



PARAMETERIZATION OF RADIOCAESIUM SOIL-PLANT TRANSFER USING SOIL CHARACTERISTICS

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Summary

A model of radionuclide soil-plant transfer is proposed to parameterize the transfer factor by soil and soil solution characteristics. The model is tested with experimental data on the aggregated transfer factor T_{ag} and soil parameters for 8 forest sites in Baden-Württemberg. It is shown that the integral soil-plant transfer factor can be parameterized through radiocaesium exchangeability, capacity of selective sorption sites and ion composition of the soil solution or the water extract. A modified technique of [FES] measurement for soils with interlayer collapse is proposed.

1. Introduction

To characterize the Cs-137 soil-plant transfer, the concept of transfer factor (T) or aggregated transfer factor (T_{ag}) has been used (Desmet et al., 1991; Konoplev et al., 1993; Zibold et al., 1995):

$$T = \frac{\text{Cs - 137 specific activity in plants, Bq / kg dry wt.}}{\text{Cs - 137 specific activity in soil, Bq / kg dry wt.}} \quad (1)$$

$$T_{ag} = \frac{\text{Cs - 137 specific activity in plants, Bq / kg dry wt.}}{\text{Cs - 137 deposition on soil, Bq / m}^2}$$

However, the main problem that exists when applying the T_{ag} or T in predictions, dose assessment and modelling of radionuclide migration is that T_{ag} or T varies considerably from soil to soil and from plant to plant. The values of T_{ag} vary by two orders of magnitude even within one geomorphologic cross-section (Konoplev et al., 1993). This high variability stems from the fact that T_{ag} or T is the net result of different specific processes: distribution of radiocaesium in the „soil-soil solution“ system and its uptake from the soil solution to the plant. The first one is influenced by the sorption and fixation abilities of the soil and the ion composition of the soil solution. The second one is also determined by the composition of the soil solution. The uncertainty of a T_{ag} or T

estimation can be decreased substantially using its parameterization through soil-soil solution characteristics based on analyses of fundamental processes.

