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Transport dynamic of strontium in groundwater: Safety Assessment study

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Abstract

One of the activities of the Safety Assessment is the evaluation of the impact of a nuclear facility on the environment. The radionuclide transport into groundwater is subjected to different phenomena (e.g. groundwater dynamic, surface stream dynamic, the interaction between surface water and groundwater, radionuclide interaction with environmental matrix, etc.) that influence the risk of contamination of water. The investigation of the source term is fundamental to understand its impact on radionuclide transport. In this paper, in situ surveys and modelling were coupled to investigate the dynamic of strontium in an Italian nuclear site. On-site measurements have identified low quantities of strontium in monitoring wells, and through the modelling, the possible migration pathway of this radionuclide was identified. For a primary safety evaluation purpose, a parametric detailed analysis was carried out to identify which hydrogeological parameters and which artificial structure present in the area could influence the dynamic of strontium in the investigated site. In particular, the effect of the Cavour artificial channel on the strontium migration and dilution was demonstrated. The coupling of monitoring activities, periodically performed in the area, and the modelling activities, focused on the detailed relationships between the Cavour artificial channel and the underground water flow, contributes to better evaluate the possible radiological risk for population and environment and to support future safety studies.

1. Introduction

In the context of nuclear decommissioning and radioactive waste management, the safety is also guaranteed by periodic assessment about the possible release of radionuclide that can represent a possible radiological risk for the public and environment. This assessment led to continuous in-depth investigations and evaluations of the nuclear site, of the area outside the nuclear plant and of the events that may influence the dynamic of possible radioactive contaminants migration. This approach is one of the elements allowing to satisfy the national legal framework and the international standards about nuclear safety.

The radionuclide transport into groundwater is one of the fundamental activities that can contribute assessing the safety of nuclear activities, especially if it is possible to integrate on-site measurement and modelling activities. For example, Lamego Simoes Filho et al. (2013) modeled tritium dynamic in surface waters in order to evaluate the radiological impact of its potential release from nuclear power plants into the environment. Jakimaviciute-Maseliene and Cidzikiene (2015) studied the modeling of tritium in underground water at the new nuclear power plants (NPPs) site in Lithuania, by means of the FEFLOW code (MIKE, 2016). Yu et al. (2016), assuming transport of radionuclides as one of the principal aspects to be evaluated in a geological and near surface radioactive waste repository from the safety viewpoint, investigated the migration and sorption of Sr in clay-sand mixture by batch experiment, column experiments and numerical simulation. Palágyi et al. (2017) focused on the determination of transport parameters of radionuclides in systems made of various solid crushed granitic and soil materials, sands and disturbed or undisturbed soil cores with synthetic or real groundwater, containing effective tracers. In particular, they investigated the behavior of strontium and cesium in a simplified modelling in dynamic column technique. Pohjola et al. (2019) carried out a probabilistic assessment of the impact of bottom sediment on doses to humans from a groundwater-mediated radionuclide release in a farm-lake scenario. In particular, they focused on the investigation of the most contributing parameters, related to the sediment layers, to the overall radiological dose for humans.

Authors performed also different studies investigating the dynamic of groundwater and related migration of radionuclides in a nuclear site. Testoni et al. (2015) studied tracer techniques coupling modelling and environmental monitoring activities, as an effective tool to characterize and to foresee the radionuclide dynamic in the environment. Testoni et al. (2015) analyzed the source term as the key point in the modeling of the transport of radionuclides in groundwater and soil applying a methodological approach, which focuses on the dynamic of the source term in space and time. Testoni et al. (2016) investigated an Italian nuclear site undergoing decommissioning, focusing on the hydrogeological aspect as a propaedeutic study for the transport of radionuclides. A step-by-step approach was developed and tested to investigate the relationship between groundwater and surface water, coupling on-site measurement and modelling activities.

In this work, authors have carried out a modelling activity to study the strontium dynamic in groundwater outside a nuclear site, in order to evaluate not only the influence of different hydrogeological parameters on the migration of contaminants in groundwater, but also of artificial surface channels. In particular, the effect of the Cavour artificial channel on the strontium migration and dilution downstream the channel itself was demonstrated, correlating the data about on-site measurements and the development of a transport model. In the context of primary safety studies, the results of the work underline the necessity to focus on all the different aspects potentially influencing the dynamic of radioactive contaminants in environment. The same results may be useful to improve the detailed understanding of mass transport in groundwater and, as a consequence, the possible future evaluation of radiological risk for population and environment, introducing new elements of reasoning about the future safety analysis of the site.

