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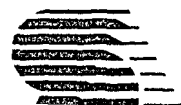
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**CESIUM-137 IN SOIL TEXTURE FRACTIONS
AND ITS IMPACT ON CESIUM-137
SOIL-TO-PLANT TRANSFER**

M.H. Gerzabek, S.A. Mohamad, K. Mück

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ABSTRACT: Field studies at two sites contaminated by the Chernobyl fallout showed $^{137}\text{Cesium (Cs)}$ soil-to-plant transfer factors in wheat, rye and potato. Transfer values ranged from 0.0017 (potato tuber) to 0.07 (wheat straw). Generally transfer coefficients in cereal grains and potato tubers were significantly below the values of the shoots. A comparison of the two sites led to the conclusion that for all plants investigated ^{137}Cs – transfer factors were higher in Lower Austria (Calcic Chernozem) than in Upper Austria (Eutric Cambisol). The specific activities of the texture fractions of the two soil types increased from sand to silt and clay. In the Calcic Chernozem the ratio of the ^{137}Cs -activity in the silt fraction to the total activity in the soil was considerably higher than in the Eutric Cambisol. At the same time extractability of ^{137}Cs from the silt fraction of the latter soil was clearly lower. Both results mainly were attributed to the differences between the soils according to the organic matter content of the silt fractions, the Calcic Chernozem being seven times higher. Therefore, the differences in the ^{137}Cs -soil-to-plant transfer can be attributed partly to these soil characteristics.

INTRODUCTION

The radionuclide release following the reactor accident in Chernobyl caused a considerable radionuclide deposition and a significant increase of ^{137}Cs levels in foodstuff in Austria. The deposition of ^{137}Cs in Austrian agricultural regions ranged from about 0.7 to more than 90 kBq/m² (1).

Exceedingly high Cs-migration rates of 0.2 to 0.3 cm/h in soils were found few days after deposition during a rain shower (2). However, generally the deposited Cs was quickly fixed in the top soil and Cs-migration in agricultural

soils decreased to values below 1 cm/a, levels which are known from the global fallout caesium (2,3). Caesium soil-to-plant transfer factors derived from field studies after the Chernobyl accident [$TF = (0.1 - 5) \times 10^{-2}$] agree with results from studies conducted earlier (4,5,6). On organic soils, Chernobyl-derived caesium shows a higher soil-to-plant transfer (7). Nevertheless, the organic matter content of agricultural soils (OM <15%) seems not to be correlating with the Cs-transfer (8). Differences in the soil-to-plant concentration ratios partly can be explained by the influence of the clay content of the soils (9). However, the correlations are not really satisfactory. Other factors, like *e.g.* the resuspension rate might have a significant influence on the Cs-activity concentration of plants (4,9).

To elucidate the above mentioned questions the present work deals with a comparison of the extractability of ^{137}Cs from soil texture fractions of two characteristic Austrian soils (Calcic Chernozem, Eutric Cambisol) with Cs-transfer factors derived from field investigations.

The chosen soils are quite similar in their pH, potassium and clay contents. Differences in Cs-soil-to-plant concentration ratios therefore cannot be attributed to any of these factors.

MATERIALS AND METHODS

Soils: Two soil samples comparatively high in ^{137}Cs activity were selected for fractionation from a large group of samples collected as a part of field studies on Cs-soil-to-plant transfer. The A horizon of a Calcic Chernozem (FAO, Unesco) characteristic for large areas of Lower Austria with low post-Chernobyl contamination was sampled in July 1986. A typical Eutric Cambisol (FAO, Unesco) of Upper Austria (higher contamination levels) was the second soil chosen for the fractionation study (sampling date: September 1986). The samples were air dried and passed through a sieve with 2-mm mesh size. The soils were characterized by means of standard analytical methods (10).

Texture Fractions: The method of choice was a simple dispersion of the samples by ultrasonification, modified after Armour et al. (11). Methods including chemical pretreatment for particle size fractionation were rejected in order to avoid the exchange of ^{137}Cs from the samples. Preceding the ultrasonification 180 g of soil (<2 mm) was shaken with 450 mL H₂O in order to extract the soluble Cs-fraction. The water was separated from the soil after 24 hours of treatment by

centrifugation. After three more successive steps of extraction and four minutes of ultrasonification (Minison, Heraeus) the sand fraction (63 μm to 2 mm) was isolated by wet sieving. The remaining particles below 63 μm were separated by settling in water (12) into the clay fraction ($<2 \mu\text{m}$) and the silt fraction (2 to 63 μm). The material not retained by the 2-mm sieve was poured into glass cylinders (1000 mL volume), resuspended by stirring and then allowed to settle for 7 hours 43 minutes according to Stoke's Law. After that time the top 10 cm of the suspension was withdrawn. This upper fraction contains only particles smaller than 2 μm . Distilled water replaced the lacking volume in the settling cylinders and the procedure was continued five additional times (if necessary more often) in order to obtain a nearly clear supernatant. Measurements of the particle size distribution according to the standard method allowed to calculate the amount of clay still present in the rest of the sample and to correct the results of the ^{137}Cs -measurements.

