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Validation of the global model for ^{90}Sr migration from the waste burial in the Chernobyl exclusion zone

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Abstract. The reported work aimed at modeling process of radionuclide migration from the near-surface waste burial (trench no.22) containing nuclear fuel particles to the unsaturated zone and aquifer. The developed global model of the waste site accounts for dissolution of reactor fuel particles, vertical radionuclide transport in the unsaturated zone, and horizontal transport in the underlying aquifer by advection-dispersion mechanisms. It is based on the following sub-models: the geostatistical model for radioactivity distribution in trench; the radionuclide source term model describing dissolution of fuel particles, radionuclide redistribution in the trench body and unsaturated zone (STERM1D) and the two- and three-dimensional contaminant transport in the aquifer (MODFLOW – MT3D). Modeling work included sensitivity analyses, calibrations and eventually validation tests for the global model by means of comparison of model predictions to experimental data. In addition, a preliminary step by step validation process was applied to "elementary" sub-models. The developed modeling methodology is mostly suited for producing spatially averaged parameter values (such as radionuclide concentrations in trench porous solution, integral radionuclide release to aquifer) and is applicable for long-term predictions on a scale of decades assuming that hydrological and geochemical conditions remain steady state.

1. INTRODUCTION

The waste dump sites created in 1986-87 during emergency clean-up activities at the Chernobyl Nuclear Power Plant (ChNPP) contain about 10^6 m^3 of low-level wastes [1]. These waste dumps largely do not satisfy regulatory requirements for low-level waste disposal facilities and pose radiological risks to the environment. Of particular concern is hydrogeologic migration of ^{90}Sr, which is known to be mobile in soils and groundwater systems. As pointed out in the appraisal by the Nuclear Energy Agency Committee on Radiation Protection and Public Health [2] "… all these waste are potential source of contamination of the groundwater which will require close monitoring until safe disposal into appropriate repository is implemented", and that "… large uncertainties remain which require a correspondingly large characterization effort".

During the period of 1999-2003 the behavior of Chernobyl fallout radionuclides has been studied jointly at the experimental site in the "Red Forest" waste dump area near the ChNPP by an international team of French and Ukrainian institutes (see author affiliations). Established in the course of the project Chernobyl Pilot Site (CPS) provided an experimental field facility for in-situ confirmation (validation) and development of theoretical models for radionuclide migration in soils and the geosphere. In addition, the CPS project was of prime importance for Ukrainian partners from the perspective of practical risk assessment and waste site remedial analyses.

The project comprised three stages: (1) site characterization; (2) development of the migration model for the waste site (from fuel particles source term to geosphere), and (3) carrying out global model validation tests [3-4-5].

The prerequisite to constructing the global model of the waste site was development of the two principal sub-models: the structural model for radioactivity distribution in the trench no.22; and the radionuclide source term model describing dissolution of fuel particles and radionuclide redistribution in the trench body and unsaturated zone [6-7]. In the following we will briefly review the approach and the main results on sub-model validation tests and on global modeling exercise.

2. APPROACH TO GLOBAL MODELING

The studied waste burial (trench no.22) represents ~70 m long, 8-10 m wide and 2-2.5 m deep unlined trench containing site clean-up wastes. The source term of radionuclide migration to the geo-environment is a heterogeneous mixture of contaminated organic materials (decomposed vegetation) and soil containing micron-size reactor fuel particles. The specific activity of ^{137}Cs and ^{90}Sr in waste is n*10^5- n*10^6 Bq/kg. The geologic section of CPS consists of the sequence of layers of Quaternary eolian and alluvial sands. The depth to groundwater table is 2-3 m (Figure 1).