First, literature analysis of hydrogeological data and strontium concentration data detected by the national environmental authority was carried out. Then, a modelling activity was developed. As a first step, the model was calibrated from the hydrogeological and mass transport point of view. As a second step, a parametric analysis was performed to assess the strontium dynamic under specific boundary conditions, first of all, considering the influence of surface stream features.

2. Site background

The investigated site is near Saluggia (VC), in the North of Italy. This site hosts several nuclear facilities: the Avogadro temporary repository for spent fuel; the Eurex plant, which is a nuclear fuel reprocessing plant under decommissioning, and a temporary radioactive waste repository. The investigated area, involving the nuclear facilities area and the neighboring area, is shown in Figure 1. The boundaries of the area are defined by the following reference points: the Dora Baltea river to the west and south-west sides, the location of the monitoring wells SP01 to the south side, the Farini channel to the north-east side.

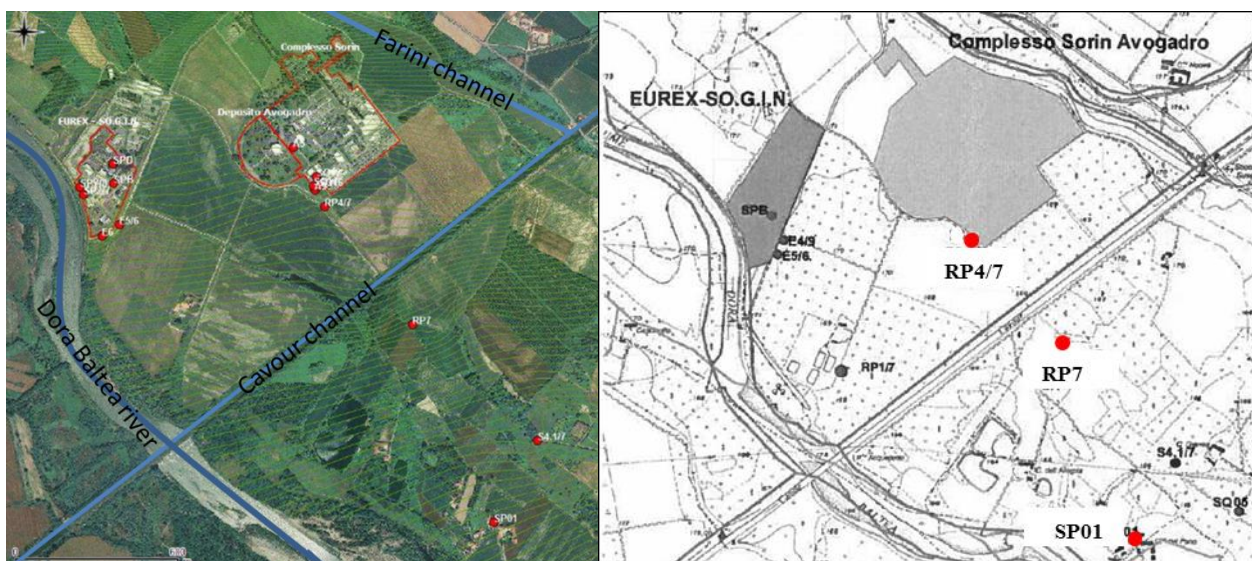


Figure 1 a) Investigated site area (ARPA Piemonte, 2013), b) location of the considered monitoring wells.

Concerning the geological aspects, the site is located in the flood plain of the Dora Baltea River. This area is characterized by alluvial sediments that overlap fluvioglacial deposits, up to a depth of about 45 m. The stratigraphy below the fluvioglacial deposits is featured by alternating sandy and silty-clayey layers of the Villafranchian age (Testoni et al., 2015). In Table 1, the representative stratigraphy of the shallow aquifer, as well as the main characteristic parameters of each layer, are reported.

For what concern the hydrogeology, the hydrogeological system is constituted by two 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 aquifer. The shallow aquifer is characterized by regional groundwater recharge, mainly due to the water coming from the Ivrea morainic amphitheater and from rice paddies during their flooding (Testoni et al., 2015). The shallow aquifer is separated from the deeper aquifer by an almost 0.5 m thick silty layer.