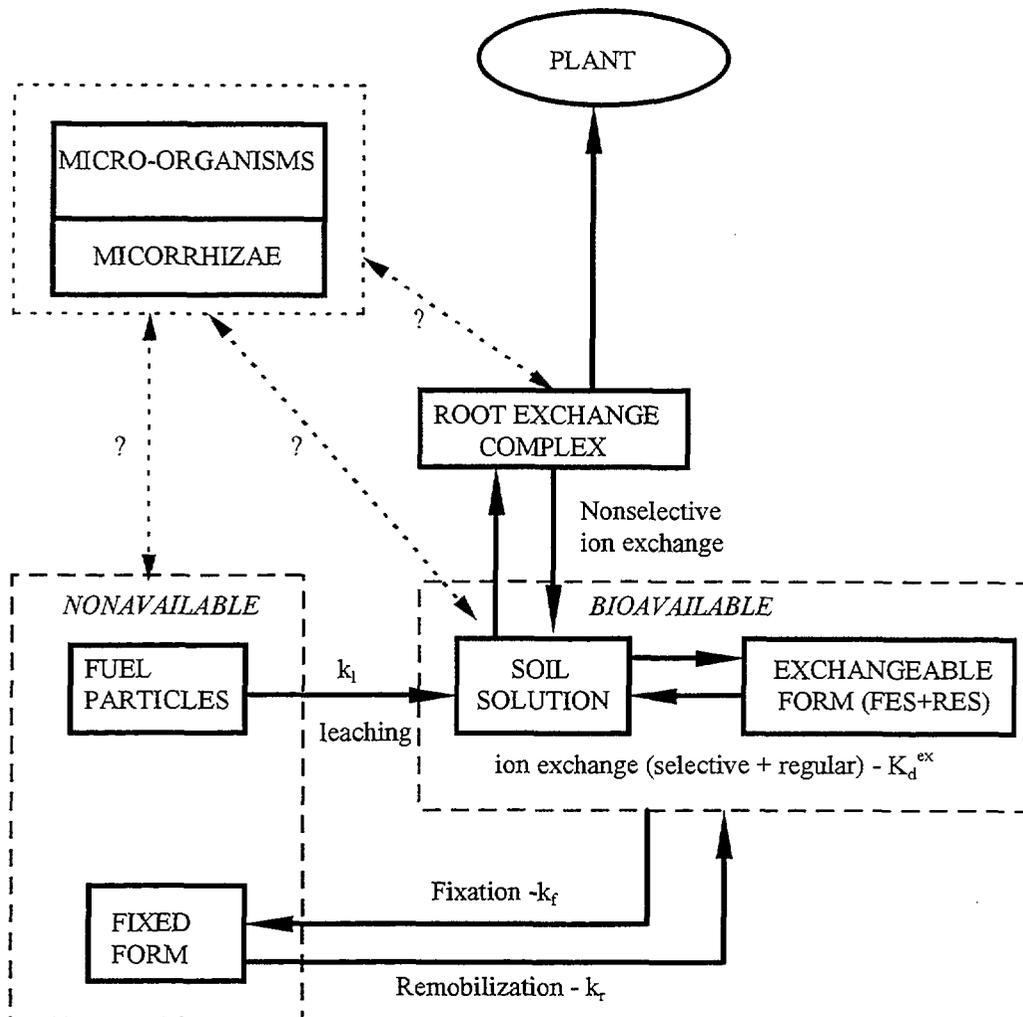


Fig. 1: Scheme of Cs-137 transfer in a „soil/soil solution/plant“ system

2. Theoretical Considerations

Root uptake of radionuclides occurs only from the soil solution and, hence, it is evident that the higher the radionuclide concentration in soil solution the greater is its concentration in the plant. With T_{ag} the radionuclide concentration in the plant is normalized to its inventory in the soil. However, for a given inventory, the concentration of Cs-137 in the soil solution can vary significantly and depends mainly on sorption properties (CEC, FES capacity, fixation ability) and on the composition of the soil solution (concentration of competitive ions). Thus, for a more adequate parameterization of the soil-plant transfer, the radionuclide concentration in the plant must be normalized to the radionuclide concentration in the soil solution (Desmet et al., 1991; Konoplev et al., 1993). Furthermore, at a given radionuclide concentration in the soil solution, T_{ag}

depends on the concentration levels of other ions in the soil solution competing with Cs-137 for root uptake.

The scheme of the main processes determining the radiocaesium soil-plant transfer is presented in Fig. 1. According to the current view of plant physiology (Smolders et al., 1996), the radiocaesium taken up in the plant is a linear function of the radionuclide loading in the root exchange complex at the plasma lemma. According to (Smolders et al., 1996), the unit ion exchange selectivity coefficient for the K/NH₄ and the Cs/K pairs can be taken. The same assumption can be used for the Ca/Mg pair. Consequently, K⁺ and NH₄⁺ can be treated as one ion M⁺, and Ca²⁺ and Mg²⁺ can be represented by M²⁺. On the basis of these assumptions, one can write (Smolders et al., 1996):

$$[\text{Cs-137}]_{\text{plant}} = \frac{k \cdot [\text{Cs-137}]_{\text{ss}} \cdot (M)}{[M]} \quad (2)$$

where (M) is the fractional loading of M⁺ in the root ion exchange complex (potassium window); [M] is the concentration of M⁺ in the soil solution; [Cs-137]_{ss} is the Cs-137 concentration in the soil solution; [Cs-137]_{plant} is the Cs-137 concentration in the plant; k is a proportionality constant linking uptake and radiocaesium level in the exchange complex, reflecting the efficiency of the transport process across the plasma lemma.

On the other hand, according to (Cremers et al., 1990) one gets:

$$K_d^{\text{ex}} = \frac{[\text{Cs-137}]_{\text{ex}}}{[\text{Cs-137}]_{\text{ss}}} = \frac{K_C^{\text{FES}}(\text{Cs/K}) \cdot [\text{FES}]}{K_C^{\text{FES}}(\text{NH}_4/\text{K}) \cdot [\text{NH}_4] + [\text{K}]} \quad (3)$$

where [NH₄] and [K] are concentrations of ammonium and potassium in the soil solution, [FES] is the frayed edge sites capacity, K_C^{FES}(Cs/K) and K_C^{FES}(NH₄/K) are selectivity coefficients on FES for correspondent ion pairs.

