Radionuclide Migration: Prediction Experience

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ABSTRACT

Many different methods of calculating radionuclide migration (transfer) with groundwater – from very simple handmade calculations to use of sophisticated computer models, - exist and are in use. There is no doubt whether we can solve a particular problem in this area; the question is how can we find means of doing this in a fast, precise and economical way.

According to practical experience of MosSIA “Radon” specialists it is useful at the first stage to assess the degree to which various parameters affect the final result. Then the relevance of modeling parameters is usually assessed.

SUE MosSIA “Radon” has applied this complex approach to assessing possible radionuclide transfer from the long term storage facilities located within one of the sites in Moscow. Questions of model verification, computer realization, the analysis of obtained results, a role and a place of these calculations in safety assessment and safety case are beyond the scope of this paper.

INTRODUCTION

A little more then ten years ago prediction calculations of radionuclide migration from repositories were the basic means of safety assessment for such objects. In time an understanding has come, that the content of radionuclides in ground and subsoil water is far from being the unique criterion of safety. An annual effective doze which is formed by direct irradiation from contaminated territory, consumption of contaminated water and food prepared from the products, brought up in the contaminated territory, inhalations of a dust and other factors depending on prospective character of activity of the person and an specific environmental conditions – such parameters can demonstrate the radiation effect from the object in more complete manner.

Accumulated experience in field of safety assessment and changes in safety requirements have shown that prediction of radionuclide migration in geosphere is only one of the safety assessment components, when from the other side safety assessment is only one part of the document set required for licensing of activity or the objects or other decision making in field of radioactive waste management. At the same time prediction of the radiation effect on the public based on the calculations of radionuclide migration in geosphere for case of natural degradation of engineered barriers still remains one of the basic reference scenarios in the long term safety assessment of the near surface disposal facilities.

Besides this, the use of radionuclide concentration in ground and subsurface water as endpoints of safety assessment allows increasing of uncertainty level caused by an unpredictable behavior of public in future that is very important for the dose calculations especially when considering hundreds and even thousand of years. This means that prediction of radionuclide migration is still one of the actual tasks in safety assessment and decision making in field of radioactive waste management.

The task of prediction of radionuclide migration from radioactive waste repositories or local contaminated areas is the one researches face with during more then one decade. Many ways of its solution are already developed, tested and widely used by present time including computer tools. In practice, the final choice of a way to be used frequently has subjective character and depends on qualification of the researcher and accessible resources, but, certainly, in many respects is determined by the purpose of the work.

It is possible to say that almost any problem in prediction of radionuclide migration can be solved now. The question, from our point of view, consists in the choice of the way of doing this quickly and precisely. In addition to the problem of available resources that can affect the choice it is important to take into account an additional aspect – uncertainties. Even in case when unpredictable human behavior is out of the scope, a lot of uncertainties still can remain in prediction calculations of radionuclide migration. They can be caused by different reasons and can lead to incorrect results or wasted time and other resources. That is why from our point of view it is very important to optimize the calculations.

OPTIMIZATION OF CALCULATIONS

Currently, there are different methods of calculating radionuclide migration (transfer) with groundwater. There is no doubt whether we can solve a particular problem in this area; the goal is to find means of doing this in a fast, precise and economical way.

When estimating the spread of contamination, we face a number of uncertainties caused by insufficient background research of some processes, multivariant behavior of the system in future, lack of data on the installation under investigation. Therefore, it is necessary to assess at the first stage the degree to which various parameters affect the final result. For this purpose it is convenient in most cases to use mathematical models based on known analytical solutions.

There are different approaches to this prior assessment. One is to define, at the initial stage, the radionuclides and routes of their migration that can a priori be excluded from calculations when solving a particular problem with given specific conditions. With this aim in view, it is necessary to estimate the period of RW specific activity reduction form the initial level to a safe one on account of natural decay, and in future to exclude the routes and mechanisms of transfer with a period of migration that exceeds estimated values. The radionuclides, whose specific activity drops to normal by the time of their transfer into the human habitation system, can also be excluded from further calculations.

To analyze the influence of particular parameters on the estimations, it is most convenient to use mathematical models based on known analytical solutions. In most cases, to assess the influence of these parameters, it is sufficient to use one-dimensional models, especially as using them in calculations leads to conservative results which is acceptable both in safety assessment of projected facilities, and at the initial stage of assessment of commissioned installations. When building such models, it is vital to determine precisely the mechanism of radionuclide release from the final waste forms, possible migration routes and calculation parameters.

