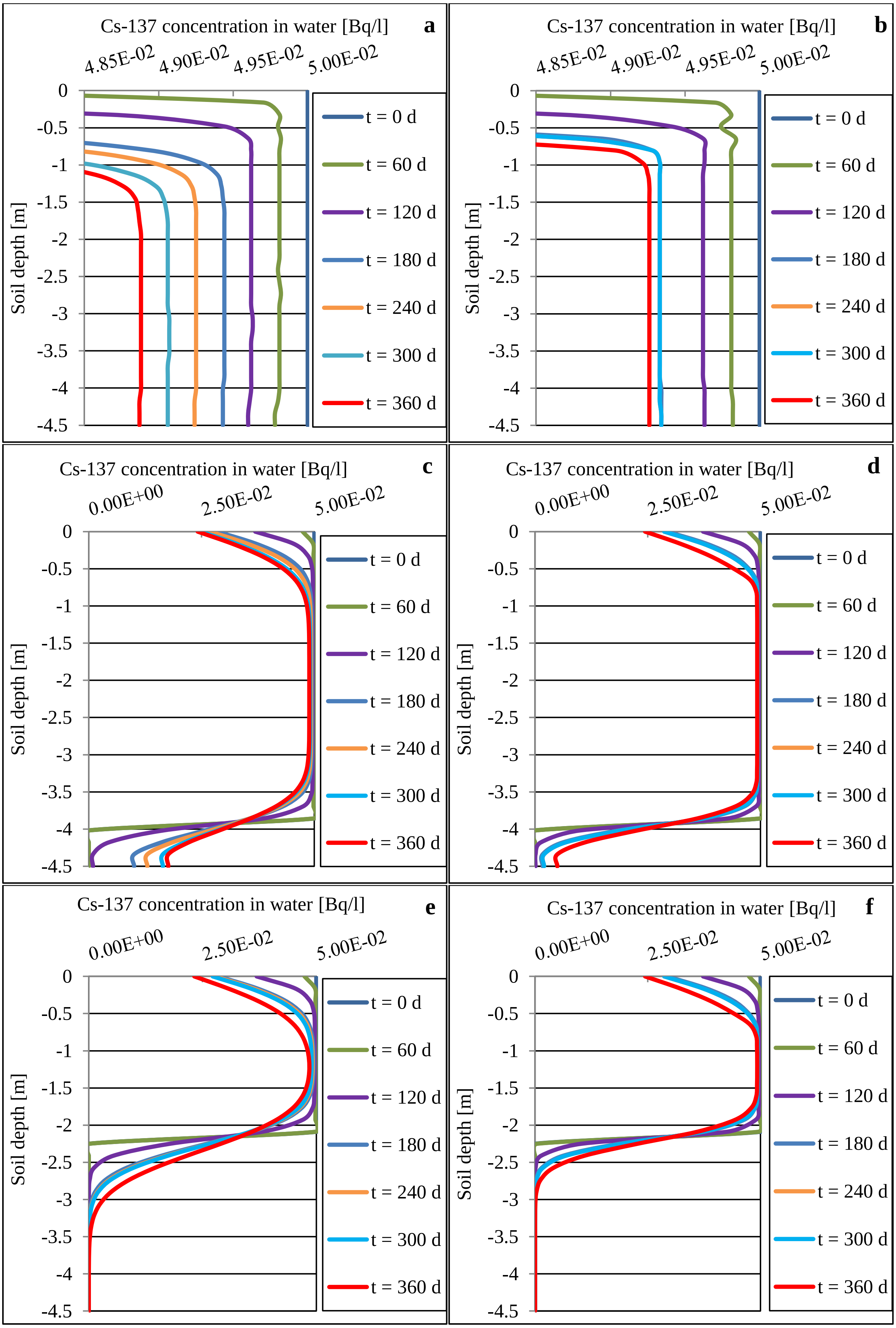

Figure 6 Cs-137 concentration in the unsaturated zone 8 with different initial conditions of source release for the case with sandy loam and sand soil profile (a, c, e) and for the case with sandy loam and clay soil profile (b, d, f).