Using [Cs]_{ex} = α_{ex} [Cs]_{tot}, where [Cs]_{ex} is the concentration of exchangeable Cs-137 in the soil, [Cs]_{tot} is the Cs-137 total concentration in soil, α_{ex} is the fraction of exchangeable Cs-137 in the soil, we get:

$$T = \frac{[\text{Cs}]_{\text{plant}}}{[\text{Cs}]_{\text{tot}}} = k \frac{(M)}{[M]} \cdot \frac{\alpha_{\text{ex}} \{K_C^{\text{FES}}(\text{NH}_4/\text{K}) \cdot [\text{NH}_4] + [\text{K}]\}}{K_C^{\text{FES}}(\text{Cs/K}) \cdot [\text{FES}]} \quad (4)$$

On the basis of eq.(5) it is possible to identify the soil parameters which can be used for the prediction of the transfer factor. Taking into account that k and K_C^{FES}(Cs/K) are constant, and the fact that for most mineral soils [K] >> [NH₄], we get:

$$T \sim \frac{\alpha_{\text{ex}}}{[\text{FES}]} \cdot (\text{K}) \quad (5)$$

It can be shown that for most soil solution compositions, (K) is proportional to $[\text{K}]_{\text{ss}}/([\text{Ca}]_{\text{ss}}+[\text{Mg}]_{\text{ss}})^{1/2}$ (Smolders et al., 1996); and [FES] can be roughly approximated by $[\text{K}]_{\text{ex}}$ (Wauters et al., 1996). In this case, we get:

$$T \sim \frac{\alpha_{\text{ex}}}{[\text{K}]_{\text{ex}}} \cdot \frac{[\text{K}]_{\text{ss}}}{\sqrt{[\text{Ca}]_{\text{ss}}+[\text{Mg}]_{\text{ss}}}} \quad (6)$$

A similar relationship can be obtained for conditions when $[\text{K}] \ll [\text{NH}_4]$.

The ratios of ion concentrations in the soil solution are assumed to be proportional to the correspondent ratios in a water extraction of the soil. Thus, the relationship (6) can be rewritten as follows:

$$T \sim A = \frac{\alpha_{\text{ex}}}{[\text{K}]_{\text{ex}}} \cdot \frac{[\text{K}]_{\text{w}}}{\sqrt{[\text{Ca}]_{\text{w}}+[\text{Mg}]_{\text{w}}}} \quad (7)$$

where $[\text{M}]_{\text{w}}$ are the dominant ion concentrations in the water extraction.

So, applying only two extractions: a water and an ammonium (1N) extraction, and measuring [Cs-137] and [K] in the ammonium extraction and [K], [Ca], [Mg] in the water extraction, the parameter A can be obtained.

3. Materials and methods

Soil samples from 8 control points of the Radiometry laboratory of the Fachhochschule Ravensburg-Weingarten have been taken from Oh horizon. The T_{ag} of Cs-137 for various plants are known for these sites (Zibold et al., 1995, Drissner et al., 1996). 50 g of each soil were finely ground in a mortar prior to use. First, the total Cs-137 activity was measured by γ -spectrometry. Then, 400 ml of a 1N ammonium acetate was added to each soil sample. The extractions were carried out for 24 hours. After that, the extract was filtered and Cs-137 was measured in the liquid. 5g of each soil was extracted by distilled water during 24 hours. The ammonium and water extracts of each soil were analysed by AAS to measure the concentrations of basic competing ions, namely Ca, Mg, K and Na.

The effect of interlayer collapse in high organic forest soils (Hird et al., 1995) makes a quantitative determination of [FES] impossible if the original methodology (Cremers et al., 1988) is used without modifications. In this work, [FES] was obtained from the

intercept of the linearization of the Langmuir isotherm in the range below the sorption maximum:

$$\frac{1}{[Cs]_{ads}} = \frac{1}{[FES]} + \frac{const}{[FES]} \cdot \frac{1}{[Cs]_{sol}} \quad (8)$$

where $[Cs]_{ads}$ and $[Cs]_{sol}$ are concentrations of adsorbed and dissolved Cs at the equilibrium. Taking the initial range of the isotherm at low caesium concentrations, the capacity of high affinity sites [HAS] was determined. To avoid an influence of Cs fixation on the measurement of [FES], an additional step of a 1N ammonium acetate extraction of exchangeable Cs in the soil after equilibration was introduced.