ASSESSING THE RELEVANCE OF MODELING PARAMETERS

Calculation results for H-3, Cs-137, Sr-90, Co-60, Ra-226, Pu-238 and Pu-239 migration carried out in the geological environment of the Science-and-Production Facility of Moscow "Radon" showed that depending on the investigated model the rate of release from the repository can vary. Nevertheless, this little affects the maximum migration of contaminated area from the repository and the period of time needed for the concentration to drop to a safe level. Thus, when calculating possible long-term (300-500 years) radionuclide migration from a repository at long range, the analyzed variants of radionuclide release from final waste forms can be regarded equivalent.

Another important constituent in calculating groundwater radionuclide migration is the model of mass transfer, including values of hydrogeological and sorption parameters specific for the examined site. To assess the effect of hydrogeological characteristics on estimation results, "Radon" specialists used an integrated parameter – the groundwater (perched water) flow rate.

Calculations showed that in the environment of the Moscow "Radon" site a single-order reduction of the groundwater flow rate causes an almost double increase of the time of short-lived radionuclides' release from the repository. In this case the period of radionuclide concentration reduction to a safe level in the contamination area slightly increases, and the maximum distance of the contamination area from the repository declines. The impact of the flow rate on the final distance between the repository and the contamination area is more pronounced for radionuclides with a shorter half-life.

On the other hand, increasing water flow rate leads to a reduction of the estimated time of radionuclide release from the repository; for radionuclides with a longer half-life this effect is more obvious. For example, if the value of the groundwater flow rate grows by an order of magnitude, the period of Co-60 transfer from the repository drops by half, and for Ra-226 it grows more than three-fold. At the same time, for radionuclides whose period of decay to a safe level exceeds the period of estimations (here - Cs-137 and Ra-226), the increase of maximum distance between the repository and the contamination area accompanied by a single-order increase of the water flow rate produces comparable results: 2.8 and 3.5 times correspondingly. On the whole, changing water flow rate by a factor of 100 alters migration estimations for Cs-137 by a factor of 7 only.

Sorption properties of the environment, that is the ability to interreact with and trap water soluble ions, also affect radionuclide transfer. These properties can often be characterized by the distribution coefficient (k_d) which is an equilibrium ratio between radionuclide concentration in the rock and liquid phase. "Radon" specialists have analyzed the impact of changing sorption parameters of rock that builds up the perched water formation area on the estimation results for Cs-137, Co-60, H-3 and Ra-226.

There is a inverse dependence between the distribution coefficient and the maximum distance from the repository to the contamination area. This dependence varies for different radionuclides and is of nonlinear character. Thus, for Cs-137 a single-order increase of the distribution coefficient reduces the distance to the contamination area by 2.5 times, and a single-order reduction leads to a 3.8 increase. An overall double-order change of the sorption properties of the environment changes the cesium migration route length by less than an order of magnitude, in which case the migration calculation results prove to be more sensitive to reduction of the distribution coefficient than to its increase.

The latter trend is even more obvious in calculations made for Co-60. A single-order reduction of the distribution coefficient causes a three-fold increase of the migration route length, and a double order reduction increases it 24 times.

A similar result has been obtained for Ra-226. A single-order reduction of the distribution coefficient for radium increases the migration route length 3.5 times.

Unlike the radionuclides mentioned above, tritium is practically not sorbed by the rock environment, therefore its distribution coefficient can be taken as 0. Nevertheless, such factors as one-side open pores and diffusion of the waterflow front can affect its move in the soil. To assess the effect of a possible delay of tritium migration on the contamination zone travel, a scenario with a distribution coefficient conventional value of 1l/kg was studied. In the given conditions, increasing kd from 0 to 1 decreases the maximum distance to the contamination area by a factor of 7 or more.

For natural rock environment, changing its filtration properties does not, as a rule, exceed a single order, therefore uncertainties of their values do not bring any significant errors into migration calculations. Since the sorption characteristics of rock exert a notable influence on migration forecast results, Moscow "Radon" specialists pay great attention to studying them, together with analyzing the geological structure and hydrogeological conditions of the examined site.

The example below can illustrate the way we use to investigate the interaction of radionuclides with site specific host rocks and obtain sorption characteristics to be used in prediction calculations.

MIGRATION ESTIMATIONS FOR RADIONUCLIDES ON THE KURCHATOV INSTITUTE GROUNDS

SUE MosSIA "Radon" has applied this complex approach to assessing possible radionuclide transfer from the interim storage facilities located on the grounds of the Russian Research Center "Kurchatov Institute". The geological and hydrogeological analysis and the results of long-term monitoring of the Center's territory allowed to determine the most probable routes of pollution migration in case of radionuclide release from the repositories. Samples of rock from potential contamination areas were taken for laboratory study of their sorption properties.

