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Abstract

Radioactive waste management is a strategic activity that is adopted in the nuclear field to safeguard the operating staff, the population and the environment from radiological risks. Studies that support the environmental assessment of a nuclear site offer insight into the understanding of the transport of radionuclides in environmental matrices (e.g. soil, groundwater, surface water, etc.). These studies, that involve the propagation of a radiological source in the environment, are conducted to identify how to approach the safety assessment of current or future nuclear facilities. This work analyzes the source term as the key point in the modeling of the transport of radionuclides in groundwater and soil. A methodological approach, which focuses on the dynamic of the source term in space and time, was applied to model the transport of radionuclides. This approach can be used to plan reliable environmental monitoring networks. The analysis was performed at the nuclear site of Saluggia, Vercelli (Italy).

Keywords: radionuclide transport; shallow groundwater; transport time; source term; environmental monitoring; radiological impact

1. Introduction

In the nuclear field, radioactive waste management is a strategic activity that is adopted to safeguard the operating staff, the population and the environment from radiological risks, whose final aim is the design and the construction of a permanent repository. Each nuclear facility is characterized by a specific inventory, in which the type of radionuclides and their radioactivity must be known in detail, and by several containment barriers, which prevent the release of radioactive material or delay the transport of radionuclides outside the facility (Yim and Simonson, 2000).
In accidental situations, these radionuclides, or some of them, can be released into the environment. In this case, they are identified as source terms. The aim of this work was to analyze the transport of radioactive pollutants through soil and groundwater. The evaluation of the transport of radionuclides is a useful tool to design a reliable environmental monitoring network or to plan adequate mitigation works or other interventions in the case of accidental events, as well as to create a background of knowledge for accidental leakages from nuclear facilities (e.g. Levenson and Rahn, 1981; Lee and Jeong, 2011).
IAEA defines source term as the magnitude, composition, form (physical and chemical) and mode of release (puff, intermittent or continuous) of radioactive elements (fission and/or activation products) released during a confinement loss from nuclear facilities (IAEA, 2008). The study of the source concerns the quantities and rates of release in groundwater, surface water, soil, etc. The

type, the time and the location of the release must also be identified. The radiological consequences can be grouped into three categories, to facilitate their assessment (IAEA, 2008): consequences inside the reactor building with the operating staff or personnel within the building being subject to doses; on-site consequences within the area of the nuclear site (outside the reactor building); off-site consequences (on population). This work examines the consequences outside the facilities and focuses on the source term and on transport mechanisms in order to evaluate the behavior of radionuclides in soil and groundwater.

Jakimavičiūtė-Maselienė and Cidzikienė (2015) have studied the modeling of tritium in underground water at the new NPP site in Lithuania by means of the FEFLOW code. Aquino et al. (2008, 2010) presented an overview of the progress achieved in the development of Eulerian-Lagrangian schemes, to approximate the transport of radionuclides in unsaturated porous media, and developed a new algorithm for the numerical simulation of radionuclide transport problems in saturated heterogeneous porous media. Lamego Simões Filho et al. (2013) modeled tritium dynamics in surface waters in order to evaluate the radiological impact of its potential release from nuclear power plants into the environment.

A methodological approach, which couples on-site measurement data and the use of suitable software, Modflow (Harbaugh, 2005) and MT3DMS (Chunmiao and Wang, 1999), was applied in this work. The results are useful to identify the main critical points in which radionuclides could accumulate and, consequently, to plan a suitable environmental monitoring of the area under investigation. All these aspects were applied and validated at the Saluggia nuclear site, Vercelli (Italy). The on-site measurements were periodically carried out by national authorities in the field of environmental monitoring of nuclear sites, ARPA and ISPRA (e.g. ARPA, 2013a). Two different transport analyses were carried out: the first in the Sogin area of the site, where four test cases were developed with a hypothetical Cs-137 as the source term; the second in the Avogadro area of the site, considering a hypothetical source of Sr-90. The choice of Cs-137 and Sr-90 was made on the basis of an accurate analysis of data published by ARPA in the 2009-2013 period.

2. Methodological approach

In the context of the environmental and safety assessment carried out in a nuclear site, the transport of radionuclides in environmental matrices (e.g. groundwater, soil, air, etc.) has to be investigated in a precise and detailed way in order to support the environmental monitoring of the nuclear site and the surrounding areas. A methodological approach is a useful tool to reach this goal. In this work, the analysis has focused on groundwater and soil, but the approach can be extended to any environmental matrix. The main steps of the methodological approach used to investigate the concentration in space and time, are:

- site description: the hydrogeological framework and the possible source terms that characterize the reference area;
- hydrogeological framework: evaluation of the hydrological features with identification of the flow lines as preferential radionuclide pathways;
- source study: the type of radionuclides and their characteristics;
- transport model: integration of hydrogeological data with transport phenomenon modeling in order to study the behavior of radionuclides in space and time;
- analysis of the results to identify any critical points in the area and to plan the management of any possible contamination.

As far as the site description is concerned, it is necessary to collect the data which characterize the hydrogeological framework: the type of aquifers, the stratigraphy of the soil profile, the topography, and the presence of rivers or canals. Human activities and the presence of potential radiological sources must be also identified.

In the second step, the collected hydrogeological data must be qualitatively analyzed to identify the phenomena (e.g. recharge, discharge, loss of water from streams, etc.) which characterize the investigated site. In this phase, the lack or the shortage of data must be considered in order to estimate the possible errors that could be made concerning the transport of radionuclides and also to establish how to direct further investigations or sensitivity analysis.
The study of the source term determines the types of radionuclides in each nuclear facility and their quantities, which represent the possible radiological risk for the site and the surrounding areas. After identification of the main hazardous radionuclides (IAEA, 2002, 2003), their different types of behavior (e.g. radioactive half-life, interaction between radionuclide and solid matrix) are evaluated.
As far as the transport evaluation is concerned, a suitable transport model has to be developed by means of appropriate software. Several realistic and possible scenarios can be considered to analyze possible critical situations, in order to evaluate the potential migration of radionuclides in the environment, to identify radioactivity accumulation areas and to evaluate the concentration of radionuclides in space and time.
The final step concerns checking the results, and analyzing the concentration of the radionuclides in space and time. This step makes it possible to identify the main migration pathways, the accumulation points of contaminants and the risky areas for the propagation of the radioactivity. All the information about artificial radionuclides in the environment must be managed, not only to try to restore the previous environmental conditions, but also to manage the contamination by means of removal and fixation, or environmental monitoring in the case of negligible contamination (IAEA, 2004).
The described approach can also be applied to plan a suitable and specific environmental monitoring network, according to the criteria imposed by national and international laws on nuclear monitoring activities.

3. Site description

The investigated area is the Saluggia nuclear site, Vercelli (Italy). The nuclear site, which is shown in Figure 1, can be divided in two main areas: the Sogin area, in which the Eurex plant, which is owned by Sogin, is located, and the Avogadro area, in which the temporary Avogadro repository is located (Porzio, 2009). Eurex is a nuclear fuel reprocessing plant which is currently being decommissioned. Some temporary new radioactive waste storage facilities and waste ponds, which collect the liquid effluents from the Eurex plant, are also located in the Sogin area. In the past, the Avogadro repository was a nuclear research reactor, but it was transformed into a temporary storage building for spent fuel.

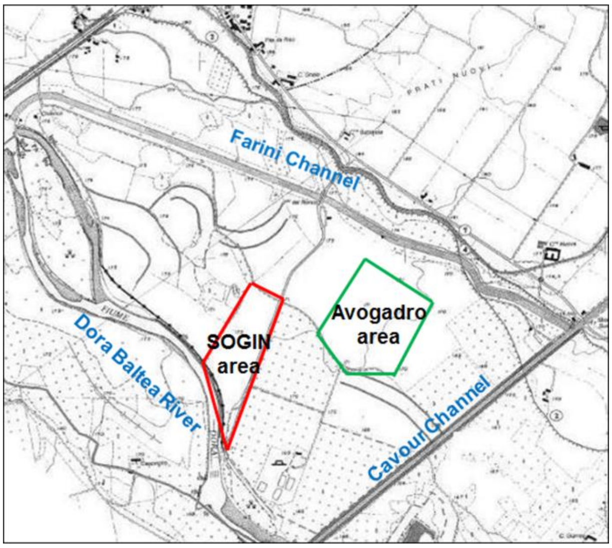

Figure 1 Saluggia nuclear site: Sogin area and Avogadro area.

All the aforementioned nuclear facilities are potential radiological source terms, because they are used to store radioactive materials. These materials can be divided into two types: conditioned materials and non-conditioned materials. The first ones are in a solid state, and they have already been treated and conditioned; the second ones can be solid materials (e.g. pump, filter, tube, etc.), which need to be reduced in volume and compacted, or liquid waste, which must be treated and conditioned before being converted into solid materials. Sogin reports show the main physical, chemical and radiochemical data of the liquid waste. The radioactive material in Saluggia, which has been divided into conditioned material and non-conditioned material on the basis of the Italian radioactive waste classification (Sogin, 2013) is shown in Table 1.

Table 1 Radioactive waste in the Saluggia nuclear site divided into non-conditioned material and conditioned material as well as the total activity with respect to the Italian radioactive waste classification (Sogin, 2013).

