INFLUENCE OF THE CHEMICAL PROPERTIES OF THE GEOLOGICAL FORMATIONS ON THE MIGRATION PARAMETERS OF THE RADIONUCLIDES FROM SALIGNY REPOSITORY♦

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Abstract: The radionuclide concentration into the disposal system compartments of Saligny near surface repository is used as safety indicator in order to demonstrate the safety. The disposal system of Saligny near surface repository consists of waste form itself, engineered barriers and natural barriers. The conceptual model of Saligny near surface repository as

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well as the natural barriers characteristics are described. For the aim of calculation, the disposal system is splitted into eight compartments: waste form, slab foundation, foundation ground, silty loess, clayey loess, red clay, pre-quaternary clay, Barremian limestone. The concentration of the ^{3}H, ^{60}Co, ^{137}Cs, ^{14}C, ^{90}Sr, ^{99}Tc and ^{129}I isotopes in each disposal compartment of Saligny repository is calculated by using AMBER computer code and graphically represented. In order to demonstrate the safety of the disposal system, the calculated values of safety indicators are compared with the regulatory limits for drinking water.

Keywords: *radioactive waste, disposal system, safety indicator, safety assessment, site characterization, distribution coefficient, host geology*

INTRODUCTION

Safety assessment is an internationally recognized process used to evaluate the safety of radioactive waste disposal facilities. Safety assessments typically evaluate the impact of a disposal facility on human health in terms of radiological dose and, in some cases, radiological risk. Other performance measures (e.g., concentrations or fluxes of radio-nuclides in different parts of the disposal system) are also often considered and, in some cases, non-radiological and wider environmental impacts are considered.

A wide range of information is acquired in the course of developing a safety assessment. Some, but not all of this information is directly used in the formulation of models and data-sets for the evaluation of system performance and potential radiological impact. The results of such quantitative evaluations provide safety indicators that are used to construct arguments in order to evaluate the performance of a disposal system. Such indicators allow a judgment to be made as to whether the disposal system is acceptably safe in terms of risk to human health and harm to the environment. Safety indicators are characteristics or consequences that can be measured or calculated and compared to rigid or more loosely defined measures or 'yardsticks' in order to formulate such arguments [1].

Table 1 provides an overview of safety indicators, according to [1]. The table lists individual indicators, their 'sources', the location within the disposal system to which the indicator applies and the yardstick against which the value assigned to the indicator may be judged. The most widely used indicators in the context of the long term safety of disposal systems are those of radiological dose and risk. The last three safety indicators could be found as complementary indicators due to the proposed system is meant to help in placing the various indicators in a logical relationship with the various elements of a safety case. The most useful application of the proposed indicators will need to be determined on a case by case basis. In particular, it must be determined which indicators are expected to be most beneficial, considering both the specific features of the waste isolation system and the audience to which the safety case is to be presented.

Table 1. *Overview of safety indicators*

Safety indicator	Source	Application	Yardstick
Risk	Assessment model results	Human beings	Risk limit or constraint
Dose	Assessment model results	Human beings	Dose limit or constraint
Environmental impact	Assessment model results	Other species	Environmental protection standards
Radionuclide concentration outside the near field	Assessment model results	Accessible environment	Levels of corresponding natural concentrations
Radionuclide fluxes outside the near field	Assessment model results	Accessible environment; geosphere-biosphere interface	Corresponding natural fluxes
Containment time	Experiments, technical specifications, and/or process model calculations	Canisters/ containers Engineered barriers or geosphere	'Crossover times' for hazard indices

MATERIALS AND METHODS

The multi-barrier concept of the Saligny repository

The basic concept related to disposal system is the defense in depth concept (multiple barrier concept), which defines the disposal system as an assembly between the disposal structures and the site. The confinement of radio-nuclides is ensured by the synergy of the engineered structures and the favorable site characteristics (geosphere and biosphere), the control of the waste form and content, adequate operational procedures and the institutional control.

The first engineered barrier proposed for the Saligny repository, according [2], consists of the waste form itself. The wastes are immobilized into a cement matrix in a disposal drum. More drums are placed into a disposal module and the free space between them is filled with gravel or sand, in order to allow retrieval of module, if case. The disposal modules are considered the second barrier; they have parallelepiped in shape and their structure consists of reinforced concrete. The material used for disposal modules has a very low corrosion rate in order to confer both a good long term operation and intrinsic safety of the containers system. Also, the disposal modules are designed to resist fall during the filling period of repository. Disposal cells together with the separated drain water systems for the cell-infiltrated water and of the meteoric waters represent the next barrier. After the cells are filled and covered with the reinforced concrete cap, and the protection is performed with bituminous waterproofing layer.

The proposed system for the long-term cover of the disposal cells consists of draining and waterproofing alternating layers from sand, geo-membrane, crushed rock and clay

or compacted loess, to prevent the surface water infiltration into the repository structures. The long term cover structures have also the role to prevent the intrusion of animals and of plant roots towards the cells with radioactive waste. The final covering layers are provided with slopes, slopes that will provide the rapid evacuation of meteoric water.
The soil superior layer is performed in order to rebuild the natural environment, after the repository is closed. The long term closing system of the disposal cells as well as the improved foundation ground represent external repository barriers, which contribute to confinement of radio-nuclides.
The safety functions for repository engineered barriers considered into conceptual design of Saligny repository are the following:

- Physical, chemical, hydraulic and biological isolation of wastes and long term minimization of radio-nuclides releases into geosphere.
- Guarantee of a chemical barrier, physical confinement, retention and retardation of radio-nuclides, as well as control of gases generation and release.
- Completion of natural barriers functions, regarding wastes confinement.
- Long-term stability of disposal facility.

