Chernobyl case study increases confidence level in radionuclide transport assessments in the geosphere

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Abstract. Results are presented from the ongoing international study (i.e., Chernobyl Pilot Site Project) on characterization and modeling of fallout nuclear fuel particles dissolution behavior and subsequent radionuclide transport from the near-surface waste disposal site in Chernobyl zone to the geo-environment. The reported experimental work includes: - characterization of radionuclide physico-chemical speciation inside the waste site (trench); - evaluation of infiltration recharge regime through the trench body; and determination of waste soil matrix sorption parameters. In order to describe radionuclide releases from the waste, the multi-component source term model accounting for several types of fuel particles (UO_2, UO_{2+X}, and U-Zr-O matrix) was developed. Application of the source-term model to the Pilot Site data has given satisfactory and encouraging results. The modeling approach has proved to be capable of rather accurately reproducing independent field characterization and monitoring data relevant to ^{90}Sr speciation inside the trench and radionuclide releases from the trench to the aquifer. The developed parameter database and modeling approach are of general value for radiological assessments of Chernobyl contaminated areas, and for possible similar accidental situations in future. Thus, acquired knowledge and data serve to increase the confidence level in radionuclide transport assessments from nuclear fuel source term to soils and geo-sphere.

1. INTRODUCTION

Important step of any environmental impact or safety assessment is connected with selection of contaminant migration model(s) and parameters. The usual approach is to use in parallel the data from the site (often very sparse) and the existing modeling methodologies and migration parameter databases from the literature. The challenge of such an approach is to evaluate the accuracy (or uncertainty) of modeling predictions with no (or very limited) practical means to test their validity (because of time or space scale of the considered phenomena). Often, the quantification of uncertainties is based on no more than a judgment of the involved experts.

Within the frame of Chernobyl Pilot Site project [1-3], which addresses a real-world “worst case” scenario of near-surface disposal of nuclear fuel materials, we get the opportunity to compare and cross-check *a-priori* (or based on preliminary site characterization) modeling analyses with the *a-posteriori* collected field data and observations. Possibility to do continued characterization of the site allows establishing permanent feedback between data collection, selection (and revision, if necessary) of radionuclide migration models, and eventually to perform model confirmation (validation) exercises in the laboratory or in the field.

At the current stage of the project (which is scheduled for completion in mid 2003) we can draw preliminary conclusions on the adequacy of models that we use to describe radionuclide migration from the dispersed nuclear fuel source term to the geo-environment, formulate some lessons learned from the Chernobyl case study, and outline research issues deserving future analyses.

2. BACKGROUND

2.1. Chernobyl Pilot Site Project

The Chernobyl Pilot Site Project is organized through an international cooperative agreement between four organizations. From Ukraine, there are the Institute of Geological Sciences (IGS) and the Ukrainian Institute of Agricultural Radiology (UIAR), and from France, the partners are the Tracer Application Office (CEA/SAT-French Atomic Energy Commission) and the Institute for Radiation Protection and Nuclear Safety (IRSN/DPRE/SERGD/LESTS).

IRSN is managing the Chernobyl Pilot Site (CPS) project from the mid 1999 for a four years duration period. The project is aimed at characterization and modeling of processes of radionuclides migration in soils and geosphere from the near-surface radioactive waste burial (trench) containing dispersed nuclear fuel. In the frame of the CPS research program, modeling and characterization techniques are used together in the issue of experimental confirmation (validation) in the laboratory and in the field of the relevant radionuclide migration models [1].

In 1999 the experimental site (Chernobyl Pilot Site) has been selected and instrumented, and the research program has been started on characterization of radionuclide migration source term and surrounding hydrogeological environment (unsaturated zone and aquifer). The main processes involved in radionuclide migration from the waste disposal site to the surrounding geo-environment give rise to two major lines of research. Firstly, the Chernobyl Pilot Site Project is focused at the study of the dissolution mechanisms of fall-out fuel particles and geochemical interactions between the soil and the dissolved radionuclides, and secondly, at the study of the hydrodynamics of water and associated dissolved radioactive elements transport in the unsaturated zone and in the aquifer underlying the waste burial (Fig.1).

The project is scheduled for 1999 – mid 2003 and comprises three main stages: (1) the site characterisation involving collection and analysis of radiological, hydrogeological and geochemical data; (2) development of a set of sub-models and global model of the waste site, and planning of model validation (confirmation) experiments, and (3) setting up, carrying out and interpretation of model validation tests.

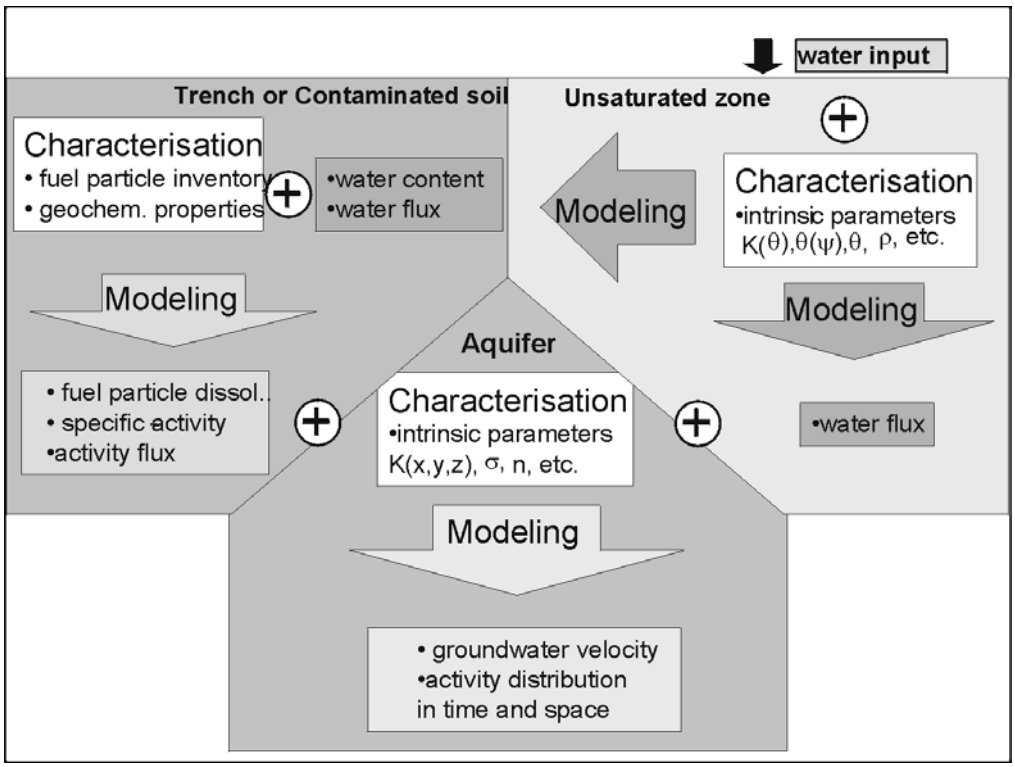


Fig.1. Block-diagram of the research issues of the Chernobyl Pilot Site project.

