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Classification of soil systems on the basis of transfer factors of radionuclides from soil to reference plants

Proceedings of a final research coordination meeting organized by the Joint FAO/IAEA Programme of Nuclear Techniques in Food and Agriculture and held in Chania, Crete, 22–26 September 2003

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FOREWORD

The IAEA Basic Safety Standards for Radiation Protection include the general requirement to keep all doses as low as reasonably achievable, taking account of economic and social considerations, within the overall constraint of individual dose limits. National and Regional authorities have to set release limits for radioactive effluent and also to establish contingency plans to deal with an uncontrolled release following an accident or terrorist activity. It is normal practice to assess radiation doses to man by means of radiological assessment models. In this context the IAEA published (1994), in cooperation with the International Union of Radioecologists (IUR), a Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Temperate Environments to facilitate such calculations. The obvious limitation of the Handbook is that the data on soil-to-plant transfer of radionuclides are strictly relevant only to temperate climates. Therefore, the IAEA, together with the United Nations Food and Agriculture Organization and the IUR, conducted a coordinated research project (CRP), to obtain similar data in tropical and sub-tropical regions.

A conclusion of this research was that some combinations of ecosystems and radionuclides do not behave as might be expected. A consultants meeting held in May 1998 produced a proposal for a CRP to address the issue of identifying such situations in temperate as well as tropical and sub-tropical conditions.

The IAEA acknowledges the contributions of all the participants to this CRP and in particular the mentoring of M. Frissel (Netherlands) and work undertaken by R.J. Hance (UK) in reviewing the individual contributions presented at the final research coordination meeting held in Chania, Crete from 22–26 September, 2003.

The IAEA officers responsible for this publication were M.A. Matin, J. Brodesser and I.G. Ferris of the Joint FAO/IAEA Programme of Nuclear Techniques in Food and Agriculture.

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SUMMARY OF THE COORDINATED RESEARCH PROJECT

1. INTRODUCTION

The IAEA, together with the United Nations Food and Agriculture Organization (FAO) and the International Union of Radiologists (IUR), conducted a Coordinated Research Project (CRP) on *Transfer of Radionuclides from Air, Soil and Freshwater to the Foodchain of Man in Tropical and Subtropical Environments*. This produced a set of values for key transfer parameters of radionuclides between the various components of tropical and sub-tropical ecosystems that can be used in dose assessment models. It concentrated on what are considered as the key parameters in assessment models—radionuclide transfer from soil to plant and from freshwater to fish.

A data bank was developed of transfer factors for radionuclides, principally ^{137}Cs and ^{90}Sr, from soil to cereals, fodder crops including grass, legumes, root crops, green vegetables, and plantation crops. Account was taken of soil properties, nature of the contamination (artificial, weapons testing fallout, Chernobyl fallout and so on) and the type of experiment (field, pot or lysimeter) that generated the values.

For soil-to-plant transfer the following conclusions were drawn:

(i) there are no systematic differences between soil-to-plant transfer factors in temperate, subtropical and tropical environments;

(ii) the effect on transfer of a) soil pH, b) nutrient status of the soil and c) time elapsed since the soil was contaminated with radionuclides, is generally independent of the climatic zone;

(iii) there exist, however, ecosystems with a relatively high or low uptake (by a factor 10 or even 100 higher or lower than average values);

iv) a higher or lower uptake condition is nuclide specific; an ecosystem may show a relatively high or low uptake for a particular radionuclide and not at all for other radionuclides;

(v) a higher or lower uptake condition is *not* crop specific. If an ecosystem shows a relatively high or low uptake for one crop, all crops show this behaviour qualitatively.

2. PROBLEM

The earlier CRP *Transfer of Radionuclides from Air, Soil and Freshwater to the Foodchain of Man in Tropical and Subtropical Environments* concluded that the generic values as published in the IUR/IAEA *Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Temperate Environments* [1] may be used as a first approximation in assessment studies, but that site specific deviations of a factor of ten or more must be expected. For more precise assessment studies an investigation to identify the conditions causing local deviations is necessary.

3. BACKGROUND

3.1. Transfer factors

The soil-to-plant transfer factor (TF), or the ratio of the concentration of radioactivity in the crop to the radioactivity per unit mass (sometimes surface area is used) of the soil, is a value used in evaluation studies on the impact of releases of radionuclides into the environment. For much of Europe and the USA, the TFs for most important agricultural products are known. For other areas, and especially the developing countries, TFs are not so readily available. The approach used hitherto, in which TF values are estimated for each crop, is very time consuming and expensive. Therefore, within this CRP, another approach is used based on the development of generic values.

3.1.1. Factors influencing Transfer Factors

The TF values of radionuclides vary enormously. The main factors which cause this variability for any particular radionuclide are the type of crop and type of soil. The length of time the radionuclide has been in the soil is also important, particularly for ^{137}Cs. Other factors are crop variety, agricultural practice (especially fertilization) and differences in the weather during the growing season (not overall climate) (Fig. 1).

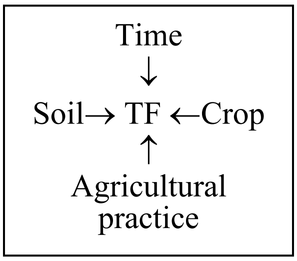


FIG. 1. Main factors affecting TF values.

Soil properties that are likely to affect TF values include mineralogical and particle size distribution, organic matter content, pH and fertility. Cation exchange capacity and the nature of exchangeable bases are important since Cs and Sr radionuclides are cations (Cs^+ and Sr^{2+}).

Comparison of different soils should be made with the same crop but this is not practical because of the range of climates represented in this CRP. The data obtained with the earlier CRP showed that, for Cs, cereals are the best choice while for Sr both cereals and broad leaved green vegetables are suitable. Because it is easier to have the same crop group for both radionuclides, cereals were chosen as the reference crop.

As the availability of Cs decreases with time, the TF for Cs depends not only on soil properties but also on the time that the radionuclide has been present in the soil. A variety of processes is involved, including fixation to soil minerals, incorporation by microorganisms and migration within the rooting zone. Consequently a reference value is contact time dependent. The TF for Cs in non-equilibrium conditions may be up to a factor of 10 higher than for equilibrium conditions. Equilibrium for Cs may take some five years to attain, sometimes longer, although the rate of decrease in TF is greatest during the first three months. The availability of Sr also decreases with time but the effect is much less pronounced than with Cs.

Aside from the inherent characteristics of a crop to take up Cs^+ and Sr^{2+}, TFs are a function of the physiological state of the plant, which is affected by soil fertility (hence agricultural practice), the weather, and its stage of development. In this CRP considering only the radionuclide content of the harvested part of the plant reduces this variability, although for crops such as Swiss chard or grass, which are cut several times a season, the TF remains a function of the stage of physiological development.

3.2. Soil Classification

The base for the classification of soils is the *World Reference Base for Soil Resources* [2] which was published in 1998. Consequently, the first version of the Experimental Protocols used earlier documentation. The changes in classification introduced in 1998 caused difficulties for some participants. Soil classification involves a range of criteria but pedological considerations dominate. It assesses the essential properties of the soil itself including the processes of formation, distribution, mineralogical composition, the organic matter content and the texture. There are 30 Soil Groups:- Acrisols; Albeluvisols; Alisols; Andosols; Anthrosols; Arenosols; Calcisols; Cambisols; Chernozems; Cryosols; Durisols; Ferralsols; Fluvisols; Gleysols; Gypsisols; Histosols; Kastanozems; Leptosols; Lixisols; Luvisols; Nitisols; Phaeozems; Planosols; Plinthisols; Podzols; Regosols; Solanchaks; Solonetz; Umbrisols; Vertisols. They are further subdivided into 121 Lower Level Units, several of which may occur in the same soil group so that 509 Group/Unit combinations are listed in Ref. [2]. It is at the Unit level that criteria include some of agricultural significance such as Eutric (fertile defined as >50% base saturation), Dystric (unfertile, <50% base saturation) and Calcaric (calcareous). However, soils influenced by human activity may be placed in the Anthrosol Group. Given the secondary level of significance allocated to agricultural properties, it cannot be assumed that this classification will be the most appropriate for the prediction of TFs but clearly it must be taken into consideration. Also [2] was designed to accommodate the total range of soils of the world so particular soils may not fit the system well as is noted in the paper of Twining et al. (p.21).[1]

4. OVERALL OBJECTIVE.

The overall objective is to improve the specificity of radiological assessment models. This would lead to:

(1) better planning for emergency response and long-term agricultural countermeasures, particularly in developing countries, through the development of generic data as well as those more relevant to local conditions;
(2) more precise information on environmental parameters to be used when setting limits for authorised discharges from nuclear installations.

5. SPECIFIC RESEARCH OBJECTIVE

The specific objective was to generate data on transfer factors of radionuclides from soil to plants in a range of soil systems so as to characterize systems in which transfer factors deviate substantially from average and to assess the extent it is possible classify soils according to the availability of radionuclides contained in them.

6. APPROACH

The TF in this report is defined as the ratio between the units of radioactivity per unit of mass dry crop and the units of radioactivity per unit of mass dry soil. If not stated otherwise, concentration in the crop refers to the concentration in the edible part at harvest. Concentration in the soil refers to the upper 20 cm of soil for all crops, with the exception of grass for which 10 cm is chosen. This follows the practice used in the IUR/IAEA *Handbook* [1]. The TF can also be based on the contamination per unit soil surface area. The comparison of TFs is not influenced by the way TFs are defined, provided the definitions are not mixed.

The approach makes three assumptions based on published data [1, 3–5] and the future FAO/IAEA/IUR database (Under http://www-naweb.iaea.org/nafa/databases-nafa.html).

i**) The TFs within a crop group, such as cereals, green vegetables, potatoes, and root crops, for a particular soil and radionuclide are the same.** This can only be an approximation because differences between species, varieties and types within one crop group are neglected.

ii) **The ratios between the TFs of different crop groups for a particular radionuclide are constant**. These ratios are called conversion ratios. They allow the TF value for one crop group to be predicted from the TF value of another crop group, provided the crops are cultivated on the same soil. Again, this is a considerable oversimplification.

iii) **Reference TF values can be established for one crop on a range of soils**. TFs depend on soil properties and, as discussed in *3.1.1.*, the TF for Cs decreases with soil residence time. Thus the reference value for Cs is contact time dependant. In practice it may be useful to distinguish between 'Equilibrium Transfer Factors' which represent data useful for routine releases and long lasting contamination and 'Non-equilibrium Transfer Factors' for the initial stages of dealing with acute

[1] All references in the summary without citations refer to papers in this Tecdoc.

contaminations [4, 5]. This CRP is concerned with the classification of soils, so should use equilibrium conditions. Therefore the experimental protocol [Appendix I] recommends equilibration times of at least two months for Cs and one month for Sr which is a compromise between what is desirable and what is practical.

7. THE CRP

The CRP involved 11 Contract holders, three Agreement holders, two IUR observers and two consultants. It lasted five years and Research Coordination Meetings were held in 1998 (Izmir), 2001 (Vienna) and 2003 (Chania).

The experimental protocol is set out in Appendix I. It contains guidance on crop and soil choices, soil preparation, soil sampling, crop production and harvesting. It permits studies both in the field and in pots or lysimeters. Standard data sheets were provided for reporting the results.

All but one participant reported experiments with artificially contaminated soil and six also reported data obtained from soils contaminated by fall-out from either the accidents at Chernobyl and Goiânia or weapons testing. All participants reported data for Cs and Sr for at least two crops on at least two soils.

Interlaboratory comparisons were carried out in 2000 and 2002/3 for the analysis of ^{137}Cs and ^{90}Sr in soil and vegetation samples provided by the IAEA AQCS. Some participants measured other radionuclides present.

8. RESULTS AND DISCUSSION

8.1. General considerations

The investigators reported almost 3000 TF values, mostly for radioisotopes of Cs and Sr but also of Mn, Zn, Po, Pb, Th, U and K, obtained in some 25 soil Lower Level Units from 16 different Soil Groups using 25 crops. Whilst this is a large body of data only about 5% of all Lower Level Units are represented, in some cases by only one set of observations. Hence only tentative steps towards a classification can be taken at this stage.

All mean TF values, per investigator and per soil, are listed in Appendix II. Geometrical means were used because data in the literature consistently show log-normal distributions. It is stressed that the mean values consider all observations even in cases where it might seem advisable to reject certain observations. This Appendix also contains data for other nuclides and includes information about soil properties and crops. The data sheets from which they were calculated are available at ftp://ftp.iaea.org/dist/rifa-trc/Crete/RCM/Frissel/raw-data/.

8.2. Sources of variability in data

8.2.1. Crop cultivation

As discussed in *3.1.1.*, agricultural practices and the weather affect TF values. However, the Protocol did not require comparisons of, for example, fertilizer regimes, cultivation methods or irrigation practices, as this would have made the number of combinations of factors unmanageable. In general, therefore, field experiments followed local agricultural practice, and pot experiments used appropriate procedures to obtain satisfactory crops. As noted in the Protocol, some differences may be expected between field, lysimeter and pot experiments but in this CRP the results from each were considered together. Some investigators did vary one or more factor and their results give an indication of the significance of those variations.

The Protocol was the latest version of one originally written in 1982. Since then the concept of *Good Laboratory Practice* (GLP) for experimental systems has been developed particularly in the context of evaluation of pesticides. There may be scope for incorporating some elements of GLP in any future protocol, especially with regard to method validation.

8.2.1.1 Soil fertilization

Sachdev et al. (p.89) measured Cs TFs at three levels of potassium fertilization on three different soils. The highest level of fertilizer reduced TFs to cereals by up to 33% and to cabbage by about 50%. Similarly Al-Oudat et al. (p.139) found that sorghum, barley, alfalfa and spinach fertilization according to local practice decreased the transfer values of ^{137}Cs by 27–33% by in one soil and by 13–31% for sorghum, barley and spinach in another. Schuller *et al.* (p.59) found K fertilizer reduced Cs TFs to Swiss chard, sweetcorn and cabbage by 30–75% in one soil and 60–75% in another but addition of Ca did not affect Sr TFs.

Soil microbiological activity has not previously been considered but Twining et al. (p.21) draw attention to this as a possible source of variation.

8.2.1.2. Weather

Weather data were not systematically collected although most investigators record some details in their reports. There is no obvious procedure with which to assess the effect of weather on the TFs measured in such a relatively small data set. Djingova (p.51) found TFs to winter cabbage were higher than those to summer cabbage, presumably because it takes longer for the winter crop to reach maturity, an indirect effect of weather. Schuller et al. (p.59) noted that TFs to Swiss chard, which is cut several times per season, declined through the year although it is not clear whether or not this should be interpreted as a weather effect.

8.2.1.3. Crop variety

Results from Robison (p.179) give an idea of the spread obtained with a specific variety within this CRP. Four types of (rather unusual) vegetables were investigated at the same time and on a soil which was contaminated long ago, so that differences caused by fixation of Cs are absent. The mean TFs from the Bikini atoll albic Arenosol for KK Cross, Amaranth, Won Bok and Kai Choi were 52, 21, 99, 24 (Bq kg^{-1} crop/Bq kg^{-1} soil), respectively, i.e., a range of 5 fold. The values for individual samples ranged from 15 to 253, a variation of a factor 17, illustrating the need to use mean values at all times. The variation is typical for this type of experiment. For example Wasserman et al. (p.39) found the TF for ^{40}K varied by a factor of 10, both for maize and for cabbage. Skarlou et al. (p.81) found that TFs to cabbage variety Brunswick from soils were 2.5, 1.3, 1.3 and 2.2 times those to variety Kozanitiko, and corresponding figures for sweetcorn cultivars Vilmorin and Elite were 1.3, 0.5, 2.8 and 2.2 so in these cases the effect was relatively small.

8.2.2 Analytical variability

TF values are calculated from measurements of radionuclide content in soil and crop so analytical reliability is crucial. Therefore inter-laboratory proficiency tests were carried out in 2000 and 2002/3. In 2000, reported analytical values for reference soil and grass samples were compared with the ranges established by the IAEA AQCS which was the practice at that time. The outcome was highly satisfactory as the data from all participants fell within the ranges.

The proficiency test in 2002/3 was run in accordance with ISO 17025 even though the new requirements obliging laboratories to express their measurement uncertainty had not yet come into force. The procedures calculate scores for accuracy and precision both of which require the laboratory to calculate the uncertainty associated with a measurement. For Cs in both spiked soil and cabbage samples 17 out of 26 measurements were acceptable. The corresponding figures for Sr were 6 out of 15, again being the same for both soil and cabbage. At first sight these results are alarming, not least because it implies laboratory performance has deteriorated between 2000 and 2003. However, it emerged at the final RCM that several people had difficulty in estimating the components that combine to make up the total uncertainty and an error in this factor has a major effect on the scores for precision and accuracy (Appendix III). This is perhaps unsurprising given that this is the first time that the new ISO 17025 requirement had been used in a CRP but it does highlight an aspect that needs attention by those concerned with analytical quality assurance and control.

8.2.3. Overview of variability

The foregoing outlines some of the factors that introduce variability into TF calculations and is not comprehensive. For analytical measurements there is a procedure for calculating uncertainty albeit one which not everyone has yet learned to apply. For the other sources of variability mentioned here there is no such procedure other than to apply classical statistical methods where appropriate. Thus, the evaluation of the results of this CRP has to recognize that there will be variability in the data of uncertain magnitude.

8.3. Cs and Sr Transfer factors for crop groups

As stated in Section 6 it is assumed that the mean TFs within a certain crop group (such as cereals, green vegetables, potatoes, root crops, etc.), for a particular soil and radionuclide are the same. This may seem over-optimistic as there are many reports in the literature of unexpected values but this CRP required observations to be made over at least three years in order to obtain representative values.

8.4. The conversion ratios of Cs and Sr.

The assumption is (Section 6) that the ratios between the mean TFs of different crop groups for a particular radionuclide and a particular soil are constant. Data obtained previously [1,3] are summarized in Table 1.

TABLE 1. RATIOS FOR Cs AND Sr FOR CONVERSION OF REFERENCE-CEREAL-TF VALUES TO TF VALUES OF OTHER CROP GROUPS [From 1,3]

	Cereals	Cab-bage	Green vegetables	Legumes, pods	Tuber	Root crops	Grass	Fodder	Onions
Cs Conversion ratio	1	7	9	5	4	3	4.5	4	1
Sr Conversion ratio	1	12	10	5	1	7	5	4	7

All conversion ratios TFcabbage/TFcereals which could be derived from the observations made in this CRP, for Cs and Sr, are listed in Tables 2 and 3, respectively. The values are averaged over time, for different soils (as specified by the various investigators). Single observations were neglected.

For Cs, a ratio of 7 was expected (Table 1) and the majority of the ratios observed range indeed between 0.9 and 14. There is a trend to higher ratios in temperate climates. The Cs-ratios reported by Sachdev et al. (p.89) are among the lowest yet the observations were made on a large number of samples, on three soils over three years and they are very consistent. The Cs-ratios observed by Nguyen (p.191) are much higher than expected. This might be caused by fixation problems, as will be discussed later but Wang (Personal Communication) also observed a higher ratio but with a soil which was labelled nine years previously. Perhaps the explanation may be related to the fact that rice was the cereal in each case, suggesting that perhaps rice should be considered separately.

For Sr, a ratio of 12 was expected (Table 1). The observations range from 0.71–203. Because fixation is not a major issue with Sr, fixation cannot be the explanation. The validity of the conversion ratio rule for Sr seems to be limited.

TABLE 2. CONVERSION RATIOS FOR TF-CABBAGE/TF-CEREALS FOR Cs

Investigator	1st crop	2nd crop	1st TF	2nd TF	Ratio	Expt. No	Tex-ture	Soil unit	Contact time
Al-Oudat	cereals	cabbages	0.0013	0.011	8.2	A101	L	Xerosol	7–30 m
Al-Oudat	cereals	cabbages	0.0022	0.014	6.2	A102	C	Yermosol	7–30 m
Djingova	wheat	cabbage	0.021	0.008	3.1	D101	L	Chromic Luvisol	9–48 m
Djingova	wheat	cabbage	0.007	0.053	7.5	D103	L	Eutric Fluvisol	9–48 m
Li Jiango	wheat	cabbage	0.001	0.035	35	L101	L	Cambisol	10–34 m
Li Jiango	maize	cabbage	0.0023	0.035	15	L102	L	Cambisol	10–36 m
Prister	wheat	cabbage	0.055	0.17	3.1	P101	C	Chernozem	1.4 y
Prister	wheat	cabbage	0.006	0.012	2.0	P102	C	Chernozem Acrisols	5 y
Prister	wheat	cabbage	0.003	0.007	2.3	P103	C	Chernozem	16 y
Prister	wheat	cabbage	0.19	0.57	3.0	P104	S	Fluvisol	1.4 y
Prister	wheat	cabbage	0.032	0.081	2.5	P105	S	Fluvisol Acrisols	5 y
Prister	wheat	cabbage	0.012	0.031	2.6	P106	S	Fluvisol	16 y
Nguyen [a]	rice	cabbage	0.0041	0.047	12	Q101	L	Eutric Fluvisols	8–36 m
Nguyen [a]	rice	cabbage	0.144	3.80	26	Q102	S	Ferric Acrisols	8–36 m
Nguyen [a]	rice	cabbage	0.0016	0.243	152	Q103	C	Thionic Fluvisols	8–36 m
Nguyen [b]	rice	cabbage	0.0012	0.005	4.3	Q104	L	Eutric Fluvisols	16–36 m
Nguyen [b]	rice	cabbage	0.0457	2.08	46	Q105	S	Ferric Acrisols	16–36 m
Nguyen [b]	rice	cabbage	0.0016	0.022	14	Q106	C	Thionic Fluvisols	16–36 m
Robison	sorghum	mixed vegs	15	44	2.8	R101	S	Albic Arenosol	46 y
Sachdev	wheat	cabbage	0.0078	0.0061	0.78	H102	S	Ferralsol	7–13 m
Sachdev	mixed	cabbage	0.0064	0.0099	1.5	H104	S	Fluvisol	7–13 m
Sachdev	wheat	cabbage	0.0083	0.0070	0.84	H106	C	Vertisol	7–13 m
Sanzharova	cereals	cabbage	0.005	0.021	4.0	Z102	C	Chernozem	40 y
Sanzharova	cereals	cabbage	0.007	0.024	3.4	Z103	C	Chernozem	15 y
Sanzharova	cereals	cabbage	0.031	0.057	1.8	Z104	P	Histosol	15 y
Sanzharova	cereals	cabbage	0.013	0.023	1.8	Z105	C	Phaeozem	15 y
Sanzharova	cereals	cabbage	0.014	0.041	2.9	Z106	C	Chernozem	15 y
Sanzharova	cereals	cabbage	0.008	0.023	2.9	Z107	C	Kastanozem	40 y
Sanzharova	cereals	cabbage	0.015	0.035	2.3	Z108	L	Chernozem	15 y
Sanzharova	cereals	cabbage	0.027	0.043	1.6	Z109	S	Podzol	15 y
Schuller	maize, grain	cabbage	0.03	0.26	8.7	C101	L	Dystric Fluvisol	5–37 m
Schuller	maize, grain	cabbage	1.02	0.19	9.5	C103	L	Umbric Andosol	5–37 m
Skarlou	cereals	cabbage	0.072	0.47	6.6	S101	L	Andosol	8–24 m
Skarlou	cereals	cabbage	0.061	0.27	4.5	S102	S	Andosol	8–24 m
Topcuoglu	maize, grain	cabbage	0.0015	0.0049	3.3	O101	L	Leptosol	4–20 m
Topcuoglu	maize, grain	cabbage	0.0013	0.0036	2.8	O102	S	Leptosol	22–39 m
Twining	sorghum	mung, seed	0.042	0.060	1.4	T101	C_L	Arenic acrisol	5–42 m
Twining	sorghum	mung, seed	0.049	0.112	2.3	T102	C_S	Arenic Acrisol	5–42 m

Wasserman	maize,grain	cabbage	0.38	1.34	3.5	W101	S	Acrisol,	20 m
Wasserman	maize,grain	cabbage	0.069	1.28	19	W102	L	Nitisol,	15 y
Wasserman	maize,grain	cabbage	0.093	0.94	10	W103	L	Nitisol, Ferralsol	8 y
Wang	rice	cabbage	0.007	0.32	46	G1	C	Ferralsol	9 y

[a] Complete sampling period [b] Samples first year not considered

TABLE 3. CONVERSION RATIOS FOR TF-CABBAGE/TF-CEREALS FOR Sr

Investigator	1st crop	2nd crop	1st TF	2nd TF	Ratio	Exp. No	Tex-ture	Soil Unit	Contact time
Al-Oudat	cereals	mixed veg	0.036	1.43	40	A202	L	Yermosol	
Al-Oudat	cereals	mixed veg	0.024	1.19	50	A201	C	Xerosol	
Li Jiango	wheat	cabbage	0.010	0.99	99	L102	L	Cambisol	18–36m
Li Jiango	maize	cabbage	0.003	0.99	330	L103	L	Cambisol	18–36m
Prister	wheat	cabbage	0.089	0.152	1.9	P121	C	Chernozem	1.4 y
Prister	wheat	cabbage	0.061	0.073	1.2	P122	C	Chernozem	8 y
Prister	wheat	cabbage	0.020	0.038	1.9	P123	C	Chernozem	15 y
Prister	wheat	cabbage	0.069	1.25	1.7	P124	S	Fluvisol	1.4 y
Prister	wheat	cabbage	0.31	0.98	3.2	P125	S	Fluvisol	8 y
Prister	wheat	cabbage	0.10	0.31	3.3	P126	S	Fluvisol	15 y
Nguyen	rice	cabbage	0.17	12	70	Q111	L	Eutric Fluvisols	3–20 m
Nguyen	rice	cabbage	0.32	29	89	Q112	S	Ferric Acrisols	3–20 m
Nguyen	rice	cabbage	0.16	6.8	43	Q113	C	Thionic Fluvisols	3–20 m
Sachdev	wheat grain	cabbage	0.3	0.39	1.3	H201	S	Fluvisol	7–60 m
Sachdev	wheat grain	cabbage	0.69	0.49	0.71	H202	C	Vertisol	7–60 m
Sachdev	wheat grain	cabbage	0.50	0.46	0.92	H203	S	Ferralsol	7–60 m
Sanzharova	barley	cabbage	0.19	0.92	4.9	Z121	L	Luvisol	50 y
Sanzharova	barley	cabbage	0.04	0.14	3.7	Z122	C	Chernozem	40 y
Sanzharova	barley	cabbage	0.05	0.34	6.6	Z123	C	Chernozem	50 y
Sanzharova	barley	cabbage	0.39	2.09	5.3	Z124	P	Histosol	15 y
Sanzharova	barley	cabbage	0.14	0.78	5.7	Z125	L	Phaeozem	15 y
Sanzharova	barley	cabbage	0.08	0.50	6.4	Z126	C	Chernozem	15 y
Sanzharova	barley	cabbage	0.10	0.69	6.9	Z127	L	Chernozem	15 y
Sanzharova	barley	cabbage	0.14	0.84	6.1	Z128	S	Podzol	15 y
Schuller	maize, grain	cabbage	0.032	3.8	119	C201	L	Dystric Fluvisol	5–37 m
Schuller	maize, grain	cabbage	0.04	7.3	183	C202	L	Umbric Andosol	5–25 m
Topcuoglu	maize,grain	cabbage	0.014	0.88	62.9	O201	L	Leptosol	4–6 m
Topcuoglu	maize,grain	cabbage	0.013	1.07	82.4	O202	S	Leptosol	4–6m
Twining	sorghum	mung seed	0.20	1.72	8.4	T103	C_L	Arenic Acrisol	5–42 m
Twining	sorghum	mung seed	0.17	1.25	7.3	T104	C_S	Arenic acrisol	5–42 m
Wasserman	maize gr.	cabbage	0.078	2.0	26	W11	S	Acrisol	4–20 m
Wasserman	maize gr.	cabbage	0.020	0.57	29	W13	S	Ferralsol	4–20 m
Wasserman	maize gr.	cabbage	0.0050	0.13	26	W15	S	Nitisol	4–20 m

8.5. The influence of soil properties on Cs reference transfer factors

Table 4 shows reference TFs derived from the IUR database and values obtained in the earlier CRP. This combined data set is larger than that obtained in this CRP so any classification developed here should therefore not conflict with this Table. Unfortunately, with a few exceptions, soil groups as defined by the FAO classification are not listed in the IUR database and most investigators are no longer available. With the earlier CRP the situation is better, but not optimal. Also in most cases the IUR database does not include exchangeable K. Only the soil texture is almost always reported, as is the notation, P, for peat soils, i.e. Histosols in the FAO classification. Further, it would be desirable to estimate the influence on TF of different conditions such as the time that radionuclide was in contact with the soil, fertilization, irrigation, soil management and the weather but these factors were not systematically recorded.

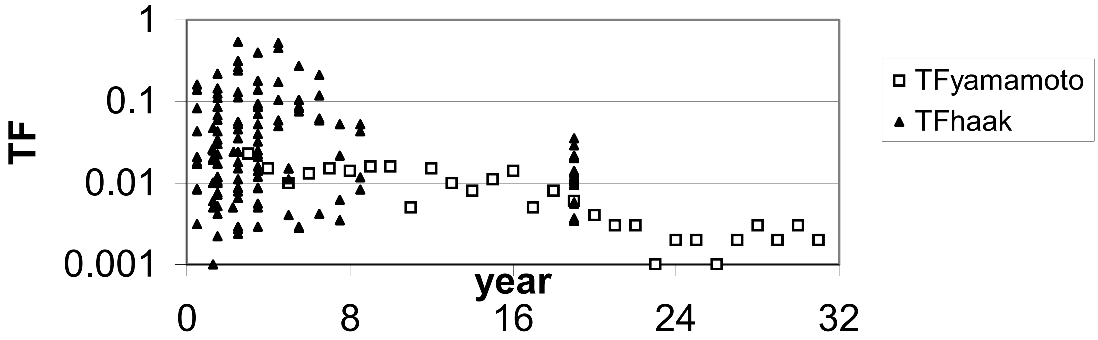
TABLE 4. REFERENCE TRANSFER FACTORS OF Cs FOR CEREALS UNDER EQUILIBRIUM CONDITIONS (Bq kg^{-1} dry crop/Bq kg^{-1} soil in the upper 20 cm of soil)

	Nutrient status of soil	Soil texture		Reference TFs of Cs	
				Expected value	Range
Soil group 1	High with pH > 4.8	All soils		0.006	0.002–0.01
Soil group2	Medium	Clay and loam soils		0.03	0.01–0.1
	pH > 4.8	Sand, peat and other soils		0.05	0.02–0.1
Soil group 3		Clay soils		0.2	0.1–0.5
		Sand and other soils		0.3	0.1–1
	Very low with pH>4.8			0.3	0.1–1
	or pH < 4.8	Peat soils	Normal moisture	0.6	0.2–2
			Wet, gleyic	3	1–10
Soil group 4	Soils with exchangeable K<0.05 cmol(+)/kg			20	10–50

8.5.1. Contact time and fixation

Fixation of Cs is a serious problem. Fixation is the partly irreversible, slow adsorption of Cs, especially to clay minerals. Usually fixation is most important in the first few years after contamination of the soil, but the amount that will be fixed differs from soil-to-soil. The process may continue for a long time but the kinetics of the process has not been characterized numerically. This CRP was not designed to address this issue but it is clearly something that requires further investigation.

One would hardly expect to see fixation effects on soils contaminated after the Chernobyl accident which occurred 13 to 15 years before the measurements were made and the results of Sanzharova et al. (p.113) and Prister et al. (p.153) are broadly consistent with this view. Even so, values reported by Prister et al. (p.153) suggest there may still be a slight decline in availability even after five years (Figs 4 and 5 in their paper). These Figs also show that Cs TFs decline between one and two orders of magnitude over five years. Similar changes occur in other data sets (Fig. 2).



Legend: TFyamamoto = FAO/IAEA/IUR Workgroup collected data by Yamasaki [7], TFhaak = IUR database (available at ftp://ftp.iaea.org/dist/rifa/Crete/RCM/Frissel) TF in (Bq kg^{-1} crop)/(Bq kg^{-1} soil)

FIG. 2. Influence of the time elapsed since the Cs contamination of soil on the TF for cereals.

Most, but not all, investigators in this CRP working with recently contaminated soil observed effects that may be caused by fixation. As the CRP covered 4–5 years, at the end of this period most of the effect of fixation should have occurred. In those cases where fixation occurred, the first year's results were obtained in conditions that were far from equilibrium so it is possible to argue that they should be omitted from the analysis. This was not done because by averaging over time variations were reduced and on balance, it was decided that reducing the data set in this way would introduce other inaccuracies. For emergency response planning it could be argued that non-equilibrium data are more useful than those obtained at equilibrium.

The results of Topcuoğlu et al. (p.145) raise an interesting complication as they found the TF to black cabbage was higher in Chernobyl contaminated soil than in recently artificially contaminated soil. They attributed this to the presence of larger quantities of stable Cs in the latter case, a possibility which was not otherwise investigated in this CRP.

8.6. Preliminary classification by Cs TFs

Figure 3 compares data from this CRP ('new') with those from the earlier CRP and the IUR database ('old') and shows reasonable concordance.

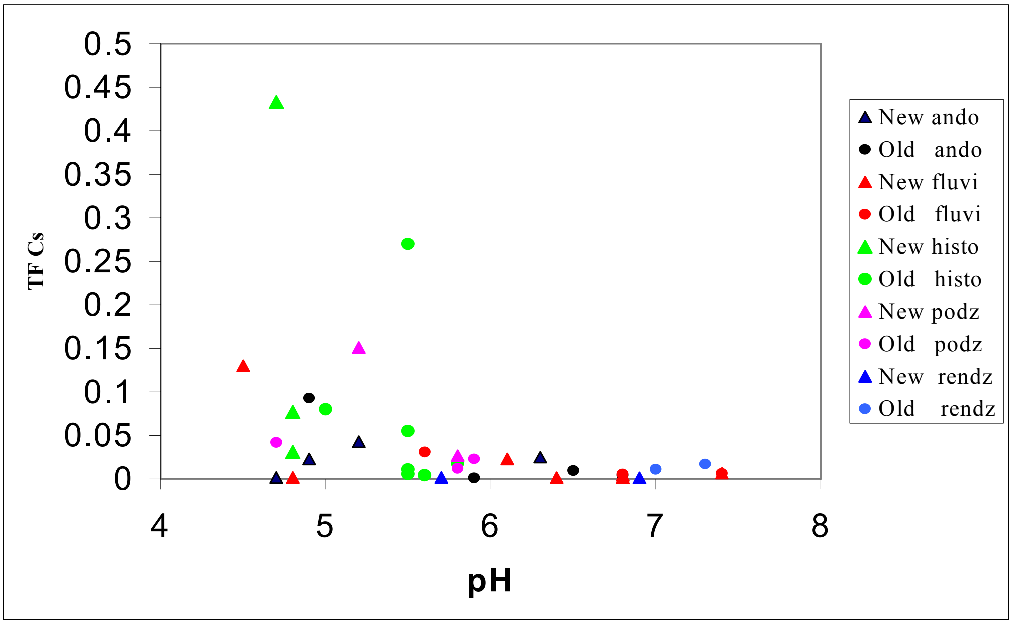


FIG. 3. Comparison of old and new Cs TFs.

As stated earlier any new classification should not conflict with Table 4. Figures 4 and 5 show that this requirement is met. They include all Cs TF values for soil units of the CRP which correspond with groups 1 and 2 of Table 4. Maximum and minimum values for expected TF values were fitted by eye (because the data set is too limited for a statistical procedure, see 8.1). They form the core of this classification. Figure 4 also shows the limits presented in Table 4. The new limits cover a slightly wider range than the old limits, undoubtedly because a wider geographical range is covered by this CRP. Some values in Figs 4 and 5 are outside the expected range, the high values may be caused by delayed fixation which is a reason for not adjusting the limits

For soils in Group 1, TF seems not to be related to texture but Group 2 TF`s, which are considerably higher, do seem to be related to the texture. This is in agreement with the groupings in Table 4 but there nutrient status was a leading criterion whereas here soil units are shown.

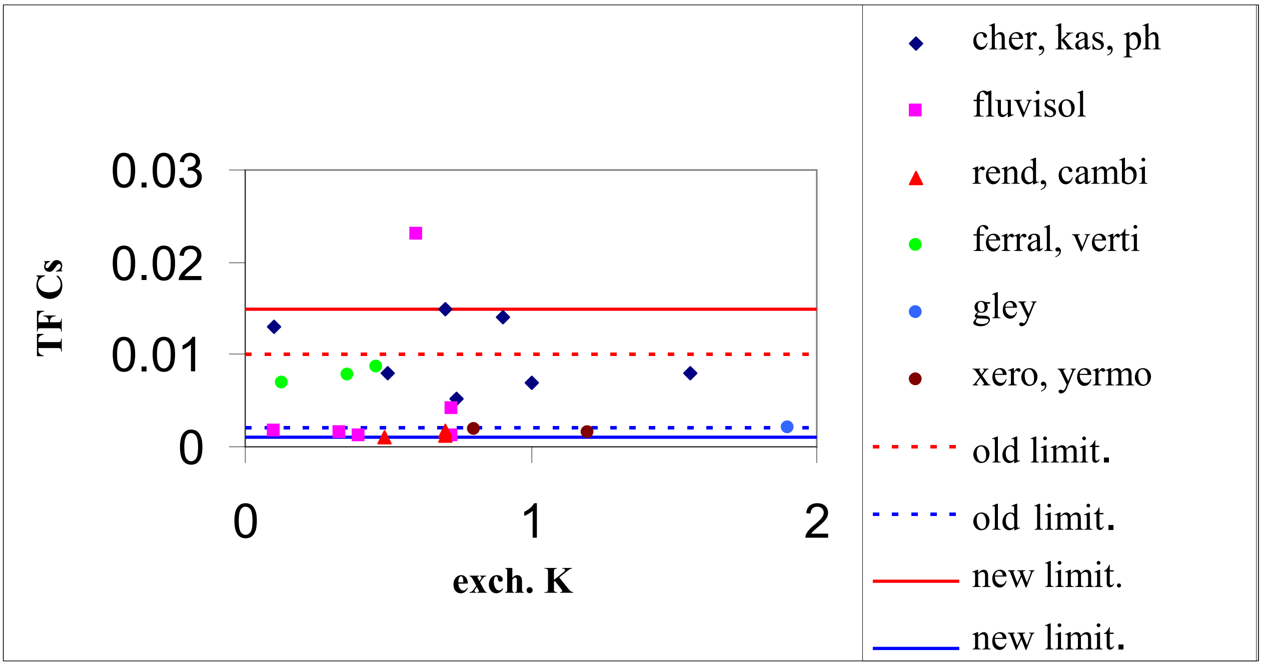


FIG. 4. Comparison of old and new Cs TF values for Soil Group 1.

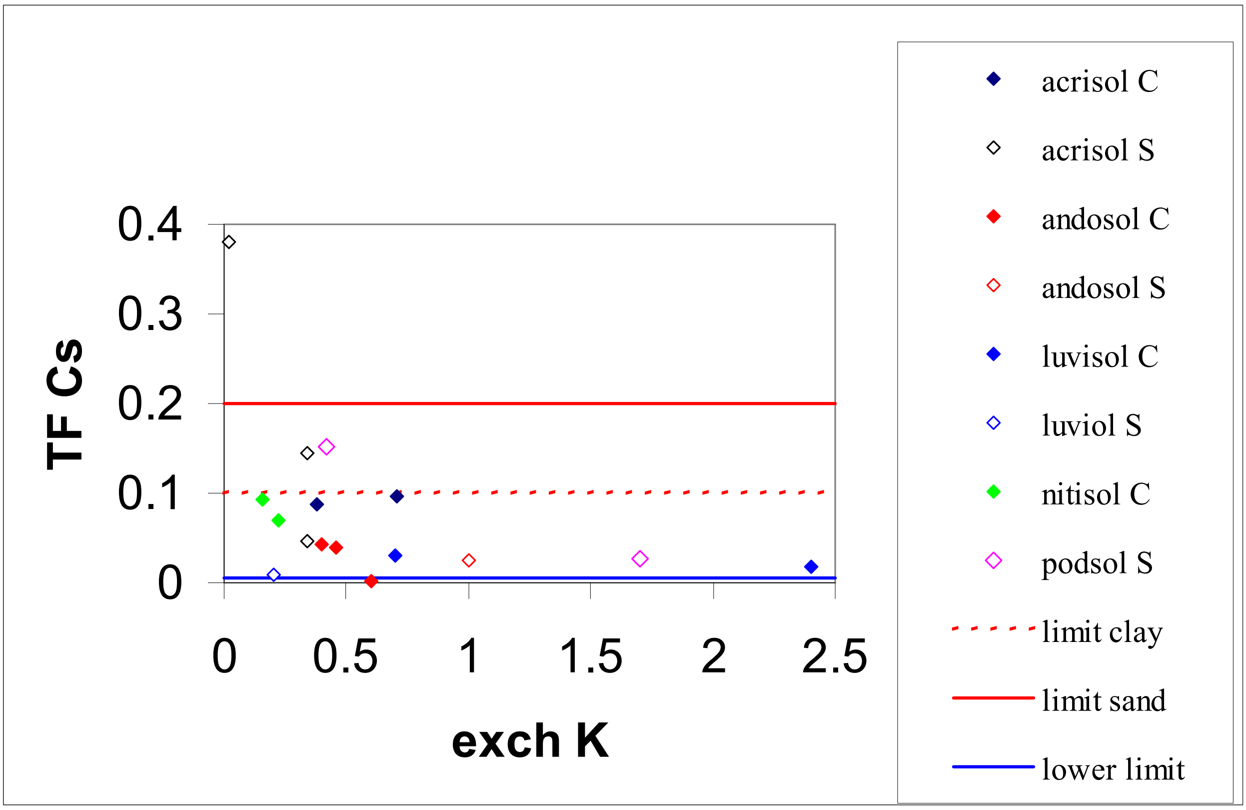


FIG. 5. Comparison of old and new Cs TF values for Soil Group 2.

The Histosols and Histic Gleysols seem to deviate from the general pattern. Therefore Histosols, Histic soils and Humic soils are treated as a separate group, Soil group 3 (hereafter all called Histosols). The Arenosol from the Bikini atoll should, because of its deviating character, be classified as a separate soil group, Soil group 4. The soil deviates because of its low exchangeable K content and probably total absence of illite. Soils which are less suitable for agricultural production (the old soil group 3), now form Soil group 5. None of the participants had a soil in this group.

These considerations lead to the following classification of soils for the uptake of Cs by cereals (Table 5).

TABLE 5. PRELIMINARY CLASSIFICATION OF SOILS BY Cs TFs TO CEREALS

Soil group		Expected transfer factors for Cs (Bq kg^{-1} dry crop/Bq kg^{-1} dry soil upper 20 cm)	
1	Cambisol Chernozem Ferralsol Fluvisol Gleysol Kastanozem Leptosol Phaeozem Vertisol	Lower limit	0.001
		Upper limit	0.015
2	Podzol Luvisol Andosol Acrisol Nitisol	Lower limit (all)	0.005
		Upper limit clay	0.1
		Upper limit sand	0.2
3	Histosol	Lower limit	0.005
	Humic soils; Histic soils	Upper limit	0.5
4	Albic Arenosol	Upper limit	5
5	Less suitable soils	Upper limit	High

As discussed earlier, this classification is imprecise because many of the TF measurements included values obtained before complete equilibrium of Cs distribution in the soil had been reached and only limited data are available for many soil groups. Consequently, the lower limits of expected TFs for Groups 2 and 3 are the same and overlap the range of Group 1. However, for planning purposes it would be prudent to use the upper limits in which case there are reasonably clear distinctions. Therefore these results give promise that soils could be classified in this way but very many additional data are required for confirmation.

8.7. The reference TF for strontium

In addition to pedological considerations, properties such as organic matter, soil acidity, calcium carbonate content and texture are used to define many Soil Groups and Lower Level Units. All these properties influence the uptake of Sr so it should be possible to group soil units on the basis of reference Sr TF values. Figure 6 shows the TFs as a function of the exchangeable Ca.

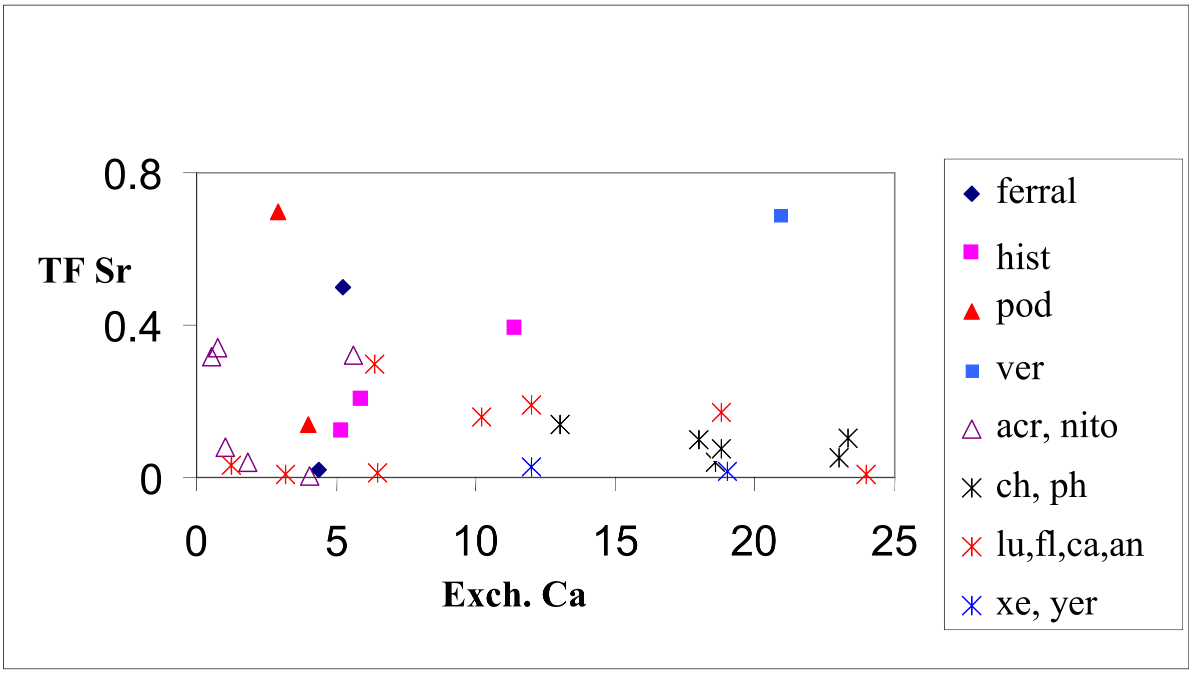


FIG. 6. Cs TFs as a function of the exchangeable Ca.

As expected the acid Acrisols and Podzols have lower TFs than soils with moderate acidity such as Chernozems. It is tempting to consider two soil groups, Ferralsols, Histosols, Podzols and Vertisols which show higher TF`s then the other soils (Acrisols, Nitisols, Chernozems, Phaeozems, Luvisols, Fluvisols, Cambisols, Andosols, Gleysols), but the number of soils investigated is too small to do this.

Moreover, the scheme in Table 4 uses clay, sand and peat contents as the main criteria. Therefore, Fig. 7 distinguishes TF values on this basis. For clay/loam and sand Histosols, it gives a reasonable fit. The Vertisol does not follow the general trend but it concerns one only soil.

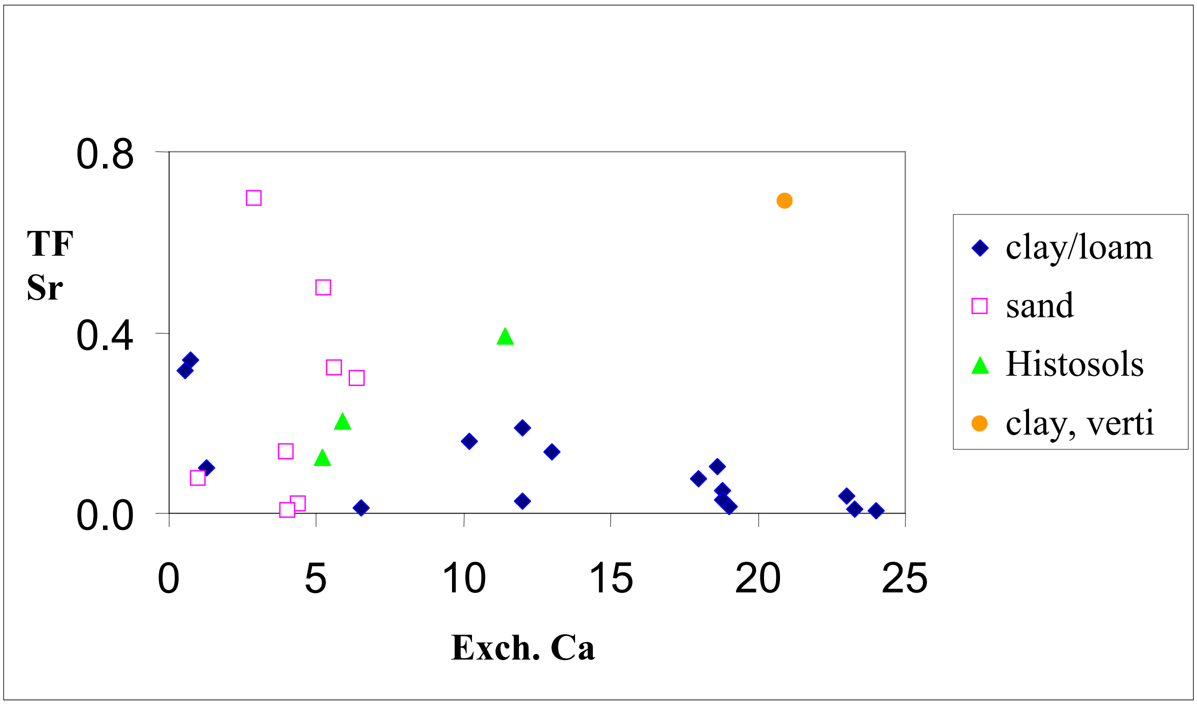


FIG. 7. Relation between soil texture and Sr TF.

Figure 7 is not very conclusive because it does not contain enough observations, therefore Fig. 8 includes those systems which could be derived from the earlier CRP and the IUR data.

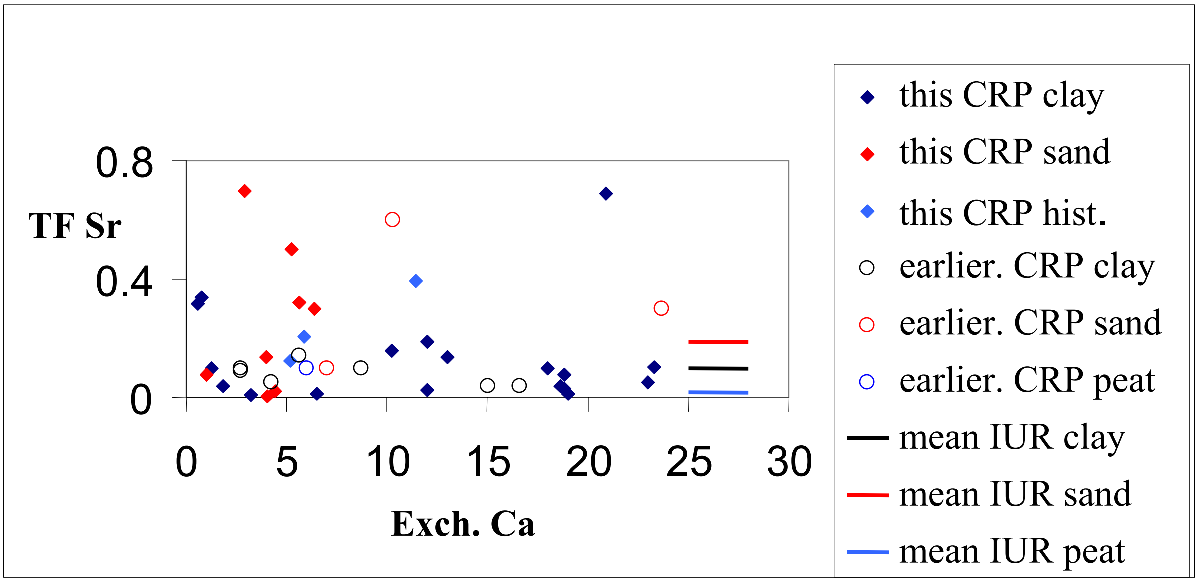


FIG. 8. Relation between soil texture and Sr TF including old data.

IUR data do not include exchangeable Ca, therefore only the mean value of the TF`s for each soil texture is given. The agreement for clay/loam and sand soils is good but it is poor for peat soils (not shown).

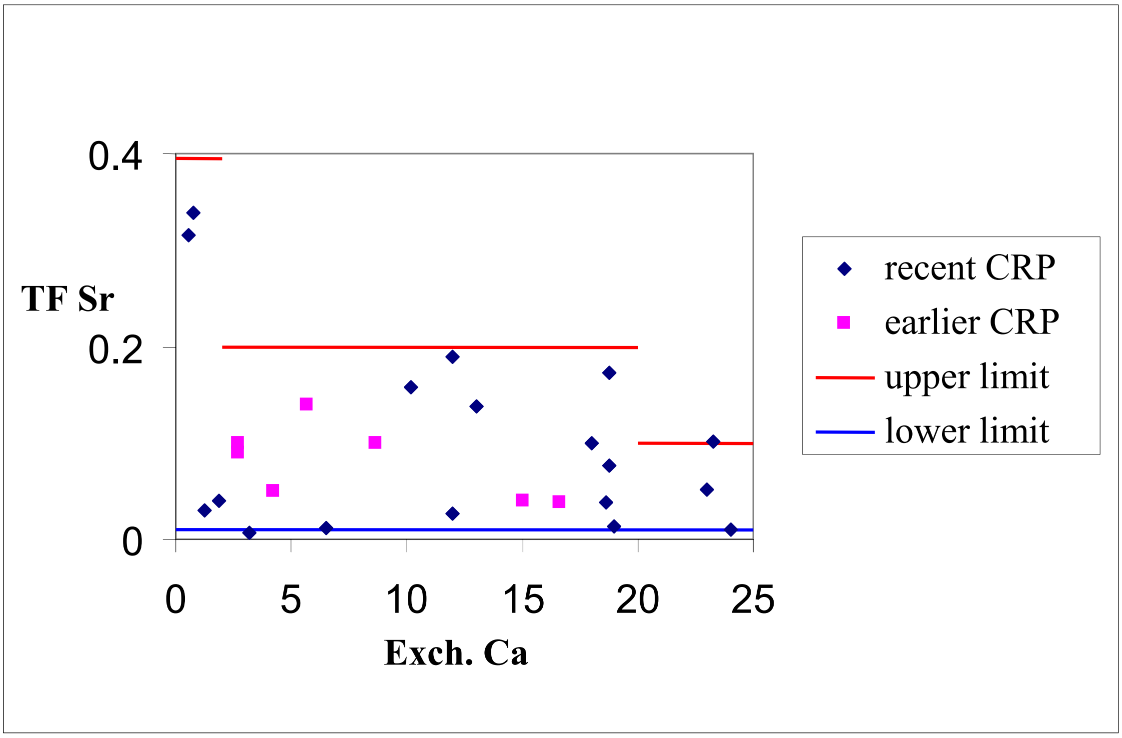


FIG. 9. Sr TFs for clay and loam soils.

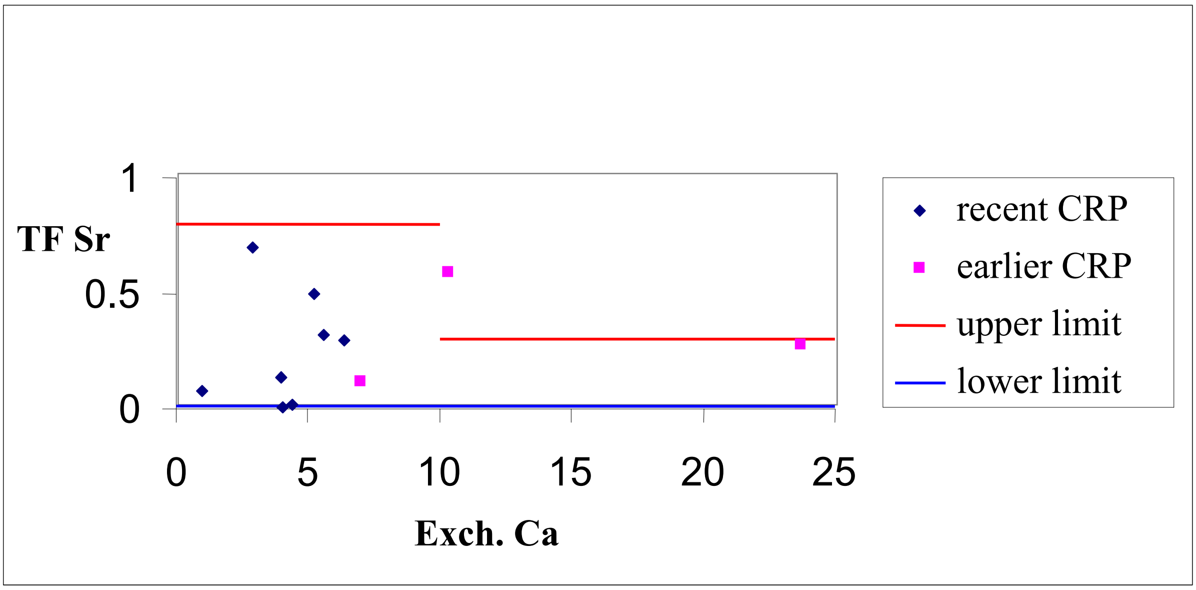


FIG. 10. Sr TFs for sandy soils.

Figures 9 and 10 show the TF values for clay and loam soils and sandy soils respectively together with the expected maximum and minimum values which are dependent on exchangeable Ca. The ranges are narrower than when using IUR data alone. For peat soils the IUR range was 0.01–0.06. The maximum (mean) value is now 0.4 for a Dystric Histosol; most IUR soils were Eutric Histosols. This might indicate that it is necessary to distinguish between Eutric systems and Dystric systems. There are insufficient data to make this distinction.

8.8. Preliminary classification by Sr TFs

The foregoing considerations lead to the following classification of soil for the uptake of Sr (Table 6).

TABLE 6. PRELIMINARY CLASSIFICATION OF SOILS BASED ON Sr TF TO CEREALS

	Expected transfer factors for Sr (Bq/kg dry crop)/(Bq/kg dry soil upper 20 cm)		
Group 1	Texture	Exch. Ca	Range TF
Acrisol Andosol	Clay and loam	<20	0.01–0.2
Cambisol Chernozem		>20	0.01–0.1
Yermosol Luvisol	Sandy soil	0–10	0.01–0.8
Nitisol Fluvisol		>10	0.01–0.3
Phaeozem Xerosol			
Gleysol Ferralsol			
Podzol			
Group 2			
Histosol			0.01–0.4
Group 3			
Vertisol			0.01–0.8

8.9. Comments on the preliminary classifications

There were enough data sets to give a fair insight into the behaviour of Cs for some soil units (Fluvisols, Luvisols, Chernozems and Podzols) but others were only represented by one set of data. It is possible that one set of data is not representative. It was assumed that all Lower Level Units belong to the same Soil Group but there is no proof. From some Units there was no representative at all. With soil group 1 (Cs classification) the majority of data refer to old contaminations, while for soil group 2) the data refer to recent contaminations; this could have unbalanced the choices made. The Sr classification is based on very limited data and, in addition, the measurement of Sr in some cases contained a large degree of uncertainty (See 8.2.1).

The range of TFs is rather wide; the higher values occurring with the most acid and Dystric classes, the lower values in the Calcaric and Eutric classes. At the moment there is not sufficient information to go into more detail. Within Soil group 2 (Cs classification) there are two Eutric and one Dystric Units listed. The TF of the latter Unit is lower that of the former two but one cannot derive a rule from three sets of observations. One might think of a statistical analysis within a Soil Group but lack of quantitative data on the individual factors affecting TF values precludes this.

Despite this, the CRP made a successful beginning with the classification and it is worthwhile to consider possible applications.

8.9.1. Predictions

Based on the results of the CRP approximate TF estimations for Cs and Sr can be made following a stepwise procedure.

(1) Define the soil and ascertain to which Soil Unit it belongs. (currently there are data for only 25 Soil Units).
(2) Decide on the soil texture as in most cases the texture together with the Soil Group defines the value of the reference TF in the classification system.
(3) Determine the Reference TF (for cereals). It depends on the purpose of the prediction if one selects the maximum value or the most probable value.
(4) For crops other than cereals apply a conversion factor to the reference cereal TF to obtain an order of magnitude estimate.

8.10. Other approaches to assess the data.

Three participants applied various mathematical procedures to their data.

Nguyen (p.191) used a stepwise multiple regression analysis to identify which parameters are most significant in explaining the variations of TF. In general, the equation constant, CEC, OM, pH, contamination time and concentration of radionuclides in soil explained more than 80% of TF variance although they are a little different from crop-to-crop. The correlation between the experimentally determined ^{134}Cs TF and soil properties was not quite linear and was improved using logarithmic TF values. These results are encouraging but as only three soils were involved they are not conclusive.

Sanzharova et al. (p.113) also used multiple regression analysis to estimate the extent to which each soil characteristic is related to other soil properties and to radionuclide TFs. They also noted that such correlations are not always linear but used linear relationships where possible. In addition they used factor analysis to consider a whole range of relations between soil characteristics and TFs to identify significant factors. Using these analyses they proposed two equivalent schemes of soil classification based on ^{137}Cs TFs to barley. Both involved three soil parameters two of which, soil group and mechanical composition, were common to both schemes. The third parameter was pH value in one case and exchangeable K in the other. For soil classification based on ^{90}Sr TFs, they identified three schemes. All included soil group plus a combination of two other parameters taken from exchangeable Ca, mechanical composition and organic matter content. These proposals are based on a large data set including soils contaminated by the Chernobyl accident and fall-out from weapons testing so are statistically robust and use observations made in equilibrium conditions.