4. Results and discussion

The approach described above is applied to analyse the variability of the Cs-137 aggregated transfer factor T_{ag} of fern for eight spruce forest sites in Baden Württemberg, previously classified into two different transfer groups (Zibold et al., 1995, Drissner et al., 1996) according to the values of T_{ag} which correlate also with some external soil properties (kind of humus, its thickness and pH). The dependence of T_{ag} on the A-parameter calculated according eq. (7) is presented in Fig. 2. It can be seen that a linear relationship exists for T_{ag} as a function of the soil parameter A within each transfer group, however the slopes of this dependence are different for different transfer groups. Two main conclusions can be derived from this analysis:

1. The classification of soils as proposed previously in terms of the Cs-137 bioavailability corresponds quite well to the physico-chemical description in terms of T_{ag} -A relationship.
2. The different magnitudes of the slopes for the two transfer groups might be caused by an additional mechanism not taken into consideration in the proposed model. This mechanism is possibly connected to the role of mycorrhizae in soil-plant transfer which is still unclear. To clarify the role of mycorrhizae in soil-plant transfer, in depth investigations of radiocaesium interaction with mycorrhizae in various physico-chemical conditions of soil should be performed.

To improve the accuracy of the determination of A, [FES] should be measured. Up to now [FES] and [HAS] of 3 forest soils were measured using the modified technique of (Cremers et al., 1988). Results are presented in Fig. 3 and Table 1. It can be seen that [FES] is relatively low for forest soils of Baden-Württemberg with a high content of

organic matter. In the case of these three forest soils [HAS] represents about 10% of the total capacity of regular [FES].

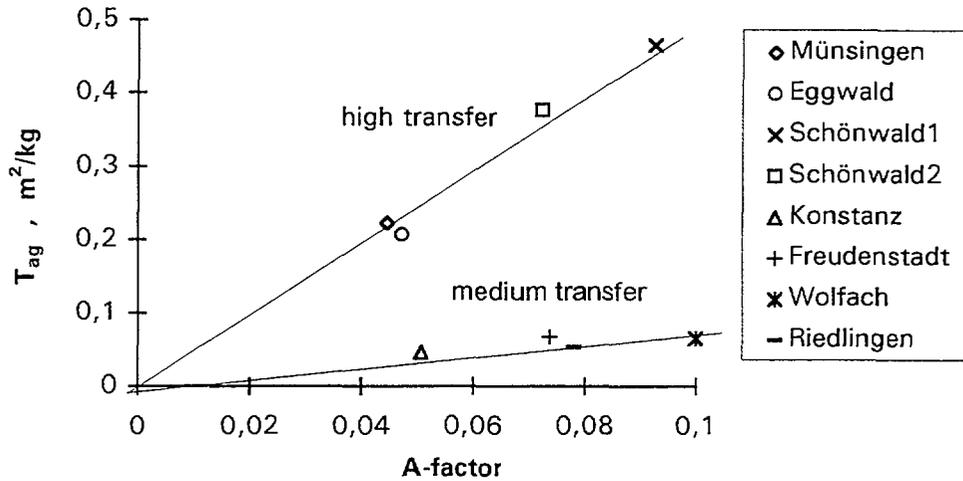


Fig. 2: Dependence of T_{ag} on A-factor for high transfer and medium transfer groups