As far as the two different unsaturated zone stratigraphies (loamy sand-sand, and loamy sand-clay) are concerned, the travel times and the retardation factors were estimated for the sand and clay layers. An approximated estimation of the travel time was performed dividing the calculated water flux in each layer for the layer thickness. In Table 6, the travel times and retardation factors in sand and clay layers are shown. This approximated analysis shows that the travel time in clay layer is higher than in sand layer. The available stratigraphy characterization allowed to estimate the travel times approximately. In (Testoni et al., 2014), the high impact on the evaluation of water flux of a detailed subsoil characterization (e.g. measurement of distribution coefficient, soil bulk density, etc.) and of the external factors, such as evapotranspiration and precipitation phenomena, and as a consequence of the travel time, was demonstrated. Future works may focus on the collection of detailed stratigraphy in terms of composition of sand, clay, silt, bulk density, etc., in order to improve the assessment of travel times. Instead, as far as the retardation factor is concerned, it was estimated for the limit conditions of residual water content and saturated water content, that characterizes the domain 8 (sand parameters are reported in Table 4; clay residual and saturated water content are 0.068 and 0.38, respectively). In fact, the retardation factor (eq. 9) depends on the subsoil characteristic, as well as on the radionuclide by means of the distribution coefficient. The retardation factor in clay layer results almost one order of magnitude higher than in sand and loamy sand layers. This analysis can be considered a preliminary and conservative assessment of the travel times and the retardation factors. The introduction of geochemical aspects, that allow to express the distribution coefficient variation in space and radionuclide transport in time, and/or the coupling of modelling analysis with experimental tests (e.g. measurement of K_d and soil characteristics, such as subsoil bulk density) could increase and improve the knowledge of the travel dynamic in the unsaturated zone.

Table 6 Travel times and retardation factors of sand and clay layers.

	Travel Times [y]	
	Sand	Clay
Mean	1.04	1.21
Minimum	0.15	0.28
Maximum	3.76	3.82
	Retardation Factor [-]	
	Sand	Clay
Minimum	$1.06 \cdot 10^2$	$3.16 \cdot 10^3$
Maximum	10^3	$1.74 \cdot 10^4$

6.3 Coupling of Unsaturated Zone and Saturated Zone from the radionuclide transport point of view

After the investigation of the hydrogeological system (unsaturated zone and saturated zone) and of the radionuclide transport in unsaturated zone, the radionuclide migration in groundwater was investigated, considering its concentration in unsaturated zone. As far as the modelling is concerned, it was developed by means of MODFLOW-MT3DMS software tools. The dynamic of Cs-137 in the unsaturated zone for the transient of 365 days was assumed as input for the transport in the groundwater. In the integrated model, the hydrogeological data obtained by the hydrogeological coupling (e.g. variation of the water table level in time) were introduced as boundary conditions. In Figure 7, the regional groundwater dynamic is shown. The continuous lines represent the equipotential lines of the hydraulic head in groundwater [m a.s.l.] in the shallow aquifer. The flow direction is perpendicular to hydraulic-head lines.