Prister et al. (p.153) adopted a different approach based on the concept of the complete estimation of soil properties (CESP). They considered that the most important factors controlling the availability of ions in the soil to plants are ion absorption capacity, organic matter content and pH, thus reducing the

CESP to three components. Because each component is measured in different units their values were normalized with respect to a neutral pH (=7) and the highest values for adsorption capacity and organic matter content of the range of soils considered (in their case 40 meq/100 g and 6%, respectively). Thus, the area of the triangle with sides of normalized values of pH, organic matter and adsorption capacity can be calculated to give a dimensionless area designated *Sef.* They showed good, though non-linear, relationships between *Sef* values and Cs TF values in lucerne, tomato fruit, potato tubers and grasses as well as Sr TFs for tomato fruit and potato tubers.

The statistical and CESP approaches are attractive because they identify and make use of factors known to be involved in controlling the availability of cations in the soil to plants. The data they need are easy to obtain routinely and require no specialist knowledge of soil classification. Their further development and validation is of great interest.

9. CONCLUSIONS

(1) This CRP has shown that it should be possible to arrange the Soil Units defined in the FAO *World Reference Base for Soil Resources* [2] into groups based on TFs for radionuclides. In the case of Cs, five groups have been tentatively identified but more data are needed to confirm this. Although the divisions between groups are not precise they will probably give a useful guide for emergency planning purposes. For Cs the classification is complicated by slow soil fixation processes that take several years to reach equilibrium. Because only limited data are available only three groups could be identified on the basis of Sr TFs. At the moment these groupings are probably too tentative to be used in emergency planning.

(2) Three assumptions made at the outset of the programme:

(i) the TFs within a crop group, such as cereals, green vegetables, potatoes, and root crops, for a particular soil and radionuclide are the same;
(ii) the ratios between the TF`s of different crop groups for a particular radionuclide are constant;
(iii) TF values for all other crops can be calculated from a reference TF for one crop.

All were justified for Cs, within experimental error, which admittedly is sometimes considerable. Thus, knowing the TF group to which a soil belongs and the reference TF (for cereals) for the group, assessments can be made of the TFs to other crops on this soil to within an order of magnitude. The available data do not justify a similar calculation for Sr.

(3) There are two promising alternative approaches to classifying soils with regard to radionuclide behaviour. One uses correlation and factor analysis to identify the most important soil properties controlling TFs which can then be used predictively. The other is a semi-empirical procedure based on the assumption that ion exchange capacity, pH and organic matter content control ion availability in the soil. A dimensionless factor derived from normalized values of these parameters shows good correlations with TF for the data sets analysed so far.

(4) The variability data of the data arises from many sources many of which can only be described qualitatively. The lack of standard validated procedures for field and pot experiments is a handicap.

10. RECOMMENDATIONS

(1) Steps should be taken to obtain sufficient data to allow soils to be classified on the basis of Sr TFs. Further data should also be obtained to improve the definition of the classification based on Cs TFs.

(2) The alternative approaches mentioned in Conclusion 3 should be applied to as wide a range of data as possible.

(3) All relevant data from this CRP and from other sources should be collected in a publicly available electronic database.

(4) Sources of variability in field and pot experiments should be explored and quantified with a view to recommending standard procedures that can be validated by analogy with, for example, chemical analytical methods.

(5) Efforts should be made to ensure that the provisions of ISO 17025 can be applied effectively in international programmes.

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TRANSFER OF RADIOACTIVE CAESIUM, STRONTIUM AND ZINC FROM SOIL TO SORGHUM AND MUNG BEANS UNDER FIELD CONDITIONS IN TROPICAL NORTHERN AUSTRALIA

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Abstract

Soil-to-plant radionuclide transfer factors for caesium (^{134}Cs), strontium (^{85}Sr) and zinc (^{65}Zn) into sorghum and mung plants grown in tropical Australia have been determined over a four-year study period. The crops were grown on two types of red earth soils. Transfer factors for Cs and Sr are not substantially different from the expected values based on previous studies, reported in the general literature and compiled in the IUR database, mainly performed within temperate climates. In contrast, the values for Zn are more than an order of magnitude greater than anticipated. Most of the radioactivity added to the soils has been retained in the top 5 cm of both soils. There has been a general decline in soil-to-plant transfer of Cs and Zn as time has increased.

1. INTRODUCTION

This is a report on a field study of the transfer of radionuclides from soils to crops in tropical Australia. Its purpose is to provide information to an IAEA/FAO/IUR coordinated research program (CRP), as well as to interested individuals and groups in Australia, and to provide project reviewers with an overview of the study.

The CRP followed on from previous international investigations that had identified: a) a substantial lack of data on radioecological parameters outside the more closely studied regions of the planet (data currently exist predominantly for cool temperate northern hemisphere environments); and b) that soil type seemed to be the predominant factor influencing plant bioaccumulation. The initial studies were undertaken because of the recognition that nuclear power was likely to become more widespread as an energy source in tropical regions as a result of economic and social development concomitant with a need to reduce greenhouse gas emissions. Details of those studies and other general background information on this study can be found in [1].

This report covers: introduction; study location, design and rationale; farming practice and application; summaries of the results of physical, chemical and biological analyses and gamma spectrometry performed on plants and soil samples; calculation of transfer factors; comparison with data from the literature; and a brief discussion of the results and their implications. A detailed evaluation of the implications of the results has not been performed in this report but will be the subject of upcoming manuscripts in the general scientific literature.

Prepared within the Australian Nuclear Science & Technology Organisation
- International Policy Interaction Project – Dr J.F. Easey, Project Leader
- Environment Division – Professor A. Henderson-Sellers, Director

2. MATERIALS AND METHODS

2.1. Study design and siting

Within the IAEA/FAO CRP it was agreed that variables between individual national studies should be reduced to enhance the likelihood of determining which factors relating to soil type were most influential in affecting soil to plant transfer of radioactivity. A standard protocol to achieve this was prepared [2] and was followed as far as was practical in this study, given local constraints. The protocol covers: selection of crops and radionuclides; modes of application of radioactivity and experimental conditions; measurements and data reporting; and quality assurance procedures.

Leafy vegetables and grains generally tend to have the highest and lowest transfer factors, respectively. Hence, these two crop types were selected by the CRP as being the best to use across all studies. The choice of species was left to the individual researcher based on normal agricultural practice in their region. At our trial site, leafy vegetables could not be grown because of their high maintenance requirements. The Australian site is within a grazing/broad acre cropping area. Hence, high-intensity horticulture, required for leafy vegetables, is not normally practised. The grain crop chosen was sorghum, *Sorghum bicolor* (L.) Moench. The second crop selected was mung, *Vigna radiata* (L.) Wilczek, which is often used in rotation in the region as a nitrogen fixer. It was believed that the broad leaves of this low-growing crop might approximate the result for leafy vegetables.

The location of the farm used for the Australian contribution is shown in Fig. 1 together with rainfall data for the selected study sites. The Douglas Daly Research Farm is (approx.) 250 km south of Darwin. It lies within the tropics and has a continental monsoonal climate. The wet season usually extends from December to March with little rain falling at other times of the year. The growing season occurs over the period of greatest rainfall.

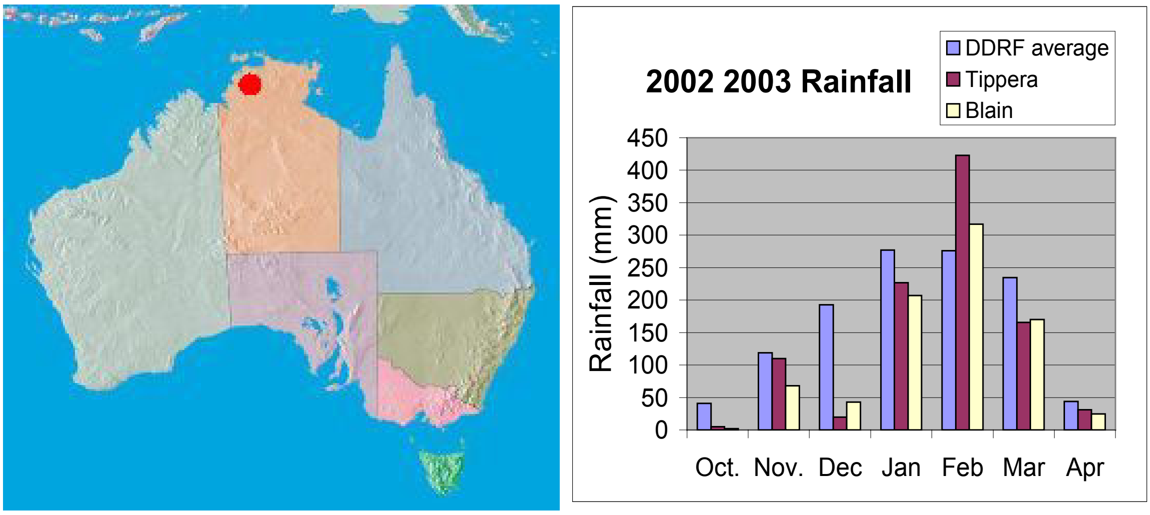

FIG. 1. Location of the study site at the Douglas Daly Research Farm and rainfall data for the study sites.

Two soil types were available at the study site, a sandy loam (Blain) and a clay loam (Tippera). These soils are both classified as Red Earths and are also described as lateritic in some soil taxonomies (e.g. [3; 4]). Classification details for these specific soils are described in [5]. Under the Soils World Reference Base (WRB) [6], both soils key out to Arenic Acrisols although this is not ideal. "*There are some problems with the WRB in that a number of major groupings are similar to the Kandosols / Red Earths developed on limestone in the Daly Basin. Most of the soils in the WRB have more strongly developed structure in the sub-soil than soils in northern Australia - Ferralsols, Lixisols, Planosols and Nitisols are all somewhat related but include profiles with a higher degree of structural development. Leptosols also come close but overlie hard rock, again with more structure in the subsoil. Even allowing for a lack of practice in applying the key, soils in tropical Australia do not* *appear to be well catered for."* Paraphrased from D. Howe, Natural Sciences Division, NT DBIRD, (pers. comm. 2003)

The sites selected are remote from the community at the farm (>2 km) and in areas not prone to flooding. To increase safety and minimise the potential for site disturbance, the plots are located within 50 m x 50 m paddocks surrounded with (1.8 m) barbed wire fencing that has also been pig-proofed at its base to inhibit incursion by feral animals. The radioactive plots are further enclosed within a chain-link-fenced area (17 m x 6.5 m) that is also covered with bird mesh during the growing season to minimise crop loss prior to harvest. The treated areas within the chain-link fence comprise two 2.5 m x 6 m subplots, nominally A and B, one for each crop type.

2.2. Addition of Radioactivity, Sowing of Crops and Sampling

Radioactivity (^{134}Cs, ^{85}Sr and ^{65}Zn) was initially added to the soils in October 1999. Due to its short half-life (65 days) ^{85}Sr was reapplied in October each year using the same technique. The method used, a watering can with a spreader bar, was designed to simulate a rain event early in the wet season. To prevent some of the activity falling into, and concentrating within, soil cracks, water was applied to swell the soil at each site over a few days prior to addition of radioactivity. This procedure had the added benefit of ensuring that the material adhered to the soil surface and, as such, was less likely to be remobilised by wind action.

Soil cores were taken to a maximum depth of 20 cm using a 25 mm (October 1999) or 20 mm diameter sampler (April 2000 and subsequently). In the first year, at least six cores were taken within each subplot and in adjacent, unlabelled areas for each soil. The cores were immediately separated into 0–5 cm, 5–10 cm and >10 cm sections. These sections were returned to ANSTO, dried and homogenised before analysis. In years 2–4, each subplot (A and B) on each soil was sub-divided into north, middle and south sectors to provide replication. In these cases, at least three cores were taken from each sector, separated into depths and then processed as in the first year.

The plots were seeded after the onset of rains, typically in December but occasionally in January. A zero-till planter was used to minimise soil disturbance. The crops were rotated between subplots in each successive season as is typical of agricultural practice for the region (i.e. sorghum was planted in subplot A on both soils in 1999 and 2001 and in subplot B on both soils in 2000 and 2002. Mung followed the opposite pattern). Crops were harvested by hand in late March or April. Mung leaves were also collected. In all cases, the crops were sampled from north, middle and south sectors of each subplot, on each soil, to provide replication. Plant populations and crop yields ranged from typical to good across all harvests (Specific details are available from the lead author). Care was taken to avoid contaminating the plant samples with soil particles. Plants growing within 0.25 m of the edge of the labelled area were not harvested so as to minimise the effect on radionuclide uptake of plants acquiring nutrients from outside the plots. Supplemental watering was provided on occasions whilst pre-emergent herbicides and post-emergent insecticides were also applied, in accord with local agricultural practice, to ensure that crop samples were obtained from the sites. Details of these applications can be obtained from the lead author.

2.3. Measurements

All samples returned to ANSTO were dried and homogenised before sub-sampling for the various analyses. Aliquots of each sample were placed into Marinelli beakers for gamma spectrometry. The spectrometer efficiencies were determined using standard additions to water/gelatine mix (for plant samples) or to an unlabelled soil. All radioactivities were corrected for decay to a standard reference date of 1 October 1999 or to the date of addition in the case of ^{85}Sr.

Particle size distributions for each depth in each soil were performed by sedimentation and using a hydrometer (e.g. [3]). Major clay mineralogy was evaluated using X-ray diffraction on the sub-2 μm fraction. Concentrations of a range of stable elements in all soils and crops were measured using inductively coupled plasma atomic emission spectrometry (Varian Vista-Pro ICP-AES; USEPA method 200.7, [7]) or inductively couple plasma mass spectrometry (Agilent 4500 ICP-MS; USEPA method 200.8, [7]). Total concentrations were evaluated following microwave digestion of 0.2 g of sample in 10 mL concentrated HNO_3 within a TFM pressure vessel (ANSTO modified USEPA method 3051, [7]). Estimates of the exchangeable fraction of the same elements in soils were determined after extraction with 1 M NH_4Cl [8]. Quality assurance was achieved using reagent blanks, duplicate analyses and analysis of standard materials. All elemental analyses included certified secondary standards with reference to NIST primary standards. Errors were typically 10% or less. Any subsets of data exceeding 10% error, or with evident blank contamination, were excluded from the results.

Other measured soil parameters included soil moisture content, cation exchange capacity, pH (at a 1:5 ratio in water and in $CaCl_2$) [8], and organic matter content using the Walkley-Black method [9].

Special techniques were applied to measure rhenium (Re) in sub-samples from those collected in 2000 and 2001. This was undertaken on the assumption that Re can be used as a natural analogue of Tc that has been in the environment for the long-term. There is some conjecture on this and it will be the subject of an upcoming paper discussing the results observed in the current study. Rhenium in soils was determined as a total concentration and also as a water-soluble concentration. Details of the analysis method for Re in 60–70 g soil samples, involving alkaline fusion, are given in [10]. The water-soluble fraction was determined by shaking 50 g of soil in 250 mL deionised water over an 8 hr period. The resultant soil solution was then obtained using a glass fibre filter (GF/A) followed by a 0.45 μm membrane filter. Re in plants was determined as a total concentration using 20–25 g samples. The analytical details are given in [11].

Soil sub-samples were also taken to estimate the radionuclide binding capacity (K_d). Details of the methodology applied for K_d determinations are given in [12]. Additional sub-samples were used for estimation of redox state by X-ray Analysis of the Near Edge Spectrum (XANES) using the X-ray fluorescence microprobe on beamline B20 at the Australian National Beamline Facility at the Photon Factory, Tsukuba, Japan. For methodology see [13]. Soil microbial populations (fungal and bacterial) were estimated in October 2000 and March 2001 to assess changes over a growing season. Details are provided in [14].

3. SAFETY ASPECTS

Prior to initiation of the fieldwork, all aspects of the study were evaluated for safety by a number of assessments. These involved ANSTO Safety Assessment Committee and representatives of the Northern Territory DPIF (now NT Department of Business, Industry and Resource Development, being responsible for the research farm). In addition, the Northern Territory Health Services were kept informed of any developments. Finally, the study also required an operational licence from the Australian Radiation Protection and Nuclear Safety Agency to proceed.

All operational procedures were applied successfully and there has been no action or occurrence that has led to unexpected radiological exposures or other adverse health effects. The radioactivities in the plots are at levels such that the plots can be used for any purpose. Nonetheless, an ongoing regime of annual sampling and TLD monitoring will be continued until such time as the radioisotopes added to the sites are no longer detectable.

4. RESULTS AND DISCUSSION

The acceptable results of the analyses and measurements carried out are summarised in the following text, tables and figures. Collated data are included in the combined CRP data sheets in the summary. More detailed data are available from the lead author.

4.1. Soil Properties

The values for particle size distributions, charge concentrations of exchangeable cations, cation exchange capacity (CEC) equilibrium pH and organic carbon content for each soil type at each depth are given in Table 1. The particle size distribution (also shown in Fig. 2) confirms the greater clay content of the Tippera soil compared to the Blain. The grading characteristics observed in this study are consistent with the studies of regional soils by [5] and thus the samples tested in this study can be expected to be typical of the soils in the field. The fractionation results are consistent with their classification as clay loam and sandy loam respectively. This difference is also reflected by the higher CEC of the Tippera soil (Table 1). The results of XRD analyses on the clay fractions are shown in Fig. 3. Clay fractions were found to contain predominantly quartz in Blain, whilst the clay fractions of Tippera soils contain 60% kaolinite and 40% muscovite. The CEC and organic matter content are slightly higher in the near surface soils relative to the deeper samples.

TABLE 1. SOIL CHEMICAL PARAMETERS FOR BLAIN AND TIPPERA SOILS FROM DALY RESEARCH STATION

Soil		Blain			Tippera		
Depth (cm)		0 to 5	5 to 10	10 to 15	0 to 5	5 to 10	10 to 15
pH of 1:5 soil / water		6.25	6.05	6.26	5.73	5.05	5.17
pH of 1:5 soil / 0.01 M $CaCl_2$		5.54	5.42	5.67	5.06	4.36	4.48
Size distribution	Sand / gravel (%)	84.5	83.5	80.4	43.1	38.1	38.4
	Silt (%)	2.9	3.6	4.2	30.1	30.0	30.4
	Clay (%)	12.6	12.9	15.5	26.8	31.9	31.3
Exchangeable cations ($cmol_c$/kg) (averaged over 2000-2003)	Ca^{2+}	1.97	1.34	1.16	2.52	1.70	1.91
	K^+	0.31	0.29	0.36	0.60	0.53	0.53
	Mg^{2+}	0.54	0.31	0.21	0.76	0.50	0.55
	Na^+	0.56	0.55	0.63	0.55	0.61	0.75
Sum of exchangeable cations (effective CEC)		3.38	2.49	2.36	4.43	3.33	3.74
Organic carbon content (%)		0.14	0.02	0.00	1.34	0.54	0.42

Note that the concentration values for cations have been charge compensated.

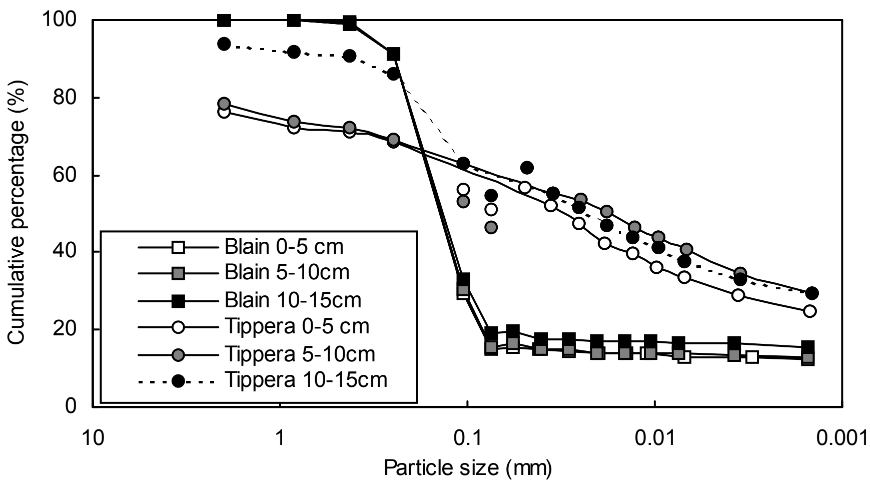

FIG. 2. Particle size analysis results. (In the Tippera soil, the kink observed between 0.048 mm and 0.075 mm is due mainly to difficulty in analysing the weathered soil samples, which disintegrated during the tests.).

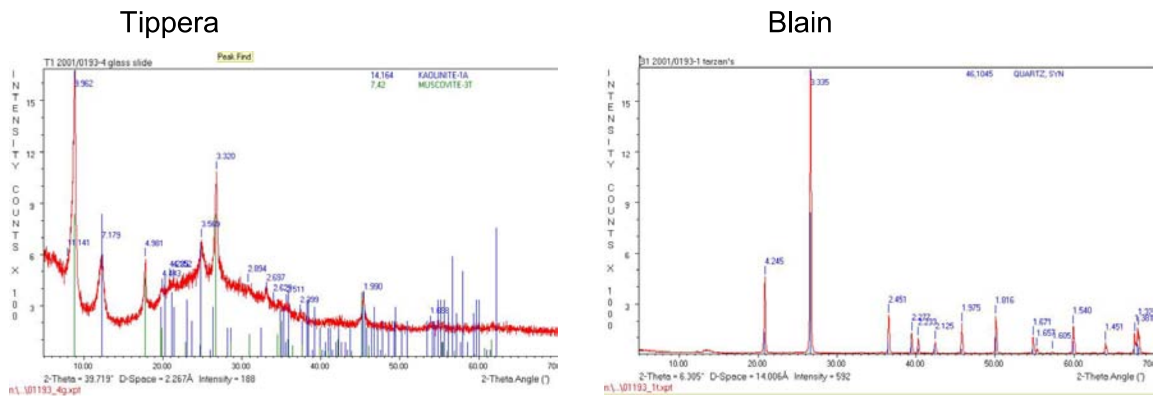

FIG. 3. Graphs of XRD results for clay fraction mineralogy in each soil.

The pH values in water (Table 1) were found to be slightly acidic and a comparable result was observed in $CaCl_2$. The pH values are similar (if slightly lower) to other results for these soils. Lucas et al. [5] reported 5.9 to 6.1 for Tippera, and Eastik (NT DBIRD, pers. comm., 2001) determined 6.6 to 6.8 for Blain.

4.2. XANES

Figure 4 shows the XANES spectra for the reference standards prepared for the study whilst Fig. 5 shows the XANES spectra for the various soil samples. Comparisons of the spectra indicate that the proportion of Mn in different oxidation states is similar in both soils within the same season (Fig. 5). Thus in October 1999, the Tippera sample contained Mn(II) 26%, Mn(III) 8% and Mn(IV) 66% and the Blain sample contained 27%, 8% and 65% respectively. In April 2000, the Tippera sample contained Mn(II) 16%, Mn(III) 8% and Mn(IV) 76% and Blain contained 19%, 10% and 71% respectively. However, the proportion of oxidised species [Mn(IV)] had increased by the end of the wet season (April) which also marks harvest time (i.e. Mn(IV) from 66 % to 76 % in Tippera sample and Mn(IV) from 65 % to 71 % in Blain sample).

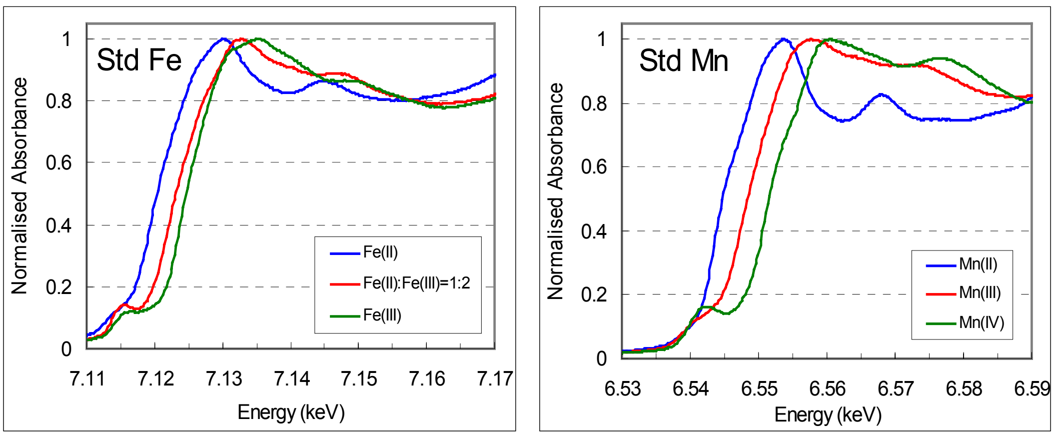

FIG. 4. XANES spectra for reference Fe and Mn materials. Note the shift in the energy to higher levels with increased redox state.

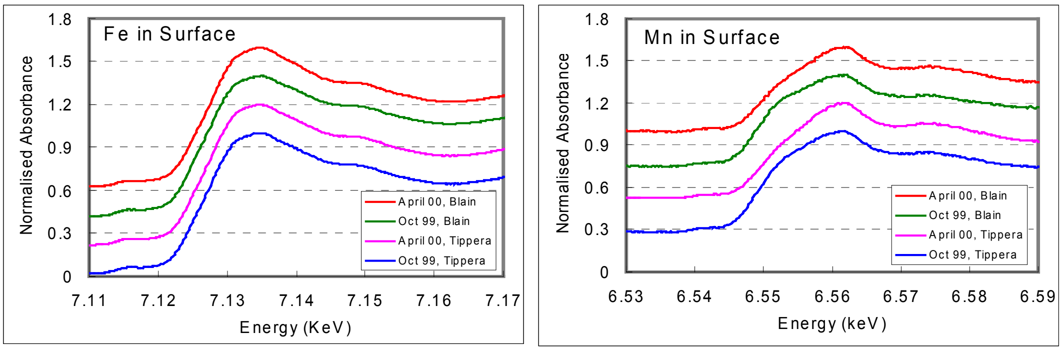

FIG. 5. XANES spectra for soils sampled from the DDRF before and after cropping in the 2000-2001 wet season. The spectra have been adjusted to different base lines for ease of comparison

Fe species are very similar for both soils and sampling periods [Fe(II) ~ 10 %] (Fig. 5). The lack of any apparent redox shift in Fe to accommodate the Mn shift noted above was not unexpected given the >10 times higher concentration of iron in the soils compared with manganese (XRD % w/w: Mn 0.13-0.29; Fe 3.6-7.9).

Several elements show some degree of redox sensitivity. Those of most interest from a radiological perspective are Tc and Pu. Given that some change was observed in redox potential (albeit small) between the beginning and end of the growing season, it would be appropriate to consider the consequential possibility of seasonal changes in plant availability of these elements in future radiological dose assessment studies. These results also have relevance to the use of results from pot studies for soil-to-plant transfer compared with those obtained from field studies. Pots will be much more likely to remain well aerated and hence be less prone to redox changes than field experiments. Lysimeters, which typically contain a large soil mass, are more likely to compare favourably with field studies in relation to redox state.

4.3. Microbiology

The viable cell count provides a guide to microbial activity in these soils although it can underestimate total microbial biomass by up to 90%. Results of microbial counts are given in Table 2. These show no appreciable change in bacterial populations over the growing period.

Despite this finding, the mix of bacterial functional groups may have altered in dominance over that time. In contrast, fungal populations increased generally in both soils over the growing period (Fig. 6).

TABLE 2. VIABLE BACTERIAL AND FUNGAL COLONY COUNTS ON NUTRIENT AGAR (NA) AND MALT EXTRACT AGAR (MEA)

			Colonies/g soil	
	Soil	Crop	Bacteria	Fungi
October 2000	Tippera	nil	8.0E + 06	6.2E + 05
	Blain	nil	2.0E + 07	2.9E + 05
April 2001	Tippera	mung	4.2E + 06	3.1E + 06
	Tippera	sorghum	1.9E+ 06	1.2E + 06
	Blain	mung	3.8E+ 06	2.1E + 06
	Blain	sorghum	4.0E+ 06	2.3E + 06

N.B. initial April samples were unreliable and these results were after storage at 4°C for 3 weeks. No loss of viability was observed.

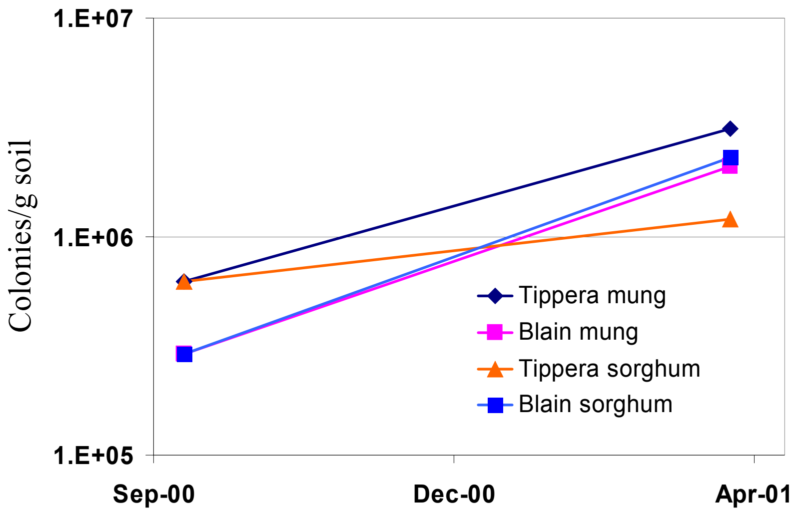

FIG. 6. Increased fungal populations in soils of the DDRF over the period of growth in the 2000-2001 wet season.

Fungi can influence soil conditions, such as pH, within the microenvironment surrounding their hyphae (Fig. 7). Several elements are more soluble at lower pH values, including Zn, P and Sr (see section on K_ds below). Radioisotopes of these elements are hence more likely to be accumulated by plants with higher soil fungi populations. The vesicular arbuscular mycorrhiza (VAM) and some other fungi associated with plant roots also facilitate nutrient uptake by greatly enhancing the effective surface area of the root system. It may be that under such circumstances radionuclides with chemical similarities to macro- and micronutrients will also be assimilated in greater quantity. Examples of this potential effect include radioactive Sr and Ra for the macronutrient Ca, Cs for K, and Tc for the micronutrient Mn. A potential corollary is that increased plant performance related to fungal symbiosis may lead to greater above ground biomass and hence lower plant concentrations or radionuclides (by biomass dilution) despite greater overall uptake of elements.

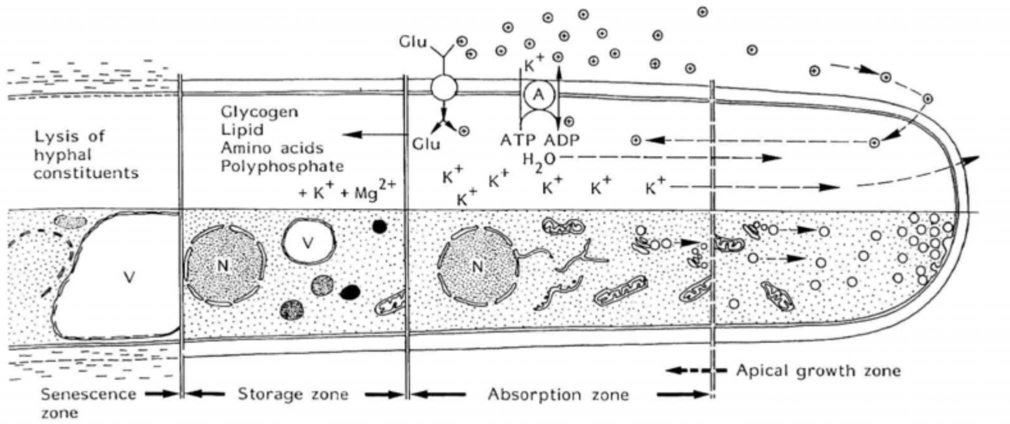

FIG. 7. Diagram showing proton pumping from a fungal mycelium into the local soil micro-environment (taken from [15]).

The two factors (redox state and microbial activity) may also be correlated. Increased fungal activity will give rise to higher respiration rates and hence, potentially, a more reducing environment. However, this does not agree with the observed trend of increased oxidation state for Mn in these soils that occurred over the same period as the fungal populations were observed to increase. It should be noted that this study has only been evaluated over a one-season period and more data are required to confirm these observations as being consistent seasonal events, or as being correlated with other seasonal factors such as rainfall and soil moisture. Nonetheless, the results obtained are not inconsistent with expectations and, hence, point to the potential for such factors to be involved in the apparent greater variability in tropical soil-to-plant transfer of radioactivity observed previously [2].

4.4. Distribution coefficients (K_d values)

The results of batch K_d experiments for ^{134}Cs are shown in Fig. 8a. The retention of Cs is very strong, indicated by K_d values of approximately 10^3 mL/g. The adsorption of ^{134}Cs onto Tippera soils (K_d of 2300–4100 mL/g) is stronger than onto Blain soils (800–1200 mL/g). As Cs is strongly adsorbed by fine particles this is attributed to the clay content, particularly the muscovite in the Tippera soil [16]. The equilibrium pH values for Blain soils are higher than the Tippera soils. However, the experiments in which the pH was varied by ± 1 unit indicate that there is no significant pH dependence of Cs sorption in either soil.

The adsorption of ^{85}Sr is much weaker than for ^{134}Cs, with K_d values of approximately 30 - 60 mL/g being measured at the equilibrium pH of the samples (Fig. 8b). There is a pH dependence of Sr adsorption, with K_d increasing with pH, ranging from 14 to 86 mL/g over the studied pH range. For a given pH value, the K_d for Sr on the Blain is higher than the Tippera samples, which can be traced to their clay mineralogy. However, the natural pH of the Blain samples is higher than the Tippera, and for this reason the measured K_d values at equilibrium pH are similar (Table 3).

The K_d data for Zn show the strongest pH dependence of the studied radionuclides, increasing by more than an order of magnitude (from about 40 mL/g to 3000 mL/g) between pH of 4.4 and 7.3 (Fig. 8c). Because of this strong pH dependence, the K_d values for Blain soils at their equilibrium pH (480 - 1630 mL/g) are higher than Tippera soils (160 - 440 mL/g). The adsorption of Zn on the Blain soils appears to slightly increase with increasing depth.

As noted above, K_d measurements provide a basis for the initial comparison of the retardation of radionuclides in the environment, under various chemical conditions and soil types. Table 3 gives a summary comparison of the radionuclide sorption K_d values obtained at equilibrium pH values in the present study with geometric mean values for different soil types reported by [17]. In general, the values are consistent with those reported for the generic soil types as categorised by [17].

TABLE 3. COMPARISON OF AVERAGE DISTRIBUTION COEFFICIENTS (STANDARD ERROR, N = 3), MEASURED AT EQUILIBRIUM pH VALUES, WITH GEOMETRIC MEAN VALUES FROM [17]. VALUES ARE K_D (mL/g)

Soil	Cs	Sr	Zn
Blain	1100 (35)	46 (6)	990 (330)
Tippera	3400 (520)	44 (6)	300 (80)
Sand	280	15	200
Loam	4600	20	1300
Clay	1900	110	2400

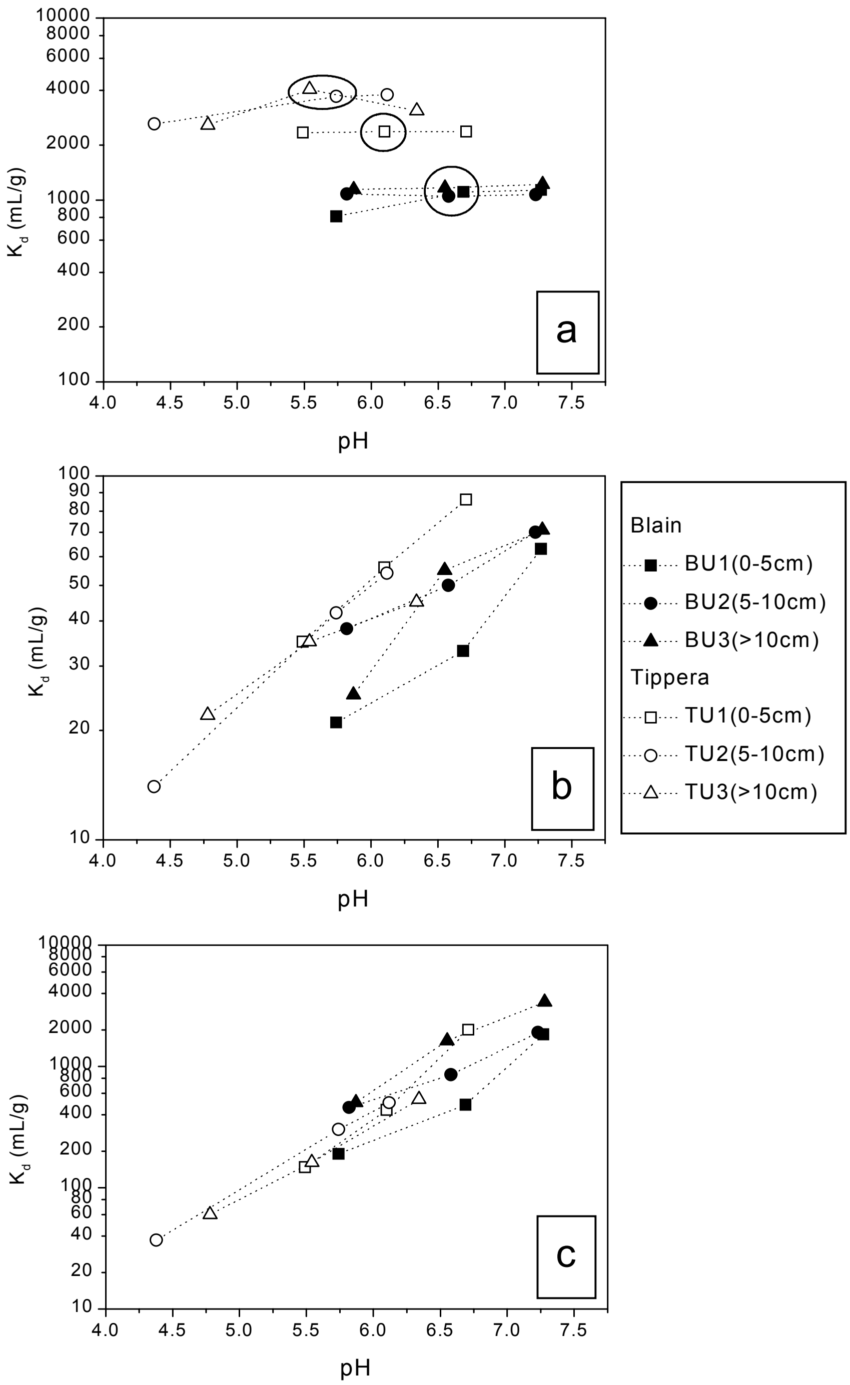

FIG. 8. Distribution coefficient (K_d) values for adsorption of radionuclides (a-^{134}Cs, b-^{85}Sr, c-^{65}Zn) on Blain and Tippera soils. The central data points in each data-set (circled in Fig. 2a) are for experiments carried out at the equilibrium pH of the samples. The other two data points for each soil were obtained at pH values approximately one unit higher or lower than this value.

4.5. Soil Chemistry–elements

Details of semi-quantitative XRF analyses on the two soils were given earlier [1]. The results showed that oxides of Si, Al and Fe predominate, comprising approximately 90% of the total mineralogy. Of these, silicates were the major component and were higher in Blain than in Tippera. These results are consistent with the XRD and particle size analyses reported earlier. Aluminium oxides were the next most abundant but in this case they were more abundant in the Tippera soil. Iron was approximately equivalent in both soils.

As has been observed by previous workers, it is important in studies of radionuclide-soil interactions to take into account the existing elemental content of the soil, as this is usually far greater (in concentration terms) than the added trace radionuclide [16];[18]. Results of ICP analyses on total digests are reported in [1] for year one. Detailed data for later years are available from the lead author. The best results were obtained for the first season samples as limit of detection problems hampered interpretation in later years. They confirm the XRF results and are consistent with general expectations. Most trace elements are 2 to 6 times more abundant in the Tippera soil. The extracted elements are also more abundant from the Tippera soil but the proportions of extractable elements to total concentrations are generally higher for the Blain soil. In the first season, the typical % extract of trace metals was ~10% for Blain and ~ 5% for Tippera with Zn being a notable exception (see Table 4). The high extractable Zn concentrations are complemented by relatively high plant uptake of ^{65}Zn from both soils (refer to later results from gamma spectrometry).

Values for Cs, Sr and Zn, the stable element analogues for the radioisotopes added into the system, are reported for each depth on both soils in Table 4. The data for Sr are similar in the two soils and show little dependence with depth. The total amounts of both Cs and Zn are higher in the Tippera samples. However, the extractable proportion of these metals is higher for the sandier Blain soil. The retention of these elements in the Tippera soil is much stronger than the Blain, as is indicated by the lower proportion extracted with NH_4Cl (except in the surface layers). The stronger binding near the surface is possibly related to the higher organic fraction in this layer.

Table 4 also compares the measured concentrations in this study with medians and ranges for those elements based on analyses of soils throughout the world [19]. All three trace elements in Tippera and Blain are towards the lower end of the global distributions, particularly for Sr and Zn, which are both more than an order of magnitude less than the median values. Hence, Tippera and Blain can both be considered to be deficient in these elements.

TABLE 4. TOTAL AND EXTRACTABLE (1M NH_4Cl) ELEMENTAL CONTENT OF UNLABELLED SOILS FOR Cs, Sr AND Zn COMPARED WITH MEDIAN AND RANGE OF MEASURED VALUES OF GLOBAL SOILS AS REPORTED IN [19]

		Caesium		Strontium		Zinc	
Soil	Depth (cm)	Total (µg/g)	Extracted	Total (µg/g)	Extracted	Total (µg/g)	Extracted
Blain	0–5	1.2	<8%	5.5	8%	7.7	22%
	5–10	1.1	80%	5.7	10%	4.6	89%
	10–15	1.4	82%	6.3	8%	4.3	46%
Tippera	0–5	3.9	<3%	10.3	8%	11.5	14%
	5–10	4.4	4%	5.0	10%	10.3	19%
	10–15	6.0	10%	9.7	6%	9.0	33%
World median		4	—	250	—	90	—
Range		0.3-20	—	4–2,000	—	1-900	—

Concentrations in soils of the Tc analogue, Re, were reported in detail elsewhere [20]. Average total concentrations in Tippera and Blain soils were 21.1 ± 2.0 pg/g and 4.9 ± 0.9 pg/g respectively, for the samples collected in 2000. The percentages of water-soluble Re in these soil samples depended on soil type, e.g., 3.6 ± 2.5% for Tippera and 12 ± 8% for Blain. The dynamic equilibrium between water-soluble and non water-soluble Re has been reached over geological time periods (as distinct from the radioactivity added freshly to the soils at the start of this study). Hence, the relatively low water-soluble fractions in these soils are not unexpected, particularly as the Re originates from the mineral matrix. Because the water-soluble fraction plays an important role in root uptake by plants, Re transfer from the soil to plants would be affected by this condition.

The Re concentrations in dry plant tissues are low, sometimes below detection limits (0.05 pg/g). Detectable levels in these samples range from 0.07 – 0.6 pg/g in sorghum and 0.06 – 0.6 pg/g in mung bean. No substantial differences have been observed between crops grown on the two soils, in either 2000 or 2001.

The average TF estimates for total and water-soluble Re from Tippera and Blain soils to sorghum grains and mung beans are shown in Table 5.

TABLE 5. TFs FOR Re FROM TIPPERA AND BLAIN SOILS TO SORGHUM GRAIN AND MUNG BEANS OVER TWO YEARS

	2000				2001			
	Total	±	water soluble	±	Total	±	water soluble	±
Tipp. sorghum	0.004	0.001	0.117	0.062	0.015	0.008	0.663	0.375
Tipp. mung	<0.003		<0.070		0.013	0.010	0.272	0.131
Blain sorghum	0.026	0.025	0.199	0.160	0.045	0.017	0.884	0.467
Blain mung	0.030	0.009	0.257	0.120	0.053	0.022	1.021	0.464

The values derive from duplicate or triplicate analyses on individual soil samples. Errors are ± 1 s.d. of the individual values above detection limit

3.6. Soil Radioactivity

After correcting for radioactive decay, there was very little depletion of the added radioactivity from the sites over the 4.5 year experimental period. This is shown in Fig. 9 which gives the average, decay corrected, activity for each nuclide in the top 20 cm of the soil profile within each subplot.

Some distributional heterogeneity of the surface-applied radioactivity obviously exists within each plot because the average value for each radionuclide varies by up to a factor of four or more over the entire period (Fig. 9). This is despite the fact that each value is representative of at least six to nine cores. Nonetheless, there is no significant overall change in activity over the experimental period in any plot. In addition, the variation between sampling periods for Cs and Zn is remarkably consistent within each plot. These isotopes were added once only in a mixed solution. This observation suggests that there has been a high degree of conservation of the radioactivity due to low radionuclide mobility together with minimal mixing and low overall site disturbance.

The low mobility of radioactivity is further demonstrated in Fig. 10 that shows the vertical profiles of ^{134}Cs at the end of the experimental period. The profiles are based on individual cores that were separated into 5 mm to 10 mm sections (Given the time since addition, ^{85}Zn was too low in activity to be accurately measured at this depth precision). These graphs show that most of the activity is still located within the top few cm of soil after more than four years of cropping under normal conditions. Some minor levels of radioactivity were detected in the surface soils adjacent to the labelled areas. This implied some horizontal movement of the activity, possibly associated with sheet flow during the rainy season or wind disturbance. However, the activity concentrations detected were relatively insubstantial and provide further support to the overall retention of radioactivity within the labelled plots, evidenced above.

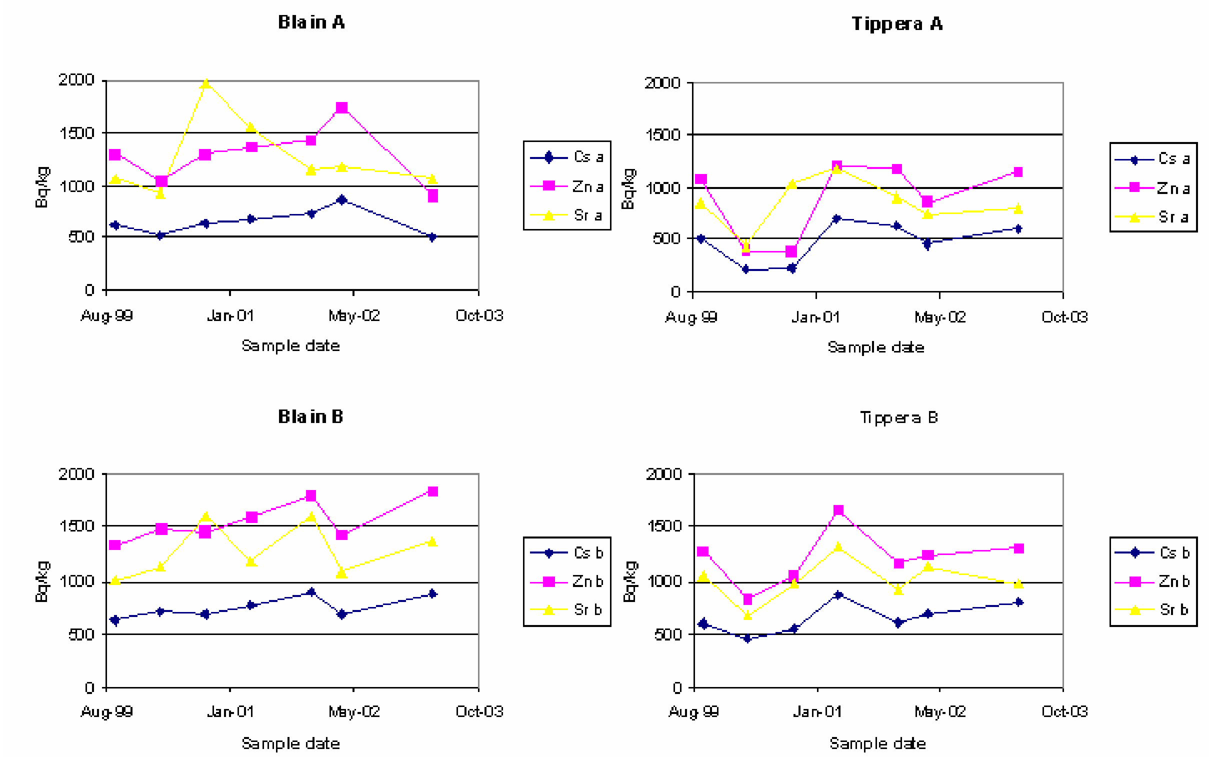

FIG. 9. Decay corrected average activity within the top 20 cm of each subplot on each soil. The ^{134}Cs and ^{65}Zn were added once only in October 1999. The ^{85}Sr was added annually in October due to its short half-life.

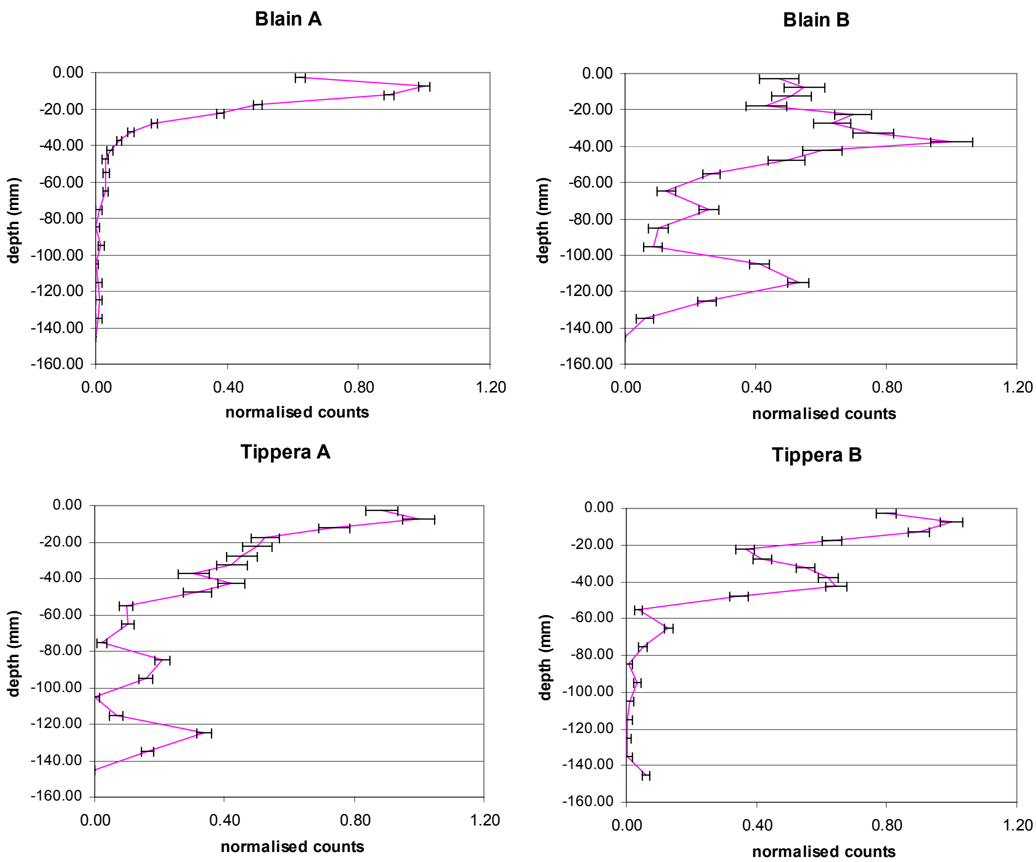

FIG. 10. Depth profiles of ^{134}Cs at the end of the experiment. Counts are corrected for sample volume and normalised to the maximum count rate in the profile. Errors were propagated from the count rate using Gammavision.

4.7. Transfer factors

The calculated dry weight radioactivity values in the soil and plant samples were used to derive transfer factors from each of the soils into each crop. The definition of a transfer factor is given by the following equation.

$$Transfer\ Factor\ (TF) = \frac{Radioactivity\ in\ crop\ (Bq/kg\ DW)}{Radioactivity\ in\ soil\ (Bq/kg\ DW)}$$

Radioactivity in soil is defined here as the average activity concentration in the top 20 cm. This is an accepted international compromise arising from alternate measures that are often based on deposition per unit area assuming atmospheric fallout. Transfer factor values in excess of one imply active bioaccumulation of activity. Values less than one imply either strong binding of the radioactivity to the soil or that the plant is not accumulating that material.

The transfer factors derived using the first and final year data, together with the IAEA recommended values for those radionuclides from sand or clay to grains, peas and leafy vegetables, are given in Table 6. The patterns of decreasing TF in each successive harvest are shown in Fig. 11. Strontium does not have the same pattern as the other radioisotopes because fresh ^{85}Sr was added to the soils each year.

TABLE 6. AVERAGE TRANSFER FACTORS FOR ARTIFICIAL RADIONUCLIDES BETWEEN CROPS AND SOIL (STANDARD DEVIATIONS IN BRACKETS, n = 3) IN SAMPLES FROM 2000 AND 2003

Soil		Blain			Tippera	
Crop	^{85}Sr	^{134}Cs	^{65}Zn	^{85}Sr	^{134}Cs	^{65}Zn
Year 1						
Sorghum	0.4 (0.1)	0.13 (0.04)	18 (3)	0.8 (0.1)	0.3 (0.05)	26 (2)
Mung bean	1.8 (0.3)	0.3 (0.1)	15 (2)	3 (0.3)	0.2 (0.01)	20 (1)
Mung leaves	37 (12)	0.4 (0.1)	15 (3)	63 (15)	0.5 (0.3)	22 (2)
Year 4						
Sorghum	0.1 (0.06)	0.03 (0.01)	3 (1)	0.15 (0.05)	0.01 (0.003)	4 (1)
Mung bean	1.2 (0.2)	0.06 (0.01)	10 (2)	4 (1)	0.04 (0.01)	7 (1)
Mung leaves	45 (12)	0.1 (0.05)	11 (3)	*24 (3)*	*0.08 (0.02)*	*6 (3)*

IAEA reference values (95% confidence intervals)

	Sand	Sand		Clay	Clay	
Grains	0.21 (0.03–1.4)	0.03 (0.003–0.3)	0.56 (0.2–1.7)	0.12 (0.2–0.6)	0.01 (0.001–0.1)	0.56 (0.2–1.7)
Peas	2.2 (0.5–9.4)	0.09 (0.01–0.7)	0.71 (0.2–2.1)	1.3 (0.3–4.9)	0.02 (0.002–0.1)	0.71 (0.2–2.1)
Leafy vegetable	3.3 (0.3–30)	0.46 (0.05–4.5)	3.3 (1.1–10)	2.7 (0.7–10)	0.18 (0.02–1.7)	3.3 (1.1–10)

N.B. IAEA reference values [21] are included for comparison. The Zn reference data were not specified for soil type, hence the same reference values have been ascribed to both soils. The last mung leaves from tippera were collected in 2002.

The calculated transfer factors show interesting results, particularly in comparison with the reference values that indicate the best estimate of the expected transfer factor based on the previous, predominantly temperate, transfer factor studies. It should be recalled that the two soils are both loams, albeit one sandier and one with more clay, and as such they fall between the classical definitions of clay and sand.

First, given the estimated uncertainties in these parameters, there may be some significant differences between plant uptake on the Blain and Tippera soils. The IAEA data indicate a higher accumulation in plants growing in sandy soil for each of the radionuclides. Despite this, any differences between the soils do not appear to be substantial for any of the radionuclides. This result is not unexpected given that both soils have a high clay component, with Blain being somewhat sandier.

Second, the IAEA data indicate higher transfer factors for all radionuclides into peas compared to grains. This was not seen as a consistent trend in the first year of sampling. However, by the final year that pattern had established itself on both soils.

Finally, the radionuclides of major concern, Cs and Sr, are generally of the same order as the recommended values in both crops, except for Cs in sorghum which was initially higher than expected but has subsequently aligned with the recommended values. On the other hand, the Zn values have remained more than an order of magnitude higher than the recommended values for the period of the study, despite declining as time progressed. The relatively consistent isotopic ratios in all plant and transfer factor results, which are different to the ratios of the solutions added to the soil or the current soil activity ratios, ensure that sample contamination was not contributing to these findings.

As noted earlier (Table 4), the soils can both be considered to be deficient in Zn. As the plants were able to grow normally and in healthy abundance, this result implies that the crops must have an efficient mechanism for accumulating Zn in these conditions. The implied high uptake efficiency is reflected in the high radionuclide TF values for Zn. This high efficiency may be linked to the observed increased abundance in fungal populations reported earlier (Table 2, Fig. 6) in that VAM fungi (in particular) are known to improve nutrient uptake efficiency in crops. The excessive transfer factors for Zn are also consistent with the observation in the chemical analyses that a high proportion of the stable zinc was readily extractable from both soils (Table 4).

5. CONCLUSIONS

The results of chemical analyses on the soils show that they are both heavily leached, with low cationic exchange capacity and organic matter content. The pHs are slightly acid but not unusually so. Both soils key out as Arenic Acrisols under the World Reference Base. XRF analyses confirmed that both soils are predominantly composed of oxides of silica with a lesser extent of oxides of aluminium and iron. The Tippera soil had higher trace metal concentrations but both soils were generally low, being deficient in both Sr and Zn when compared with worldwide data. Zinc was unusually more highly extractable than other metals in both soils.

XANES analysis indicated that there was a slight shift in Mn redox state to more oxidised species over the wet (=growing) season. Similarly, microbial analyses identified an increase in fungal populations over the same period. These results have implications with respect to the availability of some trace minerals to plants. However, the results were obtained over one harvest cycle only and hence should be treated with caution.

Adsorption of the radionuclides to the soils in question showed that binding of Cs was very strong in both soils, more so for Tippera. There was no significant pH dependence. In contrast, Sr is much less strongly adsorbed and does have a pH dependency. The relative binding strength of the two elements is reflected in the lower TF values of ^{134}Cs compared to ^{85}Sr. At any specific pH Tippera tended to bind ^{85}Sr more strongly than did Blain, however, as there was a slight difference in pH between the two soils there was no apparent difference in ^{85}Sr binding capacity at their natural pH values. Zinc was the most pH dependent of the three isotopes. The K_d values increased over 3 orders of magnitude between pH 4 and pH 8. There was no apparent difference between the soils in the ability to adsorb Zn at any pH value.

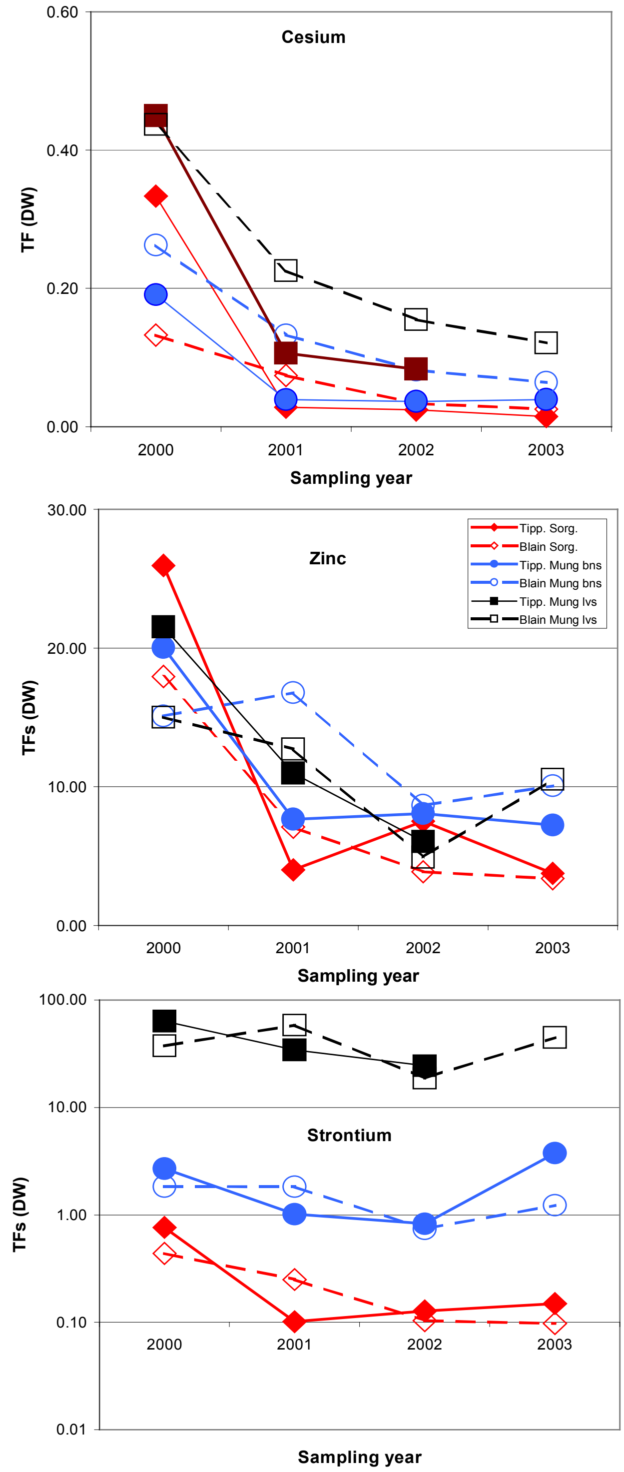

FIG. 11. Transfer factor values calculated for Cs, Zn and Sr accumulation into sorghum and mung from Blain and Tippera soils for the years 2000–2004. Both Cs and Zn were added once only in October 1999, Sr was reapplied each year.

The study highlighted very low mobility of the ^{134}Cs and ^{65}Zn in these soils. There has been no significant decline in the decay-corrected radioactivity in the top 20 cm of either soil over the duration of the study. Detailed sections of cores collected at the finish of the study confirm that most of the ^{134}Cs is still located within the top few cm of the soil profile on both Blain and Tippera. This finding also implies that the agricultural practices applied at the site are excellent at minimising soil loss from this system.

Transfer factors for Cs and Sr are not substantially different to the reference values provided by the IAEA. In contrast, the values for Zn are more than an order of magnitude greater than anticipated. This result is supported by the stable element analyses referred to above. There has been a general decline TF values with time throughout the study.