	I Category	II Category	III Category	Total
Non conditioned nuclear waste [m^3]	1232	998	317	2547
Conditioned nuclear waste [m^3]	0	166	25	191
Total activity [GBq]	0.33	$5.89 \cdot 10^4$	$3.3 \cdot 10^8$	

An analysis of the Italian radiological inventory of each nuclear facility according to the data of the Italian National Agency for New Technologies, Energy and Sustainable Economic Development shows the main α-emitters (U-234, U-235, U-238, Pu-238, Ra-226, Am-241, Th) and the main β-γ emitters (Cs-137, Sr-90, H-3, Co-60, Ni-63, Sm-151, Kr-85, Fe-55, C-14, Pm-147, Ni-59, Pu-241, Ru-106, Sb-125), in the Avogadro repository and in the Eurex plant. The α-emitters have a radioactive half-life of the order of one thousand years or more; the β-γ emitters, except C-14, Ni-63, Ni-59 and Sm-151, have a radioactive half-life of the order of some tens of years, or less. The elaboration of these data shows that the second waste category is constituted by almost 17% of generic α-emitters, 44% of β-γ emitters, 27% of Cs-137 and 7% of Sr-90; instead, the third waste category is constituted by almost 49% of β-γ emitters, 29% of Cs-137 and 21% of Sr-90.

4. Geological and hydrogeological framework

As far as the geological framework is concerned, the site is located in the flood plain of the Dora Baltea River, and is laterally confined by fluvioglacial terraces, which, to the North of the site, have an elevation of almost 9 meters above the flood plain, which then drops slightly toward the southern sector. In the flood plain, alluvial deposits overlap fluvioglacial deposits, to a depth of about 45 m; it is not possible to distinguish the precise boundary between the two lithologies, since they are both constituted by sand and gravel, with small interlayers of sandy silt. The stratigraphy below the fluvioglacial deposits is made up of alternating sandy and silty-clayey layers of the Villafranchian age. An analysis of other stratigraphies, which was performed in the Vercelli area closer to the Saluggia nuclear site (Varalda et al., 2006), shows a similar situation. The hydrogeological system is made up of two distinct aquifers (Iezzi et al., 2009; De Maio and Fiorucci, 2008): a shallow aquifer, with a mean thickness of 45 m in the examined area, and a deeper artesian aquifer (Fig. 2).

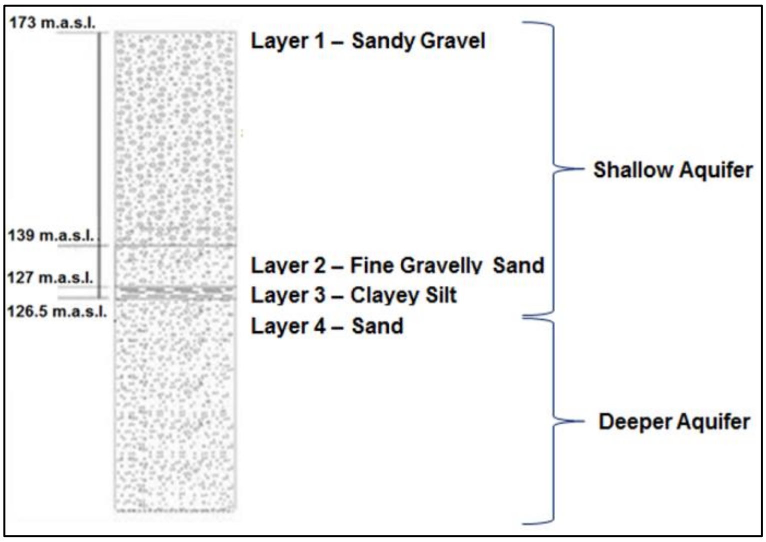

Figure 2 Soil stratigraphy of the nuclear site.

As far as the hydrology is concerned, the shallow aquifer is characterized by a regional groundwater recharge, mainly due to the water coming from the Ivrea morainic amphitheatre (Gianotti et al., 2015) and from rice paddies during their flooding. The local recharge is related to surface water, such as that of the Dora Baltea River, and several artificial canals in the investigated area; the precipitation effect is instead rather negligible. Adorni-Braccesi et al. (2001) studied the indirect infiltration of the rainfall, comparing physical methods and geochemical approaches, and found that locally the groundwater recharge is due more to the lateral recharge from the river than to the vertical recharge due to rainfall. The precipitations influence the surface water flow at a regional level and indirectly influence the groundwater recharge.

The shallow aquifer is characterized by a free water table, whose mean depth from the ground surface is about 10.5 m at the terrace and 3.5 m in the flood plain. Its flow direction is North-South, and it is characterized by local variations due to interaction with surface waters. The shallow aquifer is separated from the deeper artesian aquifer by an almost 0.5 m thick silty layer. The deeper aquifer is located in up to 100 m thick sandy layers. No detailed information is available about the deeper aquifer, thus this work has only considered the dynamics of the radionuclides in the shallow aquifer and evaluated the need to conduct further studies in the deeper aquifer.

The qualitative analysis of the piezometric head data and of literature studies on the hydrogeological framework (De Maio and Fiorucci, 2008; Iezzi et al., 2009) has made it possible to

identify two dominant phenomena which regulate the groundwater flow: the regional recharge, due to glaciers and snow melting in spring and summer, which increases the water table level, and the stream flows; the dynamics of the Dora Baltea River, which drains the groundwater in the flood plain most of the year.
The recharge and discharge effects due to the river are more evident closer to the river itself, whereas they gradually decrease toward the fluvioglacial terraces. The effect on the groundwater near the terrace is only due to regional phenomena. The river level, measured by means of a hydrometer, varies according to the water table oscillations measured by a piezometer near the river. Instead, no correlation has been found between the river level and the water table on the terrace, which changes due to the influence of the regional groundwater dynamics. The maximum water table level on the terrace is measured in summer; the maximum river head is instead observed in spring. In summer, the canals are tapped for agricultural purposes and in spring there are precipitations that influence the stream flows and the groundwater, although to a lesser extent.

5. Radiological monitoring network

The activities conducted at the Saluggia site, as well as in each nuclear site, have been planned, designed, realized and monitored in order to minimize the radiological and conventional risk for the population and the environment. Sogin has developed its environmental monitoring network over the years and organized the periodical monitoring of each environmental matrix. ARPA Piemonte, the regional environmental protection agency, periodically gathers several samples in each environmental matrix to conduct environmental controls. The monitoring results are also sent to ISPRA, the national nuclear authority and the national environmental protection agency.
As far as the transport of radionuclides in the groundwater is concerned, ARPA carries out environmental monitoring activities in some specific monitoring wells in the area. The most frequently encountered radionuclides in the groundwater are: alpha emitter, beta emitter, Cs-137, Co-60, Sr-90, H-3, Am-241. The monitoring wells in which water samples are collected are shown in Figure 3; the H2 and S_{pond} points were introduced as target for the transport simulation and the source term in correspondence to the waste pond-719, respectively.

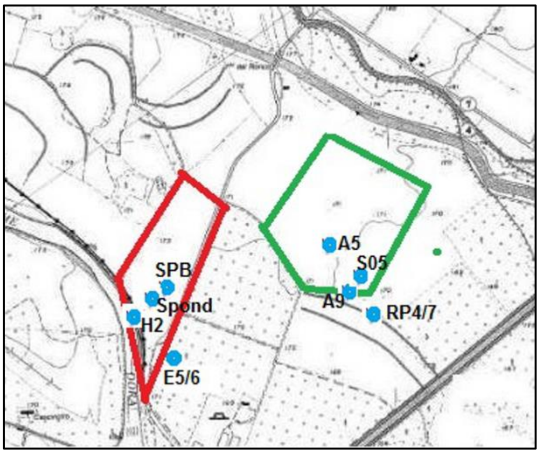

Figure 3 Monitoring wells in which water samples are collected in the Saluggia site.

Studies and simulations on transport dynamics in porous media must be coupled with on-site measurements, in order to establish the accuracy of the simulations and of the forecasting. The sensitivity of the measurement is specified by a key parameter, that is, the Minimum Detectable Activity (MDA). This parameter also immediately offers a rough idea about the importance of the data collected from the measurements and about the radiological consequences. The MDA that was obtained from the measurements which were carried out in the groundwater of the studied site

with ARPA instrumentations is shown in Table 2 (Porzio, 2009). In Figure 4, histograms have been used to show the percentage fraction on the total number of samples collected by ARPA during the 2009 – 2013 period in which the radioactivity measurement resulted larger or smaller than the MDA in the different monitoring wells in the area. No information is available on the samples collected in monitoring well S05 for the year 2013. In the Sogin area, Cs-137 was detected in E5/6. An in depth analysis of the ARPA reports showed contamination of Cs-137 in the soil samples which were measured in April 2013 (ARPA, 2013b). Starting from these data, test cases that involved a source of Cs-137 were developed in the Sogin area. Sr-90 was instead detected in the Avogadro area over the entire 2009-2013 period in each of the monitoring wells. Test cases which considered a Sr-90 source or a background concentration of Sr-90 in the environment were conducted for the Avogadro area.
An analysis of the histograms and of the measured concentrations, when a higher value than the MDA was detected, has shown a negligible radiological impact for the population and the environment.

