

Radionuclides from Soil to Aquifer in the Chernobyl Pilot Site

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1 INTRODUCTION

Models representing the transfer of radionuclides from a source term to an outlet can be used to devise strategies for monitoring contaminated sites and contribute to safety assessments. Numerous models representing basic phenomena have been developed and applied to explain the behaviour of radionuclides in the environment.

These simplified models constitute partial representations of more complex models and have to be validated by means of experiments. Furthermore, several questions arise when a contaminated site or proposed repository has to be characterised: where should samples be taken, what are the most important parameters to be measured etc.?

In the wake of the Chernobyl accident, studies have been carried out in the environment around the site, particularly since 1994, providing a unique opportunity to develop and explore new modelling and characterisation techniques. One zone in particular was selected for carrying out experiments, namely a trench for the disposal of Red Forest materials contaminated by fuel particles released during the accident.

The behaviour of radionuclides in the soil around the damaged Chernobyl plant is being studied jointly by the Institute for Nuclear Safety and Protection and the Ionising Radiation Applications and Metrology Department of the Atomic Energy Commission in France and the Institute of Agricultural Radiology and the Institute of Geological Science in the Ukraine. This study is of prime importance for the Ukrainian partners and is equally important for the Institute for Nuclear Safety and Protection; the Chernobyl Pilot Site provides it with a tool for on-site validation, and even upgrading, of the theoretical models used by the Institute in its various impact studies to represent the migration of radionuclides through soil.

2 THE PROGRAMME AND ITS OBJECTIVES

Several systems are covered by the experimental programme. Firstly, the trench and top layer of soil constitute the source term of the radionuclides likely to migrate through the environment. They contain fuel particles which dissolve in the environment. Then there is the unsaturated zone which stretches over the entire site and in which water movements are primarily vertical, with varying intensities. Finally, there is the aquifer in which the radionuclides are transported horizontally and dispersed due to interaction with rock.

The main processes involved give rise to two major lines of research. Firstly, the study of the geochemical interactions between the soil and the dissolved radionuclides in the unsaturated

zone and the aquifer and secondly, the study of the mobility of water and dissolved matter in the same two zones. In both cases, the aim is to validate basic models.

These two aspects are related since geochemistry requires that water fluxes be estimated and available mass flux and possible interactions have to be estimated if liquid-phase transport is to be studied. There are other links, for example water flux in the aquifer and infiltration flux; they are illustrated in Figure 1.