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PLANT UPTAKE PROCESSES RELATED WITH THE GEOCHEMICAL BEHAVIOUR OF RADIONUCLIDES IN SOME BRAZILIAN SOIL

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Abstract

Soil to plant transfer factors (TF) for ^{90}Sr, ^{137}Cs and ^{60}Co were measured for maize and cabbage grown in lysimeters containing a Ferralsol, a Nitisol and an Acrisol. These soils are representative of large agricultural areas in Brazil. In this study a sequential chemical extraction protocol was used to evaluate the potential mobility of radionuclides: 1) slightly acidic phase containing readily bioavailable elements; 2) readily reducible phase containing elements bound to Mn oxides; 3) oxidizable phase containing elements bound to labile organic matter; 4) alkaline phase containing mainly elements bound to Fe compounds; 5) resistant phase not potentially available to crops. This sequential procedure showed that the main soil factors influencing the ^{137}Cs transfer were exchangeable K, pH, organic matter and iron oxides content, while ^{90}Sr transfer was influenced by exchangeable Ca and soil pH and ^{60}Co transfer was influenced by manganese oxides content. The integration of the results obtained in the laboratory with the results obtained in field experiments seems to confirm that the ^{137}Cs TFs of Ferralsol, Acrisol and Nitisol are higher than those in the IUR (International Union of Radioecologists) databank whereas values for ^{60}Co and ^{90}Sr were similar or lower than those in the IUR databank for temperate climate soils.

1. INTRODUCTION

The study of radionuclides in agricultural areas is of general concern because, once in the soil, most of them can recycle within the biota, similar to nutrients. Interactions between radionuclides and soil depend on the chemical form of element and some soil properties such as pH, mineralogical composition, organic matter content and nutrient status [1–3]. Plant uptake of radionuclides will be dependent on these interactions and will also depend on the metabolic and physiological characteristics of the species. Knowledge of the mobility of radionuclides in soils is needed to assess potential hazards from radionuclide inputs into the food chains. One way to assess the potential mobility of radionuclides in soils is the use of sequential extraction procedures to describe the distribution of radionuclides in soil as a function of some physico-chemical conditions [4].

This work generated data on transfer factors of radionuclides (^{137}Cs, ^{60}Co and ^{90}Sr) from soil to reference plants in a range of soil systems (kaolinite and Fe-Al oxide rich soils, very acid soils with low nutrient contents) where transfer factors deviate substantially from the average established for temperate soils. The observed differences between soil-to-plant transfer factors obtained in some Brazilian soil systems are discussed in the light of pedology and geochemical partitioning procedures.

This work intends to provide parameters for environmental assessment models related with Brazilian soil conditions and to contribute to the identification of ecosystems that are more vulnerable to radioactive contamination, so that emergency response planning can be optimized.

2. MATERIALS AND METHODS

The transfer factor (TF) for reference plants (corn and cabbage) was determined following the IUR protocols [5]:

$$TF = A_p/A_s$$

Where, A_p = Activity in the edible part of the plant (Bq kg^{-1} dry weight) and A_s = Activity in the soil (Bq kg^{-1} dry weight).

Maize (*Zea mays*, L.) and cabbage (*Brassica oleracea,* L. var. capitata) were grown in masonry lysimeters (1 m^3 each) installed in a restricted area of the Institute for Radioprotection and Dosimetry (CNEN/Brazil).

Table 1 shows the main clay mineral type and ^{137}Cs, ^{60}Co and ^{90}Sr activities in the studied soils.

TABLE 1. SOIL TYPE (FAO CLASSIFICATION), CLAY MINERAL TYPE (IN ORDER OF PREDOMINANCE), ^{137}Cs, ^{60}Co AND ^{90}Sr ACTIVITY (Bq/kg DW± SD) OF STUDIED SOILS

Soil type	*Main clay mineral type*	*^{137}Cs in soil*	*^{60}Co in soil*	*^{90}Sr in soil*
Ferralsol 1	gibbsite, kaolinite	7.78E+03 ± 2.39E+02	*	4.74E+02 ± 2.02E+01
Ferralsol 2	gibbsite, kaolinite	7.47E+03 ± 2.33E+02	*	4.06E+02 ± 1.37E+01
Ferralsol 3	gibbsite, kaolinite	8.46E+03 ± 2.61E+02	*	
Ferralsol 4	gibbsite, kaolinite	6.10E+03 ± 1.89E+02	*	7.08E+02 ± 2.97E+01
Ferralsol 5	gibbsite, kaolinite	5.66E+03 ± 1.75E+02	*	1.10E+03 ± 4.55E+01
Nitisol	hematite, goethite vermiculite	6.77E+03 ± 2.10E+02	2.62E+03 ± 1.41E+02	9.88E+02 ± 4.18E+01
Ferralsol A	hematite, goethite	8.26E+03 ± 2.55E+02	2.80E+03 ± 8.49E+01	4.74E+02 ± 2.06E+01
Ferralsol B	hematite, goethite	6.06E+03 ± 1.90E+02	2.35E+03 ± 7.11E+01	3.79E+02 ± 1.79E+01
Goiânia A	gibbsite, kaolinite	1.36E+03 ± 4.40E+01	*	
Goiânia B	gibbsite, kaolinite	1.47E+03 ± 4.57E+01	*	*
Goiânia C	gibbsite, kaolinite	3.07E+03 ± 9.77E+01	*	*
Acrisol	kaolinite	3.34E+03 ± 4.21E+02	1.28E+03 ± 3.90E+01	3.04E+02 ± 1.40E+01

Soil treatments included: five lysimeters filled with an Al rich soil (Ferralsol 1 to 5), artificially contaminated with ^{137}Cs in 1992 and with ^{90}Sr in 2000; one lysimeter filled with an Fe rich soil (Nitisol), artificially contaminated for the experiment with ^{60}Co and ^{137}Cs in 1996; two lysimeters filled with another type of iron rich soil (Ferralsol A and B), artificially contaminated for the experiment with ^{60}Co, ^{90}Sr and ^{137}Cs in 2000; one lysimeter filled with the Acrisol soil (Kaolinite rich), artificially contaminated for the experiment with ^{60}Co, ^{90}Sr and ^{137}Cs in 2000; and three lysimeters filled with Goiânia soil, another Ferralsol (Al rich) was collected in the city of Goiânia and a site where a radiological accident with ^{137}Cs occurred in 1987 [6].

Soil was artificial contaminated with Cs and Co by spraying a treating solution to every two cm of soil, layer-by-layer, up to 40 cm. A layer of 40 cm of uncontaminated soil was beneath. Soil contamination of ^{90}Sr was by direct application onto the lysimeter soil surface.

Plants and soils were dried, ground and sieved through a 2 mm screen before direct measurement of the of ^{137}Cs and ^{60}Co activity by gamma ray spectrometry using a Ge detector. Efficiency was about 10% for the geometry used (pot of 250 g). Counting errors were less than 5%.

Sequential extractions were performed in soils samples as described in Table 2. Details of the analytical protocol can be found in [4]. All the extracts were analyzed by gamma ray spectrometry using a Ge detector, for the determination of ^{137}Cs and ^{60}Co.

TABLE 2. PROTOCOL OF SEQUENTIAL EXTRACTION CONSIDERING GEOCHEMICAL SIGNIFICATION BY PHASE

Phase	*Experimental conditions*	*Geochemical signification*
Slightly acidic	CH_3COOH + CH_3COONa 1:1; pH4.7; at room temperature (RT).	readily bioavailable
Readily reducible	$NH_2OH.HCl$ (0.1 M); pH2; RT.	mainly bound to Mn oxides
Oxidizable	H_2O_2 (30%) + CH_3COONH_4 (1M); pH2; RT.	bound to labile organic matter
Alkaline	NaOH (0.1 M) ; pH12; At room temperature	bound to Fe compounds
Resistant	Aqua regia + HF. Heat to 50°C/ 30 min	not mobilized in previous experimental condition

The Soils Office of the Brazilian Agricultural Research Corporation (EMBRAPA-Solos) performed soil analyses and clay mineralogy determinations according to their standards manual [7].

The amount of ^{90}Sr in soil and plants was determined by measuring the beta activity of ^{90}Y in a Berthold Low Background Proportional Counter (detector diameter 5 cm) [8].

3. RESULTS AND DISCUSSION

3.1. Chemical and physical properties of studied soils

The main chemical and physical properties of the studied soils are presented in Table 3. The Ferralsol (1 to 5) presents a sandy clay loam texture, low cation exchange capacity (CEC) soil. The contents of exchangeable K and Ca and the pH value of this soil are higher than normally observed in the field and is probably due to the application of fertilizers and lime in this experiment. The mineralogical analyses indicate the predominance of gibbsite and presence of kaolinite in the clay mineral fraction.

TABLE 3. PEDOLOGICAL ANALYSES OF ARTIFICIALLY CONTAMINATED SOILS AND GOIÂNIA SOIL

FAO classification	*Exch. K in soil (cmol kg^{-1})*	*Exch. Ca in soil (cmol kg^{-1})*	*CEC in soil (cmol kg^{-1})*	*pH in KCl*	*OM (%)*	*Clay (%)*	*Sand (%)*	*Al_2O_3 (%)*	*Fe_2O_3 (%)*
Ferralsol 1	0.13	4.1	5.1	6	2.3	18	71	11	3
Ferralsol 2	0.10	3.3	4.1	5.5	2.1	18	71	11	3
Ferralsol 3	0.10	3.5	4.5	6.6	2.0	18	71	11	3
Ferralsol 4	0.15	4.3	5.3	6.4	2.1	18	71	11	3
Ferralsol 5	0.13	4.4	5.2	7.1	2.2	18	71	11	3
Nitisol	0.18	4.0	6.1	4.2	2.1	49	10	10	13
Ferralsol A	0.09	0.8	5.2	4.5	1.6	77	19	30	23
Ferralsol B	0.04	0.8	5.2	4.5	1.6	77	19	30	23
Goiânia A	0.11	12.0	12.8	8.22	2.0	16	68	11	5
Goiânia B	0.09	11.1	11.8	8.24	1.9	16	68	11	5
Goiânia C	0.10	13.6	14.4	8.49	2.5	16	68	11	5
Acrisol	0.08	1	6.3	5.4	0.5	9	82	4	2

The Nitisol is an acid silty clay loam soil, with low nutrient content and low CEC. The mineralogical analysis indicates the presence of hematite, goethite and traces of vermiculite. It was fertilized with urea, potassium chloride and calcium phosphate.

The Ferralsol (A and B) is an acid soil, rich in iron oxides, with clay texture, low nutrient content and low CEC. The mineralogical analysis indicates the predominance of hematite and goethite in the clay mineral fraction.

The Goiânia soil is also a Ferralsol rich in aluminium oxides, but it is an urban soil with a higher nutrient content and CEC. Only urea has been applied in these lysimeters when necessary for a better development of the crops. The mineralogical analyses of the clay fraction indicate the presence of gibbsite and kaolinite.

The Acrisol is an acid sand soil with low nutrient content and low CEC. The mineralogical analysis indicates the predominance of kaolinite. It was fertilized with urea, potassium chloride and calcium phosphate.

The Ferralsol together with Acrisol, represents more than 60% of Brazilian agricultural soils. Their main restriction for some crops is the low nutrient contents and acidity. These properties are also expected to produce high soil-to-plant transfer of radionuclides such as Cs and Sr.

3.2. Soil to Plant Transfer Factor for ^{137}Cs in Brazilian soils

Figure 1 shows that higher values of ^{137}Cs TF for maize occurred mainly associated with soil recently contaminated (Ferralsol A and B and Acrisol). Lower TFs occurred mainly with the Nitisol. In this figure it is possible to identify variations in the TF between the different years. These variations could be associated with changes in the plant metabolism due to environmental stresses, for example higher temperature in summer or injuries caused by insects.

FIG. 1. [137]Cs TF values for maize in three consecutive years.

Higher values of ^{137}Cs TF for cabbage occurred mainly associated with recently contaminated soil, Ferralsol A, B and Acrisol (see Fig. 2). The other soils produced very similar TFs, showing the same pattern for different years. As with maize, it is possible to identify variations in the TF between different years which again seem related to plant responses to stress.

The ^{137}Cs TF values observed for maize and cabbage in all soils were higher than recommended TF values for maize and cabbage cultivated in temperate soils [5].

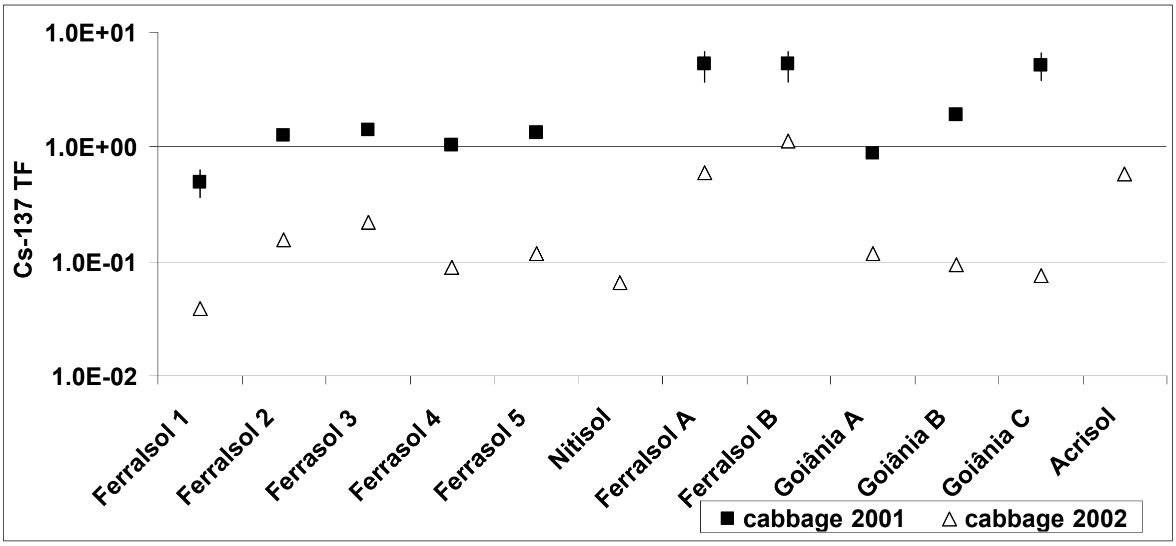

FIG. 2. [137]Cs TF values for cabbage in two consecutive years.

3.3. Soil to Plant Transfer Factor for ^{60}Co in Brazilian soil

TF values for ^{60}Co determined for maize in two consecutive years are shown in Fig. 3. Again, higher values of ^{60}Co TF for maize occurred mainly with recently contaminated soil (Ferralsol A and B and Acrisol). Lower TFs occurred mainly in the Nitisol.

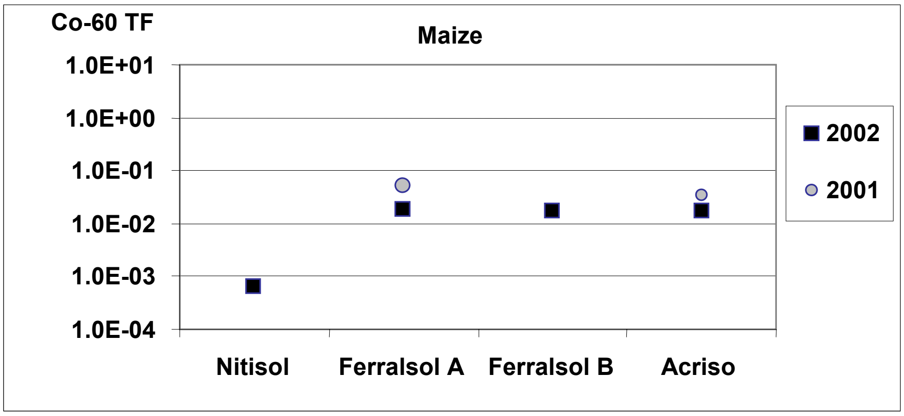

FIG. 3. ^{60}Co TF values for maize in two consecutive years.

As for maize, higher values of ^{60}Co TF occurred mainly with recently contaminated soil (Ferralsol A and B and Acrisol) and lower TF occurred in the Nitisol (Fig. 4). Figures 3 and 4 show that no significant variations in the TF occurred between the different years. These results seem to confirm that ^{137}Cs responds to some environmental changes, possibly due to its similarity with K and its role in the plant metabolism, since this behaviour was not observed for ^{60}Co.

The ^{60}Co TF values observed for maize and cabbage in all studied soil were similar to recommended TF values for maize and cabbage cultivated in temperate soils [5].

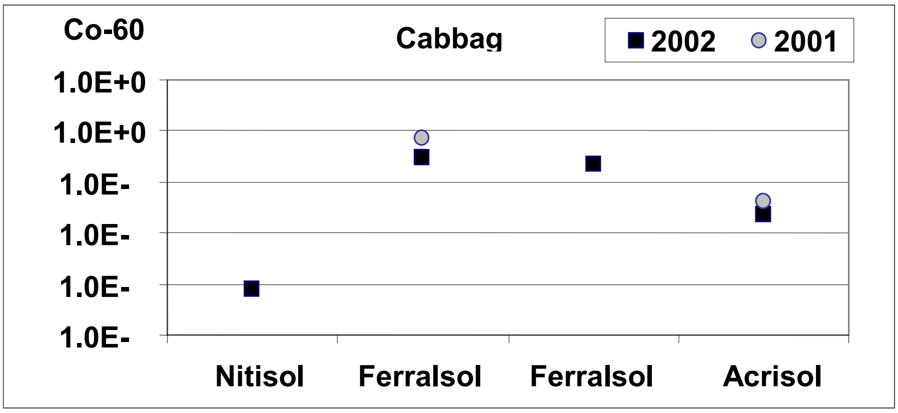

FIG 4. ^{60}Co TF values for cabbage in two consecutive years.

3.4. Soil-to-Plant Transfer Factor for ^{90}Sr in Brazilian soil

Preliminary TF values for ^{90}Sr determined for cabbage and maize are shown in Figure 5.

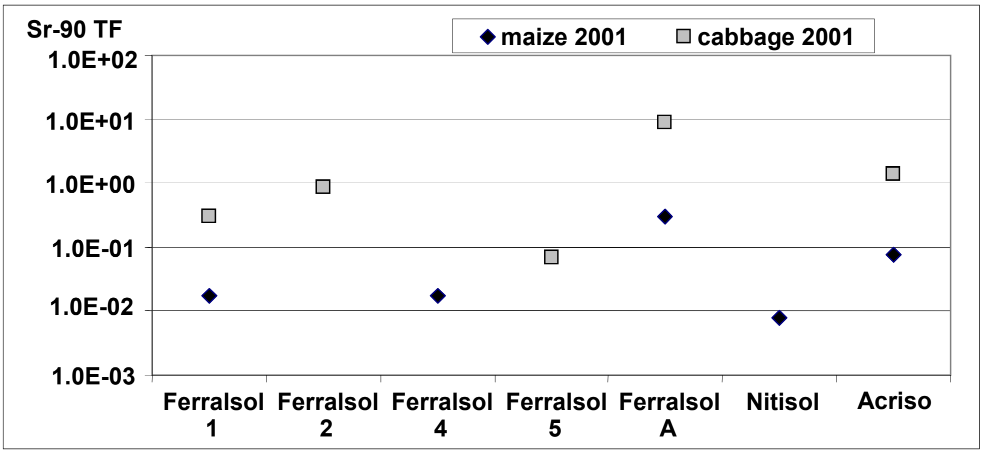

FIG. 5. ^{90}Sr TF values for cabbage and maize.

The TFs for ^{90}Sr varied up to two orders of magnitude for cabbage. This could not be accounted for by the differences in the soil activity for ^{90}Sr (see Table 1) nor by time elapsed after contamination, since all soils were contaminated in 2000. The variations for ^{90}Sr TF to maize cultivated in 2001 where greater than one order of magnitude and followed the same pattern as that for cabbage. More information is necessary to understand these results.

The ^{90}Sr TF values observed for maize and cabbage in all soils were lower than recommended TF values for maize and cabbage cultivated in temperate soils [5].

3.5. Soil-to-Plant Transfer Factor for ^{40}K in Brazilian soil

TF values for ^{40}K determined for maize are shown in Fig. 6. It shows that higher and lower values of ^{40}K TF for maize occurred in the same type of soil (Ferralsol 1 to 5).

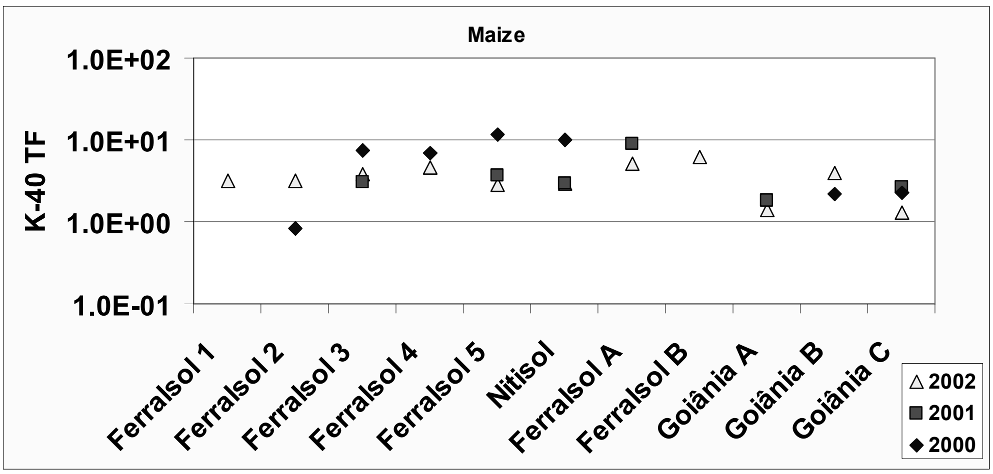

FIG. 6. ^{40}K TF values for maize in three consecutive years.

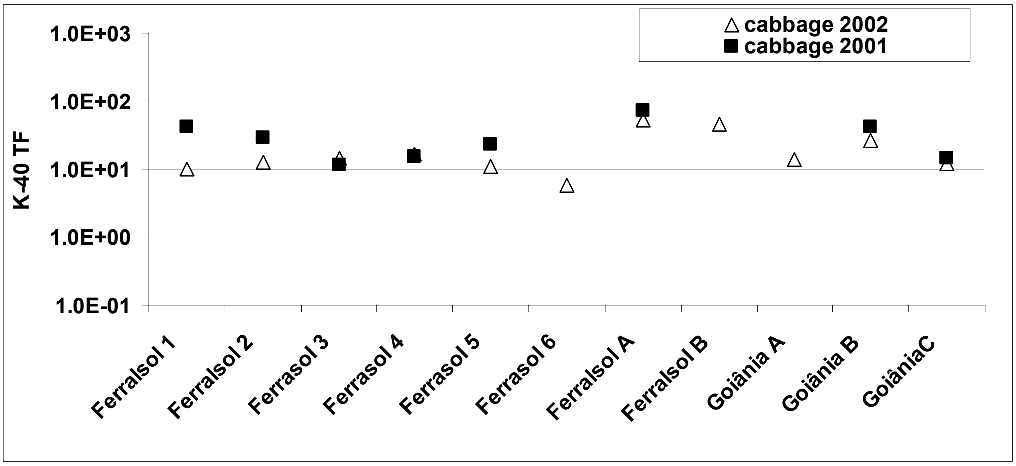

FIG. 7. ^{40}K TF values for cabbage in two consecutive years.

TF values for ^{40}K determined for cabbage in two consecutive years are shown in Fig. 7. These results did not show a specific behaviour among the different types of soils. These results were not correlated with the activity of ^{40}K in the soil that varied as follows: Ferralsol (1 to 5), 3.88E+01 ± 8.01E+00 Bq/kg of soil (n=5); Nitisol, 5.10E+01 Bq/kg of soil (single data); Ferralsol (A and B), 2.52E+01 ± 2.76E+00 Bq/kg of soil and Goiânia, 6.86E+01 ± 2.52E+01 Bq/kg of soil.

3.6. Transfer Factor Related with the Soil Parameters

Compared with Goiânia soil and Ferralsol (1 to 5), the other studied soils (Nitisol, Acrisol and Ferralsol A and B) differ in some properties, low pH, low exchangeable K and or low clay content, which influence the ^{137}Cs TF.

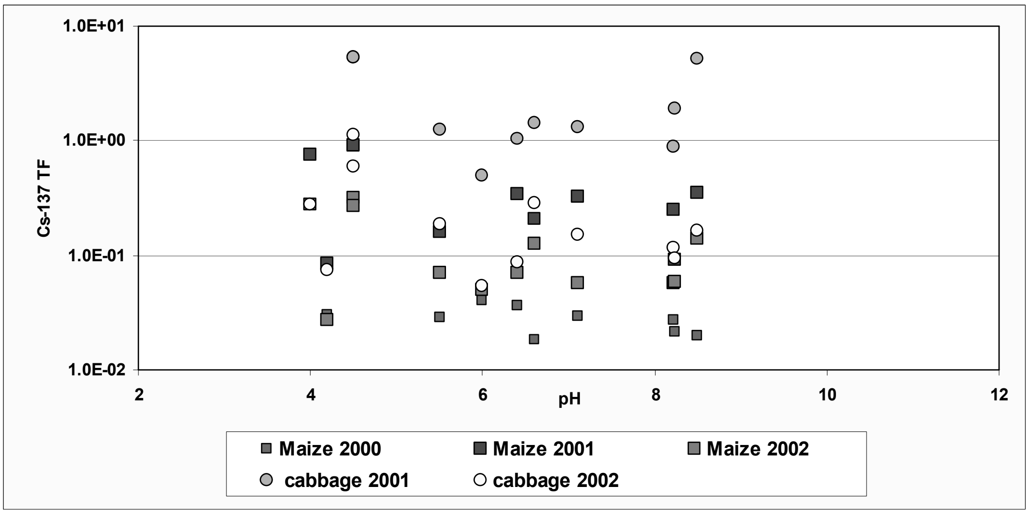

FIG. 8. Cs TF values for maize and cabbage versus pH.

Generally, there is a negative correlation between the Cs TF and the pH [1,2]. Frissel et al. [9] suggest that cereals growing in very acid soils (pH<4.8) are expected to present very high Cs TFs. Above pH4.8, TF for ^{137}Cs cannot be explained on the basis of pH. In this study, high ^{137}Cs TF values were observed for maize in acid soils (Acrisol and Ferralsol A and B) but not in the Nitisol (see Fig. 8).

Besides the influence of pH and variations due to season, as discussed in **3.2**, it is known that the clay mineral type can also influence the TF. In the presence of a 2:1 clay mineral type, the fixation of ^{137}Cs in the internal faces can occur a short time after contamination (less than three years), reducing transfer to plants [10]. The Nitisol is the only soil studied that contains traces of vermiculite and it was contaminated with ^{137}Cs in 1996, which allows enough time for Cs fixation. This can partly explain why, despite the lower pH (see Table 3), the ^{137}Cs TF was lower for maize and cabbage growing in the Nitisol compared with the Acrisol and the Ferralsol A and B (see also Figs 1 and 2). In previous work, a lower ^{137}Cs TF was also observed for radish cultivated in Nitisol compared with other acid soils [11].

Plants cannot discriminate between potassium and caesium [3]. Thus, soils containing higher concentrations of exchangeable potassium should decrease caesium root uptake, due to competition for carriers involved in root uptake.

Figure 9 shows that only soil with very low exchangeable K content (<0.10 cmol kg^{-1}) produced a high TF. Frissel et al. [11] suggest that cereals growing in soils with exchangeable K^+ concentrations smaller than 0.5 cmol kg^{-1} are expected to produce very high Cs TFs, above that limit, other factors seem to determine the TF. Soils with low fertility due to soil acidity, very limited fertilization and/or an extremely low exchangeable K content, are those more sensitive to ^{137}Cs contamination because of higher TFs than other soils [11].

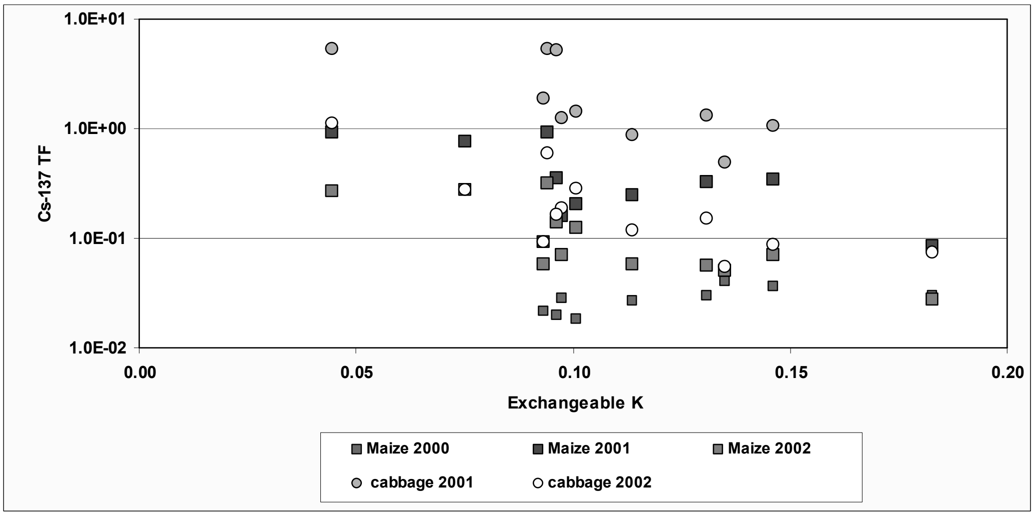

FIG. 9. ^{137}Cs TF values for maize and cabbage versus exchangeable K (cmol kg^{-1}).

The results presented in Figs 8 and 9 corroborate partially Frissel's findings in relation to pH and exchangeable K.

3.7. Geochemical partitioning of ^{137}Cs

The ^{137}Cs distribution on Goiânia soil and on the Ferralsol (Al rich) shows that similarities in soil properties determine similarity in the geochemical partitioning despite of differences in total concentration and time elapsed from the contamination (see Table 3 and Fig. 10). More than 10 years after contamination, about 70% of the total ^{137}Cs in these soils is distributed among the mobile geochemical phases, so that changes in the physical-chemical conditions can liberate Cs to the soil solution, and 10% of the total ^{137}Cs in these soils is potentially readily available for root uptake.

The ^{137}Cs readily available for plants (*slightly acidic phase*) was highest in the Ferralsol (Fe rich) and the Acrisol (Fig. 10). For the Ferralsol (Al rich) and Goiânia soil, the ^{137}Cs in the *slightly acidic phase* were similar as was the TF to maize (three seasons) and to cabbage (two seasons). This fraction was lowest in the Nitisol (3%) and the TFs in this soil were generally lower than in the other soils, as discussed previously. This corroborates the hypothesis that Cs fixation occurs in this soil that contains traces of vermiculite, thus reducing transfer to plants. It seems clear that Cs fixation is not a process that occurs systematically. Since 1996, about 10% of total ^{137}Cs concentration present in Goiânia soil remains readily available for root uptake. Studies with radish since the contamination in 1987 until

2000, showed that no significant difference in the TF in this soil, while increasing the pH in the Ferralsol reduced transfer to radish [10].

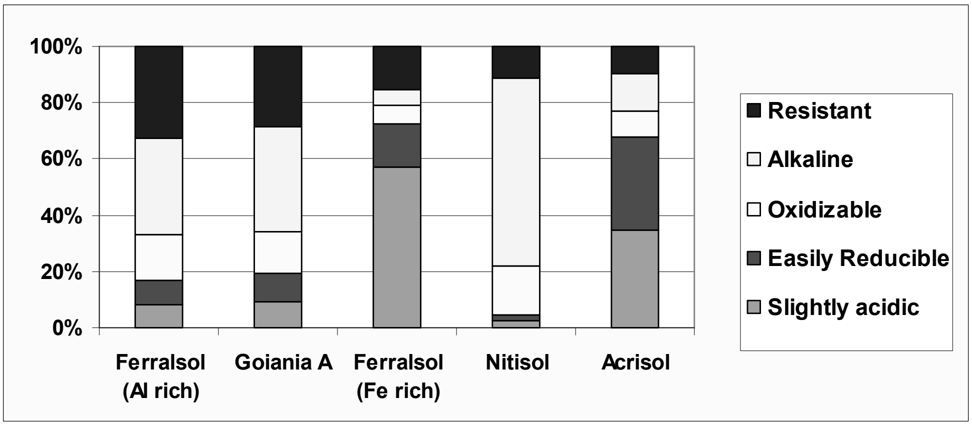

FIG. 10. Geochemical partitioning of ^{137}Cs in tropical soils.

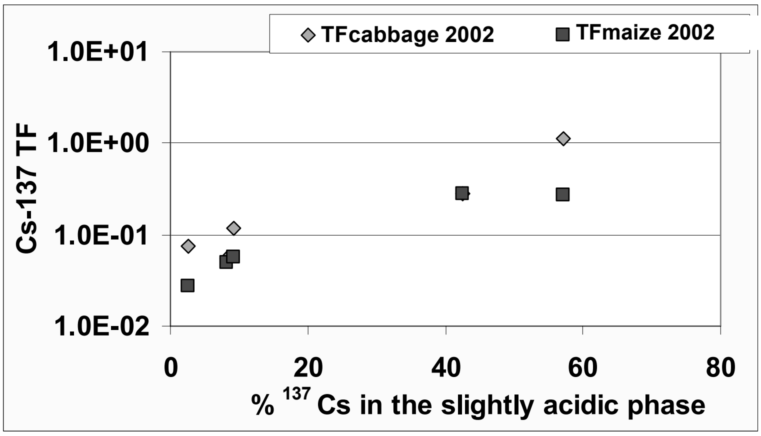

FIG. 11. ^{137}Cs in the slightly acidic phase (%), correlated with TF.

The *slightly acidic phase* correlates linearly with TF, showing that it serves as a good indicator of plant availability (Fig. 11): $maize_{2002}$ r=0.95 (n=12; p=0.999 r >0.80) and cabbage $_{2002}$ r=0.85 (n=12; p=0.999 r >0.80).

The ^{137}Cs levels in the *slightly reducible phase* did not correlate with any soil property. This phase is associated with manganese oxides that are present as traces compounds in the majority of tropical soils. The ^{137}Cs associated with this phase was generally below 20%.

The ^{137}Cs in the *oxidizable phase* (organic bound), for the Ferralsol (Al rich), Goiânia soil and the Nitisol were similar and are consistent with the similar organic C content in these soils. Despite the low carbon content in the tropical soils, more than 10% of the ^{137}Cs was bound to organic compounds (see Fig. 10).

Iron oxides are important compounds in most Brazilian soils. More than 60% of the ^{137}Cs was mobilized in the *alkaline phase*, showing that these compounds are an important sink for ^{137}Cs (see Fig. 8).

The ^{137}Cs in the *resistant phase* corresponds mainly to resistant compounds not destroyed in the previous phases and so is possibly not available for transfer to soil water. The soils with old contamination (Ferralsol, Al rich and Goiânia soil) contained the higher values of ^{137}Cs in this phase (see fig. 10).

3.8. Geochemical partitioning of ^{60}Co

Figure 12 shows the ^{60}Co distribution for the Nitisol, Ferralsol (Fe rich) and the Acrisol and it is clear that ^{60}Co is mainly associated with Mn oxides (*readily reducible phase*).

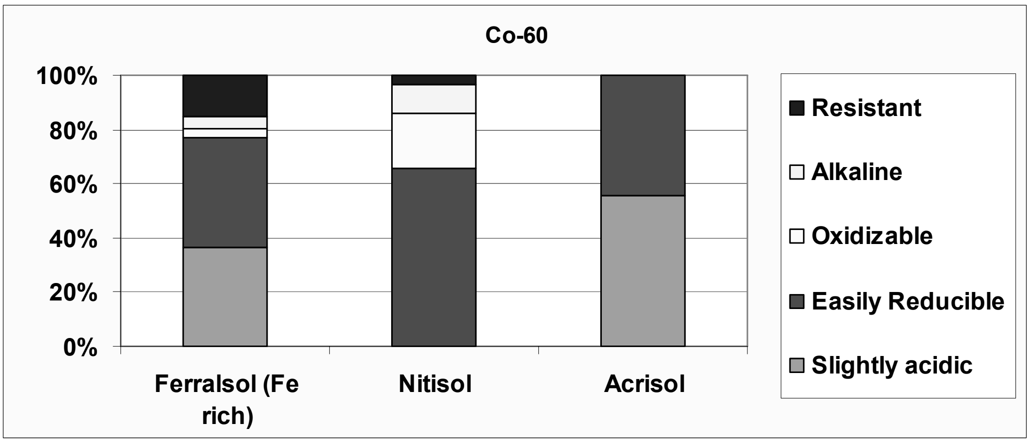

FIG. 12. ^{60}Co distribution in the tropical soil.

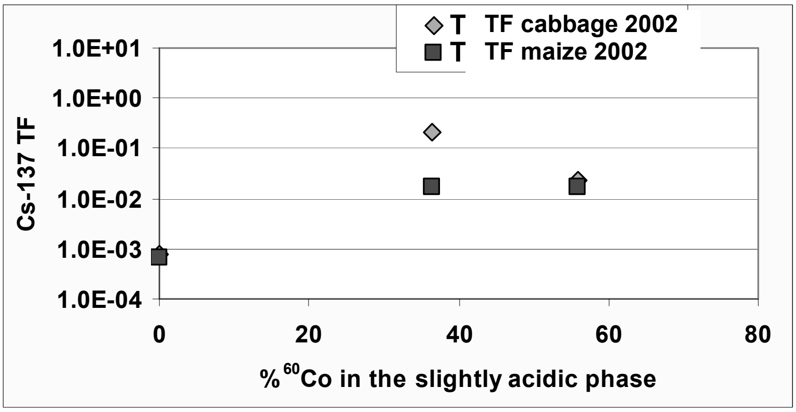

FIG. 13. ^{60}Co in the slightly acidic phase (%), correlated with TF.

There was no significant ^{60}Co in the *bioavailable phase* of the Nitisol, while this phase contained more than 40% of the ^{60}Co present in the Ferralsol (Fe rich) and in the Acrisol (see Fig. 12). This result was consistent with plant results, since little or no ^{60}Co was detected in the plants (grain or leaves) for the Nitisol while the TF was high to plants growing in the Ferralsol (Fe rich) and in the Acrisol. As observed for ^{137}Cs, the results for ^{60}Co in the *slightly acidic phase* correlate linearly with TF, showing that it serves as a good indicator of plant availability (Fig. 13): $maize_{2002}$ r=0.95 (n=3; p=0.999).

4. CONCLUSIONS

Although it is clearly difficult to identify which soil property determines a given TF, the levels of ^{137}Cs and ^{60}Co obtained in the slightly acidic phase correlated well with the TFs. These results were also coherent with some of the soil properties recognized in the literature as involved in the mechanisms of sorption of Cs (e.g. exchangeable K, organic matter and iron oxides content) and Co (e.g. manganese oxide). The ^{137}Cs distribution in soil showed that Fe oxides are the main sink for this element in all type of soil and 14 years after contamination the ^{137}Cs was still available for plants in the Ferralsol. In the Nitisol, five years after contamination, the ^{137}Cs and ^{60}Co was not considered as bioavailable (in the slightly acidic phase) and little or none of these elements was found in plants. In the Nitisol, it is possible that the low ^{137}Cs transfer be also associated with fixation in the internal faces of 2:1 clay mineral type. The integration of experimental results obtained in the laboratory with results obtained in the field seems to confirm that some Brazilian soils have high ^{137}Cs TFs. These results can be useful for risk assessment in the case of radionuclide contamination.

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INFLUENCE OF SOIL ACIDITY ON THE TRANSFER OF CAESIUM-137 AND OTHER RADIONUCLIDES FROM SOIL TO REFERENCE PLANTS

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Abstract

The transfer factor is a value used in evaluation studies on the impact of different releases of radionuclides to the environment. It accounts for the uptake of nuclides via the root and is defined as the ratio of units of radioactivity per unit of mass dry crop and the units of radioactivity per unit of mass dry soil. Two types of soils (Eutric Fluvisol and Chromic Luvisol) and two crops (wheat and cabbage) were investigated for determination of the transfer of ^{137}Cs, ^{54}Mn and ^{60}Co from soil to plant. Measurements were performed using gamma-spectrometry. Results for the soil characteristics, transfer factors of the radionuclides, and conversion factors (cabbage/wheat) were obtained. The transfer of ^{137}Cs is higher for Chromic Luvusol for both plants. The behaviour of the other radionuclides is different. A statistically significant dependence between the TF of ^{137}Cs and its concentration in soil was established for cabbage.

1. INTRODUCTION

The transfer factor is a value used in evaluation studies on the impact of different releases of radionuclides to the environment. It accounts for the uptake of nuclides via the root and is defined as the ratio of units of radioactivity per unit of mass dry crop and the units of radioactivity per unit of mass dry soil.

2. MATERIALS AND METHODS

2.1. Soils

Two agricultural soils (Eutric Fluvisol and Chromic Luvisol) representative of south-western Bulgaria were chosen. They were taken were taken from the Ap horizons in the vicinity of the villages of Sclave (Eutric Fluvisol) and Novo Delchevo (Chromic Luvisol) near the town of Sandanski - altitude 147 m, 21°17' longitude and 45°34' latitude. The average annual temperature is 13.9°C, (January average. 2.1°C, July average 24.9°C), average humidity 66% and average annual precipitation 533 mm. Agricultural crops grown on these soils are corn, vegetables, trees and some southern plants.

2.2. Experimental procedures

The experimental site is situated on the Experimental station of the Institute for Soil Sciences, Sofia.

Plants were grown in vessels with dimensions 70×40×35 cm and a volume of about 80 dm^3. About 50–55 kg soil was placed in each vessel to a depth of 22 cm. Twenty four vessels were filled with Eutric Fluvisol, 24 with Chromic Luvisol and there were six controls without radioactivity.

A premix was prepared with 1.5 kg of soil from each vessel and 10 ml of an aqueous solution of the mixed radionuclides ^{137}Cs, ^{60}Co and ^{54}Mn. These premixes were then mixed thoroughly with the soil in the vessels. The activity was 1500 Bq/kg from each of the three nuclides to give a total specific activity of 4500 Bq/kg. The radioactivity was applied August 10, 1999.

After a period for equilibration, cabbage was planted in half of the vessels (both soil types) and wheat in the rest. All the vessels are irrigated according the normal agricultural practice. During the first year, only half the vessels were fertilized. After that it was necessary to fertilize all vessels. The vessels were situated in the open and precautions taken to prevent contamination of the environment.

Wheat was planted in October 1999, 2000, 2001, 2002, and harvested in June 2000, 2001, 2002 and 2003, respectively. Cabbage was planted in June 2000, 2001, 2002 and October 2000, 2001, 2002 and harvested in September 2000, 2001, 2002 and May 2001, 2002, 2003, respectively.

2.3. Preparation of samples and measurement of radioactivity

2.3.1. Soils

Soil samples were taken at the time of crop harvest. An average sample of about 0.5 kg from each vessel was taken. Roots, small stones and other extraneous materials were removed by hand. The samples were air dried, homogenized in a ball mill and stored for analysis in plastic containers at 4^{o}C.

2.3.2. Wheat

Grain and straw were air dried, dried in an oven at 100^{o}C for 1 h and ground in a ball mill. Samples were stored in plastic containers at 4^{o}C.

2.3.3. Cabbage

Outer leaves of cabbage were washed with water to remove soil particles. The samples were weighed fresh and after drying. The samples were lyophilized and stored in plastic containers at 4^{o}C.

2.3.4. Measurement of radioactivity

Samples were pressed into standard 50 mL vessels. The soil samples weighed about 100 g and the plant samples about 40–50 g.

Gamma-spectrometry of the samples was performed using a Canberra model 7221 HPGe detector (efficiency 16% and energy resolution 1.9 KeV at 1332.5 KeV) connected to a Canberra series 85 multi-channel analyzer.

The detector was calibrated using IAEA standard reference materials and national reference materials with appropriate density and the same measurement geometry. Quality assurance and control was performed by analysis of standard reference materials and participation in round-robin tests.

2.3.5. Speciation studies

During the last two years of the experiment, speciation studies were carried out to determine the chemical form of the radionuclides in the soils. Dr. A. Wasserman, a CRP participant, kindly supplied the speciation scheme that was followed.

3. RESULTS

3.1.Characterization of soils

Soil I was a Eutric Fluvisol (FAO 1989) formed from alluvial river deposits. Soil II was a Chromic Luvisol (FAO 1989) formed from old quaternary carbonate deposits.

The morphological characteristics of the soils are given in Tables 1 and 2 and the textures of the soils are given in Table 3.

TABLE 1. CHARACTERISATION OF EUTRIC FLUVISOL

Horizon	Depth [cm]	Description
Ap	0–24	10 YR hue, 4/3 dry, middle loam sandy texture, loose
A_I	21–41	10 YR hue, 4/3 fresh, middle loam sandy texture, slightly hard
C_{1g}	58–80	10YR hue, 5/3 loam sandy texture, blocky structure
C_2	80–110	10 YR hue, 4/6 moist, coarse sand fraction with coarse aggregates, structureless

TABLE 2. CHARACTERISATION OF CHROMIC LUVISOL

Horizon	Depth [cm]	Description
Ap	0–22	7.5 YR hue, 5/4 dry, middle loam sandy texture, singular blocky structure, hard, single coarse aggregates
AB	22–50	7.5 YR hue, 4/4 dry, middle loam sandy texture, hard, middle to finely prismatic structure
B_{1t}	50–75	5 YR hue, 3/3 middle loam sandy texture, blocky prismatic structure, very hard
B_{2t}	75–106	7.5 YR hue, 5/8 fresh, middle loam sandy texture, blocky prismatic structure, very hard
Ck	106–140	10 YR hue, 6/8 old quaternary carbonate material, loam sandy texture, fresh, structureless

TABLE 3. MECHANICAL PROPERTIES [% of DW soil]

Horizon depth [cm]	Loss after treatment with HCl	Name and dimensions of particles [mm] Skeleton	Sand			Dust		Illite	Clay
Soil I		>1	1–0.25	0.25–0.05	0.05–0.01	0.01–0.005	0.005-0.001	<0.001	<0.001
Ap 0–24	1.0	0.5	23.9	27.9	17.6	4.9	1.3	22.9	29.1
A_I 24–41	1.1	0	15.6	31.2	20.6	5.4	3.6	22.5	31.5
A_{II} 41–58	1.5	0.3	15.9	29.8	17.7	6.5	4.8	23.5	34.8
C_Ig 58–80	0.7	0	30.7	38.8	11.6	3.6	1.7	12.9	18.2
C_2 80–110	0.5	24.6	48.7	17.7	2.3	2.0	1.5	2.7	6.2
Soil II									
Ap 0–22	1.3	3.3	18.4	25.8	15.5	3.9	2.5	29.3	35.7
AB 22–50	1.2	0	21.2	25.0	16.2	4.9	2.0	29.5	36.4
B_It 50–75	1.4	3.0	16.6	27.1	12.5	4.5	2.5	32.4	39.4
B_2t 75–106	0.8	23.6	12.9	26.4	7.7	3.9	2.5	22.2	28.6

Some general chemical properties of the soils are presented in Table 4, and the cation exchange capacities in Table 5.

TABLE 4. GENERAL CHEMICAL PROPERTIES OF THE SOILS

Horizon and depth [cm]	Humus %	pH KCl	pH H_2O	Total N (%)	Total P (%)	C:N ratio
Eutric Fluvisol						
Ap 0–24	1.04	6.1	6.6	0.064	0.161	9.4
A_I 24–41	0.59	5.9	6.8	0.034	0.087	10.0
A_{II} 41–58	0.56	5.9	6.9	0.040	n.d.	8.1
C_{Ig} 58–80	n.d.	6.4	7.0	n.d.	n.d	n.d.
C_2 80–	n.d.	6.1	6.5	n.d.	n.d.	n.d.
Chromic Luvisol						
Ap 0–22	1.09	4.7	5.6	0.065	0.101	9.7
AB 22–50	0.65	5.2	6.3	0.043	0.083	8.7
B_{1t} 50–75	0.50	5.3	6.6	0.037	n.d.	7.8

TABLE 5. CATION EXCHANGE CAPACITY [mequiv/100 g soil]

Horizon	Ca	Mg	Na	K
Eutric Fluvisol				
Ap	14.2	3.88	0.26	0.60
A_I	15.5	1.95	0.26	0.40
A_{II}	14.7	2.15	0.48	0.58
Chromic Luvisol				
Ap	19.7	9.30	0.04	7.80
AB	18.2	9.10	0.04	10.80
B_{1t}	22.1	10.10	0.04	8.10
B_{2t}	18.5	9.70	0.03	5.40

ICP-MS was used to characterize the total element composition of the investigated soils and the results are presented in Table 6.

TABLE 6. CHEMICAL COMPOSITION OF THE INVESTIGATED SOILS [mg/kg]

Element	Eutric Fluvisol	Chromic Luvisol	Element	Eutric Fluvisol	Chromic Luvisol
Al	5020±150	2600±100	La	4.7±0.2	9.8±0.1
As	7.41±0.14	7.37±0.07	Lu	0.24±0.01	0.22±0.02
B	20±2	8.5±0.9	Mg	1650±50	1150±20
Ba	230±25	360±30	Mn	510±20	350±30
Be	1.37±0.02	2.89±0.03	Na	1340±10	1110±10
Bi	0.79±0.01	0.78±0.01	Nd	19±1	10.2±0.2
Cd	1.01±0.08	0.65±0.07	Ni	31.2±0.3	27.4±0.3
Ce	42.5±0.5	23.1±0.2	Pb	20±1	31±1
Co	10.8±0.9	7.0±0.9	Pr	4.5±0.6	2.5±0.2
Cr	76±1	62±1	Rb	52±1	41±1
Cs	68±2	53±4	Sb	10.1±0.9	7.0±0.8
Cu	32±2	24±1	Sm	4.1±0.1	2.3±0.1
Dy	3.51±0.04	2.44± 0.02	Sr	152±2	144±2
Eu	1.04±0.01	0.63±0.01	Tb	0.59±0.05	0.38±0.01
Er	1.90±0.02	1.49±0.02	Th	10.1±0.7	5.9±0.3
Fe	3200±100	2200±200	Tl	0.33±0.01	0.69±0.07
Ga	19.2±0.2	30±1	U	3.3±0.03	2.32±0.02
Gd	7.67±0.08	4.21±0.12	V	68±2	72±4
Ho	0.65± 0.07	0.48±0.08	Y	16.7±0.9	11.0±0.9
K	3400±500	2500±600	Yb	1.72±0.02	1.51±0.02
			Zn	72±3	65±3

3.2. Activity measurements and determination of transfer factors

The individual results from the measurement and transfer determinations are presented for all crops and years together with the results of the other participants in Appendix 2.

4. DISCUSSION

4.1. Changes of the TF with the time after contamination

4.1.1. Wheat

The overall means for TF for ^{137}Cs are presented in Table 7 and Table 8 shows the change in TF of ^{137}Cs for wheat (grain and straw) during the four years of the experiment.

As expected the TF for straw is higher than for grain. The variation with the years (relative standard deviation), calculated from the results indicates the relative stability of the TF (16 to 35%) and keeping in mind the large intervals reported for the TF it indicates that for the two types of soils investigated in this study the changes in the TF for the first 3–4 years after contamination are small.

TABLE 7. GEOMETRIC MEANS FOR THE TF OF ^{137}Cs TO WHEAT

	Chromic Luvisol	Eutric Fluvisol
Grain	0.020±0.007	0.0069±0.0010
Straw	0.0375±0.009	0.023±0.0012

TABLE 8. CHANGES OF THE TF OF ^{137}Cs FOR WHEAT GRAIN AND STRAW WITH TIME AFTER CONTAMINATION

Year	Chromic Luvisol		Eutric Fluvisol	
	Grain	Straw	Grain	Straw
2000	0.012	0.03	0.056	0.029
2001	0.032	0.054	0.0084	0.054
2002	0.018	0.032	0.0064	0.032
2003	0.020	0.034	0.0059	0.034

4.1.2.Cabbage

Table 9 presents the changes in the transfer factor for cabbage. For both types of soils there is an increase of the TF after the first year and decrease after the second and third years. For Chromic Luvisol the increase and decrease after that are substantial. For Eutric Fluvisol the changes are not so drastic.

TABLE 9. CHANGES IN THE TF FOR ^{137}Cs IN CABBAGE WITH TIME AFTER CONTAMINATON

Year	Chromic Luvisol	Eutric Fluvisol
2000	0.055	0.030
2001	0.18	0.061
2002	0.037	0.050
2003	0.037	0.046
Overall geometric means	0.080±0.049	0.052±0.015

4.2. Changes of the ^{137}Cs TF in cabbage with the season of growing and the stage of development

Cabbage in Bulgaria can be grown not only during the summer but also during the winter. The results comparing summer and winter crops for cabbage grown on the Chromic Luvisol are presented in Table 10. For all radionuclides the TF for winter cabbage is higher than for the summer crop. The explanation might be that the vegetation period (6 months) of winter cabbage is twice as long as that of summer cabbage (3 months). However, for cabbage grown the on Eutric Fluvisol there was no statistically significant difference in the TF for winter and summer cabbage although the TF values are slightly higher for the winter crop (results not shown).

In addition cabbage was collected before the formation of the heads (only leaves) and the TFs were higher in comparison to fully developed plants (results not shown).

TABLE 10. COMPARISON OF THE TFs FOR WINTER AND SUMMER CABBAGE GROWN ON CHROMIC LUVISOL

Radionuclide	Winter cabbage	Summer cabbage
^{137}Cs	0.432	0.183
^{54}Mn	0.228	0.083
^{60}Co	0.094	0.015
^{40}K	1.772	0.906

4.3. Dependence of the TF of ^{137}Cs on different factors

4.3.1. Dependence on the concentration of ^{40}K

The comparison of the concentrations of ^{40}K and ^{137}Cs as well as of the TF's for both nuclides did not indicate any statistically significant dependence.

4.3.2.Dependence on the concentration of ^{137}Cs in the soil

There was a statistically significant dependence between the TF of ^{137}Cs and the concentration of ^{137}Cs in the soil for cabbage but not for wheat. Figure 1 presents the results for cabbage.

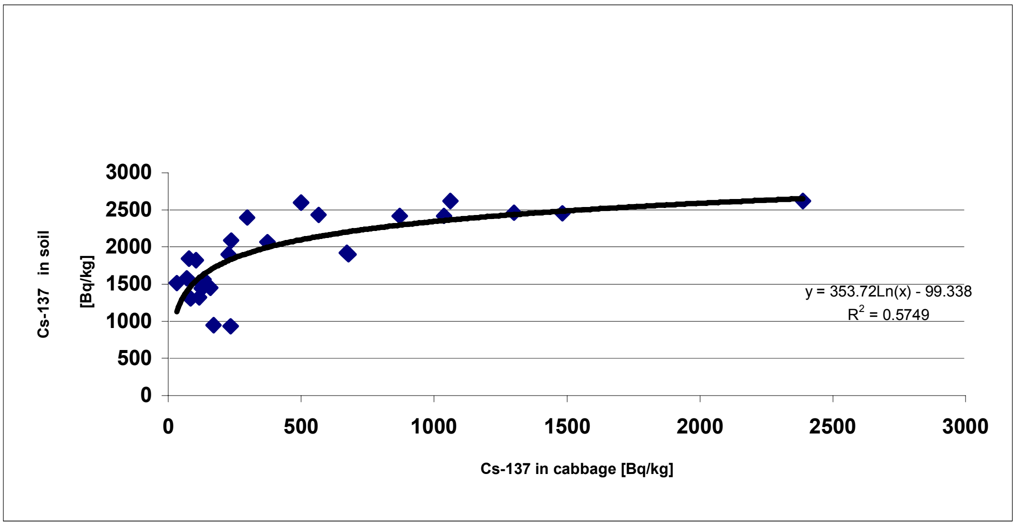

FIG. 1. Dependence between the concentrations of Cs-137 in cabbage and soil.

4.3.3.Effect of NH^{4+} ions

In half of the vessels (from each soil) 30 mg/kg NH^{4+} was applied just before planting. The comparison of the TF's between fertilized/non-fertilized plants for straw and cabbage shows no significant difference. In grain, however, there is about 40% decrease in TF.

4.4. Dependence of the TF of ^{137}Cs on soil properties

For both crops (wheat and cabbage) the TFs of ^{137}Cs are higher for plants grown on Chromic Luvisol than on Eutric Fluvisol. It is worth mentioning that all TFs determined in the present study coincide with the overall average TF for the respective soils [2–4]. The difference between the TFs is highest for wheat grain where the TF for Chromic Luvisol is 7 times higher than for Eutric Fluvisol. For wheat straw and cabbage the TFs for Chromic Luvisol are about 2 times higher that for Eutric Fluvisol.

The speciation experiments for both types of soils indicated that after the third year about 80% of the ^{137}Cs was already in a form not readily available to the plants. The Chromic Luvisol contains 10 times more available K than the Eutric Fluvisol. Therefore the result obtained contradicts the concept that the higher the available K in the soil the lower the transfer of ^{137}Cs to the plant. Obviously other factors are more important. In addition to available K the two soils differ in several other parameters (see the Tables above). Chromic Luvisol has substantially lower available Na, lower pH, higher available Ca and Mg, higher illite and clay content. It might be presumed that the soil texture, clay content, pH value and the combination between the available ions is more important for uptake of ^{137}Cs than the content of available K only.

4.5. Conversion factors

The conversion factors between cabbage and wheat are different for the two types of soils. For the Chromic Luvisol the average conversion factor cabbage/wheat grain is 3.85 about half the value for the Eutric Fluvisol (where it is 7.1). The conversion factors cabbage/wheat straw are very similar for both soils (2.1 and 2.6 respectively).

4.6. TFs for ^{54}Mn and ^{60}Co

The main aim of the program has been determination of the TFs for ^{137}Cs and ^{90}Sr. Therefore the results for the transfer of ^{54}Mn and ^{60}Co will be discussed very briefly (they are not being determined by the other participants). The transfer of both radionuclides is higher for plants on the Chromic Luvisol than on the Eutric Fluvisol but the difference is not so significant as for ^{137}Cs. The TF for Mn is always higher that for Cs while that for Co is lower or similar to Cs. The changes in the TFs with the time after contamination follow the same trend as for Cs.

5. CONCLUSIONS

The results may be summarized as follows:

The transfer of ^{137}Cs from soil to plants is dependant mostly on soil pH and texture while available K seems not to play a significant role. There is an expected decrease in the TF with the time after contamination of the soil however in some cases there was a temporary increase after the first year. This is valid for both wheat and cabbage.

The transfer of ^{137}Cs from soil to cabbage is dependant on the vegetation period. For winter cabbage (6 months vegetation period) the TF was higher especially for cabbage grown on the more acidic soil.

The conversion factors (cabbage/wheat) are dependant on the type of soil being lower for the slightly acidic soil.

The dependences that have been established between different parameters and the TFs for ^{137}Cs and the other nuclides studied under the present contract need further confirmation.

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SOIL TO CROPS TRANSFER OF RADIOCAESIUM AND -STRONTIUM IN DIFFERENT ALLOPHANIC SOILS FROM THE LAKE REGION, CHILE

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Abstract

The transfer factor (TF) of radiocaesium and -strontium from soil to Swiss chard (*Beta vulgaris var. cicla*), sweet corn (*Zea mays var. saccharata*) and cabbage (*Brassica oleracea var. capitata*) was studied in two different allophanic soils (Umbric Andosol and Dystric Fluvisol) characteristic of the Lake Region, an important agricultural region situated in central-south Chile. The TFs for both radionuclides were generally within the ranges in the literature. Cs TFs showed no effect of "ageing". There was a pronounced decrease of TFs in sequential harvests of chard during the course of the season by a factor of 3–4 for Cs and 4–8 for Sr. With all crops application of fertilizer containing 90 kg ha^{-1} of K reduced all Cs TFs. Application of fertilizer containing 100 kg ha^{-1} of Ca did not affect Sr TFs.

1. INTRODUCTION

In Chile, the highest areal activity densities of global fallout ^{137}Cs (up to 5400 Bq m^{-2}) occur in the Lake Region, because of its mid-latitudinal position (39–43ºS) and, predominantly, because of high rainfall (1500–4000 mm y^{-1}) [1]. The Lake Region is one of the most important agricultural regions of Chile. The transfer factors (TF) of different radionuclides from soil to edible plants need to be studied under the site-specific conditions and for the representative soil types of this Region. This is because of the higher potential risk of anthropogenic fallout from the atmosphere, compared to other areas from the Southern Hemisphere, and because the soils are predominantly allophanic. No data were available on long-lived radionuclide TFs to edible crops for this area before the present research.

Only a few studies related to this task have been made in Chile. During 1982-1990, global fallout ^{137}Cs TFs soil-to-pasture grass were determined for different soil types of the Region. They ranged from <0.02–0.40 in Palehumult (clay soil), from 0.05–0.62 in Hapludand (loamy soil), being highest from 0.52–5 in the flooded Placandept (loamy soil) [2]. The time dependency of the ^{137}Cs in the soil-pasture grass-milk pathway was also observed in seven dairy farms in the lake Region. The rate of decrease of ^{137}Cs concentration in the plants, showed an effective half life of 5.6 y from 1982 to 1990 and became slower between 1991 and 1997 with an effective half life of 12 y [2, 3].

The objectives of this research were to study the TF of radiocaesium and -strontium from soil to Swiss chard (*Beta vulgaris var. cicla*), sweet corn (*Zea mays var. saccharata*) and cabbage (*Brassica oleracea var. capitata*) in two different characteristic allophanic soils of an agricultural region of central-south Chile, an Umbric Andosol and a Dystric Fluvisol; to study the influence of K-fertilization on the radiocaesium transfer and of Ca-fertilization on the radiostrontium transfer; to observe the time course of the TF to Swiss chard and to sweet corn.

2. MATERIALS AND METHODS

2.1. Study area

The study sites are located at Santa Rosa farm, an Experiment Station of the Universidad Austral de Chile located in Valdivia, Chile (39º 46'S, 73º 14'W). Two of the main soils for crop production in south central Chile occur on this farm: Umbric Andosol and Dystric Fluvisol. The latter is flooded during autumn, winter, and spring. The mineralogy of the Umbric Andosol is dominated by allophane in the upper horizon, with an increase of gibbsite and metahallosysic clays in deeper horizons [4]. The mean annual rainfall in the area is about 2300 mm y^{-1}, with the heaviest rates between April and

October (autumn-winter period). Figure 1 illustrates the mean monthly rainfall estimated for the Experiment Station during the years 1960 to 2002 [5].