In the figure 1 is presented the conceptual design of the Saligny repository, according to reference [2].

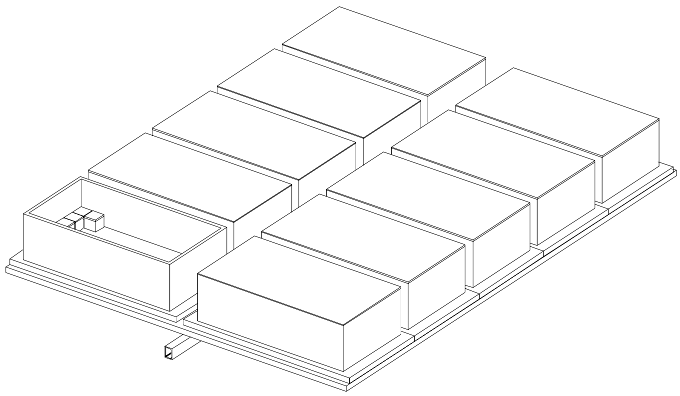

Figure 1. *Conceptual design of the Saligny repository*

Description of Saligny site characteristics

The Saligny site is located within the exclusion area of Cernavodă Nuclear Power Plant. The site is located in the Constanța County, in the South-Eastern part of Romania, in the South Dobrogea region. It is situated at around 2 km SE far from Cernavodă town and 1.5 km NW from Saligny village [2].
Geologically, the Saligny site belongs to the Dobrogean part of the Moessic platform, placed at south of Ovidiu – Capidava fault in the South – Dobrogea platform. The main characteristic of this zone is the deep crystalline foundation covered by thick sedimentary layers. Saligny site structure consists of the sequence of the following geological units: silty loess, clayey loess, Quaternary red clay, Pre-Quaternary clay, Barremian limestone, Vallanginian clay, Jurassic limestone, Paleozoic sediments and the crystalline foundation of the green slates. The units significant for the long-term

dose and risk assessment for the population in the surrounding area are the silty loess, the clayey loess, the Quaternary red clay, the Pre-Quaternary clay and the Barremian limestone. In the safety assessment report developed in order to provide confidence in safety of disposal system the units received the following codifications: horizon A – representing the silty loess, horizon B – representing the clayey loess, horizon C – representing the red clay, red sandy clay and the red clayey sand, horizon D – representing the pre-quaternary complex, consisting in different kind clays lenses of sand, gravel, limestone and sandstone.

From hydro-geological point of view unsaturated (vadose) zone includes horizons A, B, C and upper part of horizon D, and the saturated zone-lower part include horizon D which includes local small aquifers as well as the main aquifer of Cernavodă.

The structural characteristics of each geological unit of the Saligny site consist of dry density, total porosity, effective porosity, water filled porosity, saturated hydraulic conductivity, soil-water retention capacity and granulometric composition. Some of these parameters, used in the assessment of the complementary safety indicators in the present paper are listed in Table 2 [2]. According to the site stratigraphy presented in [2], the Horizon B is also split in four sub-layers: Iab1, Ib (upper), Iab2 and Ib (lower), having different clay content.

Table 2. *Characteristic parameters of Saligny site, as input data for complementary safety indicators assessment [2]*

Horizon	Average values of dry density [kg·m^{-3}]	Average values of water filled porosity [%]	Depth (avg. value of the geological layer) [m]
Horizon A – silty loess (layer Ia)	1540	12.30	1
Horizon B – clayey loess:			
- layer Iab1	1780	25.68	6
- upper layer Ib	1570	14.03	4
- layer Iab2	1720	25.13	2
- lower layer Ib	1690	25.20	6
Horizon C – quaternary clay	1760	32.38	8
Horizon D– pre-quaternary clay	1760	30.67	10

From geochemical point of view, Saligny site is characterized by pH values in range 8.5 to 9. The cation exchange capacity (CEC) increases with depth, having the highest values in C and D horizons. The average values for pH and CEC for main horizons of Saligny site are presented in Table 3 [2].

Table 3. *Average values of pH and CEC for the main horizons of Saligny Site*

Horizon	pH	CEC [meq./100 g]
A – dusty loess	9.26 ± 0.32	8.74 ± 2.38
B – clayey loess	9.52 ± 0.20	10.31 ± 3.76
C – quaternary clay	9.00 ± 0.43	14.76 ± 8.64
D – pre-quaternary clay	8.53 ± 0.48	13.39 ± 6.46

The distribution coefficients as well as diffusion coefficients are very dependent on the radionuclide and the geological unit. The values of distribution coefficients which have been obtained experimentally for the samples collected from Saligny site for some radio-nuclides ^{137}Cs, ^{60}Co, ^{90}Sr and ^{14}C for different horizons are presented in the Table 4 [2]. These values are the average values calculated taking into account all samples collected from many boreholes during the site characterization process.

Table 4. *Representative distribution coefficients (k_d) for the main horizons of Saligny site*

Radio-nuclide	k_d in the Saligny Horizons [$m^3 \cdot kg^{-1}$]			
	Horizon A Silty loess	**Horizon B Clayey loess**	**Horizon C Red Clay**	**Horizon D Pre-Quaternary clay**
^{137}Cs	0.774	1.131	4.131	2.366
^{60}Co	0.033	0.030	0.031	0.030
^{90}Sr	0.006	0.011	0.012	0.012
^{14}C	0.003	0.005	0.008	0.005

Calculation of safety indicators

The safety aspects of the disposal system are the purpose of specific safety analyses, which represents the main element in the licensing process of a repository. These analyses have to be developed systematically, following traceable and reproducible methodologies, easy to check and to justify, which allow the assessment of long term evolution of the repository. As specified in [3], the radioactive waste disposal has to be conducted to ensure the protection of workers, public and environment against the radiological risks resulted from the disposed wastes.