Table 2 MDA for the main radionuclides measured in the groundwater using ARPA instrumentations (Porzio, 2009).

Radionuclides	α	β	Cs-137	Co-60	Am-241	H-3	Sr-90
MDA [Bq/l]	0.1	0.2	0.005	0.005	0.01	4	0.005

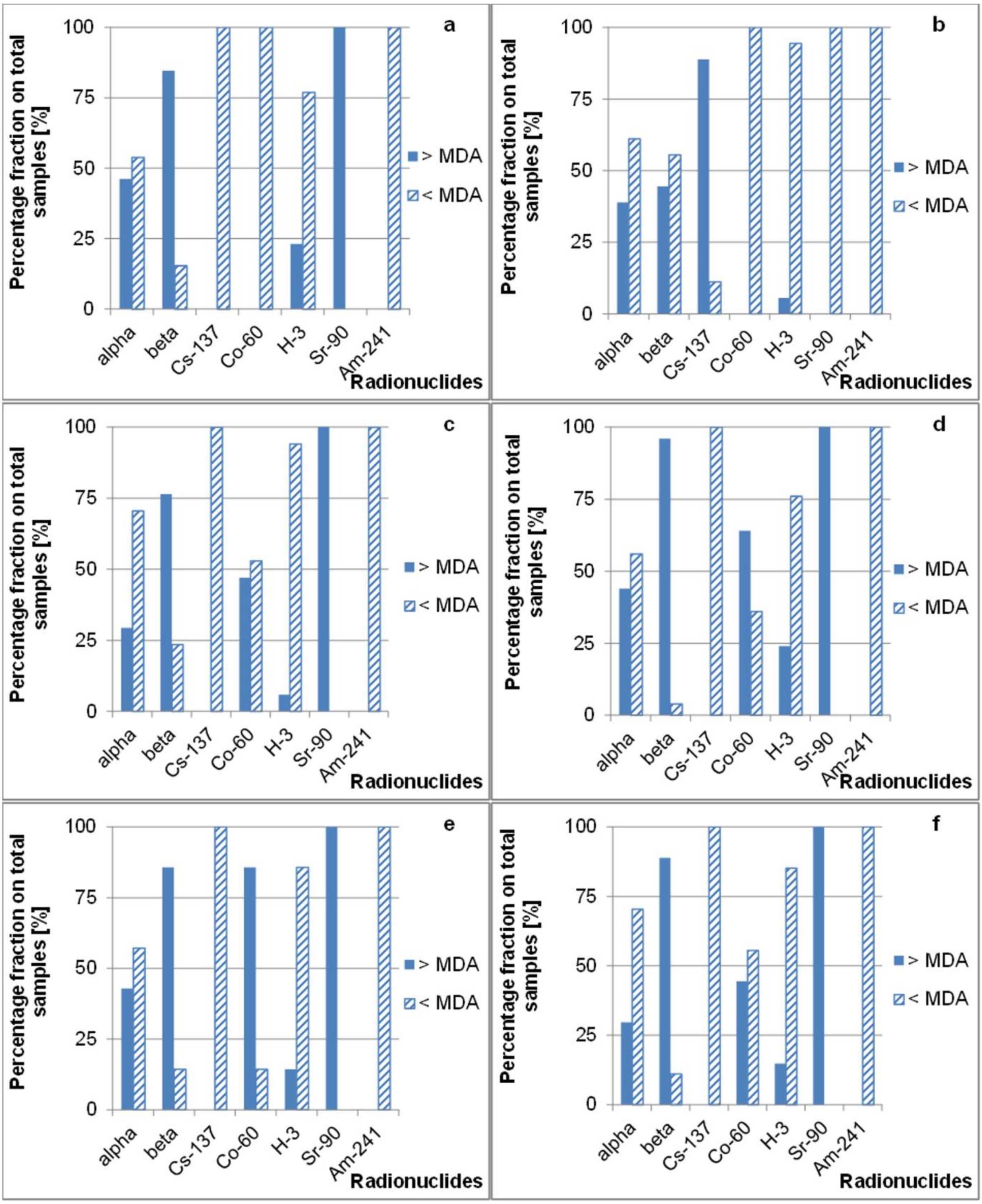

Figure 4 Histograms showing the percentage fraction on the total samples during the 2009 – 2013 period in which the radioactivity measurement resulted larger or smaller than the MDA for the different monitoring wells in the studied area: a) SPB, b) E5/6, c) A5, d) A9, e) S05, f) RP4/7.

6. Hydrogeological Model and Transport Time Estimation

The hydrogeological model was implemented by means of the MODFLOW code (Harbaugh, 2005) in order to evaluate the groundwater flow behavior. MODFLOW is a computer code which numerically solves the three-dimensional groundwater flow equation for a porous medium using a finite-difference method. The model was developed considering an almost 7 km^2 area, surrounded by a railway bridge to the North-West, by the Dora Baltea River to the South-West and West, by

the Cavour Canal to the South-East and by the Farini Canal, which runs almost parallel to the fluvioglacial terrace, to the North-East (Fig.1). The model focuses on the behavior of the shallow aquifer (Fig.2).
The model was discretized considering North-East – South-West as the x-direction and North-West – South-East as the y-direction. The z-direction corresponds to the stratigraphy of the system. The model grid consists of 62 rows and 54 columns, with a grid spacing of 20 m to 100 m in the x-direction and of 50 m in the y-direction. The z-direction consists of three layers, with the grid spacing shown in Figure 2. The grid spacing was defined by means of an iterative approach, until the convergence of the results was reached.
The soil stratigraphy, the hydraulic conductivities of the layers, the water table level, the piezometer measurements, the river flow data, as well as the characteristics of the river and canals (e.g. streambed head, streambed hydraulic conductivity, bank roughness, canal roughness, geometrical sections, sediment thickness, etc.) were collected to develop this model. A model calibration was performed over a transient period from May to August 1999, that is, a period in which groundwater and river head information was available. The model was calibrated by minimizing the difference between the measured pressure head data and the simulated pressure head data.
The simulated dynamics of the groundwater on the shallow aquifer surface is shown in Figure 5, and this can be considered a representative situation of the mean annual dynamics of the groundwater flow. The continuous lines represent the hydraulic head profile; the arrows represent the direction of the flow lines.

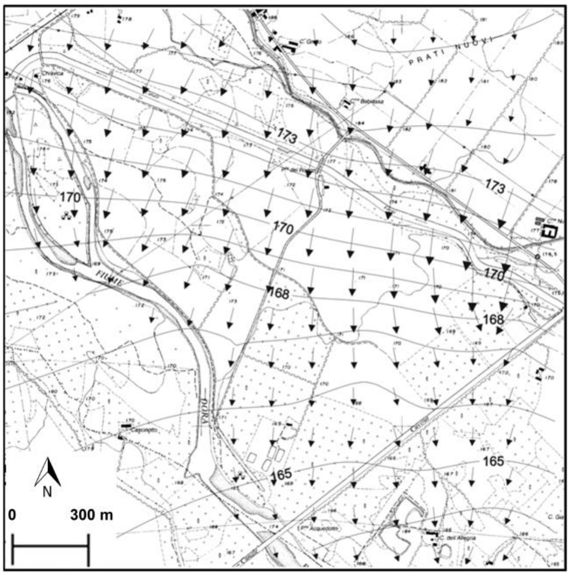

Figure 5 Dynamics of the groundwater at the end of the transient period from May to August 1999 on the shallow aquifer surface. The continuous lines represent the hydraulic head profile and the labels identify the hydraulic head value in meters above sea level. The arrows represent the flow lines.

The mean transport time between a source and a monitoring well in the area or between two possible target points, can be estimated, from the radiological risk point of view, after identification of the groundwater dynamics.
The quantitative hydrogeological analysis allows the transport time of a particle between two points to be estimated. By considering Darcy's law:

$$q = -K\frac{\Delta h}{\Delta s} = -Ki \qquad (1)$$

where: q represents Darcy's velocity [L/T]; K is the hydraulic conductivity [L/T]; Δh is the hydraulic head difference between two assessment points [L]; Δs represents the distance between the two points [L]; i is the hydraulic gradient [-]; it is possible to obtain the effective velocity, v_e:

$$v_e = \frac{q}{\theta_e} \qquad (2)$$

where θ_e is the medium effective porosity.
Thus, the transport time can be calculated as:

$$t = \frac{s}{v_e} \qquad (3)$$

This hydrogeological model is able to identify the variation in the hydraulic head in the groundwater in time. These hydraulic heads were elaborated by means of a Matlab code, which was set up to calculate the transport times over the entire period of simulation through equations (1-3). The range of the simulated transport time is related to the variation in Darcy's velocity, which depends on the variation in the hydraulic head in the groundwater during the transient. ARPA publishes water table levels periodically measured in specific monitoring wells. The difference between the topographical level and the water table level makes it possible to obtain the hydraulic head at specific points. In this way, the transport time was estimated at some specific times and for SPB-E5/6, A5-A9 and A9-RP4/7. The transport time was only estimated at specific times because the entire series of water table level data was not available. A comparison between the range of simulated transport times and the mean transport times estimated from the ARPA data are shown in Table 3. The transport times refer to the horizontal distance between two points. The transport times estimated from the ARPA data for the SPB-E5/6 and A5-A9 cases are within the range of the simulated transport times. The transport time, for the A9-RP4/7 case, is slightly less than the simulated transport time; this study has identified the necessity of knowing the entire series of the periodically measured water table levels so as to be able to develop an accurate estimation of the transport times, because a small change in the water table value can lead to a non-negligible change in the estimation of the transport time. However, the preliminary estimation and the comparison of the transport times, which are summarized in Table 3, have shown a good agreement between the simulation and the on-site measurements.
These results validate the correctness of the implemented hydrogeological system, and it is therefore possible to roughly estimate the mean transport times between a source and a monitoring well. After the analysis of the ARPA data, a source term was identified in correspondence to waste pond-719 in the Sogin area (ARPA, 2013b); the transport times between this source and the closest monitoring wells (SPB, E5/6, H2) were estimated. The H2 target was introduced to evaluate the transport time along a preferential migration direction and to compare it with the transport time between the source and wells outside the preferential direction. Another source term was hypothesized closer to the Avogadro area (S_{AV}) and the transport times were evaluated between this source and monitoring wells A9, S05, RP4/7.
The distance between source and target, Darcy's velocity, the effective velocity and the transport time were evaluated. The estimated data are shown in Table 4. A comparison between the transport times which were estimated for S_{pond}-SPB and S_{pond}-H2 shows that the second one is smaller than the first one, since it is along a preferential flow direction. Hypothesizing a path of the same length, the same result was obtained. The different times of S_{AV}-A9 and S_{AV}-S05, where the monitoring wells are almost at the same distance from the source term, are also evident. This difference is due to the hydraulic gradient.

Table 3 Comparison of the simulated and ARPA data estimated transport time for the SPB-E5/6, A5-A9 and A9-RP4/7 monitoring wells.

	Simulated transport time range [d]	Mean transport time estimated from the ARPA data [d]
SPB-E5/6	4.5 ÷ 5.6	5.5
A5-A9	9.5 ÷ 12.6	9.6
A9-RP4/7	3.6 ÷ 4.9	2.3

Table 4 Estimated transport times between the sources and monitoring wells.

	q [m/d]	ve [m/d]	t [d]	Distance between source and target [m]
S_{pond}-SPB	7.89	31.56	2.9	91.74
S_{pond}-E5/6	2.51	10.03	11.92	119.54
S_{pond} -H2	11.68	46.7	0.93	43.3
S_{AV} -A9	4.51	18.04	11.64	209.34
S_{AV} -S05	1.49	5.96	37.13	216.70
S_{AV} -RP4/7	4.47	17.88	14.85	264.86

7. Radionuclide Transport Model

The hydrogeological model was integrated with the transport model using the MT3DMS code (Chunmiao and Wang, 1999; Zheng and Wang, 1999), which makes it possible to analyze the contaminant concentrations, in space and time, in the groundwater and subsoil. MT3DMS simulates advection, dispersion, diffusion, source and sink terms and chemical reactions. In this work, several test cases were developed, focusing on the Saluggia nuclear site.

In routine conditions, the features of engineering barriers, used for the confinement of radioactive waste, change during their lifetimes. (ISAM, 2000) suggested different material degradation steps, which were evaluated if periodical and specific maintenance works and monitoring actions are performed during the life of barriers. If it is assumed that the radionuclide release from the engineering barriers is only due to the leaching process, and the release due to dissolution and diffusion is neglected, the leaching release occurs when the infiltration water removes radionuclides from the surface of the conditioned waste. The characteristics of concrete suggested by (Marseguerra et al., 2003) were here assumed as being similar to the characteristics of the engineering barriers of the analyzed nuclear facilities. The estimated leaching rates are indicated in Table 5 as a function of the material degradation, which is related to different timeframes of the lifetimes of the engineering barriers. These leaching rates were used to define some realistic source release rates of the engineering barriers for the developed test cases.

Table 5 Order of magnitude of the leaching rate for the different material degradation steps.

Temporal period [y]	State of material	Leaching rate [y^{-1}]
0 – 200	Intact	$10^{-6} \div 10^{-5}$
200 – 500	Partially degraded	$10^{-4} \div 10^{-3}$
> 500	Totally degraded	$10^{-2} \div 10^{-1}$

As already mentioned, the nuclear site is divided in two areas: the Sogin area and the Avogadro area. These two areas were considered separately (Fig.1), because the ARPA data show two possible different source terms in the sandy gravel layer. These sources were assumed to be located in the groundwater; the buffer effect of the unsaturated zone was not implemented into the model. This choice can be considered more cautionary and conservative, because the radionuclide migration could involve a smaller area and could also be slowed down due to the unsaturated zone, which is able to capture some of the radionuclides released from a nuclear facility.

The test cases were mostly performed considering a transient of 1000 days, in order to compare the simulated concentration under different conditions. Simulations of 1000 days were performed because the aim of the work was to estimate how to plan or improve an environmental monitoring network in order to manage and eventually mitigate the radiological impact of possible radionuclide migration in the short-term and mid-term.

Four test cases were conducted in the Sogin area, where a Cs-137 source was assumed near waste pond-719. The waste pond collects liquid effluents, thus it is a potential source term. Periodical measurements performed by ARPA did not detect any contamination near the pond, except in April 2013 (ARPA, 2013b). The contamination activity due to Cs-137, measured in April 2013 near the waste pond, was equal to 898±91 Bq/kg in the soil samples. This concentration was assumed as a source term for test cases 1, 2 and 3. Inspections, which were carried out by ISPRA and ARPA in 2012 on liquid and sediments stored inside the pond, measured concentrations that were consistent with the threshold values established for the plant, but they were not congruent with the criterion established by ARPA concerning a non-radiological impact during a discharge event (ARPA, 2013c). Therefore, the sediments inside the ponds have to be treated as radioactive waste. Four sediment samples were collected in the corners of the pond; their radiological characterization is shown in Table 6. A mean contamination value was assumed as the source in test case 4.

Table 6 Radiological characterization of the sediment samples [Bq/kg] in waste pond-719 (ARPA, 2013c).

	Am-241	Co-60	Cs-137	Sr-90	Pu-238	Pu-239/240
Sample 1	$3.41 \cdot 10^4 \pm 3.8 \cdot 10^3$	$<1.23 \cdot 10^3$	$6.38 \cdot 10^5 \pm 3.4 \cdot 10^4$			
Sample 2	$1.62 \cdot 10^5 \pm 9.3 \cdot 10^3$	$<5.27 \cdot 10^3$	$9.38 \cdot 10^5 \pm 4.8 \cdot 10^4$			
Sample 3	$5.20 \cdot 10^4 \pm 4.3 \cdot 10^3$	$<1.03 \cdot 10^3$	$2.06 \cdot 10^6 \pm 1.0 \cdot 10^5$	$2.43 \cdot 10^4 \pm 2.3 \cdot 10^3$	$8.38 \cdot 10^3 \pm 4.4 \cdot 10^2$	$3.64 \cdot 10^4 \pm 1.7 \cdot 10^3$
Sample 4	$5.72 \cdot 10^4 \pm 4.0 \cdot 10^3$	$<9.09 \cdot 10^2$	$1.68 \cdot 10^6 \pm 8.5 \cdot 10^4$			

The four test cases conducted for the Sogin area are:

- Test case 1 - the source term was imposed for 1 day. The transient was simulated over a period of 70 days.
- Test case 2 - the source term was imposed for the entire transient period of 70 days.
- Test case 3 - the source term was imposed for 70 days. The simulation was performed over a transient of 1000 days.
- Test case 4 - the source term was imposed for 365 days. The simulation was performed over a transient of 1000 days.

A second source was hypothesized in the Avogadro area. Sr-90 was detected in the 2009-2013 period by ARPA in monitoring wells A5, A9, S05 and RP4/7. The mean measured concentration of Sr-90 was assumed as the initial concentration for the developed test cases. No specific information on the radionuclide presence inside the facilities in the area was available, and it was therefore not possible to implement specific test cases which would have considered the real concentration of Sr-90 inside one or a part of these facilities. The investigated test cases for the Avogadro area are:

- Test case 5 - the source term was imposed for 70 days. The simulation was performed over a transient period of 1000 days. Two hypothetical sources were assumed: one of 0.2 Bq/l near monitoring well A9 and one of 0.1 Bq/l near monitoring well A5. The constant presence of Sr-90 measured by ARPA in A5 and A9 in the 2009-2013 period was assumed to be limited to the monitoring wells.
- Test case 6 - a hypothesis was made of a background contamination in the area, near monitoring well A9, of 0.2 Bq/l and, in the area near monitoring well A5, of 0.1 Bq/l; a source term of 0.1 Bq/l near A5 for 70 days was also assumed. This hypothesis was considered because a constant

concentration of Sr-90 has been measured in the monitoring wells in the 2009-2013 period. A source term near well A5 was also introduced in order to study a more critical situation. The simulation was performed over a transient period of 1000 days.