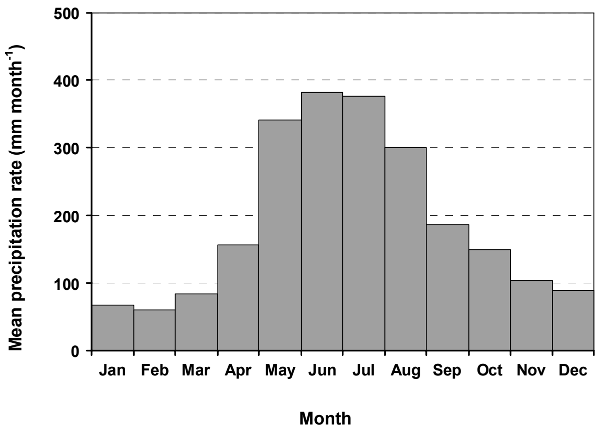

FIG. 1. Mean monthly precipitation at Santa Rosa Experiment Station (39º46'S, 73º14'W).

2.2. Preliminary trial

A preliminary field experiment was performed during the growing period 1999–2000 in the Umbric Andosol to estimate the amount of radiocaesium to be incorporated to the soil that would not cause the concentration in the plants exceed the derived intervention level of 1500 Bq kg^{-1} given for green vegetables in Chile [6]. The time-course of the radiocaesium TF soil-to-Swiss chard (whole plants), with and without K-fertilization, was also observed.

2.3. Plot installation

At the end of 2000, plots and check plots (without radiocaesium-labelling) of 6.3 m x 1.2 m were installed for all radiocaesium treatments in both soils. The upper 0 – 20 cm soil layer was removed from each plot and sieved. The plots were contaminated with 100 kBq ^{134}Cs m^{-2} (~800 Bq ^{134}Cs kg^{-1}): the Umbric Andosol on November 14 2000 and the Dystric Fluvisol on January 3 2001. To obtain an almost homogeneous distribution of the radionuclide in the soil, the ^{134}Cs was incorporated down to 20 cm by pouring 1/10 of the total amount of CsCl in aqueous solution[a] over each 2 cm layer with a watering can. To minimize the adhesion of radionuclides to plant leaves [7,8], the upper 2 cm soil layer was not contaminated.

Similar plots and check plots of 1.0 m x 1.2 m (according to the amount of radiostrontium available for the contamination of the plots) were installed for Sr and Sr-Ca. During two consecutive years (2001 and 2002), as soon as the rainy season ended, the plots were contaminated with approximately 100 kBq ^{85}Sr m^{-2} (~800 Bq ^{85}Sr kg^{-1}) using $SrCl_2$ in 0.5 M HCl[b].

Every spring, the plots were prepared for cultivation using the local agricultural practices and procedures. The soil was manually inverted, treated with weed killers and fertilized before planting or seeding. The soil surface was covered again with a 2 cm deep uncontaminated soil layer to avoid the deposition of contaminated soil on plant leaves.

2.4. Soil fertilization

P and N were added to the soil, in order to improve plant growth under the natural low nutrient status of the selected soils, as observed in a preliminary trial. K-fertilizer was added to half of the Cs

[a] Provided by Amersham Pharmacia Biotech UK Limited, Amersham Place, Little Chalfont, Buckinghamshire HP7 9NA, UK
[b] Provided by New England Nuclear GmbH, Spandlgasse 58, 1220 Vienna, Austria

contaminated plots and Ca-fertilizer to half of the plots labelled with Sr. The amounts of fertilizers added to the top 10 cm were:

Triple superphosphate (all plots) (300 kg ha^{-1})
Sodium nitrate (all plots) (200 kg ha^{-1})
Potassium sulphate (only for plots with ^{134}Cs-K treatment) (200 kg ha^{-1} ≈ 90 kg K ha^{-1})
Soprocal (only for plots with ^{85}Sr-Ca treatment) (400 kg ha^{-1} ≈ 100 kg Ca ha^{-1})

2.5. Plants selected

Swiss chard (*Beta vulgaris* L. var. *Cicla* L.*)* was selected for observing the time course of the TF, because it is harvested several times per season. This hardy biennial plant is grown exclusively for the large, fleshy and glossy leaves. It has a taproot with many deep secondary roots down to 0.20 m, exploring a great soil volume. It produces abundant basal large green leaves during spring, summer and part of autumn, becoming smaller and less numerous through winter. After wintering, chards produce stalks with caulinary leaves which are still edible, and also useful for foliar analyses.

The Swiss chard plants were transplanted avoiding the contamination of leaves by leaving the upper soil layer (2 cm) without Cs or Sr; the plants were irrigated according to needs and weather conditions. The species were rotated annually simulating the conventional regional practises.

Sweet corn was selected because it is widely grown and generally accepted by Chilean consumers. In addition, there is ample knowledge of TFs to maize for other soil types to permit appropriate comparisons.

Cabbage was cultivated during the last growing period, because it is often classified in a separate crop group [9, 10].

2.6. Sample collection

For observing the change with time of TFs to Swiss chard, plants were harvested sequentially, every 1–2 months, from late summer to the end of winter in the Umbric Andosol and to the beginning of the flooding season (autumn) in the Dystric Fluvisol. For the Cs treatment, in most cases there was enough material for three replicates (each from 1/3 of the plot), separated into external and internal leaves and external and internal stalks.

The sweet corn and cabbage were harvested when they were ripe for human consumption. They were harvested earlier in the Dystric Fluvisol when a flood shortened the growing season.

Three replicate soil samples were collected every year at the end of the harvest period along the plant rows from the upper 20 cm soil layer for measurement of ^{137}Cs and ^{85}Sr. Incremental 2 cm soil samples were also collected down to 30 cm to check for possible vertical migration of Cs below the treated layer. Additionally, soil bulk samples were collected annually from the upper 20 cm layer to determine the physical and chemical soil properties.

2.7. Gamma analyses

Swiss chard and cabbage leaves were washed before performing gamma analysis. The radionuclide concentration in plants was determined in the fresh material. Afterwards, the vegetables were dried at room temperature and later at 105ºC. The ^{134}Cs concentration in soil and vegetable samples was measured using a Canberra GC2518 gamma detector, in 500 mL Marinelli beakers and in 81.3 mL Petri dishes, depending on the available sample volume. The efficiency was determined for the Marinelli beaker and Petri dish geometries, for several soil and vegetable matrices of different density using a gamma standard solution supplied by PTB[c]. Spectra were analysed using Accuspec B software from Nuclear Data[d]. The counting time for each sample was set between 6 and 20 h, depending on the sample activity. Under these conditions the detection limit for ^{134}Cs was about 0.2 Bq kg^{-1} in soil (Marinelli beaker), and ranged from 0.3 Bq kg^{-1} (Marinelli beaker) to 0.8 Bq kg^{-1} (Petri dish) for fresh vegetable samples. The detection limit for ^{85}Sr concentration in fresh vegetables varied from 0.5 Bq kg^{-1} in the Marinelli beaker to about 2.0 Bq kg^{-1} in the Petri dish.

3. RESULTS

3.1. Soil analyses

The chemical properties of both soils are given in Table 1. The Umbric Andosol is a silty loam with about 23.8% clay, 68.2% silt, and 8.0% sand; the Dystric Fluvisol is a loam soil with about 11.4% clay, 42.3% silt and 46.3% sand.

Less than 1% of the total ^{134}Cs had migrated below the labelling depth of 20 cm in the Umbric Andosol and less than 8% in the Dystric Fluvisol 700 days after soil contamination.

TABLE 1. CHEMICAL CHARACTERISTICS OF THE SOILS

Soil type	Umbric Andosol						Dystric Fluvisol					
Crop type	Sw. chard, cabbage			Sweet corn			Sw. chard, cabbage			Sweet corn		
Fertilization	None	+K	+Ca	None	+K	+Ca	None	+K	+Ca	None	+K	+Ca
pH (1:2.5) in water	5.5			5.7			5.3			5.2		
pH (1:2.5) in $CaCl_2$ 0.01M	4.7			4.9			4.5			4.4		
Organic matter (%)	19			18			4.8			4.8		
Available N (μg/g $N\text{-}NO_3$)	28			19.6			11.2			15.4		
Available P (μg/g)	14			17			93			87		
Exchangeable K (meq/100 g)	0.3	0.6		0.5	1.0		0.2	0.5		0.3	0.7	
Exchangeable Na (meq/100 g)	0.09			0.2			0.2			0.06		
Exchangeable Ca (meq/100 g)	1.7		2.1	1.8		1.8	1.1		2.0	0.9		1.3
Exchangeable Mg (meq/100 g)	0.4	0.5		0.4			0.2			0.3		
Exchangeable cations (meq/100 g)	2.3	2.7	2.9	3.1	3.7	3.1	1.7	2.1	2.7	1.1	2.2	1.8
Exchangeable Al (meq/100 g)	0.4			0.4			1.8			2.7		
Al saturation (%)	15			11			47			60		
CEC (meq/100 g)	41			39			18			20		

3.2. Seasonal variation of the ^{134}Cs transfer factor soil-to-Swiss chard

3.2.1. Umbric Andosol

The time-course of the radiocaesium TF soil-to-Swiss chard in the Umbric Andosol during the first two years after soil contamination is represented in Fig. 2 for the soil without K-fertilizer (left) and with K-fertilizer (right), respectively.

[c] Physikalisch-Technische Bundesanstalt, Gieselweg 1, D-38110 Braunschweig, Germany.

[d] ND Instrumentation Division, Golf and Meacham Roads, Schaumburg, Illinois 60196, USA.

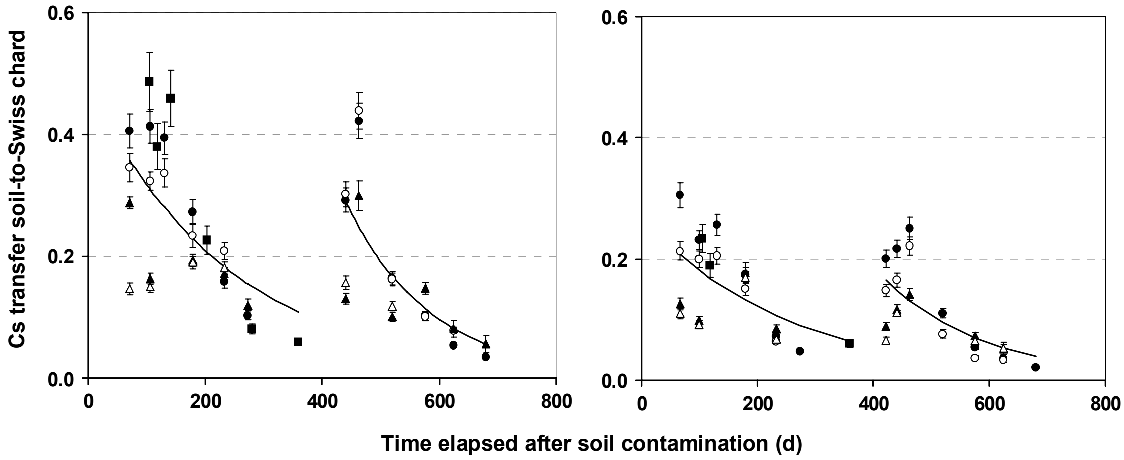

FIG. 2. Seasonal variation of the radiocaesium transfer factor soil-to-Swiss chard (● external leaves, ○ internal leaves, ▲ external stalks, Δ internal stalks, ■ leaves and stalks) at the Umbric Andosol without K-fertilization (left) and with K-fertilizer (right).

3.2.1.1. Difference of the transfer factors between various plant parts

Because the TFs were determined separately for several plant parts of Swiss chard, it was necessary to check first whether there were significantly different TFs between external and internal leaves and between external and internal stalks. For this purpose the Wilcoxon matched pair test was applied. There was no difference in the TFs obtained in the soil without K-fertilization (p>0.05, two sided). In the soil with K-fertilization, there was a significant difference only between the external and internal leaves (p<0.01, two sided) where the TF of the external leaves was slightly larger than that of the internal leaves by a factor of 1.2 (median; 95% confidence limits 1.1–1.4).

3.2.1.2. Effective half-life of the transfer factors

To characterize the observed decrease of the TF with time (see Fig. 2), the effective half-life T_{eff} of the TF was evaluated by using nonlinear regression analysis of the function.

$$TF = A \exp\left[-\ln(2)t / T_{eff}\right]$$

The TFs of all edible plant parts (leaves and stalks) for each sampling date were used. For the first harvest period, t represents the time elapsed since the date of soil contamination. For this period, the TFs determined during the preliminary trial were included in the regression curve. For the second harvest period, the parameter t represents the time elapsed since one year after the soil contamination date. The resulting summary of these regressions is given in Table 2 (columns 1 and 2 for the two periods in the soil without K-fertilization and columns 4 and 5 for the periods in the soil treated with K-fertilization). All these fits are highly significant (p<0.01). Also the values of T_{eff} are strongly significant (p<0.01).

TABLE 2. SUMMARY STATISTICS OF THE EXPONENTIAL FIT OF THE RADIOCAESIUM TRANSFER FACTOR (TF) ACCORDING TO $TF = A\ exp(-ln(2)\ t / T_{eff})$ FOR UMBRIC ANDOSOL AND DYSTRIC FLUVISOL UNDER DIFFERENT SOIL FERTILIZATION PRACTICES

Soil type	Umbric Andosol					
Harvest period,	First year,	Second year,	First year,	Second year,	Both years combined,	Both years combined,
soil fertilization	no K	no K	with K	with K	no K	with K
R	0.6485	0.7353	0.6135	0.7060	0.6513	0.6125
Sign. level of *r*: (*P*> F)	0.0003	0.0003	0.0024	0.0002	< 0.00001	0.00001
A	0.482	0.486	0.270	0.226	0.474	0.242
95%-confidence limits of A	0.31–0.65	0.24–0.74	0.16–0.37	0.14–0.31	0.33–0.61	0.175–0.309
$T_{1/2,eff}$ (d)	167	101	174	124	139	156
95% conf. limits of $T_{1/2,eff}$ (d)	67–266	38–163	47–300	46–201	77–200	78–235
Sign. level of $T_{1/2,eff}$ P =	0.002	0.003	0.0098	0.0032	0.00004	0.0002
Statistical error of $T_{1/2,eff}$ (d)	48.2	29.6	60.88	37.18	30.4	38.9

Soil type	Dystric Fluvisol	
R	0.7968	0.7641
Sign. level of *r*: (*P*> F)	0.0058	0.00618
A	0.738	0.211
95% confidence limits of A	0.13–1.34	0.11–0.30
$T_{1/2,eff}$ (d)	47	61
95%-conf. limits of $T_{1/2,eff}$ (d)	12–82	16–105
Sign. level of $T_{1/2,eff}$ p =	0.015	0.013
Statistical error of $T_{1/2,eff}$ (d)	15.1	19.7

3.2.1.3. Difference of T_{eff} between the first and second year

The different values shown in Table 2 for T_{eff} for the different harvest periods (soil without K-fertilization: first year 167 d, second year 101 d; soil with K-fertilization: first year 174 d, second year 124 d) suggest at first glance that these values are smaller in the second year than in the first. However, the 95%-confidence limits of T_{eff} are rather large. Application of a test [11] for detecting differences between two regression coefficients showed that the differences in the T_{eff} between two succeeding harvest periods are not significant (P>0.05 for both fertilized and unfertilized plots).

3.2.1.4. Difference of the transfer factors between the first and second harvest

The Wilcoxon matched pair test was used to compare TFs from the first and second harvest periods. Because not all plant samples were collected at the same date during the first and second harvest period, each period was divided in intervals of 30 d and the median TF observed in all plant parts in each period was used in this test. There is no significant difference between the TF in either the first year and the second year (P>0.05, two sided) in both fertilized and the unfertilized soil.

Because the absolute values of the TF and of the T_{eff} are not significantly different for the successive harvest periods, it is possible to combine the corresponding TF to obtain an average T_{eff} for each fertilization treatment. By applying nonlinear regression to these combined values one obtains the values shown in column 5 of Table 2 for the soil without K fertilization (139 d, 95% conf. limits 77–200 d), and in column 6 for the soil with K fertilization (156 d, 95% conf. limits 78–235 d). Using the same test as above that these two values are not significantly different (*P*>0.05). Considering the large 95% confidence limits, this result is not surprising.

3.2.1.4. Effect of K-fertilization of T_{eff}

There was no effect of K-fertilization on the effective half-lives using the same test (p>0.05 for T_{eff}.

3.2.1.5. Effect of K-fertilization on the transfer factors

To examine the effect of K-fertilization on the absolute values of the TF the ratio $TF_{without\ K}$ /$TF_{with\ K}$ was calculated for each 30 d sampling period of both years (see above) using the corresponding median TF of all plant parts. For the median of these ratios a value of $TF_{without\ K}$ /$TF_{with\ K}$ = 1.8 (95%-confidence limits 1.5–1.9) was obtained. Application of the Wilcoxon matched pair test shows that this difference is also statistically significant (*P*<0.05).

3.2.2. Dystric Fluvisol

The time-course of the ^{134}Cs TF soil-to-Swiss chard in the Dystric Fluvisol without using K fertilization (left) and applying K-fertilizer (right) is shown in Fig. 3.

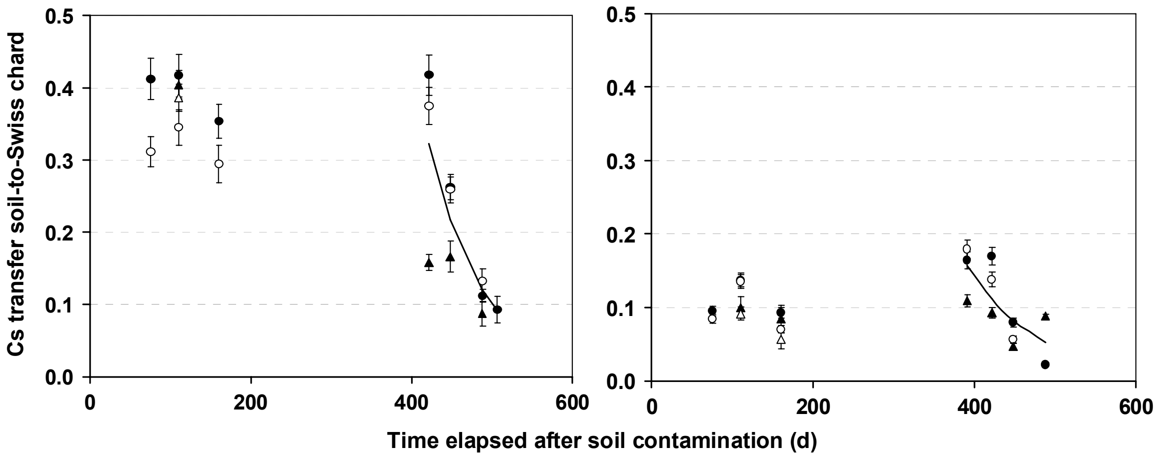

FIG. 3. Seasonal variation of radiocesium transfer factor soil-to-Swiss chard (● external leaves, ○ internal leaves, ▲ external stalks, Δ internal stalks) at the Dystric Fluvisol without K-fertilization (left) and with K-fertilizer (right).

3.2.2.1. Difference of the transfer factors between various plant parts

The same method as described above was used to test whether differences between the TF of the external and internal plant parts was present. Such a difference was, however, not detectable (*P*>0.05) neither for the leaves nor for the stalks. The number of available data pairs was in this case rather small (n ≤ 6) and it cannot be excluded that such a difference could be observable if more data were available.

3.2.2.2. Effective half-life of the transfer factors

Because of insufficient data for the first harvest period, regression analysis was applied only to the data from the second period. These results are shown also in Table 2. The effective half-life of the TF for this soil without K-fertilization was 47 d (95%-confidence limits 12–82 d) and with K-fertilization 61 d (95%-confidence limits 16–105 d).

3.2.2.3. Effect of K-fertilization on T_{eff}

There was no effect of K-fertilization on the effective half-lives of the transfer factor (P>0.05).

3.2.2.4. Effect of K-fertilization on the transfer factors

To examine the effect of K-fertilization on the absolute values of the TF the ratios $TF_{without\ K}$ / $TF_{with\ K}$ was calculated for each data pair from the same sampling date of the second harvest period using the corresponding median TF of all plant parts. The median of these ratios was $TF_{without\ K}$ / $TF_{with\ K}$ = 2.9 (95%-confidence limits 1.0–5.0). The Wilcoxon matched pair test shows that this difference is also statistically significant (P< 0.018).

3.2.3 Comparison between the transfer factors observed for both soil types

Because the sampling dates of the Swiss chard were quite different for the two soils, a direct comparison of the TF at a given time was not possible. For this reason the TF from both soils were calculated using the parameters A and T_{eff} for the Umbric Andosol (both harvest periods combined) and for the Dystric Fluvisol (second harvest period only) for various times (60, 90, 120, and 150 d) see Table 2. These results are given in Table 3. A comparison of these values shows that for each sampling date the TFs for the Dystric Fluvisol are always smaller than those for the Umbric Andosol. This is the case in the absence or presence of a K-fertilization. Without K-fertilization, this difference is, however, initially smaller and increases only with time to a factor of about two. In the presence of K-fertilization the TFs observed in both soils differ for all sampling dates by a factor of about two.

TABLE 3. TRANSFER FACTORS OF RADIOCAESIUM SOIL-TO-SWISS CHARD CALCULATED FROM REGRESSION ANALYSIS FOR VARIOUS DATES AFTER BEGINNING OF THE EXPERIMENT FOR BOTH SOILS WITH AND WITHOUT K-FERTILIZATION

Time (d)	Umbric Andosol with K	Dystric Fluvisol with K	Umbric Andosol no K	Dystric Fluvisol no K
60	0.185	0.106	0.351	0.303
90	0.162	0.075	0.302	0.194
120	0.142	0.053	0.26	0.125
150	0.124	0.038	0.223	0.080

Finally, it is evident from Table 2 that the effective half-life of the TF is considerably smaller for the plants grown on the Dystric Fluvisol as compared to the Umbric Andosol (factor of about three).

3.3. Seasonal variation of the ^{85}Sr transfer factor soil-to-Swiss chard

The data obtained for the ^{85}Sr TF soil-to-Swiss chard correspond with the first observation year after soil labelling.

3.3.1. Umbric Andosol

The time-course of the ^{85}Sr TF soil-to-Swiss chard in the Umbric Andosol during the first year after soil contamination is represented in Fig. 4 for the soil without Ca fertilizer (left) and using Ca fertilizer (right), respectively.

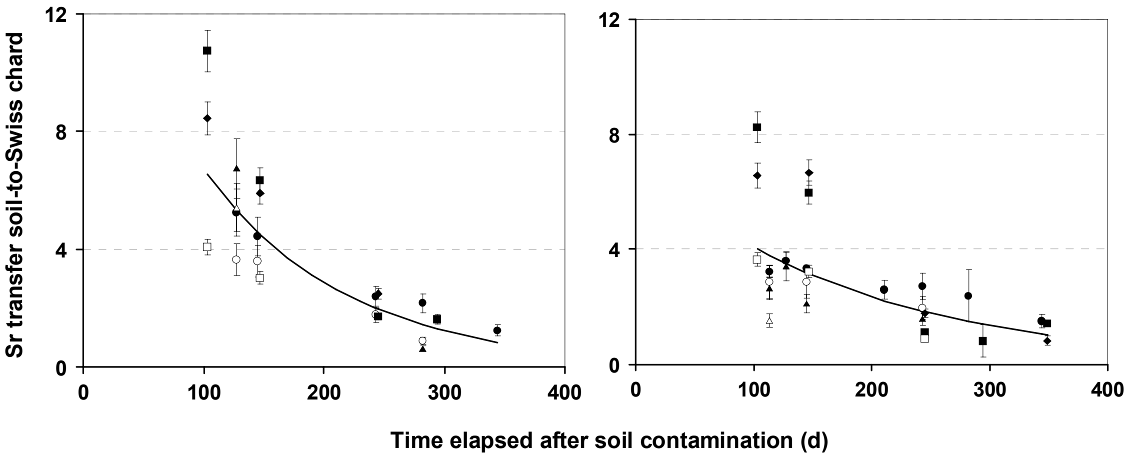

FIG. 4. Seasonal variation of the radiostrontium transfer factor soil-to-Swiss chard at the Umbric Andosol without Ca-fertilization (left) and with Ca-fertilizer (right) Year 2001: ● external leaves, ○ internal leaves, ▲ external stalks, Δ internal stalks Year 2002: ■ external leaves □ internal leaves, ♦ stalks.

As described for ^{134}Cs, the T_{eff} of the TF was evaluated, using the ^{85}Sr TFs to all edible plant parts (leaves and stalks) for each sampling date. The T_{eff} for the non-fertilized soil was about 81 d and for the Ca-fertilized one about 126 d. As shown in Fig. 4, by using a normal amount of Ca-fertilizer there was a very small reduction of the ^{85}Sr TF only at the beginning of the harvest period.

3.3.2. Dystric Fluvisol

Because of the brief plant growing period, and the reduced plant growth at the Dystric Fluvisol as a result of the flooding of this soil, only a small number of measurements of the TF were obtained during the two replicates of the experiment carried out in two sequential growing periods. The results are represented in Fig. 5.

The T_{eff} observed for the soil without using Ca-fertilization (about 82 d) is very similar to the T_{eff} obtained under similar treatment for the Umbric Andosol. The number of data pairs available for the soil treated with Ca-fertilizer (n=4) was to low for estimation of the corresponding T_{eff}.

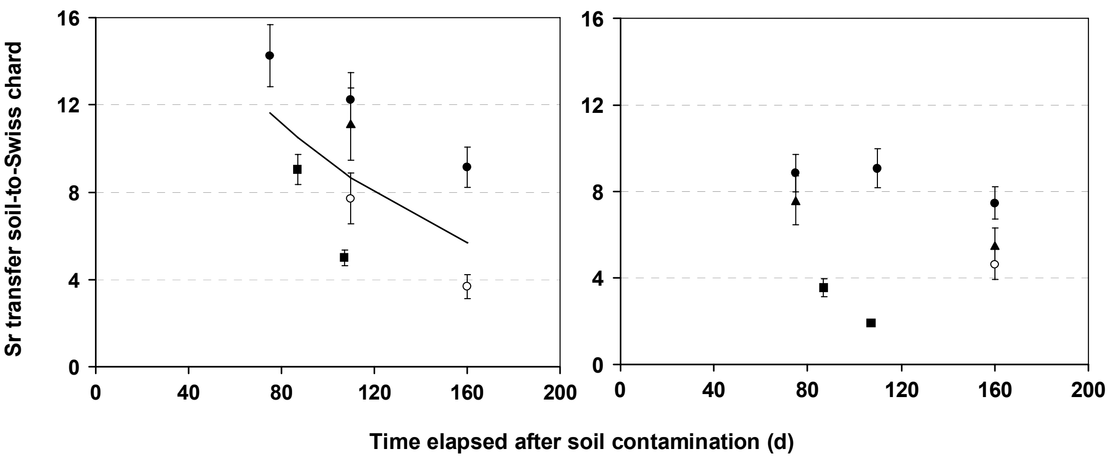

FIG.5. Seasonal variation of the radiostrontium transfer factor soil-to-Swiss chard at the Dystric Fluvisol without Ca-fertilization (left) and with Ca-fertilizer (right) Year 2001: ● external leaves, ○ internal leaves, ▲ stalks. Year 2002: ■ leaves and stalks.

3.4. Time-course of the radiocaesium and –strontium transfer factor to Swiss chard.

The results discussed above show that for the two soils the radiocaesium and –strontium concentration in Swiss chard decreased considerably during each growing season. Many perennial plants translocate mineral nutrients from their aboveground parts to belowground storage organs before they sprout in spring [12]. An additional factor in the seasonal decrease could be the growth of some chard roots below the contaminated soil layer observed at the end of the growing periods. The seasonal behaviour of the Swiss chard TF must be considered in mathematical models based on this parameter to estimate other radioecological parameters of public concern (radioecological sensitivity of an ecosystem, dose affecting population and others).

No decrease of the absolute values of the radiocaesium TF to Swiss chard was observed during the two consecutive observation years after soil contamination, when compared in corresponding seasonal periods, for soils without and with K-fertilizer. Following local management practices, the soil was manually inverted at the beginning of each growing period. This procedure could have contributed to a more homogeneous distribution of the radionuclide within the 0–0.20 m soil layer and in this way to provide a better access for the available radionuclide by the roots during the second growing period. According to Nisbet and Woodman [9] the age of contamination in the soil does not need to be taken into account when deciding best estimates of TF for radiocaesium or strontium for arable systems in the medium term after an accident. Nevertheless, there are other authors reporting an effect of age of contamination on the TF [13, 14, 15].

3.5. Radiocaesium transfer soil-to-sweet corn

There was no decrease of the TF of ^{134}Cs maize grains (sweet corn), for either the unfertilized or for the K-fertilized soil during the three sequential harvest periods after soil contamination (2002–2003) (Fig. 6). Therefore, the median value of the ^{134}Cs TF and the corresponding 95%-confidence limits were reported (Table 4).

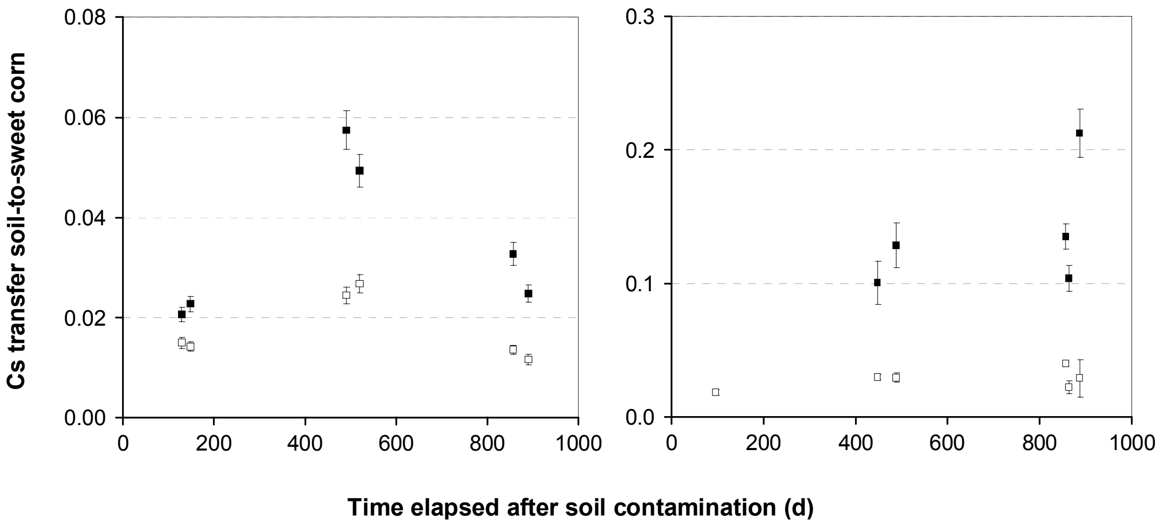

FIG. 6. Time-course of the radiocaesium transfer factor soil-to-sweet corn grains, during three harvest periods after soil contamination, at the Umbric Andosol (left) and Dystric Fluvisol (right), without K-fertilization (■) and with K-fertilization (□).

The ^{134}Cs TF represent a median of the values measured during three consecutive years after soil labelling. The ^{85}Sr TF represent the median of the TF determined in the first growing period after soil contamination.

TABLE 4. RADIOCAESIUM AND -STRONTIUM TRANSFER SOIL-TO-SWEET CORN AT THE UMBRIC ANDOSOL AND DYSTRIC FLUVISOL

Soil type	Radio nuclide	Fertilization	Median TF	95 % con. lim.	n	TF no fert/. TF with fert	95 % con. lim	Recommended value	
								Ref [13]	Ref [9]
Umbric Andosol	^{134}Cs	No K	0.029	0.021–0.057	6	2.0	1.4–2.4		
Umbric Andosol	^{134}Cs	K	0.015	0.012–0.027	6			0.01	0.14
Dystric Fluvisol	^{134}Cs	No K	0.13	0.10–0.21	5	4.3	3.3–7.3		
Dystric Fluvisol	^{134}Cs	K	0.029	0.019–0.040	6			0.01	0.14
Umbric Andosol	^{85}Sr	No Ca	0.027	0.025–0.056	4	0.8	0.6–1.1	0.12	0.15
Umbric Andosol	^{85}Sr	Ca	0.040	0.028–0.053	4				
Dystric Fluvisol	^{85}Sr	No Ca	0.035	0.027–0.044	2	1.0	0.8–1.1	0.12	0.15
Dystric Fluvisol	^{85}Sr	Ca	0.038	0.023–0.054	2				

N.B. The ^{134}Cs TFs are median values of the three consecutive years after soil contamination; the ^{85}Sr TFs are the median values for the first growing period after contamination.

To examine the effect of the K-fertilization on the absolute value of the TF, the ratio $TF_{without\ K}/TF_{with\ K}$ was calculated for each sweet corn harvest. For the median of these ratios a value of $TF_{without\ K}/TF_{with\ K}$ = 2.0 (95%-confidence limits 1.4–2.4) was obtained in the Umbric Andosol and $TF_{without\ K}/TF_{with\ K}$ = 4.3 (95%-confidence limits 3.3–7.3) at the Dystric Fluvisol. As a result of a normal K-fertilization the radiocaesium transfer from soil-to-sweet corn was lowered by a factor of 2 at the Umbric Andosol and by a factor of about 4 in the Dystric Fluvisol. This soil had a lower exchangeable K content than the Umbric Andosol before K-fertilization.

The median values of the radiocaesium TF sweet corn observed at the Umbric Andosol and Dystric Fluvisol treated with normal amounts of K-fertilizer are within the 95% confidence limits of the recommended transfer values to cereals [9, 13] (Table 4).

3.6. Radiostrontium transfer soil-to-sweet corn

The median ^{85}Sr TFs to sweet corn obtained at the Umbric Andosol and Dystric Fluvisol are also summarized on Table 4. The results correspond to the first harvest period after soil labelling and were obtained during two replicates of the experiment carried out in two successive years (2001 and 2002). For both soils the radiostrontium TF to sweet corn is lower than the recommended values reported for loam soils, but included within the lower limit of the 95%-confidence [9, 13].

As described above, the effect of the Ca fertilization on the absolute value of the TF was examined calculating the ratio $TF_{without\ Ca}/TF_{with\ Ca}$ for each maize harvest. The median of the values is $TF_{without\ Ca}/TF_{with\ Ca}$ = 0.8 (95%-confidence limits 0.6–1.1) in the Umbric Andosol, and $TF_{without\ Ca}/TF_{with\ Ca}$ = 1.0 (95%-confidence limits 0.8–1.1) in the Dystric Fluvisol. These results indicate that a usual fertilization of 100 kg Ca ha^{-1} did not affect radiostrontium TF to sweet corn.

3.7. Radiocaesium transfer soil-to-cabbage

The ^{134}Cs TF soil-to-cabbage at the Umbric Andosol and Dystric Fluvisol was studied in the third year after soil contamination. The results in Table 5 correspond with the transfer to the internal leaves of the cabbage plants.

To examine the effect of the K-fertilization on the absolute value of the ^{134}Cs TF, the ratio $TF_{without\ K}/TF_{with\ K}$ was calculated. The ratios were 5.7 and 3.7 for the Umbric Andosol and Dystric Fluvisol, respectively. These ratios show that K- fertilization reduced the radiocaesium transfer to cabbage in these soils.

TABLE 5. RADIOCAESIUM AND -STRONTIUM TRANSFER SOIL-TO-CABBAGE AT THE UMBRIC ANDOSOL AND DYSTRIC FLUVISOL

Soil type	Radio nuclide	Fertilization	Median TF	95 % con. lim.	n	TF no fert/. TF with fert	$TF_{cabbage}/TF_{maize\ grain}$	Recommended value	
								Ref [13]	Ref [9]
Umbric Andosol	^{134}Cs	No K	0.86	0.41–1.31	2	5.7	29.7		
Umbric Andosol	^{134}Cs	K	0.15	0.15–0.22	3		11.5	0.18	0.028
Dystric Fluvisol	^{134}Cs	No K	0.71	0.65–0.77	2	3.7	5.1		
Dystric Fluvisol	^{134}Cs	K	0.19	0.16–0.23	2		6.3	0.18	0.028
Umbric Andosol	^{85}Sr	No Ca	3.1		1	0.7			
Umbric Andosol	^{85}Sr	Ca	4.5		1			2.7	2.2
Dystric Fluvisol	^{85}Sr	No Ca	2.3		1	1.6			
Dystric Fluvisol	^{85}Sr	Ca	1.4		1			2.7	2.2

N.B. The ^{134}Cs TFs were measured during the third year after soil labelling (field experiment); the ^{85}Sr TFs were measured during the first year after soil contamination (pot experiment).

For both soils, the median values of the TFs in the soils treated with normal amounts of K-fertilizer are similar to the TF value recommended for green vegetables for loam soils [13]. The lower 95%-confidence limit of the median value is higher than the upper limit of the 95%-confidence interval reported for brassicae by Nisbet and Woodman [9] in loam soils.

The ratio between the radiocaesium TF to cabbage and the TF to sweet corn grains as reference cereal was calculated for the third harvest period after soil contamination. The ratios $TF_{cabbage}$ /TF_{maize} in Umbric Andosol are about 2–3 times higher than the ratios obtained for the Dystric Fluvisol (Table 5). For both soils treated with K-fertilizer, the ratios are within the range of ratios (3–11) reported by Frissel et al. [10].

3.8. Radiostrontium transfer soil-to-cabbage

The ^{85}Sr TF to cabbage (internal leaves) at the Umbric Andosol and Dystric Fluvisol was studied in pot experiments during the first harvest period after soil contamination. The results are summarized in Table 5. Similarly as described for the transfer to maize, there was no effect of Ca fertilization on the radiostrontium TF ($TF_{without\ Ca}$ /$TF_{with\ Ca}$ = 0.7 at the Umbric Andosol and $TF_{without\ Ca}$ /$TF_{with\ Ca}$ = 1.6 at the Dystric Fluvisol).

The TF values soil-to-cabbage obtained in pot experiments are similar to the strontium TF value for brassicas in loam soil [9] and also similar to the TF to green vegetables [13]. The ratio $TF_{cabbage}$ /TF_{maize} was not calculated for radiostrontium, because the measured TF to cabbage and maize were not comparable, since they were determined in pot and field experiments, respectively.

4. CONCLUSIONS

In both soils the radiocaesium and –strontium TF to Swiss chard (perennial plants) decreased considerably (by a factor of about 3–4 for radiocaesium and of about 4–8 for radiostrontium) during each growing period. This seasonal decrease of the TF to perennial plants has to be considered in mathematical models used to estimate other radioecological parameters of public concern.

The sequential ^{134}Cs TF to chard values measured during the first two consecutive harvest periods after soil contamination showed no significant differences during the course of the harvest periods. This implies there was no significant decrease (ageing effect) of the ^{134}Cs TF to Swiss chard during the two years.

The results for chard consider mainly the transfer to the plants by root uptake. A deposition of the radionuclide on the plant leaves and subsequent foliar absorption was minimized by sealing the surface of the contaminated soil with an uncontaminated 2 cm soil layer. The possibility that contaminated soil can be deposited on green vegetable leaves by rain-splash under the high precipitation rates of the agricultural Lake Region of Chile requires study.

As for chard, there was no decrease of the radiocaesium TF to sweet corn grain during the three harvest periods after soil contamination.

Normal K-fertilization decreased the radiocaesium TF to Swiss chard, sweet corn and cabbage in both soils. There was no significant effect of Ca fertilization on ^{85}Sr TFs.

Under normal K-fertilization, most of the median values of the radiocaesium TF obtained are within the 95%-confidence intervals of the recommended TF values reported [9, 13] for loam soils. Nevertheless, the lower 95%-confidence limit of the radiocaesium TF to cabbage was higher than the upper 95%-confidence limits of the value recommended for brassicas in loam soils [9].

Similarly, the median values of the ^{85}Sr TF are within the 95%-confidence intervals of TF values recommended for radiostrontium in loam soils. The median radiostrontium TF to sweet corn in both soils are lower than the recommended values but included within the lower limit of the 95%-confidence interval reported for loam soils [9, 13].

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A FIELD STUDY OF SOIL-TO-PLANT TRANSFER OF STRONTIUM-90 AND CAESIUM-137 BASED ON A CALCARIC CAMBISOL IN SUB-TROPICAL SOUTHWEST CHINA

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Abstract

Soil-to-plant transfer of ^{90}Sr and ^{137}Cs in field conditions was measured in Sichuan Province in 1999, 2000, 2001 and 2002. The soil involved is a Calcaric Cambisol that was contaminated artificially with ^{90}Sr and ^{137}Cs in 1999. The plants were wheat, maize, Chinese cabbage, broad bean, spinach, potato, radish, tomato and lettuce. Each TF (soil-to-plant transfer factor) was determined from five replicates of crop and related soil samples taken at harvest time. Soil characteristics are analyzed and other parameters important to the experiments are provided.

1. INTRODUCTION

Root uptake is one of the most important pathways of radionuclide transfer from a contaminated environment to the terrestrial food chain. For this pathway the transfer factor (TF), which is defined as "concentration of radionuclide per unit weight of plant organ" divided by "concentration of radionuclide per unit weight of dry soil", is necessary for dose assessment models. In China assessment generally uses TF values derived from American or European countries [1]. In 1994, IAEA published the Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Temperate Environments [2]. The data are mostly drawn from North America and Europe, much of which was compiled through projects of IUR and EU. However, there are only limited data for crops cultivated in Asian countries, especially in China. Since TF changes with factors such as soil properties, crop species, and climatic characteristics, it is necessary to obtain TF data on radionuclides for major agriculture crops grown in typical soils in different regions or countries. In this study we investigated the transfer of ^{90}Sr (half-life: 28.78 y) and ^{137}Cs (half-life: 30.07 y) from a Calcaric Cambisol to wheat, maize, broad bean, spinach, Chinese cabbage, potato, radish, tomato and lettuce.

2. MATERIALS AND METHODS

2.1. Experimental field

The field is located in the suburbs of Guangyuan city, Sichuan Province (southwest China where the climate is sub-tropical and humid). The soil (80 m^2) was contaminated by adding a solution containing 9.25 MBq ^{90}Sr (as $Sr(NO_3)_2$) and 9.25 MBq ^{137}Cs (as CsCl) in 1999 for this study. Both ^{90}Sr and ^{137}Cs are important products of nuclear facilities.

2.2. Cropping

To obtain TF data on a range of crops, we planted maize, potato, spinach, wheat, radish, Chinese cabbage, tomato, lettuce and broad bean in 1999, 2000, and 2001. In 2002 only wheat, maize, tomato, Chinese cabbage and lettuce were planted. Plot sizes were 20 m^2 for wheat, 18 m^2 for tomato and 14 m^2 for the others. No crop was planted in the same plot in consecutive years. The plants were irrigated with tap water to supplement natural rainfall as necessary. The soil was fertilized with carbamide. The planting and sampling times of crops are listed in Table 1.

TABLE 1. PLANTING AND SAMPLING SCHEDULE OF CROPS

Crop	Planting or seeding	Sampling	Experimental period (days)
Maize	Apr. 8, 1999	Aug. 2, 1999	117
Maize	Apr. 20, 2001	Aug. 28, 2001	129
Maize	May 12, 2002	Sep. 4, 2002	115
Potato	Apr. 1, 1999	Jun. 21, 1999	81
Spinach	Aug. 20, 1999	Nov. 12, 1999	84
Wheat	Oct. 29, 1999	May 17, 2000	201
Wheat	Oct. 25, 2002	May 15, 2003	202
Chinese cabbage	Sep. 18, 2000	Nov. 20, 2000	62
Chinese cabbage	Oct. 11, 2001	Jan. 11, 2002	92
Chinese cabbage	Sep. 26, 2002	Nov. 28, 2002	63
Radish	Sep., 10, 1999	Jan. 24, 2000	136
Tomato	April 15, 2000	July 12, 2000	88
Tomato	May 10, 2001	Sep. 13, 2001	126
Tomato	May 22, 2002	Sep. 4, 2002	105
Lettuce	Nov.21, 2000	April 14, 2001	144
Lettuce	Oct. 5, 2002	April 4, 2003	181
Broad bean	Oct.12, 2001	May 8, 2002	208

2.3. Analytical methods

Analysis of ^{90}Sr involved oxalate precipitation, di(2-ethylhexyl)orthophosphoric acid (HDEHP) chromatography and beta counting. ^{137}Cs was measured by direct non-destructive gamma-spectrometry. Brief descriptions of the methods are listed in Table 2.

TABLE 2. RADIONUCLIDE ANALYTICAL METHODS FOR SOIL AND PLANT SAMPLES

Nuclides	Samples	Brief description of methods	Detection limit
^{90}Sr	Plants	Oxalate pptn., HDEHP chromatography, β counting	8.0E-4 Bq/g (ash)
	Soil	Leaching with HCl, cation exchange, β counting	0.01 Bq/kg dry soil
^{137}Cs	Plant	non-destructive γ spectrometry (Model S-95)	0.01 Bq/kg dry plant*
	Soil	non-destructive γ spectrometry (Model S-95)	0.01 Bq/kg dry soil*

N.B. Detection limit varies with measuring time. The measuring efficiency of the γ spectrometry was 35%.

2.4. Sampling procedures

At harvest time, plant and soil samples were collected simultaneously. Soil samples were randomly taken. Only the edible parts of the crops were sampled. Potato, spinach, Chinese cabbage and radish were washed to remove soil contamination. Maize and wheat grains were not washed. For each TF measurement five replicate crop and soil samples were taken.

For ^{90}Sr analysis, plant samples were dried at 105°C, carbonized in an electric stove, then ashed in a muffle furnace at 450 °C ready for analysis. The corresponding soil samples were air-dried for 7 days and then passed through a 2 mm sieve, ready for analysis.

For ^{137}Cs plant samples were dried at 105 °C and then ground into powder for analysis. Soil samples were treated as for ^{90}Sr.

3. RESULTS AND DISCUSSION

TABLE 3. SOIL CHARACTERISTICS AND OTHER PARAMETERS OF THE EXPERIMENT

Parameters	Unit	1999*	2000*	2001*	2002
pH(water)		7.4	7.5	7.5	7.5
Exch K^+	Cmol/kg	25.9*	17.9*	20.5*	16.0*
Exch Ca^{2+}	Cmol/kg	8.0*	7.8*	10.0*	7.0*
CEC	Cmol/kg	41.1*	23.3*	37.2*	27.5*
Org. carbon	%	4.3	5.7	6.9	3.6
Rooting depth	cm	18	18	18	18
Bulk density	(kg/m^3)	1650	1650	1650	1650
Crop yield					
Maize	(kg/m^2)	0.33		0.53	0.62
Potato	(kg/m^2)	0.47			
Spinach	(kg/m^2)	0.43			
Chinese cabbage	(kg/m^2)		0.3	2.85	9.28
Wheat	(kg/m^2)		0.25		0.25
Radish	(kg/m^2)		1.4		
Tomato	(kg/m^2)		0.5	0.63	0.27
Lettuce	(kg/m^2)			0.65	1.81
Broad bean	(kg/m^2)			0.52	
Moisture content of crop					
Maize	%	54.5		57.4	44.5
Potato	%	79.8			
Spinach	%	94.2			
Chinese cabbage	%	—	92.1		95.4
Wheat	%	—	—		—
Radish	%	—	92.4		
Tomato	%	—	92.9	95.4	94.7
Lettuce	%			93.7	96.1
Broad bean	%				

Soil properties, pH (in water), organic carbon content, exchangeable K in soil (cmol(+)/kg), exchangeable Ca in soil (cmol(+)/kg) and CEC were determined yearly by the methods described in [3]. Other parameters related to the experiments were measured, including moisture content of crop before drying, rooting depth (estimate, cm), bulk density at sampling times (kg/m^3), crop yield (kg/m^2). These soil characteristics and the parameters concerned are given in Table 3.

The concentrations of ^{90}Sr and ^{137}Cs in soil and crop samples are given in Tables 4 and 5.

TABLE 4. THE CONCENTRATION OF ^{90}Sr AND ^{137}Cs IN SOIL SAMPLES (Bq/kg dry soil)

	Crop	N	^{90}Sr	^{137}Cs
1999				
	Maize	5	5.50E+02±1.21E+02	6.16E+02±2.59E+02
	Potato	5	6.59E+02±1.09E+02	3.91E+02±4.79E+01
	Spinach	5	7.50E+02±1.50E+02	5.33E+02±2.75E+02
2000				
	Wheat	5	4.52E+02±1.19E+02	4.92E+02±1.70E+02
	Chinese Cabbage	5	4.03E+02±1.46E+02	5.41E+02±2.77E+02
	Tomato	5	5.50E+02±1.16E+02	3.86E+02±1.45E+02
	Radish	5	5.10E+02±1.28E+02	4.21E+02±9.73E+01
2001				
	Lettuce	5	3.91E+02±2.08E+02	7.86E+02±3.93E+02
	Tomato	5	3.58+E02±6.11E+01	4.95E+02±2.06E+01
	Maize	5	5.58E+02±1.67E+02	7.20E+02±2.42E+02
	Chinese cabbage	5	5.01E+02±1.24E+02	3.98E+02±1.20E+02
	Broad bean	5	4.33E+02±1.24E+02	3.75E+02±8.92E+01
2002				
	Maize	5	2.98E+02±1.13E+02	3.45E+02±1.33E+02
	Tomato	5	3.36E+02±4.67E+01	2.76E+02±4.02E+01
	Chinese	5	3.50E+02±7.21E+01	2.76E+02±8.59E+01
	Wheat	5	2.75E+02± 9 .73E+01	3.99E+02±7.07E+01
	Lettuce	5	1.68E+02± 4 .04E+01	3.27E+02±4.96E+01

N.B. The data are the average±SD.

TABLE 5. CONCENTRATION OF RADIONUCLIDE IN EDIBLE PARTS OF CROPS*
(Bq/kg dry crop)

Crop		N	^{90}Sr	^{37}Cs
1999				
	Maize	5	1.59E+00±6.01E-01	1.16E+00±6.57E-01
	Potato	5	4.97E+00±7.31E-01	8.80E- 01±1.41E-01
	Spinach	5	1.84E+01±1.09E+0	6.06E+01±3.86E+01
2000				
	Wheat	5	2.91E+00±1.08E+00	5.23E-01±2.12E-01
	Chinese cabbage	5	5.69E+02±2.02E+02	1.67E+01±8.34E+00
	Tomato	5	2.17E+01±1.13E+01	8.88E+00±7.90E-01
	Radish	5	3.88E+02±6.07E+01	3.80E+00±1.07E+00
2001				
	Lettuce	5	1.65E+01±9.95E+00	1.74E+01±8.93E+00
	Tomato	5	9.58E+00±3.24E+00	1.21E+01±1.10E+00
	Maize	5	7.05E-01±1.73E-01	1.25E+00±8.19E-02
	Chinese cabbage	5	4.13E+02±6.73E+01	6.56E+00±2.35E+00
	Broad bean	5	4.40E+00±1.39E+00	5.50E+00±1.89E+00
2002				
	Maize	5	2.47E-01±2.16E-02	1.29E+00±5.72E-02
	Wheat	5	3.00E+00±4.94E-01	3.55E-01±2.83E-03
	Tomato	5	7.25E+00±6.31E-01	4.14E+00±4.26E-01
	Chinese cabbage	5	2.86E+02±6.15E+01	2.35E+01±6.63E+00
	Lettuce	5	1.10E+01±2.61E+00	6.29E+00±1.90E+00

* See note of Table 4.

TF values of ^{90}Sr, ^{137}Cs for experimental crops (average and standard deviation) are given in Table 6.

TABLE 6. FIELD TFs OF ^{90}Sr AND ^{137}Cs IN 1999, 2000, 2001 AND 2002*
(Bq/kg dry crop)/(Bq/kg dry soil)

	Crops	N	^{90}Sr	N	^{137}Cs
1999					
	Maize	5	2.97E-03±1.31E-03	5	2.27E-03±1.79E-03
	Potato	5	7.64E-03±1.11E-03	5	2.25E-03±2.56E-04
	Spinach	5	2.34E-02±1.22E-02	5	6.74E-03±2.72E-03
2000					
	Wheat	5	6.50E-03±2.28E-03	5	1.18E-03±4.48E-04
	Chinese cabbage	5	1.47E+00±4.04E-01	5	3.90E-02±2.74E-02
	Radish	5	7.80E-01±1.12E-01	5	8.93E-03±5.75E-04
	Tomato	5	4.26E-02±2.00E-02	5	2.61E-02±1.73E-02
2001					
	Lettuce	5	5.32E-02±5.05E-02	5	2.30E-02±5.50E-03
	Tomato	5	2.79E-02±1.19E-02	5	2.45E-02±2.63E-03
	Maize	5	1.29E-03±1.76E-04	5	1.97E-03±8.81E-04
	Chinese cabbage	5	8.57E-01±2.36E-01	5	1.79E-02±9.04E-03
	broad bean	5	1.10E-02±5.02E-03	5	1.49E-02±4.74E-03
2002					
	Maize	5	9.08E-04±2.64E-04	5	4.16E-03±1.39E-03
	Wheat	5	1.17E-02±3.81E-03	5	9.17E-04±2.68E-04
	Tomato	5	2.21E-02±4.78E-03	5	1.53E-02±2.92E-03
	Chinese cabbage	5	8.69E-01±3.21E-01	5	9.27E-02±3.98E-02
	Lettuce	5	6.65E-02±1.24E-02	5	1.91E-02±4.19E-03

For ^{90}Sr, Chinese cabbage and radish (only one harvest year) have the highest TFs while maize has the lowest TF. For ^{137}Cs, maize, wheat, potato and spinach have the lowest TFs.

As mentioned above, maize, Chinese cabbage and tomato were grown for three harvest years. The comparison of ^{90}Sr and ^{137}Cs TF data for these crops in different years are shown in Figs 1–6. Figure 1 shows that uptake of ^{90}Sr by maize decreased yearly whereas uptake of ^{137}Cs by maize increased in 2002 (Fig. 2).

Figure 3 shows TF of ^{90}Sr for Chinese cabbage in 2000 is higher than those for 2001 and 2002 while Fig. 4 shows that ^{137}Cs TF for Chinese cabbage is highest in 2002. We noticed that the yields of Chinese cabbage increased during the years of the experiment so the higher TFs might be differences in plant development.
There are individual samples in the first year with relatively high TF values (see Figs 1, 2, 5 and 6), that might indicate higher variation in the first year of the study but Figs 1–6 also show that TFs for the same type of crop and the same radionuclide in different harvest years are generally of the same order. Mean values of ^{90}Sr and ^{137}Cs TF are presented in Tables 7 and 8.

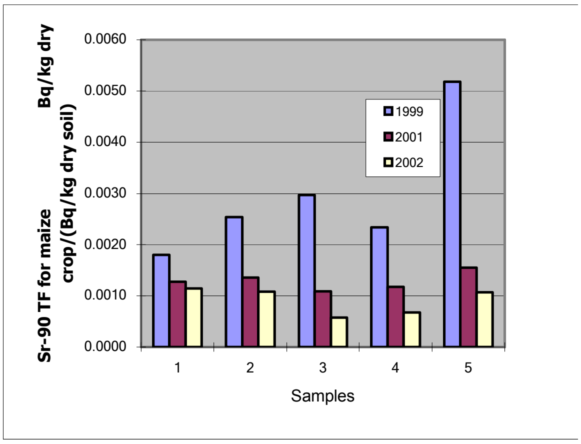

FIG.1. Comparison of Sr-90 TFs for maize in 1999, 2001 and 2002.

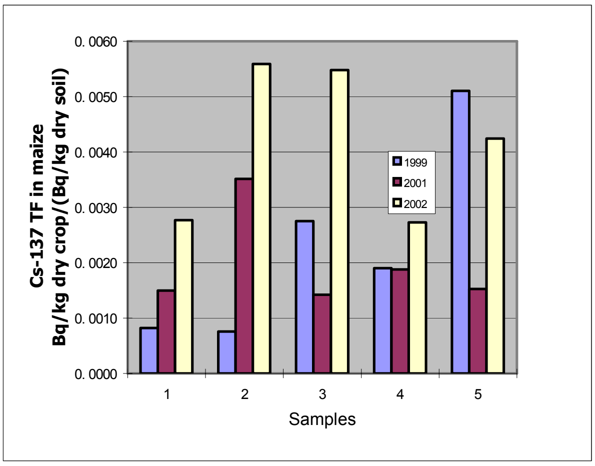

FIG. 2. Comparison of Cs-137 TFs for maize in 1999, 2001, 2002.

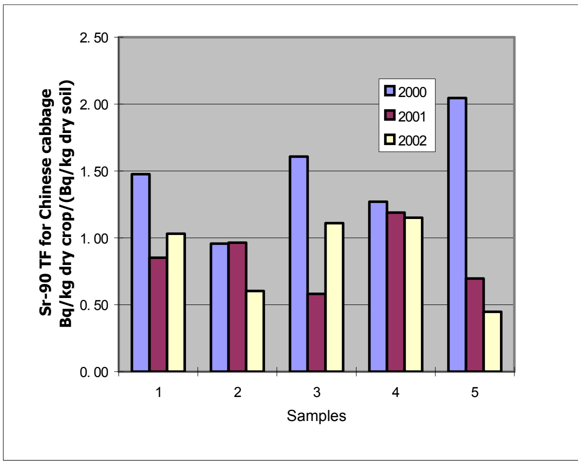

FIG. 3. Comparison of Sr-90 TFs for Chinese Cabbage in 2000, 2001 and 2002.

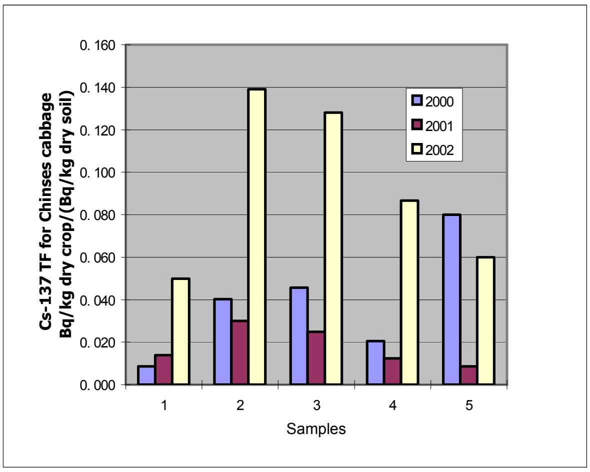

FIG. 4. Comparison of Cs-137 TFs for Chinese cabbage in 2000, 2001 and 2002.

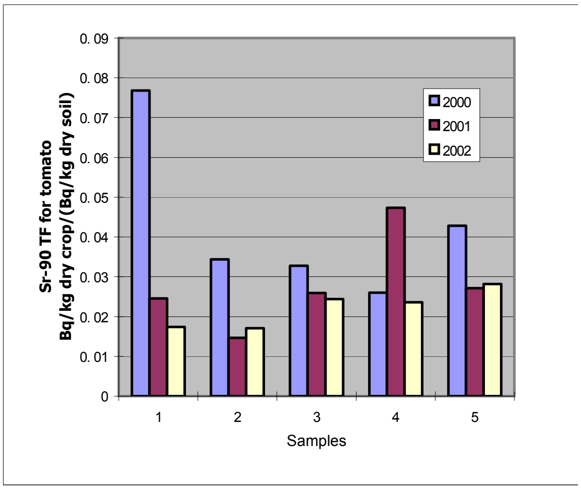

FIG. 5. Comparison of Sr-90 TFs for tomato in 2000, 2001 and 2002.

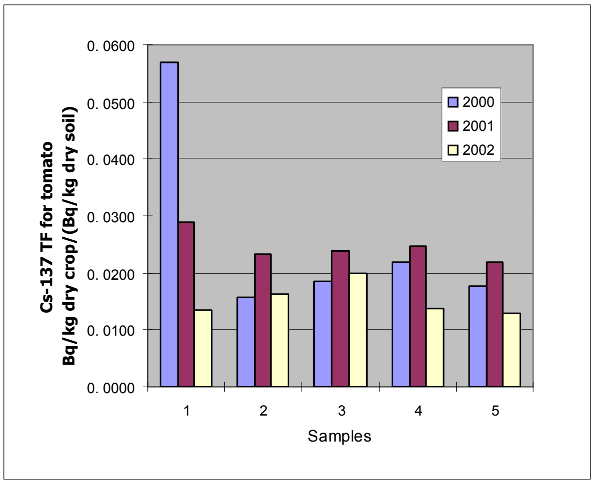

FIG. 6. Comparison of Cs-137 TFs for tomato in 2000, 2001 and 2002.

TABLE 7. TFs OF ^{90}Sr (Bq/kg dry crop)/(Bq/kg dry soil)

Crops	N	Geo. mean	Average±SD	Range	
Wheat	10	1.05E-02	1.35E-02±1.30E-02	5.04E-03	4.89E-02
Maize	15	3.14E-03	5.58E-03±5.53E-03	5.72E-04	1.55E-02
Broad bean	5	1.00E-02	1.10E-02±5.02E-03	5.02E-03	1.64E-02
Spinach	5	3.37E-01	4.16E-01±2.94E-01	1.47E-01	8.52E-01
Chinese cabbage	15	9.88E-01	1.07E+00±4.24E-01	4.48E-01	2.04E+00
Potato	5	7.57E-02	7.64E-03±1.11E-03	6.13E-03	8.77E-03
Radish	5	7.73E-01	7.80E-01±1.12E-01	6.17E-01	9.01E-01
Tomato	15	2.82E-02	3.09E-02±1.55E-02	1.46E-02	7.68E-02
Lettuce	10	5.03E-02	5.99E-02±3.52E-02	1.49E-02	1.38E-01

TABLE 8. TFs OF ^{137}Cs (Bq/kg dry crop)/(Bq/kg dry soil)

Crops	N	Geo mean	Average±SD	Range	
Wheat	10	1.00E-03	1.05E-03±3.68E-04	6.19E-04	1.89E-03
Maize	15	2.34E-03	2.83E-03±1.64E-03	7.55E-04	5.59E-03
Broad bean	5	1.39E-02	1.49E-02±5.12E-03	9.34E-03	2.14E-02
Spinach	5	1.06E-01	1.24E-01±5.49E-02	2.96E-02	1.71E-01
Chinese cabbage	15	3.49E-02	4.99E-02±4.18E-02	8.61E-03	1.39E-01
Potato	5	2.24E-03	2.25E-03±2.54E-04	1.91E-03	2.53E-03
Radish	5	8.92E-03	8.93E-03±5.75E-04	8.19E-03	9.60E-03
Tomato	15	2.03E-02	2.20E-02±1.07E-02	1.30E-02	5.69E-02
Lettuce	10	2.05E-02	2.11E-02±5.06E-03	1.47E-02	2.97E-02

^{90}Sr TFs for maize, wheat, broad bean and potato are generally lower in than those of Chinese cabbage, radish and lettuce. ^{137}Cs TFs for wheat, maize and potato are usually lower than those of vegetables. Chinese cabbage showed the highest ability to accumulate ^{137}Cs.