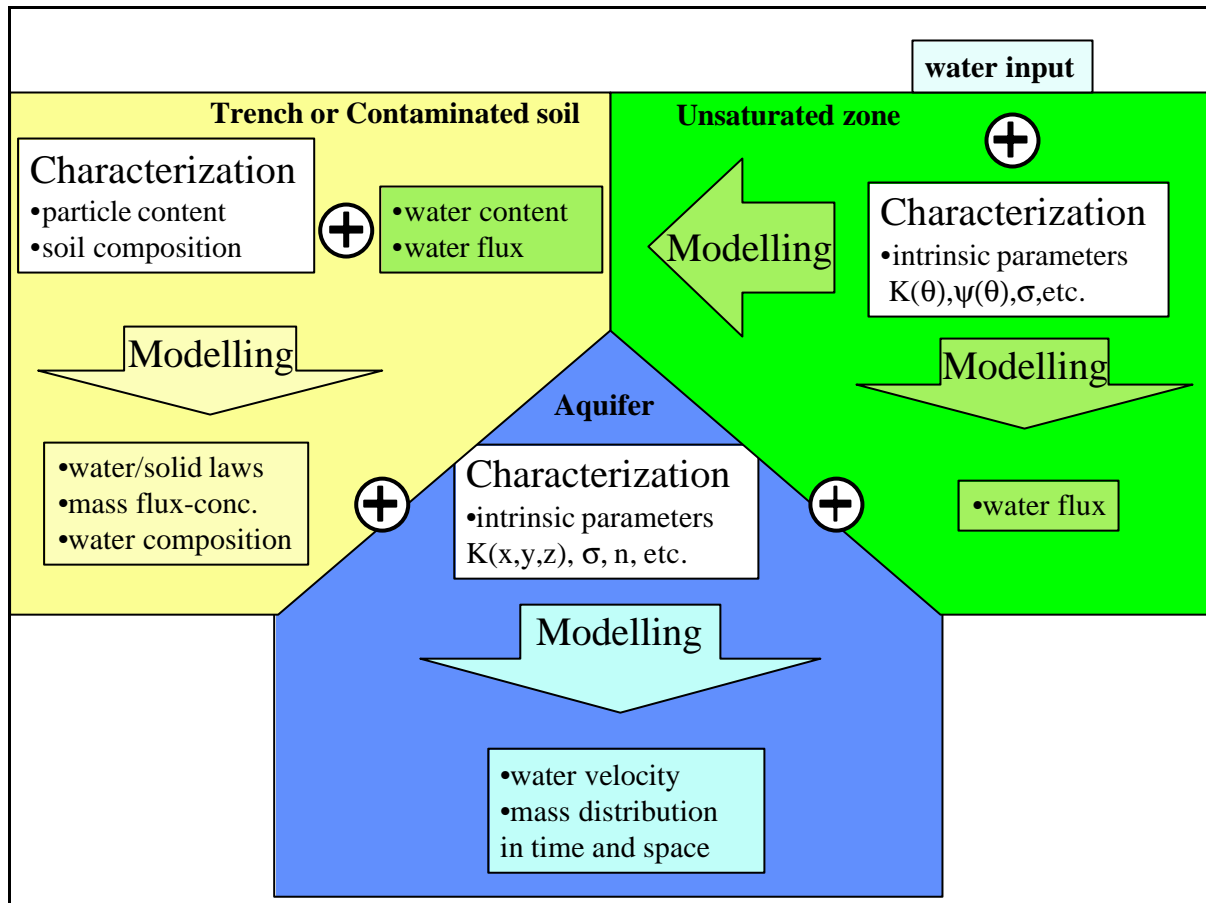


Figure 1 - Illustration of interconnected studies

The project will begin in 1999 and is scheduled for completion in late 2002. It is the continuation of a previous programme (Gri *et al* 99, Guilloux *et al* 99) set up to characterise the governing parameters for the behavior of fuel particles.

It comprises three main stages:

- an additional characterisation stage involving analysis of existing data and preliminary modelling,
- a stage involving modelling and partial validation, determination of supplementary characterisation operations and final validation experiments,
- setting up, carrying out and interpretation of validation tests.

3 DESCRIPTION OF THE SITE

The study site is 2.5 kilometres south west of Chernobyl Nuclear Power Plant in Sector 2.1, known as the Red Forest. In 1987, this sector was earmarked for the interim storage of surface soil and contaminated tree trunks from the Red Forest (Site of Temporary Localization of Radioactive Wastes). These materials can be found in trenches a few metres deep. Since then, the waste has been subjected to the effects of groundwater and infiltration water to varying degrees, depending on the trenches involved. Both the characterisation and modelling studies carried out to date indicate that Trench T22 is the most suitable for carrying out experiments to validate the theoretical models representing the transfer of radionuclides in soils and aquifers.

3.1 Facts and figures

Trench T22 lies in a sedimentary layer of quaternary aeolian and alluvial sand which contains slightly clayey beds of sand (Dzhepo *et al.* 1994). The most abundant mineral in this layer is quartz (around 80 to 90%), followed by feldspar (around 10 to 20%) and secondary minerals such as tourmaline, ilmenite or corundum. Particle size varies with depth, finer particles being found in interbedded layers. Not far from Trench T22, the geological characteristics around Trench T24 (Bugai *et al.* 1995) are very similar and indicate a certain horizontal uniformity.