The transport model requires the introduction of specific parameters to describe the transport phenomena. The soil characteristics (bulk density, effective porosity, diffusion coefficient) and the interaction between radionuclide and solid matrix (distribution coefficient) are shown in Table 7. These data were obtained from a literature analysis (e.g. Varalda et al., 2006; Iezzi et al., 2009).

Table 7 Main transport parameters in the different layers representing the investigated area.

Layer	Soil Type	Bulk Density [kg/m^3]	Effective Porosity [-]	Diffusion Coefficient [m^2/y]	Sorption Parameter Kd [m^3/kg]
					Cs-137
1-2	Sandy gravel & Fine gravelly sand	1800	0.25	0.00221	0.03
3	Clayey silt	2500	0.08	0.0000315	0.08
					Sr-90
1-2	Sandy gravel & Fine gravelly sand	1800	0.25	0.00221	0.015
3	Clayey silt	2500	0.08	0.0000315	0.2

All the test cases were undertaken to analyze the concentration in space and time in the investigated area and in some specific targets (monitoring wells). In the Cs-137 test cases, monitoring wells SPB and E5/6 were considered targets, as a fictitious well, H2, which was added along a preferential flow direction. The H2 position allows the major solute transport to be evaluated along the direction of the river. In fact, if the radionuclides were to reach the river, they would travel to a great distance from the point of release and they could contaminate large areas far away from the site. As far as the Sr-90 cases are concerned, monitoring wells S05 and RP4/7 were imposed as targets. On the other hand, A9 and A5 were considered near the hypothetical source terms in test case 5, while A5 was considered near the source term in test case 6.

8. Results

8.1 Cs-137 transport test cases

The transport of Cs-137 was analyzed through four test cases. The simulated concentration of Cs-137 in water is shown in Figure 6 at the end of the transient for each simulation. The letter S in the figure represents the source term, which is located in the first layer and in correspondence to waste pond.

Test cases 1 (Fig.6a) and 2 (Fig.6b) show the behavior of the simulated Cs-137 concentration in the Sogin area. In these situations, the concentration appeared closer to the waste pond. The concentration has been shown to not reach particularly risky values for the population. Test cases 3 (Fig.6c) and 4 (Fig.6d) show more extended contamination areas than cases 1 and 2. These cases directly influence the Dora Baltea River, even though the radioactivity does not show significant radiological risks. In this situation, radionuclides could be transported along the river and reach distant areas from the site. The simulated Cs-137 concentration in monitoring wells E5/6 and SPB and in the fictitious target H2 was analyzed for test cases 3 and 4. The simulated concentrations did not appear relevant from the radiological point of view, and even though radionuclides could reach the river, dilution and solubility phenomena would reduce the concentration during the radionuclide migration. The sediments in the river bed could also capture some of the radionuclides, thus both the radioactivity and the distance travelled would be reduced.

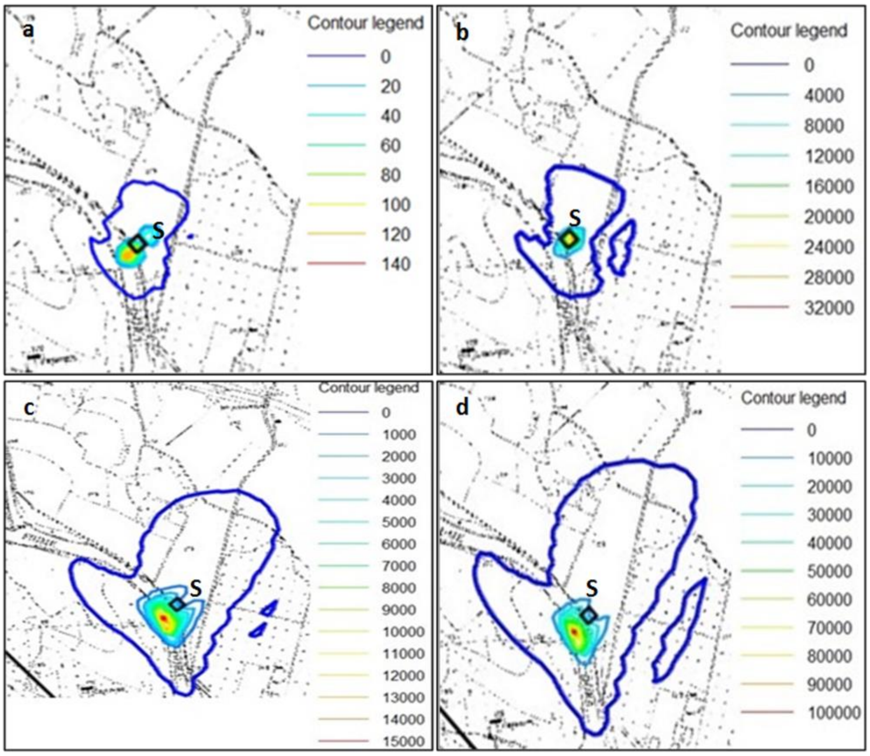

Figure 6 Cs-137 concentration in the groundwater [Bq/m^3] on the shallow aquifer surface with the source term in correspondence to waste pond 719, at the end of the simulated transients for test cases 1 (a), 2 (b), 3 (c) and 4 (d).

The ratios between the simulated concentration in the monitoring wells and the source term C_0 for test case 3 are shown for each layer in Figure 7. The highest concentration in each layer is detected in the H2 target, which is located near the source and along a preferential migration direction. In the first layer (Fig.7a), the concentration profile is similar to a Gaussian profile, with the peak located some days after the end of the release of the source. In the second layer (Fig.7b), the peak was reached on day 370 in the H2 target and on day 445 in the SPB monitoring well. The peak in monitoring well E5/6 will be reached around day 3500 after the start of the simulation, if the same boundary and initial conditions assumed for this test case are hypothesized. The second layer was assumed to be without any initial contamination: the different behavior from the first layer is due to the retardation phenomenon and to the downward infiltration. Instead, in the third layer (Fig.7c), the concentration profile can be seen to increase in the H2 well, but the values are lower than in the second layer where the peaks has already been reached. In monitoring wells SPB and E5/6, the concentration increases slightly compared to the H2 target, because H2 is on a preferential pathway. The peaks in this layer will be reached around day 4000 in H2, on day 5000 in SPB and on day 10000 in E5/6, under the hypothesis of the same initial and boundary conditions implemented in test case 3. Starting from a non-contaminated layer, the concentration peaks in the third layer will reach a lower concentration than in the second layer. The Cs-137 concentration in the third layer is very low, thus simulations which investigate the shallow aquifer are sufficient to establish the transport and the radionuclide migration in the groundwater. On-site investigations and the extension of the simulations to the deeper aquifer are not necessary. The detailed study of the radionuclide transport in the shallow aquifer is sufficient to safeguard the population and the environment from a radiological risk. Comparing the simulated ratio of the concentration and the ratio between the MDA and the source term ($1.64 \cdot 10^{-4}$), it can be seen that a periodical environmental monitoring should be performed, at least until the concentration decreases, because it is higher than the MDA, except in the third layer in monitoring well E5/6. In this way, it is possible to assess whether the radiological risk is negligible.