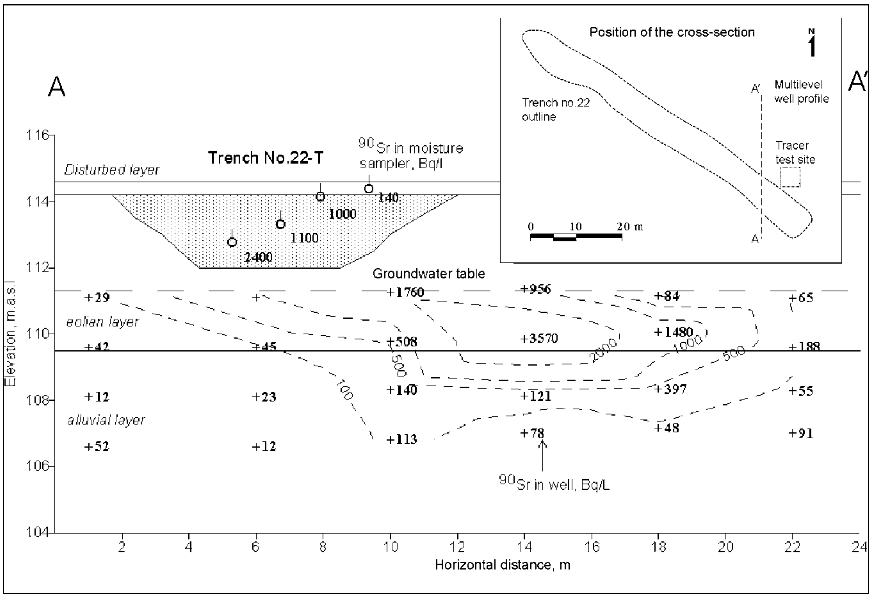

Figure 1. ^{90}Sr distribution in cross-section of the aquifer at the CPS in June 2002 (analytical error 10-20%). A-A' is the line of hydrogeologic section.

During the nearly two decades following the waste disposal, dissolution of fuel particles has occurred, radionuclides have been released from the uranium oxide matrix of particles and redistributed to soil matrix and trench porous solution. Radionuclides in ion-exchangeable form have been leached subsequently 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 of above process, ^{90}Sr concentration in groundwater in the upper part of the aquifer in the vicinity of the trench at present time varies between n*100 and n*10,000 Bq/l, and the ^{90}Sr plume has spread some 10 m downstream from the source. Detailed data on hydrogeology conditions of the study site, radioactivity distribution in the trench no.22, and on physical and chemical characteristics and dissolution parameters of fuel particles can be found in [8-9].

The waste site can be conceptually represented as a system incorporating three major compartments: the trench body containing dissolving fuel particles, the unsaturated soil zone below the trench down to the groundwater table, and the aquifer underlying the waste site system. For each of the listed compartments, the purposeful characterization program has been carried out aimed at determination of key hydro-physical and geo-chemical parameters [8-9].

The global model for radionuclide transport from waste site to geosphere for the Chernobyl Pilot Site incorporates the following "elementary" sub-models (Figure 2):

I. Structural model for initial radionuclide activity distribution (^{137}Cs, ^{90}Sr) in trench in 3D. The relevant geostatistical model used kriging interpolation technique [6].

II. Model for dissolution of fuel particles in waste material and subsequent vertical one-dimensional radionuclide transport by advection-dispersion in the trench body and unsaturated zone below the trench. The source term model was implemented in the computer code STERM1D [7].

III. Model for water flow and radionuclide transport in the aquifer. To predict radionuclide transport in the aquifer we utilized several models of a different level of complexity: 1D (using specially written computer code), and 2D-3D advection-dispersion model [10] utilizing MODFLOW–MT3D groundwater modeling software [11-12].

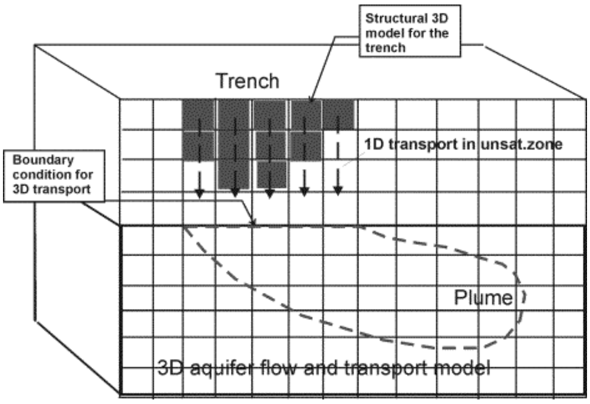

Figure 2. Schematic representation of the numerical realization and data exchanges of the global radionuclide transport model for Chernobyl Pilot Site.