The Dora Baltea river and the local artificial channels deeply influence the local recharge. The main characteristics of the Dora Baltea river and of the artificial channels, Farini and Cavour channels, are reported in

Table 2. Adorni-Braccesi et al. (2001) have demonstrated, using a comparison between physical methods and geochemical approaches, that the local groundwater recharge is due to surface water, such as the Dora Baltea river and the artificial channels; instead, the precipitations are quite negligible. The free water table has a mean depth of 3.5 m from the ground surface. The flow direction is north-south, and it is characterized by local variations due to interaction with surface waters. This work is focused on the transport dynamic of strontium in the shallow aquifer.

Table 1 Representative stratigraphy of the area and main characteristic parameters.

Layer	Depth [m above sea level]	Hydraulic conductivity [m/s]	Porosity [-]	Bulk density [kg/m ³]
Gravel	173-139	0.0192	0.25	1800
Sand	139-127	0.0066	0.25	1800
Clay	127-126.5	1.50E-08	0.08	2500

Table 2 Mean characteristics of the Dora Baltea river, Farini channel, and Cavour channel.

Stream	Flow rate [m ³ /s]	Streambed sediments hydraulic conductivity [m/s]	Streambed thickness [m]	Upstream streambed elevation [m]	Downstream streambed elevation [m]
Dora Baltea River	300	0.0192	1	173.10	172.00
Farini channel	8	0.0096	1	172.00	170.00
Cavour channel	14.67	0.0064	6	170.30	170.20

3. On-site measurement and previous works

The investigated area is subjected to periodical monitoring by ARPA Piemonte, the Italian Regional Agency for the Protection of the Environment. Concerning the groundwater monitoring, ARPA Piemonte reported the level of the groundwater in monitoring wells until December 2013. The groundwater level data in the monitoring wells RP4/7 and SP01 were considered (Figure 1). Others data and considerations about the hydrogeological framework of the area were derived by the model carried out by Testoni et. al (2016). In that work, a calibration model was developed identifying the main parameters that influence the groundwater dynamic, such as streams sections profile of the Dora Baltea River. Concerning the radiological monitoring, ARPA Piemonte periodically collects samples on the basis of the matrix to analyze: groundwater is monitored every 4 months, as well as the surface water; instead, water for human purposes is controlled each month. Soil and vegetables are monitored every six months, instead, the air monitoring is performed in continuous. Annually reports are published on the ARPA Piemonte web page

(<http://www.arpa.piemonte.it/approfondimenti/temi-ambientali/radioattivita/siti-nucleari/saluggia>) with the data and information about the monitoring activities.

For what concerns radioactive contaminants in the groundwater of the investigated site, three monitoring wells were considered: RP4/7, RP7, and SP01. In all these wells, Sr-90 concentration was always detected in quantity below the radiological limits: 0.17 Bq/l as no-radiological relevance defined by ARPA Piemonte, and 0.4 Bq/l as radiological relevance defined by Italian law (D. Lgs. 28/2016, 2016). The behavior of Sr-90 concentration measured from April 2011 to December 2016 is reported in Figure 2. The available data were averagely measured every 3 months.

From the qualitative point of view, it is possible to state the following considerations. First, in the monitoring well RP4/7, the Sr-90 concentration shows a slow and steady decrease in concentration that stabilizes during 2014 and 2015; minimum concentrations occur in late autumn and late winter, probably related to the concomitant effects of the recharge due to high flow of the Dora Baltea river and of regional groundwater recharge. Second, in the monitoring well RP7, the concentration behavior is similar but mitigated respect to that one described for RP4/7. This is probably due to the leakage of water by the Cavour channel streambed to the groundwater in the spring-summer period, that dilutes the concentration of the contaminant. Thus, if the mean concentration in the two monitoring wells is compared, a value of 0.06 Bq/l characterizes the mean concentration in RP4/7 and a value of 0.015 Bq/l represents the mean concentration in RP7. This corresponds to a dilution factor of 4, that supports the hypothesis that the Cavour channel can influence the local dilution of concentration in RP7. Third, hypothesizing the Sr-90 concentration measured in the monitoring well RP4/7 as source term, analyzing the groundwater flow lines it is possible to suppose the propagation of Sr-90 from the monitoring well RP4/7 to RP7 and to SP01.