The individual annual effective dose both for workers and public has been identified and evaluate for Saligny repository as main safety indicator. The radionuclide concentration into the disposal system compartments, and containment time values have been identified and preliminary evaluated as complementary safety indicators in references [2] and [4]. The present paper describes the second iteration of the complementary safety indicators assessment, namely the distribution of radionuclide concentration in the geological layers of Saligny site, as complementary safety indicator of the proposed disposal system.

The evaluation of radionuclide concentration into disposal compartments was used the AMBER computer code, version 5.0. A simplified conceptual model of Saligny repository has been developed using the interaction matrix method in accordance with the requirements of the evaluation computer code. The AMBER computer code uses a compartment model approach and it allows all the system components, migration processes and exposure mechanisms to be represented using a single code. The system was split into a series of assumed homogenous compartments and the transfer processes between the compartments were expressed as transfer coefficients that represent the fraction of the activity in a particular compartment transferred from that compartment to another one, per unit time. The mathematical representation of the inter-compartmental transfer processes takes the form of a matrix of transfer coefficients that allow the compartmental amounts to be represented as a set of first order linear differential equations. For the i^{th} compartment, the rate at which the inventory of radio-nuclides in a compartment changes with time is given by:

$$\frac{dN_i}{dt} = \left(\sum_{j \neq i} \lambda_{ji} N_j + \lambda_N M_i + S_i(t) \right) - \left(\sum_{j \neq i} \lambda_{ij} N_i + \lambda_N N_i \right) \quad (1)$$

where i and j indicate compartments, N and M are the amounts (Bq) of radio-nuclides N and M in a compartment (M is the precursor of N in a decay chain). $S(t)$ is a time dependent external source of radionuclide N ($Bq \cdot y^{-1}$). λN is the decay constant for radionuclide N (y^{-1}) and λ_{ji} and λ_{ij} are transfer coefficients (y^{-1}) representing the gain and loss of radionuclide N from compartments i and j. For simplicity, the above equation assumes a single parent and daughter. However, AMBER allows the representation of multiple parents and daughters.

The mathematical model developed in order to assess the distribution of radionuclide concentration in the geological layers of the Saligny site, is mainly based on the release, migration and transport of radio-nuclides from waste considering only decay, adsorption, dispersion and advection processes. The transport equation for unsaturated and saturated geological layers was simplified in order to be solved using AMBER computer code. The dispersion was considered by the discretisation of the disposal facility compartments.

Radioactive decay is represented through the decay rate (λ, in y^{-1}), which is given by the equation (2):

$$\lambda = \frac{\ln 2}{t_{1/2}} \quad (2)$$

where $t_{1/2}$ is the half-life period of the radionuclide (y).

Adsorption is described through the retardation phenomenon, which, for a given compartment, is dependent on the radionuclide. The retardation factor R (dimensionless) specific for a compartment is calculated using equation (3):

$$R = 1 + \frac{\rho \cdot k_d}{\vartheta_w} \quad (3)$$

where ρ is the dry bulk density of the compartment ($kg \cdot m^{-3}$), k_d is the distribution coefficient of the element in the compartment ($m^3 \cdot kg^{-1}$) and ϑ_w represents water-filled porosity of the analyzed compartment (dimensionless).

For the unsaturated transfers, the advective transfer rate of radio-nuclides (λ_{flow}, in y^{-1}) is given by equation (4):

$$\lambda_{flow} = \frac{q}{L \cdot \vartheta_w \cdot R} \quad (4)$$

where q represents the annual flow rate through the compartment ($m \cdot y^{-1}$), L represents the length of the compartment on the direction of the water flow (m), ϑ_w represents the water-filled porosity of the compartment (dimensionless), and R represents the retardation of the compartment (dimensionless). The parameter q ($m \cdot y^{-1}$) represents the infiltration rate of water through the repository compartments and geosphere (unsaturated zone).

For the saturated zone, the water infiltration rate is given by equation (5):

$$q = K \frac{\partial H}{\partial x} \qquad (5)$$

where K represents the hydraulic conductivity of the compartment ($m \cdot y^{-1}$) and $\partial H/\partial x$ represents the hydraulic gradient (dimensionless).

Assessment Hypotheses

The conceptual model of the Saligny disposal facility considered in the assessment on the present paper is presented in Figure 2. The compartments which represent the engineered barriers of repository (Waste form, Slab Foundation and Foundation Ground), as well as the compartments Silty Loess, Clayey loess, Red Clay and the Upper part of Pre-Quaternary clay were considered as unsaturated layer. The Barremian limestone, which is the host of the site main aquifer, is considered as saturated zone.