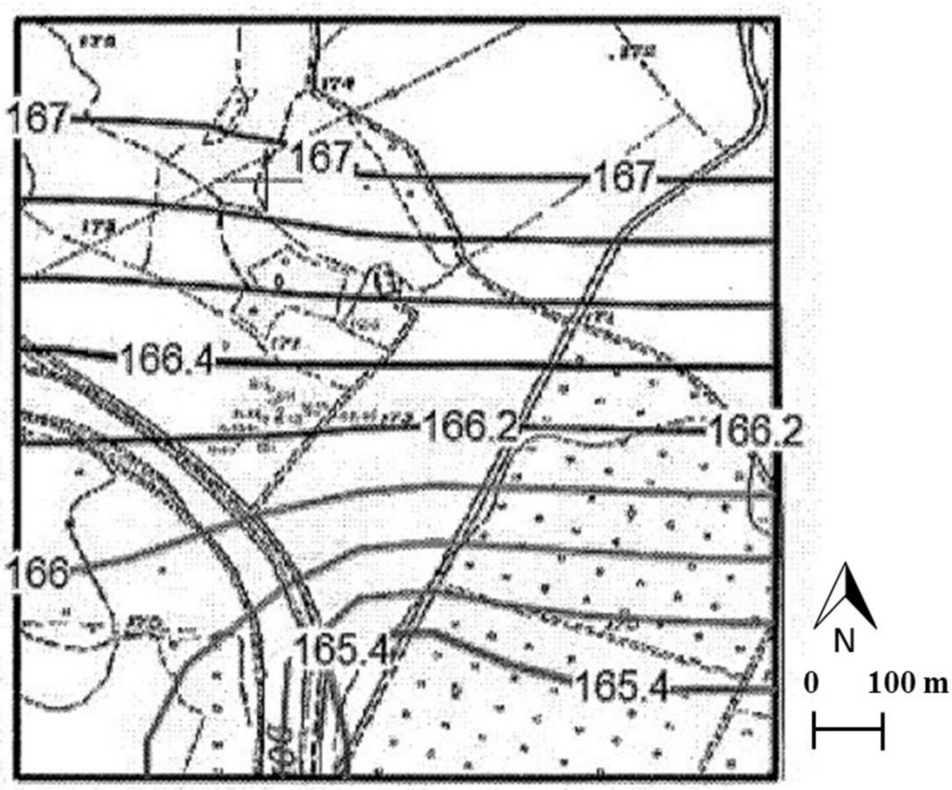

Figure 7 Regional groundwater dynamic of the investigated site. Continuous lines represent the hydraulic head line [m a.s.l.].

In Figure 8, the Cs-137 concentration in the shallow groundwater at the end of the transient is shown for the first two hypothesized contamination conditions (see paragraph 6.2). The test cases, that involve an hypothesized release in the first 2 m of the soil depth (cases e and f), were not taken into account due to the negligible concentration estimated at the interface between unsaturated zone and saturated zone. From a qualitative and quantitative point of view, the comparison between cases a and b shows a very similar concentration distribution in the area. The impact of clay layer instead of sand layer is more evident comparing cases c and d. In fact, the area involved by the Cs-137 migration is larger for the real stratigraphy with sand, due to the higher hydraulic conductivity than the clay layer. When a nuclear facility must be installed, the choice of site is fundamental for what concern the migration of radionuclide in accidental conditions. All investigated cases involve a concentration of cesium negligible from the radiological risk point of view. In any case, the measurement of concentration must be carried out to control the level of radioactivity, because it results higher than the minimum detectable activity by the monitoring devices.