Extraction Procedure: In order to get more information about the binding of ^{137}Cs to the three particle size fractions, the dried sand, silt, and clay were extracted by three different resins following each other:

- Barium-triethanolamine (0.05 M BaCl_2 + 0.036 M triethanolamine, pH = 8.1), to characterize the exchangeable caesium,
- EDTA (0.1 M EDTA, set on pH = 10) to extract Cs bound to the humic substances, and
- HCl (0.5 M HCl) for the carbonate-bound caesium.

The soil resin ratio was 1:2.5. The samples were shaken 24 hours and then separated from the extractant by centrifugation. Each resin was used four times successively. The large volumes of solution obtained were reduced by evaporation to approximately 75 mL prior to being measured by the γ -spectrometry. The sand, silt, and clay fractions were oven-dried after the extraction procedure.

γ -Spectrometry: The ^{137}Cs activity concentrations were determined by gamma spectrometry with a high purity Ge-detector of 30% relative efficiency in a well-shielded configuration in a low-level laboratory. By applying long measurement times of 100.000 sec the limit of detection between 0.0005 and 0.001 Bq/g for sample volumes of 50 to 100 mL was obtained. The values of ^{134}Cs were

measured, but are not shown in the present paper. From the ratio of ^{134}Cs to ^{137}Cs it is evident that no significant contribution of pre-Chernobyl caesium was still present in the soil samples investigated.

Transfer Factors: The transfer factors were obtained by field studies in Upper (Eutric Cambisols) and Lower Austria (Calcic Chernozems). ^{137}Cs contents of soil and plant samples were measured by the means of the gamma spectrometry. The transfer factors were calculated as follows:

$$\text{TF} = \frac{\text{Bq } ^{137}\text{Cs} / \text{kg plant fresh weight}}{\text{Bq } ^{137}\text{Cs} / \text{kg soil dry weight}}$$

The mean standard deviation of the transfer factors due to measuring errors is approximately 13.7%.

RESULTS AND DISCUSSION

Cesium Extraction: The characteristics of the two investigated soils are demonstrated in Table 1. The main differences between the soils appear in their C_{org} and CaCO_3 values, soil No. II being considerably higher in both. The pH-values, clay contents and the cation exchange capacity are very similar.

The results of the Cs-fractionation are shown in Table 2. As it has to be expected from literature (12,13), ^{137}Cs -activities of the soil texture fractions increased from sand to silt and clay (residual, Table 2). In both cases the relative Cs-concentrations in silt fractions were more than 10 times higher compared to the sand. The silt fraction of soil No. II turned out to be considerably higher in caesium relative to the total ^{137}Cs -activity of the soil in comparison to soil No. I. Therefore the increase of the ^{137}Cs -activity towards the clay fraction of soil No. II was less pronounced. This result may be due to either the high CaCO_3 -content of soil No. II (Table 3) and the lack of a chemical pretreatment, which may have led to an incomplete dispersion of the sample and therefore to a not neglectable clay content of the silt fraction or a higher Cs-adsorption capacity of the silt fraction of soil No. II due to a higher organic matter content compared to soil No. I. Clay and silt fractions of soil No. II have nearly the same C_{org} content (Table 3). Other investigators showed the importance of humic materials for caesium binding (13).

Cesium contents of the water extract and of all other solutions from sand and clay fractions were below the detection limit of the gamma-spectrometer. This is

TABLE 1
Soil characteristics

parameter	soil number	
	I	II
pH CaCl ₂	7.78	7.40
% C _{org}	2.24	5.29
% CaCO ₃	3.8	34.4
% sand	2.7	17.8
% silt	69.9	58.0
% clay	27.4	24.2
meq K _{BT} /100 g	0.42	0.90
meq Ca _{BT} /100 g	24.70	22.00
meq Mg _{BT} /100 g	1.23	2.20
meq Na _{BT} /100 g	0.07	0.03
CEC meq /100 g	26.42	25.13
Bq ¹³⁷ Cs/kg	1389	324

BT barium-triethanolamine

supposed to be due to the fact, that the sand fractions contained less than 1% of the total caesium and therefore no measurable amounts of ¹³⁷Cs could be extracted. Furtheron the chosen resins were not able to extract significant amounts of ¹³⁷Cs from the clay, probably due to its fixation properties. However, remarkable differences between the soils arose in the silt fraction. The relative amount of caesium extracted from silt No. II was more than three folds compared to soil No. I. Especially ¹³⁷Cs in the humic substances (EDTA-extract) was considerably higher in the Calcic Chernozem (soil No. II). The latter soil has a seven times higher C_{org} content of the silt fraction. This is a further clue to the influence of humic materials on caesium adsorption properties of soils.