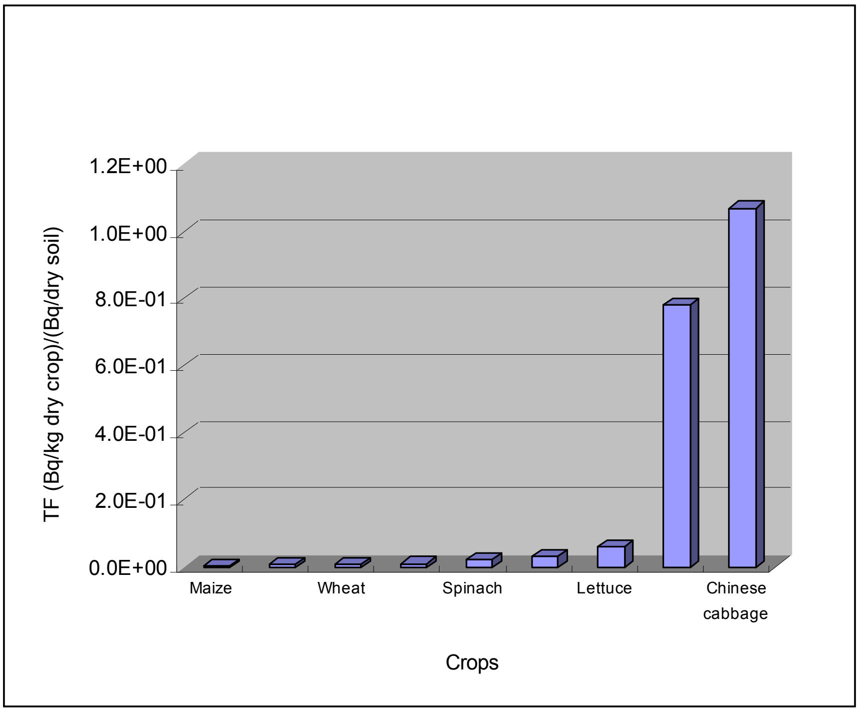

FIG. 7. Soil to plant TF of 90Sr for crops in Calcaric Cambisol in China.

The ranges of ^{90}Sr and ^{137}Cs mean TF values for different crops are shown in Figs 7 and 8.

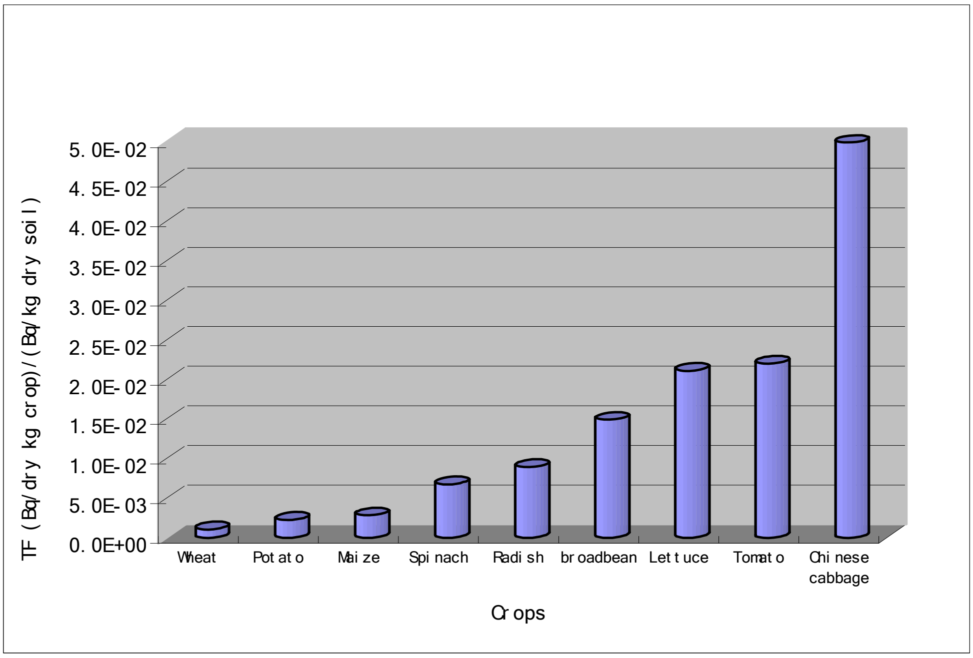

FIG. 8. Soil to plant TF of 137Cs for Crops in Calcaric Cambisol in China.

4. CONCLUSION

The variability of the ^{90}Sr and ^{137}Cs TFs is rather wide. The year-to-year variations suggest climatic factors may be important. For both radionuclides, the TFs to cereals were close to, or below, the bottom of the range predicted on the basis of IUR data and the previous CRP. There was no clear trend for TF to change with time for either ^{90}Sr or ^{137}Cs.

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TIME DEPENDENT CAESIUM-134 TRANSFER FACTORS FOR CROPS GROWN ON DIFFERENT GREEK SOIL TYPES

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Abstract

Corn and cabbage plants were grown in a greenhouse pot experiment in two volcanic and two 'representative' Greek soils artificially contaminated with ^{134}Cs for a three year period. Results for the 3rd year of experimentation as well as the main findings for the whole period are presented in this paper. In the third year of the project, two sweet corn (*Elite* and *Vilmorin*) and two cabbage varieties, (*Kozanitiko* and *Brunswick*) were tested. These varieties, except *Vilmorin*, were also grown on the same soils the previous years. ^{134}Cs transfer factors (TFs) were significantly higher in the two volcanic soils than in the other soils, in agreement with previous years' data. Irrespective of crop species or variety, the clay-calcareous soil continued to show the lowest TF values. Comparing ^{134}Cs uptake by the two corn varieties, it was found that *Vilmorin* plants absorbed approximately two times more ^{134}Cs than *Elite* plants. No such difference was observed for the cabbage varieties. ^{134}Cs TFs for both seeds and vegetative part of *Elite* variety significantly decreased compared to the respective values of the previous year. A similar trend, rarely significant, was observed for the two cabbage varieties. The ratio TF cabbage/TF corn (seeds) in the four soils increased this year and ranged from 6–16 with a mean value of 9. This increase is due to the higher reduction of corn ^{134}Cs TFs than that of cabbage TFs. For all studied varieties and in all possible combinations the mean value of the ratio TF cabbage/TF corn was 8, coinciding with the respective values of the previous two years (7 and 6 respectively). During the three years of experimentation, three different corn varieties were tested and mean reference TFs for corn seeds in the studied soils were calculated.

1. THE OBJECTIVE

The main objective of the current Coordinated Research Programme is the classification of soil ecosystems on the basis of specified radionuclides, specified soil types and reference crops. A classification of soil ecosystems might be a way to reduce uncertainties due to the enormous range of uptake parameters. Our contribution is to produce data on transfer factors of ^{134}Cs from soil-to-reference plants in a range of Greek soil systems to characterize systems in which TFs might differ substantially from what would be regarded as normal.

The purpose of the present work was to study ^{134}Cs uptake by corn and cabbage plants grown on two "representative" and two volcanic Greek soils during three years. Conversion factors for cabbage using corn as a reference crop were calculated. Based on corn TFs in the different soils reference TFs for all corn varieties studied are presented.

2. MATERIALS AND METHODS

2.1. Soils

The soils were carefully selected to have soil systems in which transfer factors might differ substantially from what would be regarded as normal. Four different soil types were selected; two from southeastern Greece (Peloponese) and two volcanic from the island of Santorini. The main characteristics of the soils are presented in Table 1.

TABLE 1. MAIN SOIL CHARACTERISTICS

Soil	Clay (%)	Silt (%)	Sand (%)	Texture	pH (1:1)	O.M. (%)	CEC $cmol_c/kg$	Exch. K^+ $cmol_c/kg$
1	8.6	18.0	73.4	SL	5.6	0.46	7.39	0.23
2	29.3	47.0	23.7	CL	7.4	2.11	17.39	0.42
3	8.6	27.2	64.2	SL	5.8	0.56	3.48	0.39
4	7.0	18.0	75.0	LS-SL	6.9	2.71	9.44	1.06

Soil 1 is classified as Luvisol. It is an acid soil of coarse–medium texture at a high level of development. Soil 2 is a medium to heavy textured calcareous Fluvisol, in its early stages of development, representing a high percentage of the Greek agricultural soils. Soils 3 and 4 are volcanic ash soils, from Santorini island, developed on pumice and volcanic ash and are classified as Andosols. The genesis of these soils is determined mainly from their age (approximately 1500 years, when a very strong volcanic eruption took place), the climate of the island, the mineral composition and the texture of the volcanic ash.

As discussed in Ref. [1], the soils in Santorini are characterized by their low clay content and most of them are neutral to alkaline. In the surface soil layers heavier particles are present e.g., gravels of aluminum-iron composition or pumice. The presence of gravels can be explained by the intense wind erosion, where the strong winds take away the fine particles of the soil surface. Gravels also prevent the loss of soil moisture by evaporation, which is crucial for plant growth considering the land on the island is not irrigated. The average rainfall in the island is low (average 350 mm/year).

The two soils from Santorini are of low clay content, but differ in other soil properties, such as the pH, the organic matter content, the cation exchange capacity and the concentration of exchangeable potassium (Table 1).

2.2. Experimental conditions

The plants were grown in pots (4 replicates) containing 14 kg air-dried soil. The size of pots is considered sufficient to provide reliable data and to prevent water shortage and nutrient deficiency problems.

2.3. Soil Contamination

Soil was contaminated with ^{134}Cs (1.9 MBq pot^{-1}) as CsCl on July 28, 1999. The soil was transferred to each pot in seven layers of approximately 2 kg of soil each. On the top of each layer 100 ml of the radioactive solution was added in the form of very small drops. This technique of soil contamination has been used successfully in previous experimentation with annual and tree crops (see Refs [2] and [3]). The distribution of the radioactive material through out the pot is well controlled by this method and loss via cracks or edges is absent. The soil in pots was moistened to field capacity and left to stand for two months for the ^{134}Cs to reach equilibrium.

2.4. Plant growth

Corn was selected as a reference crop followed each year by cabbage to check the constant ratio, which was supposed to exist between the TFs of the two plants in the different soils.

2.4.1. Plant varieties

The following varieties were grown:
2000–2001 — corn var. Corduna and cabbage var. local-Kozanitiko;
2001–2002 — corn var. Corduna, sweet corn var. Elite, cabbage var. local-Kozanitiko and var. Brunswick;
2002–2003 — sweet corn var. Elite and sweet corn var. Vilmorin, cabbage, var. local-Kozanitiko and var. Brunswick.
For the last year of the experiment, the corn variety Corduna was replaced by the sweet corn variety Vilmorin, because of its poor growth on some soils.

In the second and the third years, in addition to NH_4NO_3, 5 g of K_2SO_4 were added in each pot.

2.5. Analytical methods

Cation exchange capacity of the soils was determined by the Na-acetate method [4]. Organic matter content was determined by the Walkley-Black procedure [4]. Mechanical composition was determined by the Bouyoukos hydrometer method [4] and the pH by glass and calomel electrodes in soil-water ratio of 1:1. Exchangeable bases were extracted by 1 M ammonium acetate.

After harvesting, plants were separated into edible and vegetative parts, where necessary (corn); representative plant samples were cut into small pieces, dried at 70°C and counted for ^{134}Cs with an HpGe detector (efficiency 22%) connected to a CANBERRA 35^{+} 4K multichannel analyzer plus a computer with software for gamma-ray spectroscopy analysis. The concentration of ^{134}Cs in plant samples was expressed in Bq kg^{-1}.

3. RESULTS AND DISCUSSION

3.1. Transfer factors

Radiocaesium soil-to-plant transfer factors (TFs) are presented for corn (Table 2) and cabbage (Table 3). Significant differences of ^{134}Cs TFs in the four soils are illustrated in Figs 1, 2 and 3. Though mean ^{134}Cs TFs were considerably different in the four soils in some cases these differences were not significant due to the high variation of the values.

In 2003, ^{134}Cs TFs for corn and cabbage varieties were significantly higher in the volcanic soils (soils 3 and 4). The light texture and the presence of pumice that enhances water retention, may explain the increased availability of ^{134}Cs in the volcanic soils. The significantly lower ^{134}Cs TF were observed again in soil 2, which is a calcareous soil with high clay content and high CEC. These factors are known to more effectively 'fix' radiocaesium compared to other soils (see Refs [5–7] and Ref. [2]). The 2003 data support the observation that despite the plant species or variety, soil-to-plant radiocaesium transfer is higher in the two volcanic soils (soils 3 and 4) and lower in the heavy textured soil 2 (see Refs [8] and [9]).

TABLE 2. TRANSFER FACTORS OF ^{134}Cs (Bq kg^{-1} DW PLANT/Bq kg^{-1} DW SOIL) FOR CORN GROWN ON THE FOUR SOILS

	Elite		Vilmorin	
Soil	Grain	Vegetative	Grain	Vegetative
1	0.009	0.036	0.012	0.038
	(0.004)*	(0.011)	(0.004)	(0.017)
2	0.002	0.010	0.001	0.007
	(0.001)	(0.002)		(0.002)
3	0.026	0.110	0.074	0.213
	(0.007)	(0.020)	(0.009)	(0.039)
4	0.017	0.085	0.038	0.117
	(0.005)	(0.023)	(0.003)	(0.052)
Mean	0.014	0.060	0.031	0.094
	(0.004)	(0.014)	(0.005)	(0.028)

* Standard deviation is given in the parenthesis.

TABLE 3. TRANSFER FACTORS OF ^{134}Cs (Bq kg^{-1} DW PLANT/Bq kg^{-1} DW SOIL) FOR CABBAGE GROWN ON THE FOUR SOILS

Soil	Kozanitiko	Brunswick
1	0.050	0.114
	(0.017)	(0.039)
2	0.012	0.008
	(0.003)	(0.004)
3	0.431	0.308
	(0.163)	(0.042)
4	0.120	0.173
	(0.016)	(0.010)
Mean	0.153	0.151
	(0.050)	(0.024)

* Standard deviation is given in the parenthesis.

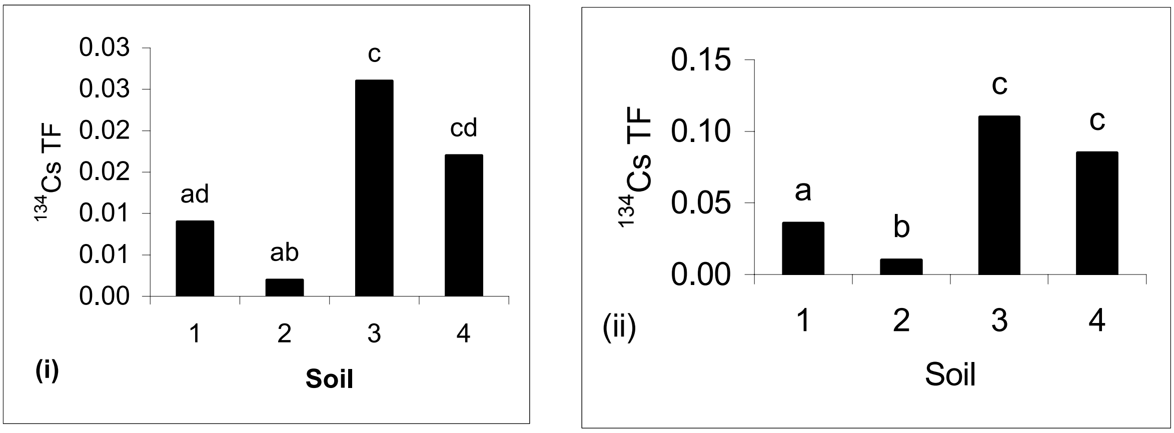

FIG. 1. ^{134}Cs TFs for sweet corn variety elite grown on the four soils
(i) grain, (ii) vegetative (t-test, same letter no significant difference, n=4).

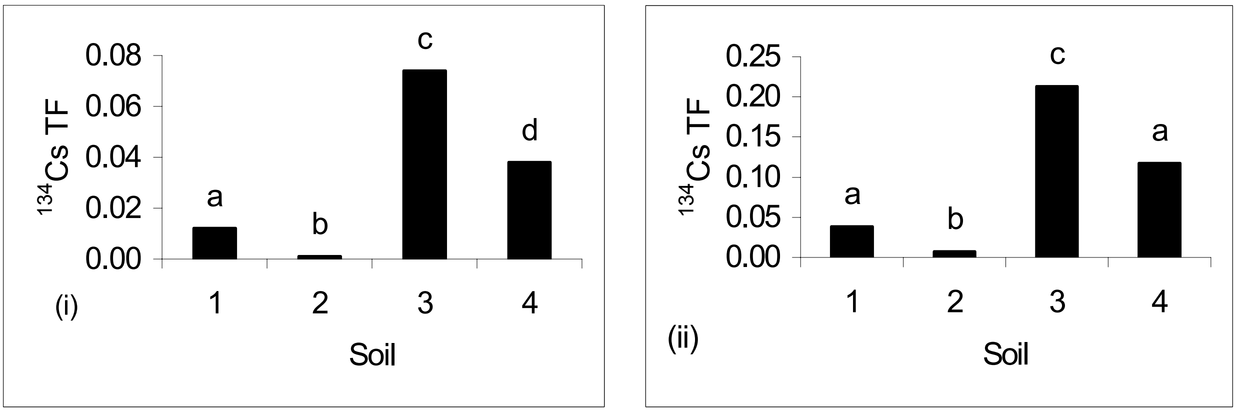

FIG. 2. ^{134}Cs TFs for sweet corn variety Vilmorin grown on the four soils
(i) grain, (ii) vegetative (t-test, same letter signifies no significant difference, n=4).

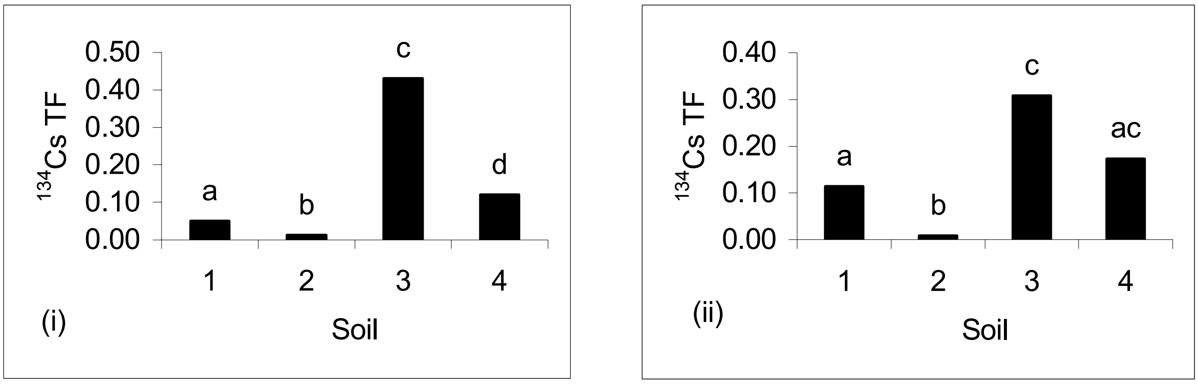

FIG. 3. ^{134}Cs TFs for cabbage plants grown on the four soils
(i) Kozanitiko, (ii) Brunswick (t-test, same letter signifies no significant difference, n=4).

Comparing the varieties of the same plant species, ^{134}Cs TFs for the grain and the vegetative part of the sweet corn variety Vilmorin were higher, not always significantly, than the corresponding values of sweet corn variety Elite (Fig. 4). As indicated by the mean ^{134}Cs TF values the plants of Vilmorin variety absorbed approximately two times more ^{134}Cs than the plants of Elite variety (Table 2).

Considering the two cabbage varieties, in most cases no significant difference of ^{134}Cs TFs was observed (Fig. 5), though in the previous year ^{134}Cs concentration in plants of Brunswick variety was significantly higher than in plants of Kozanitiko variety [9].

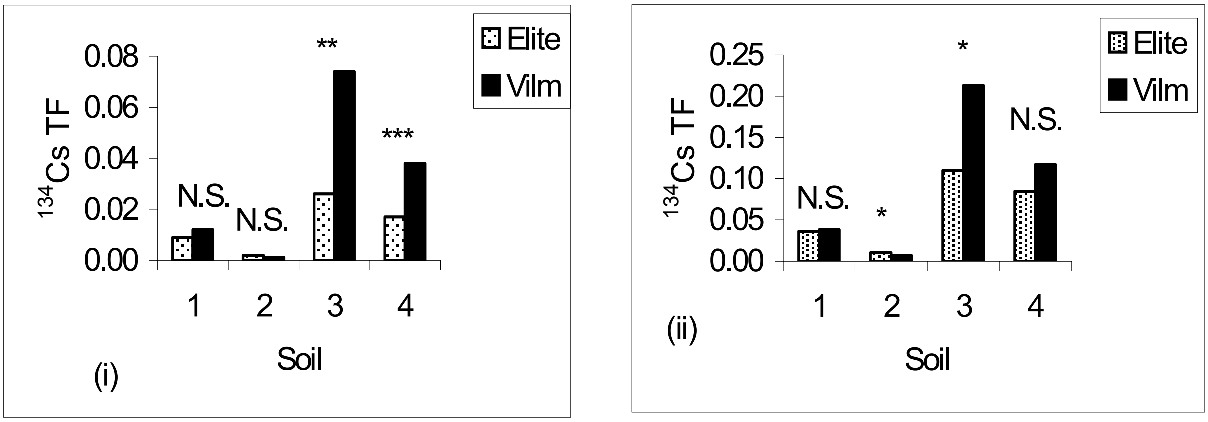

FIG. 4. Comparison of ^{134}Cs TFs for sweet corn varieties Elite and Vilmorin
*(i) grain, (ii) vegetative (t-test, *=P<0.05, **=P<0.01, ***=P<0.001,*
N.S.= no significant difference, N=4).

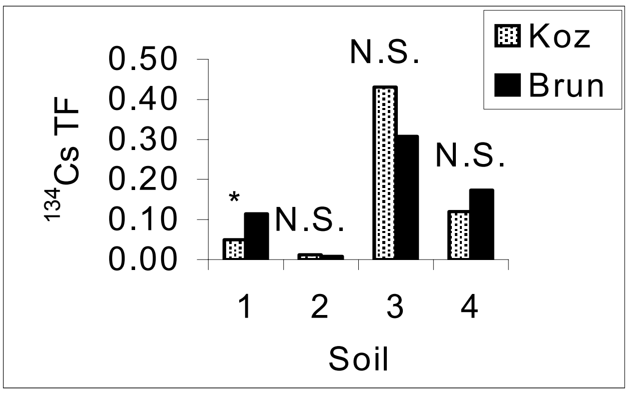

*FIG. 5. Comparison of ^{134}Cs TFs for cabbage varieties Kozanitiko and Brunswick (t-test, *=P<0.05, N.S.= no significant difference, N=4).*

In all soils, ^{134}Cs TFs of corn plants variety Elite significantly decreased in the second growth period (2002), by a factor of 3–4, both in the grain and the vegetative part (Fig. 6). A similar trend was observed for the cabbage varieties in most cases (Fig. 7). This agrees with Ref [10], that reported transfer of caesium to edible parts of several plants decreased by a factor of four over a seven year period. Similar data are also reported in Ref. [11]. On the other hand, Refs [7] and [12] observed no significant change or a consistent trend in radiocaesium plant uptake with time.

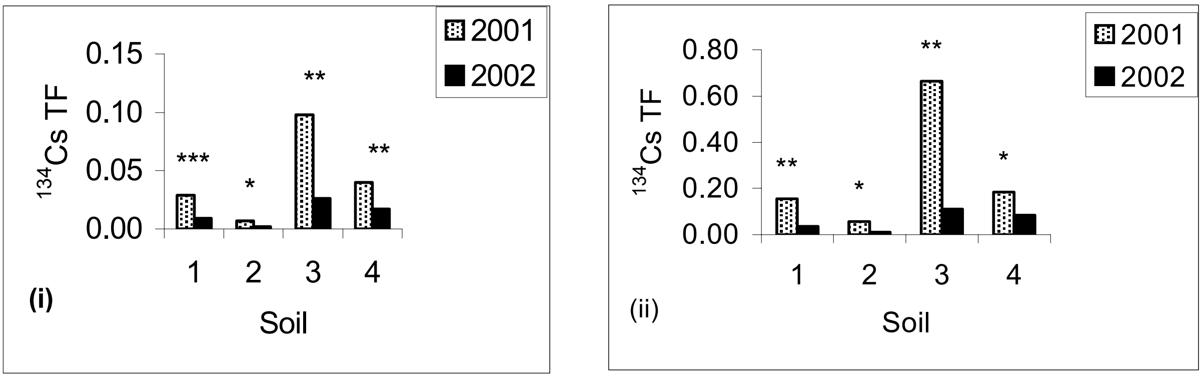

*FIG. 6. ^{134}Cs TFs of sweet corn, variety Elite, grown on the four soils for the years 2001 and 2002 (i) seeds, (ii) vegetative (t-test, *=P<0.05, **=P<0.01, ***=P<0.001, N=4).*

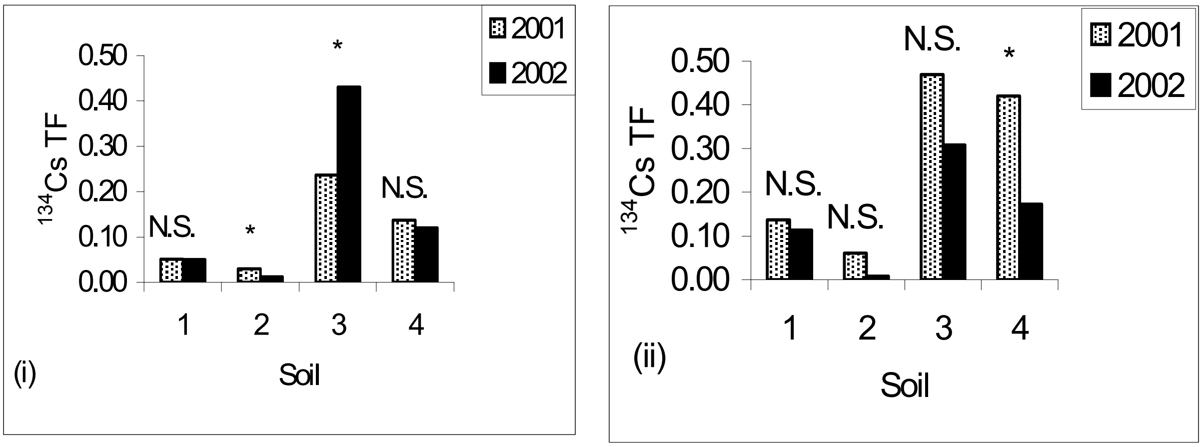

*FIG. 7.^{134}Cs TFs for cabbage grown on the four soils for the years 2001 and 2002 (i) Kozanitiko, (ii) Brunswick (t-test, *=P<0.05, N.S.= no significant difference, N=4).*

3.2. Conversion factors and reference TFs

In Ref. [13] a method is described correlating radiocaesium uptake as a function of the soil type. It is based on observations that most individual species of a crop group show almost the same soil-to-plant uptake factors on the same soil and that between crop groups uptake factors appear more or less constant. A reference TF is derived for a reference crop depending solely on soil properties. Crops are divided into groups, depending on the characteristics of the group. Cereals serve as the reference group. Thus TFs of other groups can be calculated by multiplying values for cereals with a conversion factor.

Using this method conversion factors for cabbage (var. Kozanitiko) were calculated for the years 2000–2002, using corn (var. Corduna) as a reference crop [9]. In 2003 Vilmorin replaced Corduna, so conversion factors for cabbage were calculated using sweet corn variety Elite as the reference crop (Tables 4 and 5). The calculated ratio ^{134}Cs TF cabbage / ^{134}Cs TF corn (grain) in the four soils increased in the last growth period and ranged from 6–16 (2–4 in the previous year) with a mean of 9

(3 in the previous year), though no significant difference was observed (Table 4). Such an increase was expected, because in 2002 ^{134}Cs TFs for corn grain were lower than before while ^{134}Cs TFs for cabbage remained in most soils more or less constant (Figs 6i,7i).

TABLE 4. CALCULATED RATIOS ^{134}Cs TF CABBAGE (KOZANITIKO) ^{134}Cs TF CORN SEEDS (ELITE)

Soil	1	2	3	4	Mean
Year 2001	2	4	2	3	3
Year 2002	6	7	16	7	9

TABLE 5: CALCULATED CONVERSION FACTORS FOR THE YEAR 2002

	Corn	Cabbage
Cs conversion factor	1	9
Range		6–16

The TF cabbage / TF corn ratios for all varieties and in all possible combinations gave mean values ranging from 3–16 with a mean value of 8 (N=16). This value is very close to those of the previous two years (mean values 7 and 6, respectively) and it coincides with the generic values reported in Ref. [13] for cereals and leafy green vegetables.

The soils were classified according to reference TFs of corn grain for the three varieties used over the three years of experimentation (Table 6).

TABLE 6. REFERENCE ^{134}Cs TFs FOR CORN (GRAIN)

	Corduna		Elite		Vilmorin	
Soil type	1st year	2nd year	2nd year	3rd year	3rd year	Mean
Fluvisol Clay-Calcareous	0.005	0.004	0.007	0.002	0.001	0.004 (0.002)
Luvisol Sand-Acid	0.015	0.015	0.029	0.009	0.012	0.016 (0.008)
Andosol Volcanic	0.100	0.066	0.069	0.022	0.056	0.063 (0.028)
Mean	0.048 (0.048)	0.028 (0.033)	0.035 (0.031)	0.011 (0.010)	0.023 (0.029)	

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THE CLASSIFICATION OF INDIAN SOILS ON THE BASIS OF TRANSFER FACTORS OF RADIONUCLIDES FROM SOIL TO REFERENCE PLANTS

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Abstract

The soil-to-plant transfer factors (TF) of ^{137}Cs, ^{134}Cs and ^{90}Sr were determined for wheat, maize, rice and cabbage plant under irrigated conditions in the greenhouse and in the field. Two greenhouse experiments were conducted with three soils collected from different agro-regions of India and the field experiment was conducted at the protected field of the institute. In both pot culture and field conditions, transfer factor values for ^{137}Cs to cabbage were higher than those for wheat and maize grown under identical conditions of crop growth and soil contamination. TF values were similar in both experiments. There was little effect of contamination level on the transfer factor of ^{137}Cs in cabbage, wheat or maize. The TF values for ^{90}Sr are 50–100 times higher than those for ^{137}Cs or ^{134}Cs in all the crops studied. The TF values of ^{137}Cs in rice or wheat straw were in general higher compared with grain. The values did not differ greatly between the three different Inceptisol soils used. There was a slightly higher value of TF for ^{137}Cs to both rice grain and straw from the soil where normally rice is grown during the monsoon season which has higher clay content compared to other two soils.

1. INTRODUCTION

Nuclear power is a major source of electricity in more than thirty one nations and in India there are ten operating nuclear reactors producing about 1600 MW of power. For radiological assessments for nuclear facilities, we need to evaluate the long term effects on human health by the radionuclides released to the environment. The terrestrial system is of primary importance in evaluating the internal dose by ingestion in India because most of our foods come from terrestrial sources. Radionuclides deposited on the soil can be ingested by humans through the food chain but presently very little information is available for Indian conditions to predict the precise internal dose. The radionuclides are deposited on the surface soils through atmospheric transport of nuclear explosion fallout products and those present in wastes of nuclear fuel processing facilities or nuclear power plants. It is important to understand those processes that control the accumulation and mobility of radionuclides in soils as these directly influence their absorption by plants [1]. The understanding of the reactivities of the soil constituents and mobilizing agents towards these radionuclides is essential to develop techniques to treat contaminated soils, thus minimizing radionuclide entry into the food chain.

Adsorption processes fix nearly the whole amount of nuclide added to the soil, thus reducing its availability at least temporarily to growing plants. Studies have shown that both soil clays and organic matter can retain ionic caesium. Illite has been found to be a more effective sorbent than montmorillonite, kaolinite or vermiculite [2]. In both terrestrial and aquatic ecosystems, strong binding of ^{137}Cs to soils and sediments reduces its mobility and consequently its assimilation by biota. In general, Cs is not a very mobile element in the soil system, but certain soil conditions can enhance the mobility and biological availability of Cs. Caesium behaves similar to potassium and is easily absorbed from soil solution by plant roots, fungi and other living systems and is efficiently absorbed in the digestive system of mammals [3]. The major limiting factor for its bioavailability is the removal from solution by adsorption. It is therefore, essential to measure or predict accurately the degree of adsorption of caesium on soils or other solids [4].

In general assessments, the amount of radionuclide transferred from soil to plant is calculated with a transfer factor (TF) which is the ratio of concentration in plant edible parts to that in soil. A study has been conducted to assess the transfer of ^{137}Cs or ^{134}Cs and ^{90}Sr from soil to cereals, wheat or maize and to a leafy plant, cabbage, in field and in pot culture conditions in different soils. Also, the effect of potassium on transfer factors of ^{134}Cs and calcium on that of ^{90}Sr was evaluated. A second pot experiment was conducted to determine the transfer factor of ^{137}Cs in rice in three Inceptisol soils.

2. EXPERIMENTAL DETAILS

2.1. Transfer factor of ^{134}Cs and ^{90}Sr in cabbage, wheat and maize as influenced by potassium or calcium application (pot experiment)

The protocols finalized in the first Research Coordination Meeting of the project held at Izmir, Turkey, (April 12–16, 1999) were followed to measure transfer parameters of ^{134}Cs and ^{90}Sr to arable crops from different soils. Transfer factors for reference crops of cabbage and wheat and maize as affected by varying levels of potassium or calcium application, respectively, were measured in a greenhouse pot culture experiment. Glazed pots 30 cm high and 20 cm diameter were filled with 8 kg of the different soils. The physicochemical properties of the soils are presented in Table 1.

The radionuclides studied were ^{134}Cs and ^{90}Sr. Soils were contaminated with the radionuclides at the rate of 37 kBq kg^{-1} soil. A premix of the soil with each radionuclide was prepared by adding the required amount of the radionuclide as 10 mL solution to 500 g of the air-dried soil. This premix was brought to 50 per cent of the water saturation value in a 600 mL glass beaker and mixed thoroughly. The premix was air-dried, ground and then mixed uniformly with the remaining quantity of the processed soil for each pot. The pots were filled with contaminated soil and water was added to bring the soil moisture content to 50% of the water saturation value. The pots were left to air-dry for equilibration of radionuclides in soil. After drying, the soil in the pots was mixed and again sufficient water was added to bring the soil moisture content to about 50% of the water saturation value. The equilibration period for which the contaminated soils in the pots were subjected to drying and wetting cycles was ten weeks.

After the equilibration period, three composite core samples of soil from each pot were collected for checking the homogeneity of radionuclide distribution. The soil samples drawn from each pot to a depth of 20 cm were air-dried, processed and ground to pass a 100 mesh in a porcelain pestle mortar.

TABLE 1. SELECTED CHARACTERISTICS OF SOIL OF POT AND FIELD EXPERIMENTAL SITES

Soil type	Red	Alluvial	Black
Soil order	Oxisol	Inceptisol	Vertisol
Location from where collected	Dharwar	New Delhi	Indore
pH (H_2O)	6.1	7.8	7.3
pH ($CaCl_2$)	6.0	7.1	7.0
Organic C (%)	0.38	0.48	1.05
CEC [cmol (p^+) kg^{-1}]	8.66	8.30	46.01
Exchangeable K [cmol (p^+) kg^{-1}]	0.30	0.33	0.44
Exchangeable Ca [cmol (p^+) kg^{-1}]	5.23	6.35	20.26
Sand (%)	76.6	81.2	8.0
Silt (%)	6.0	8.6	41.0
Clay	17.4	10.2	51.0
Texture	Sandy-loam	Sandy-loam	Clay loam

The first crop taken in pots was wheat. In total, there were 54 pots with three soils, two radionuclides, three levels of potassium or calcium each and three replications. The recommended doses of N and P were applied to wheat as urea and single superphosphate. Potassium was applied at 0, 75 and 150 kg K_2O as potassium chloride to the ^{134}Cs contaminated pots and calcium was applied at the 0, 75 and 150 kg CaO as calcium chloride to the ^{90}Sr contaminated pots. The wheat crop was grown under irrigated conditions in the winter season (*Rabi* season 1999–2000).

At maturity, the wheat crop was harvested and separated into grain and straw and these were ground in a centrifugal mill. Care was taken to avoid cross contamination of samples by grinding a large amount of uncontaminated grain or straw between the samples. Also soil samples were taken from the pots by mixing the whole soil and withdrawing a representative sample. The soil samples were air-dried and were ground in pestle and mortar.

The protocols developed by IUR and IAEA were used for determining the transfer factors and the methods outlined in the IAEA technical report *Measurement of Radionuclides in Food and Environment - A Guidebook* [5] were used for radioassay. The ^{134}Cs activity was measured directly by counting in a well type NaI(Tl) scintillation detector connected to Canberra Accuspec 8K multichannel analyzer. For ^{90}Sr activity measurement in plant and soil samples radiochemical separation was carried out and the activity was counted in a Geiger Mueller counter.

In the next monsoon season (*Kharif* 2000) cabbage was planted in the same pots in July 2000 with the application of recommended doses of N and P and varying levels of K or Ca as in the previous season. Cabbage was harvested in October 2000 and the plant sample was dried in an oven. After harvesting the cabbage, soils were sampled and the activity of ^{134}Cs and ^{90}Sr measured in both cabbage and soil samples. During the subsequent five seasons, cabbage was sown in the same pots:
(1) cabbage—winter *Rabi* season (2000–01); (2) cabbage—monsoon *Kharif* season (2001); (3) cabbage—winter *Rabi* season (2001–02); (4) cabbage—monsoon *Kharif* season (2002); (5) cabbage—winter *Rabi* season (2002–03). In each of the seasons the effect of varying levels of potassium on ^{137}Cs and varying levels of calcium on ^{90}Sr transfer factor were determined.

2.2. Field Experiment

A field experiment was conducted with ^{137}Cs and ^{90}Sr in plots that were contaminated in 1996 and had been used earlier for similar studies. Separate microplots were freshly contaminated in September 1999 at 18.5 kBq kg^{-1} soil and 37 kBq kg^{-1} soil in the upper 5 cm soil layer. In both cases the experiment was laid-out with 24 plots 3 m x 3 m (9 m^2) each containing a microplot of 1 m x 1 m (1 m^2) for radionuclide application. There were 12 microplots each for ^{137}Cs and ^{90}Sr. For application of radionuclides, the top five cm soil from the microplots was dug-out and allowed to air-dry on a polythene sheet. One litre of solution of radionuclide containing the required application level was sprayed onto 75–80 kg air-dry soil and the soil was mixed thoroughly taking care that no lumps were formed. The contaminated soil was air-dried and replaced in the microplots. After leveling, the microplots were irrigated to bring the soil moisture content in upper 10 cm soil layer to about 50% of the water saturation value. The soil in microplots was allowed to equilibrate for eight weeks with radionuclides. During this period, moisture was kept at about 50% water saturation by frequent addition of water. After ten weeks of equilibration, soil samples were taken from four spots in the microplots to a depth of 20 cm. ^{137}Cs was measured as before.

In the plots contaminated with ^{137}Cs and ^{90}Sr in 1996 a cabbage crop was taken in the winter (*Rabi*) season (1998-99), followed by maize in the monsoon (*Kharif*) season (1999). In both old and freshly contaminated plots during the subsequent seven seasons cabbage and wheat / maize crop was planted in three replications.

The harvested wheat and maize crops from field experiments were separated into grain and straw / stover and ground. Also, soil samples collected from microplots from 0–20 cm depth were analyzed for ^{137}Cs as before. The soil samples drawn from the field experiment as well as from the different soils used for the greenhouse experiment were processed and analyzed for pH (H_2O), pH ($CaCl_2$), electrical conductivity, exchangeable K, Ca etc.

2.3. Transfer Factor of ^{137}Cs in rice (pot experiment):

Glazed pots 30 cm high and 20 cm diameter were filled with 8 kg of three different soils (Inceptisols) collected from the Indian Agricultural Research Institute farm. The physicochemical properties of the soils are presented in Table 2. The soils were artificially contaminated with ^{137}Cs at 37 kBq kg^{-1} soil. A premix of the soil with radionuclide was prepared by adding the required amount in 10 ml solution to 500 g of the air-dried soil. This premix was brought to 50% of the water saturation value in a 500 ml polypropylene beaker and mixed thoroughly. The premix was air-dried, ground and then mixed uniformly with the remaining quantity of the processed soil for each pot. The pots were filled with contaminated soil and water was added to bring the soil moisture content to 50% of the water saturation value. The soils were contaminated in May 2001.

The pots were left to air-dry then the soil was mixed again and sufficient water was added to bring the soil moisture content to about 50% of the water saturation value. The contaminated soils in the pots were subjected to drying and wetting cycles for ten weeks. After the equilibration period, three composite core samples of soil from each pot were collected for checking the homogeneity of radionuclide distribution. The soil samples drawn from each pot to a depth of 20 cm were air-dried, processed and ground to 100 mesh in a porcelain pestle mortar.

The first rice crop was grown from July to November 2001. The rice crop was grown as transplanted and under flooded conditions in the monsoon season (*Kharif* season 2001). In total, there were 24 pots with three soils in eight replications. The recommended doses of N, P and K were applied to rice crop as urea, single superphosphate and potassium chloride.

TABLE 2. SELECTED CHARACTERISTICS OF SOILS USED IN THE POT EXPERIMENT WITH RICE

	1	2	3
Soil type	Alluvial	Alluvial	Alluvial
Soil order	Inceptisol	Inceptisol	Inceptisol
Location from where collected	IARI MB8C	IARI 1ABC	IARI MB14C
pH (H_2O)	7.8	8.0	8.1
pH ($CaCl_2$)	7.1	7.5	7.7
Organic C (%)	0.43	0.47	0.45
CEC [cmol (p^+) kg^{-1}]	9.14	7.53	9.34
Exchangeable K [cmol (p^+) kg^{-1}]	0.25	0.33	0.31
Exchangeable Ca [cmol (p^+) kg^{-1}]	6.84	5.86	6.38
Sand (%)	76.4	49.9	46.0
Silt (%)	11.8	37.3	33.0
Clay	11.8	12.8	21.0
Texture	Sandy-loam	Silty-loam	Clay loam

At maturity, the rice crop was harvested and separated into grain and straw and these were ground in a centrifugal mill. Care was taken to avoid cross contamination of samples by grinding a large amount of uncontaminated grain or straw between the samples. Also soil samples were taken from the pots by mixing the whole soil and withdrawing a representative sample. The soil samples were air-dried and were ground in pestle and mortar. ^{137}Cs activity was measured directly in both plant and soil samples.

In the next winter season (*Rabi* 2001–02) wheat was planted in December 2001 in the same pots with application of recommended doses of N, P and K as in the previous season. The wheat crop was harvested in April 2002 and again rice was planted from July to November 2002 in the same pots followed by wheat till April 2003 with recommended application of NPK fertilizers.

3. RESULTS

3.1. Greenhouse experiment with wheat and cabbage

The transfer factors for ^{134}Cs ranged from 6.37 to 12.8x10^{-3} in grain and from 7.81 to 14.44x10^{-3} in straw, respectively (Table 3). With increasing level of potassium fertilizer application the TF values both in grain and straw in all the soils decreased significantly.

TABLE 3. TRANSFER FACTORS (x 1000) OF ^{134}Cs TO WHEAT AND CABBAGE AS INFLUENCED BY POTASSIUM FERTILIZER APPLIED IN FIRST CROP

K level (kg K_2O/ha	Soil			Mean		Sem (±)	CD at 5%	CD at 1%
	Red	Alluvial	Black					
	Winter 1999–2000 season (Wheat Grain)							
0	8.85	11.93	12.82	11.20	Soil	0.28	0.59	0.81
75	7.49	9.16	7.78	8.14	K	0.28	0.59	0.81
150	7.12	9.33	6.37	7.61	Soil x K	0.49	1.02	NS
	7.82	10.14	8.99					
	Winter 1999–2000 season (Wheat Straw)							
0	9.78	13.41	14.44	12.54	Soil	0.30	0.64	0.88
75	7.79	9.68	9.68	9.05	K	0.30	0.64	0.88
150	7.21	9.62	8.33	8.39	Soil x K	0.53	NS	NS
	8.26	10.90	10.82					
	Monsoon 2000 season (Cabbage)							
0	9.57	10.91	12.09	10.86	Soil	0.21	0.44	0.60
75	6.43	6.66	6.95	6.68	K	0.21	0.44	0.60
150	5.23	5.16	5.60	5.33	Soil x K	0.36	NS	NS
	7.08	7.57	8.22					
	Winter 2000–2001 season (Cabbage)							
0	7.05	8.91	9.56	8.51	Soil	0.16	0.34	0.47
75	5.86	7.66	7.70	7.07	K	0.16	0.34	0.47
150	4.90	6.89	5.94	5.91	Soil x K	0.28	NS	NS
	5.94	7.82	7.73					
	Monsoon 2001 season (Cabbage)							
0	7.05	8.52	9.07	8.21	Soil	0.18	0.38	0.51
75	5.53	7.49	5.48	6.16	K	0.18	0.38	0.51
150	4.63	6.02	3.98	4.88	Soil x K	0.31	NS	NS
	5.73	7.34	6.18					
	Winter 2001–2002 season (Cabbage)							
0	4.60	4.75	5.26	4.87	Soil	0.08	0.18	NS
75	3.33	3.26	3.16	3.25	K	0.08	0.18	0.24
150	2.94	2.62	2.87	2.81	Soil x K	0.15	NS	NS
	3.62	3.54	3.76					
	Monsoon 2002 season (Cabbage)							
0	3.74	3.40	4.29	3.81	Soil	0.07	0.14	0.19
75	2.61	2.12	2.74	2.49	K	0.07	0.14	0.19
150	2.14	1.73	2.30	2.06	Soil x K	0.11	NS	NS
	2.83	2.42	3.11					
	Winter 2002–2003 season (Cabbage)							
0	3.23	3.07	3.83	3.38	Soil	0.08	0.16	0.22
75	2.25	2.20	2.27	2.24	K	0.08	0.16	0.22
150	1.85	1.83	1.84	1.84	Soil x K	0.13	NS	NS
	2.44	2.37	2.65					

In the third crop of cabbage taken after contamination the TF values ranged from 4.90 to 9.56x10^{-3} with increasing level of potassium fertilizer application and these values decreased significantly in all the soils. There were significant differences in transfer factors of ^{134}Cs in different soils. The lowest mean TF values were obtained in red soil and the highest in black soil without application of potassium. In the next monsoon season the cabbage crop, fourth in sequence, showed the similar trend although the values were lower than that obtained in the previous crop of cabbage. Here again potassium application significantly decreased the TF values and the differences in mean values in different soils were statistically significant. In subsequent three seasons the TF values of ^{134}Cs in cabbage decreased progressively and in all the crops and soils potassium application lowered the values. In the seventh crop taken after contamination of soil with ^{134}Cs the TF values ranged from 1.83×10^{-3} to 3.83×10^{-3}, which is nearly three times less than those of the first cabbage crop.

The data in Table 4 show that the TF of ^{90}Sr in wheat (grain and straw) and cabbage were significantly different in the soils studied. However, they were not influenced by calcium application like those of ^{134}Cs by potassium. Also they were 50–100 times higher than those for ^{134}Cs. The transfer factor of ^{90}Sr in cabbage in the following two seasons (winter, 2000–01 and monsoon, 2001) were significantly different in the soils studied and the calcium application had a slight but significant effect in lowering the TF values in all the soils. In the fourth season the cabbage crop had nearly five times lower values of TF compared to that in the first crop of wheat grown immediately after contamination. Also, as in the first two crops, in the third and fourth crop the TF values of ^{90}Sr were 50–100 times higher than those for ^{134}Cs. There was a trend of continuous decrease of TF values in the each successive crop of cabbage and also the calcium application significantly decreased the TFs which was not found in the first two crops (wheat or cabbage) taken immediately after contamination of soils.

TABLE 4. TRANSFER FACTORS (x 10) OF ^{90}Sr TO WHEAT AND CABBAGE AS INFLUENCED BY CALCIUM FERTILIZER APPLIED IN THE FIRST CROP OF WHEAT UNDER GREENHOUSE CONDITIONS

K Level (kg K_2O/ha	Soil			Mean		Sem (±)	CD at 5%	CD at 1%
	Red	Alluvial	Black					
			Winter 1999–2000 season (Wheat Grain)					
0	5.19	4.88	5.08	5.05	Soil	0.14	0.29	0.40
75	5.04	4.86	6.82	5.52	Ca	0.14	NS	NS
150	6.93	6.82	6.96	6.91	Soil x Ca	0.24	NS	NS
	5.72	5.52	5.73					
			Winter 1999–2000 season (Wheat Straw)					
0	6.30	5.95	6.15	6.13	Soil	0.14	0.29	0.40
75	6.22	6.10	6.13	6.15	Ca	0.14	NS	NS
150	7.58	7.49	7.95	7.67	Soil x Ca	0.24	NS	NS
	6.70	6.51	6.74					
			Monsoon 2000 season (Cabbage)					
0	4.67	4.64	4.49	4.60	Soil	0.10	0.21	0.29
75	4.05	4.12	4.11	4.09	Ca	0.10	NS	NS
150	5.34	5.30	5.21	5.28	Soil x Ca	0.17	NS	NS
	4.69	4.69	4.60					
			Winter 2000–2001 season (Cabbage)					
0	3.10	3.63	4.33	3.69	Soil	0.07	0.14	0.19
75	3.07	3.11	4.11	3.43	Ca	0.07	0.14	0.19
150	2.63	2.64	4.16	3.14	Soil x Ca	0.11	NS	NS
	2.94	3.12	4.20					
			Monsoon 2001 season (Cabbage)					
0	2.89	2.60	3.10	2.86	Soil	0.07	0.14	0.20
75	2.51	1.87	2.93	2.43	Ca	0.07	0.14	0.20
150	2.49	1.89	2.75	2.38	Soil x Ca	0.12	NS	NS
	2.63	2.12	2.92					
			Winter 2001–2002 season (Cabbage)					
0	2.67	2.38	3.80	2.95	Soil	0.06	0.14	0.19
75	2.54	2.40	3.68	2.87	Ca	0.06	0.14	0.19
150	2.43	2.23	3.56	2.74	Soil x Ca	0.11	NS	NS
	2.55	2.34	3.68					
			Monsoon 2002 season (Cabbage)					
0	2.65	2.49	3.92	3.02	Soil	0.06	0.13	0.17
75	2.50	2.52	3.80	2.94	Ca	0.06	0.13	0.17
150	2.20	2.34	3.68	2.74	Soil x Ca	0.10	NS	NS
	2.45	2.45	3.80					
			Winter 2002–2003 season (Cabbage)					
0	2.39	2.20	3.45	2.68	Soil	0.04	0.09	0.12
75	2.28	2.22	3.41	2.64	Ca	0.04	0.09	0.12
150	2.00	2.10	3.30	2.46	Soil x Ca	0.07	NS	NS
	2.22	2.17	3.39					

3.2. Field experiment

Transfer factors of ^{137}Cs From the soil contaminated in 1996 to crops (cabbage, wheat or maize) taken from the 1998–99 winter season to 2002–03 winter monsoon season showed a decreasing trend but the values were much higher in cabbage than in wheat or maize grain (Table 5).

TABLE 5. TRANSFER FACTORS OF ^{137}Cs IN CABBAGE, WHEAT AND MAIZE UNDER IRRIGATED FIELD CONDITIONS

Crop		Contamination level (kBq kg^{-1} soil)	
		18.5	37.0
		TF x 1000 ± SD	TF x 1000 ± SD
Cabbage (1998–99)	Leaves	8.18 ± 0.64	8.39 ± 0.55
Maize (1999)	Grain	3.19 ± 0.40	4.22 ± 0.33
	Straw	5.39 ± 0.42	5.34 ± 0.43
Cabbage (1999–2000)	Leaves	5.79 ± 0.49	5.78 ± 0.24
Wheat (1999–2000)	Grain	3.91 ± 0.34	4.20 ± 0.30
	Straw	4.25 ± 0.23	4.95 ± 0.55
Cabbage (2000)	Leaves	6.01 ± 0.56	7.29 ± 0.20
Maize (2000)	Grain	4.43 ± 0.74	4.95 ± 0.63
	Straw	7.02 ± 0.47	7.11 ± 0.48
Cabbage (2000–01)	Leaves	6.07 ± 1.34	7.06 ± 0.81
Wheat (2000–01)	Grain	2.93 ± 0.28	6.34 ± 0.45
	Straw	4.39 ± 0.27	7.04 ± 0.46
Cabbage (2001)	Leaves	4.48 ± 0.47	5.90 ± 0.81
Maize (2001)	Grain	3.36 ± 0.30	5.70 ± 0.31
	Straw	2.62 ± 0.30	6.23 ± 0.43
Cabbage (2001–02)	Leaves	4.73 ± 0.74	6.51 ± 0.86
Wheat (2001–02)	Grain	2.71 ± 0.27	4.16 ± 0.25
	Straw	3.02 ± 0.45	4.55 ± 0.20
Cabbage (2002)	Leaves	3.67 ± 0.47	5.73 ± 0.79
Maize (2002)	Grain	2.76 ± 0.34	3.30 ± 0.48
	Straw	3.01 ± 0.32	3.68 ± 0.48
Cabbage (2002–03)	Leaves	3.39 ± 0.32	4.58 ± 0.47
Wheat (200203)	Grain	2.41 ± 0.16	2.63 ± 0.16
	Straw	2.97 ± 0.29	3.37 ± 0.42

The numbers after ± are standard deviations

The value of soil to plant transfer factor of ^{137}Cs decreases slowly over the seasons and this decrease is comparatively very much less than that observed in our earlier studies under rainfed conditions in the same soil.

The transfer factor values for ^{90}Sr are much lower than those recorded for the first crop after contamination, as has been the case in earlier investigations in rainfed conditions (Table 6). In case also of ^{90}Sr the TF values in cabbage were higher than those in wheat and maize grain or straw.

TABLE 6. TRANSFER FACTORS OF ^{90}Sr IN CABBAGE, WHEAT AND MAIZE UNDER IRRIGATED FIELD CONDITIONS

Crop		Contamination level (kBq kg^{-1} soil)	
		18.5	37.0
		TF x 10 ± SD	TF x 10 ± SD
Cabbage (1998–99)	Leaves	4.68 ± 0.42	6.12 ± 0.34
Maize (1999)	Grain	2.18 ± 0.25	2.59 ± 0.24
	Straw	3.26 ± 0.35	3.55 ± 0.15
Cabbage (1999–2000)	Leaves	2.75 ± 0.33	2.65 ± 0.13
Wheat (1999–2000)	Grain	1.97 ± 0.24	2.12 ± 0.05
	Straw	2.65 ± 0.37	3.12 ± 0.21
Cabbage (2000)	Leaves	2.03 ± 0.26	1.88 ± 0.10
Maize (2000)	Grain	1.47 ± 0.23	1.46 ± 0.14
	Straw	1.86 ± 0.10	1.97 ± 0.05
Cabbage (2000–01)	Leaves	1.72 ± 0.29	1.90 ± 0.16
Wheat (2000–01)	Grain	1.34 ± 0.21	1.54 ± 0.06
	Straw	1.83 ± 0.05	2.14 ± 0.20
Cabbage (2001)	Leaves	1.58 ± 0.09	1.63 ± 0.10
Maize (2001)	Grain	1.27 ± 0.11	1.24 ± 0.31
	Straw	1.29 ± 0.06	1.55 ± 0.04
Cabbage (2001–02)	Leaves	1.47 ± 0.06	1.61 ± 0.09
Wheat (2001–02)	Grain	1.26 ± 0.12	1.18 ± 0.12
	Straw	1.24 ± 0.03	1.29 ± 0.04
Cabbage (2002)	Leaves	1.18 ± 0.19	1.48 ± 0.07
Maize (2002)	Grain	0.88 ± 0.13	1.07 ± 0.19
	Straw	1.04 ± 0.13	1.10 ± 0.23
Cabbage (2002–03)	Leaves	1.10 ± 0.10	1.37 ± 0.14
Wheat (2002–03)	Grain	1.12 ± 0.18	1.02 ±0.14
	Straw	1.29 ± 0.19	1.03 ± 0.15

The TFs of ^{137}Cs and ^{90}Sr from the field plots contaminated in 1999 are presented in Table 7. The values for ^{137}Cs for cabbage, even in the fourth season, were nearly three times higher than those observed in the same season for wheat or maize, grain or straw. The level of contamination of ^{137}Cs did not have much effect on values for cabbage but did affect those to maize and wheat grain or straw. The TF values under irrigated field conditions were very much higher than those for the successive crops in the same soil grown under rainfed conditions in our earlier studies.

TABLE 7. TRANSFER FACTORS OF ^{137}Cs AND ^{90}Sr IN CABBAGE, WHEAT AND MAIZE UNDER IRRIGATED FIELD CONDITIONS

		Contamination level (kBq kg^{-1} soil)	
		18.5	37.0
Crop		TF x 1000 ± SD	TF x 1000 ± SD
		^{137}Cs	
Cabbage (1999–2000)	Leaves	21.91 ± 2.58	29.46 ± 2.60
Wheat (1999–2000)	Grain	9.21 ± 0.62	16.19 ± 0.92
	Straw	13.25 ± 0.95	15.36 ± 0.85
Cabbage (2000)	Leaves	22.59 ± 1.80	24.09 ± 1.67
Maize (2000)	Grain	10.22 ± 0.92	10.86 ± 0.84
	Straw	13.47 ± 1.55	12.44 ± 0.67
Cabbage (2000–01)	Leaves	24.58 ± 2.03	27.79 ± 0.93
Wheat (2000–01)	Grain	8.68 ± 0.73	11.28 ± 1.01
	Straw	9.97 ± 0.73	11.87 ± 1.02
Cabbage (2001)	Leaves	21.33 ± 1.41	24.91 ± 0.20
Maize (2001)	Grain	8.62 ± 0.88	10.13 ± 0.40
	Straw	8.51 ± 0.72	11.30 ± 1.08
Cabbage (2001–02)	Leaves	19.76 ± 1.26	21.89 ± 0.36
Wheat (2001–02)	Grain	5.42 ± 0.72	6.80 ± 0.61
	Straw	6.10 ± 0.61	7.64 ± 0.56
Cabbage (2002)	Leaves	17.71 ± 0.87	17.31 ± 0.64
Maize (2002)	Grain	4.29 ± 0.50	6.42 ± 0.39
	Straw	5.10 ± 1.01	6.71 ± 0.41
Cabbage (2002–03)	Leaves	18.16 ± 0.64	12.36 ± 0.48
Wheat (2002–03)	Grain	4.12 ± 0.73	5.48 ± 0.53
	Straw	5.11 ± 0.32	5.00 ± 0.46
		^{90}Sr	
Cabbage (1999–2000)	Leaves	9.71 ± 0.23	9.89 ± 0.08
Wheat (1999-2000)	Grain	7.42 ± 0.30	7.79 ± 0.13
	Straw	8.00 ± 0.57	8.41 ± 0.49
Cabbage (2000)	Leaves	7.14 ± 0.25	8.03 ± 0.27
Maize (2000)	Grain	5.67 ± 0.40	5.94 ± 0.47
	Straw	7.04 ± 0.17	7.00 ± 0.15
Cabbage (2000–01)	Leaves	7.15 ± 0.38	7.90 ± 0.29
Wheat (2000–01)	Grain	5.49 ± 0.33	5.95 ± 0.06
	Straw	6.10 ± 0.44	6.89 ± 0.17
Cabbage (2001)	Leaves	4.30 ± 0.26	6.14 ± 0.24
Maize (2001)	Grain	2.91 ± 0.31	4.23 ± 0.26
	Straw	3.86 ± 0.06	5.69 ± 0.14
Cabbage (2001–02)	Leaves	4.06 ± 0.07	5.57 ± 0.29
Wheat (2001–02)	Grain	2.53 ± 0.32	3.87 ± 0.23
	Straw	3.25 ± 0.26	4.49 ± 0.17
Cabbage (2002)	Leaves	3.55 ± 0.31	5.03 ± 0.28
Maize (2002)	Grain	2.33 ± 0.16	2.78 ± 0.34
	Straw	2.34 ± 0.35	3.04 ± 0.29
Cabbage (2002–03)	Leaves	3.74 ± 0.47	5.07 ± 0.24
Wheat (2002–03)	Grain	2.56 ± 0.16	2.61 ± 0.26
	Straw	2.64 ± 0.35	2.72 ± 0.36

The TF values for ^{90}Sr in the fourth cabbage crop decreased by a factor greater than two compared with those in the first crop after contamination of soil. The values in the same season were generally higher in cabbage in wheat and maize grain or straw by a factor of 2–3. In the first two seasons there was little difference in ^{90}Sr TF values from the two levels of contamination und but in later seasons the TF values for the higher level contamination were greater.

3.3. Greenhouse experiment with Rice

Transfer factors of ^{137}Cs in rice grain and straw in different soils are given in Table 8. The TF values were higher to straw than to grain. The values did not differ greatly between the three different Inceptisol soils. There was a slightly higher value of TF for ^{137}Cs in both rice grain and straw from the soil where normally rice is grown during the monsoon season which has higher clay content than the other two soils.

TABLE 8. TRANSFER FACTOR OF ^{137}Cs IN RICE GRAIN AND STRAW IN THREE ALLUVIAL SOILS

Soil	Rice (2001)			
	Grain		Straw	
	TF x 1000	SD (±)	TF x 1000	SD (±)
1	7.65	0.55	9.73	0.43
2	7.64	0.62	10.26	0.66
3	8.70	0.46	10.70	0.55
	Wheat (2001–02)			
	Grain		Straw	
1	5.14	0.54	6.73	0.39
2	5.11	0.58	7.25	0.68
3	6.20	0.47	7.71	0.56
	Rice (2002)			
	Grain		Straw	
1	5.45	0.55	6.98	0.40
2	5.42	0.59	7.52	0.70
3	6.53	0.48	7.99	0.57
	Wheat (2002–03)			
	Grain		Straw	
1	4.03	0.55	4.55	0.38
2	4.00	0.59	5.08	0.72
3	5.12	0.48	5.56	0.58

In the following crop of wheat taken in the winter season (2001–02) the TF values in both grain and straw were lower than those found in the first rice crop. In the next monsoon season the rice crop showed slightly higher values than the previous wheat crop but they were lower than in the rice crop grown a year before, immediately after soil contamination. The fourth crop of wheat taken in the next season had still lower values of ^{137}Cs transfer factors. The soil having higher clay content (soil No. 3) consistently showed higher values of transfer factors to all the four crops.