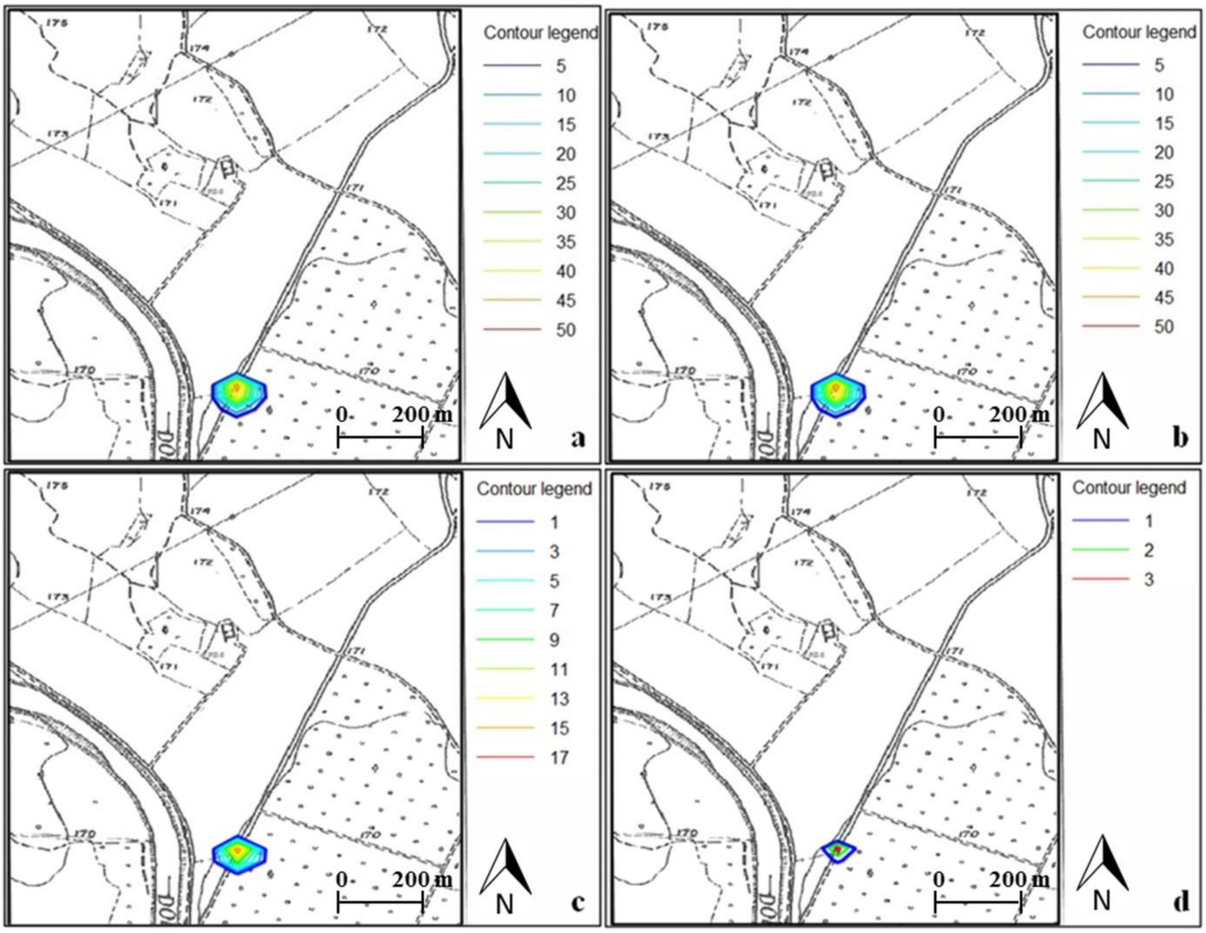

Figure 8 Cs-137 concentration in 10^{-3} Bq/l in the shallow groundwater at the end of the simulated transient (365 days) for the first contamination condition and for the two different stratigraphy of the unsaturated zone (cases a, and b), and for the second contamination condition for the two different stratigraphy of the unsaturated zone (cases c, and d).

To demonstrate, the qualitative and quantitative effectiveness of unsaturated zone, another case was implemented. The Cs-137 concentration in the shallow groundwater without taking into account the unsaturated zone at the end of the transient was assessed. As example, the first contamination condition considering the real stratigraphy of the unsaturated zone (loamy sand and sand) was compared with the initial radioactive source without implementing the unsaturated zone. This comparison is shown in Figure 9. The case without unsaturated zone (Fig.9b) appears more cautionary from the contamination distribution point of view. It represents a worse situation than the case that takes into account the unsaturated zone. However, Figure 9a underlines the barrier effect of the unsaturated zone. The area involved in the radionuclide migration is larger in the case of absence of unsaturated zone. This means that the unsaturated zone can be a natural barrier (barrier effect) to delay the cesium migration into groundwater, protecting groundwater from possible radiological risks. The implementation of unsaturated zone describes a realistic system: if unsaturated zone is neglected, the environmental system is oversimplified and the results are partially to plan a suitable environmental monitoring network and/or to identify restoration activities in accidental cases. Analyzing the simulated concentration lines, this observation is confirmed. The concentration line of $5 \cdot 10^{-3}$ Bq/l is taken as reference, because this concentration represents the lowest detectable quantity from the common used monitoring devices (Porzio L., 2009). Thus, it can be observed that the maximum distance between this concentration level and the source term is almost 65 m in the groundwater flow direction for the case with unsaturated zone, while it is almost 80 m for the case which is neglecting the effect of the unsaturated zone. These results show that the barrier effect of the unsaturated zone can have an impact on the delay of radionuclide migration in the groundwater.