2.2. Instrumentation and characterization of experimental site

The study site is located 2.5 kilometers South-West of the ChNPP in the so-called “Red Forest” radioactive waste dump site (Fig.2). In 1987 in the course of clean-up works radioactive materials, made of topsoil layer and contaminated tree trunks from the dead pines “red forest” killed by extreme radiation levels in 1986, have been bulldozed *in-situ* in a few meters deep trenches [4].

A particular waste burial (trench no.22), namely Chernobyl Pilot Site (CPS), was selected since 1998 by the joint French-Ukrainian scientist team for radiological characterization and monitoring studies reported below. The burial represents ~70 m long, 8-10 m wide and 2-2.5 m deep unlined trench (see Fig.2). The source term of radionuclide migration to the geo-environment is a heterogeneous mixture of contaminated organic materials and soil containing micron-size reactor fuel particles. The specific activity of ^{137}Cs and ^{90}Sr in waste is $n\times10^5$- $n\times10^6$ Bq/kg.

The geologic section of CPS consists of the sequence of sedimentary layers of Quaternary eolian and alluvial sands with beds of sandy loam material. The depth to groundwater table is 2-3 m. The unconfined sandy aquifer is bounded at a depth of 30 m by a low permeable Eocene marl layer. For 15 years following disposal, radionuclides have been leached from the trench by meteoric water (average annual rainfall is 550-650 mm), and have been penetrating the underlying unsaturated soil and the aquifer. As a result, ^{90}Sr concentration in groundwater in the upper part of the aquifer in the vicinity of the trench varies between $n\times100$ and $n\times10{,}000$ Bq/l, and the radiostrontium plume has spread some 10 m downstream from the source [3]. In accordance with field evidences of migration, the CPS project is primarily focused on characterization and modeling of migration behavior of strontium-90 (^{90}Sr), which is a mobile radioactive contaminant, and has migrated in significant quantity from the trench to the surrounding geo-environment.

In the course of the project the CPS was equipped by series of piezometers and multilevel groundwater quality observation wells to the aquifer, ceramic moisture samplers to unsaturated soil profile, on-site laboratory facility, automated weather station and special pit instrumented with sensors for monitoring water flow and radionuclide migration process in the unsaturated zone (see Fig.2).

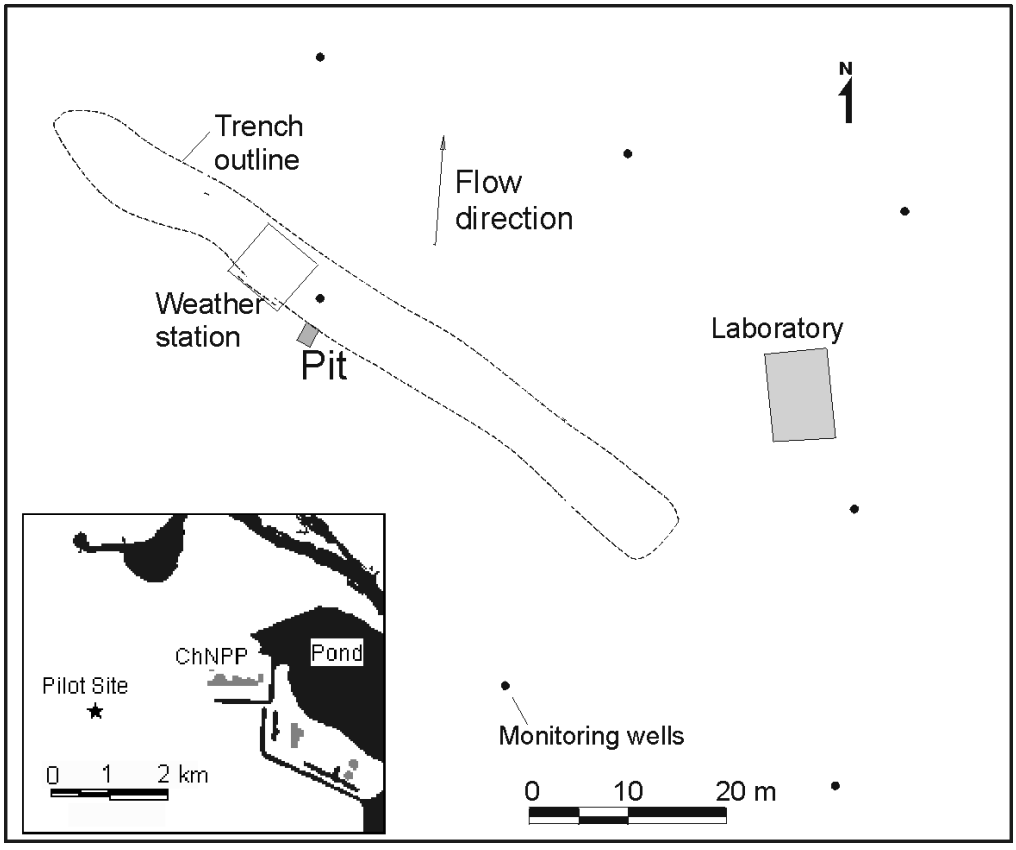


Fig.2. Map of the Chernobyl Pilot Site.

During the characterization phase of the project (1999-2000) data has been acquired on the general characteristics of the waste site, including: - trench no.22 geometry and radioactivity inventory (^{137}Cs, ^{90}Sr); - lythology properties of sediments and geology model of the site; - groundwater flow regime in the aquifer; - ^{90}Sr distribution in the subsurface environment and parameters of interaction (for instance sorption process) with the aquifer sediments. Results from above studies are reported in [1-3].

In this paper we present results on characterization and preliminary modeling of the CPS radionuclide migration source term. Following conclusions of the preliminary project phase (see above paragraph), the subsequent experimental studies has focused on in-depth analysis of a number of the key questions, in particular: physico-chemical properties and dissolution parameters of fuel particles; infiltration flow regime in the unsaturated zone; and sorption parameters for the waste matrix. The developed improved conceptual understanding of fuel particle dissolution mechanisms and acquired hydro-physical and geo-chemical parameters were integrated to the mathematical model describing radionuclide releases from the fuel particles to the waste matrix, and further to the surrounding geo-environment. Presented below comparison of modeling predictions with independent field observations provides experimental confirmation of the developed model for the Chernobyl radionuclide migration source term.

3. SOURCE TERM CHARACTERIZATION

3.1. Physico-chemical speciation and dissolution of fuel particles

The Chernobyl fall-out radionuclides comprising migration source-term were initially associated with micron-size dispersed nuclear fuel “hot particles” [5-6], which were not readily dissolvable in natural waters. Therefore, in order to quantify the rate of fission product (^{90}Sr and ^{137}Cs) migration to the geo-environment, it is necessary to determine the radionuclide inventory and dissolution rates of fuel particles (FP).