Transfer Factors: ¹³⁷Cs soil-to-plant transfer ratios for wheat, rye and potato are given in Figure 1. Generally transfer coefficients into cereal grains and potato

TABLE 2
 ^{137}Cs in soil texture fractions of an Eutric Cambisol (I)
 and a Calcic Chernozem (II)
 % of total and [(Bq ^{137}Cs /kg fraction) . 100 / (Bq ^{137}Cs /kg soil total)]

soil no. I	total : 100 ± 5 % /-		water soluble : < 0.02 /-		
	sand		silt	clay	
BT-extraction	< 0.02	/ < 0.75	< 0.02	/ < 0.03	< 0.04 / < 0.1
EDTA-extraction	< 0.03	/ < 1.04	0.04±11.4%	/ 0.05	< 0.06 / < 0.2
HCl-extraction	< 0.02	/ < 0.88	0.12± 9 %	/ 0.17	< 0.01 / < 0.0
residual	0.07±8.4%	/ 2.51	9.82± 5 %	/ 26.36	93.47±5% / 284.8
soil no. II	total : 100 ± 5.2 % /-		water soluble : < 0.86 /-		
	sand		silt	clay	
BT-extraction	< 0.07	/ < 0.38	0.10±12 %	/ 0.18	< 0.15 / < 0.6
EDTA-extraction	< 0.09	/ < 0.48	0.26±10.7%	/ 0.44	< 0.21 / < 0.8
HCl-extraction	< 0.09	/ < 0.48	0.14±13 %	/ 0.23	< 0.14 / < 0.5
residual	1.01±8.9%	/ 5.25	37.7 ± 5.3%	/ 56.79	68.5 ±5% / 254.9

TABLE 3

 C_{org} - and lime-contents of the soil texture fractions

	sand		silt		clay	
	% C_{org}	% $CaCO_3$	% C_{org}	% $CaCO_3$	% C_{org}	% $CaCO_3$
soil no. I	39.2	n.d.	0.88	n.d.	2.14	n.d.
soil no. II	1.60	25.3	6.13	34.1	6.38	41.9

n.d. not detected

tubers were significantly below the values for the shoots, wheat straw, rye straw, and potato top being on average 3.7, 2.4, and 5.2 times higher, respectively. Caesium is supposed to be a mobile element similar to potassium. Known from literature it is evident that potassium is more concentrated in vegetative plant parts (14). A second source for the higher ^{137}Cs -concentrations in cereal straw and potato tops may be the contamination due to resuspension or soil splash (15). Caesium bound to the plant surface by soil adhesion is only partly available for uptake and translocation.

The observed values cope perfectly well with mean literature transfer factors (16) for ^{137}Cs into cereal grains (0.005). The literature mean value for potato tuber (0.015) is a factor of two higher than observed (16). In general Cs-transfer coefficients derived after the Chernobyl-fallout incline to be lower than those investigated during weapons testing and by artificial contamination (17).

Comparing the two soil types it becomes evident that the Calcic Chernozem in all cases showed higher ^{137}Cs -transfer factors than the Eutric Cambisol. Average transfer values on Calcic Chernozem exceeded those of the Eutric Cambisol by a factor of 2.8. In general two explanations may be considered, a higher root uptake and translocation of ^{137}Cs or/and a decrease of outer contamination and ^{137}Cs -uptake through the leaves with higher deposition levels due to differences in resuspension.