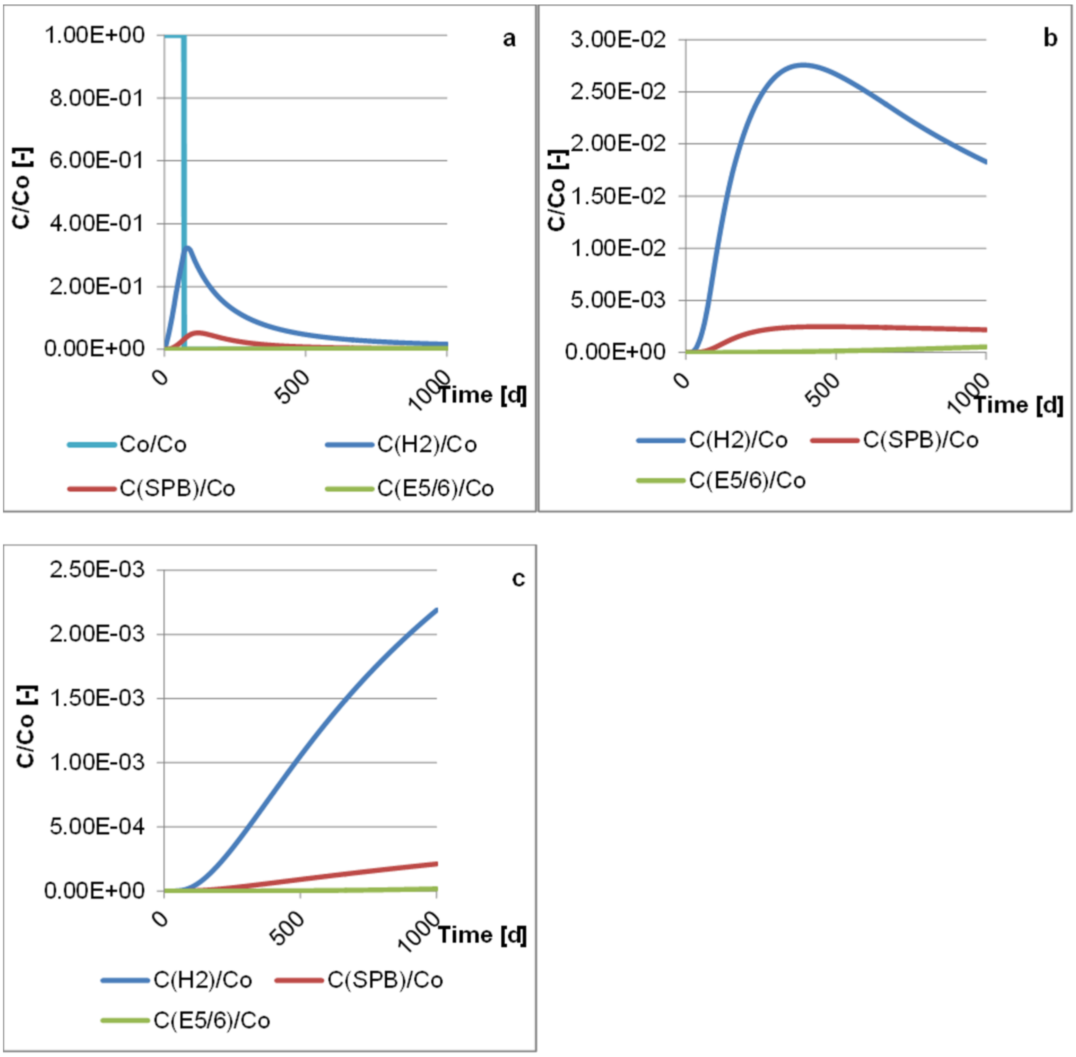

Figure 7 Ratio between the Cs-137 concentration and the source term for each monitoring well in the Sogin area in test case 3, for layers 1 (a), 2 (b) and 3 (c). The distance between the source term and monitoring wells H2, SPB and E5/6 is almost 43 m, 90 m, 120 m, respectively.

The ratios between the maximum simulated concentration in the monitoring wells and the source term are shown in Table 8. The ratios diminish in the soil by up to $10^{-4}\div10^{-5}$ orders of magnitude in the third layer. The ratios highlight the effect of the adsorption phenomenon on the solid matrix, which increases with the depth and the distance.

Table 8 Ratios between the maximum Cs-137 concentration in the monitoring wells and the source term for test case 3.

	$C(H2)/C_o$	$C(SPB)/C_o$	$C(E5/6)/C_o$
Layer 1	$7.45\cdot10^{-2}$	$5.25\cdot10^{-2}$	$3.58\cdot10^{-3}$
Layer 2	$2.76\cdot10^{-2}$	$2.49\cdot10^{-3}$	$5.46\cdot10^{-4}$
Layer 3	$2.19\cdot10^{-3}$	$2.11\cdot10^{-4}$	$1.71\cdot10^{-5}$

The ratios between the simulated concentration and the source term for each layer and in each monitoring well pertaining to test case 4 are shown in Figure 8. In the first layer (Fig.8a), the concentration in H2 reaches a peak, when the release of Cs-137 finishes (365 days), as does the concentration in SPB. Instead, in monitoring well E5/6, the concentration will reach a peak after about 1800 days, under the hypothesis of the same initial and boundary conditions assumed for this test case. In the second layer (Fig.8b), the maximum concentration is reached on day 568 in the H2 target and on day 625 in SPB. Assuming the same initial and boundary conditions implemented in test case 4, the peak in monitoring well E5/6 will be reached around day 3800. In the third layer (Fig.8c), the Cs-137 profile in H2 increases continuously and it is relevant; the concentration increment is instead slower in SPB and E5/6. The Cs-137 concentration can be seen to increase, because this layer was not initially contaminated, and the dynamic of the concentration is related to the migration time (infiltration in the deeper layer) and retardation processes. The concentration peaks will be reached around day 4000 in the H2 well, on day 5100 in SPB and around day 10000 in E5/6. These results are valid under the hypothesis of the same initial and boundary conditions imposed for the performed test case. Thus, it is necessary to continue the groundwater monitoring, at least till the concentration begins to decrease, in order to evaluate any possible radiological risk. The comparison between the simulated ratio of the concentration and the ratio between the MDA and the source term ($1.5 \cdot 10^{-5}$) shows that it is only in the third layer and in monitoring well E5/6 that the concentration is lower than the MDA. However, the contamination has no radiological impact on the population or on the environment, because of the very low concentration of Cs-137.

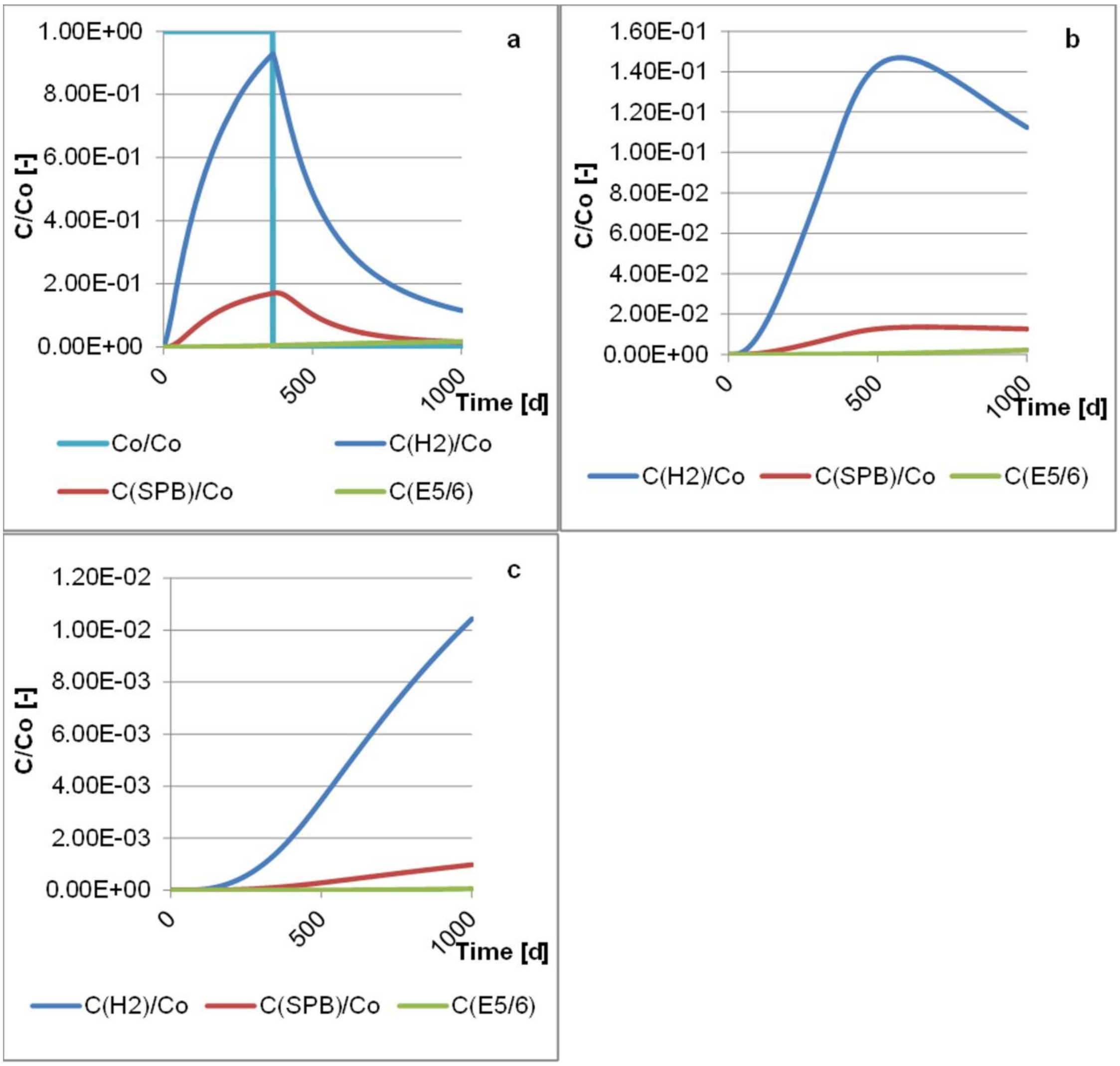

Figure 8 Ratio between the Cs-137 concentration and the source term for each monitoring well in the Sogin area in test case 4, for layers 1 (a), 2 (b) and 3 (c). The distance between the source term and monitoring wells H2, SPB and E5/6 is almost 43 m, 90 m, 120 m, respectively.

The ratios between the maximum simulated Cs-137 and the source term are shown in Table 9 for the different monitoring wells and for each layer. The orders of magnitude are higher than those of test case 3 (Tab.8), but the maximum simulated concentration does not represent a radiological risk for the population. The ratios highlight the relevance of the adsorption phenomenon on the solid matrix, which increases with the depth and the distance.

Table 9 Ratios between the maximum Cs-137 concentration in the monitoring wells and the source term for test case 4.