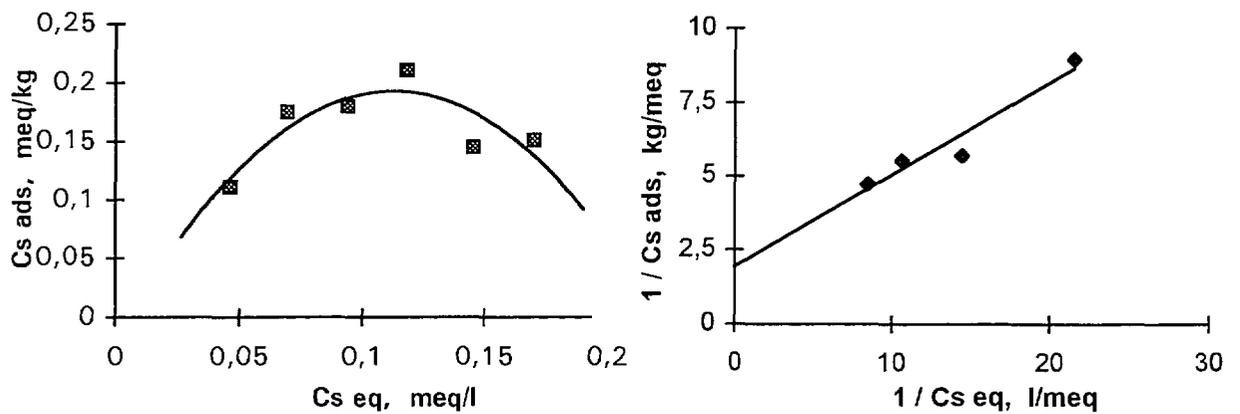


Fig. 3: [FES]-determination of forest soil (Wolfach): Langmuir isotherm (left) with collapse due to breakdown of clay mineral particle interlayers and linearisation (right). [FES] = 0.52 meq/kg is the reciprocal of the intercept.

Tab. 1: FES and HAS capacities of spruce forest soils with high organic content as measured with NH_4 extraction and Langmuir extrapolation.

Site	[FES], meq/kg	[HAS], meq/kg
Konstanz	0.12	0.012
Wolfach	0.52	0.055
Freudenstadt	0.08	0.008

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5. References

- Cremers A., Elsen A., De Preter P., Maes A. (1988). Quantitative analysis of radiocaesium retention in soils. *Nature* **335**, 247-249
- Cremers A., Elsen A., Valcke E., Wauters J., Sandalls F.J., Saudern S.L. (1990). The Sensitivity of Upland Soils to Radiocaesium Contamination. *In: Transfer of Radionuclides in Natural and Semi-natural Environments. Ed. by G. Desmet, P. Nassimbeni, M. Belli*, 238-248.
- Desmet G.M., L.N. Van Loon, B.Howard (1991). Chemical Speciation and Bioavailability of elements in the environments and their relevance to radioecology. *Sci. Total Environ.*, **100**, 105-124.
- Drissner J., Klemt E., Bürmann W., Enslin F., Heider R., Schick G., Zibold G. (1996). Availability of Caesium Radionuclides for Plants - Classification of Soils and Role of Mycorrhiza. *J. Environ. Radioactivity* (submitted).
- Hird A.B., Rimmer D.L., Livens F.R. (1995). Total Caesium-Fixing Potentials of Acid Organic Soils. *J. Environ. Radioactivity*, **26**, 103-119
- Konoplev A.V., Viktorova N.V., Virchenko E.P., Popov V.E., Bulgakov A.A., Desmet G.M. (1993). Influence of Agricultural countermeasures on the Ratio of Different Chemical Forms of Radionuclides in Soil and Soil Solution.- *Science of the Total Environment*, **137**, 147-162.
- Smolders E., Sweeck L., Merckx R., Cremers A. (1996). Interionic Effects in Radiocaesium Uptake in Plants: A Quantitative Interpretation. *Sci. Tot. Environ.* (in press).
- Sweeck L., Wauters J., Valcke E., Cremers A. (1990). The Specific Interception Potential of Soils for Radiocaesium. *In: Transfer of Radionuclides in Natural and Seminatural Environments. Ed. by G. Desmet, P. Nassimbeni, M. Belli*, 249-258.
- Wauters J., Elsen A., Cremers A., Konoplev A.V., Bulgakov A.A., Comans R.N.J. (1996). Prediction of solid/liquid distribution coefficients of radiocaesium in soils and sediments. Part one: A simplified procedure for solid phase speciation of radiocaesium. *Applied Geochemistry* (in press).
- Zibold G., Bürmann W., Drissner J., Enslin F., Heider R., Klemt E., Schick G. (1995). Availability of Cesium Radionuclides for Plants on Different Spruce Stands in South-West Germany. *Proceedings of the XXVIth Annual Meeting of ESNA. Working Group 3, Soil-Plant-Relationships. Ed. by Gerzabek M.*, 151-157.