Model **I** was used to estimate initial distribution of ^{90}Sr in trench at time t=0 [6]. This is a fully 3D model, accounting for the heterogeneity of initial distribution of radioactivity within the trench. Model **II** was used to calculate radionuclide activity as a function of time in infiltration water entering the aquifer below the trench. This is a one-dimensional model. The 1D transport calculations were carried out for each column of the 3D numerical grid, describing initial distribution of radioactivity within the trench. The output of model **II** (radionuclide concentration in infiltrating water out-flowing from the unsaturated zone to groundwater table) served a boundary condition for the model **III**.

3. MAIN RESULTS AND DISCUSSION

3.1 The structural model for radioactivity distribution in trench 22 using kriging technique

We applied the geostatistical analysis [13] of the data set of ^{137}Cs field measurements to develop a structural model of ^{137}Cs distribution in trench in nodes of a regular grid in the 3D space. We then estimate ^{137}Cs values and then correlated ^{90}Sr activities [6].

A number of preliminary data treatment procedures (including filtering of peripheral points and logarithmic transformation) carried out on the initial data set of ^{137}Cs activity measurements (Acs) [14] in trench no.22 have lead to approximately symmetric and normally distributed data set.

Geostatistical analyses using variogram function have established that radioactivity distribution in trench no.22 was characterized by regular spatial correlation patterns. The $\log_{10}(Acs)$ data set was found to be essentially anisotropic. The correlation length along the trench (X) is approximately 15-20 m while it is approximately 4 m across the trench (Y). For the vertical direction (Z) the asymptotic sill for relevant variogram was not reached, as the trench had limited depth (less than 3 m). However, experimental variogram for Z direction showed correlation pattern generally similar to X direction. By scaling in directions of X and Z axis the $\log_{10}(Acs)$ data set was rendered approximately isotropic and the spatial structure of the data set was described by a spherical model variogram.

The adequacy of the proposed structural model for ^{137}Cs distribution in the trench no.22 in 3D was checked in the course of a validation exercise which compared results of kriging interpolation (Figure 3) to the independent cross-validation data set [6]. The relevant cross-validation statistical criteria were satisfied. In all cases cross-validation points fitted with predicted 95% confidence intervals. When back transformed to original values of ^{137}Cs activity, actual kriging errors translated to the average interpolation error of less than 30%.