The caesium extraction showed significant differences between the two soil types, the Chernozem being four times higher in extractable ^{137}Cs than the Cambisol (Table 2) which corresponds to the investigated transfer factors. Thus it

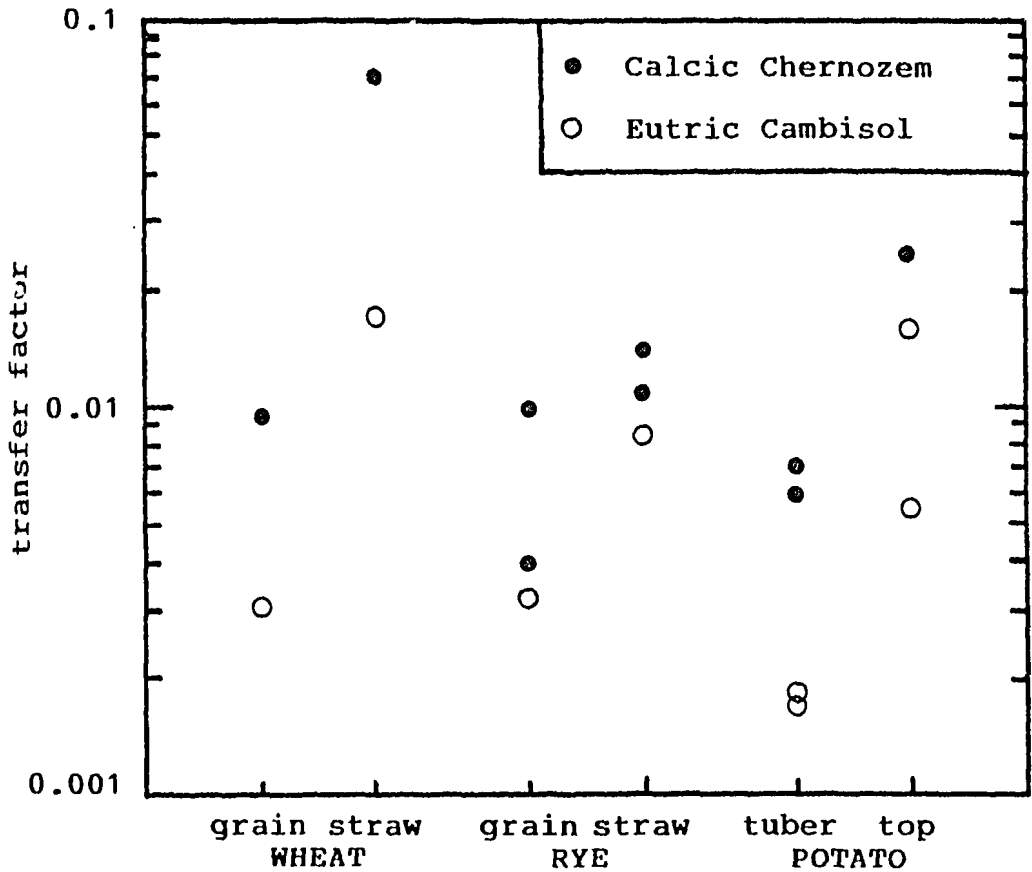


FIGURE 1. The Influence of the Soil Type on the ^{137}Cs Soil-to-Plant Transfer Factor.

can be assumed that soil properties in this case have a distinct influence on ^{137}Cs -uptake into plants. Considering the poor correlations between soil properties and ^{137}Cs -transfer published earlier (4) and the present results it should be concluded that there are significant influences of the soil, but they may be overlapped by the phenomena of outer contamination and foliar uptake due to resuspension and soil splash (15).

There is some evidence for the latter factor (17). After The Chernobyl fallout it has been proven that ^{137}Cs -resuspension diminished with increasing deposition (18). Correlation analyses showed a significant decrease of the ^{137}Cs soil-to-plant transfer factor with growing ^{137}Cs -concentration in the soil (4,9).

CONCLUSION

The present work clearly demonstrates the variation of site in relation to soil-to-plant transfer factors. A ^{137}Cs -extraction experiment resulted in significant differences between the two investigated soil types. Extractability and ^{137}Cs -transfer factors were higher at the Calcic Chernozem than at the Eutric Cambisol sites. However, the differences in the soil-to-plant transfer can only partly be attributed to soil parameters. From the literature (15) and previous investigations (4,9,17), re-suspension should be considered as being a second parameter influencing ^{137}Cs -concentrations in edible plant parts.