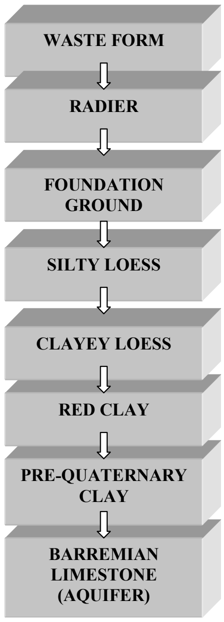

Figure 2. *Conceptual model of the Saligny disposal system considered in the evaluation*

The dry density and the water filled porosity for the geological layers are the values from Table 2. For the other compartments of the conceptual model, the values for density and water filled porosity were taken from literature [5]. Thus, the waste form and the slab foundation were considered concrete type (dry density = 2500 $kg \cdot m^{-3}$ and water filled porosity = 0.15). The Barremian limestone were considered as aquifer type, having a dry density of 1800 $kg \cdot m^{-3}$ and water filled porosity = 0.30. The foundation ground is considered to have the physical and chemical properties as clay. Another very important parameter to be considered in the calculation is the infiltration rate in the natural layers of the site. For the Saligny site, for the natural ground, an infiltration rate of about 20 mm/year is specified in [2]. The value was obtained during the site characterization process. In the present calculation, for the first 300 years after the repository closure, the infiltration rate in the disposal structures was considered zero. Between 300 and 500 years, the infiltration rate in the repository was considered about

10% from the natural infiltration rate and, after 500 years, the infiltration rate in the disposal structures was considered the same like in the natural ground.
The main radio-nuclides considered in the evaluation are the characteristic radio-nuclides from a CANDU Power Plant, namely ^{3}H, ^{60}Co, ^{137}Cs, ^{14}C, ^{90}Sr, ^{99}Tc and ^{129}I. The Waste Form compartment is considered to be a generic disposal cell, with an inventory (table 5) equal with the total inventory evaluated to be disposed in the Saligny repository [2]. The half-life periods are from literature.

Table 5. *Inventory considered in evaluation*

Radionuclide	Half life periods [y]	Total activity [Bq]
^{3}H	12.4	7.6 E+14
^{14}C	5730	3.0E+14
^{60}Co	5.27	2.9E+11
^{90}Sr	29.1	2.7E+14
^{99}Tc	2.13E+05	8.9E+10
^{129}I	1.57E+07	8.9E+09
^{137}Cs	30	8.9E+14

For ^{137}Cs, ^{14}C, ^{60}Co and ^{90}Sr, the distribution coefficients considered in calculation for the geological layers were the coefficients specified in [2] (experimentally determined for Saligny site). The distribution coefficients for above mentioned radio-nuclides, for the other compartments of the disposal system, as well as the distribution coefficients for ^{3}H, ^{99}T and ^{129}I for whole disposal system were taken from literature [5] and [6]. The time period considered in evaluation was 1E+6 years. In the first 300 years the regulatory control period was considered.
The regulatory limits of contaminants for drinking water specified in [7] and [8] and presented in table 6 were considered as yardsticks against which the obtained values of radionuclide concentrations may be judged.

Table 6. *Regulatory limits fore drinking water*

Radionuclide	Concentration limit in drinking water [Bq m^{-1}]
^{137}Cs	1.48 E+00
^{90}Sr	3.70 E+00
^{129}I	3.70 E-03
^{14}C	9.26 E+04
^{3}H	7.41 E+05
^{99}Tc	3.33 E+04

RESULTS AND DISCUSSIONS

For all radionuclides taken into account is the assessment, the concentration distribution in the disposal system compartments is presented in the figures 3÷8. The compartments are considered from the upper level to the lower level. The deeper compartment was considered the lower part of pre-quaternary clay, under the saturated zone of Saligny site. The figures 9÷11 present some special cases of ^{14}C, ^{137}Cs and ^{3}H.

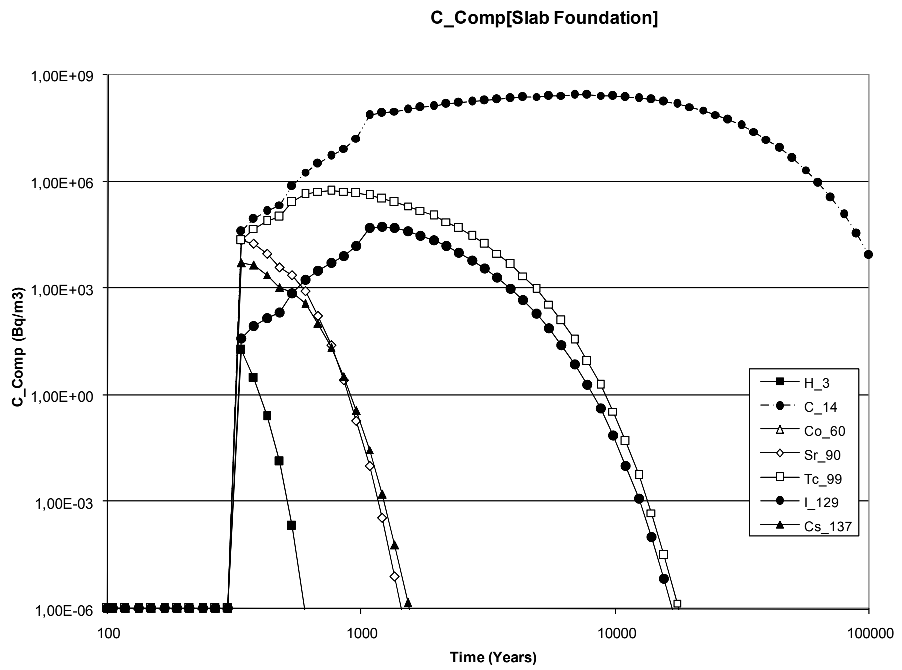

Figure 3. *Radionuclide concentration distribution in the Slab Foundation compartment*