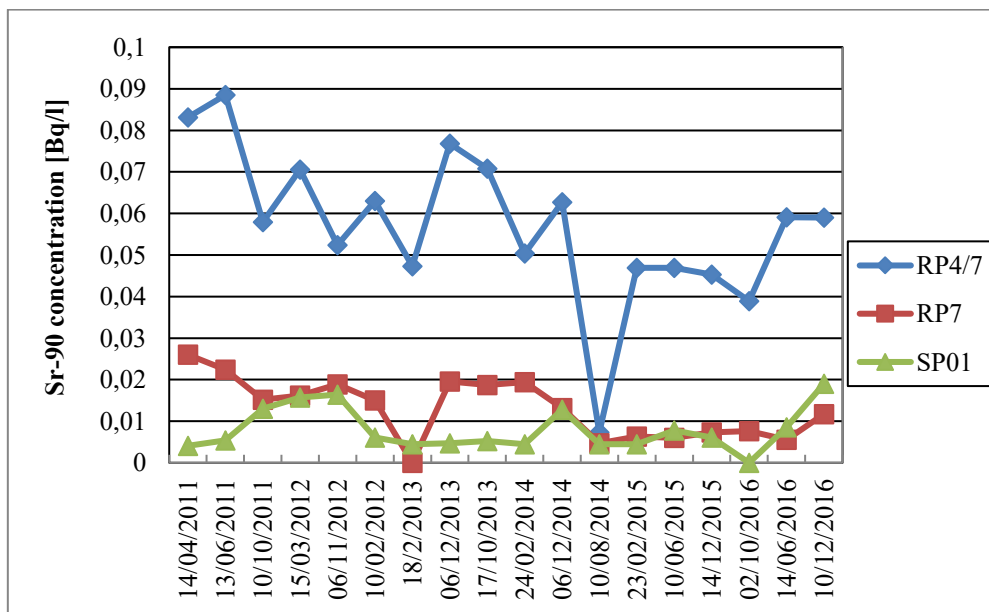


Figure 2 Measured Sr-90 concentration in the monitoring wells RP4/7, RP7, and SP01.

4. Modelling

The modelling activity involves two steps, regarding the groundwater dynamic and the radionuclide transport. The analyzed site was simplified in a conceptual model that was represented mathematically by means of the Modflow code (Harbaugh, 2015), for what concern the groundwater dynamic, and by means of the MT3DMS tool (Zheng and Wang, 1999), for what concern the radionuclide transport. Modflow and MT3DMS codes are coupled by means of the Graphical User Interface (GUI) ModelMuse (Winston R.B., 2009). ModelMuse is constituted of a main module and additional modules, called packages. These packages make it possible to represent in detail the boundary conditions; wells, river, recharge and discharge, contaminant transport, geochemical reactions, etc. may be introduced. Each of these packages are represented by specific equations that characterize the module. It is also possible to manage these equations introducing formulas that vary in space, and time. For example, El-Zehairy et al. (2018) studied the interactions of artificial lakes with groundwater applying an integrated MODFLOW solution. Morway et al. (2013) developed different scenarios modeling variably saturated subsurface solute transport through MODFLOW-UZF and MT3DMS. Instead, Zhang (2013) applied MT3DMS software tool and geographic information system in order to evaluate the groundwater contamination in the Sherwood Sandstone Aquifer (United Kingdom).

5. Results and discussion

The modeling activity is divided in two steps: first the groundwater model representing the pressure heads and the strontium transport is calibrated by means of RP4/7, RP7 and SP01 monitoring wells; then a parametric analysis on the strontium transport is performed, investigating the main influencing parameters (e.g. flow rate in Cavour channel).

5.1 Calibration of the model

The calibration of the model was preliminarily carried out implementing the hydrogeological aspects; the calibration started from the previous works proposed by Testoni et al. (2015 and 2016), extending the investigated site to the area south of the Cavour channel (Figure 1). Thus, the strontium transport was introduced.

First, the groundwater dynamic model was calibrated involving two main features: the water level in the river and the artificial channels, and the groundwater hydraulic head. These two aspects were studied simultaneously, as they represent the interaction between the surface water and the groundwater (Testoni et al., 2016).