Historically, dissolution of FP and releases of radioactive contaminants to the environment in post-Chernobyl studies have been described by a single phenomenological first order rate equation with empirical coefficient [5,7-8]. It was established by Kashparov *et al.* [5, 10] that principal factors controlling fuel particle dissolution rate are pH of soil media, and FP oxidation state. The last one is mainly determined by the degree of FP high temperature annealing by fire in the Unit 4 during the Chernobyl accident.

The adequacy of the traditional approach [e.g., 7,8] to modeling dissolution of FP was questioned by the data of the scanning electron microscopy (SEM) studies of fuel particles carried out by Ahamdach [9] in the frame of CPS project. It was established that the CPS source term consists of two types of FP: 1) composed of uranium (U) and oxygen (O), and 2) composed of U, O and zirconium (Zr). The last type of particles accounts for significant part of the activity inventory. The microphotographs showed that U-O particles have significant secondary porosity due to chemical weathering, while Zr-U-O particles apparently demonstrate chemical stability. Thus, some portion of activity in the trench “bound” to Zr-U-O particles is practically unavailable for hydrology migration and bio-uptake process. Therefore, to model releases of radioactivity from the waste, a multi-component source model should be used with different dissolution kinetic coefficients for different types of FP.

Based on new knowledge and for purposes of further modelling analyses we sub- divide fuel particles (separate grains and crystallites of uranium oxide of a median diameter of 4-6 μm) and their conglomerates in the trench no.22 into 3 types:

I. Oxidised fuel particles (UO_{2+x}), which are relatively chemically low stable, and were formed as a result of oxidisation of nuclear fuel in a period 26.04.86-5.05.86.
II. Non-oxidised fuel particles (UO_2) of the first release on 26.04.86, which are relatively chemically stable, and were formed as a result of mechanical destruction of nuclear fuel.
III. Chemically extra-stable presumably uranium-zirconium matrix particles (U-Zr-O), which were formed as a result of high-temperature annealing of UO_2 in presence of zirconium in Unit 4 construction materials

It should be noted, that subdivision of the fuel particles into the three above types is conditional, because no clear borders (i.e., based on degree of nuclear fuel oxidation and zirconium content) can be drawn between various types of particles. From the fact that the CPS is located at the narrow Western fuel trace of the radioactive fallout, which was mainly formed at the initial moment of the accident on 26.04.86, it can be expected that radioactivity of waste in the trench no.22 to a great extent is caused by

UO_2 and U-Zr-O fuel particles. Based on the literature data of Bobovnikova *et al*. [11] and model experiments on mechanical destruction and annealing of irradiated nuclear fuel by Kashparov *et al*. [10], it can be deduced that only about 2±1% of initial ^{90}Sr inventory in the fallout was in mobile (condensed fraction) form.

In order to further characterize contributions to the source-term and dissolution rate coefficients for different FP types, purposeful studies were carried out in 2000-2001 which included: (1) chemical extraction procedures on waste samples, and (2) laboratory fuel particle dissolution experiments.

Fraction of ^{90}Sr activity associated with chemically extra-stable particles was estimated via application to a set of 200 g waste samples from the trench no.22 repeated extraction procedure involving 2-hour heating in 6M HNO_3. Results of this experiment are shown in Table 1. It was established also, that the leaching residue material is significantly depleted in ^{137}Cs in comparison with ^{90}Sr, which may be indicative of a high temperature of formation of this material (particles). Apparently residue may be composed of U-Zr-O matrix particles, because a high temperature is needed for U-Zr-O melt formation, which explains leakage of volatile ^{137}Cs. It can be concluded, that the ^{90}Sr activity fraction in waste associated with the U-Zr-O particles is about 20±10%.

Table 1. ^{90}Sr activity in the residue after two stages of waste samples leaching in heated 6M HNO_3.

Sample no.	Activity fraction in the residue after the 1 stage of leaching, %		Activity fraction in the residue after the 2 stage of leaching, %	
	m	σ	m	σ
1	35	2	27	2
2	12	2	09	1
3	19	1	14	1
Average	22	12	17	9

One other issue addressed by waste leaching experiments was estimation of the present (year 2001) fraction of non-dissolved FP (including FP with UO_2 and UO_{2+x} matrix) in waste material. For this purpose the sequential extraction procedure developed earlier by Kashparov *et al.* [5] for contaminated soils in Chernobyl zone was applied, utilizing 1 M ammonia acetate solution (for exchangeable ^{90}Sr) and 6M HNO3 extraction (for total "soluble" ^{90}Sr, including UO and UO_{2+x} matrix FP). Few changes were made to the initial protocol in order to account for the presence of non-dissolved U-Zr-O particles. The resulting data on chemical speciation of ^{90}Sr activity in waste are as follows (for 2001): 36±13% of activity is in mobile (exchangeable form), while 64±13% is associated with fuel particles with different matrix (UO_2, UO_{2+x}, U-Zr-O).

To parameterise source-term model, information is needed on kinetic dissolution rate coefficients for different FP types. For the U-Zr-O particles it can be assumed that the relevant rate coefficient is zero (α_3=0). Based on previous work [10], rate constant for UO_{2+x} particles is expected to be one order of magnitude higher than for UO_2 particles (dissolution period of UO_{2+x} particles in the trench conditions, at pH of soil solution ≈5, is about 2.5 years). Therefore, at present moment (15 years after the waste disposal), practically all UO_{2+x} particles presumably have already dissolved, and FP in the trench are represented only by U-Zr-O and UO_2 particles. Dissolution of UO_2 particles is comparatively slow process, and it can be hardly estimated during a laboratory test. Therefore, decision was made to dissolve waste samples in more aggressive conditions (at pH=3-4) accelerating the fuel particles transformation. Then obtained dissolution parameters were extrapolated back to the natural (trench) conditions, using data on the real trench pH and empirical dependence of FP dissolution rate constant from pH [10]. Studies were carried out on waste samples from 3 trench locations, using 100 g mass aliquots in two replications. Before leaching, the mobile ^{90}Sr was washed away from waste samples by 2M ammonia acetate solution. The duration of waste dissolution experiment at pH=3 to 4 was 216 days. The resulting dissolution rate coefficients for fuel particles from the trench (Fig.3) are in good agreement with dissolution rate parameters of real UO_2 particles of irradiated Chernobyl nuclear fuel in simulated conditions [10]. Therefore, using of FP transformation rate coefficients obtained in our previous model experiments on fuel particles of different oxidation state (UO_2 and UO_{2+x}) [10] is justified. Corrections for the real acidity of the soil solution in the trench (pH ≈5) provided estimates of dissolution rate coefficients for the fuel particles in the trench (Table 2).