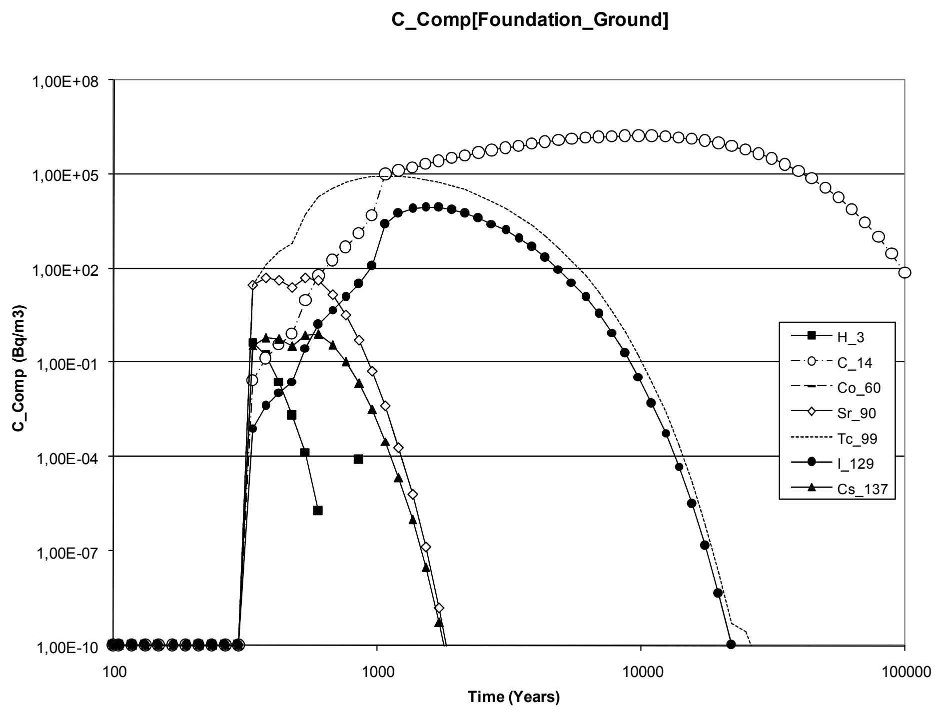

Figure 4. *Radionuclide concentration distribution in the Foundation ground compartment*

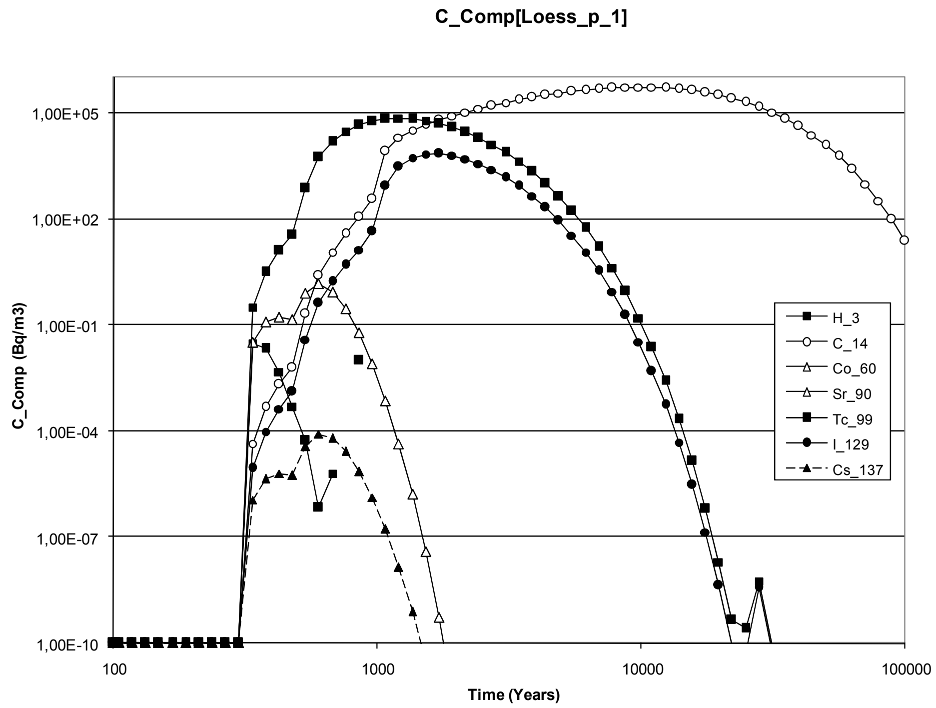

Figure 5. *Radionuclide concentration distribution in the Silty loess compartment*