It was found that Cs-137 is fairly tightly held by soil particles; the main absorbed dose of this radionuclide is in an nonexchangeable form, i.e. does not interreact with groundwater.

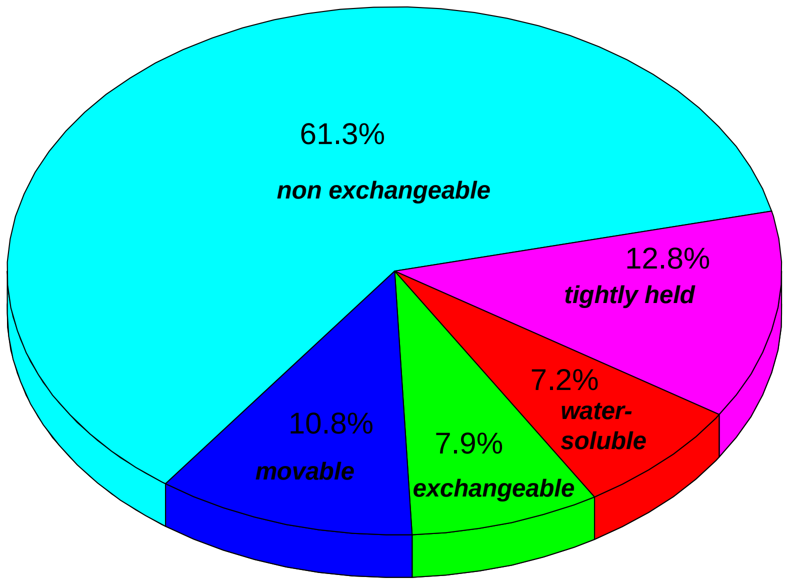

Fig. 1. Forms of Cs-137 in contaminated ground

To determine the sorption properties, natural water artificially contaminated with Cs-137 and Sr-90 radionuclides was used. It was discovered that the ion-exchange equilibrium in the “soil-solution” system is reached within 6 hours for Cs-137 and 30 hours for Sr-90, and hereby the sorption isotherm for both radionuclides is of linear character. The distribution coefficient for Cs-137 was 70 ± 5 ml/g, and for Sr-90 9 ± 1 ml/g.

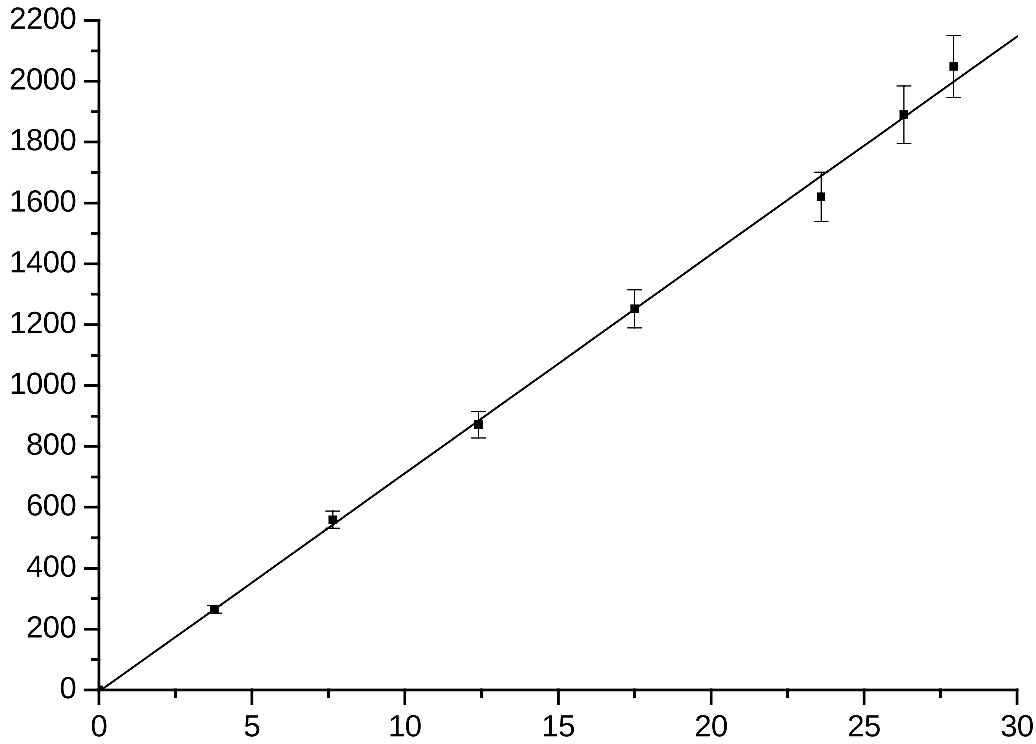

Fig. 2. Isotherm of Cs-137 sorption from the solution with constant concentration