The presented above data on inventory of ^{90}Sr inside the trench for the year 2001 in U-Zr-O particles, and in mobile (exchangeable) form, and on dissolution rate coefficients for different components of the fallout, allow us to estimate using simple analytical developments contributions of

different types of fallout components to the initial (for year 1986) activity of ^{90}Sr in waste inside the trench. The resulting parameters are summarized in Table 2.

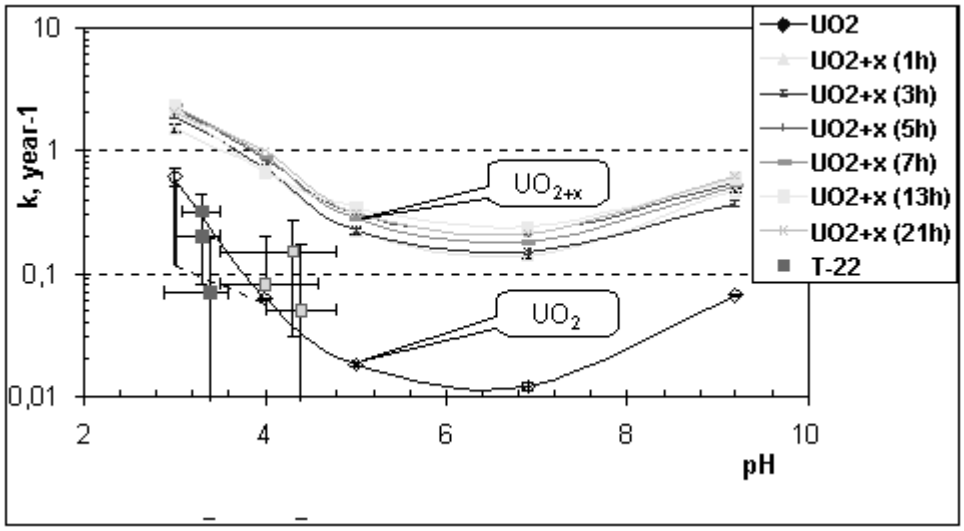


Fig.3. Results of model experiments on dissolution of nuclear fuel particles (UO_2 and UO_{2+x}, obtained by annealing in air of UO_2), and of waste material from the trench no.22.

Table 2. Estimated dissolution rate constants and contributions to initial trench ^{90}Sr activity inventory for fuel particles of different type.

Type of fuel particle	Dissolution rate coefficient, y^{-1}	Contribution to the initial trench inventory (1986), %
UO_2	0.018±0.001	57±15
UO_{2+x}	0.28±0.05	21±15
U-Zr-0	≈0	20±10
Condensation component	∝	2±1

3.2. Infiltration flow regime

Radionuclide migration in the trench body and unsaturated zone is driven by flow of infiltrating meteoric water. Therefore characterization of water fluxes in unsaturated soil profile is important pre-requisite for quantification and modeling of radionuclide releases to from the waste site to the surrounding geo-environment.

To characterize infiltration flow regime at CPS we have used “hydro-physical” method developed by Sytnikov [12]. The method is based on direct measurements of moisture pressure in the soil profile using tensiometer sensors, and subsequent calculation of water flux using Darcy law for unsaturated media:

$$q_{i+1/2}(t) = K(\psi_{i+1/2})\frac{h_{i+1}(t) - h_i(t)}{z_{i+1} - z_i}, \quad h_i(t) = z_i + \psi_i(t)$$

$$\varepsilon = \int_o^T q_{i+1/2}(t)dt$$

where z_i is depth of tensiometer sensor “*i*” installation (m); ψ_i is soil moisture suction pressure head at point “*i*”(m of water column); h_i is total hydraulic head at point “*i*” (m of water column); $K(\psi)$ is hydraulic conductivity of unsaturated soil (m/day); $q_{i+1/2}$ is moisture flux through soil section at depth $z_{i+1/2}$= 0.5 (z_i+z_{i+1}) (m/day); ε is cumulative infiltration recharge flux during (m of water column) the time t (day).

Monitoring data on distribution of soil moisture suction pressure in waste site profile were provided by the specially designed pit, constructed at CPS in June 2000 (Fig.4). One side of the pit partially cuts the body of the trench with radioactive waste, while the opposite side provides access to soil profile outside of the trench. Such an arrangement of the pit allows parallel study of processes of moisture flow in the body of the trench and outside the trench in the undisturbed soil profile. The pit is

equipped by arrays of sensors for measuring moisture content, suction pressure and soil temperature, and data logger to automatically store sensor readings (see Fig.4).
The other parameter, which is necessary to apply formula (3.1), is functional dependence of hydraulic conductivity of the unsaturated soil from suction pressure - $K(\psi)$. This functional characteristic for CPS soil was determined in laboratory column experiment using "steady state" method [13]. Derived using the described above input data and parameters information on recharge regime of Pilot Site for the period from 1.11.2000 to 1.10.2001 is shown at Fig.5.