4. CONCLUSIONS

Under both pot culture and field conditions, the transfer factor values for ^{137}Cs and ^{90}Sr are higher for cabbage than those for wheat and maize. Also in the successive crops of cabbage the TF values are similar to those found in the first crop after contamination. The TF values for ^{137}Cs and ^{90}Sr are 50–100 times higher than those for ^{137}Cs. In rice the soil to plant transfer factor for ^{137}Cs is similar to those observed for upland cereal crops of wheat or maize and from soil having a higher clay content the ^{137}Cs transfer factor was greater.

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VARIATION OF TRANSFER FACTORS OF RADIONUCLIDES FOR FOOD CROPS IN JAPAN

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Abstract

To obtain the TFs of ^{137}Cs and ^{90}Sr from paddy soils to rice, analyses of global fallout ^{137}Cs and ^{90}Sr in both rice and the associated soils were carried out. Paddy soils were collected nationwide from 12–15 sampling sites from 1987–2001, and rice plants grown on these soils were also collected. The geometric-mean-TFs of ^{137}Cs were 0.0026 for brown rice and 0.0011 for polished rice, and for ^{90}Sr they were 0.015 and 0.0059, respectively. When the distribution ratios of ^{137}Cs and ^{90}Sr in polished rice to brown rice were calculated, the average ratios were 0.35 for ^{137}Cs and 0.36 for ^{90}Sr, indicating that these radionuclides were mainly associated with the bran. Then the TF of ^{137}Cs was particularly focused on in this study because the available data was sufficient for statistical analysis (i.e. the variation in the TFs for Cs from paddy soils to rice grain). The variation of TFs of ^{137}Cs for polished rice at each site year-to-year was very small, being within the range of two orders of magnitude, whereas the variation of ^{137}Cs-TFs for polished rice was somewhat greater between sample sites across Japan. The relationships between TF ^{137}Cs and soil properties (pH, exchangeable-potassium, cation exchange capacity and clay content) were also investigated. No correlations were found for ^{137}Cs-TFs with these properties, which was difficult to explain. More data must be collected for both TF and soil properties.

1. INTRODUCTION

The ingestion of contaminated crops and livestock products is the most important pathway through which radionuclides are taken into the human body and contribute to internal radiation dose. Radiation dose estimation is usually made with the aid of mathematical models in which model parameters express transfers of radionuclides from one environmental compartment to another. These parameters are often described as the concentration ratios of radionuclides between two compartments, when the system is in equilibrium. For example, transfers of nuclides from soil to crops, from grass to milk and from grass to meat have been estimated by using the parameters TF, F_m and F_f, respectively [1].

From the viewpoint of more precise radiological assessment, it is necessary to obtain the variations of transfer parameters that are used in the models. Among the parameters, a soil-to-crop transfer factor (TF) is the key parameter that directly affects the internal dose assessment for the ingestion pathway. Eating habits, however, differ between countries. For example, in European and North American countries, livestock products including meat, eggs and milk make a big contribution, whereas, cereals and vegetables are the major dietary components in Asian and South American countries. In Japan, rice is the most commonly consumed cereal and is therefore the most important crop for internal radiation dose assessment.

We obtained the TFs of Cs and Sr from paddy soils to rice by analyses of fallout ^{137}Cs and ^{90}Sr in both rice and the associated soils. It is illegal to use any kind of radioactive materials in open fields in Japan, hence our analyses used radioactive fallout. In this study, we particularly focused on the variation in the TFs for Cs from paddy soil to rice grains.

2. MATERIALS AND METHODS

2.1. Paddy soil and rice samples

Paddy soils (ploughed soil layer: 0–20 cm) were collected nationwide from 12–15 sampling sites (National and Prefectural Agriculture Experiment Stations) from 1961–2001, and rice plants grown on these soils have also been collected in the harvesting season. The sampling sites are shown in Fig. 1. About 5 kg of the soil were collected from each site. The soil samples in their natural moist conditions were sent to the National Institute of Agro-Environmental Sciences (NIAES) for analysis. They were air-dried, then passed through a 2 mm sieve and stored at room temperature until used.

Rice varieties grown traditionally in Japan are classified as short grain-types. About 10 kg of the unhusked seed plant sample were collected from each site and they were also sent to NIAES. Each rice sample was processed into brown rice (with bran) and polished rice (without bran). The average weight ratio of the polished rice to the brown rice was 0.9 (90% yield).

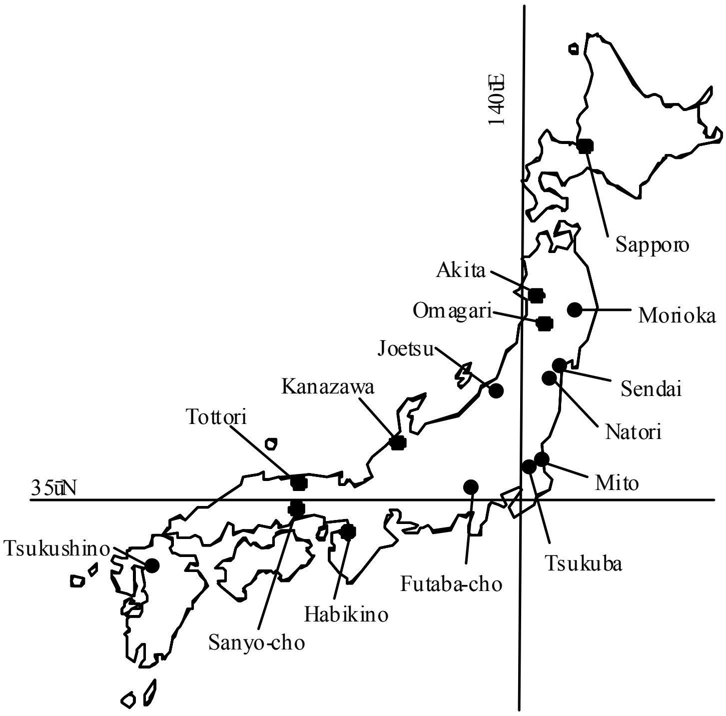

FIG. 1. Sampling sites of rice and soils for ^{137}Cs and ^{90}Sr measurements by National Institute of Agro-Environmental Sciences (NIAES).

2.2. Analytical methods

The analytical methods for ^{137}Cs and ^{90}Sr in the soil and plant samples were as follows. For ^{137}Cs measurement, a 40 to 60 g aliquot of the sieved soil sample was transferred into a plastic vessel and ^{137}Cs activity was measured with Ge semiconductor detector systems. For the rice grain samples, 1 kg of the brown rice and 3 kg of the polished rice were used. Each sample was incinerated at 500°C in an electric oven to concentrate low levels of the radionuclide. The incinerated sample was transferred into a plastic vessel to measure its ^{137}Cs activity with the Ge detector systems.

For ^{90}Sr analysis, 200 g of the sieved soil sample, 1 kg of the brown rice sample and 3 kg of the polished rice samples were each used. After incineration of the samples, a radiochemical separation of ^{90}Sr was carried out. The beta activities in each sample were counted with a low background gas flow counter [2].

3. RESULTS AND DISCUSSION

3.1. TFs of ^{137}Cs and ^{90}Sr for rice

The concentrations of fallout ^{137}Cs in brown rice, polished rice and paddy soils collected in 1999 are shown in Table 1; these are reproduced from Ref. [3]. The TFs were calculated for the present study from the reported data. They are also listed in the same table. Direct contamination of the rice seeds was considered negligible, because the radioactive fallout levels observed over the last decade were extremely low compared with the total fallout level. The maximum TFs for brown rice and polished rice were 0.02 and 0.008, respectively, while the minimum TFs were 0.0009 (brown rice) and 0.0002 (polished rice). The geometric-mean-TFs were 0.0026 (brown rice) and 0.0011 (polished rice).

Table 2 shows the concentrations of ^{90}Sr in brown rice, polished rice and paddy soils collected in 1990 (reproduced from Ref. [4]). The TFs were calculated for the present study from the reported data. They are listed in the last two columns of Table 2. The geometric-mean-TFs for ^{90}Sr were 0.015 for brown rice and 0.0059 for polished rice. The ^{137}Cs and ^{90}Sr distribution ratios in the polished rice to brown rice were calculated. The average ratios were 0.35 for ^{137}Cs and 0.36 for ^{90}Sr, indicating that most of the activity from these two radionuclides was associated with the bran. About 35% of each nuclide was distributed in the polished rice.

As shown in Tables 1 and 2, the variation in TFs was within two orders of magnitude for both ^{137}Cs and ^{90}Sr taken up by rice. For example, the ratios of the maximum values of TF for the polished rice to the minimum ones were 31 for ^{137}Cs and 12 for ^{90}Sr.

It would be interesting to compare the TFs of both these radionuclides for further discussion. However, the radiochemical analysis of ^{90}Sr is complicated and the concentrations in rice plant samples are extremely low. The numbers of available ^{90}Sr data were too limited to ensure reliable statistical analysis. Thus, in the following section, only the ^{137}Cs TF is discussed.

TABLE 1. CONCENTRATIONS OF ^{137}Cs IN SAMPLES (BROWN RICE, POLISHED RICE AND PADDY SOILS) COLLECTED IN 1999, AND TFs

Sampling sites	Cs-137			Transfer Factor	
	Brown rice (mBq kg^{-1})	Polished rice (mBq kg^{-1})	Paddy soil (Bq kg^{-1})	Brown rice	Polished rice
Sapporo, HOKKAIDO	16 ± 2.2	15 ± 1.8	0.52 ± 0.11	3.08E-02	2.88E-02
Akita, AKITA	59 ± 4.0	19 ± 1.3	12.5 ± 0.3	4.72E-03	1.52E-03
Omagari, AKITA	147 ± 5.2	64 ± 2.1	6.6 ± 0.3	2.23E-02	9.70E-03
Joetsu, NIIGATA	26 ± 2.5	19 ± 1.8	16.2 ± 0.5	1.60E-03	1.17E-03
Kanazawa, ISHIKAWA	-	4 ± 0.2	3.6 ± 0.2	-	1.11E-03
Tottori, TOTTORI	60 ± 2.7	23 ± 2.2	13.9 ± 0.5	4.32E-03	1.65E-03
Morioka, IWATE	36 ± 2.4	23 ± 2.1	11.8 ± 0.3	3.05E-03	1.95E-03
Natori, MIYAGI	40 ± 2.4	15 ± 0.8	7.9 ± 0.4	5.06E-03	1.90E-03
Mito, IBARAKI	10 ± 0.6	2 ± 0.3	0.65 ± 0.08	1.54E-02	3.08E-03
Tsukuba, IBARAKI	7 ± 0.7	10 ± 0.7	4.7 ± 0.3	1.49E-03	2.13E-03
Tachikawa, TOKYO	25 ± 1.6	20 ± 2.0	15.4 ± 0.5	1.62E-03	1.30E-03
Futaba-cho, YAMANASHI	-	4 ± 0.9	2.9 ± 0.1	-	1.38E-03
Habikino, OSAKA	6 ± 0.9	3 ± 0.3	4.8 ± 0.3	1.25E-03	6.25E-04
Sanyo-cho, OKAYAMA	15 ± 1.3	2 ± 0.1	6.3 ± 0.3	2.38E-03	3.17E-04
Tsukushino, FUKUOKA	-	85 ± 2.3	9.2 ± 0.4	-	9.24E-03
Number of samples	12	15	15	12	15

TABLE 2. CONCENTRATIONS OF Sr-90 IN SAMPLES (BROWN RICE, POLISHED RICE AND PADDY SOILS) COLLECTED IN 1990, AND TFs

Sampling sites	Sr-90			Transfer Factor	
	Brown rice (mBq kg^{-1})	Polished rice (mBq kg^{-1})	Paddy soil (Bq kg^{-1})	Brown rice	Polished rice
Sapporo, HOKKAIDO	33	10	4.22	7.8E-03	2.4E-03
Akita, AKITA	77	30	3.51	2.2E-02	8.5E-03
Omagari, AKITA	23	10	1.97	1.2E-02	5.1E-03
Joetsu, NIIGATA	83	22	5.72	1.5E-02	3.8E-03
Kanazawa, ISHIKAWA	9	3	1.07	8.4E-03	2.8E-03
Tottori, TOTTORI	45	15	1.83	2.5E-02	8.2E-03
Morioka, IWATE	4	3	0.91	4.4E-03	3.3E-03
Natori, MIYAGI	26	8	4.17	6.2E-03	1.9E-03
Mito, IBARAKI	22	10	1.81	1.2E-02	5.5E-03
Tsukuba, IBARAKI	30	10	1.19	2.5E-02	8.4E-03
Futaba-cho, YAMANASHI	3	3	0.13	2.3E-02	2.3E-02
Habikino, OSAKA	22	8	0.63	3.5E-02	1.3E-02
Sanyo-cho, OKAYAMA	14	5	0.74	1.9E-02	6.8E-03
Tsukushino, FUKUOKA	15	7	0.55	2.7E-02	1.3E-02
Number of samples	14	14	14		

3.2. Variation of Cs TF for polished rice

3.2.1. Nationwide variation

Figure 2 shows example results of ^{137}Cs TFs for polished rice collected in Akita (Ref. [5]). Although samples have been collected since 1961, we assumed that the soil-to-rice plant system was not in equilibrium before 1987.

The reasons why we did not use the samples before 1987 were as follows. In 1986, samples were collected in the typical harvest months of September and October. However, we have to take into account the Chernobyl accident. Most of the ^{137}Cs released from the reactor was deposited in the northern hemisphere during the spring of 1986. In Japan, the amount of fallout ^{137}Cs from the Chernobyl reactor did not greatly affect the total ^{137}Cs content in soil. However, rice is typically planted in April and May. When we discuss the TF of ^{137}Cs for rice plant samples collected in 1986, the direct deposition of ^{137}Cs onto plant had to be considered. Additionally, the TF data for the period 1961–1985 were excluded, because global fallout of ^{137}Cs from nuclear tests by USA, USSR and China was found. Thus, again, direct deposition of ^{137}Cs onto plant samples would probably have to be included in the calculated TFs during that time.

Thus, the ^{137}Cs-TF data obtained from 12–15 sites nationwide between 1987–2001 were used for this analysis. There were 169 samples measured. Figure 3 shows a probability distribution for TF of ^{137}Cs for polished rice collected in 1987–2001. The 169 samples showed a log-normal distribution of TFs. The variation of TF was about three orders of magnitude and the median was 0.002. The maximum TF was 0.029 and the minimum was 0.00005, giving a ratio of maximum TF to minimum TF of 550. The geometric and arithmetic means were 0.0019 and 0.0033, respectively.

3.2.2. Year-to-year variations of ^{137}Cs TFs for each sample site

The variations of ^{137}Cs TFs for polished rice obtained from 1987–2001 at eight sampling points are shown in Fig. 4 as a box-whisker plot. The ends of the box are the 25 and the 75 percentiles, also known as quartiles, and the distance between the quartiles is the interquartile range. The line across the middle of the box identifies the median sample value. Each box has lines that show the minimum and maximum values.

The medians were 0.0025 for Sapporo, 0.0033 for Akita, 0.0015 Joetsu, 0.0036 for Morioka, 0.0043 for Sendai, 0.0014 for Mito, 0.0013 for Tsukuba and 0.0035 for Tsukushino. From these results, it was clear that the TF values observed at these sampling sites were almost the same. At each site, the variation of TFs, that is, the ratios of the maximum to the minimum TF, were very small, within the range of two orders of magnitude. Maximum TF variation of 50 was found in Tuskushino, followed by Sapporo and Sendai (44 for both sites). In particular, we noted the variation ratio in Akita, Joetsu, Morioka, Mito and Tsukuba were less than 10. The reasons were not clear why there were large differences in the TFs for some fields and almost no differences for others, though a possible cause might be different sampling points in the same sites. Generally, paddy field conditions are well controlled so that it is expected that the soil properties would be almost uniform in one paddy field. One of the reasons for the variations of ^{137}Cs-TFs obtained in this study is the extremely low concentration of ^{137}Cs in polished rice. Further research is needed to obtain more precise information.

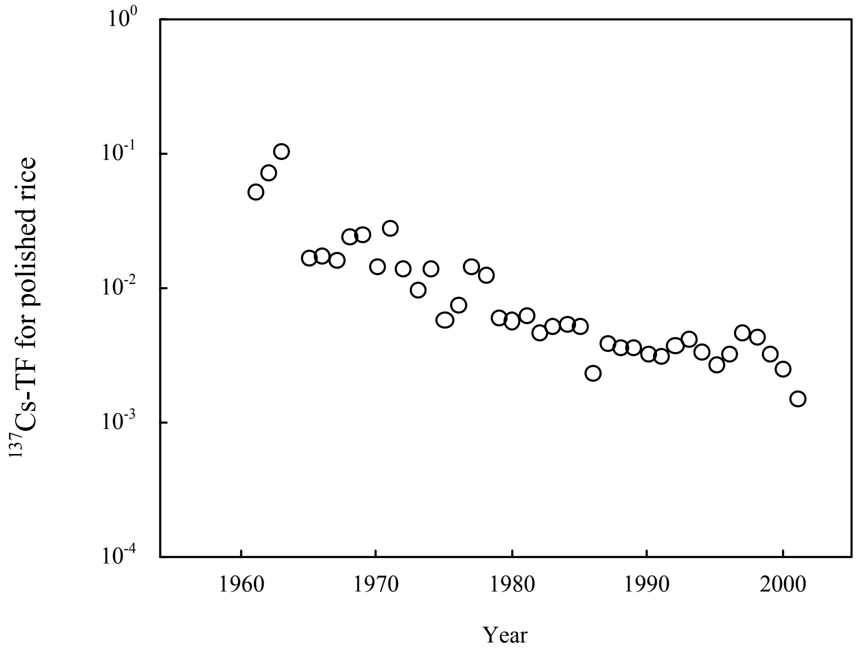

FIG. 2. Variations of ^{137}Cs-TFs for polished rice collected in Akita as a function of time.

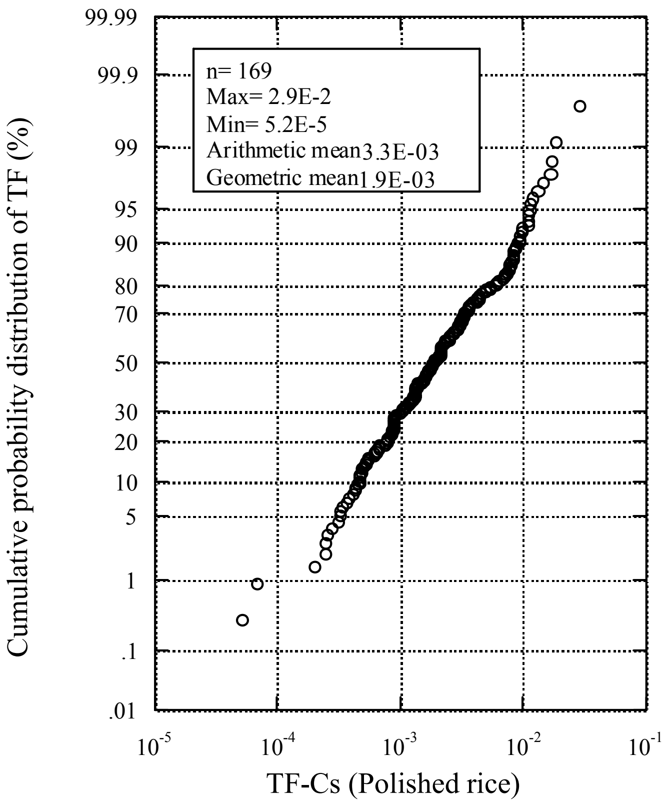

FIG. 3. Probability distribution of ^{137}Cs-TFs from paddy soil to polished rice.

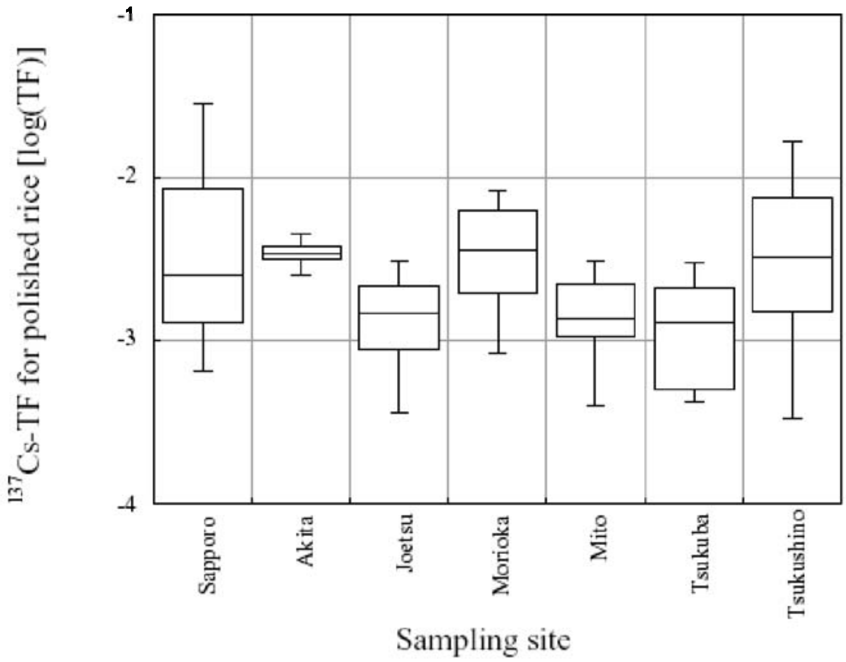

FIG. 4. Variation of TFs of ^{137}Cs for polished rice obtained from 1987–2001 in each.

3.2.3. Site-to-site variation of TFs for ^{137}Cs each year from 1987 to 2001

Figure 5 shows variations of ^{137}Cs TFs for the polished rice obtained in each year as a box-whisker plot. The median of TFs ranged from 0.00086–0.0027. The TF distributions seemed to be log-normal ones, but it was difficult to reliably identify the distribution patterns based on the relatively small sample size (n=12–15).

Although the variation in TFs was almost within the two orders of magnitude found for the year-to-year variation, the site-to-site variations found in 1992 and 1993 were greater than two orders of magnitude, being 331 and 139, respectively. This meant that the variation of ^{137}Cs TF for polished rice was somewhat greater between sample sites across Japan than from year-to-year at any sample site.

3.3 Relationship between TFs for ^{137}Cs and soil properties

Figure 6 shows relationships between TF of ^{137}Cs and some soil properties, such as pH (H_2O), exchangeable-potassium (cmol kg^{-1}), cation exchange capacity (cmol kg^{-1}) and clay content (%). No good correlations were found for ^{137}Cs-TFs with these soil properties. The TF of ^{90}Sr showed negative correlations with clay content, cation exchange capacity and humic acid content as discussed in Ref. [6]. Presumably, the extractabilities of ^{137}Cs and ^{90}Sr in the soil samples differed from each other. For example, distribution coefficients (Kds) of ^{137}Cs using Japanese agricultural soils [7] showed little correlation with soil properties except for exchangeable-K, but Kd-^{85}Sr showed good correlations with CEC, water content, total Al, Fe and C.

However, it is still difficult to explain why the TF for ^{137}Cs had no correlations with these soil properties. We need to collect more data for both TF and soil properties. It is also useful to compare the TFs of ^{137}Cs with those of stable Cs [8]. Recently, inductively coupled plasma mass spectrometry has become a powerful tool to measure low-levels of elements including stable Cs in plant and soil samples. We should be able to obtain more TF data not only for stable Cs, but also other elements in the future.

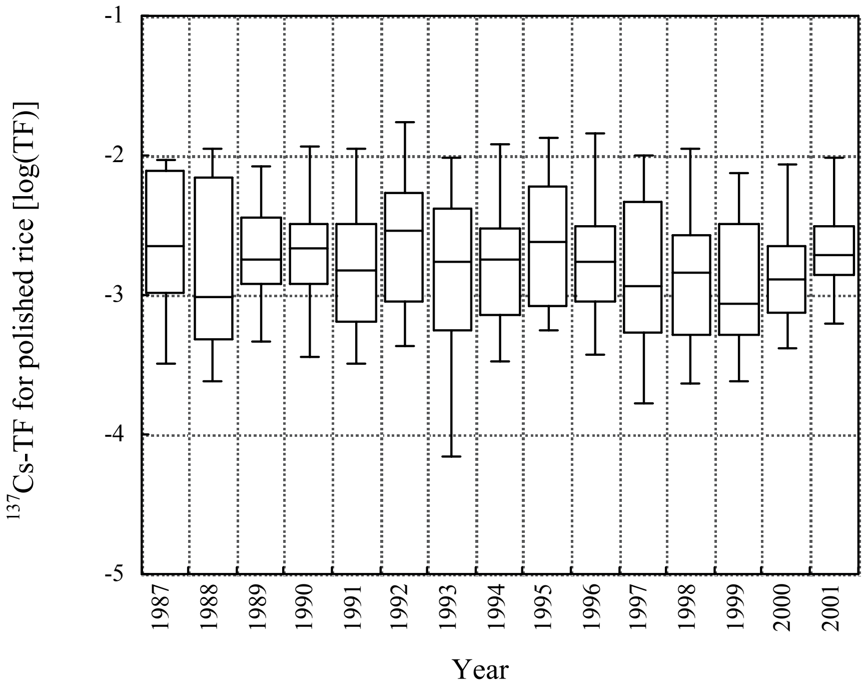

FIG. 5. Variations of ^{137}Cs-TFs for polished rice obtained from 1987 to 2001.

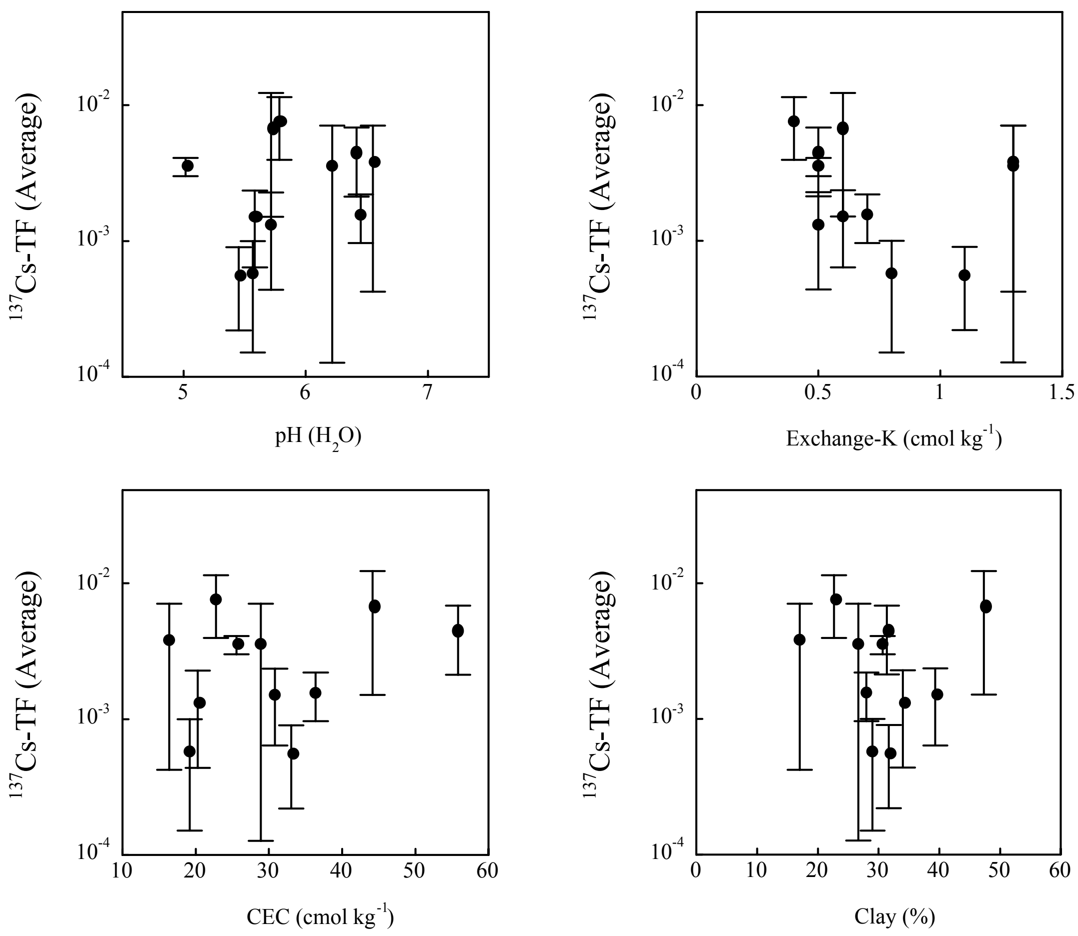

FIG. 6. Relationships between TFs of ^{137}Cs and some soil properties.

3.4. Conversion factors (CF)

Frissel et al. [9] proposed a new parameter, conversion factor (CF), to obtain generic values of TFs. We can estimate an unknown TF value from the TF value of the reference crop by using the CF. Tsukada measured ^{137}Cs concentrations in vegetables collected in Aomori Prefecture [10–12]. The TFs for potato, cabbage and polished rice are listed in Table 3. The TFs showed log-normal distributions. The TFs in the table are geometric means. From these data, we obtained the CFs. For volcanic ash soils, the CFs calculated are 9.4 for potato/polished rice and 9.1 for cabbage/polished rice. For non-volcanic ash soils, the CFs are 22 for potato/polished rice and 13 for cabbage/polished rice.

TABLE 3. TRANSFER FACTORS OF ^{137}Cs IN POTATOES, CABBAGE AND POLISHED RICE

Soil type	Potato	Cabbage	Polished rice
Volcanic ash soil	0.033 (n=16)	0.032 (n=6)	0.0035 (n=6)
Non-volcanic ash soil	0.025 (n=10)	0.015 (n=2)	0.0012 (n=14)

We also calculated the CFs based on the TFs of brown rice by using the ratio of the TFs for brown rice/polished rice. For volcanic ash soils, the calculated CFs are 3.3 for potato/brown rice and 3.2 for cabbage/brown rice, respectively. For non-volcanic ash soils, the factors are 7.6 for potato/polished rice and 4.6 for cabbage/brown rice, respectively.

4. CONCLUSIONS

(1) TFs (geometric means) of ^{137}Cs and ^{90}Sr for rice grain are 0.0043 and 0.015 for brown rice respectively and 0.0021 and 0.0059 for polished rice, respectively.
(2) The 169 data of the ^{137}Cs TFs showed a log-normal distribution. The geometric mean is 0.0019 and 95% confidence interval is from 0.00024 to 0.016.
(3) The year-to-year variations of ^{137}Cs TF for each sample site are within two orders of magnitude and site-to-site variations are greater than two orders of magnitude.
(4) No correlations are found between the TFs and some soil properties (CEC, pH, clay content, exchangeable-K).
(5) The ratios of ^{137}Cs and ^{90}Sr for polished rice to brown rice are 0.35 and 0.36, respectively.
(6) The conversion factors (CFs) are 9.4 for potato/polished rice and 9.1 for cabbage/polished rice for volcanic ash soils and the CFs for non-volcanic ash soils are 22 for potato/polished rice and 13 for cabbage/polished rice.

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THE CLASSIFICATION OF RUSSIAN SOIL SYSTEMS ON THE BASIS OF TRANSFER FACTORS OF RADIONUCLIDES FROM SOIL TO REFERENCE PLANTS

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Abstract

A database of TFs for ^{90}Sr and ^{137}Cs for crops from different Russia soils was developed. Among the key factors influencing TFs, soil properties and biological properties of crops were important. The differences of ^{137}Cs TFs for different soil types are up to 28.1 times for barley and 5.2 times for cabbage. The differences between barley and cabbage in the accumulation of radionuclide are from 1.1 to 7.0 times. Differences in means of TFs ^{90}Sr were 6.4 times for barley and 5.5 times for cabbage. The differences between barley and cabbage in the accumulation of radionuclide varied from 1.1–2.0 times. The accumulation of ^{90}Sr by crops was higher 2.6–14.3 times than that of ^{137}Cs for barley and 8.1–32.0 times for cabbage. Soil characteristics that largely influence the accumulation of radionuclides by plants were identified. Transfer factors were used for radioecological classification of soils. On the basis of the statistical processing of experimental data two schemes of soil classification have been developed based on ^{137}Cs transfer factors in barley and three schemes of soil classification based on the rate of ^{90}Sr uptake by plants.

1. INTRODUCTION

The accumulation of radionuclides in farm crops is a complicated process dependent on many factors. Soil properties are a factor influencing radionuclide accumulation. One of the integral indicators of radionuclide behaviour in soils of different types is the transfer factor (TF). The differences in TFs for different soil types can reach two orders of magnitude [1].

The use of transfer factors to describe the uptake of radionuclides by vegetation has been subject to a number of criticisms [2–4]. Initially designed for application in agricultural ecosystems, transfer factors may be particularly unsuitable for use in natural pastures where the soil is unploughed and the radionuclides being studied are therefore not uniformly distributed with soil depth. However, aggregated transfer factors, defined in terms of deposition (Bq m^{-2}) have been shown to be less variable than those defined in terms of activity (Bq kg^{-1}) in soil [5] and have been used with success in other radiological investigations [6, 7].

After the Chernobyl accident a database for transfer factors from different soil types to plants was developed in the Russian Institute of Agricultural Radiology and Agroecology. There are approximately 3,000 measurements of radionuclide content in samples of soils and agricultural crops (winter rye, potato, barley, oat, spring and winter wheat, maize, perennial grasses, natural grasses, beet) collected in 11 regions in Russia [8–13]. These data were used for evaluation of radionuclide behaviour in agricultural systems and for prediction of countermeasure effectiveness [11, 14–16].

There are more than 300 soil types and sub-types within Russia. The soil classification system in Russia is rather complex. The taxonomic system of the soil-geographic territory division consists of the following sub-units [17, 18]: soil-bioclimatic area; soil-bioclimatic region; soil zone; soil province; soil district; and soil sub-district.

The soil-geographic regions are identified by zones from north to south. The whole territory of Russia is divided into soil-geographic regions homogeneous in soil cover.

The accumulation of radionuclides in farm crops varies considerably for different soil-climatic zones being largely dependent on the soil properties. The highest availability of radionuclides is reported for peaty-swamp, sod-podzolic sandy and sandy loam soil. Soils with higher fertility and heavier texture retain radionuclides more strongly. Grouping of transfer factor data according to the soil-geographic division of the territory has shown that in general TF tends to decrease from north to south because of the change from low fertility soils of light texture to fertile heavy ones. The radionuclide transfer factors to farm crops for these soils are one order of magnitude lower than for low-fertility soils (Table 1).

TABLE 1. MEANS FOR ^{90}Sr AND ^{137}Cs AGGREGATED TRANSFER FACTORS TO CROPS IN DIFFERENT SOIL-BIOCLIMATIC REGIONS [19]

Soil-Bioclimatic region	Type of soil	Cereal		Perennial grasses	
		^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs
Taiga-forest region	Sod-podzolic sandy and sandy loam	0.41	0.10	2.4	1.5
	Sod-podzolic medium loam	0.40	0.04	1.5	0.31
	Sod-gley sandy loam	-	-	-	1.8
	Gley medium loam	-	-	2.7	0.49
	Soddy-calcareous medium loam	0.19	0.02	1.3	0.39
	Peaty-swamp	-	-	-	6.6
Central forest-steppe and steppe region	Sod-podzolic sandy loam	0.95	0.29	2.33	2.6
	Grey forest light loam	0.8	0.31	2.73	1.78
	Dark grey forest medium sandy loam	0.53	0.14	1.9	1.38
	Chernozem podzolized medium sandy loam	0.47	0.20	1.95	1.80
	Chernozem leached medium sandy loam	0.13	0.08	1.13	0.55
	Chernozem typical heavy loam	0.09	0.04	0.35	0.12
Steppe region	Chernozem ordinary heavy loam	0.14	0.09	1.07	0.35
	Chernozem south heavy loam	0.12	0.03	0.66	0.32
Dry steppe region	Chestnut and dark chestnut heavy loam	0.14	0.05	0.46	0.12
	Chernozem south clay	0.12	0.04	0.56	0.16

Thus, studies of radionuclide migration within the soil-plant system have obtained data that can be the basis for the classification of soils by radioecological criteria. Such a classification makes it possible to predict the contamination of agricultural products and to determine intervention levels in the event of radioactive contamination of agricultural land.

Development of a classification of soils on the basis of transfer factors of radionuclides from soil to different crops was the overall objective of this the Coordinated Research Project. The database of TFs for ^{90}Sr and ^{137}Cs was developed using data obtained from 1999–2002. Soil characteristics that largely influence the accumulation of radionuclides by plants were identified and the transfer factors were used for radioecological classification of soils.

2. MATERIALS AND METHODS

2.1. Experimental plots

The experimental plots were chosen in different soil-climatic zones. In 1999 sites were selected in the Voronezsh, Kursk, Orel, Tula and Bryansk regions. In the Voronezsh region, experimental plots are located on different sub-types of chernozem (Chernic Chernozem and Haplic Chernozem). In the Kursk region Grey-Luvic Phaeozem, Luvic Chernozem and Molli-Gleyic Fluvisols are present. The experimental plots in the Orel and Tula regions are located on Luvic Chernozem, Albic Luvisols, Grey-Luvic Phaeozem and Umbric Podzols. Experimental plots in the Bryansk region are located on Umbric Podzols.

To expand the spectrum of the soils studied, plots were added in 2000–2002 in the Rostov and Saratov region where Haplic Kastanozems and Calcaric Chernozems dominate the soil cover. Fourteen plots were in added the Bryansk region where the soil cover is dominated by peat soil (Umbric Podzols, Dystric Histosols and Eutri-Histic Gleysols).

The experimental plots are located on agricultural land. The crop cultivation techniques can influence the accumulation of radionuclides in plants. To minimize this, plots were selected on fields where conventional local cultivation methods were used.

^{90}Sr and ^{137}Cs accumulation in barley and cabbage were studied. Contamination of experimental plots in the Voronezsh, Kursk, Orel, Tula and Bryansk regions resulted from the Chernobyl fallout and in Rostov and Saratov regions is due to nuclear weapons fallout.

Descriptions of the experimental plots, which were 100 m^2, are presented in Table 2.

2.2. Methods of sampling and analysis

At each plot four soil and plant samples were taken from a sampling area of 1–2 m^2. Barley was sampled in July and cabbage - in September.

Soil agrochemical properties were determined by conventional procedures [20]. ^{137}Cs was determined by high-resolution gamma spectrometry using a coaxial Ge(Li) detector (ORTEC GMX-series) connected to a multi-channel analyser (IN-1200, France). ^{90}Sr was determined by a radiochemistry method. Activity of the daughter ^{90}Y was measured with low-level beta-counter (Canberra 2404).

3. RESULTS AND DISCUSSION

3.1. Characteristics of soils

3.1.1. Agrochemical parameters of soils

Agrochemical soil parameters are presented in Table 3. Characteristics differ significantly within each type of soil. To analyse the results, the soils are divided into two groups—mineral and organic. The first group includes different types of Chernozems, Haplic Kastanozems, Grey-Luvic Phaeozem, Albic Luvosols, Umbric Podzols and Molli-Gleyic Fluvisols. The second group includes Dystric Histosols and Eutri-Histic Gleysols.

TABLE 2. DESCRIPTION OF EXPERIMENTAL PLOTS

Plot	Region	District	The nearest village	Type of soil	Crop
1-2	Rostov	Zumlyansk	Podgorenskaya	Haplic Kastanozems	Barley, cabbage
3-4			N.Zhukovka		Barley, cabbage
5-6	Saratov	Balakovo	Natal'ino	Calcaric Chernozem	Barley, cabbage
7-8			Balakovo		Barley, cabbage
9-10	Voro-	Kashira	L. Rossosh	Chernic Chernozem	Barley, cabbage
11	nezsh		Kolodeznoye		Cabbage
12			Mosalskoye	Haplic Chernozem	Cabbage
13			Hvorostan	Chernic Chernozem	Barley
14			Drakino		Cabbage
15			Davudovka		Cabbage
16		Ostrogozshsk	Storozshevoye	Haplic Chernozem	Barley, cabbage
18			Boldurevka		Cabbage
19		Hoholsk	Archangelskoye	Chernic Chernozem	Barley
20			Hoholsk		Cabbage
21			Hohol		Cabbage
22	Kursk	Kurchatov	Komyakino	Luvic Chernozem	Barley
23			Dichnya		Cabbage
24-25			Dronyaevo	Grey-Luvic Phaeozem	Barley, cabbage
26			Lukashevka	Molli-Gleyic Fluvisols	Cabbage
27-28	Orel	Bolhov	Scherbinskye	Albic Luvosols	Barley, cabbage
29-30			Cherno	Umbric Podzols	Barley, cabbage
31-32	Tula	Plavsk	Molochnye Dvory	Luvic Chernozem	Barley, cabbage
33			Rahmanovo	Grey-Luvic Phaeozem	Barley
34-35	Bryansk	Novozybkov	VIUA	Umbric Podzols	Barley, cabbage
36-37			Katichi	Dystric Histosols (Dried peat)	Barley, cabbage
38-39			Demenka	Umbric Podzols	Cabbage, barley
40-41			Katichi	Umbric Podzols	Cabbage, barley
42			S.Bobovitch	Umbric Podzols	Cabbage
43					Cabbage
44					Barley
45				Eutri-Histic Gleysols	Barley
46-47				Dystric Histosols	Cabbage, barley
48-49			N.Bobovitch	Umbric Podzols	Cabbage, barley
50-51				Dystric Histosols	Cabbage, barley
52					Barley

TABLE 3. AGROCHEMICAL CHARACTERISTICS OF SOILS OF EXPERIMENTAL PLOTS

Plot	Type of soil	Crop	pH_{kcl}	OM (%)	K_{exch}	Ca_{exch}	CEC
					$cmol_c kg^{-1}$		
1	Haplic Kastanozems	Barley	7.9	1.9	1.94	19.2	29.5
2		Cabbage	7.9	2.3	2.21	20.4	29.9
3		Barley	6.5	1.9	1.56	18.0	24.9
4		Cabbage	7.6	2.1	1.7	20.0	27.0
5	Calcaric Chernozem	Barley	7.2	2.65	1.1	20.0	36.8
6		Cabbage	7.0	3.44	0.88	18.5	26.2
7		Barley	7.1	2.74	0.74	18.6	26.5
8		Cabbage	7.3	3.72	1.11	22.75	38.8
9	Chernic Chernozem	Barley	6.3	2.92	1.1	27.9	37.8
10		Cabbage	6.7	2.9	0.8	25	38.5
11		Cabbage	6.0	2.8	0.88	24.7	36.2
12	Haplic Chernozem	Cabbage	6.0	2.9	0.78	24.1	38.7
13	Chernic Chernozem	Barley	5.9	2.4	0.94	24.8	36.5
14		Cabbage	7.1	3.1	1.22	27.8	38.9
15		Cabbage	7.4	3.1	1.75	31.3	44.8
16	Haplic Chernozem	Barley	6.3	2.1	0.92	18.8	31.3
17		Cabbage	7.0	2.9	1.44	28.9	42.5
18		Cabbage	6.3	3.1	1.4	30.4	42.1
19	Chernic Chernozem	Barley	5.9	2.8	1.3	26.6	39.1
20		Cabbage	6.6	2.9	1.12	29.9	44.1
21		Cabbage	6.5	3.0	1.05	30.6	40.6
22	Luvic chernozem	Barley	5.9	1.7	0.89	20.0	30.3
23		Cabbage	6.5	1.9	0.91	22.1	36.4
24	Grey-Luvic Phaeozem	Barley	6.3	1.6	0.44	14.5	21.1
25		Cabbage	5.8	1.4	0.6	12.2	18.6
26	Molli-Gleyic Fluvisols	Cabbage	7.2	1.9	1.77	14.8	32.6
27	Albic Luvosols	Barley	5.6	0.9	0.7	8.2	12.3
28		Cabbage	5.5	1.1	0.84	10.6	18.1
29	Umbric Podzols	Barley	6.3	1.3	0.44	7.9	13.9
30		Cabbage	5.8	1.3	0.34	9.1	13.8
31	Luvic chernozem	Barley	5.8	2.1	0.68	16.6	22.4
32		Cabbage	5.6	2.6	0.67	18.9	28.8
33	Grey-Luvic Phaeozem	Barley	5.5	1.6	0.73	10.4	18.7
34	Umbric Podzols	Barley	5.7	1.5	0.45	5.0	11.1
35		Cabbage	5.9	0.9	0.50	6.1	13.1
36	Dystric Histosols	Barley	4.8	15.6	0.18	5.7	30.7
37	(Dried peat)	Cabbage	5.3	25.4	0.13	6.9	65.9
38	Umbric Podzols	Cabbage	5.3	1.3	0.23	6.9	10.3
39		Barley	4.6	1.31	1.07	2.4	3.6
40	Umbric Podzols	Cabbage	6.7	1.97	1.42	6.0	6.8
41		Barley	4.8	0.8	0.19	4.9	10.8
42	Umbric Podzols	Cabbage	5.75	1.75	1.69	6.4	11.2
43		Cabbage	7.0	0.85	0.50	4.0	8.3
44		Barley	4.7	1.0	0.31	5.4	12.5
45	Eutri-Histic Gleysols	Barley	4.8	16.76	0.18	5.9	34.7
46	Dystric Histosols	Cabbage	5.5	25.2	0.59	20.5	86.0
47		Barley	5.1	19.0	0.50	22.4	47.5
48	Umbric Podzols	Cabbage	5.9	1.47	0.77	4.0	10.6
49		Barley	4.9	1.0	0.19	4.9	13.0
50	Dystric Histosols	Cabbage	4.6	6.67	0.13	7.0	17.1
51		Barley	4.7	12.5	0.34	7.9	32.5
52		Barley	4.8	18.3	0.27	11.4	43.0

Ranges of data for mineral soils are : pH_{KCl} 5.3–7.9; OM 0.9–3.44%; K_{exch} 0.23–2.21 $cmol_c kg^{-1}$; Ca_{exch} 5.0–31.3 $cmol_c kg^{-1}$; CEC 10.3–44.8 $cmol_c kg^{-1}$. Parameters for Eutri-Histic Gleysols and Dystric Histosols are: pH_{KCl} 4.6–5.5; OM 6.67-25.4%; K_{exch} 0.13–0.59 $cmol_c kg^{-1}$; Ca_{exch} 5.7–22.4 $cmol_c kg^{-1}$; CEC 17.1–86.0 $cmol_c kg^{-1}$.

Differences in the soil parameters vary over a wide range. Ranges of data for barley soils are: pH_{KCl} – 1.7 times, OM – 21.1 times; K_{exch} – 10.8 times, Ca_{exch} - 11.6 times and CEC – 10.9 times. Ranges for cabbage are: 1.7, 29.6, 17.0, 7.8 and 12.6 times, respectively**.**

3.1.2. Composition of soils

Particle size analysis of soils was performed by N.A. Kachinsky's method [21]. The soils studied are highly variable in particle size distribution. Thus, the content of particles <0.01 mm varies from 9.7–68.12%. The content of the silt fraction (content of particles <0.001 mm) varies from 1.3–35.89% (Table 4).

3.2. Estimation of radionuclides accumulation by crops from different soils

^{90}Sr and ^{137}Cs in soils and crops were determined. The uptake of radionuclides by crop is reported as the specific activity in crop divided by the specific activity in soil:
TF = (Bq/kg plant)/(Bq/kg soil).

3.2.1. Accumulation of ^{137}Cs by crops from different soils

The results illustrate the influence of soil properties and biological peculiarities of crops on ^{137}Cs TFs. For the period 1999–2002 the differences in means of TFs ^{137}Cs for different soil types reach 28.1 times for barley and 5.2 times for cabbage (Table 5). The differences between barley and cabbage are from 1.1–7.0 times.

The highest availability of ^{137}Cs is reported for peat (Dystric Histosols and Eutri-Histic Gleysols), Molli-Gleyic Fluvisols and sod-podzolic sandy and sandy loam soil (Umbric Podzols). Soils with higher fertility and higher content of clay retain ^{137}Cs more strongly. The studies have shown that radionuclides are accumulated less by plants from Hapric Kastanozems and different chernozems than from Grey-Luvic Phaeozem, Umbric Podzols and Dystric Histosols.

3.2.2. Accumulation of ^{90}Sr by crops from different soils

Differences of ^{90}Sr TFs for different soil types were 6.4 times for barley and 5.5 times for cabbage. The differences between barley and cabbage in the accumulation of ^{90}Sr varied from 1.1–2.0 times (Table 6). The highest availability occurred in Dystric Histosols, Eutri-Histic Gleysols, Umbric Podzols, Albic Luvosols, Grey-Luvic Phaeozem and Molli-Gleyic Fluvisols.

The accumulation of ^{90}Sr by crops was higher than that of ^{137}Cs. The differences averaged 2.6–14.3 times for barley and 8.1–32 times for cabbage (Tables 5,6).

The behaviour of ^{137}Cs and ^{90}Sr is different, because of different mechanisms of radionuclide fixation in soil. ^{137}Cs exists in soil mainly in the non-exchangeable form, whereas ^{90}Sr sorption is characterized by a cation exchange.

TABLE 4. SOIL TEXTURES

Plot	Type of soil	Soil type codes	Particle size composition (%)		
			>0.01 mm	<0.01 mm	<0.001 mm
1	Haplic Kastanozems	C	39.24	60.76	33.96
2		C	38.14	61.86	32.07
3		C	37.87	62.13	35.87
4		C	39.65	60.35	32.67
5	Calcaric Chernozems	C	33.57	66.43	34.51
6		C	32.85	67.15	31.12
7		C	37.21	62.79	32.24
8		C	31.88	68.12	34.44
9	Chernic Chernozem	C	39.23	60.77	33.98
10		C	39.23	60.77	33.98
11		C	37.87	62.13	35.89
12	Haplic Chernozem	C	39.15	60.85	29.59
13	Chernic Chernozem	C	40.85	59.15	26.84
14		C	39.65	60.35	32.67
15		C	38.62	61.38	31.55
16	Haplic Chernozem	C	38.14	61.86	32.07
17		C	38.14	61.86	32.07
18		C	46.56	53.44	27.18
19	Chernic Chernozem	C	54.54	45.46	29.29
20		C	51.68	48.32	28.91
21		C	46.46	53.54	28.88
22	Luvic Chernozem	C	40.32	53.68	27.17
23		C-L	59.38	40.62	18.91
24	Grey-Luvic Phaeozem	C-L	58.85	41.15	21.15
25		L-C	62.32	37.68	20.08
26	Molli-Gleyic Fluvisols	C	52.46	47.54	28.15
27	Albic Luvosols	L	71.93	28.07	13.74
28		L	73.85	26.15	14.15
29	Umbric Podzols	L	69.35	30.65	15.21
30		L	69.35	30.65	15.21
31	Luvic chernozem	L-C	60.16	39.84	19.62
32		L	65.75	34.25	17.28
33	Grey-Luvic Phaeozem	C	54.65	45.35	23.15
34	Umbric Podzols	S	89.14	10.86	1.70
35		S-L	86.46	13.54	1.78
36	Dystric Histosols	S-L	85.4	14.60	2.66
37		L	79.39	20.61	2.82
38	Umbric Podzols	S	86.34	13.66	1.76
39		S	90.22	9.78	2.6
40	Umbric Podzols	S	89.68	10.32	1.8
41		S	89.16	10.84	1.8
42	Umbric Podzols	S-L	87.95	12.05	2.2
43		S	89.57	10.43	1.3
44		S-L	85.92	14.08	2.5
45	Eutri-Histic Gleysols	S-L	82.53	17.47	4.7
46	Dystric Histosols	L-S	76.08	23.92	6.0
47		L-S	76.36	23.64	7.5
48	Umbric Podzols	S	90.23	9.77	1.6
49		S	88.96	11.04	3.0
50	Dystric Histosols	S-L	84.94	15.06	7.1
51		L-S	78.89	21.11	3.5
52		S-L	82.11	17.89	3.0

TABLE 5. TFs OF ^{137}Cs BY CROPS FROM DIFFERENT SOILS (Bq/kg plant)/(Bq/kg soil), 1999–2002

Type of soil	1999		2001		2002	
	Average	SD	Average	SD	average	SD
Barley. grain						
Haplic Kastanozems			0.0072	0.0025	0.0076	0.0013
Calcaric Chernozems			0.0040	0.0010	0.0051	0.0002
Chernic Chernozem	0.006	0.005	0.0117	0.0064	0.0081	0.0015
Haplic Chernozem	0.0043	0.006	0.0093	0.0021	0.0047	0.0036
Luvic Chernozem	0.0111	0.0028	0.0146	0.0065	0.0145	0.0078
Grey-Luvic Phaeozem	0.0074	0.0034	0.0233	0.0133	0.0154	0.0062
Albic Luvosols	0.017	0.004	0.0294	0.0079	0.035	0.0090
Umbric Podzols	0.023	0.0004	0.0450	0.0200	0.0246	0.0137
Dystric Histosols			0.0860	0.0280	0.0228	0.0105
Eutri-Histic Gleysols	0.055	0.019			0.10	0.0033
Cabbage. head						
Haplic Kastanozems			0.0080	0.0024	0.0007	0.0001
Calcaric Chernozems			0.0041	0.0027	0.0021	0.0002
Chernic Chernozem	0.005	0.006	0.0054	0.0029	0.0026	0.0017
Haplic Chernozem	0.006	0.005	0.0064	0.0021	0.0021	0.0006
Luvic Chernozem	0.002	0.0012	0.0065	0.0081	0.0027	0.0010
Grey-Luvic Phaeozem	0.0028	0.0006	0.0036	0.0007	0.0014	0.0007
Molli-Gleyic Fluvisols	0.017	0.02	0.0227	0.0044	0.0008	0.0074
Albic Luvosols	0.001	0.0005	0.0120	0.0030	0.0015	0.0010
Umbric Podzols	0.015	0.001	0.0086	0.0028	0.0050	0.0036
Dystric Histosols			0.0101	0.0039	0.0054	0.0036

3.2.3. Influence of soil properties on uptake of ^{137}Cs and ^{90}Sr by the crops

Distinctions in ^{137}Cs and ^{90}Sr accumulation from different soil types result from the influence of soil characteristics, which are responsible for the biological availability of radionuclides.

A preliminary data analysis has shown that ^{137}Cs accumulation is inversely proportional to the contents of exchangeable potassium and calcium, cation exchange capacity, content of particles <0.01 mm (clay) (Figs 1–3,6,7–9,12). There are differences between crops with clear-cut relationships for barley but these are not so well pronounced for cabbage.

There is a trend towards declining in TFs ^{137}Cs to barley and cabbage with the increasing pH (Fig. 4,10). It should be noted that the effect pH depends partly on the response of different crops to pH. Thus, cabbage grows well only with neutral or weak alkaline reaction (pH7-8) but barley grows well only with neutral or weak acidic reaction (pH6-7).

There appeared to be no influence of organic matter (Fig. 5, 11).

TABLE 6. TFs OF ^{90}Sr BY THE CROPS FROM DIFFERENT SOILS (Bq/kg plant)/(Bq/kg soil), 1999-2002

Type of soil	1999		2001		2002	
	average	SD	Average	SD	average	SD
Barley (grain)						
Haplic Kastanozems			0.078	0.032	0.0895	0.0474
Calcaric Chernozems			0.054	0.027	0.0765	0.0332
Chernic Chernozem	0.037	0.013	0.059	0.018	0.0540	0.0028
Haplic Chernozem	0.122	0.019	0.053	0.032	0.05	0.024
Luvic Chernozem	0.086	0.011	0.080	0.023	0.073	0.016
Grey-Luvic Phaeozem	0.102	0.03	0.103	0.048	0.125	0.050
Albic Luvosols	0.163	0.038	0.158	0.056	0.219	0.049
Umbric Podzols	0.093	0.039	0.126	0.071	0.141	0.058
Dystric Histosols			0.280	0.144	0.355	0.109
Eutri-Histic Gleysols	0.122	0.034			0.28	0.087
Cabbage (head)						
Haplic Kastanozems			0.055	0.032	0.0585	0.0092
Calcaric Chernozems			0.033	0.005	0.032	0.0099
Chernic Chernozem	0.038	0.017	0.046	0.021	0.033	0.0102
Haplic Chernozem	0.0509	0.0011	0.054	0.021	0.0447	0.0021
Luvic Chernozem	0.108	0.050	0.073	0.043	0.041	0.019
Grey-Luvic Phaeozem	0.093	0.026	0.074	0.014	0.086	0.032
Molli-Gleyic Fluvisols	0.0709	0.018	0.143	0.019	0.169	0.034
Albic Luvosols	0.078	0.019	0.090	0.032	0.107	0.021
Umbric Podzols	0.0468	0.0304	0.103	0.038	0.0821	0.0182
Dystric Histosols			0.159	0.062	0.1995	0.0804

For ^{90}Sr, the dependence of the TFs on various soil properties is similar to that of ^{137}Cs but the relation between ^{90}Sr accumulation in crops and pH_{KCl} (Fig. 16, 22) and content of OM (Fig. 17, 23) is weaker. At the same time, ^{90}Sr TFs tend to decrease with increase in exchangeable potassium (Fig. 13, 19), exchangeable calcium (Fig. 14, 20), cation exchange capacity (Fig. 15, 21) and clay content (Fig. 18, 24).

Thus, soil properties influence the behaviour of ^{90}Sr and ^{137}Cs in the soil-plant system differently. Statistical methods need to be applied to estimate the effects of individual soil parameters on the radionuclide migration [22].

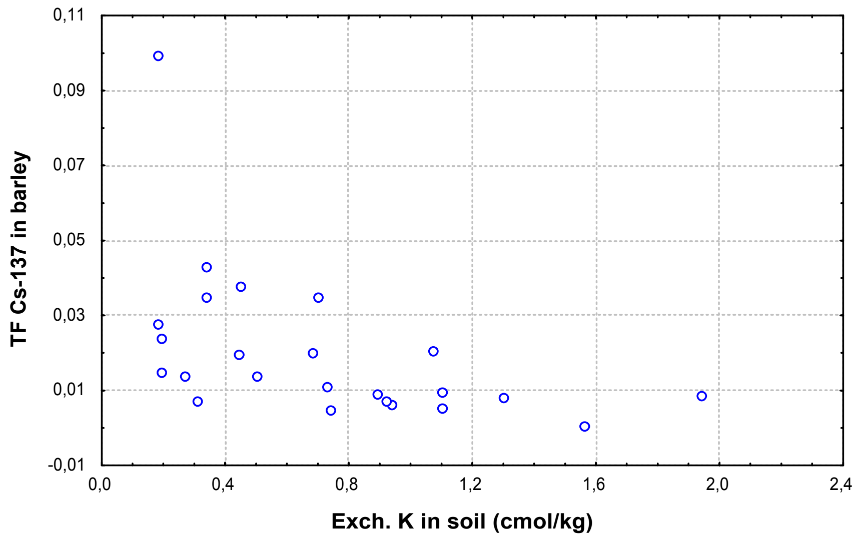

Fig. 1. The relationship between K and the ^{137}Cs TF for barley.

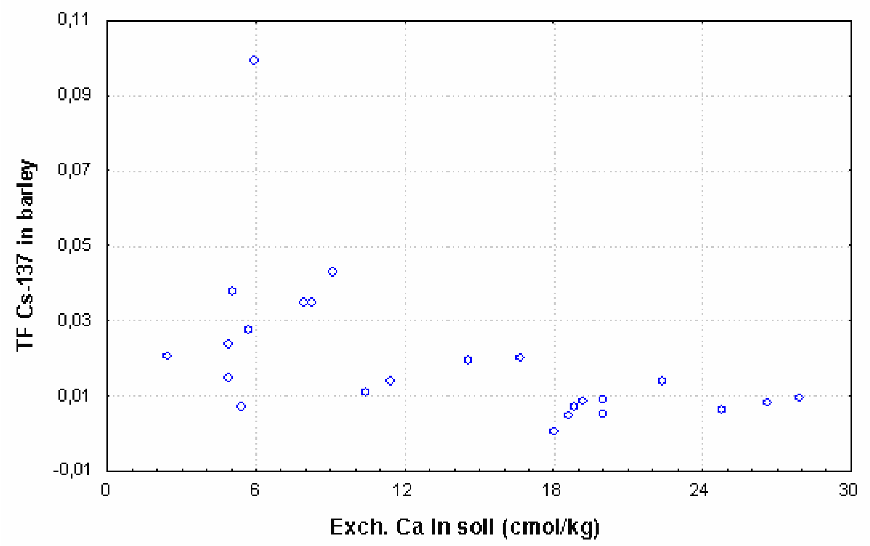

Fig. 2. The relationship between Ca and the ^{137}Cs TF for barley.

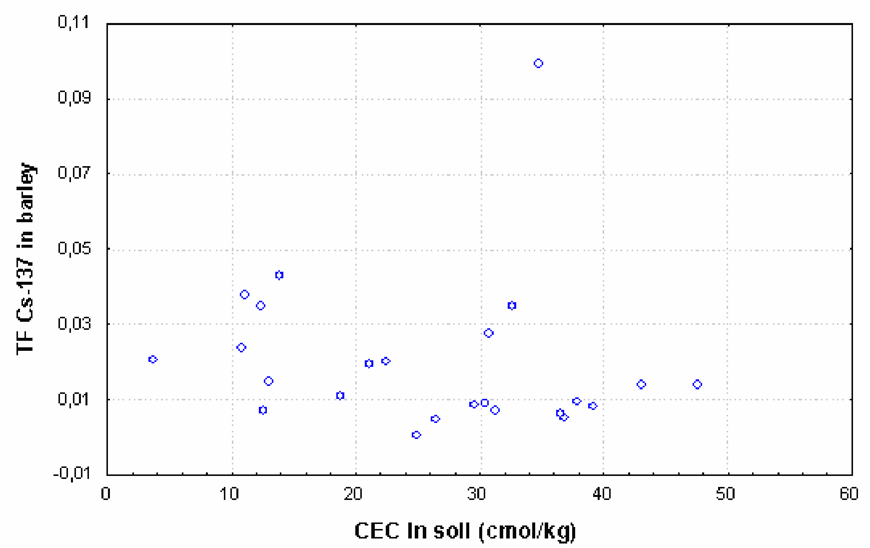

Fig. 3. The relationship between CEC and the ^{137}Cs TF for barley.