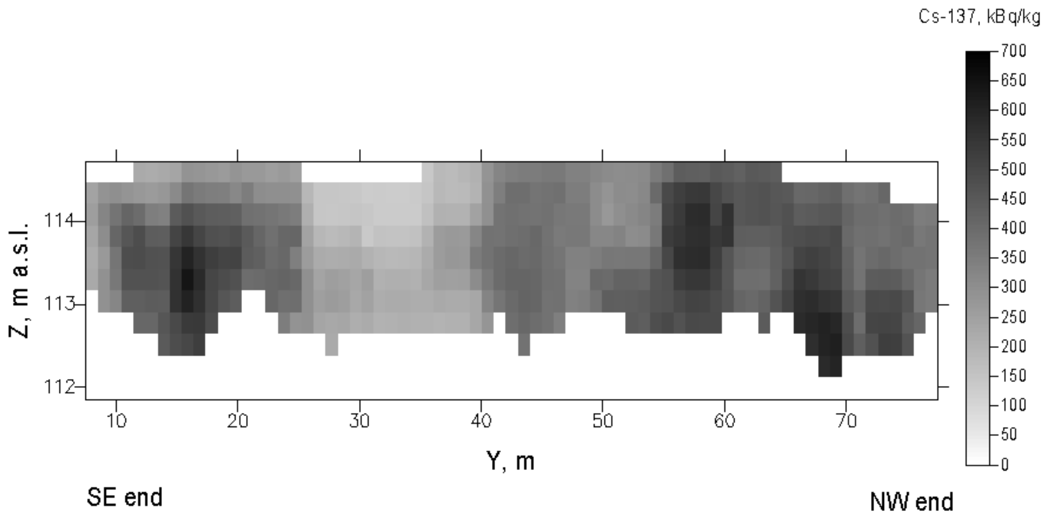

Figure 3. Kriging interpolation of radioactivity distribution in a vertical slice along the trench no.22.

3.2 Validation tests for the model describing the ^{90}Sr migration source-term

Validation tests for the source-term model consisted in comparison of model predictions with independent field observations. Calculations were carried out within a probabilistic framework using the Monte-Carlo method [7], accounting for uncertainties in migration parameters.

The first validation test consisted in modeling of ^{90}Sr concentration in trench porous solution for t=15 y since waste disposal (Figure 4). Validation data set corresponded to selected ^{90}Sr concentrations profiles in soil water samplers (SWS) installed to the trench no.22. Predicted by radionuclide migration model confidence intervals (p=0.9) for radionuclide concentration in SWS agreed with observation data for 9 of 10 data points. The failure for one point could be explained by variability in observation data [7]. It should be noted, however, that: (1) predicted expected values of ^{90}Sr concentrations in few cases essentially differed from relevant mean observation data (up to the factor of 6; and on average by a factor of 2), and (2) predicted confidence intervals were rather large (a lower bound of confidence interval differed from an upper bound by a factor of 20 to 28).

Relatively large uncertainties in model predictions resulted from: (1) essential uncertainty in estimating initial ^{90}Sr distribution in trench, and (2) spatial variability of radionuclide migration parameters in trench no.22. However the Monte-Carlo simulation has given the expected value of

9,020 Bq/L for averaged ^{90}Sr concentration in trench porous solutions, which closely matched the observed mean ^{90}Sr concentration of 10,400 Bq/L. This result shows that Monte-Carlo simulation has provided unbiased estimation of ^{90}Sr concentration within the trench.

The second validation test consisted in modeling of cumulative release of ^{90}Sr from the trench no.22 to the geo-environment for the same time t=15 y. Monte-Carlo modeling predictions (expected value of release is 4%) have provided close fit with experimental estimates (e.g., 2-5% based on integration of radionuclide concentrations in the aquifer) [7].

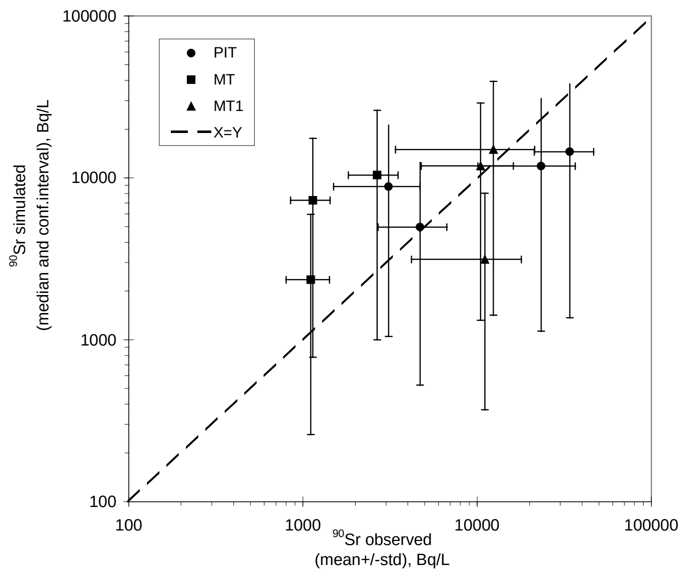

Figure 4. Predicted from Monte-Carlo simulation ^{90}Sr concentrations compared to SWS observation data. (Vertical bars represent confidence intervals. Horizontal bars represent standard deviation in observation data.)