Concerning the initial condition implemented, piezometric head data were introduced. As far as the boundary condition is concerned, particular attention was dedicated to the representation of the river and the artificial channels, which were characterized with different sections and with different stream hydraulic heads along the longitudinal axis. Different scenarios were featured setting the following main parameters: mean flow, stream hydraulic head, streams section, hydraulic conductivity of the streambed sediment, streambed elevation, streambed thickness, channel roughness. These data were provided by the Italian National Agency for New Technologies, Energy and Sustainable Economic Development (ENEA), and they are reported in (Testoni et al., 2016). The Stream-Flow Routing Package (Prudic et al., 2004) was applied to evaluate the influence of the Cavour channel, the largest one, because of its permeable containment structure which produces huge water losses to the groundwater. At the first stage, the flow in the stream was hypothesized constant along all the year; subsequently, a seasonal variation of the flow in the channel was imposed. A maximum flow equal to 110 m³/s was considered every year from May to September, while a null flow was considered every year from October to April, as revealed by ENEA data (personal communication).

Secondly, the strontium transport was introduced in the model to compare measured data with the simulated data, and to evidence the effect of the surface stream on the mass transport of groundwater. The source term was imposed as a step-source in the monitoring well RP4/7, introducing the measured data of ARPA (<http://www.arpa.piemonte.it/appfondimenti/temi-ambientali/radioattivita/siti-nucleari/saluggia>). A

background concentration was assumed considering the measured data in the neighboring monitoring wells. A first model without setting a background concentration underestimated the strontium concentration in the area. For this reason, an in-depth investigation on the historical collected data in the monitoring wells of the area was performed and a background concentration of the site was identified and implemented. With this approach, the comparison between measured concentrations and simulated concentrations in the two monitoring wells, RP4/7 and SP01, was carried out. The result is reported in Figure 3. From a qualitative point of view, the Sr-90 concentration simulated in both monitoring wells shows a good agreement with the measured data. It is possible to notice a slightly minor correlation between measured and simulated data in the first 900 days in the RP4/7 monitoring well; this is probably due to the high variability of concentration that would require more measured data to improve the calibration model. The comparison between measured and simulated data in RP7 is not reported; the lack of data about the groundwater level in that monitoring well influences a lot the strontium transport behavior.

From a quantitative point of view, the available data provide a mean relative error of the order of 25% for the RP4/7 monitoring well, and of 35% for the SP01 monitoring wells. These values are related to the available data, that were collected each 2-3 months. The possibility to increase the number of data collected will improve the model calibration. However, the qualitative goodness of the obtained Sr-90 concentration profiles makes it possible to discuss the importance of the source term and to perform a parametric analysis too.

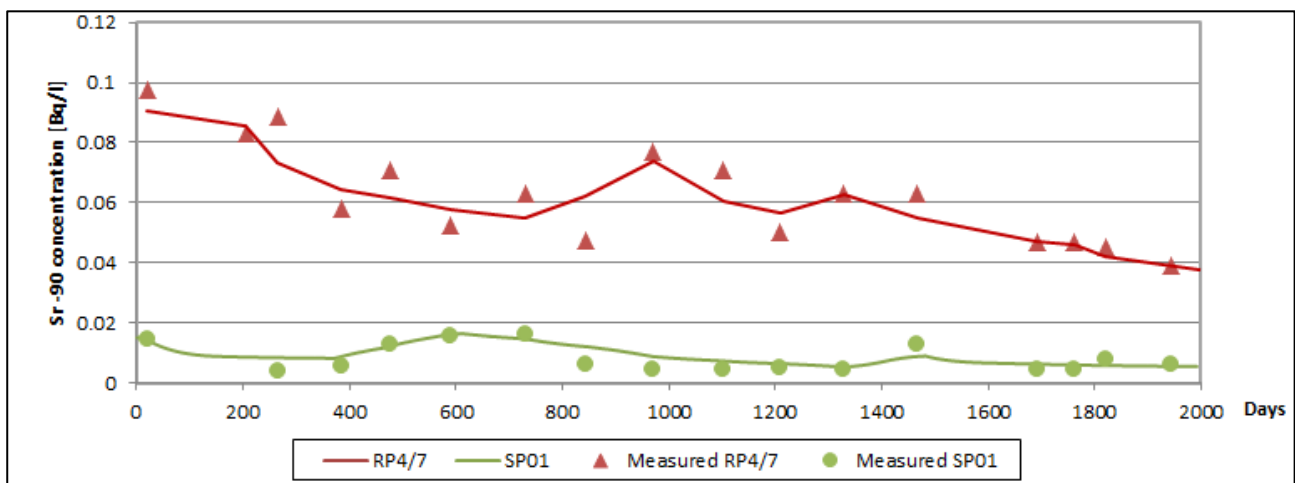


Figure 3 Comparison between measured and simulated Sr-90 concentration in the monitoring wells RP4/7 and SP01.