REFERENCES:

1. Horak, O. and Gerzabek, M.H. 1988. Basisdaten zur regionalen Prognose der Strahlenbelastung des Menschen nach dem Model I ECOSYS. OEFZS-Rep.-4447, Austrian Research Centre Seibersdorf.
2. Schimmack, W., Bunzl, K., and Zelles, L. 1989. Initial rates of migration of radionuclides from the Chernobyl fallout in undisturbed soils. *Geoderma* 44: 211-218.
3. Meisel, S., Gerzabek, M.H., and Muller, H.K. 1991. Influence of plowing on the depth distribution of various radionuclides. *Z. Pflanzenernaehr. Bodenk.* 154: 211—215.
4. Gerzabek, M.H., Horak, O., and Muck, K. 1990. Cs-137 soil to plant transfer studies and their implications on parameters used in the Austrian version of ECOSYS, pp. 611-618. IN: Desmet, G., Nassimbeni, P. and Belli, M. (eds.) *Transfer of Radionuclides in Natural and Seminatural Environments*. Elsevier Applied Science, New York, NY.
5. Haisch, A., Capriel, P., and Forster, S. 1985. Cesiumverfugbarkeit fur Pflanzen auf drei verschiedenen Boden. *Transferfaktoren Boden - Pflanze. Landwirtsch. Forschung* 38: 229-236.
6. Steffens, W., Mittelstaedt, W., Fuhr, F., Forstel, H., and Klaes, J. 1986. Abschätzung der Aufnahme des abgelagerten Cs-137 und Sr-90 über die Wurzel. *Atomwirtschaft* July: 389-392.
7. Colgan, P.A., McGee, E.J., Pearce, J., Cruickshank, J.G. Mulvany, N.E., McAdam, J.H. and Moss, B.W. 1990. Behaviour of radiocesium in organic soils - some preliminary results on soil-plant transfers from a semi-natural ecosystem in Ireland, pp. 341-354. IN: Desmet, G., Nassimbeni, P. and Belli, M. (eds.) *Transfer of Radionuclides in Natural and Seminatural Environments*. Elsevier Applied Science, New York, NY.

8. Frissel, M.J., Noordijk, H., and Van Bergeijk, K.E. 1990. The impact of extreme environmental conditions, as occurring in natural ecosystems, on the soil-to-plant transfer of radionuclides, pp. 41-47. IN: Desmet, G., Nassimbeni, P. and Belli, M. (eds.) *Transfer of Radionuclides in Natural and Seminatural Environments*. Elsevier Applied Science, New York, NY.
9. Artner, C., Gerzabek, M.H., Horak, O. and Muck, K. 1990. Ermittlung praxisbezogener Transferfaktoren für ^{137}Cs und ^{90}Sr aus dem Fallout des Reaktorunfalles in Tschernobyl. *VDLUFA-Schriftenreihe* 32:881-886.
10. Blum, W.E.H., Spiegel, H., and Wenzel, W.W. 1989. Bodenzustandsinventur; Konzeption, Durchführung und Bewertung. Bundesministerium für Land- und Forstwirtschaft, Vienna.
11. Armour, J.D., Ritchie, G.S.P., and Robson, A.D. 1990. Extractable zinc in particle size fractions of soils from Western Australia and Queensland. *Aust. J. Soil Res.* 28: 387-397.
12. Livens, F.R. and Baxter, M.S. 1988. Particle size and radionuclide levels in some West Cumbrian soils. *The Science of the Total Environment* 70:1-17.
13. Maule, C.P. and Dudas, M.J. 1989. Preliminary identification of soil separates associated with fallout ^{137}Cs . *Can. J. Soil Sci.* 69: 171-175.
14. Gerzabek, M.H., Ullah, S.M., and Muck, K. 1989. Cs-137 transfer into plants from contaminated Austrian soils, pp. 196-208. IN: Gerzabek, M.H. (ed.) *Proceedings 19th ESNA Conference Vienna, 1988*. Austrian Research Centre Seibersdorf, Seibersdorf, Austria.
15. Pietrzak-Flis, Z., Szybkowska, R., Indeka, L., and Sunderland, N.R. 1990. Effect of resuspension on transfer of ^{137}Cs to plants, pp. 170-181. IN: 7th Report of the IUR Working Group Soil-to-Plant Transfer Factors. RIVM, Bilthoven, The Netherlands.
16. Haunold, E., Horak, O., and Gerzabek, M. 1987. Umweltradioaktivität und ihre Auswirkung auf die Landwirtschaft. I. Das Verhalten von Radionukliden in Boden und Pflanze. *Die Bodenkultur* 38:95-118.
17. Horak, O., Muck, K., and Gerzabek, M.H. 1990. ^{137}Cs soil to plant transfer factors derived from pot experiments and field studies, pp. 29-36. IN: *Proceedings of an International Symposium on Environmental Pollution Following a Major Nuclear Accident, Vienna, 1989*. International Atomic Energy Agency, Vienna.
18. Garland, J.A. and Pattenden, N.J. 1989. Resuspension and the Chernobyl Accident - A Preliminary Review. Joint CEC-IAEA Meeting on Validation of Model Predictions (VAMP), December 5-8, 1989, Vienna.

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