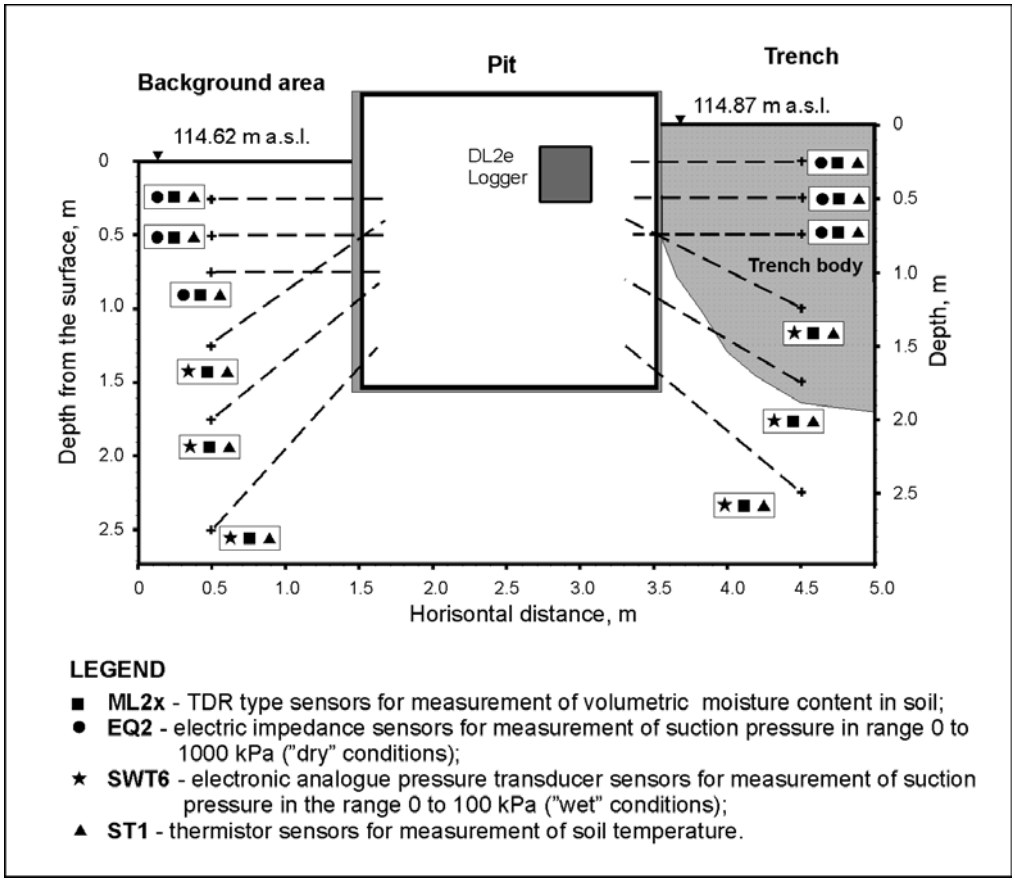


Fig. 4. Cross-section scheme of the pit for unsaturated zone monitoring.

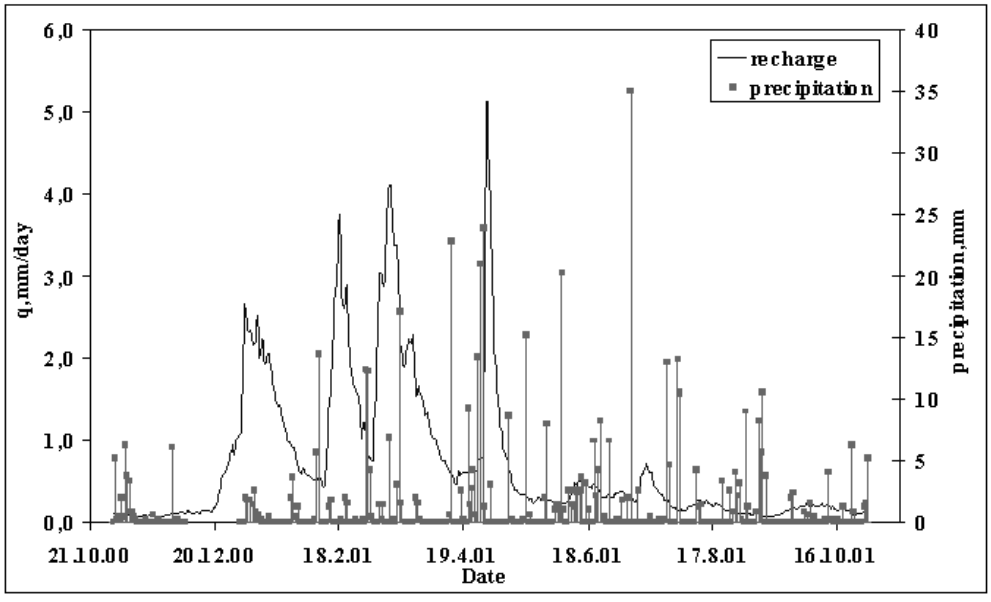


Fig.5. Infiltration recharge regime at Pilot Site for the period from 1.11.2000 to 1.10.2001.

The cumulative infiltration recharge during 12 month is ≈260 mm or ~40% of precipitation. It is clearly seen that the meteoric water inflow to the aquifer is an event-based process, where most recharge occurs during winter thaws and spring snowmelt, and occasionally by large rainstorms during the summer-autumn period. During the rest of the year the rainfall mostly is consumed by evapotranspiration process.

3.3. Sorption parameters for the trench body

Once radionuclide was leached from fuel particles, and has entered the ambient waste-soil system, one of key parameters governing radionuclide release rate from the waste burial is radionuclide distribution coefficient (Kd) of the waste-soil matrix [14]. To determine waste matrix K_d we have conducted column experiments in dynamic conditions using ^{85}Sr isotope label (tracer solution). The methodology offers number of advantages. During each experiment on waste material several research questions were addressed, including: ^{85}Sr tracer Kd (main focus of this paragraph); hydraulic parameters of waste; and leaching behavior of waste (regarding ^{90}Sr, ^{137}Cs; this last issue will be considered elsewhere).

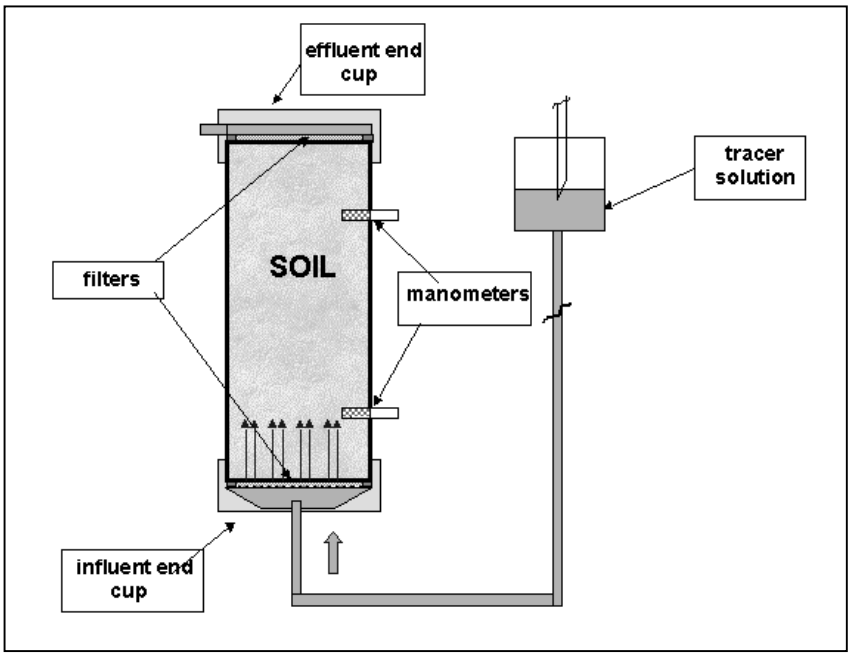
Scheme of the column apparatus (10 cm length and 5.2 cm diameter plastic tube) is shown in Fig.6-a. Hydraulic head gradient through the column has been controlled by manometers. The 2-L bottle with a Mariotte siphon was used as a tracer reservoir. The chemical composition of synthetic tracer carrier solution was close to the soil solution, which was collected at 1.5 m depth in the trench by ceramic moisture sampler: [Ca]=10 mg/l, [Sr stable]=2 mg/l, and pH=5.