5.2 Parametric analysis

Starting from the calibrated model, a parametric analysis was carried out to understand the strontium transport in the groundwater in the investigated site and in particular in the area south of the Cavour channel. The effect of the hydraulic conductivity of the streambed, of the flow rate and of the high permeability of the Cavour channel were investigated. The channel water flow may influence the dynamic of contaminants in groundwater, due to the water leaks from the streambed; results of this evidence could introduce new elements of reasoning about the safety analysis of the site.

Concerning the hydraulic conductivity, different conditions were considered from 0.00192 m/s to 0.192 m/s, using the hydraulic conductivity of the calibrated model as a mean value. This range of hydraulic conductivity data corresponds to a sand/gravel streambed configuration, representing the Cavour channel streambed. Simulations evidence that this parameter does not influence the concentration behavior. This was expected for the SP01 monitoring well due to the distance from the Cavour channel, but the same behavior appears also for RP7 monitoring well. This means that the variation of hydraulic conductivity is mitigated by

the elevation of Cavour channel respect to the ground. For this reason, no figures concerning this analysis are reported.

The second investigated parameter is the flow rate in the Cavour channel. In this survey, extreme conditions of dryness ($1.47 \text{ m}^3/\text{s}$) and flooding ($146.67 \text{ m}^3/\text{s}$) were considered, taking into account the very slight effect of the streambed hydraulic conductivity. In this range of flow rates, different simulations were performed. In Figure 4 and Figure 5, some cases with a different flow rate in Cavour channel are shown for RP7 and SP01 monitoring wells, respectively. As expected, a higher flow rate in the Cavour channel represents a direct vertical flux of water from the streambed toward the groundwater. As a consequence, it is possible to notice a slight dilution of concentration in the nearest investigated monitoring well increasing the flow rate. In particular, with a low flow rate up to the initial condition imposed of $14.67 \text{ m}^3/\text{s}$, no influence on strontium transport is evident. Moving to flooding condition, a reduction of Sr-90 concentration occurs. These results evidence the link between the strontium dynamic in the groundwater and the Cavour channel dynamic. Instead, in the monitoring well SP01, no direct influence of the different flow rates in the Cavour channel appears.