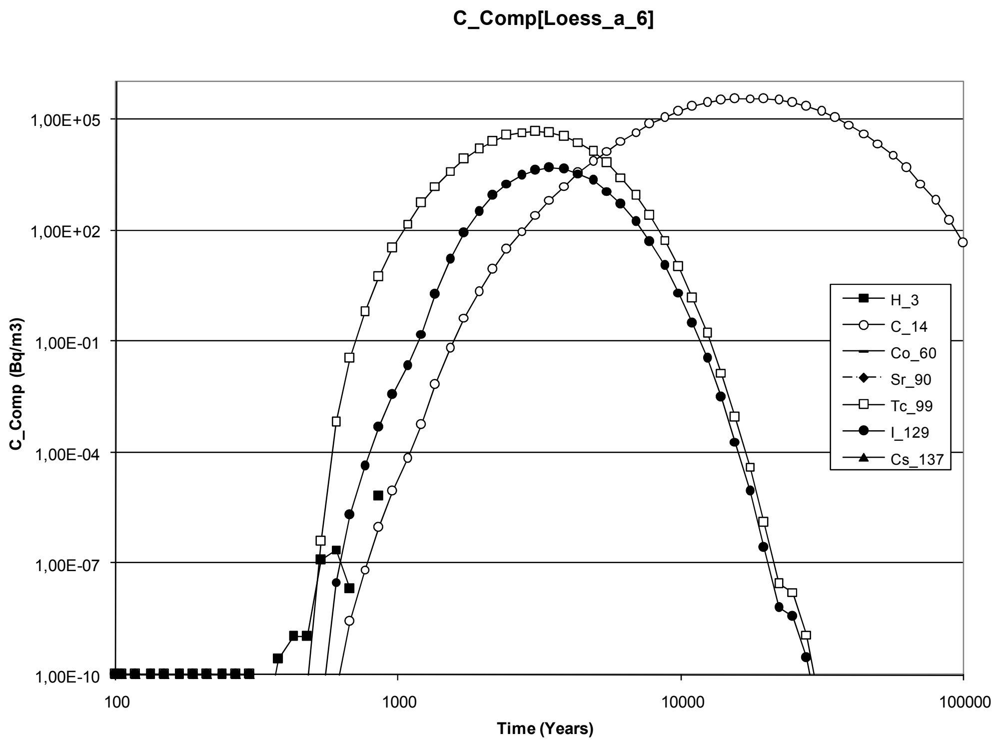

Figure 6. *Radionuclide concentration distribution in the Clayey loess compartment*

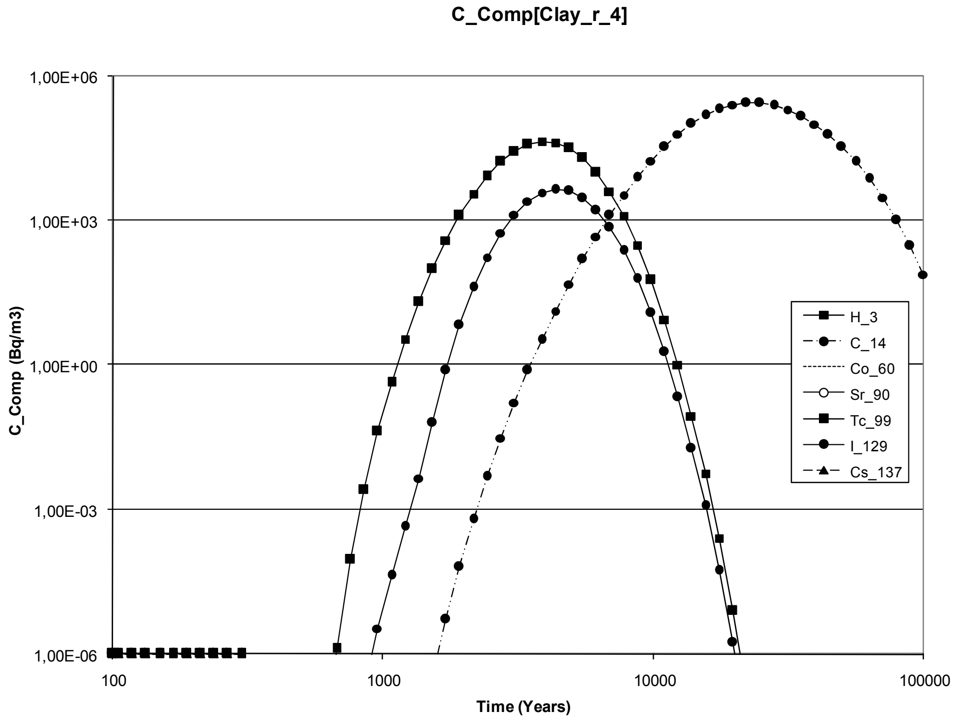

Figure 7. *Radionuclide concentration distribution in the Red clay compartment*

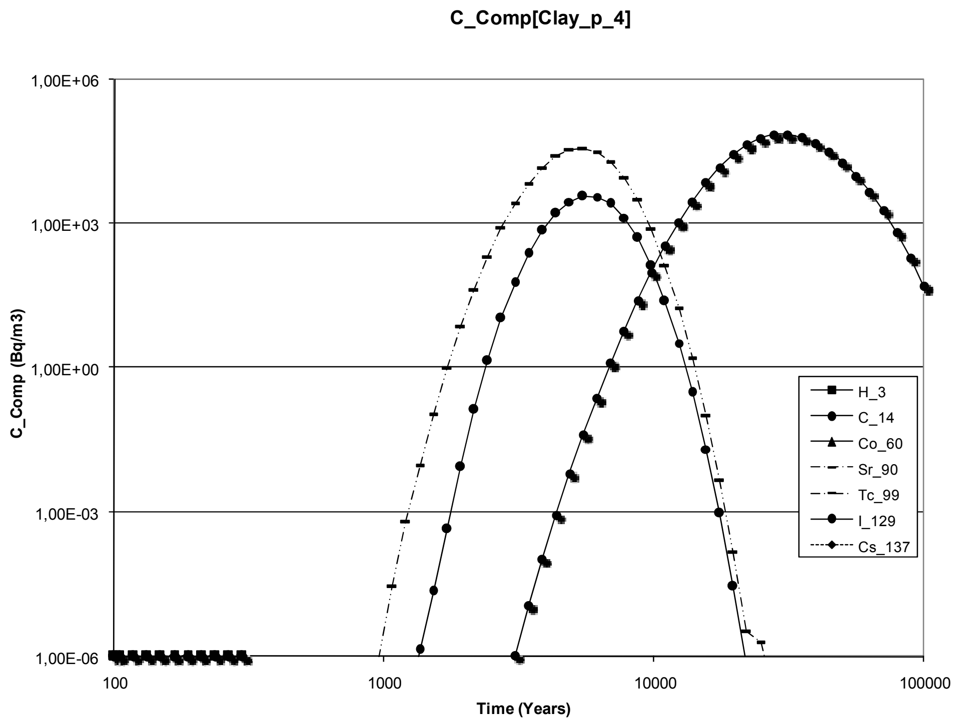

Figure 8. *Radionuclide concentration distribution in the Pre-quaternary clay compartment*