Column experiments were carried on 3 waste-soil samples (sample no.1 in 2 replications) collected at the different locations of the trench no.22. The bulk density of samples was 1.66-1.71 g/cm^3; porosity was 0.33-0.36. Interpretation of experimental data was done using classical advection-dispersion model with K_d-dependent retardation coefficient. The effective ^{85}Sr K_d was determined by adjusting the retardation factor in a such way, that the C/C_0=0.5 of simulated curve matched that of measured curve (Fig.6-b). Results of column experiments are summarized in Table 3. Radiostrontium distribution coefficient for waste material (soil) fall into the range from 0.8 to 4.5 ml/g with an average value of ≈2 ml/g. Variations in Kd reflect significant heterogeneity of waste material.

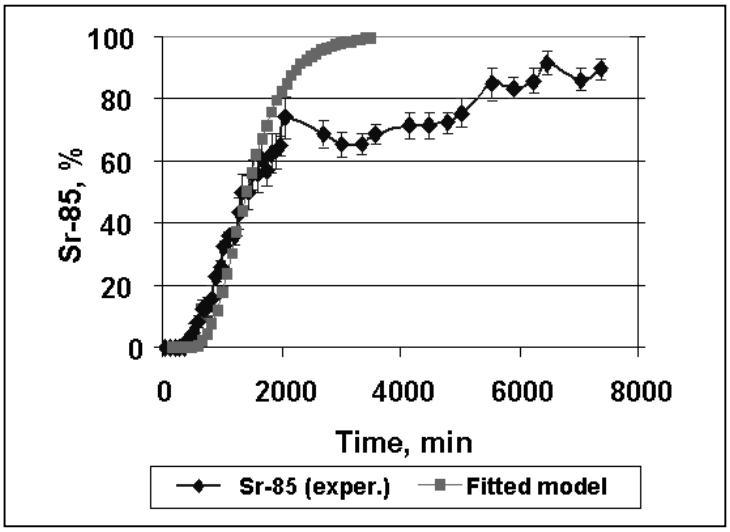
The trench Kd values compare with the determined earlier Kd-s for the eolian sand from Pilot Site hosting the trench no.22, i.e ≈1-3 ml/g by different methods [3]. However, it can be expected that the effective radiostrontium Kd for the waste matrix on average should be somewhat higher than for the eolian sands due to higher content of the clay particles and organic matter in the trench material compared with surrounding geology sediments [2]. The cation exchange capacity (CEC) of eolian sands at CPS based on large statistics on average is 0.7 meq/100g, while waste matrix has CEC of 1.2 meq/100g.

Table 3. Effective parameters of ^{85}Sr transport in column experiments on waste material.

Soil sample	Effective ^{85}Sr transport velocity, cm/s	^{85}Sr K_d, ml/g
1(a)	$6.8 \cdot 10^{-4}$	1
1(b)	$2.9 \cdot 10^{-4}$	0.8
2	$4.6 \cdot 10^{-5}$	4.5
3	$1.2 \cdot 10^{-4}$	1.6



(a) - Setup



(b) - Typical ^{85}Sr effluent curve

Fig.6. Column experiments on waste material.

4. SOURCE TERM MODELING

4.1. Radionuclide migration model

The hydrogeology, geochemistry and radiochemistry conceptual understanding of the waste site and parameters described in previous paragraphs were integrated into the model for the trench source term of radionuclide migration to the geo-environment. The modeling work pursued following of objectives:

- to check "compatibility" and consistency of derived hydrological and radiochemical parameters (infiltration recharge rate, soil K_d, data on ^{90}Sr chemical forms in waste, etc.)
- to reproduce via modeling independents field characterization and monitoring data (i.e., carry out model "confirmation" exercise);
- to assess long-term radionuclide leaching dynamics from the trench (e.g., for safety assessment purposes).

The modeling approach for radionuclide transport in the trench body and unsaturated zone is based on advection-diffusion equation with time-constant coefficients. This approach is most suitable for long-term time-averaged source term modeling as a part of global model for the waste site. The distinctive feature of the model is that it accounts for various types of fuel particles with different dissolution rates composing the radioactivity source-term. The mathematical model encompasses the unsaturated zone from the ground surface to the groundwater level including the trench body (Fig.7). Model includes the body of the trench ($0 < Z < L_{tr}$; L_{tr} is trench depth), and unsaturated zone below the trench ($L_{tr} < Z < L_{gw}$; L_{gw} is depth to groundwater level). The model schematization assumes that moisture flow in the trench body and underlying unsaturated zone is one-dimensional (downward) and steady state. The lower boundary of the model corresponds to the average position of the groundwater level.

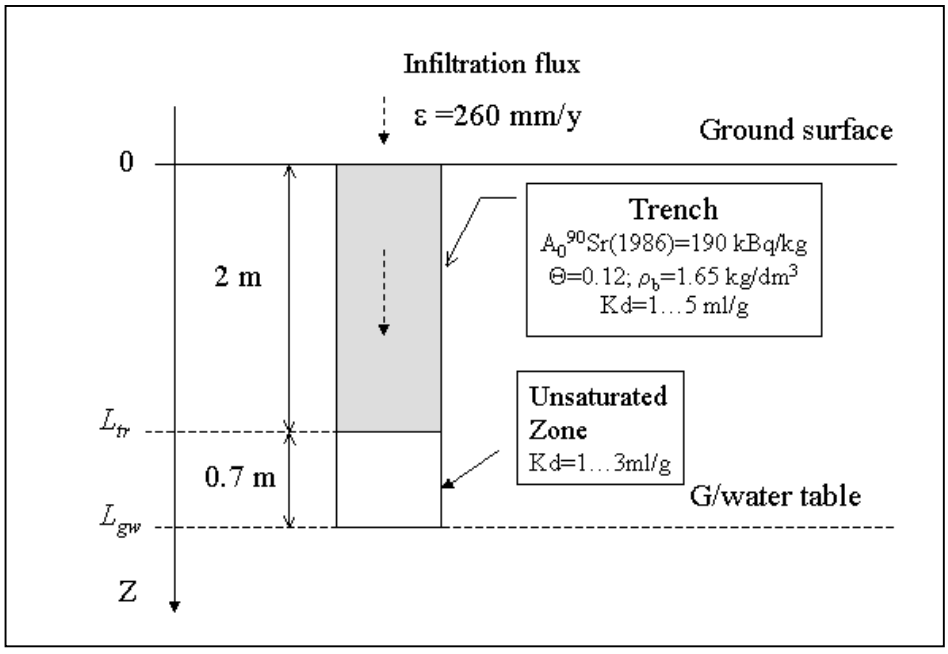


Fig.7. Source term model schematization.