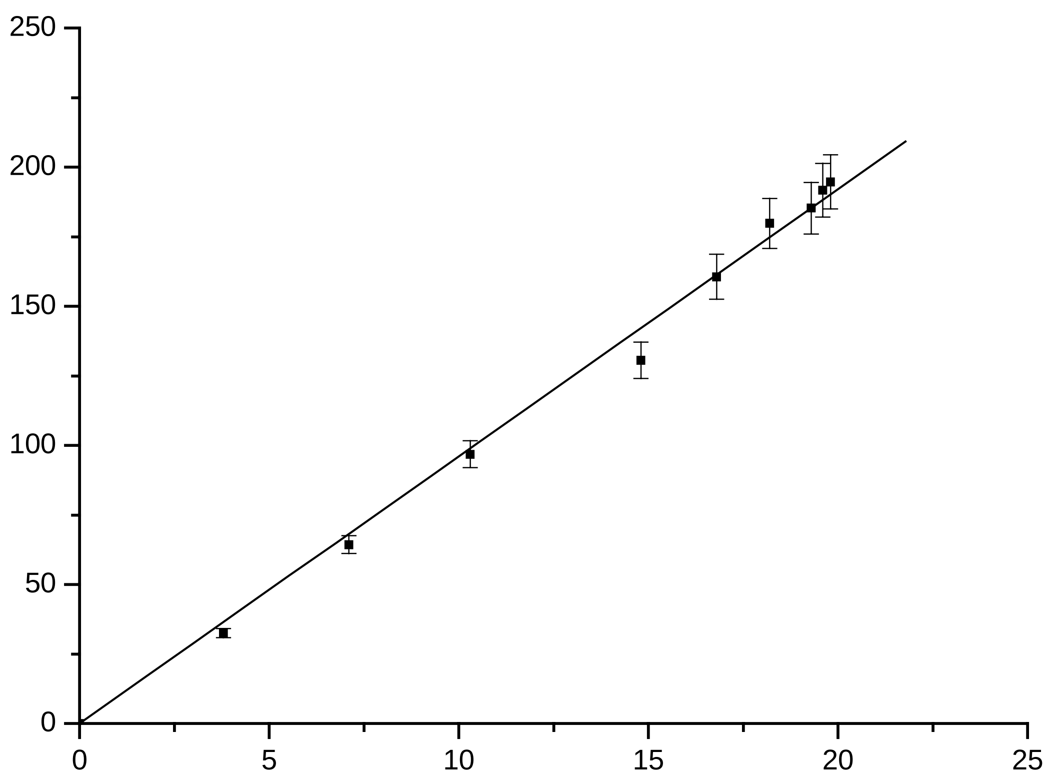

Fig. 3. Isotherm of Sr-90 sorption from the solution with constant concentration

Research of Cs-137 desorption (washing) with natural water from contaminated soil with a specific activity of 45 Bq/g showed that only a minor part of Cs-137 transfers into the liquid phase. The equilibrium concentration of Cs-137 in a desorbing solution did not exceed 0.12 Bq/ml, and the total desorption degree – 0.45 %.

The obtained values were used as benchmark data to predict radionuclide migration form interim RW storage facilities. Calculations for 13 scenarios with different hydrophysical and reservoir parameters were carried out.

Migration prediction did not reveal any significant move of contamination area or high radionuclide concentrations – this can be explained by intensive dilution of groundwater with precipitation and absorption of Sr-90 by underlying moraine loam.

Due to good sorption properties of the rock environment, Cs-137 will not be exported beyond the boundaries of the interim storage site. According to the estimations, the value of Cs-137 contamination area transfer with the groundwater flow will not exceed 3.2 m.

The obtained results were confirmed by the groundwater monitoring data that showed no Cs-137 exceeding background levels.

CONCLUSION

The work carried out by SUE MosSIA "Radon" demonstrates that the calculations must include possible migration routes, taking into account the real hydrogeological and climatic settings and using the calculated parameters peculiar to particular sites. If the direction and routes of possible migration are determined correctly, the effectiveness of the one-dimensional prediction model is comparable to a sophisticated three-dimensional one, built on the basis of long-term monitoring.

The way and results presented above are not a recipe and reference values or parameters. In each specific case they can depend on site specific conditions and the main goal of the work. It is only the illustration of our approach and one can make his own conclusions from it.