The hydrogeological system (Dzhepo *et al.* 1994) is fairly simple between ten and one hundred metres, with a free surface aquifer bounded at a depth of around thirty metres by a low permeable clay marl layer. The free surface of the superficial aquifer is at a depth of between one and three metres, in the area of Trench T22. The hydrodynamic characteristics of this aquifer are known to be following: a permeability of around 1 to 10 m.day⁻¹ (1 to 10x10⁻⁵m.s⁻¹) and a hydraulic gradient of between 2 and 3x10⁻³ (based on both *in situ* and in the laboratory estimations). Trench T22 is located above the water table recharge zones and the water flows to the North East towards the River Prypiat, which drains the region. Infiltration recharge is estimated by means of chloride analysis, at between 150 and 300 mm.year⁻¹ whereas annual rainfall is between 550 and 600 mm.year⁻¹. After deforestation in 1987, the amplitude of piezometric fluctuations was sufficient for the water table level to reach the bottom of Trench T22. This phenomenon was curtailed somewhat after surface replanting.

The source term of the radionuclides in the environment is a heterogeneous mixture of contaminated tree trunks and soil containing fuel particles. The total amount of stored activity is estimated at 1.3x10¹² Bq of caesium-137 and 9.0x10¹¹ Bq of strontium-90; these figures were obtained by drilling through the trench and in the immediate vicinity. The horizontal spatial distribution of this activity must vary along the length of the trench, due to the types of materials stored and in the surrounding soil due to deforestation and decontamination operations (Prister *et al.* 1997, Bugai *et al.* 1995).

For ten years, radionuclides (see Figure 2) have been penetrating the unsaturated soil and the water table. Therefore, the ⁹⁰Sr concentration in the water table varies between 10 and 13,000 Bq.l⁻¹ and the plume is seen to spread over a few tens of metres downstream of the trench. A depth of around five to seven metres is seriously affected. This short migration distance seems to imply that the site being studied has not been affected by Trench T24 located 70 m upstream of the discharge.