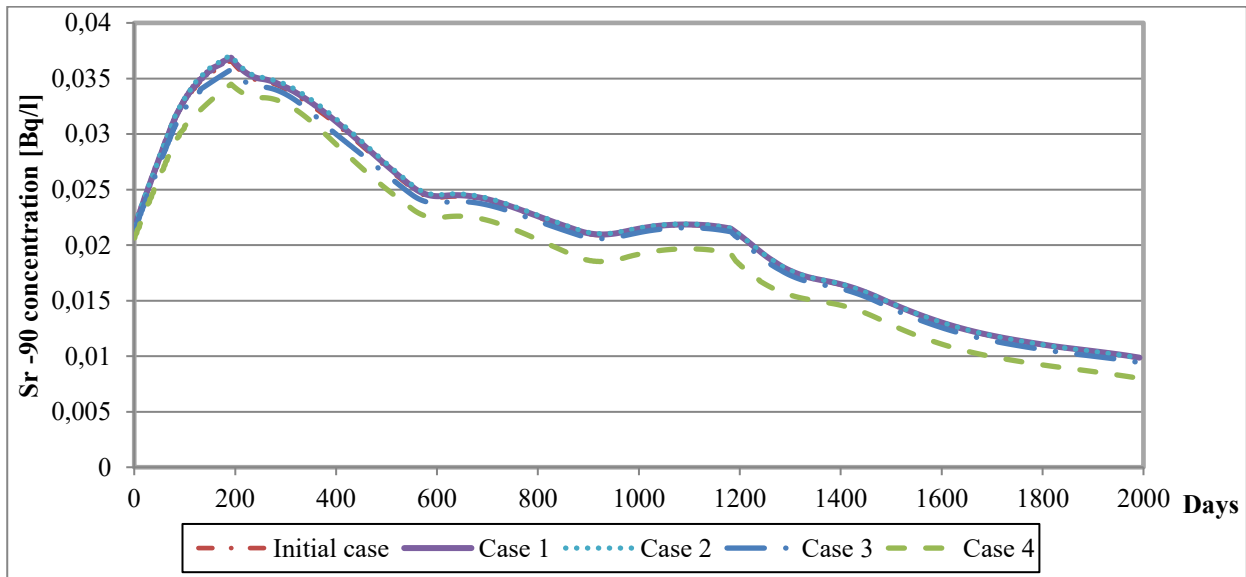


Figure 4 Sr-90 concentration in RP7 monitoring well for flow rates in Cavour channel equal to $14.67 \text{ m}^3/\text{s}$ (initial case), $7.33 \text{ m}^3/\text{s}$ (case 1), $1.47 \text{ m}^3/\text{s}$ (case 2), $44 \text{ m}^3/\text{s}$ (case 3), $146.67 \text{ m}^3/\text{s}$ (case 4).



Figure 5 Sr-90 concentration in SP01 monitoring well for flow rates in Cavour channel equal to $14.67 \text{ m}^3/\text{s}$ (initial case), $7.33 \text{ m}^3/\text{s}$ (case 1), $1.47 \text{ m}^3/\text{s}$ (case 2), $44 \text{ m}^3/\text{s}$ (case 3), $146.67 \text{ m}^3/\text{s}$ (case 4).

The third investigated aspect is the high permeability of the Cavour channel, that influences the leakage of water from the surface to the groundwater system, indeed the channel is raised of almost 5-6 m above the ground surface and the streambed is featured by highly permeable sandy and gravelly sediments. This condition was set up in the model. Several cases were carried out imposing, at the beginning, constant flow leakage for the entire investigated period in order to understand the capacity of the model to implement this condition. Then, this condition was improved with a maximum flow leakage, in the period from May to September, and a minor flow leakage, in the period from October to April, corresponding to the maximum and the minimum flow rate in the Cavour channel. The flow leakage was hypothesized of the order of magnitude of a meter per day, as estimated by means of Darcy's law. In Figure 6, the Sr-90 concentration in the monitoring well RP7 under different flow leakage conditions (min 10 m³/s; max 30 m³/s) of the Cavour channel is reported.

No measured data are available concerning the water leakages, however, a comparison with the measured concentration in RP7 was performed to understand the goodness of this analysis. The case 6, that corresponds to a flow leakage variable along the year, reflects the real monitored Sr-90 concentration profile from both qualitative and quantitative point of view. As in the other two parametric studies, in monitoring well SP01, no direct influence of the different flow leakage from the Cavour channel occurs.