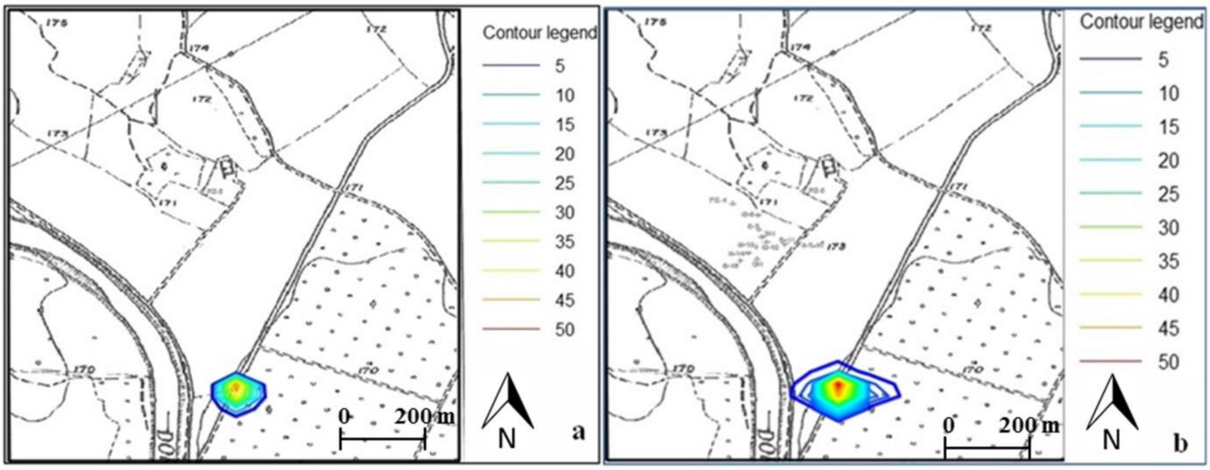

Figure 9 Cs-137 concentration in 10^{-3} Bq/l in the shallow groundwater at the end of the simulated transient (365 days) taking into account the unsaturated zone (a) and without implementing unsaturated zone (b).

In our study, we considered unsaturated zone and saturated zone both from the hydrogeological and radionuclide transport point of view as a unique system. In this way, it is possible to obtain an overview of the system under investigation, to plan a more suitable environmental and radiological monitoring network both for the unsaturated zone and groundwater, and also to take better decision in accidental situations. In particular, the developed test cases showed the importance of the unsaturated zone as natural barrier to protect the groundwater from the radiological risk. In the studied cases, unsaturated zone can delay the migration towards groundwater. Specific devices could be located in the unsaturated zone to monitor it, especially in a soil zone made up of sand. In situ, cesium measurement techniques can be conducted with portable survey instruments, such as Geiger-Muller detector, and gamma ray spectrometer. In addition, analytical methods for measuring Cs-137 in environmental samples can be applied. For what concern the analysis of water and soil samples, instrumental neutron activation analysis or radiochemical techniques, which require a sample preparation, provide good detection sensitivities, precision and bias. Detailed explanation of the different applicable techniques is stated by IAEA (e.g. IAEA, 2011), American Society for Testing and Materials International (e.g. ASTM, 2014), Italian National Institute for Environmental Protection and Research (e.g. ISPRA, 2014), etc. Thus, the detection of radionuclides makes it possible to plan a restoration or a mitigation action in order to avoid or delay the radionuclide migration in the groundwater.

7. Conclusions

The coupling of unsaturated zone and saturated zone from the hydrogeological and radionuclide transport point of view through HYDRUS 1D and MODFLOW/MT3DMS was proposed in this work. This approach was tested developing a case study on an Italian nuclear site. The comparison between measured and simulated data was performed in order to validate the proposed approach. As far as the hydraulic head is concerned, a relative error lower than 1 % was obtained modeling the dynamic of water flux coupling HYDRUS 1D and MODFLOW. For what concern the radionuclide transport, different test cases, that involved Cs-137, were developed analyzing the radionuclide migration starting from concentration data measured by ARPA in the area. In addition, an estimation of travel times and retardation factors in the unsaturated zone was performed. This analysis has highlighted the barrier effect of the unsaturated zone in case of accidental release from a nuclear facility. The unsaturated zone may represent a natural delay and/or capture system for radionuclide into the environment. This natural system can be used to improve monitoring activities and/or restoration activities.

The coupling of hydrogeological and radionuclide transport aspects in the unsaturated and saturated zone makes it possible to obtain an overview of the contamination migration. A detailed model of the transport can be implemented with this approach. In particular, the effect of the unsaturated zone is important to understand possible radiological risks for the health of human beings due to groundwater use. In fact, groundwater represents a fundamental natural resource for the population, that must be safeguarded. These analyses can improve the Safety Assessment studies on the environmental impact of a nuclear facility.

In future works, improvements on the proposed approach may be introduced. First, starting from the characterization data of the site, an optimization method to identify the sufficient number of domains with similar soil and hydrogeological properties may be proposed to better represent the unsaturated zone. Then, how many time steps are necessary to collect hydrogeological features (e.g. water table level) may be identified in order to implement a more realistic model. Another important aspect that has to be investigated is the water table level near the top surface that could create instability in the numerical model of the coupling. Finally, the introduction of further geochemical aspects to assess the travel times in the unsaturated zone with higher accuracy should be also investigated, in order to increase the understanding of the barrier effect for the radionuclide transport.

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