	$C(H2)/C_o$	$C(SPB)/C_o$	$C(E5/6)/C_o$
Layer 1	$9.29\cdot10^{-1}$	$1.72\cdot10^{-1}$	$1.75\cdot10^{-2}$
Layer 2	$1.47\cdot10^{-1}$	$1.36\cdot10^{-2}$	$2.16\cdot10^{-3}$
Layer 3	$1.04\cdot10^{-2}$	$9.80\cdot10^{-4}$	$5.79\cdot10^{-5}$

8.2 Sr-90 transport test cases

The Sr-90 transport was analyzed through two test cases. The simulated concentration of Sr-90 in water at the end of the simulation transient is shown in Figure 9 for test cases 5 (Fig.9a) and 6 (Fig.9b). The letter S in the figure represents the position of the hypothesized source terms. The hypothesis was deduced from the ARPA data, which show a continuous detection of Sr-90 in monitoring wells A5 and A9 in the 2009-2013 period.
The areas involved in the Sr-90 transport are not restricted to the Avogadro area, but they are more extended there. In test case 5, the maximum simulated concentration value is far from the source terms. In test case 6, the maximum concentration is near monitoring well A9, where the constant concentration background was imposed.

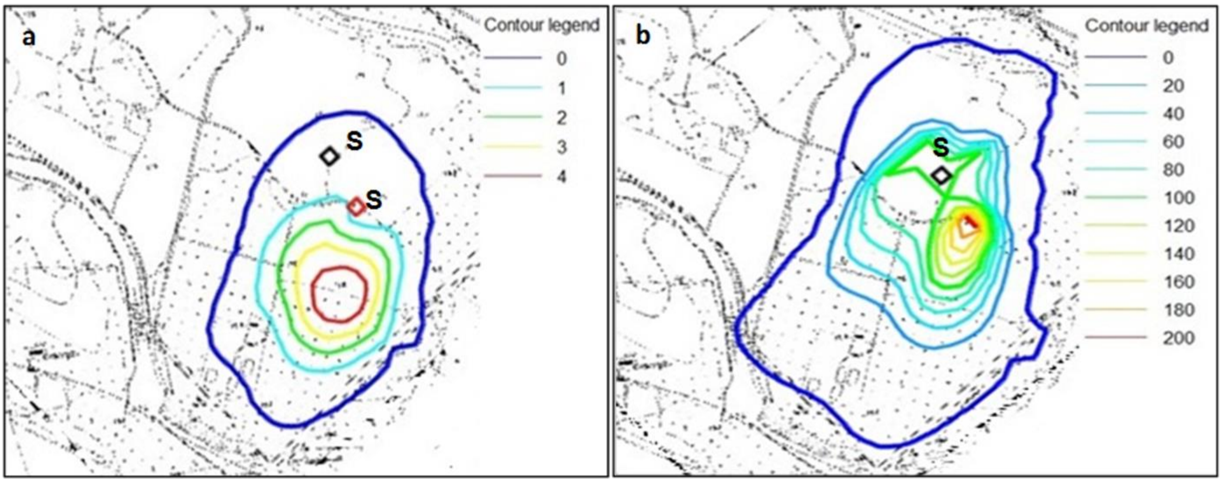

Figure 9 Sr-90 concentration in the groundwater [Bq/m^3] on the shallow aquifer surface at the end of the simulated transients (1000 days) for test cases 5 (a) and 6 (b).

In Figure 10, the Sr-90 concentration can be observed in each layer and in monitoring wells RP4/7 and S05 for test case 5. In the first layer (Fig.10a), the concentration shows the same behavior in both wells, with a peak in correspondence to the end of the source release. In the second layer (Fig.10b), the concentration profile is similar to that in the first layer. A growing profile, which is less steep in monitoring well S05 than in monitoring well RP4/7, is shown in the third layer (Fig.10c). The peaks will be reached between day 1500 and day 1700, if the same initial and boundary conditions imposed in test case 5 are hypothesized. This different kind of behavior is due to the infiltration and the retardation phenomena. In this case, the concentrations in the second and third layers are below the MDA, thus there is not contamination risk for the deeper aquifer. The analysis and the simulations that were focused on the shallow aquifer, which has been considered to be hydrologically separated from the deeper aquifer, are sufficient in order to carry out a correct evaluation of the radionuclide transport and to minimize the possible radiological impact on the population and the environment.

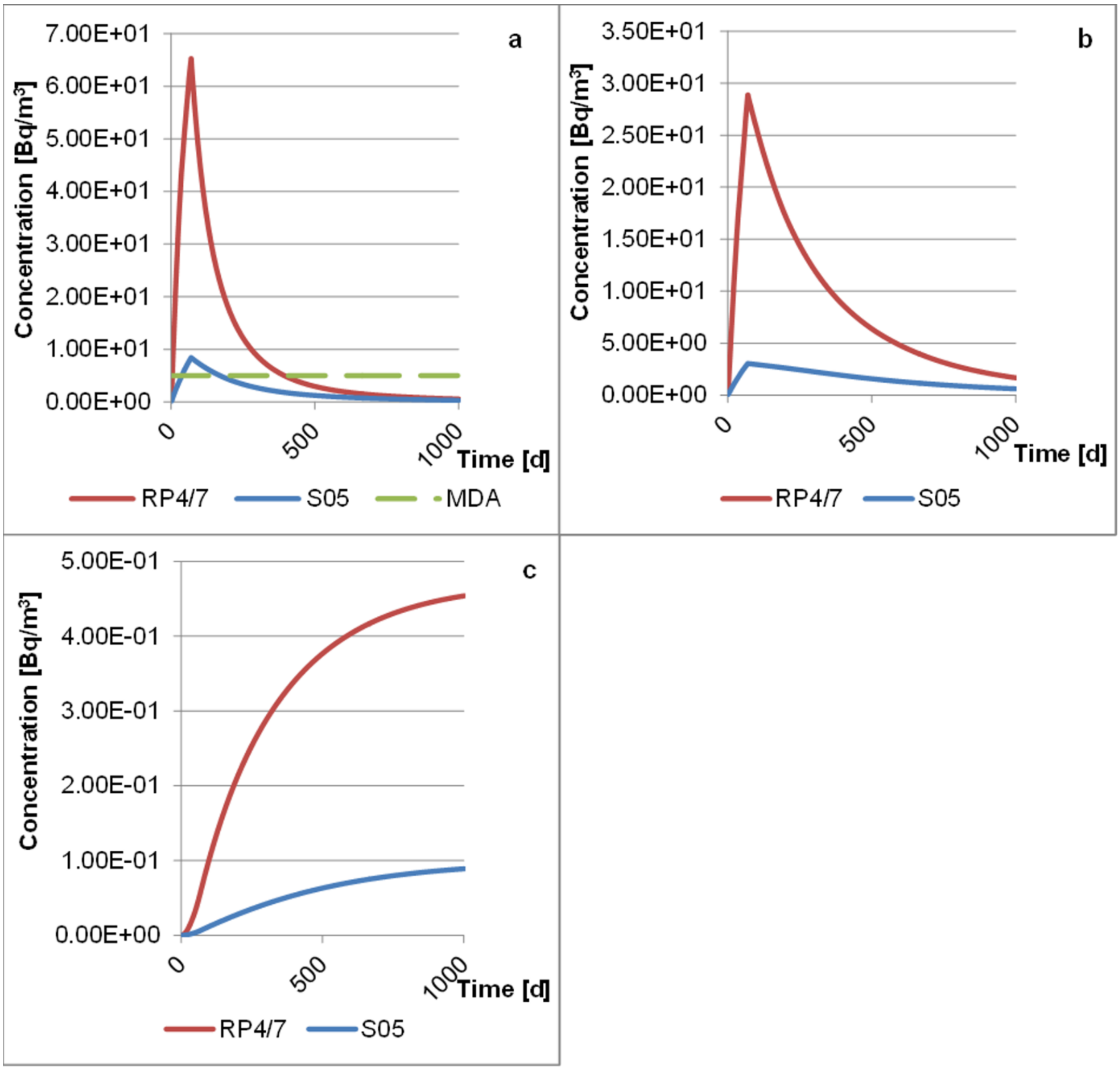

Figure 10 The Sr-90 concentration for each monitoring well in the Avogadro area in test case 5, for layers 1 (a), 2 (b) and 3 (c).

The ratios between the maximum simulated concentration in the monitoring wells and the source terms, for test case 5, are shown in Table 10. The orders of magnitude of the ratios decrease with the soil depth, reaching values of 10^{-3}÷10^{-4} in the third layer.

Table 10 Ratios between the Sr-90 concentration in the monitoring wells and the source terms for test case 5.