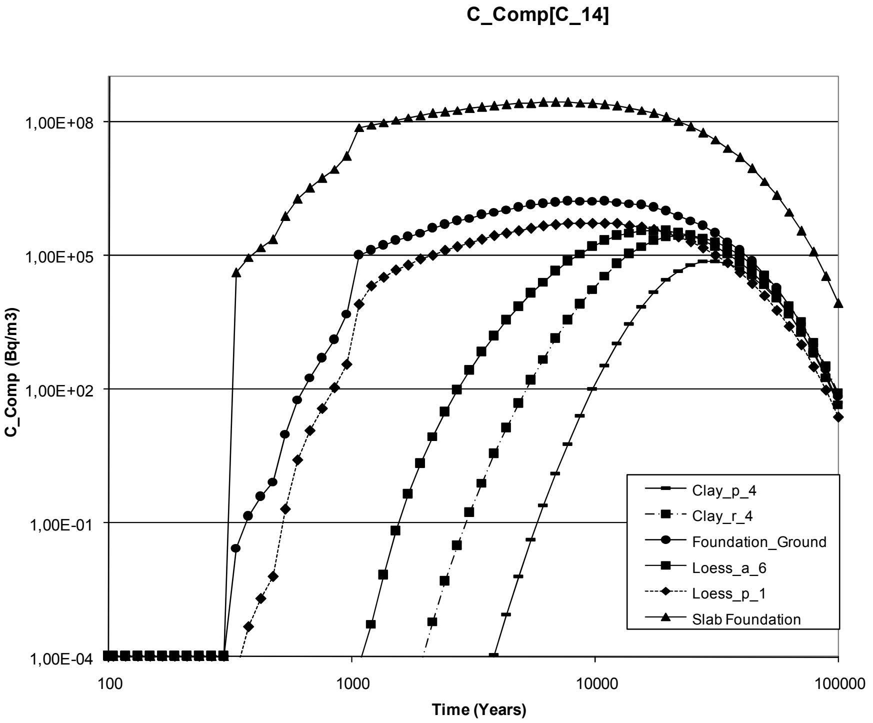

Figure 9. *C-14 concentration distribution in the disposal system compartments*

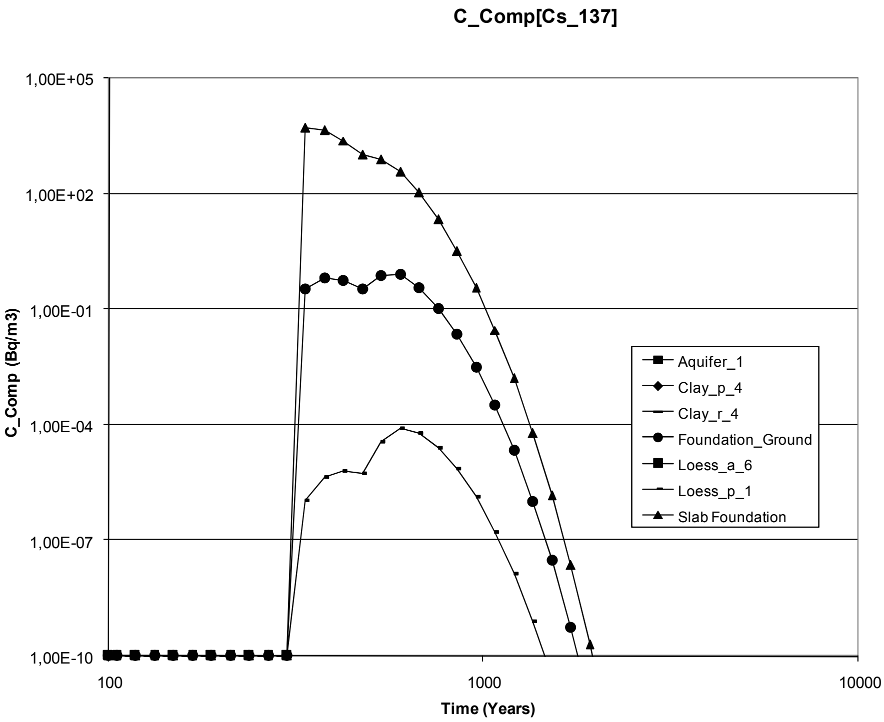

Figure 10. *Cs-137 concentration distribution in the disposal system compartments*