The main equation of the model describing evolution in space and time of the concentration of radionuclide in soil porous solution is as follows:

$$\frac{\partial C}{\partial t} = D\frac{\partial_2 C}{\partial z_2} - \frac{V}{\theta R(z)}\frac{\partial C}{\partial z} - \lambda C + \frac{F(t,z)}{\theta R(z)}\ ,\ R(z) = 1 + \frac{\rho}{\theta}K_d(z)$$

where C is radionuclide concentration in porous solution (Bq/dm^3); Θ is volumetric moisture content in unsaturated zone (dimensionless); D is effective dispersion coefficient (accounting for sorption) (m^2/day); V is infiltration recharge rate (m/day); λ is radioactive decay coefficient (day^{-1}); $F(t,z)$ is source term which describes process of radionuclide leaching from fuel particles inside the trench (Bq/(dm^3 day), see below); $R(z)$ – is sorption retardation coefficient (dimensionless; see below); $\rho(z)$ is soil bulk density

(kg/dm^3); $K_d(z)$ is varying in space sorption distribution coefficient of radionuclide (dm^3/kg); z is space coordinate (m; see Fig.7); *t* is time (day).

The process of radionuclide leaching from fuel particles in waste material is described by following equations:

$$F(t,z)=\sum_{i=1}^{3} f_i(t,z), \quad f_i(t,z)=\alpha_i M_i(t,z),$$

$$M_i(t,z)=\begin{cases} M_{i,0}\exp(-(\lambda+\alpha_i)t), & 0<z<L_{tr} \\ 0, & L_{tr}<z<L_{gw} \end{cases}; \qquad M_{i,0}=E_i A_0$$

where $F(t,z)$ is integral flux of activity per unit volume of porous medium (waste material) due to dissolution of FP (Bq/(dm^3 day)); $f_i(t,z)$ is flux of activity per unit volume of porous medium (waste material) due to dissolution of FP of type "*i*" (Bq/(dm^3 day)); $M_i(t,z)$ is concentration in trench of FP of type "*i*" at time *t* (Bq/dm^3); α_i is the dissolution rate coefficients of FP of type "*i*" (day^{-1}); $M_{i,0}(t,z)$ is concentration in trench of FP of type "*i*" at initial time *t=0* (Bq/dm^3); A_0 integral specific activity (in all chemical forms) of radionuclide in waste at initial time *t=0* (Bq/dm^3); E_i is fraction of radionuclide activity associated with FP of type "*i*" at initial time *t=0* (fraction of unit, dimensionless).

The model assumes that 3 different types of FP characterized by different dissolution rate compose the source-term (see §3.1 for detail): *i*=1 – oxidized (annealed) FP with matrix from UO_{2+X}; *i*=2 – non-oxidized FP (UO_2 matrix); *i*=3 – hot particles with U-Zr-O matrix. Fractions of described above types of FP in integral inventory of activity in waste at initial time *t=0* (A_0) are defined by coefficients E_1, E_2 and E_3 (fractions of unit). In addition, it is assumed that at initial time *t=0* waste contained fraction E_0 of radionuclide in mobile (ion-exchangeable) form. It is obvious that the following equation holds:

$$E_0 + E_1 + E_2 + E_3 = 1.$$

Initial and boundary condition for equation are written as follows:

$$C(0,z)=\begin{cases} C_0, 0<z<L_{tr} \\ 0, \quad L_{tr}<z<L_{gw} \end{cases}, \qquad C_0=\frac{E_0 A_0}{\Theta R};$$

$$\left(D\frac{\partial C}{\partial z}(z,t)-\frac{V}{\theta R}C(z,t)\right)\Big|_{z=0}=0, \qquad D\frac{\partial C}{\partial z}(z,t)\Big|_{Z=Lgw}=0$$

The described above boundary value problem was solved numerically using explicit finite-difference scheme and programmed in Fortran language.

4.2. Model confirmation

The evaluated modeling scenario assumed that hydrogeology and radiochemistry parameters were representative of the "average" trench conditions. The initial activity of ^{90}Sr in waste was calculated based on the trench inventory of 2.9×10^{11} Bq (for year 2000) and volume of waste of 1300 m^3 [3]. Initial distribution of ^{90}Sr activity between fuel particles belonging to different categories (UO_2, UO_{2+X}, U-Zr-O) and respective dissolution rate constants were based on data of §3.1 of this paper. Flow system geometry (L_{tr}= 2 m, L_{gw}-L_{tr}= 0.7 m) corresponds to average position of groundwater level relative to trench bottom during last 10 years [2]. Infiltration recharge rate estimates were based on studies presented in §3.2. Of particular interest is selection of representative ^{90}Sr sorption distribution coefficient values (K_d-s) for the trench matrix and unsaturated zone sediments below the trench. The K_d is known to be a very sensitive parameter with regard to model output. As Kd estimates vary for the trench material from ≈1 to 5 ml/g (see §3.1), and for the eolian sand below the trench from 1 to 3 ml/g [2,3], we have carried sensitivity analysis of model results to Kd values.

A number of "target" parameters exist at present time (year 2001) in order to evaluate performance ("validity") of the model. Amount of ion-exchangeable ^{90}Sr in trench material is estimated by various experimental method at ~ 30±5% (see§ 3.1). Release of ^{90}Sr outside the trench to the unsaturated zone and aquifer was estimated at 7±5% using data on ^{90}Sr/^{154}Eu ratio in trench material and comparing with Chernobyl Unit 4 fuel ratio [3]. Alternative methodology (integration of ^{90}Sr groundwater concentrations data) results in estimated radionuclide release to the aquifer of 1.5-4.5% (unpublished 2002 data by the IGS). (The last estimate does not account for ^{90}Sr in unsaturated zone). Lastly, arrays of monitoring data exist on ^{90}Sr concentrations in trench porous solutions and groundwater below the trench [2,3], the "typical" trench – averaged value being ≈10±5 kBq/L.

In the course of the numerical experiment model output was checked against the validation criteria described above. Modeling scenarios, output and target parameters are described in Table 4. In scenarios I – III soil Kd value was systematically increased from 1 to 3 ml/g. For simplicity, same sorption properties were assumed for the trench material and for unsaturated zone sediments underlying the trench. It can be clearly seen (Table 4), that for K_d=1 ml/g we get over-depletion of trench by mobile ^{90}Sr (only 14% instead of ~30% from field data) and respectively over-estimation of amount of ^{90}Sr in the unsaturated zone and aquifer outside the trench (25% instead of 7%). Also, ^{90}Sr concentration at outflow boundary exceeds the expected value (37 kBq/L instead of 10±5 kBq/L). The value of K_d=2 ml/g results in better fit for proportion of exchangeable ^{90}Sr inside the trench (25%), however amount of ^{90}Sr outside the trench is still higher than it should be (13 %), and the same is true for predicted ^{90}Sr groundwater concentration at outflow boundary (~14 kBq/L). Lastly, with K_d=3 ml/g we get nearly perfect fit (29%) for proportion of mobile ^{90}Sr in trench, but radionuclide release to the aquifer is clearly underestimated (0.8% instead of 1.5-4.5%).

