# Estimation of speciation and distribution of long-lived radionuclides in soils after irrigation with contaminated well water

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## Introduction

A current approach to estimate radiological doses and risks to humans living far in the future in the vicinity of nuclear waste disposals is the concept of so-called "Reference Biospheres" Within this framework, the irrigation of agricultural soils by contaminated well water and the subsequent transfer of radionuclides within the biosphere is an important pathway to be considered

The transfer of radionuclides into the food chain is strongly influenced by their speciation. distribution and transport within the soil. These processes depend on soil parameters such as pH value, mineralogical composition and organic matter. These parameters, in turn, may vary considerably with soil type and agricultural conditions. In this study it is shown that the distribution of radionuclides in soils after irrigation can be satisfactorily estimated by a CA (component additivity) model implemented into a geochemical code

The concept of Reference Soils (e.g. the German Refesol system [1]) offers an attractive possibility of choosing representative soils that may be used to characterize a Reference Biosphere. By definition, Reference Soils are thoroughly specified and particularly their basic physicochemical properties are known. Our model is intended to be applied to these soils.

# **Description of the model**

Modelling was done using the code PHREEQC [2], which has frequently been employed for geochemical calculations

The model comprises the most important soil components that determine the distribution of radionuclides introduced into the soil:

- · soil solution
- oxalate extractable hydrous ferric oxides (HFO)
- clay (illite as representative material, including frayed edge sites)
- immobile organic matter dissolved organic matter (DOM)

The parameters for the calculation of exchange and complexation were taken from the work of Bradbury and Baeyens [3, 4] (illite), Dzombak and Morel [5] (hydrous ferric oxides) and Tipping [6] (immobile and dissolved organic matter).

The thermodynamic data base used here is an extension of the NAGRA database [7].

## **Model verification**





Figure 1. Comparison of measured and calculated U concentrations in soil solution

#### Caesium

In another batch eauilibrium experiment, Nisbet [9] treated <sup>134</sup>Cs contaminated soils with a solution of 11.5 m potassium sulphate. The <sup>134</sup>Cs concentration in the soil increased due to replacement by potassium on the surfaces. The partial PHREEQC model for illite is able to excellently predict this behaviour

Figure 2. Activity of 134Cs in soil solution before and after treatment with 11.5 m potassium

Vandenhove et al. [8] studied the influence of soil parameters on soil solution concentration of uranium. which is sorbed by clay, HFO and organic matter as well.

They performed batch equilibrium experiments with 18 different soils that covered a comparatively wide range of soil parameters such as pH, cation exchange capacity, clay content, HFO content and percentage of organic Four matter. weeks after contamination of these soils with 238U soil parameters were measured and the soil solutions were analysed.

DOM was not measured; therefore, an average content of 36 mg/l was used for the PHREEQC simulations.

The maximum deviation of the calculated U concentration compared to the measurement is by a factor of 2.5



# Applications

The model has been applied to two soils from the Refesol system (Luvisol, 02-A and Podzol, 04-A). The distribution coefficients ( $K_D$ ) for the nuclides  $^{135}Cs$ ,  $^{63}Ni$ ,  $^{238}U$  and  $^{79}Se$  were are comparable to the new best estimates for loam and sand published by Gil-García et al. and Vandenhove et al. [10]





Figure 3. Comparison of the calculated  $K_D$ -values with best estimates (geometric mean values) from the literature [10] (top: loam, bottom: sand). The range of literature values is indicated by the whisker plots

#### Conclusions

The calculations presented above show that our model is capable of reasonably estimating the content of uranium and cesium in soil solution after contamination with these elements. This opens the possibility to predict the influence of soil parameters on the distribution of radionuclides in soils

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