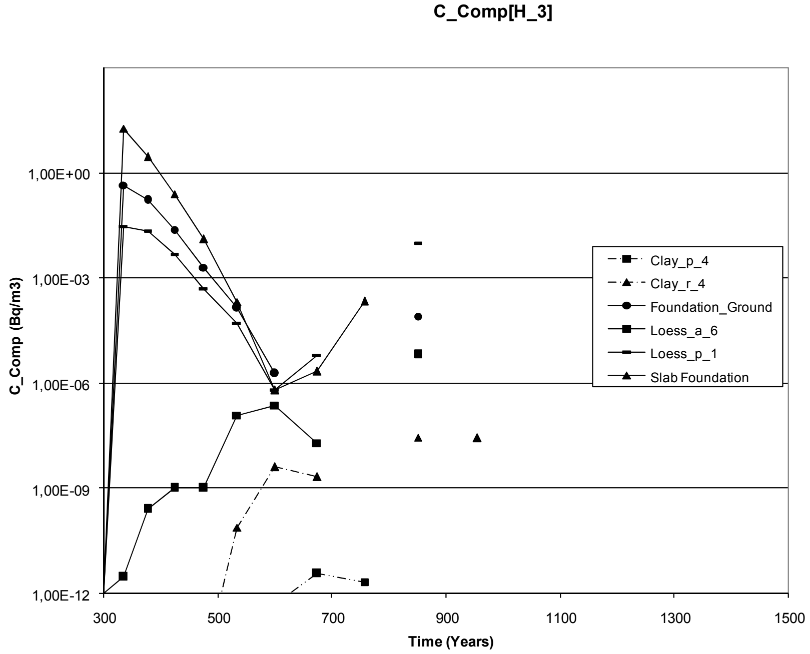

Figure 11. *H-3 concentration distribution in the disposal system compartments*

The figures 3 to 8 show the role of the engineered barriers and geological layers regarding the retardation and retention of radionuclides. Therefore, all the radionuclides considered in the assessment reach the upper layers of the disposal system (namely Slab Foundation, Foundation Ground and Silty Loess), their concentrations having values higher than 1·E-6 Bq·m^{-3} (exception for short lived radionuclide ^{60}Co). The maximum extension of concentration plume for ^{60}Co is only in slab foundation and foundation ground, and the peak values are lower than 1·E-06 Bq·m^{-3}.
But in the deeper layers of site, and, especially, at the upper limit of saturated zone, only the long lived and very mobile radionuclides (^{14}C, ^{99}Tc, ^{129}I) may have a concentration comparable with the regulatory limit for drinking water. Especially in the pre-quaternary clay layer, at the upper limit of the Saligny site saturated zone, the peak values are reached after 5,000 years from the repository closure for the very mobile radionuclides ^{99}Tc and ^{129}I. The peak value for ^{14}C is reached only after 30,000 years, due to the good retention of that radionuclide in the concrete engineered barriers, and, also, in some natural layers.
The figure 8 shows that the concentrations of ^{14}C and ^{99}Tc in the pre-quaternary clay are at the same order of the regulatory limit for drinking water. Only ^{129}I reaches the saturated zone in a concentration higher than the maximum admitted limit for drinking water, but the present assessment have a lot of limitation regarding the iodine release and transport. Thus, dilution in aquifer as well as the gas phase release was not considered, and, in the same time, the distribution coefficient of iodine was not determined for the geological layers of Saligny site and will be the subject of the future experimental research activity).
Also, it is necessary to highlight the movement in time of peak values from the upper layer (silty loess) to the deeper layer (pre-quaternary clay) and in the same time, the decreasing of the correspondent peak values for each to the large time moment.

Figure 9 presents the situation of ^{14}C, a long lived radionuclide, having a good retention on engineered barriers of disposal system (concrete type compartments) and a significant retention on some geological layers (as is presented in table 4). The figure highlights the movement in time of peak values from the upper layer (silty loess) to the deeper layer (pre-quaternary clay) and in the same time, the decreasing of the correspondent peak values.
The extension of ^{137}Cs concentration plume is also in the upper part of loessoid layers (Figure 10). Taking into account the calculation assumptions, even in the silty loess the ^{137}Cs concentration is lower that the admitted limit in drinking water (see table 6). The cesium migration on geological media of the site is mainly influenced by the presence of clayey minerals, especially montmorillonit that leads to a very strong adsorption of radionuclide in unsaturated zone. The extension of the ^{90}Sr concentration plume is the same as the ^{137}Cs.
For the very mobile radionuclide, tritium, the proposed disposal system seems to be very adequate. Taking into account the calculation assumptions, in the first 300 years after the repository closure, tritium will be released from the disposed waste only by diffusion and in the gas phase. Therefore, the migration in the aqueous phase (after 300 years) will have as result a plume with low extension, only in the upper part of saturated zone. The peak concentration will be well lower that the maximum admitted limit for drinking water.

CONCLUSIONS

The engineered barriers and geological layers have a very important role in the retardation and retention of radionuclides.
The deeper layers of the Saligny site, especially at the upper limit of saturated zone the long lived and very mobile radionuclides (^{14}C, ^{99}Tc, ^{129}I) may have a concentration comparable with the regulatory limit for drinking water.
The long lived radionuclide ^{14}C, has a good retention on engineered barriers of disposal system and a significant retention on some geological layers.
The tritium concentration into aqueous phase is lower that admitted limit after 300 years.

REFERENCES

1. IAEA-TECDOC-1372: *Safety indicators for the safety assessment of radioactive waste disposal*, IAEA, Vienna, **2003**.
2. RI-1547: *Safety Report for Siting a Near Surface Repository at Saligny Site*, SITON, **2007**;
3. NDR-05: *Norms for near surface disposal of radioactive waste*, approved by CNCAN Order no. 400/2005, and published in Official Monitor no. 345/17.04.2006
4. Niculae, O., Durdun, I., Ionita, Gh.: Safety indicators used to prove the role of natural barrier for Saligny near surface disposal system, *Proceedings of the 13th*

International Conference "Progress in cryogenics and isotopes separation", Calimanesti, Romania, **2007**;
5. ISAM: *Safety Assessment Methodologies for Near Surface Disposal Facilities*, Results of a co-ordinated research project, Volumes 1 and 2, IAEA, **2004**;
6. Park, J.B., Park,J-W., Lee, E-Y., Kim, C-L.: Experiences from the source-term analysis of a low and intermediate level radwaste disposal facility, *Proceedings of WM'03 Conference*, February 23-27, Tucson, AZ, **2003**;
7. EPA- U.S. Environmental Protection Agency: *Radionuclides in Drinking Water - A Small Entity Compliance Guide*, Office of Ground Water and Drinking Water (4606M), EPA 815-R-02-001, 2002;
8. US NRC: Federal Code 40 CFR 141.16.