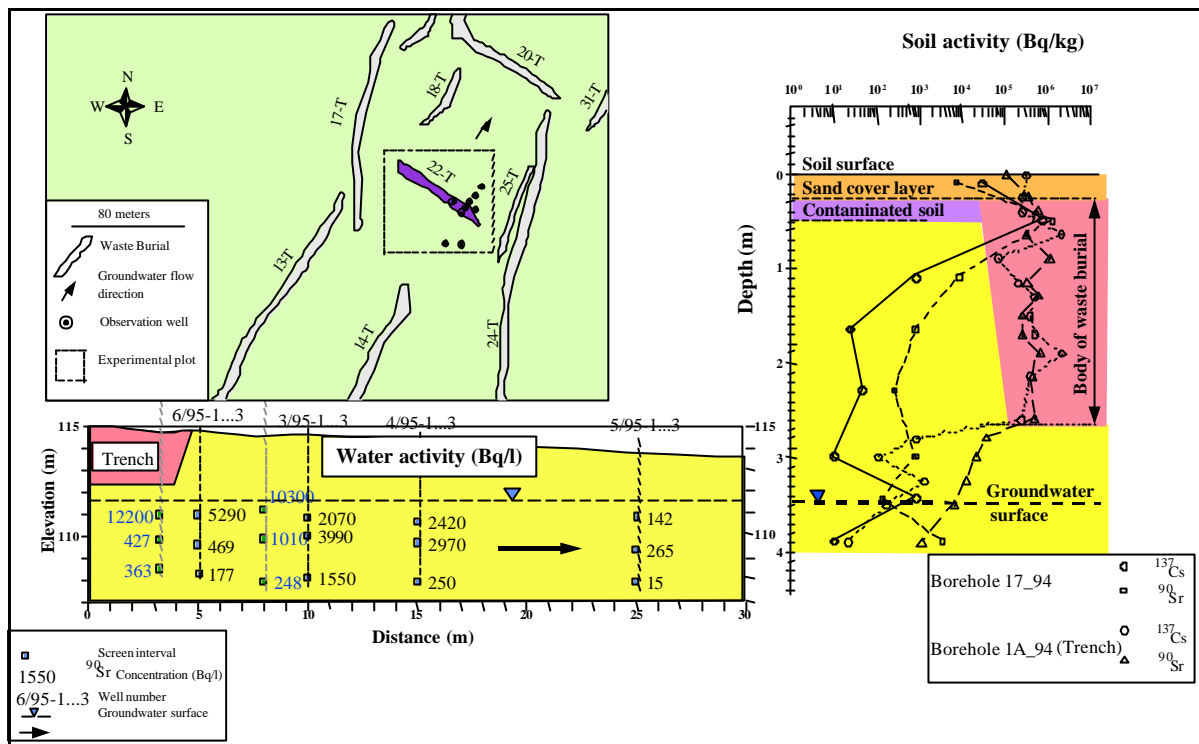


Figure 2 - Partial view of site and strontium and caesium contamination

3.2 Additional characterisation data obtained in 1999

3.2.1 Quantification of the source term

In 1987, the creation of the waste repository in Trench T22, in what were difficult radiological conditions, was not accurately documented and there is still a degree of uncertainty as to the position of the trench and its radiological contents. Several exploration techniques were tested on the site in an attempt to clarify the situation. Surface methods involving radiation and geophysical measurements were used, as were direct measurements of soil activity. Over an on-site area 80 metres by 100 metres, levelled stakes were sunk 20 metres apart on a square grid, with dividers every five metres. The grid was the basis for the various measurements.

The geophysical method used by the Institute for Nuclear Safety and Protection is used to measure apparent electrical conductivity. The tree trunks mentioned in the literature were expected to provide sufficient contrast between the surrounding soil and the trench as to cause an anomaly. After several profiles had been produced, no significant discrepancy was observed. This method was abandoned forthwith.

The teams from the Ukrainian Institute of Agricultural Radiology and the Ionising Radiation Applications and Metrology Department measured the gamma radiation on the surface and a few centimetres underground. Both methods were intended to delimit the trench by pinpointing the difference between the overall signal and the background noise. To this end, the two probes used were positioned in such a way that they did not detect interference from above or from the side and measurements were also made at the bottom of smaller bore holes to eliminate the shield created by the surface layer.

Current analyses show the measured values to be highly non uniform, with numerous points where the activity is more intense. The signal is very noisy and there is no sign that the activity is distributed in the way expected of the trench.

These measurements were supplemented by samples taken at a depth of two to three metres in the trench zone. Since contaminated soil was used to fill the trench, there is clearly a difference between the surrounding soil and that in the trench itself. This can be seen by a change in colour; the contaminated soil is brown whereas the natural soil is white. This visual characterisation was backed up by measurement of the gamma ray emission profiles in the sample holes by the Ukrainian Institute of Agricultural Radiology (see Figure 3) and the beta ray emission profiles by the Institute for Nuclear Safety and Protection using core samples, both on site and in the laboratory.