3.3 Global model for the ^{90}Sr transport in the aquifer at Chernobyl Pilot Site

The discussed above sub-models have provided mass input for “horizontal” transport calculations in the aquifer (using MODFLOW – MT3D software). The described above coupling approach resulted in the so-called “global model” of the waste site. The aquifer transport has been analyzed using the following sequence of models of an increasing degree of complexity: the specially developed 1D model (transport along path-line) was used for preliminary calibrations and sensitivity analyses with respect to sorption distribution coefficients, and to check consistency of global model parameters with source-term model calibration studies; the 2D model was used for calibrations with respect to the dispersivity coefficients; then the fully 3D model was applied for a longer term (t=50 y) illustrative predictive calculations of ^{90}Sr transport in the aquifer.

The 1D model calibrations using observation data from selected wells for the period from 1995 to 2002 resulted in the following fitted parameter values: the ^{90}Sr sorption distribution coefficient for the trench was Kd, tr=2-8 ml/g, and distribution coefficient for the eolian sand aquifer was Kd,aq=1 ml/g. Thus, effective parameters of the global model fitted using the data set on ^{90}Sr concentration in the aquifer, were consistent with previous calibrations for the source term model using SWS data[7]. For the 2D model, the best fit with observation data from multilevel well profile for the period of 2001-2002 was achieved with Kd,tr=5 ml/g, Kd,aq=0.5 ml/g, and dispersivity coefficients a_L=10 cm, a_T=1 cm. Again consistency was reached with respect to effective sorption distribution coefficients for each of the system components and dispersivities with respect of experimental data [8-9].

The numerical analyses using the 3D model has shown that the developed modeling methodology is generally applicable to a 3D case. The 3D modeling results allowed visually examine some peculiarities of the ^{90}Sr transport from the trench no.22 to the aquifer in 3D (Figure 5).

The numerical experiments have shown that by means of global model calibration with respect to listed above key parameters (sorption distribution coefficients, dispersivities) a reasonable agreement can be reached between aquifer transport modeling results and field observations. Importantly, the fitted parameter values have fallen to the intervals which were *a-priori* predicted from independent laboratory experimental studies [8-9], geochemical modeling analyses [15] and "elementary model" calibration studies [6-7].

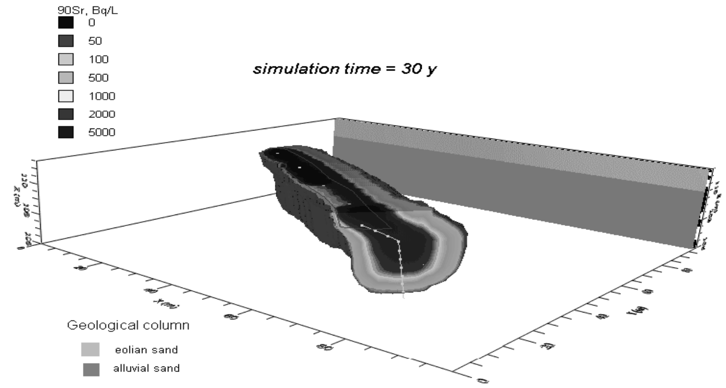

Figure 5. Simulated ^{90}Sr plume in the aquifer for t = 30 years.

4. CONCLUSIONS

In view of the experimental site features (relatively simple geology, well-defined hydrogeological conditions, accurately characterized radionuclide source term, etc.) the modeling approach has given generally good results. The comprehensive multi-disciplinary characterization work on the activity distribution in the trench, dissolution behavior of fuel particles and on sorption interactions in "soil-solution" system provided bases for the quality of the modeling work. The validation tests for the global model serve to increase confidence level in the derived in the course of the Chernobyl Pilot Site project conceptual understanding of radionuclide migration process from nuclear fuel source-term to geo-sphere. Lastly, it should be noted that the developed modeling methodology is mostly suited for producing spatially averaged parameter values, and for long-term predictions on a scale of years and decades assuming that hydrological and geochemical conditions remain steady state.

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