The above numerical experiment shows that single Kd value for the trench material and eolian sand in unsaturated zone is not capable to provide perfect fit with all model validation criteria. In fact, we can expect that trench material is characterized by higher sorption value compared to eolian sand (see §3.3). Therefore in the last evaluated scenario (scenario IV; Table 4) we have used Kd=4 ml/g for the trench and Kd=2 ml/g for the unsaturated zone sand. As a result, modeling provides a quite satisfactory fit with experimental data for all criteria.

Table 4. Source term model sensitivity analyses with regard to ^{90}Sr K_d values.

	Input parameters		Output parameters (for 2001; t=15 years)				
Scenario	Trench Kd, ml/g	Unsatur. zone Kd, ml/g	Amount of exch. ^{90}Sr in trench, % activity	Amount of ^{90}Sr in unsat.zone, % tot. inv.	Amount of ^{90}Sr in aquifer, % tot. inv.	Total ^{90}Sr outside trench, % tot. inv.	^{90}Sr outflow conc. , kBq/L
I	1	1	13.6	10.8	14.4	25.2	37
II	2	2	25.4	10.1	3.2	13.3	13.9
III	3	3	28.9	8.2	0.8	9	4.7
IV	4	2	30.6	5	1.6	6.6	6.8
Target value			**30±5**	**-**	**1.5-4.5**	**7±5**	**10±5**

An illustrative long-term (300 years) time dynamics of ^{90}Sr concentration in leaching solution from the trench is shown at Fig.8. The ^{90}Sr concentration in porous solution decreases to the level of 2 Bq/L (Ukrainian drinking water standard for ^{90}Sr) in approximately 240 years. This type of modeling result is of interest from the perspective of safety assessment of “Red Forest” waste dumps, and demonstrates the time scale of radiological significance of the waste dump problem.

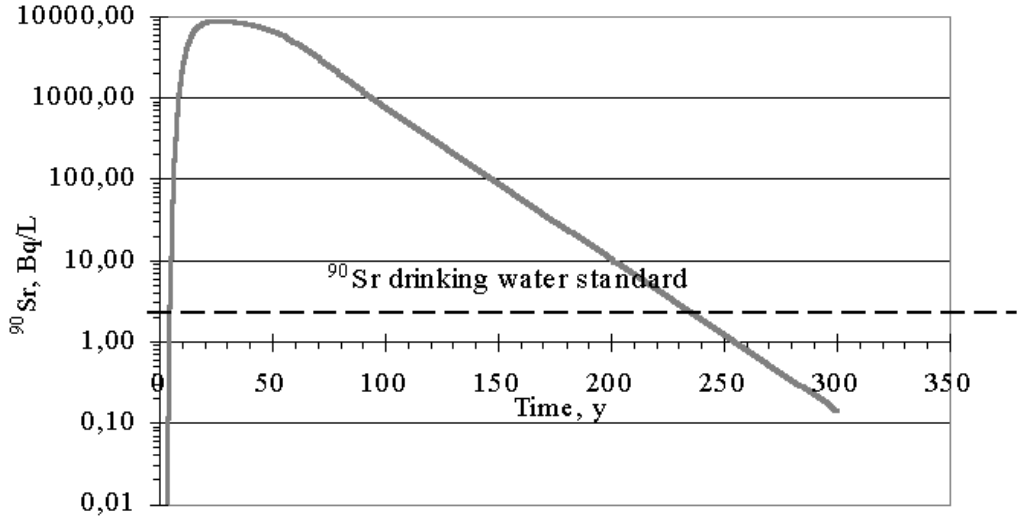


Fig.8. Long-term dynamics of 90Sr concentration in leaching solution from the trench (scenario IV; Table 4).

5. CONCLUSIONS

At the beginning of the Pilot Site project the project team already disposed site database from previous works by Ukrainian institutes. It was believed that the site is relatively “simple” in terms of hydrogeology and radiology conditions, and thus quite suitable for model validation exercises [1]. The above considerations, in fact, have governed selection of the site as an experimental ground for future studies.

However, the early conceptual model of the site has undergone in the course of the project essential development and revision. This concerns both hydrogeology and radiochemistry understanding of the site. For example, the groundwater flow regime in the aquifer appeared to be transient and fully 3-dimensional [2,3] (i.e., much more complex contrary to early 1-D steady state schematizations). As discussed above, the projected has lead, also, to a significant evolution of understanding of the Chernobyl radionuclide migration source term (i.e., dispersed Unit 4 nuclear fuel particle characteristics and dissolution behavior). This example evidences that concepts based on sparse data sets and heavily relying on expert judgment, rather than careful in-depth multidisciplinary research, are a vulnerable and rather unreliable basis for radionuclide transport and safety assessments in nuclear waste disposal practices.

Application of the presented above Chernobyl radionuclide fallout source-term model to the Pilot Site data set can be considered quite satisfactory and encouraging. When combined into the comprehensive transport model, radiochemistry and hydrogeology parameters for the CPS appeared to be consistent with each other and with independent characterization/monitoring data. The modeling approach has proved to be capable of rather accurately reproducing field characterization and monitoring data relevant to ^{90}Sr speciation inside the trench and radionuclide releases from the trench to the aquifer. Model provides useful tool for safety assessment of “Red Forest” waste dumps. The developed knowledge and modeling approach are of general value for radiological assessments of Chernobyl contaminated areas (especially in the near-zone of the ChNPP), and for possible similar accidental situations in future.

Lastly, the project has identified a number of issues, where current understanding is insufficient, and future radioecology research is warranted, in particular: - transient effects of water regime on unsaturated soil solution geochemistry and sorption interactions (the last aspect is of particular importance for radionuclides with more complex chemistry, compared to ^{90}Sr, e.g. actinides); - influence of geo-chemical evolution of organics-rich source term (e.g., due to oxidation process) on radionuclide release rate from waste site; - seasonal temperature and biological effects on soil solution pH and on dissolution behavior of fuel particles, etc.

It should be stressed that the presented data and analyses can be considered no more than a *partial* confirmation of the Chernobyl radioactivity source-term model (as models in earth sciences “can not be ever fully validated”, because of non-uniqueness of solutions, possible compensating errors, unknown future stresses on the modeled system etc. [15,16]; see also unresolved issues outlined in the previous paragraph). Nevertheless, we believe that the Chernobyl Pilot Site project has resulted in a substantial progress in understanding of process governing radionuclide releases and subsequent migration from the Chernobyl fuel particles, and thus serves to increasing the confidence level in radionuclide transport assessments from nuclear fuel source term to soils and geosphere. The developed experimental techniques on fuel particles characterization, dissolution rate measurement, chemical interaction in soils, as well as relevant parameter databases and models could serve in environmental impact assessment in case of accidental contamination by nuclear materials, or resulting from use of depleted uranium ammunitions, or terrorist “dirty nuclear bomb” attack.

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