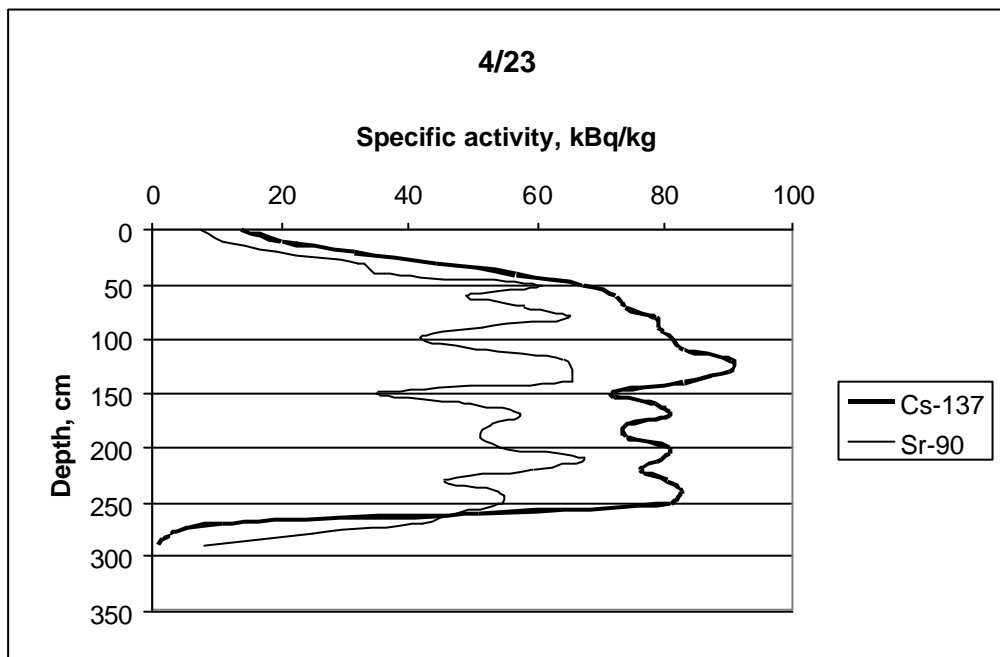


Figure 3 - An example of the activity profile through the trench (hole 4/23)

The two methods, which were carried out 50 centimetres to one metre apart in the horizontal direction and 10 to 20 centimetres apart in the vertical direction, were used to determine the three-dimensional geometry of the trench and quantify its radiological contents. Despite being longer to set up, preference was subsequently given to this type of characterisation.

The change in the radiological contents of the trench over time was also dealt with by investigating the particle dissolution mechanisms. These mechanisms were characterised and modelled by combining chemical dissolution measurements and a description of the type and morphology of the particles contained in the soils. Electron microscope techniques can also be used to provide phenomenological explanations (Kashparov *et al.* 1999) for the differences observed between models based on particles taken directly from fuel and collected in the soil. The material fluxes likely to migrate can be directly determined from this type of model.

3.2.2 The aquifer

In addition to what is generally known about the aquifer, the research programme includes tracer tests in condition of natural drainage. The experiments are carried out in a limited space and only a few tens of metres are available for characterising the direction and intensity of groundwater flow. The site therefore had to be fitted out with piezometers before the two parameters could be quantified.

The existing equipment, managed by the Institute of Geological Science, comprised a series of small-diameter multiple piezometers screened on a few tens of centimetres high. Although this installation was perfectly suitable for three-dimensional characterisation of radionuclide concentration in the water table, it could not be used for the required measurement scale. New piezometers were therefore drilled on site, screened at lengths ranging from one to several metres. The new installations were used to measure the hydraulic head gradient and local permeability in two dimensions over several tens of metres. The gradient was calculated using the water levels measured in the various piezometers; permeability was determined by hydraulic tests or estimated on the basis of the sand particle size curve. Darcy's law was applied to these two values to obtain the intensity of the water flux; the direction was determined by the geometry of the piezometer surface (see Figure 4). This measurement represents one instant in a transient situation and has to be supplemented if the change in the parameters over time is to be monitored. Four of the installation have therefore been fitted with an automatic piezometric data acquisition system.

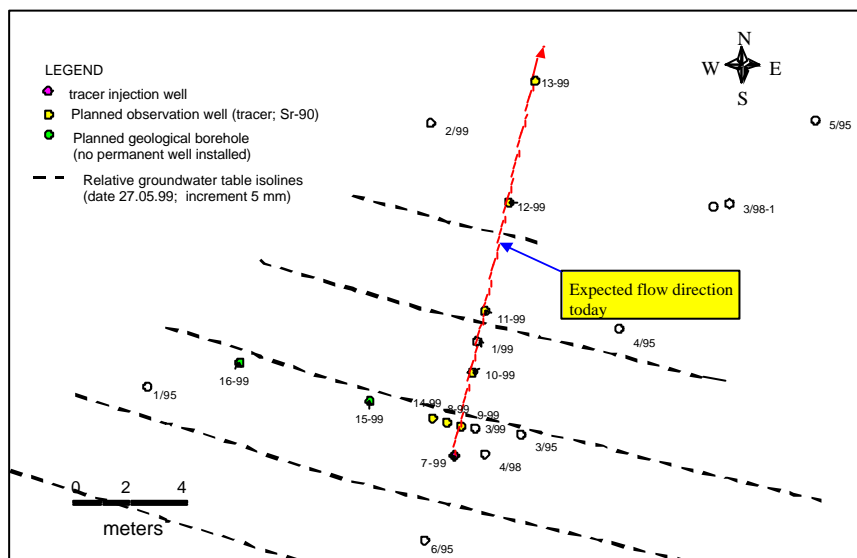


Figure 4 – Piezometric map and mesh of wells

The estimated water flux characteristics were compared with a direct on-site flux measurement. This method, which is known as a single hole dilution test, involves measuring the rate at which a homogenised, injected to the well tracer disappears due to mixing with groundwater flow through the well screen installed to the aquifer. At the expense of a number of hypotheses on the quality of the contact between the piezometer and the aquifer, and on the characteristics of the homogenisation system, this rate is linked to the Darcy velocity (see Figure 5) perpendicular to the measuring chamber.