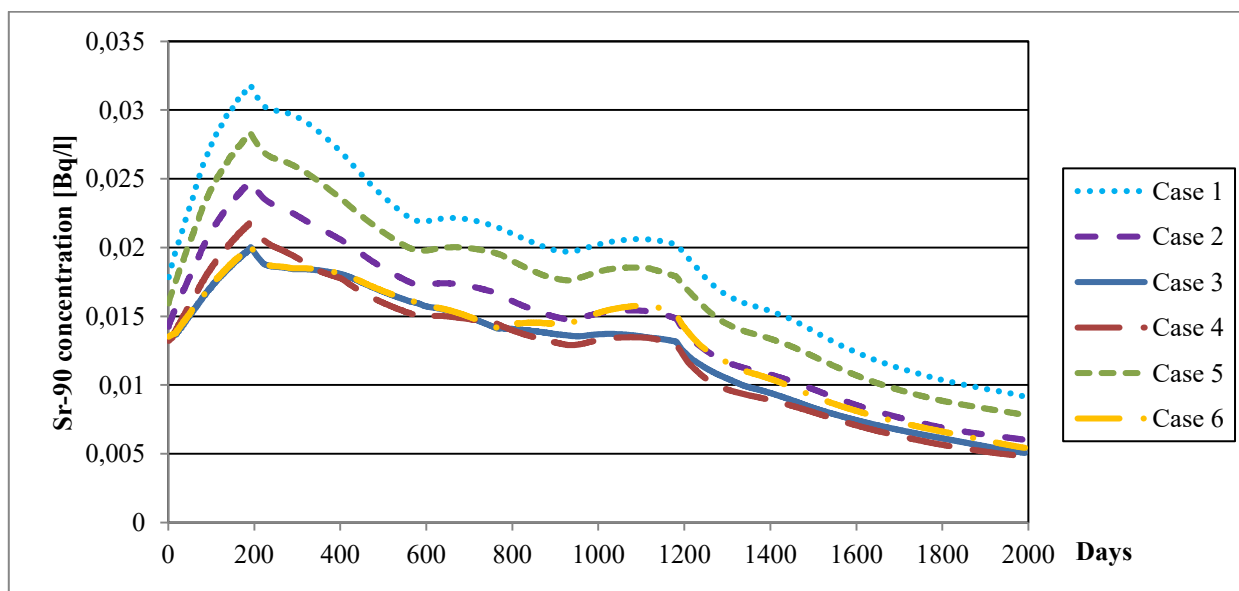


Figure 6 Sr-90 concentration in the monitoring well RP7 under different flow leakage conditions of the Cavour channel.

The performed parametric analysis underlines that if an in-detail survey on-site will be carried out concerning the main hydrogeological parameters (e.g. water table level, hydraulic conductivity of the streams, permeability of the streams and flow rate in the streams), a goodness prediction transport dynamic of Sr-90 can be performed. In particular, the effect of water flow rate in the surface channels influences the migration of strontium in the groundwater. The evident influence is not detectable in monitoring well far from it, such as SP01, where the parametric analysis has identified no variation respect to the calibrated model but it is locally determinable near the surface channel. This impact may add a new element of reflection to be considered in the safety analysis about the nuclear activities in the area.

However, the situation analyzed has evidenced that it does not represent a risk for population and environment, from a safety point of view. In fact, the detected quantities are far below the radiological risk and in some cases below the threshold of instrumental detection; the current safety standards and monitoring activities adopted in the nuclear site, as the current safety management of nuclear plants, allow to prevent or eventually mitigate, any release of the radionuclides, in quantity that may represent a radiological risk for population and environment.

Conclusions

Safety assessment activities of a nuclear site involve the survey of the radionuclide dynamics. This activity allows estimating the possible radiological risk for the population and environment. In this work, a nuclear site in Italy was analyzed, that hosts nuclear facilities under decommissioning. This site is monitored by the national environmental authorities (i.e. ARPA Piemonte). Authors have coupled on-site measurement published by the ARPA Piemonte with modelling, in order to understand the strontium dynamic in the hydrological and hydrogeological context of the investigated site. The presence of surface streams influences the pollutant transport in groundwater. First, the on-site measurements were analyzed to understand the surveyed strontium concentration in the monitoring wells located in and outside the nuclear site. The detected concentrations are far below the radiological limit, and in some cases below the threshold of instrumental detection, but the investigation of its dynamics is useful to add knowledge elements to the safety analysis framework. Second, this information was used to carry out a modelling activity. As a first step, a calibration model was developed evaluating the groundwater dynamic and the strontium transport in two of the main monitoring wells, RP4/7 and SP01. Then, a parametric analysis was performed to integrate previous studies on the groundwater (Testoni et al., 2016). The main parameters analyzed in this context were the hydraulic conductivity of the streambed, the flow rate of the Cavour channel and the high flow leakage from the Cavour channel. As result, the variation of the hydraulic conductivity of the streambed has no influence on the concentration in the investigated monitoring wells outside the nuclear site. Instead, the variation of flow rate has a direct influence on the concentration in the nearest monitoring well and no influence on the other one. The flow leakage from the Cavour channel has an evident influence in the monitoring well RP7 near the same channel. The water leakage from the Cavour channel dilutes the concentration of possible pollutants coming from the nuclear site, in an immediate neighborhood of the same channel; this is determined by the on-site measurement, analysis of groundwater dynamic and modeling. With respect to the Dora Baltea river, its local effect in the investigated site is due to the drainage of the groundwater for most of the year. From the safety viewpoint, this means that the risk of a relevant quantity of radionuclide in groundwater is reduced by the hydrogeological configuration of the area. The coupling of this modelling analysis with the monitoring activities periodically performed in the area contributes to support the safety study and may be useful to better evaluate the possible radiological risk for population and environment.

Future works could be carried out to improve this study, measuring the groundwater behavior in specific points, such as all the monitoring wells in the area, at shorter time intervals and characterizing more in detail the Cavour channel features (e.g. flow rate, flow leakage).

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