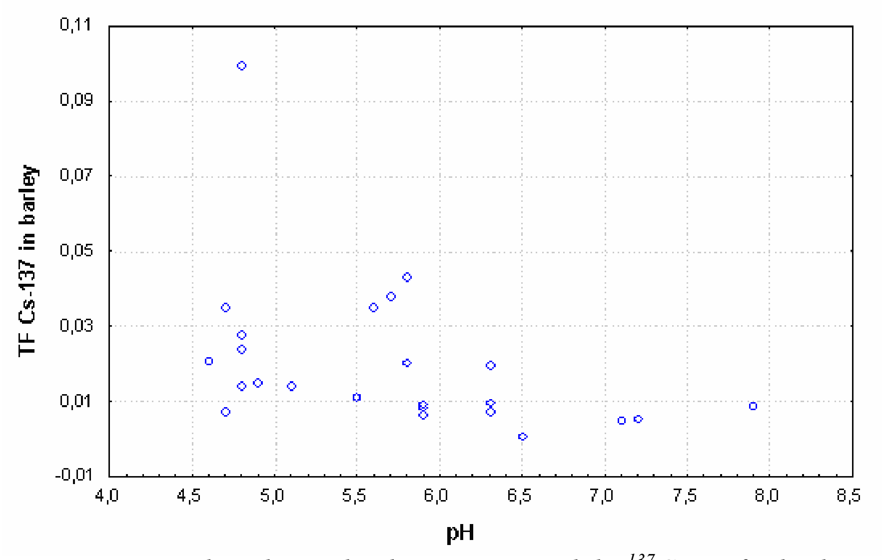

Fig. 4. The relationship between pH and the ^{137}Cs TF for barley.

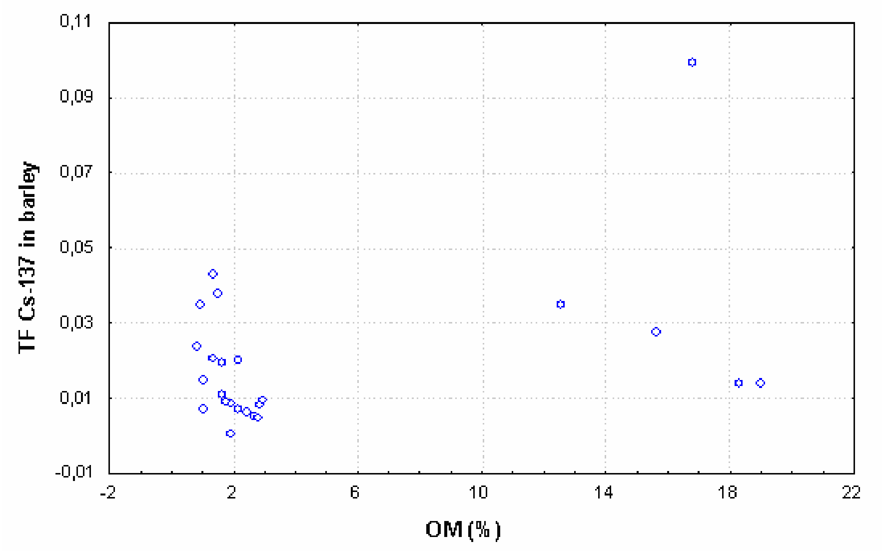

Fig. 5. The relationship between OM and the ^{137}Cs TF for barley.

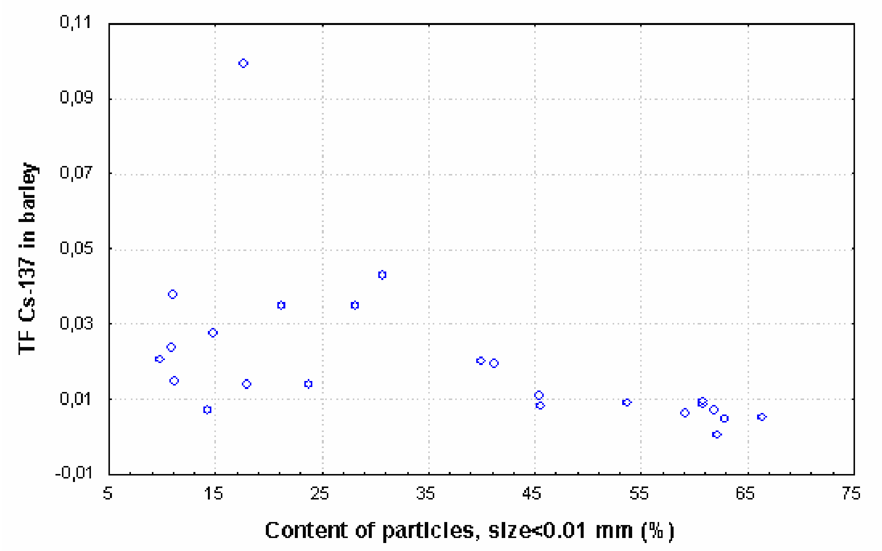

Fig. 6. The relationship between the content of particles (<0.01 mm) and the ^{137}Cs TF for barley.

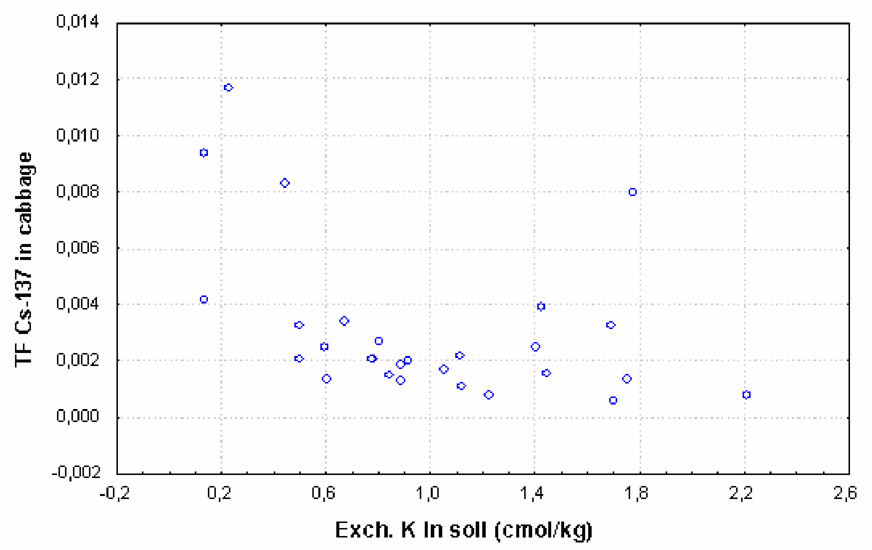

Fig. 7. The relationship between K and the ^{137}Cs TF for cabbage.

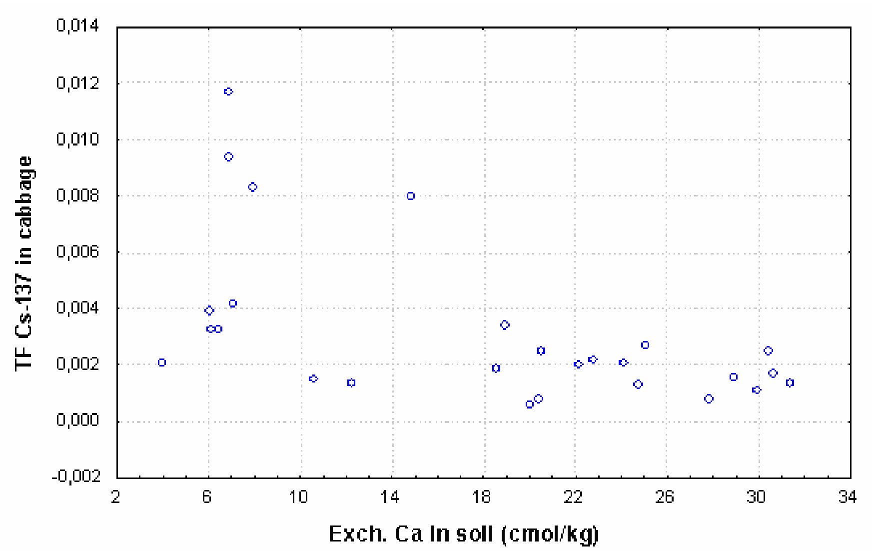

Fig. 8. The relationship between Ca and the ^{137}Cs TF for cabbage.

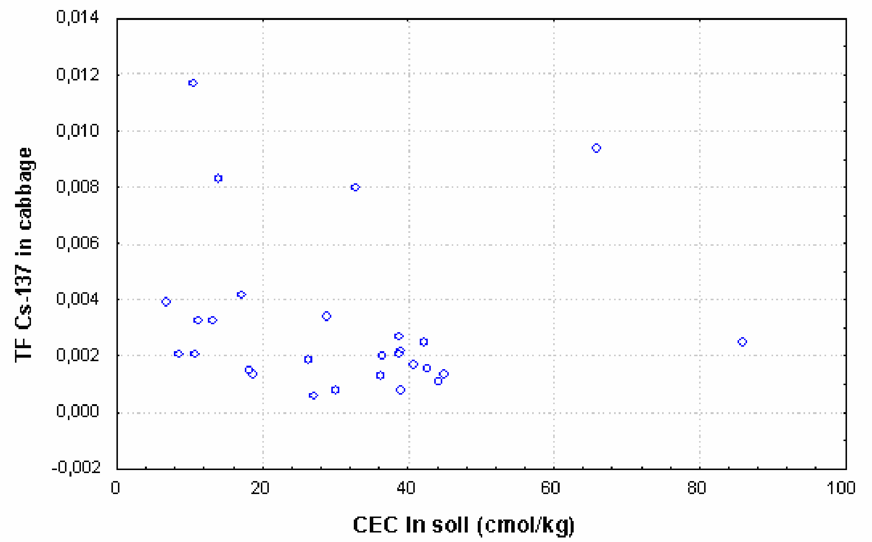

Fig. 9. The relationship between CEC and the ^{137}Cs TF for cabbage.

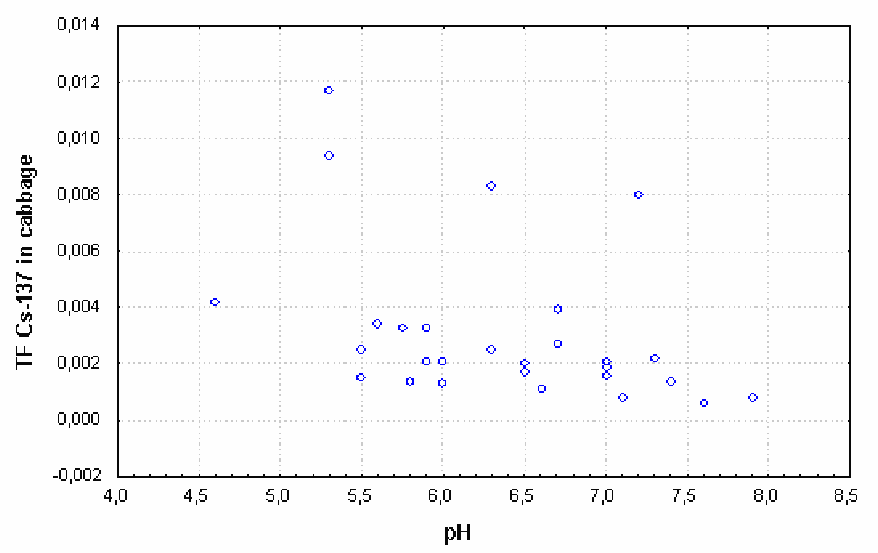

Fig. 10. The relationship between pH and the ^{137}Cs TF for cabbage.

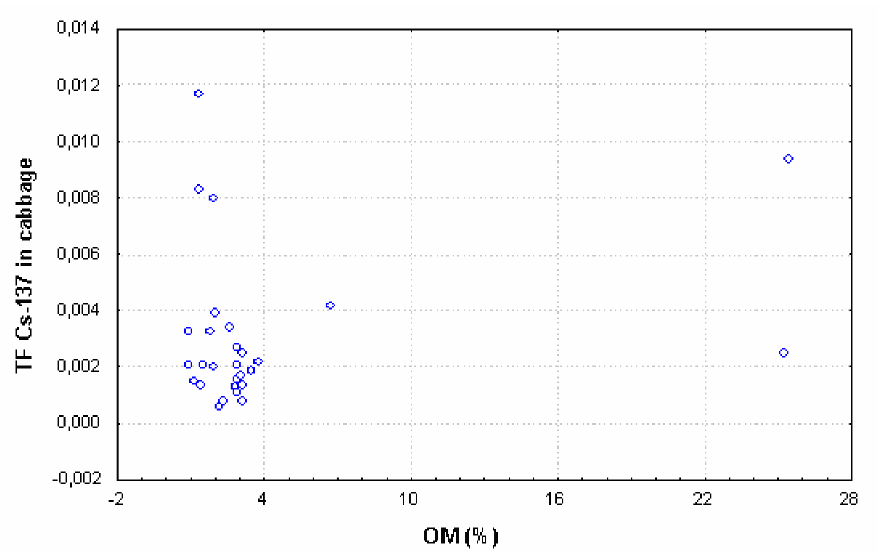

Fig. 11. The relationship between OM and the ^{137}Cs TF for cabbage.

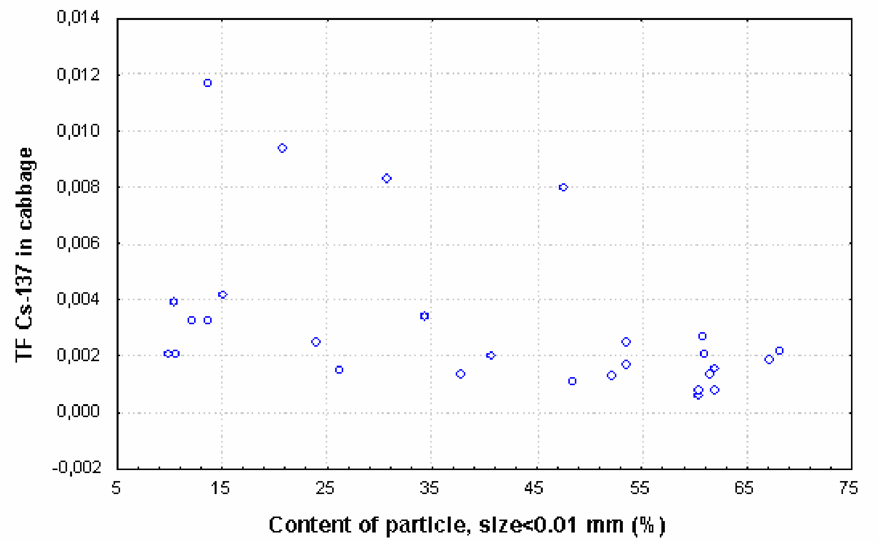

Fig. 12. The relationship between the content of particles (<0.01 mm) and the ^{137}Cs TF for cabbage.

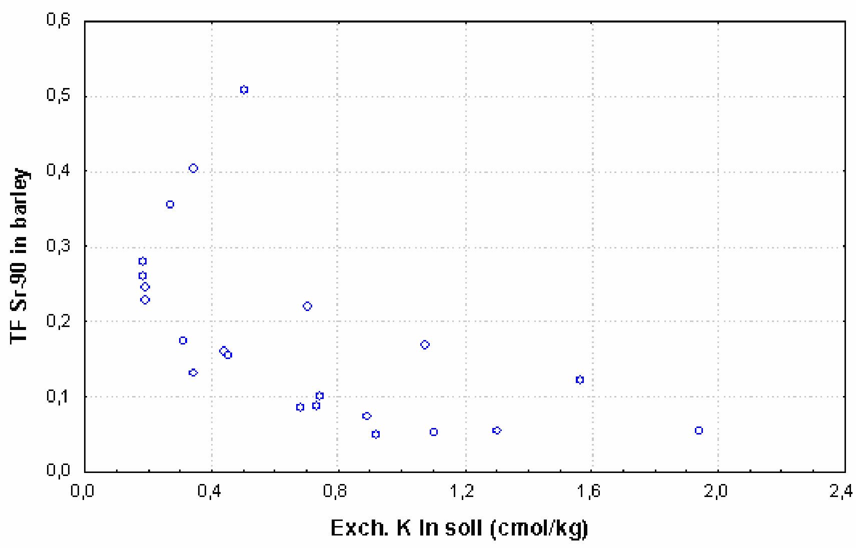

Fig. 13. The relationship between K and the ^{90}Sr TF for barley.

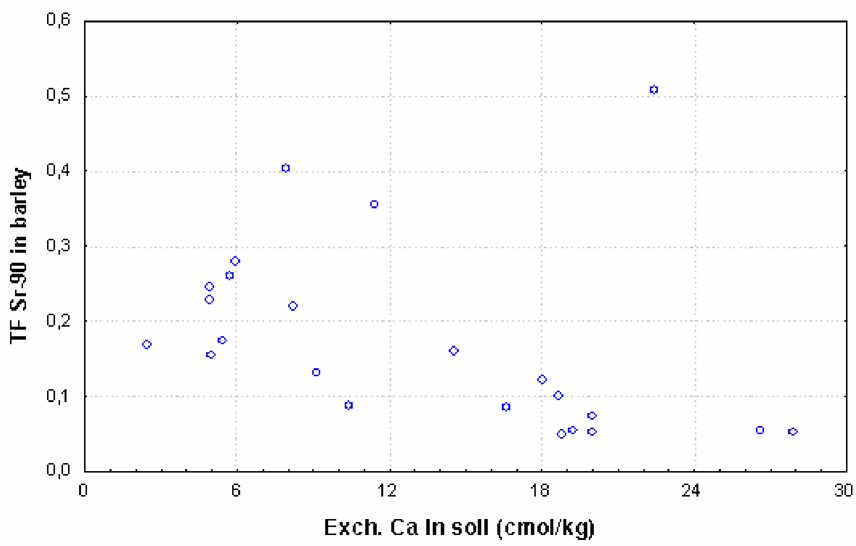

Fig. 14. The relationship between Ca and the ^{90}Sr TF for barley.

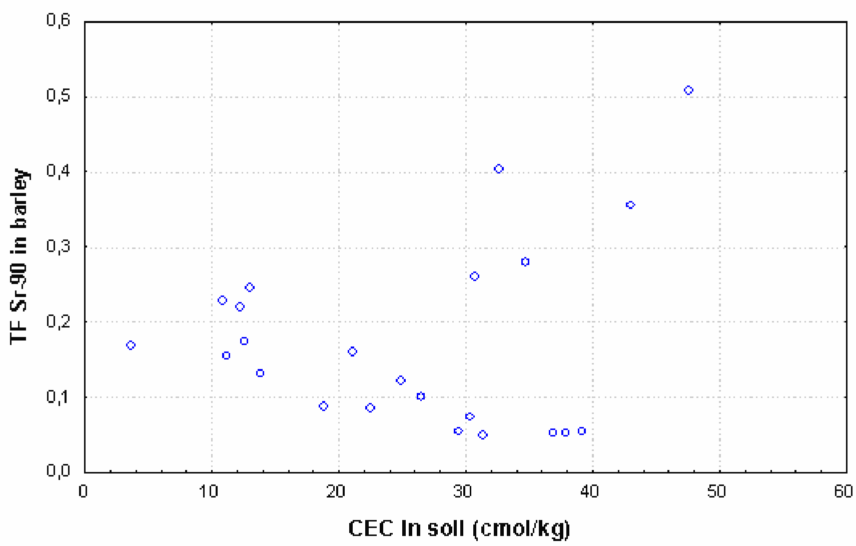

Fig. 15. The relationship between CEC and the ^{90}Sr TF for barley.

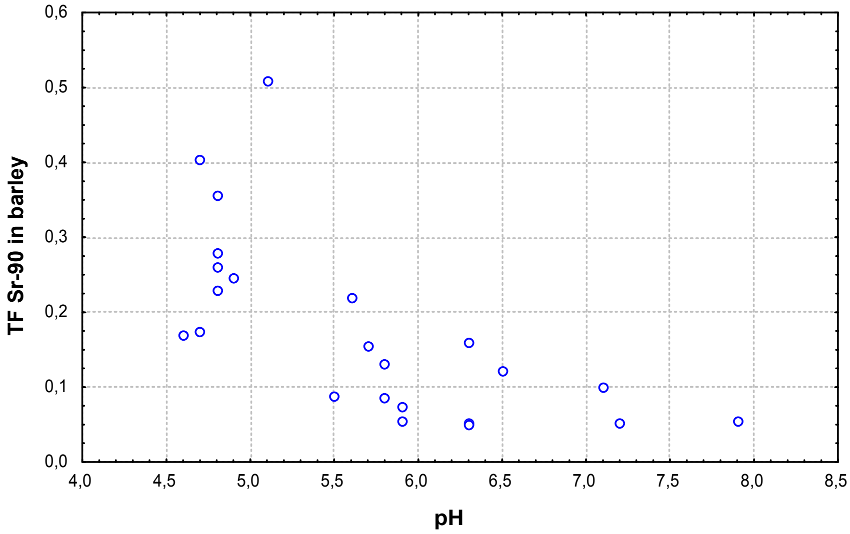

Fig. 16. The relationship between pH and the ^{90}Sr TF for barle.y

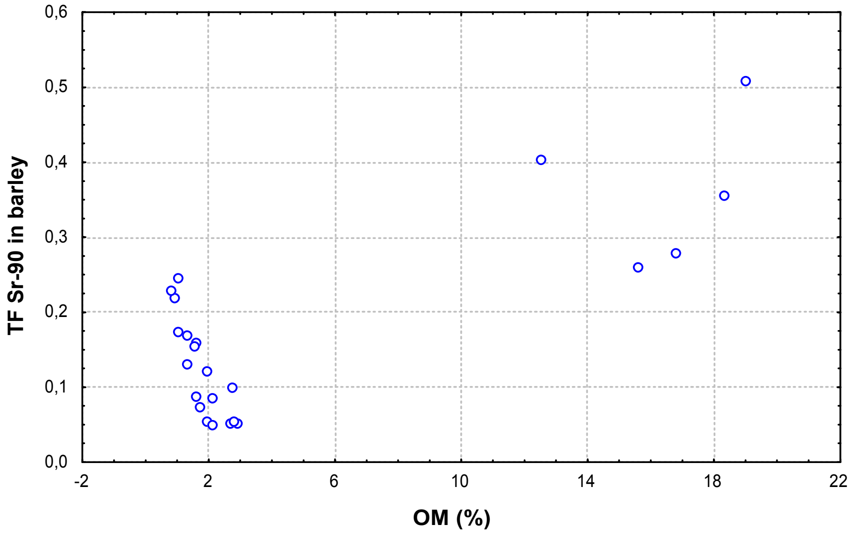

Fig. 17. The relationship between OM and the ^{90}Sr TF for barle.y

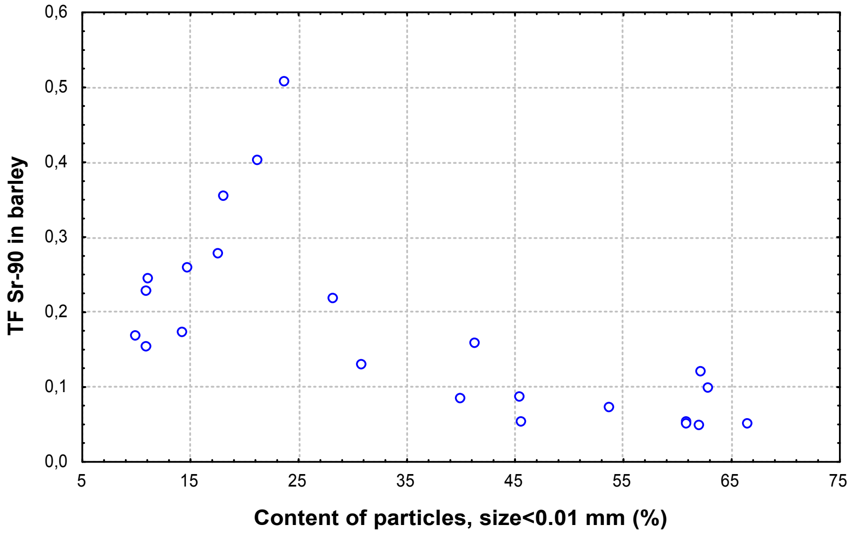

Fig. 18. The relationship between content of particles (<0.01 mm) and the ^{90}Sr TF of barley.

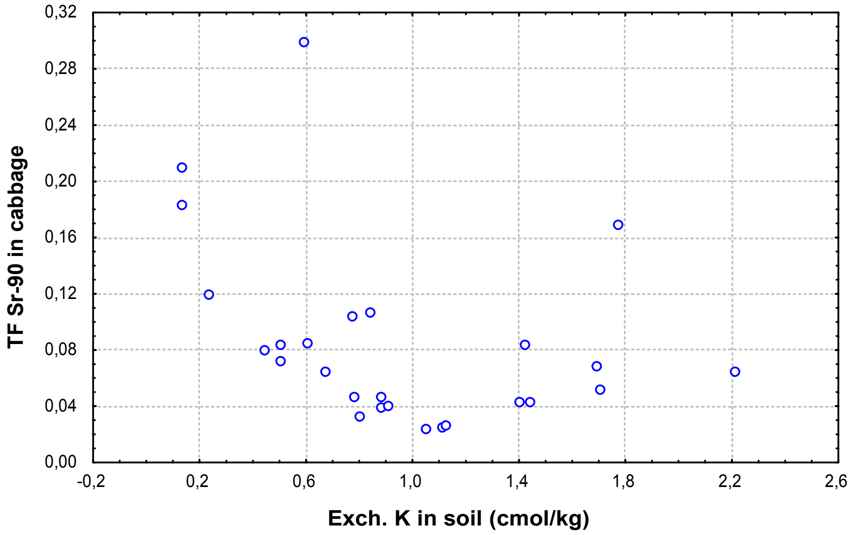

Fig. 19. The relationship between K and the ^{90}Sr TF for cabbage.

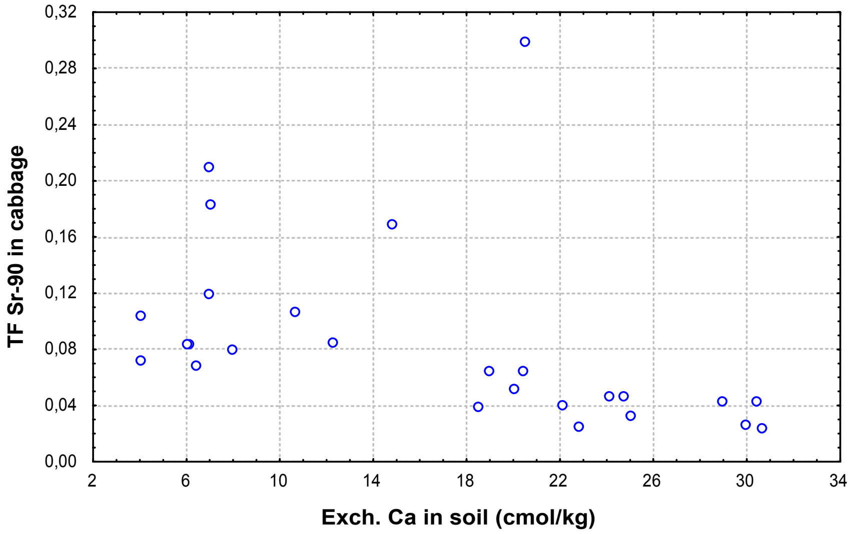

Fig. 20. The relationship between Ca and the ^{90}Sr TF for cabbage.

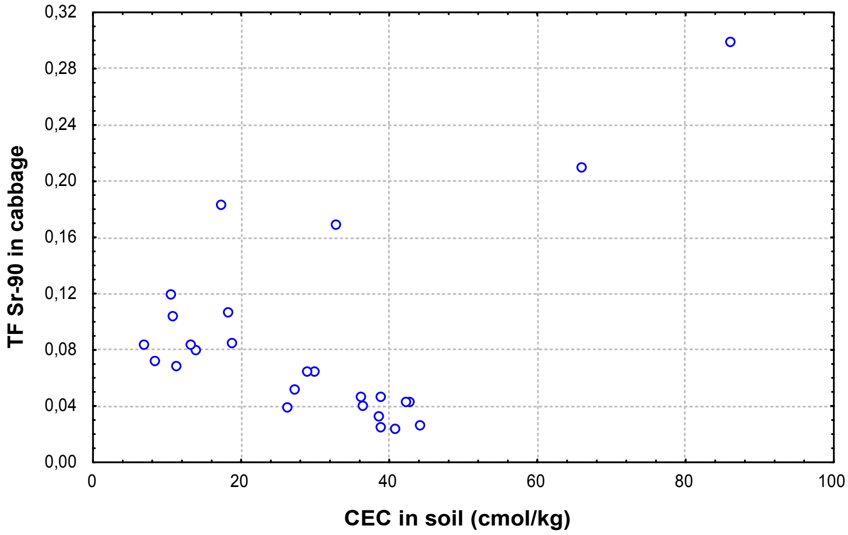

Fig. 21. The relationship between CEC and the ^{90}Sr TF for cabbage.

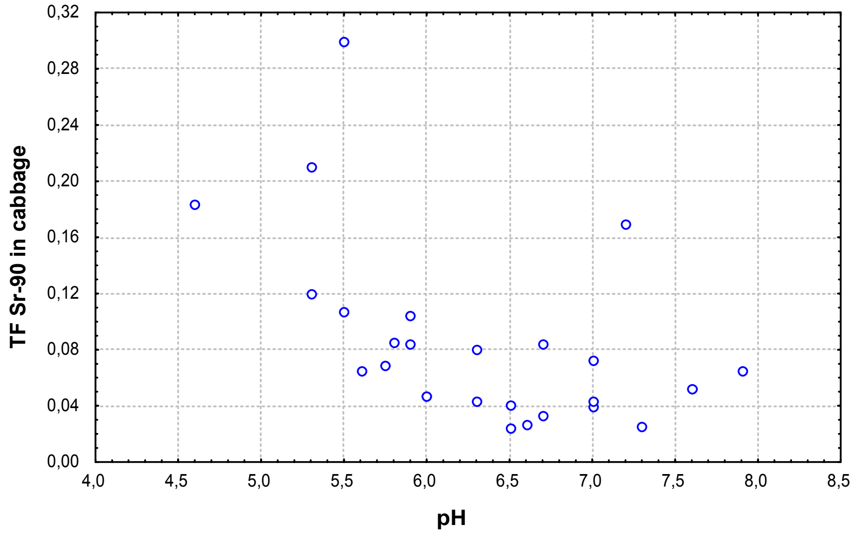

Fig. 22. The relationship between pH and the ^{90}Sr TF for cabbage.

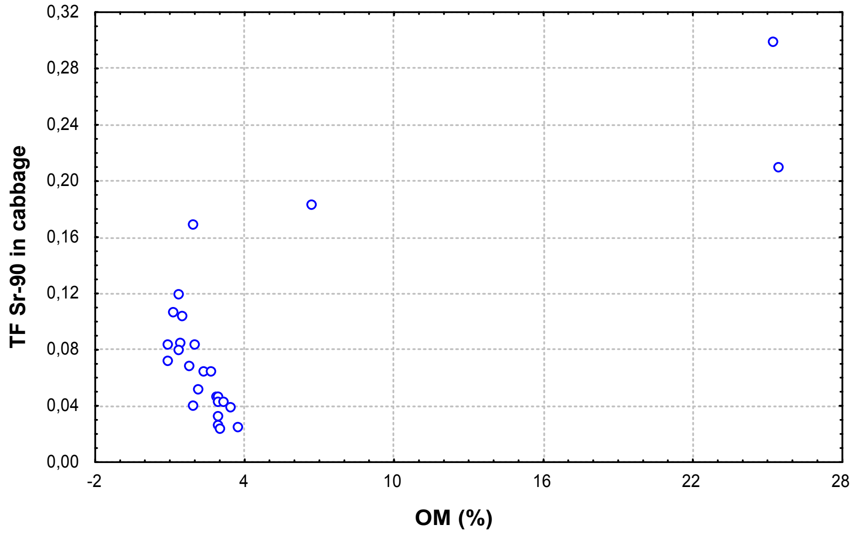

Fig. 23. The impact of OM on the ^{90}Sr TF of cabbage.

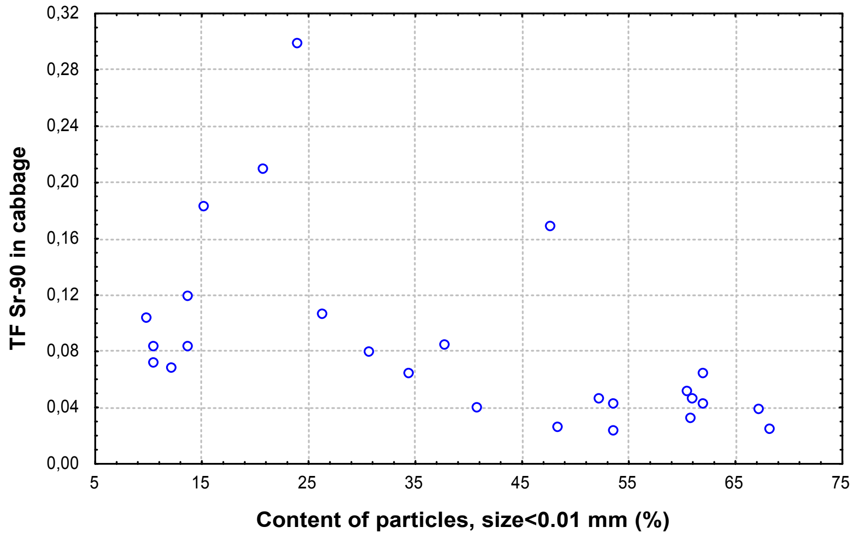

Fig. 24. The relationship between the content of particles (<0.01 mm) and the ^{90}Sr TF of cabbage.

4. IDENTIFICATION OF QUANTITATIVE RELATIONSHIPS BETWEEN RADIONUCLIDE TRANSFER FACTORS IN CROPS AND SOIL PARAMETERS

4.1. Linear regression dependencies of ^{137}Cs TFs to plants and soil characteristics

Different regression models can be used to establish regression functions. The simplest is the linear (multiple regression) model:

$$TF = a + \sum_{i=1,n} b_i X_i \qquad (1)$$

where
TF is the transfer factor; and
X_i is the *i*-th soil agrochemical characteristic.

At an early stage of the study, regression models were devised that describe relations between ^{137}Cs and ^{90}Sr to barley and cabbage TFs over the range of soil characteristics. Tables 7–8 show parameters of the regression models. Multiple correlation factors (R) are rather high for all the cases considered.

TABLE 7. PARAMETERS OF REGRESSION EQUATIONS REFLECTING RELATIONSHIPS BETWEEN ^{137}Cs TFs TO PLANTS AND SIX SOIL VARIABLES

Crop	*a*	b_i						R
		Ex. K	Ex. Ca	CEC	pH_{KCl}	OM	Clay	
Barley	0.0228	-0.0052	-0.0008	0.0000	0.0029	0.0014	-0.0003	0.72
Cabbage	-0.001776	0.0011675	-0.0005599	0.0003758	0.0013268	-0.000721	-3.713E-05	0.46

TABLE 8. PARAMETERS OF REGRESSION EQUATIONS REFLECTING RELATIONSHIPS BETWEEN ^{90}Sr TFs TO PLANTS AND SIX SOIL VARIABLES

Crop	*a*	b_i						R
		Ex. K	Ex. Ca	CEC	pH_{KCl}	OM	Clay	
Barley	0.175	-0.00092	0.00311	-0.00488	0.00453	0.019	-0.00149	0.86
Cabbage	0.150	0.015	-0.005	0.003	-0.010	-0.001	-0.001	0.84

4.2. Non-linear regression dependencies of ^{137}Cs and ^{90}Sr TF to barley and cabbage on soil characteristics

Graphical analysis of transfer factor dependencies on soil properties suggests that in some cases these relationships are non-linear. Therefore, a possibility of using non-linear models needs to be assessed:

$$TF = A \cdot X_1^{B_1} \cdot X_2^{B_2} \cdot ... X_n^{B_n} \qquad (2)$$

Linear transformation reduces this equation to the following form:

$$\ln(TF) = \ln A + \sum_{i=1,n} B_i \ln(X_i) \qquad (3)$$

To find parameters of this model, a procedure of multiple regression analysis was applied. Tables 9–10 contain parameters of the regression models.

TABLE 9. PARAMETERS OF THE REGRESSION MODELS DESCRIBING RELATIONSHIPS BETWEEN LOGARITHMS OF ^{137}Cs TFs TO PLANTS AND LOGARITHMS OF SOIL CHARACTERISTICS

Crop	$\ln A$	b_i						R
		Exch. K	***Exch. Ca***	CEC	pH_{KCl}	OM	Clay	
Barley	-2.13	-0.50	-0.03	-0.41	-0.33	0.23	-0.21	0.73
Cabbage	-8.27	-0.08	-0.77	1.13	1.33	-0.35	-0.33	0.32

TABLE 10. PARAMETERS OF THE REGRESSION MODELS DESCRIBING RELATIONSHIPS BETWEEN LOGARITHMS OF ^{90}Sr TFs TO PLANTS AND LOGARITHMS OF SOIL CHARACTERISTICS

Crop	$\ln A$	b_i						R
		Exch. K	Exch. Ca	CEC	pH_{KCl}	OM	Clay	
Barley	0.062	-0.102	-0.417	-0.217	-0.409	0.315	-0.046	0.76
Cabbage	0.005	0.137	-1.016	0.695	-1.131	-0.017	-0.037	0.76

Multiple correlation factors (R) for non-linear functions (Tables 9–10) are rather high. However, comparison of these factors with the R-values listed in Tables 7–8 demonstrates that in some cases linear regression models are preferable for describing the available experimental data.

5. DEVELOPMENT OF APPROACH FOR CLASSIFICATION OF RUSSIAN SOIL ON THE BASIS OF RADIONUCLIDE TRANSFER FACTORS FROM SOIL TO CROPS

The preliminary analysis of information contained in the database has shown a high TF variability for each vegetation species depending on the soil conditions. Therefore, the experimental data have been classified based on the soil properties. The final stage of the data processing solved the following tasks:

- identification of soil characteristics that largely influence the accumulation of radionuclides by plants and can be used for soil radioecological classification;
- development of a classification of Russian soils on the basis of radionuclide transfer factors from soil to crops.

5.1. Identification of soil characteristics, which can be used for soil radioecological classification

Correlation analysis was used to estimate the extent to which each soil characteristic is related to other soil properties and radionuclide transfer factor to plants (estimation of significance of individual relations).

Factor analysis was used to consider a whole range of relations between soil characteristics and transfer factors in order to identify of significance factors. Its main objective was to detect latent general factors that explained relations between the observed variables.

For radioecological classification the experimental data were divided into two groups, those for mineral soils and those for organic soils (peat). Organic soils (peat) were identified as an independent group, because they have some peculiarities and show rather high mobility of radionuclides.

A set of standard soil parameters is not sufficient to describe differences in organic and mineral soils in terms of influence on the radionuclide behaviour. Neglecting some factors may result in errors in developing classification systems.

5.1.1. Correlation analysis

5.1.1.1. The relationships between soil properties

The first stage was to use correlation analysis to identify the relationship between soil properties. Results for mineral soils are shown in Table 11.

TABLE 11. CORRELATION COEFFICIENT MATRIX FOR SOIL PROPERTIES

	Exch. K	Exch. Ca	CEC	pH_{KCl}	OM	Clay
Exch. K	1.00	0.54	0.55	0.64	0.33	0.56
Exch. Ca	0.54	1.00	0.97	0.49	0.69	0.82
CEC	0.55	0.97	1.00	0.50	0.70	0.82
pH_{KCl}	0.64	0.49	0.50	1.00	0.33	0.61
OM	0.33	0.69	0.70	0.33	1.00	0.58
Clay	0.56	0.82	0.82	0.61	0.58	1.00

The results from the correlation analysis can be used to identify the most significant (in terms of soil radioecological classification) variables. Soil characteristics that closely correlate with the largest number of other properties have been identified. On the other hand, attention should be paid to the soil characteristics that poorly correlate with the other properties.

Analysis of the information presented in the correlation matrix offers the following conclusions.
(i) Clay, Exch. Ca and CEC show the highest correlation among each other. These can be interchanged in soil classification.
(ii) The highest correlation coefficent (0.97) was obtained for Exch. Ca and CEC. Either of the two can be used in soil classification.

Parameters such as pH_{KCl}, Exch. K, OM do not correlate as well with other variables. These variables can be defined as most "independent" among all the soil properties.

5.1.1.2. The relationship between radionuclide TFs and soil properties

A relation was found between ^{137}Cs and ^{90}Sr transfer factors to crops and soil properties (Tables 12, 13). Table 14 shows a close correlation between TF ^{137}Cs in barley and clay content, exchangeable cations (Exch. Ca and Exch. K) and CEC. Very low correlation coefficients between TF ^{137}Cs for cabbage and soil characteristics exclude the use of the "^{137}Cs cabbage" data sample for further processing and analysis. The highest correlation factors were obtained between ^{90}Sr TF for barley and soil properties such as CEC, Exch. Ca and Clay (Table 12).

The correlation coefficient between ^{90}Sr TF for cabbage and soil properties are similar. This is consistent with correlations coefficients reported for barley. However, the use of the "^{90}Sr-cabbage" sample for factor analysis seems less effective then that the use of the "^{90}Sr-barley" sample.

TABLE 12. CORRELATION COEFFICIENTS BETWEEN ^{137}Cs TFs IN PLANTS AND SOIL PROPERTIES

Crop	Exch. K	Exch. Ca	CEC	pH_{KCl}	OM	Clay
Barley	-0.60	-0.62	-0.62	-0.40	0.01	-0.71
Cabbage	0.16	-0.17	-0.09	0.17	-0.19	-0.11

TABLE 13. CORRELATION FACTORS BETWEEN ^{90}Sr TFs IN PLANTS AND SOIL PROPERTIES

Crop	Exch. K	Exch. Ca	CEC	pH_{KCl}	OM	Clay
Barley	-0.57	-0.79	-0.81	-0.49	-0.5	-0.74
Cabbage	-0.23	-0.65	-0.55	-0.30	-0.19	-0.58

The information presented in the correlation matrix (Tables 11, 12) shows that correlation coefficients between TFs and some soil parameters have similar values. Thus other statistical procedures need to be invoked. Also it is advisable to use only values derived for barley for further processing.

5.1.2. Factor analysis

To determine the relationship between the above parameters, factor analysis was applied. Its main objective is to detect latent general factors that explain relations between the observed variables. At the first stage, a set of initial data included only agrochemical soil properties. The choice of a number of factors was based on the "screen criterion". To make use of this criterion, a plot of eigenvalues was constructed as a function of their numbers. The co-ordinate of the point at which reduction of eigenvalues is maximally retarded determines the number of factors. The factors were identified by the method of principal components. Factor rotation was performed by the Varimax method. Factor loads for two datasets incorporating soil agrochemical parameters are summarized in Table 14.

TABLE 14. FACTOR LOAD FOR A DATASET INCORPORATING AGROCHEMICAL VARIABLES OF MINERAL SOILS

Variables	Factor 1	Factor 2	Factor 3	Factor 4
Exch. K	0.288	0.321	0.100	0.895
Exch. Ca	0.870	0.147	0.344	0.249
CEC	0.860	0.164	0.362	0.249
pH_{KCl}	0.252	0.901	0.113	0.308
OM	0.400	0.118	0.902	0.091
Clay	0.805	0.418	0.191	0.178
The share of variance explained by Factor	0.408	0.192	0.187	0.177

The factor analysis made it possible to identify four factors. The maximum factor loads fall at the main factor, Factor 1, due to parameters Clay, Exch. Ca and CEC. Factor 2 is determined by the pH_{KCl} value. The maximum factor loads on Factor 3 are caused by the influence of the OM variable and on Factor 4 by the Exch. K variable.

Factor 1 is difficult to interpret because of the high factor loads of three variables. The correlation matrix (Table 11) reveals a high correlation coefficient between the variables Exch. Ca and CEC (0.97). This gives grounds to exclude one of the factors from consideration. Subsequent analysis excluded the CEC variable. This allowed unloading of Factor 1. Table 15 summarizes results of the factor analysis (factor loads) for a range of data including soil parameters and TFs of ^{137}Cs in barley.

TABLE 15. FACTOR LOAD FOR A DATASET INCORPORATING AGROCHEMICAL VARIABLES OF MINERAL SOILS AND ^{137}Cs TFs TO BARLEY

Variables	Factor 1	Factor 2	Factor 3	Factor 4
TF	-0.923	0.078	-0.167	-0.252
Exch. K	0.336	0.068	0.251	0.897
Exch. Ca	0.592	0.464	0.307	0.465
pH_{KCl}	0.189	0.065	0.950	0.200
OM	-0.001	0.979	0.066	0.058
Clay	0.616	0.226	0.577	0.398
The share of variance explained by Factor	0.288	0.207	0.237	0.214

The factor analysis identified four factors, with the maximum factor loads due to variables Clay, OM, pH_{KCl}, Exch. K. The factors can be ranked by values of factor loads due to the TF value as follows: Factor 1>Factor 4> Factor 3>Factor 2.

5.1.3. Classification systems on the basis of factor analysis

The soil variables for soil radioecological classification using ^{137}Cs as a marker radionuclide can be ranked as follows: content of clay; content of exchangeable K; pH value; and content of organic matter.

When classifying the available experimental data, it is expedient to use two variables, as using more requires a larger datasets. The factor loads due to TF on Factor 3 and Factor 4 (Table 14) differ insignificantly. Therefore, for soil radioecological classification with ^{137}Cs as a marker radionuclide, two systems of variables can be used.

Classification system 1:
soil group (organic–peat, mineral),
mechanical composition (physical clay content),
pH value.

Classification system 2:
soil group (organic–peat, mineral),
mechanical composition (physical clay content),
content of exchangeable K.

Table 16 summarizes results of the factor analysis (factor loads) for a dataset incorporating soil parameters of mineral soils and ^{90}Sr transfer factors to barley.

TABLE 16. FACTOR LOADS FOR A DATASET INCORPORATING AGROCHEMICAL VARIABLES OF MINERAL SOILS AND ^{90}Sr TFs TO BARLEY

Variables	Factor 1	Factor 2	Factor 3	Factor 3
TF	-0.831	-0.327	-0.188	-0.203
Exch. K	0.382	0.021	0.325	0.864
Exch. Ca	0.838	0.223	0.227	0.317
pH_{H2O}	0.269	0.050	0.920	0.257
OM	0.238	0.968	0.037	0.018
Clay	0.740	0.048	0.546	0.291
The share of variance explained by Factor	0.369	0.183	0.223	0.173

The factor analysis identified for factors, with the maximum factor loads due to the variables Clay, OM, pH_{H2O}, Exch. K. The factors can be ranked by values of factor loads due to the TF parameter as follows: Factor 1>Factor 2>Factor 4>Factor 3.

Significant factor loads on Factor 1 are from two variables - Exch. Ca and Clay. There is a high correlation coefficient between these variables (0.83) which suggests that the two variables are interchangeable. On this basis two systems of variables can be employed for soil radioecological classification with ^{90}Sr as a representative radionuclide: classification system 1 using soil group (organic–peat, mineral), content of exchangeable Ca, content of organic matter; and classification system 2 using soil group (organic–peat, mineral), particle size composition (physical clay content), content of organic matter.

However, there may be another interpretation of the factor analysis results (Table 15). For soil classification, soil properties can be used with the maximum factor loads on the most significant factor (Factor 1). In this case the classification system will include the following parameters: soil group (organic–peat, mineral); particle size composition (physical clay content); content of exchangeable Ca. A larger set of variables could be used for soil classification but broader classification systems require larger bodies of experimental data.

6. DEVELOPMENT OF SOIL CLASSIFICATION ON THE BASIS OF RADIONUCLIDE TRANSFER FACTOR TO BARLEY

6.1. Characteristics of data samples for ^{137}Cs and ^{90}Sr transfer factors in the classification systems developed

The characteristics of samples for ^{137}Cs and ^{90}Sr transfer factors in barley were assessed for each cluster of the classification systems. To this end, distributions of values for classification characteristics (exchangeable K, pH, etc.) were analysed. In the event of bimodal distribution, the initial cluster that contained TF values was divided into two clusters. Differences in the geometric means for transfer factors between different clusters are readily explained with current knowledge of the mechanisms responsible for radionuclide behaviour in soil. The information presented in Tables 17–21 confirms the legitimacy of all the classification systems developed. To identify the best system, requires further investigation on a larger dataset.

TABLE 17. CHARACTERISTICS OF SAMPLES OF ^{137}Cs TFs IN BARLEY FOR EACH CLUSTER OF THE SOIL TYPE – EXCH. K CLASSIFICATION SYSTEM

Soil characteristics			Transfer factor		
Soil group	Soil type	Exch. K	Geometric mean	95% conf. level	Range
Mineral soils	Clay	0.44–0.94	0.0083	0.0029	0.004–0.023
		1.1–1.94	0.0058	0.0016	0.00067–0.011
	Loam	0.34–0.4	0.026	0.018	0.012–0.043
		0.6–0.7	0.020	0.0068	0.012–0.035
	Sand	0.1–0.19	0.031	0.021	0.023–0.055
		0.31–1.07	0.024	0.018	0.007–0.055
Organic soils	Peat	0.18–0.53	0.034	0.029	0.014–0.1

TABLE 18. CHARACTERISTICS OF SAMPLES OF ^{137}Cs TFs IN BARLEY FOR EACH CLUSTER OF THE SOIL TYPE –pH_{KCL} CLASSIFICATION SYSTEM

Soil characteristics			Transfer factor		
Soil group	Soil type	pH_{H2O}	Geometric mean	95% conf. level	Range
Mineral soils	Clay	5.2–6.5	0.0075	0.0020	0.00067–0.023
		7.1–7.9	0.0051	0.0016	0.004–0.0085
	Loam	5.4–5.6	0.026	0.010	0.017–0.035
		5.8–6.9	0.021	0.010	0.012–0.043
	Sand	4.6–4.9	0.021	0.020	0.007–0.053
		5.7–5.9	0.036	0.018	0.023–0.055
Organic soils	Peat	4.8–5.1	0.034	0.029	0.014–0.1

TABLE 19. CHARACTERISTICS OF SAMPLES OF ^{90}Sr TFs IN BARLEY FOR EACH CLUSTER OF THE EXCH. Ca – ORGANIC MATTER (OM) CLASSIFICATION SYSTEM

Soil characteristics			Transfer factor		
Soil group	Exch. Ca	Organic matter	Geometric mean	95% conf. level	Range
Mineral soils	2.4–10.4	0.6–1	0.20	0.031	0.16–0.25
		1.3–6.7	0.13	0.021	0.089–0.17
	14.5–20	1.6–1.9	0.098	0.037	0.056–0.21
		2.1–2.7	0.07	0.015	0.049–0.12
	24–28	2.3–2.9	0.047	0.0093	0.047–0.064
Organic soils	5.7–22.4	12.5–19	0.34	0.077	0.26–0.51

TABLE 20. CHARACTERISTICS OF SAMPLES OF ^{90}Sr TFs IN BARLEY FOR EACH CLUSTER OF THE SOIL TYPE – ORGANIC MATTER (OM) CLASSIFICATION SYSTEM

Soil characteristics			Transfer factor		
Soil group	Soil type	Organic matter	Geometric mean	95% conf. level	Range
Mineral soils	Clay	1.6–1.9	0.098	0.029	0.056–0.21
		2.1–2.9	0.055	0.021	0.022–0.12
	Loam	0.6–0.9	0.17	0.039	0.15–0.21
		1.3–2.1	0.10	0.020	0.085–0.13
	Sand	0.8–1.3	0.20	0.037	0.17–0.25
		1.5–6.7	0.13	0.036	0.093–0.17
Organic soils	Peat	12.5–19	0.34	0.077	0.026–0.51

TABLE 21. CHARACTERISTICS OF SAMPLES OF ^{90}Sr TFs IN BARLEY FOR EACH CLUSTER OF THE SOIL TYPE - EXCH. Ca CLASSIFICATION SYSTEM

Soil characteristics			Transfer factor		
Soil group	Soil type	Exch. Ca	Geometric mean	95% conf. level	Range
Mineral soils	Clay	10.4–19.2	0.089	0.026	0.049–0.21
		20–28	0.052	0.0079	0.054–0.073
	Loam	6–9.1	0.16	0.032	0.12–0.22
		16.6–20	0.087	0.0025	0.085–0.089
	Sand	2.4–4.9	0.173	0.068	0.093–0.25
		4.9–7	0.155	0.025	0.12–0.17
Organic soils	Peat	5.7–22.4	0.34	0.077	0.26–0.51

7. CONCLUSIONS

(1) In the soil-plant system, soil properties influence the behaviour of ^{90}Sr and ^{137}Cs in various ways. The main parameter, which describes the rate of radionuclides accumulation in farm crops, is the transfer factor (TF). Therefore, the transfer factor can be used for radioecological classification of soils.

(2) Regression equations have been derived that describe the effects of soil properties on ^{137}Cs and ^{90}Sr TFs to barley and cabbage with high multiple correlation cofficients. When developing such regression models, linear relationships are preferable but it is advisable to test for non-linearity.

(3) Based on statistical (correlation and factor) analyses, ratings have been established for variables that can be used for soil radioecological classification, which reflects the rates of ^{137}Cs and ^{90}Sr uptake by crops.

(4) On the basis of the statistical analyses of data, two schemes of soil classification have been developed based on ^{137}Cs transfer factors for barley: classification system 1 using soil group (organic–peat, mineral), mechanical composition (physical clay content) and pH value; classification system 2 using soil group (organic–peat, mineral), mechanical composition (physical clay content) and content of exchangeable K.

(5) For soil radioecological classification based on the rate of ^{90}Sr uptake by plants, three schemes of soil classification have been developed: classification system 1 using soil group (organic–peat, mineral), content of exchangeable Ca and content of organic matter; classification system 2 using soil group (organic–peat, mineral), mechanical composition (physical clay content) and content of organic matter; and classification system 3 using soil group (organic–peat, mineral), mechanical composition (physical clay content) and content of exchangeable Ca.

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TRANSFER FACTOR OF CAESIUM-137 IN ARID AND SEMI-ARID REGIONS

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Abstract

Transfer factors (TFs) for ^{137}Cs were measured in two soils with a range of annual crops and olives. The values were towards the lower end of the ranges reported in the literature. Fertilization reduced TFs by 13–33%. TFs to the edible parts of arable crops were in the order cereals < broad beans, cucumber < leafy crops ≤ potato tubers. Olive oil contained a lower ^{137}Cs concentration than any of the other food products.

1. INTRODUCTION

The possibility exists of an accident that may release radioactive material into the environment. As nuclear facilities are not normally sited in densely populated areas, the adjacent land is likely to be agricultural or at least rural. Furthermore, the accident at Chernobyl in 1986 showed that agricultural practices could be affected within hundreds and even thousands of kilometers from the accident site. Therefore, contingency plans are needed to consider countermeasures that can be used to reduce contamination of agricultural products, even in countries without nuclear power.

The two main considerations in preparing an agricultural counter measures strategy are to:

- protect human health by reducing radioactive contamination of agricultural products; and
- return the land to normal usage as far as possible.

Different countermeasures are available to reduce or prevent transfer of radionuclides through the food chain. However the countermeasures vary in both effectiveness and their economic, ecological and social impact [1].

One of the countermeasures to be applied in the medium and long term is change of land use [2]. For example alternative crops could be grown that accumulate lower levels of the contaminating radionuclide than the crops usually grown in the areas under consideration. Another possibility is to grow crops such as olive trees where the edible product accumulates low amounts of radionuclides that are further reduced by processing.

There is little information on the transfer of anthropogenic radionuclides to plants and the food chain in semi–arid and arid regions, compared to humid and temperate regions. The climate in Syria varies from humid to extra–arid and soil properties also differ, depending on geological and climate conditions. It is therefore important to evaluate Syrian agricultural products as sources of internal dose by ingestion. As a part of this process the present research was carried out to determine the transfer factor of ^{137}Cs from different soils to different crops in Syria.

2. MATERIALS AND METHODS

2.1. Climate

The climate of the experimental region is arid Mediterranean type with the mean annual precipitation of 150 mm and mean temperature of 7.2°C in January and 27.2°C in August.

2.2. Soils

Two types of soils were selected:

(1) poor soil (Aridisol, Lethic Xerothents = Yermosol according to the FAO–UNESCO classification); and
(2) the major agricultural soil (Inciptisol, Calcixerolic Xerocrept = Xerosol according to FAO–UNESCO).

2.3. Field preparation and soil labeling

The experimental field was sited in the agricultural station of the Atomic Energy Commission of Syria (AECS), southeast Damascus (33° 25' longitude, 33° 21' latitude). About 2500 m^2 (50 × 40 m) was isolated as a protected area, and about 36 m^2 (6×6 m) in its centre was isolated for contamination. This area was ploughed many times to a depth of about 20 cm, to ensure soil homogeneity. Samples from five places were collected for soil analysis. Two different amounts of ^{137}Cs were each mixed with 1 g of CsCl and dissolved in water. The resulting solutions were spread over the two experimental areas after first saturating the soil with water. A surface dosimeter was used to assure the homogeneity of surface distribution. After five days, the areas were nearly dry and then ploughed five times to a depth of about 20 cm using a cultivator before samples were taken for homogeneity.

2.4. Soil and plant sampling

Plant samples were collected from the centre of each plot at harvest. Edible and vegetative parts were separated, washed with water, chopped and weighed. Sub samples were dried at 70°C for 48 h, milled to 50 mesh and then analyzed. Soil samples were also collected on the same dates as the plant samples. These were taken to a depth of 20 cm from three locations in each plot. The soil samples were air dried, passed through a 0.5 mm sieve, mixed again and analyzed.

2.5. Soil analysis

The properties of soil such as pH, electrical conductivity, particle size distribution, organic matter, cation exchange capacity (CEC), exchangeable cations, and pH were determined by standard procedures [3].

2.6. Radioactivity measurements

The milled plant and soil samples were used to measure Cs radioactivity using gamma spectroscopy where samples were counted in fixed calibrated geometry using a HPGe detector. The transfer factor (TF) was calculated as the ratio between the concentration of the radionuclide in the dry mass of the annual crops and that of the dry soil in the upper 20 cm, or 40 cm for trees.

2.7. Sowing

The radionuclides were allowed to equilibrate in soil for two months. During this period, the soil moisture was kept at over 50% of field capacity. After equilibrium, the crops were sown and fertilizers were applied in accordance with local practice.

3. RESULTS

3.1. Soil characteristics

Tables 1a,b show the physical and chemical composition of the soils. The Aridisol is a silty-loam soil and the Inciptisol is a silty-clay soil.

TABLE 1A. PHYSICAL CHARACTERISTICS OF SOILS

Soil type	Aridisol			Inciptisol		
Composition	Sand	Silt	Clay	Sand	Silt	Clay
Depth (cm)						
0-10	36.8±2.2	49.9±2.1	13.2±0.0	11.7±1.3	55.5±2.5	32.8±2.2
10-20	35.9±1.2	50.0±0.0	14.1±1.2	8.97±2.5	46.4±3.8	44.6±6.5
20-40	33.3±2.4	49.9±2.1	16.7±3.3	9.8±0.0	49.2±7.6	41.5±7.9

TABLE 1B. CHEMICAL CHARACTERISTICS OF SOILS

Parameters	Aridisol Values±SD	Insiptisol Values±SD
pH ($CaCl_2$)	7.91±0.04	8.1±0.04
CEC (meq/100 g)	20.7±0.5	38.9±2.8
Exchangeable potassium (meq/100 g)	0.86±0.05	1.18±0.11
Exchangeable calcium (meq/100 g)	12.3±0.2	19.6±0.15
Organic matter (%)	1.4±0.1	0.71±0.02
Available phosphorus (mg/kg)	5.1	6.88

3.2. Transfer factors of ^{137}Cs

3.2.1. Annual crops

Data in Tables 2 and 3 show large variations in average TF values. The TF values for cereal grains (0.001–0.0033) were substantially lower than to the edible parts of the other crops, which were 0.0075 for broad bean seeds, 0.016 for leafy crops (cabbage, alfalfa and spinach) and 0.027 for potato.

In general these TFs are towards the lower limits of the values reported for other areas [4]. This might be due to two major factors: soil properties and climatic conditions. The soils used (Table 1) have a clay loam or silty clay structure (clay of 14 and 39%) with high pH (7.9–8.1) and high exchangeable potassium (0.9–1.2 meq/100 g) and calcium (12.3–19.6 meq/100 g). These properties can explain the TFs of ^{137}Cs [5–8]. The climatic conditions in the area, with high light intensity, high temperature and low air humidity may reduce the uptake of ^{137}Cs compared with temperate and humid areas [9, 10].

TABLE 2. TRANSFER FACTOR VALUES FOR ^{137}Cs AT HARVEST (Cs CONCENTRATION IN THE SOIL ABOUT 20000 Bq/kg DW)

Crop	Aridisol Edible	Aridisol Veg.	Aridisol Ratio	Inciptisol Edible	Inciptisol Veg.	Inciptisol Ratio
Sorghum	0.0033	0.0055	1.7	0.0016	0.0037	2.3
Maize	0.0017	0.009	5.3	0.0008	0.0038	4.8
Wheat	0.0016	0.0032	2.0	0.001	0.0023	2.3
Barley	0.0016	0.0027	1.7	0.0014	0.0023	1.6
Mean for cereal	0.0021	0.0051	2.4	0.0012	0.003	2.5
Broad bean	0.0075	0.0125	1.7	0.0047	0.011	2.3
Potato	0.027	0.028	-	0.011	0.02	1.8
Cucumber	0.01	0.054	5.4	0.0074	0.046	6.2
Leafy*	0.016		-	0.011		
Unfertilized						
Sorghum	0.0046	0.0077	1.7	0.0022	0.0047	2.1
Barley	0.0022	0.0038	1.7	0.0016	0.0037	2.3
Leafy*	0.024	-	-	0.016	-	-

* throughout the season

TABLE 3. TRANSFER FACTOR VALUES FOR ^{137}Cs* (Cs CONCENTRATION IN THE SOIL ABOUT 5000 Bq/kg DW)

Crop	Edible	Veg.	Ratio
Sorghum	0.0032	0.009	2.8
Wheat	0.0018	0.01	5.6
Barley	0.0021	0.015	7.14
Mean for cereal	0.0024	0.11±0.003	4.6
Broad bean	0.0068	0.025	3.7
Cabbage	0.014	-	-
Chick pea	0.007	0.05	7.1

*All the crops were fertilized, and the soil was Aridisol.