The deviations between the deduced velocity from permeability and gradient values, ranging from a factor of five to a factor of ten, and the measured values are currently being analysed.

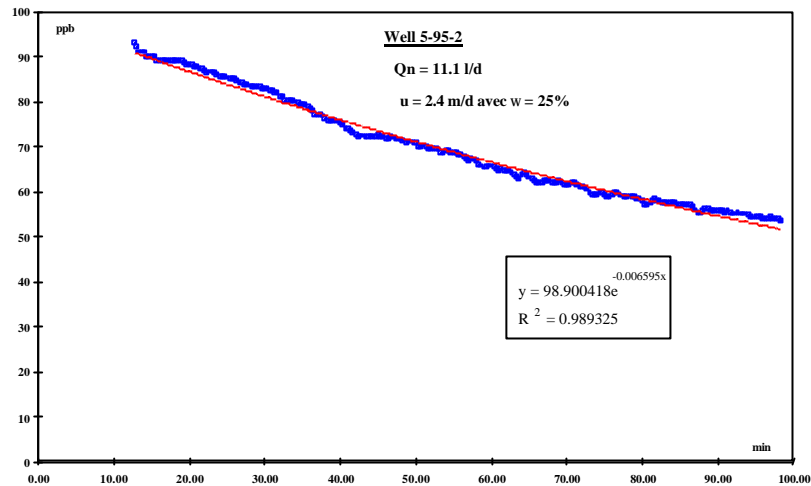


Figure 5 - Dilution curve from Well 5-95-2 and velocity analysis

4 THE PROGRAMME IN THE FUTURE

Characterisation focused on delimiting the waste trench and determining local conditions of infiltration recharge to the water table. Numerous resources have been and will continue to be implemented to obtain the information required for setting up tracer experiments. Even at this very early stage, it is possible to obtain facts about the site itself and the methods used to finalise characterisation.

As concerns describing the source term for, amongst other things, quantifying the radiological contents of the trench, the various surface exploration methods used have not provided any tangible results. The direct sampling approach is by far the most effective and we now have the data required for this study. The geometry of the interface between waste and host environment has been described and facts are emerging about the spatial distribution of β and γ activity.

As regards the water table, drilling test methods and particle size analysis are suitable for calculating the permeability of the sand formation. Likewise, piezometer monitoring means that the direction and intensity of the hydraulic gradient can be estimated fairly accurately. Moreover, single hole tracer test back up these interpretations for determining the local water velocity. It is thus possible to sketch out the well drilling configuration to be used to carry out natural gradient tracer experiments using the knowledge acquired of the intensity and direction of water flux.

The project is being carried out in close collaboration with our Ukrainian colleagues from the Institute of Agricultural Radiology and the Institute of Geological Science, as well as those from the Ionising Radiation Applications and Metrology Department at the French Atomic Energy Commission, and is scheduled for completion in 2002. The Institute for Nuclear Safety and Protection will then have experimental methods for carrying out its expert appraisal operations and modelling the behaviour of radionuclides in the environment. In 1999, work focused mainly on *in situ* characterisation; in 2000, characterisation studies will continue in the laboratory and *in situ* validation of the concepts and models developed is scheduled for 2001. All the results will be collated by late 2002 and an attempt will be made to recreate by modelling means the behaviour of radionuclides at the experimental site.

The Ukrainian Institute of Agricultural Radiology and the Institute of Geological Science are charged with assessing the evolution of the contamination in the vicinity of Chernobyl Nuclear Power Plant; this work will be gaining increasing importance in the future and both Institutes are eagerly awaiting the results.

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