	C(S05)/C(A9)	C(S05)/C(A5)	C(RP4/7)/C(A9)	C(RP4/7)/C(A5)
Layer 1	$4.21\cdot10^{-2}$	$8.42\cdot10^{-2}$	$3.26\cdot10^{-1}$	$6.53\cdot10^{-1}$
Layer 2	$1.52\cdot10^{-2}$	$3.04\cdot10^{-2}$	$1.44\cdot10^{-1}$	$2.89\cdot10^{-1}$
Layer 3	$4.45\cdot10^{-4}$	$8.90\cdot10^{-4}$	$2.27\cdot10^{-3}$	$4.54\cdot10^{-3}$

Test case 6 was developed considering a background contamination of Sr-90 in the area, because ARPA detected an almost constant concentration of Sr-90 in monitoring wells A5 and A9 in the 2009-2013 period. The ratio between the simulated concentration and the source term for each layer is shown in Figure 11. As far as the first layer is concerned (Fig.11a), the concentration in A9 is constant, due to the background contamination. In monitoring well RP4/7, the concentration increases and reaches an almost constant concentration, because of the Sr-90 background. In well S05, the concentration grows, due to the background contamination and to the hypothesized source. Hypothesizing the same initial and boundary conditions as the implemented test case over a longer transient, a constant concentration will be reached in monitoring well S05 almost after 2000 days. In all three monitoring wells, the concentration ratio is higher than the ratio between the MDA and the initial source (0.05). The Sr-90 concentration in the second layer (Fig.11b) can be seen to increse in all the monitoring wells, due to the concentration background. In this case, the concentration is higher than the MDA value, thus the monitoring activity should be continued until a constant concentration is reached, which should take place on day 3500, if the same initial and boundary conditions imposed to develop this case are hypothesized over a longer period. A possible radiological risk should be evaluated analyzing the maximum concentration that has been reached. In the third layer (Fig.11c), the concentration can be seen to increase, but more slowly than in the second layer; the concentration is lower than the MDA. The maximum constant concentration will be reached over a longer period than the 1000 days of the simulation performed (almost 50000 days in each well), due to infiltration and retardation phenomena and due to the background contamination. If no inspections are undertaken to identify the origin of Sr-90 or to recover the area, the monitoring activity will detect the presence of Sr-90 over a longer period because of the interaction between Sr-90 and the solid matrix. If Sr-90 interacts with the solid matrix, there will be no radiological risks for the population, except in particular cases, for example, during drilling or excavation activities which could expose workers to Sr-90 contamination. If Sr-90 is in the solid matrix, variations in water table level or in chemical-physical conditions of the groundwater could induce its transport to the aquifer. In this case, Sr-90 could be detected in the groundwater in samples taken in the future.

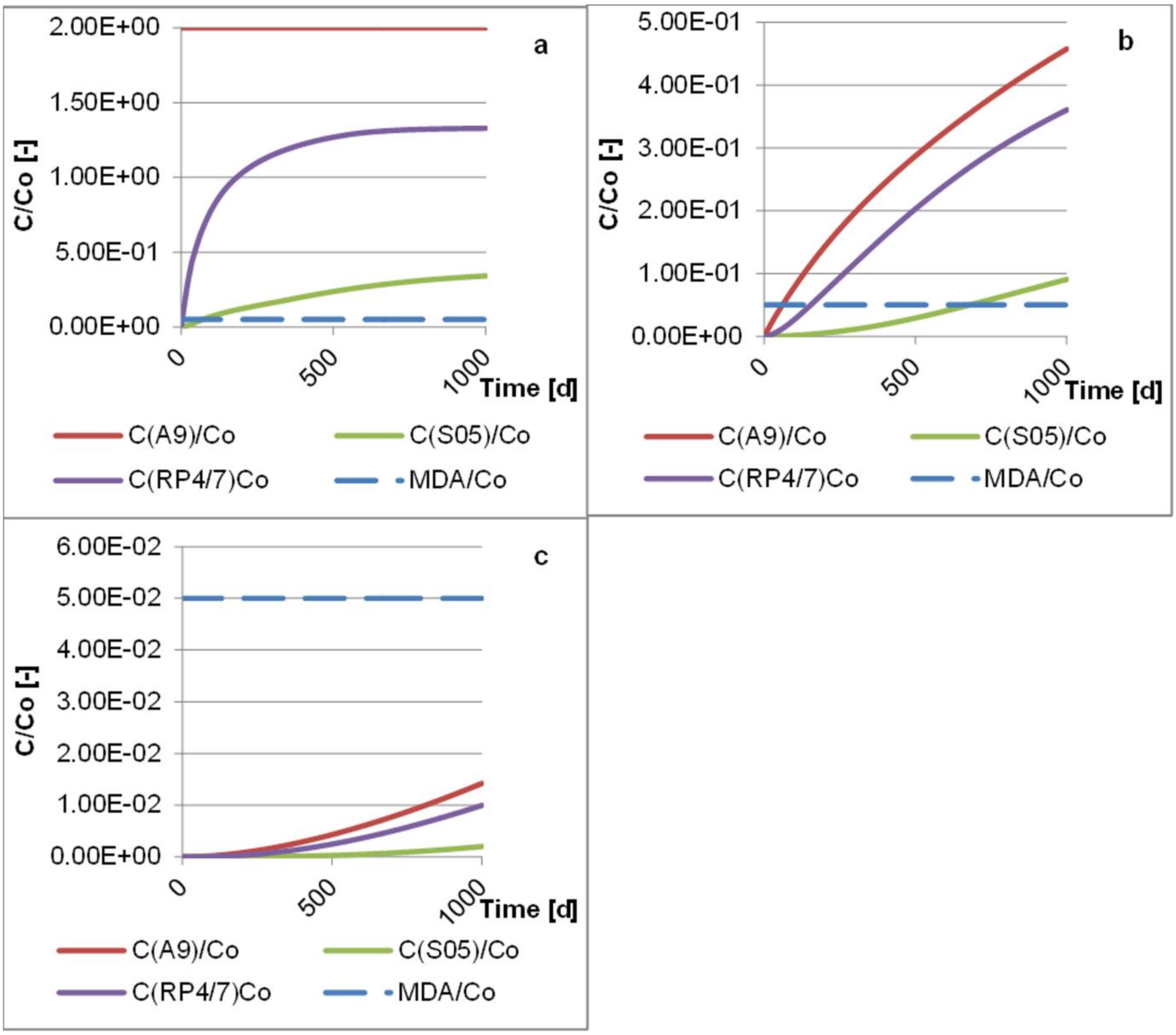

Figure 11 Ratio between the Sr-90 concentration and the source term for each monitoring well in the Avogadro area in test case 6, for layers 1 (a), 2 (b) and 3 (c). The distance between the source term and monitoring wells A9, S05 and RP4/7 is almost 210 m, 217 m, 265 m, respectively.

The ratios between the maximum simulated concentration and the source term for test case 6 are shown in Table 11. The orders of magnitude are higher than in test case 5 (Tab.10), because of the Sr-90 background. Again in this case, the ratios highlight the effect of the adsorption phenomenon on the solid matrix, which increases with the depth.

Table 11 Ratios between the maximum Sr-90 concentration in the monitoring wells and the source term for test case 6.

	$C(A9)/C_o$	$C(S05)/C_o$	$C(RP4/7)/C_o$
Layer 1	$2.00 \cdot 10^{0}$	$3.41 \cdot 10^{-1}$	$1.33 \cdot 10^{0}$
Layer 2	$4.58 \cdot 10^{-1}$	$9.06 \cdot 10^{-2}$	$3.61 \cdot 10^{-1}$
Layer 3	$1.42 \cdot 10^{-2}$	$2.01 \cdot 10^{-3}$	$9.97 \cdot 10^{-3}$

9. Conclusions

Radioactive waste management is a strategic activity that is adopted in the nuclear field for the safeguarding of the operating staff, of the population and of the environment. Safety studies on the management of current nuclear activities and on the design of a new nuclear facility should also involve the assessment of the environmental impact. In the context of these activities, the migration of radionuclides in the geosphere must be evaluated. This work has in particular focused on the transport of radionuclides through groundwater. The transport of radionuclides in groundwater has been investigated, focusing on source terms and applying a methodological approach. The assessment has involved an analysis of the Italian nuclear site in Saluggia (VC), coupling on-site measurements, carried out by national authorities (ARPA, ISPRA) in monitoring wells, and the use of modeling codes. This can be considered a propaedeutic activity to plan or improve environmental monitoring networks or to plan adequate mitigation or other intervention following accidental situations. This work has analyzed a nuclear site which has an existing environmental monitoring network.

First, information on the geological and hydrogeological framework, the radioactive waste located in the nuclear facilities and the environmental monitoring network were collected and elaborated for the purpose of the work. Hydrogeological data were implemented in a model which was calibrated to represent the groundwater dynamics under a common environmental condition. A Matlab code was set up to elaborate the data obtained with the hydrogeological model and to extract the transport times between the sources and monitoring wells (targets). The possibility of elaborating transport times can lead to an increase in the knowledge on the migration dynamics of radionuclides in the environment pertaining to the case of leakage from a nuclear facility. Then, the transport of Cs-137 and Sr-90 was examined through different test cases, starting from contamination measurements that have been carried out by ARPA and ISPRA in monitoring wells.

The simulated concentration at the interface between the shallow aquifer and the deeper aquifer has not resulted to be relevant, from the radiological point of view, in any of the test cases. The simulation results have confirmed that the existing monitoring network has been set up correctly.

The performed test cases have also shown that the areas involved in the migration of radionuclides are restricted to the nuclear site.

The detailed analysis of a nuclear site, from the hydrogeological and the radiological point of view, is of fundamental importance to minimize the radiological risk for the population and the environment. Thus, with reference to the analyzed test cases, it can be stated that the conducted in-depth and detailed study of the shallow aquifer has also proved to be sufficient to protect the deeper aquifer from the radiological contamination point of view.

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