The TF values were 75% higher from the Aridisol than the Inciptisol in the edible parts of cereals (Table 2). This may be due to the less available ^{137}Cs in the Inciptisol (60%) than that in the Aridisol. The difference in available ^{137}Cs between the Aridisol and the Inciptisol could be attributed to different clay contents, about 39% in the Inciptisol and about 14% in the Aridisol.

Fertilization of sorghum, barley, alfalfa and spinach decreased the transfer values of ^{137}Cs in both soils by 28%, 27% and 33% for Aridisol and by 27%, 13% and 31% in Aridisol for sorghum, barley and leafy crops, respectively (Table 2).

Data in Tables 4 and 5 show that at harvest plants grown on the Aridisol absorbed more ^{137}C than plants grown in the Inciptisol. The average uptake over two years of ^{137}C in the edible parts of fertilized crops were 39–421 Bq/kg DW in the Aridisol and 23–194 Bq/kg DW in the Inciptisol. For the unfertilized crops the absorbed quantities of ^{137}Cs were 27–33% higher in the Aridisol and 13–31% in the Inciptisol.

TABLE 4. Cs CONCENTRATION (Bq/kg DW) IN PLANT PARTS AT HARVEST (Cs CONCENTRATION IN THE SOIL ABOUT 20000 Bq/kg DW)

Soil type	Aridisol		Inciptisol	
Part	Edible	Vegetative	Edible	Vegetative
Sorghum	59	99	28	65
Maize	35	182	18	82
Wheat	29	59	18	42
Barley	31	52	26	44
Mean for cereal	39±14	98±60	23±5	59±19
Broad bean	177	295	100	221
Potato	421	490	194	355
Cucumber	209	1120	129	821
Leafy	303*		189*	
Unfertilized crops				
Sorghum	102	173	38	83
Broad bean	44	75	30	65
Leafy	446*		278*	

* Cs concentration throughout the season

TABLE 5. Cs CONCENTRATION (Bq/kg DW) IN DIFFERENT PARTS OF CROPS GROWN ON SOIL WITH Cs CONCENTRATION IN THE SOIL OF ABOUT 5000 Bq/kg DW

Part	Edible	Vegetative
Sorghum	20	80
Wheat	13	77
Barley	18	76
Mean for cereals	17±3.6	78±2
Broad bean seeds	55	141
Cabbage	77	-

All the crops were fertilized, and the soil was Aridisol.

A correlation was found between crop uptake and concentration of ^{137}Cs in soil (Tables 4 and 5). This suggests there is a relation between ion uptake by plant roots and ion concentration in the soil. However, Horak et al. [11] reported a non-linear correlation between ion uptake by plant roots and ion concentration in the rooting medium.

The concentration of radionuclide differed widely between various plant organs. This will obviously influence the selection crops since only certain plant parts enter the human food chain. The average transfer factors in Tables 2 and 3 were calculated for important plant species or plant group (cereal) of similar properties.

The concentration of ^{137}Cs was higher in straw than in the grain of all crops, and the activity ratios of grain:straw in barley, sorghum and wheat (1.7–2 fold) were less than in maize (5.3 fold). Our results were in agreement with those obtained by Horak et al. [11] in which there were lower grain:straw activity ratios in barley and wheat compared to maize.

There is evidence that potassium is concentrated more in vegetative parts than seeds [11]. This may account for the lower transfer factors for cereal grain than straw, since caesium behaves similar to potassium [12].

3.2.2. Tree crops (olives)

The transfer factor of ^{137}Cs to the flesh of olive fruits ranged between 0.0034–0.004, and the concentration of ^{137}Cs in the extracted olive oil was about 4 Bq/kg oil in the first fruiting year and decreased in the next season. Olives are major tree crop in the Mediterranean region, and its fruits and oil are an important foodstuff.

3.2.3 Alternative food crops

Replacement of one food crop with another is an option to maintain crop production in a contaminated area. Differences in soil-plant transfer factors and concentration factors in the edible parts may be sufficient to produce food with acceptable levels of radioactivity. Our results (Table 6) show that average (over two seasons) transfer of ^{137}Cs to different plants on the same soil was in the order: cereal (1) < broad bean (3.6) < cucumber (4.8) < leafy crops (7.6) < potato tubers (12.9) in the Aridisol and cereals (1) < broad bean (3.9) < cucumber (6.2) < leafy crops (9.2) ≤ potato tubers (9.2) in the Inciptisol. Results from areas of the former Soviet Union affected by the Chernobyl accident showed that inter-species differences in radiocaesium uptake were up to 10 fold [2]. However, olive oil is the food product with the lowest levels of ^{137}Cs.

TABLE 6. IMPACT AND EFFECTIVENESS OF CHANGING LAND USE:(Cs CONCENTRATION IN THE SOIL OF ABOUT 20000 Bq/ kg DW)

Change	*Reduction factors	
	Aridisol	Inciptisol
Broad bean to cereals	4.5	4.4
Cucumber to cereals	5.4	5.6
Leafy to cereals	7.8	8.2
Potato to cereals	10.8	8.4
Unfertilized		
Leafy to cereals	6.1	8.1
(Cs concentration in the soil of about 5000 Bq/kg DW)		
Broad bean to cereals	3.2	-
Cabbage	4.6	-
Chick pea	2.2	-

* Reduction factor = Cs concentration in alternative product/Cs concentration in original product

4. CONCLUSION

In general changes in land use through substitution of cereals and/or olive trees for other crops provide effective measures to reduce ^{137}Cs contamination in the food chain. Other measures such as the application of fertilizers are less effective but reduced the transfer factor of ^{137}Cs up to 33% in the Aridisol and 31% in the Inciptisol.

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TRANSFER OF CAESIUM-137, STRONTIUM-90 AND POLONIUM-210 FROM SOIL TO MAIZE AND BLACK CABBAGE CROPS

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Abstract

This report provides experimental information on the transfer of ^{137}Cs, ^{85}Sr and ^{210}Po radionuclides from soil to reference plants in artificially and Chernobyl contaminated soils. Two types soil were used in the artificially contaminated field experiment in both open and sheltered plots. In general, the transfer factors of the radionuclides in soil taken from a flood control area are higher than in local soil. The mean TF values for ^{137}Cs in black cabbage were 0.006, 0.004 and 0.005 in flood area, local and sheltered local soil samples, respectively. The lowest TF value of ^{85}Sr was 0.357 in lettuce at the local soil type. In contrast, the highest value was 2.9 in parsley at the flood area soil. The data indicated that atmospheric deposition is the main source of ^{210}Po in the black cabbage crop. The mean TF values of the ^{137}Cs in the black cabbage samples from Chernobyl contaminated soils were 0.356 and 0.360 in 2001 and 2002 samples, respectively. We also determined stable caesium concentrations in artificially contaminated field and Chernobyl contaminated soil samples. The result indicated that the TF values were reduced when soil stable caesium concentrations increased from 3–7 mg kg^{-1}**.** The annual effective dose from consumption of black cabbage in 2002 in the Chernobyl contaminated area was calculated to be 761 nSv per person.

1. OBJECTIVES

The objectives were: (1) to determine transfer factor (TF) values of ^{137}Cs for black cabbage, lettuce, white cabbage, maize, sunflower and parsley crops in different soil types in open plots and a sheltered plot in artificially contaminated field conditions; (2) to determine TF values of ^{85}Sr for black cabbage, lettuce, white cabbage, maize and parsley crops in different soil types under artificially contaminated field conditions; (3) to determine TF values of ^{210}Po for black cabbage in different soil types in open plots and a sheltered plot under field conditions; (4) to determine TF values of ^{137}Cs for black cabbage, chard and maize in the region at the east of the Black Sea contaminated by the Chernobyl accident; and (5) to assess the dose from the consumption of black cabbage.

2. MATERIALS AND METHODS

2.1. Artificially contaminated field experiment

2.1.1. Preparation of test area

The test site was prepared in a segregated area at the Cekmece Nuclear Research and Training Center. The experimental area (about 40 m^2) was divided two parts. One part of the field was open, whereas the other part by sheltered by a polyethylene sheet. However, the open part was covered by fish net.

Two types of soil (one flood area soil and one local soil) were collected for this project. The flood area soil sample (hereafter termed 'flood soil') was taken from the 0–10 cm layer (about 800 kg) from a flood control area in Istanbul. The local soil (about 1600 kg) removed (0–20 cm) from the experiment area. The soil samples were air dried and screened to pass through a 3 mm sieve.

2.1.2. Addition of radioactivity

The soil samples were contaminated by the addition of ^{137}Cs radionuclide at 10 kBq kg^{-1} on 15th June 1999. This radionuclide was obtained from the Amersham Radiochemical Center as CsCl in aqueous solution. The stock solution was diluted approximately for addition to soil (~70 kg soil) in a big mixer. This was repeated 10 and 20 times for flood and local soil samples, respectively. After mixing, the contaminated soil was transferred into three holes (each hole is 3 m^2 with depth 20 cm) prepared in the open (two holes) and sheltered (one hole) parts of the experimental area. About 700 kg flooded soil was placed in the hole in the open field. The local soil (about 1400 kg) was divided two parts, one part was placed in the hole in the open area and the second part placed in the other hole in the sheltered area. The homogeneity of contaminated soil samples was checked and the coefficient of variation was found to be 11%.

About 50 kg of soil was taken from both ^{137}Cs contaminated flood and local soil holes from the top 5 cm. The soil samples were dried and then contaminated by the addition of ^{85}Sr at 60 kBq kg^{-1} on 29th September 2000. The contamination of the soils was done using a direct method [1]. This radionuclide was obtained from the A/O "Techsnabexport", 109180 Moscow, Russia, as a solution of Sr(II) in HCl with concentration of 0.5 mol L^{-1}. The concentration of the radionuclide was 0.22 GBq mL^{-1}. The activity concentrations of the radionuclide were measured during the contamination procedure to provide a homogeneous soil label.

2.2. Field experiment in soil contaminated by the Chernobyl accident

Black cabbage and maize are produced on the eastern Black Sea coast, especially near Rize. The highest Chernobyl contamination in Turkey occurred in the Rize region [2]. Black cabbage, chard and maize grain and soil samples were collected from the Musadağı, Çayeli, Pazar, Işıklı and Rize stations from this region in 2001 and 2002. These stations are indicated on the map (Fig. 1).

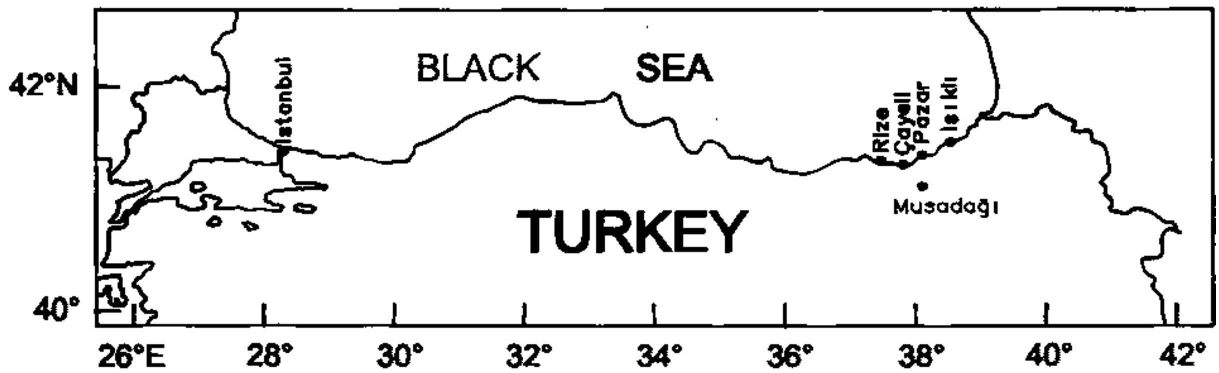

FIG. 1. Collection sites.

2.3. Radioactivity measurement and chemical analysis

The ^{137}Cs and ^{85}Sr activities in the samples were determined by a gamma-ray spectrometer. The sample powder was pressed by hand into the plastic cup of the instrument. The cup was placed directly on the detector of HPGe and its resolution (FWHM) was 1.8 keV (at 1332.5 keV for ^{60}Co). The energy calibration was done with a solid mixed gamma reference source of Amersham. The efficiency of the detector was determined using standard radioactive sources with the same geometry as the sample. In addition, the internal reference standards of the radionuclides were used to correct counting geometries and physical decays.

The measurement of ^{210}Po was made using a standard method using a standard addition of a known activity of ^{209}Po as isotopic tracer. For soil, samples were completely dissolved with mineral acids (HNO_3, HCl, H_2O_2, $HClO_4$). After evaporation, polonium was plated onto a silver disc in 0.5 M HCl in the presence of ascorbic acid. The silver discs were counted with surface barrier detectors in a multi-channel analyzer (Model BU-019-300AS).

The textural and chemical parameters of the soil samples were analyzed by soil laboratories in Istanbul and Agricultural Faculty of Ege University using standard methods [3].

The nitrate, nitrite, ammonia and phosphate levels were determined in rain and tap waters using a Hatch spectrometer. The precision of the nutrient analysis of the water samples was checked for each parameter using Strickland and Parson methods [4].

For neutron activation analysis, about 0.2 g soil samples were placed in 1 cm^3 polyethylene tubes and irradiated for 4 h at a thermal neutron flux of about 2 x 10^{13} n cm^{-2} s^{-1} in the TR-2 Reactor of Çekmece Nuclear Research and Training Center. IAEA CRM SL-1 was used as the standard. The gamma spectra of the samples and standards were measured by using a gamma-ray spectrometer consisting of a 4K analyzer (Canberra, S 85) and a high purity Ge detector (Ortec, GMX) which has a resolution of 1.9 keV at 1333 keV of ^{60}Co.

2.4. Sample preparation

Soil samples were taken at the time of harvest. The soil samples were dried at room temperature and ground to pass a 2 mm sieve. The crop leaves were rinsed under running tap water. The leaf or grain samples were dried below 85°C and were passed through a Braun laboratory homogenizer and their dry weights were determined.

2.5. Other conditions

The insecticide parathion-methyl (as the formulation Folidol M EC 360) was applied monthly throughout the summer in the field experiment according to local agricultural practice. Artificial (NPK) fertilizer was also applied twice a year in 1999 and 2000 at 100 g m^{-2} according to local application. At the same time barnyard manure was also applied twice in 1999 and 2000 at 0.5 kg m^{-2}. Following contamination, the sheltered field area was irrigated twice a week. The open field plots were also irrigated once a week if there was no rain.

On May 2002, soil was collected from the uncultivated site at Pazar in the eastern Black Sea region for determination of the vertical distribution of the ^{137}Cs. The soil cores were collected with a soil corer of radius 8 cm and length 45 cm. The cores were cut into 2 cm slices.

3. RESULTS

3.1. Artificially contaminated field experiment

3.1.1. Transfer factors of ^{137}Cs to black cabbage leaves

The TF value of ^{137}Cs radionuclide to black cabbage leaves in sheltered local soil was significantly higher than in flood and local soil samples in June 2000 but in September 2000 the TF values in flood and sheltered local soil samples are higher than in the local soil. However, the TF values in flood soil are slightly lower than those in local and sheltered local soil from October 1999 to February 2000. On the other hand, the TF values in flood soil increased after March 2000. The mean TF values were 0.006, 0.004 and 0.005 in flood, local and sheltered local soils, respectively.

The TF values of ^{137}Cs in black cabbage seed samples in June 2000 were higher from the flood soil than from the local soil. However, the value in the sheltered local soil was significantly the highest.

3.1.2. Transfer factors of ^{137}Cs for maize, sunflower, white cabbage and lettuce crops

TF values of ^{137}Cs in maize grain and straw samples from the flood soil are slightly higher than from local soil samples. On the other hand, the TF values are higher in the maize grain and maize straw samples harvested from the sheltered hole.

The TF values of ^{137}Cs in black cabbage, sunflower grain and lettuce in flood soil are higher than in local soil, whereas the TF in white cabbage leaves is highest in the local soil.

3.1.3. Transfer factors of ^{85}Sr in black cabbage, white cabbage, lettuce, parsley and maize crops

The TF values of ^{85}Sr in black cabbage and white cabbage leaves in the local soil are higher than in the flood soil. On the other hand, the TF values in lettuce and parsley leaves are higher in flood soil. The TF values of the ^{85}Sr in black cabbage in the same soil types decreased from February to April. The lowest TF value was 0.357 in lettuce and the highest was 2.9 in parsley.

The TF values of ^{85}Sr in maize were significantly higher in straw than grain while the TF of the straw sample in the local soil type is significantly lower than in the flood soil**.**

3.2. Transfer factors of ^{210}Po in black cabbage

The ^{210}Po concentration in rain and tap water samples were 0.030±0.001 and 0.0010±0.0001 Bq L^{-1}, respectively. The nutrient levels in rain and tap waters were also determined (Table 1). The TF values for black cabbage in the sheltered plot are slightly lower than those grown in the open holes. The data indicate that atmospheric deposition is the main source of ^{210}Po in the black cabbage.

TABLE I. THE NUTRIENT LEVELS IN RAIN AND TAP WATERS (mg L^{-1}).

	rain water January 2000	rain water February 2000	rain water March 2000	tap water March 2000
Nitrate	1.0	1.3	0.8	1.6
Nitrite	0.012	0.010	0.008	0.002
Ammonia	0.12	0.56	0.75	0.01
Phosphate	0.42	0.01	0.33	0.11

3.3. Transfer factors of ^{137}Cs in black cabbage, chard and maize crops in Chernobyl contaminated soils

TF values were determined in 2001 and 2002 in black cabbage and chard samples collected from the Işıklı, Çayeli, Pazar and Musadağı stations near Rize from soil contaminated by Chernobyl. The TF values of the black cabbage from Çayeli and Musadağı stations were higher than from Işıklı and Pazar. The mean TF values were 0.356 and 0.360 in 2001 and 2002, respectively. The TF value in chard was 0.146. We were not able to determine the TF value in the maize grain.

3.4. Vertical distributions of ^{137}Cs radioactivity in an uncultivated site in the Chernobyl contaminated area of the eastern Black Sea

The vertical distributions of ^{137}Cs, ^{238}U, ^{232}Th and ^{40}K radionuclides in the Pazar station in May 2002 are seen in Table 2. Approximately 75% of the total ^{137}Cs activity was present in top 10 cm of the Pazar soil in 2002.

TABLE 2. VERTICAL DISTRIBUTION OF SOME RADIONUCLIDES IN UNCULTIVATED SOIL OF THE PAZAR STATION DURING MAY 2002 (Bq kg^{-1} d.w.)

Soil section	^{137}Cs	^{238}U	^{232}Th	^{40}K
0–2 cm	273±26	ND	24±17	325±135
2–4 cm	263±18	ND	ND	229±90
4–6 cm	222±27	ND	ND	215±81
6–8 cm	266±39	ND	46±21	264±77
8–10 cm	234±34	ND	51±19	210±60
10–12 cm	126±28	ND	37±17	187±72
12–14 cm	173±25	ND	26±15	>180
14–16 cm	49±10	ND	40±20	208±81

3.5. Influence of stable caesium on accumulation of ^{137}Cs in black cabbage

The influence of stable caesium concentration in soil on TF values of ^{137}Cs in black cabbage is shown in Fig. 2. The data suggest that the TF values were decreased when soil stable caesium concentrations increased from 3–7 mg kg^{-1}.

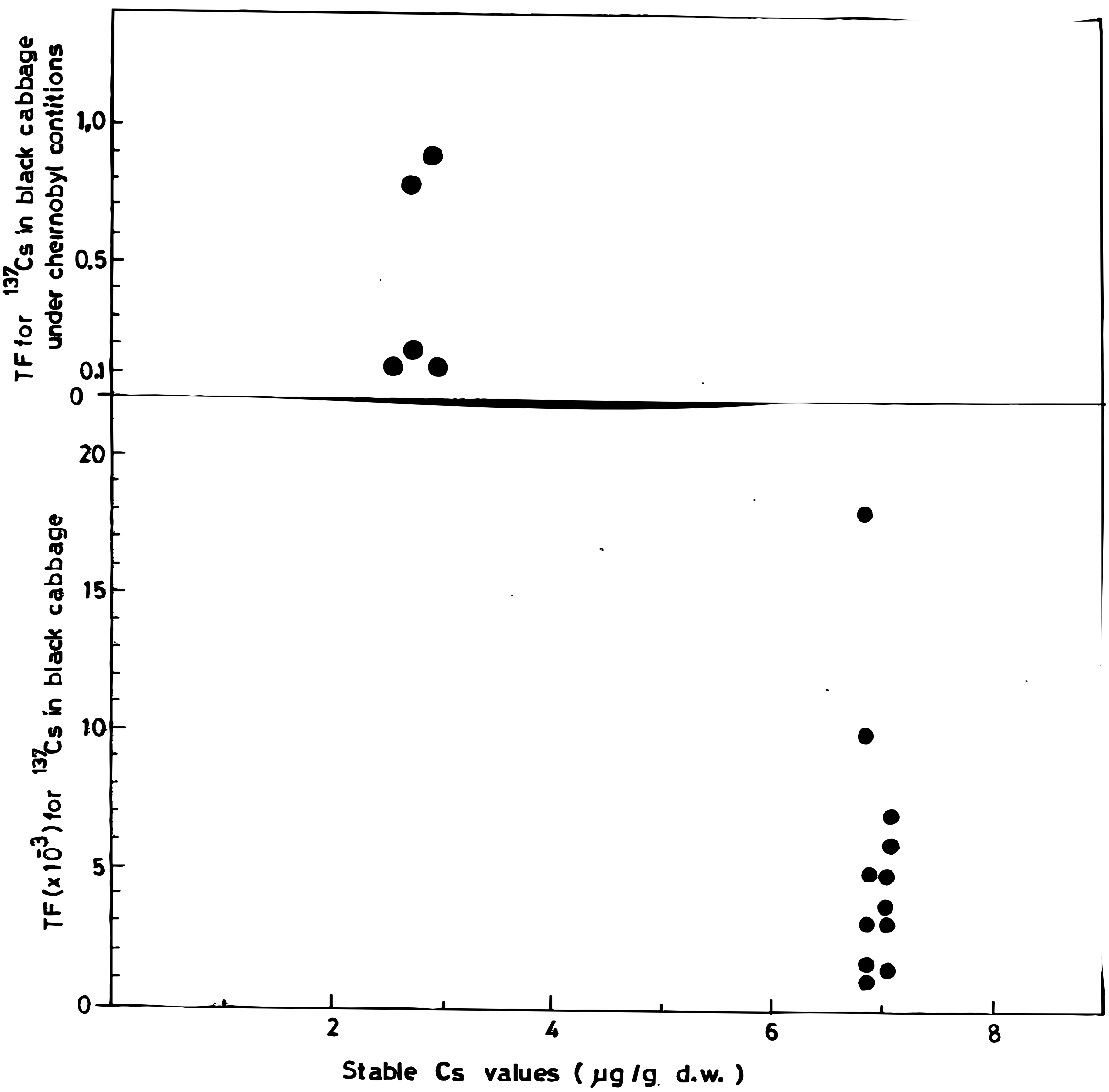

FIG. 2. The relationship between stable caesium on the TF of ^{137}Cs in black cabbage leaves.

3.6. Dose assessment

The dose from consumption of black cabbage was calculated by two different methods, using the determined activity concentrations of ^{137}Cs in soil samples (Bq/kg d.w.) for 2002 in Rize and applying TF values from the Chernobyl contaminated soils (Method 1) and also using concentrations in black cabbage samples collected from the same region in the same year (Method 2) and applying the consumption rate in the region.

3.6.1. Calculation of doses

Method 1.

The dose was calculated using the following formula:

$$D_{Cs} \text{ (Black cabbage)} = C_s\, C_f\, F_p\, F_h\, F_e\, D_f\, 1.2 \cdot\times 10^{-8} \text{ (man Sv) [5]} \quad (1)$$

where

D_{Cs} is the collective committed effective dose from ^{137}Cs by consumption of black cabbage from intake during 2002,

C_s is the ^{137}Cs mean activity of soil samples,

C_f is the mean value of the Transfer Factor of ^{137}Cs in black cabbage samples,

F_p is the production of black cabbage in the region from the regional commercial association (about 20 000 000 kg per year),

F_h is the fraction of the production which goes for regional human consumption (assumed to be 0.9),

F_e is the fraction actually eaten (assumed to be 0.8),

D_f is the delay factor between production and consumption time and is calculated to be 0.9,

$1.2 \cdot\times 10^{-8}$ is the factor used for the committed effective dose calculation [6],

and D_{Cs} (black cabbage) $= 135 \times 0.36 \times 20\,000\,000 \times 0.9 \times 0.8 \times 0.9 \times 1.2 \times\cdot 10^{-8}$
$= 7.55$ (man Sv)
$= 755$ nSv/person

Method 2.

The dose was calculated using the following formulae:

$$E_{ingk} = C_{Csk}\, H_k\, DF_{ing} \text{ [7]} \quad (2)$$

where

E_{ingk} is annual effective dose from consumption of ^{137}Cs radionuclide in black cabbage (k) for 2002 (Sv yr^{-1}),

C_{Csk} is mean concentration of ^{137}Cs radionuclide in black cabbage (k) (Bq kg^{-1} w/w),

H_k is consumption rate for black cabbage (kg yr^{-1}) from the regional commercial association,

DF is dose coefficient for ingestion of Cs radionuclide (Sv Bq^{-1})($1.3 \cdot 10^{-8}$ is factor used for the annual effective dose calculation), and

E_{ing} (black cabbage) $= 6.6 \times 12 \times 1.3 \cdot\times 10^{-8}$
$= 0.000000767$ Sv
$= 767$ nSv /person

4. DISCUSSION

The TF value of ^{137}Cs in black cabbage in the sheltered plot was significantly higher in June 2000 than those under open conditions with same or different soils. During summer the temperature in the sheltered plot about was 5ºC higher than in the open plot. In previous studies ^{137}Cs concentration, especially in vegetative plant parts, increased with increasing temperature during the growing season [8–9]. On the other hand, the TF value in flood soil is significantly higher than those obtained in local and sheltered local soil samples in February 2001. However, the TFs were lower in September 2002. This implies that the biokinetics (uptake and loss) of ^{137}Cs in black cabbage also depends on climatic conditions. The effect of temperature also showed the TFs in maize under sheltered conditions.

In general, The TF values in maize grain, maize straw, sunflower grain and lettuce are higher in the flood soil than in the local soil. However, the TF value in white cabbage of the local soil is significantly higher than that from the flood soil. It is well known that variation in growth rate and in root surface area is important for radiocaesium uptake [10]. It is also well known that the TF value depends on soil properties such as pH, exchangeable K, organic matter content, mineral composition, etc [11]. The TF values in black cabbage did not show similar trends under artificially and Chernobyl contaminated conditions. This high variation may depend on the stable Cs concentrations (Fig. 2). Furthermore, the ^{137}Cs concentrations in the Chernobyl contaminated area were lower than in the artificially contaminated field plots. Therefore it was suggested that the ^{137}Cs in the Chernobyl contaminated area was carrier-free. For this reason, the mean TF value of the ^{137}Cs in the black cabbage samples under Chernobyl conditions was found to be 60 times higher than artificially contaminated ^{137}Cs experiment with carrier. A previous study indicated that there was correlation between TF for radiocaesium and stable caesium [12]. Another study also indicated that the distribution coefficient, k_d was reduced after a small addition of stable caesium [13].

In 1986, the highest value of ^{137}Cs in black cabbage leaves was 80 Bq kg^{-1} (wet weight) from the eastern Black Sea region of Turkey [14]. In the present report, the mean ^{137}Cs concentration was 6.6 Bq kg^{-1} (wet weight). We calculated the annual effective dose from consumption of black cabbage in 2002 as 755 or 767 nSv/person. In previous studies, we also calculated the annual effective doses from tea and anchovy consumption for ^{137}Cs and ^{210}Po in 1986 and 1997 which were 0.4 and 0.2 mSv/person, respectively [15–16].

The TF values of ^{85}Sr in studied crops in the flood soil decreased in the order: parsley > black cabbage > white cabbage > lettuce > maize straw > maize grain. The TF values in the local soil crops were in the order: black cabbage > parsley > white cabbage > lettuce > maize straw > maize grain. The exchangeable calcium level in local soil was higher than in the flood soil. For this reason, the TF values in parsley and maize straw in the local soil are significantly lower than in the flood soil. On the other hand, this relationship in black cabbage, white cabbage, lettuce and maize grain samples not significant. The TF values in black cabbage in the two types soils significantly decreased from February to April in 2001. Previous studies indicated that the availability of soil ^{90}Sr to plants gradually decreased with time and also depends on the climatic conditions [8, 17].

5. CONCLUSIONS

Sixteen years after the deposition from the Chernobyl accident, the ^{137}Cs is still located in the upper soil layers (0–10 cm). The vertical migration appears to be slow. On the other hand, about 10% of the radionuclide has been transferred into plants, especially green vegetables in 2001 and 2002. At the same time, isotopic exchange had not been extensive due to the low concentration of the stable caesium (^{133}Cs). We conclude that the determination of stable Cs concentrations in soil and plant samples may necessary for the prediction of ^{137}Cs uptake.

Ingestion of radionuclides in foods is an important part of the total dose received by a person [7]. For this purpose, we calculated annual effective dose (use of TF value) by consumption of black cabbage in the Chernobyl contaminated area.

In general, soil to plant transfer factors of ^{137}Cs, ^{85}Sr and ^{210}Po were higher from the flood soil than the local soil and the sheltered local soil. The mean TF values of ^{137}Cs in black cabbage were calculated to be 0.006, 0.004 and 0.005 in flood, local and sheltered local soil samples, respectively. The lowest TF value of ^{85}Sr was 0.357 in lettuce in the local soil type whilst the highest value was 2.9 in parsley in the flooded soil type.

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THE CLASSIFICATION OF UKRAINIAN SOIL SYSTEMS ON THE BASIS OF TRANSFER FACTORS OF RADIONUCLIDES FROM SOILS TO REFERENCE PLANTS

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Abstract

Transfer factors (TFs) for ^{137}Cs and ^{90}Sr from soil to plants were measured for oats potatoes and beetroot at three sites contaminated by the Chernobyl accident over four years (1999–2002). In addition, data collected from many sites since 1987 were considered. The TF values were independent of the level of radionuclide contamination in the soil but declined with time after contamination. The decline followed a two-component model for ^{137}Cs and a one-component model for ^{90}Sr. Ratios of TFs between different crops varied from year to year but averaged over several years such reference ratios could be used to give an acceptable comparison of crops in different regions. A semi-empirical method was developed in which normalised values of soil pH, organic matter and adsorption capacity are used to characterise soils so that TF values may be compared. It is proposed that this could be the basis for a radioecological soil classification.

1. INTRODUCTION

Radionuclide transfer in a soil-plant system depends on three main groups of factors, the characteristics of the radionuclide, soil properties and biological features of plants. The problem has been studied for a long period and a large amount of information has been obtained, analysed and generalized. Much of the experimental data on the behaviour of long-lived radionuclides in food chains, including those obtained by us, has been accumulated and collected in computer databases after the Chernobyl NPP accident. Application of the Transfer Factor, TF, as an integral characteristic of radionuclide behaviour in food chains for classification of soils, is an attempt to arrange diverse data so they can be used to draw fundamental conclusions.

Classification of soils by radionuclide TF would be useful in the creation of predictive models for radionuclide transfer and accumulation in components of biocaenoses (including agrocaenoses). Our experiments in relation to this project were carried out over several years on soils in the forest-steppe ("Lesostep") zone of Ukrainian Polessje with widely varying mineral composition and agrochemical properties. The soils were contaminated with radionuclides ejected from the nuclear power plant during the Chernobyl accident in 1986. Data from our previous studies carried out in Ukraine in 1986–2001 have also been included for consideration.

Variations in experimental conditions during the project have complicated generalization of the results, i.e., different levels of soil contamination, different residence periods of radionuclides in a soil and a great variability of agrochemical and physical soil properties within the same classification group.

2. MATERIALS AND METHODS

2.1. Experimental sites

Studies were carried out in accordance with the protocols discussed and approved at Co-ordination meetings in Izmir (1999) and Vienna (2000). Field experiments were performed in accordance with local practices including crop rotation. Sites are located in three regions of the Ukraine contaminated with radionuclides of the Chernobyl accident, thus the soils have been contaminated since 1986. There are three main soil types of contrasting agrochemical properties in the experimental areas. The soils are typical for Polessje zone of Ukraine and many agricultural regions of Europe.

Site 1 is located in the Rovno region, Dubrovitsa district, vil. Miljach, where the soil is a Histosol (peat soil). The ^{137}Cs contamination varies from 130–178 kBq m^{-2}; and the ^{90}Sr contamination varies from 4.1–4.8 kBq m^{-2}. Site 2 is located in the Zhitomir region, Narodichi district, vil. Selets, where the soil is a Podzoluvisol (sod-podzolic) soil. The ^{137}Cs contamination varies from 150–227 kBq m^{-2}; and the ^{90}Sr contamination varies from 6.7–8.1 kBq m^{-2}. Site 3 is located in the Kiev region, Tarascha district, vil. Kivshovatoje, where the soil is a Chernozem. The ^{137}Cs contamination varies from 97–160 kBq m^{-2}; and the ^{90}Sr contamination varies from 8.8–9.9 kBq m^{-2}.

The variability of soil contamination between replicates reaches 25% but the contamination is distributed relatively homogeneously within each site.

The soils vary in fertility, have high and middle acidity and lack some microelements (Cu, Zn, I). Therefore these soils, especially, the sod-podzol and the peat need improvement before planting crops. Appropriate fertilisers were therefore applied before sowing. Experimental sites of 250 m^2 were divided into 36 parts for experiments performed in 1999–2002 as shown in Fig. 1.

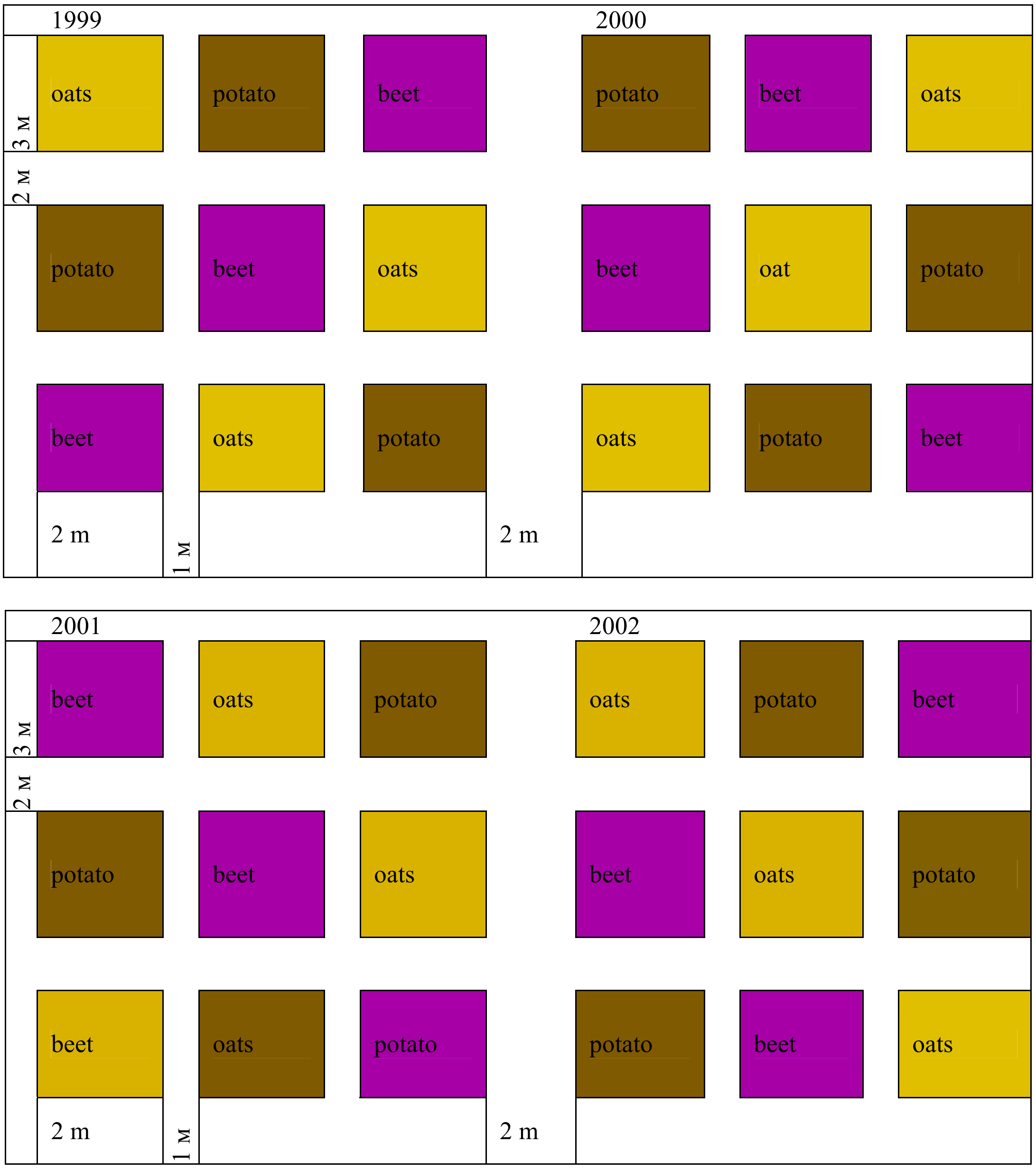

FIG.1. Scheme of experimental sites and crop rotation.

Crops and varieties were chosen which are typical for Polessje and forest-steppe zones. Seeds of oats, beet and potato seed tubers were sown manually according to local practice. The varieties of crops grown were: spring oat, variety 'Jubilee', potato variety 'Nevsky', beetroot, variety 'Pobeditel'.

2.2. Sampling

Five individual soil samples were taken on the envelope pattern from each replicate of the experiment. Soils were sampled immediately before sowing in 1999 and at harvest in the following years by soil corer (diameter 3.7 cm) to a depth of 20 cm. Samples were air-dried, passed through a 1 mm sieve and sub-samples taken for spectrometric, agrochemical and radiochemical analysis.

Crops were harvested at full maturity from the total plot area. Spectrometric and agrochemical analysis were performed on threshed oat grains, potato and beetroot tubers (both washed, cut and air-dried).

2.3. Agrochemical analysis

The agrochemical properties of the soils under investigation were determined in 1999 before experiments began and in the following years at the harvesting time. Standard methods accepted in CIS countries were used [1] as described previously [2]. Agrochemical properties of soils from three stations under investigation over three years are presented in Table 1.

TABLE 1. AGROCHEMICAL CHARACTERISTICS OF SOILS IN EXPERIMENTAL SITES IN 1999–2002, AVERAGE DATA OF NINE REPLICATES

Type of soil	Year	pH_{salt}	K_2O	P_2O_5	Ca^{2+}	Exch. base	Organic matter (%)
			mg/100 g		meq/100 g		
Peat-bog	1999	4.8±0.02	7.7±0.06	9.8±0.08	5.1±0.05	10.4±0.16	11.3±0.09*
(Histosols)	2000	4.7±0.04	12.7±0.23	11.9±0.08	5.4±0.06	10.3±0.07	12.1±0.08*
	2001	4.9±0.09	10.8±0.56	11.5±0.74	4.8±0.24	9.6±0.48	11.6±0.41*
	2002	4.7±0.07	9.3±0.48	10.8±0.69	5.2±0.06	9.8±0.24	11.8±0.13*
Average, M±m§		**4.8±0.06**	**10.1 ±0.33**	**11.0±0.40**	**5.1±0.10**	**10.1±0.24**	**11.7±0.18***
Sod-podzolic	1999	4.9±0.02	10.5±0.5	7.9±0.08	2.86±0.03	3.4±0.04	0.96±0.03
(Podzoluvisols)	2000	4.9±0.03	17.6±0.36	17.0±0.32	3.5±0.23	4.7±0.09	1.0±0.02
	2001	5.0±0.03	15.3±0.80	12.5±0.68	3.1±0.15	4.2±0.21	0.93±0.04
	2002	5.2±0.04	16.8±0.51	13.3±0.40	2.9±0.11	3.6±0.17	1.0±0.05
Average, M±m§		**5.0±0.03**	**15.1±0.54**	**12.7±0.37**	**3.09±0.13**	**4.0±0.13**	**0.97±0.04**
Chernozem typical	1999	6.6±0.05	21.3±0.63	13.4±0.40	23.9±0.54	34.3±0.67	3.8±0.05
(Chernozems)	2000	6.5±0.06	20.7±0.6	13.7±0.10	22.9±0.49	32.1±0.57	3.95±0.06
	2001	6.4±0.07	20.5±1.03	13.0±0.52	23.0±1.04	33.8±1.59	3.7±0.17
	2002	6.5±0.10	20.1±1.20	12.9±0.25	23.3±0.61	34.1±0.64	3.8±0.09
Average, M±m§		**6.5±0.07**	**20.7±0.87**	**13.3±0.32**	**23.3±0.67**	**33.6±0.87**	**3.8±0.09**

§ - mean ± 68% confidence interval
* - content of ash in organic soils (%)

2.4. Analysis of radionuclides

Radionuclide species in soils were analysed following a sequential extraction procedure [3]. ^{137}Cs content was detected by gamma spectrometry on a low-background high resolution gamma spectrometer with passive shielding and a semi-conducting detector of pure germanium GEM-30185, Ge(Li) detector GMX-series (EG&G ORTEC) with multichannel analyzers (ADCAM-300, USA; IN-1200, France). Marinelli vessels of 500 mL were used.

For ^{90}Sr analysis soil samples (weight up to 200 g) were incinerated at 500°C for four hours and treated with 6 M HNO_3 for two hours with constant stirring and heating. The soil sediment was removed by filtration, and Ca and Sr oxalates precipitated from the filtrate (stable strontium was added in advance, as a carrier). By adding an excess of oxalic acid at pH 4.2, Fe and Al oxalates stay in the solution under these conditions. The precipitate was incinerated at 900°C until carbonates were formed. The latter were left for two weeks to establish equilibrium with ^{90}Y and analysed on a low radiation beta-radiometer (Canberra-2400).

2.5. Experimental conditions and crop yields.

Fluctuations of weather during vegetation periods are presented in Table 2.

TABLE 2. METEOROLOGICAL CONDITIONS AT THE EXPERIMENTAL SITES IN 2000 AND 2002

Month	Average temperature (^oC)		Precipitation (mm)	
	2000	2002	2000	2002
Site 1 Rovno region				
May	14.7	16.5	19.6	14.9
June	17.2	17.5	49.5	59.9
July	17.2	22.2	199.3	48.9
August	18.2	19.8	20.4	29.2
September	13.9	13.4	72.5	29.4
Site 2 Zhitomir region				
May	14.4	15.5	22.6	40.6
June	17.1	17.3	31.8	56.8
July	17.5	22.6	129.4	58.7
August	18.6	18.9	16.2	68
September	12.5	12.9	47.8	111
Site 3 Kiev region				
May	15.1	16	24.8	51.6
June	16.8	17.6	39.3	160
July	17.6	22.6	110.3	30.7
August	18.9	19.4	28.5	80.3
September	14.3	13.3	61.3	61.3

Yield is the main criterion of soil fertility depending upon soil properties and climatic conditions. Rainfall and temperature during a vegetative period affect yield significantly. Oats is a crop with a short vegetation period (90–100 days) which grows well on mineral soils. Maximum yields are obtained in regions with sufficient rain during the initial period of plant development (June–July for this zone). Potato has a low water requirement at the initial stage but moisture is needed at the stages of budding, flowering and tuber formation. Peat and chernozem are the most favourable soils for potatoes. The critical vegetative stages determining yields lasts 70–80 days (May–July) for oats, 30–45 days (June–July) for potato and 80–100 days (June–August) for beet.

Rainfall in 2002 was favourable for crop growth on the 2nd and 3rd sites (Table 2). Yield of oats in the Zhitomir (sod-podzolic soil) and Kiev (chernozem) regions was 1.5–2.7 times higher, beetroot 2.2 times higher and potatoes 2 times higher than the yield of these crops grown in the Rovno (peat) region (Table 3) because precipitation rate and temperature during the maximum yield formation were optimum (Table 2).

TABLE 3. YIELD[§] OF EXPERIMENTAL CROPS OBTAINED ON VARIOUS SOILS (kg m^{-2})

Year	Type of soil		
	Peat-bog (Histosols)	Sod-podzolic (Podzoluvisols)	Chernozem (Chernozems)
Oats			
1999	0.15±0.006	0.10±0.004	0.20±0.01
2000	0.18±0.007	0.13±0.004	0.22±0.007
2001	0.23±0.009	0.15±0.008	0.28±0.01
2002	0.13±0.003	0.19±0.007	0.35±0.01
Potato			
1999	1.29±0.016	0.82±0.03	1.43±0.014
2000	1.30±0.02	1.16±0.01	1.46±0.01
2001	1.18±0.07	0.89±0.06	1.28±0.09
2002	1.06±0.01	0.91±0.02	1.93±0.017
Beetroot			
1999	1.52±0.022	0.98±0.018	1.82±0.01
2000	1.60±0.03	1.50±0.02	1.90±0.01
2001	1.45±0.08	1.18±0.07	1.90±0.01
2002	1.28±0.02	1.19±0.015	2.17±0.013

[§] - data are the mean of nine replicates ± 68% confidence interval

Research teams from 15 countries collaborated in this project. Some experimental details varied as well as the length of time the radionuclides had been in the soil. In this situation, comparison of data needs information on radionuclide binding with soil and their rate of transfer to plants. Our experiments in the project were begun in only 1999 and were carried out for four years. Some participants of the project applied soluble forms of caesium and strontium in a soil at the beginning of experiments, whereas we observed radionuclides behaviour in a soil-plant system on the 13^{th} and the following years after Chernobyl NPP fallout. To understand and compare the data it seems useful and necessary to consider in addition observations on Cs and Sr TF dynamics obtained by ourselves during 15 post-accident years in the Ukraine.

Studies of ^{137}Cs and ^{90}Sr behaviour were carried out on 31 farms from 1987 until 1991 and on 15 farms located on sod-podzolic sandy and sandy-loam, chernozem and peat-bog soils from 1991 until 2001. Hence it has been possible to analyse the dynamics of radionuclides accumulation in main crops on various types of soil. The main characteristics of typical soils in the Ukraine and Europe are presented in Table 4. It is clear that values of soil properties vary up to 2 times even within the range of a single type.

To compare our data with results of other participants of the project it is necessary to prove that TF value is not affected by variability of soil contamination. The development of a soil classification based on radionuclide TFs should take account of different periods of radionuclide-soil interaction in the experiments of various authors.

TABLE 4. DISTRIBUTION OF FARMS AND SOIL CHARACTERISTICS

Type of soil		Region	District centre	pH	Humus (%)	S (m-eq/100 g soil)	K (g/100 g soil)	Silt (%)
FAO-UNESCO	CIS classification							
Podzolu-visols	Sod-podzolic sandy	Kiev	Ivankov, Poljesskoje	4.5¬5.9	0.5¬0.9	6.2¬18.1	6.4¬8.4	5.0¬7.1
		Chernigov	Kozelets, Chernigov,	5.2¬5.6	0.6¬0.8	6.0¬8.0	4.8¬6.0	6.3¬7.2
Podzolu visols	Sod-podzolic gleic	Zhitomir	Narodichi	5.4¬6.6	0.9¬1.7	6.2¬15.9	3.2¬8.4	17.6¬19.3
Podzolu-visols	Sod-light podzolic sandy sand	Kiev	Borodjanka, Vyshgorod, Brovary, Makarov	4.7¬6.2	0.5¬1.1	6.1¬15.5	3.2¬9.2	8.2¬11.6
		Rivno	Dubrovitsa	4.5¬6.0	0.5¬0.9	6.0¬16.3	4.0¬9.2	6.2¬8.4
Podzolu visols -	Sod-light podzolic sandy	Chernigov	Repki	4.8¬5.5	0.7¬0.9	10.5¬13.2	6.0¬7.6	6.8¬7.8
Arenosols	Sod sandy sand gleic	Rivno	Sarny, Zarechnoje	5.6¬6.5	0.7¬1.2	17.1¬18.1	9.6¬10.0	13.8¬14.2
Arenosols	Sod clay sandy	Rivno	Rokitnoje	5.8¬6.7	1.0¬1.2	17.1¬18.3	8.4¬9.6	14.5¬16.1
Chernozem	Chernozem typical	Kiev	Boguslav, Baryshevka, Fastov, Jagotin, Perejaslav Khmelnitskij, Tetijev, Skvira, Tarascha, Kagarlyk, Vasil'kov	6.3¬7.4	1.8¬3.6	10.5¬28.7	15.2¬32.4	24.7¬27.2
Chernozem	Chernozem podzolised	Kiev	Kiev-Svjatoshin	6.2¬7.3	1.7¬2.7	16.3¬25.3	9.2¬20.4	25.7¬32.4
Histosols	Peat-bog	Rivno	Dubrovitsa	3.8¬5.6		2.3¬9.3	6.0¬12.6	
		Zhitomir	Narodichi	4.6¬5.4		4.0¬12.5	4.5¬13.0	

3. RADIONUCLIDE TRANSFER IN FIELD EXPERIMENTS IN ACCORDANCE WITH THE PROGRAM FOR 2000-2002)

3.1. Radionuclides transfer from various types of soil to crops

Concentrations of ^{137}Cs and ^{90}Sr in soil and crops was determined as earlier. Data on radionuclide content in components of the chain and calculated TF_{ag} and TF (accumulation) values are presented in Tables 5–7. All calculations were based on fresh weight of plants.

TABLE 5. RADIONUCLIDE TRANSFER FROM SOIL TO GRAIN OF OATS[†], 2002

Type of soil	Rep-lica	Soil contamin. kBq m^{-2}		Concn. in plants Bq kg^{-1}		TF_{ag} Bq kg^{-1} /kBq m^{-2}		TF (accumulation) Bq kg^{-1}/Bq kg^{-1}	
		^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr
Peat-bog* (Histosols)**	1	160.1	3.79	139.29	0.64	0.87	0.17	0.10	0.02
	2	130.8	4.12	119.03	0.87	0.91	0.21	0.11	0.03
	3	156.1	4.31	123.32	1.03	0.79	0.24	0.09	0.03
Average, M±m[§]		**149.0 ±3.7**	**4.07 ±0.06**	**127.21 ±2.5**	**0.85 ±0.05**	**0.86 ±0.01**	**0.21 ±0.008**	**0.10 ±0.002**	**0.03 ±0.002**
Sod- podzolic* sandy loam (Podzoluvisol)**	1	158.8	6.75	15.88	4.12	0.10	0.61	0.03	0.16
	2	149.1	7.43	17.899	3.94	0.12	0.53	0.03	0.14
	3	216.9	5.93	28.20	4.33	0.13	0.73	0.04	0.20
Average, M±m[§]		**174.9 ±8.6**	**6.7 ±0.18**	**20.66 ±1.6**	**4.13 ±0.05**	**0.12 ±0.003**	**0.62 ±0.02**	**0.03 ±0.001**	**0.17 ±0.006**
Chernozem typical* (chernozem)**	1	106.3	9.11	0.74	0.82	0.017	0.09	0.002	0.03
	2	98.5	8.31	0.79	0.58	0.018	0.07	0.003	0.02
	3	147.3	10.32	1.03	1.03	0.017	0.10	0.002	0.03
Average, M±m[§]		**117.4 ±6.2**	**9.24 ±0.24**	**0.85 ±0.036**	**0.81 ±0.05**	**0.017 ±0.001**	**0.09 ±0.004**	**0.002 ±0.0004**	**0.03 ±0.0004**

[†] - moisture content 15%
[§] - mean ± 68% confidence interval
Here and elsewhere:
* - name of soil classification accepted in CIS
**-name of soil according to FAO-UNESCO classification

^{137}Cs and ^{90}Sr soil to plant transfer factors (TF) depend on soil pH, humus, potassium and calcium contents, and biological peculiarities of crops. Increases of pH from 4.7–6.5 and potassium content in soil solution from 9.3 to 20.1 mg-eq/100 g (see Table 1) lead to decreases of ^{137}Cs accumulation in oats up to 7.2–122 times, in potatoes to 5.2–52 times and beet to 5–32 times (see Tables 5–7).

TABLE 6. RADIONUCLIDE TRANSFER FROM SOIL TO BEETROOT†, 2002

Type of soil	Rep	Soil contamin. kBq m^{-2}		Concn. in plants Bq kg^{-1}		TF_{ag} Bq kg^{-1} /kBq m^{-2}		*TF* (accumulation) Bq kg^{-1}/Bq kg^{-1}	
		^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr
Peat-bog* (Histosols)**	1	129.7	4.79	127.11	2.25	0.98	0.47	0.12	0.06
	2	152.1	3.85	138.41	1.58	0.91	0.41	0.11	0.05
	3	129.6	3.69	124.42	1.96	0.96	0.53	0.12	0.06
Average, M±m§		**137.1 ±3.06**	**4.11 ±0.14**	**129.98 ±1.75**	**1.93 ±0.08**	**0.90 ±0.02**	**0.47 ±0.01**	**0.12 ±0.002**	**0.06 ±0.001**
Soddy-podzolic* sandy loam (Podzoluvisol)**	1	153.9	7.52	29.24	8.80	0.19	1.17	0.05	0.32
	2	213.1	7.05	42.62	6.42	0.20	0.91	0.05	0.25
	3	150.1	7.51	25.52	6.68	0.17	0.89	0.05	0.24
Average, M±m§		**172.4 ±8.33**	**7.36 ±0.06**	**32.46 ±2.12**	**7.30 ±0.31**	**0.19 ±0.004**	**0.99 ±0.04**	**0.05 ±0.00**	**0.27 ±0.01**
Chernozem typical* (chernozem)**	1	101.3	8.99	3.14	3.69	0.03	0.41	0.01	0.12
	2	153.8	8.75	5.23	3.33	0.03	0.38	0.01	0.11
	3	91.6	8.05	2.93	2.98	0.03	0.37	0.01	0.11
Average, M±m§		**115.6 ±7.89**	**8.60 ±0.08**	**3.77 ±0.30**	**3.33 ±0.08**	**0.03 ±0.00**	**0.39 ±0.036**	**0.01 ±0.00**	**0.11 ±0.002**

† - moisture content of 86%   § - mean ± 68% confidence interval

TABLE 7. RADIONUCLIDE TRANSFER FROM SOIL TO POTATO TUBERS†, 2002

Type of soil	Rep	Soil contamin. kBq m^{-2}		Concn. in plants Bq kg^{-1}		TF_{ag} Bq kg^{-1} /kBq m^{-2}		*TF* (accumulation) Bq kg^{-1}/Bq kg^{-1}	
		^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr
Peat-bog* (Histosols)**	1	151.3	4.79	84.73	0.67	0.56	0.14	0.07	0.02
	2	153.8	4.05	81.51	0.49	0.53	0.12	0.06	0.01
	3	129.1	3.65	61.97	0.37	0.48	0.10	0.06	0.01
Average, M±m§		**144.7 ±2.9**	**4.16 ±0.14**	**76.07 ±2.9**	**0.51 ±0.04**	**0.52 ±0.01**	**0.12 ±0.005**	**0.06 ±0.002**	**0.01 ±0.001**
Soddy-podzolic* sandy loam (Podzoluvisol)**	1	178.8	7.15	17.88	3.22	0.10	0.45	0.03	0.12
	2	142.1	6.75	12.79	2.50	0.09	0.37	0.02	0.10
	3	156.9	7.38	18.83	3.10	0.12	0.42	0.03	0.11
Average, M±m§		**159.3 ±4.35**	**7.09 ±0.08**	**16.50 ±0.56**	**2.94 ±0.09**	**0.10 ±0.004**	**0.41 ±0.01**	**0.03 ±0.002**	**0.11 ±0.002**
Chernozem typical* (chernozem)**	1	157.9	9.25	1.58	0.54	0.01	0.06	0.003	0.02
	2	101.8	8.25	1.02	0.41	0.01	0.05	0.003	0.02
	3	91.5	8.15	0.82	0.33	0.01	0.04	0.003	0.01
Average, M±m§		**117.1 ±8.4**	**8.55 ±0.14**	**1.14 ±0.09**	**0.43 ±0.025**	**0.01 ±0.002**	**0.05 ±0.002**	**0.003 ±0.000**	**0.02 ±0.0001**

† - moisture content of 79%   § - mean ± 68% confidence interval

^{90}Sr intake into crops from soil is determined by exchangeable calcium content and, to a lesser extent, by humus content. When calcium content changes from 2.9 (sod-podzolic) to 23.3 (chernozem) meq/100 g, and humus content from 1 to 1.8%, ^{90}Sr accumulation in oats decreases from 2.9 to 6.7 times, in potatoes from 3.4 to 10 times and in beet from 2.1 to 2.5 times (see Tables 5–7)

Analysis of the data allows some general conclusions to be drawn. According to data obtained during studies carried out in 1999–2002 both ^{137}Cs and ^{90}Sr transfer depended on climatic conditions, and agrochemical characteristics of soil. It should be noted that variation of TF reaches 10–20% in all cases but does not exceed 30% of the average. That allows the data to be used for prediction and the creation of analytical functions.

Data on the dynamics of radionuclide TFs during the period of study are presented in Table 8 and show that the minimum accumulation of ^{137}Cs in potatoes and beetroot occurred in 2000 and 2002. These years are characterised by optimum values of temperature and moisture in the period vegetative growth (June–July), that lead to high yields (see Table 2 and 3) and a lower radionuclide accumulation by 1.3 times (peat), 2.8–3.0 (sod-podzolic) and 2–3 times (chernozem).

TABLE 8. CHANGES IN RADIONUCLIDE TFs WITH TIME

Years	^{137}Cs and ^{90}Sr TF (mean±SD)					
	Peat-bog		Sod-podzolic		Chernozem	
	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr
	Grain of oats (moisture 15%)					
1999	0.11±0.002	0.03±0.001	0.04±0.01	0.36±0.01	0.005±0.0002	0.06±0.002
2000	0.09±0.001	0.02±0.002	0.04±0.001	0.31±0.01	0.004±0.0001	0.05±0.002
2001	0.08±0.004	0.02±0.001	0.05±0.004	0.26±0.02	0.005±0.0004	0.032±0.003
2002	0.10±0.002	0.03±0.002	0.03±0.001	0.17±0.01	0.002±0.0004	0.03±0.0004
	Beetroot (moisture 86%)					
1999	0.15±0.001	0.04±0.001	0.11±0.004	0.28±0.01	0.03±0.0001	0.15±0.003
2000	0.11±0.001	0.06±0.002	0.04±0.002	0.32±0.003	0.01±0.001	0.14±0.003
2001	0.11±0.01	0.05±0.004	0.05±0.004	0.29±0.01	0.02±0.004	0.15±0.01
2002	0.12±0.002	0.06±0.001	0.05±0.002	0.27±0.01	0.01±0.004	0.11±0.002
	Potato tubers (moisture 79%)					
1999	0.08±0.003	0.03±0.002	0.12±0.005	0.10±0.002	0.007±0.001	0.03±0.001
2000	0.05±0.001	0.01±0.0003	0.03±0.001	0.11±0.001	0.005±0.001	0.02±0.001
2001	0.06±0.004	0.01±0.001	0.04±0.004	0.12±0.007	0.006±0.001	0.03±0.0001
2002	0.06±0.002	0.01±0.001	0.03±0.002	0.11±0.002	0.003±0.001	0.02±0.0001

A similar effect was observed for oats on chernozem in 2002 and on peat soil in 1999. Meteorological characteristics were optimum for the crop growth on peat in 1999 and on sod-podzolic soil and chernozem in 2002. Studies of radiocaesium transfer to various crops confirmed this trend.

It is clear that values of caesium TF are lower for all crops investigated on more fertile soils. Using the decrease of caesium TF to experimental plants as the criterion, soils can be put in a following order: peat>sod-podzolic>chernozem. Cs transfer is dependent on exchangeable potassium content in a soil solution and pH (see Table 1).

A different picture is observed for strontium TF. Ca and organic matter content in soil determine strontium transfer—the higher these values, the lower strontium TF figures. It should be noted that this dependence is correct at Ca concentrations below 25 meq/100 g. Based on these characteristics soils can be arranged in the following order: chernozem>peat>sod-podzolic. Precipitation rate did not affect values of strontium TF.

Crops can be put in the following order of ^{137}Cs TF: beetroot>oats>potatoes. Differences of TF are explained by biological peculiarities of crops in terms of vegetation and physiological development. As seen in Table 8 our data belong to the tail-end of curves characterising the dynamics of caesium and strontium TF.

There is a clear inverse proportional relationship between agrochemical properties and TF of ^{90}Sr and ^{137}Cs for all three crops grown on podzoluvisol and chernozem. TF values decrease in the order beetroot > oat grain >potato tubers. ^{90}Sr TF values are significantly higher than ^{137}Cs TF values. In contrast, ^{137}Cs TF values on the organic peat-bog soil are higher than those for ^{90}Sr, as the latter makes stable complexes with fulvic acids. Complex formation in a peat soil is a more important factor then soil pH in determining radionuclides migration. Probably the formation of radionuclide-organic complexes determines differences of ^{90}Sr TF between podzolic soil and chernozem.

The data permit at least two groups of soil properties determining absorption capacity to be defined, i.e. CEC, OM, pH, and complexation ability. It was shown earlier that hydrological regime (automorphous and hydromorphous) and soil particle size composition can be considered as principal characteristics. The last determines the availability of absorbing surface and its quality, and correlates with the exchangeable cations.

Evidently, the quantitative relationship between radionuclide TF value and soil properties requires the use the use of a complete estimate of soil properties accounting for the characteristics of the main phases.

4. DEPENDENCE OF RADIONUCLIDES ACCUMULATION IN PLANTS ON LEVEL OF SOIL CONTAMINATION AND TIME AFTER THE CHERNOBYL ACCIDENT

The task of this investigation is a comparative assessment of radionuclide behaviour in various types of soil under different conditions. The influence of residence time of radionuclides in soil on their availability to different crops is a subject of special interest.

The direct dependence of radionuclide accumulation in biological chains on their quantity is the paradigm of radiobiology. The dependence of ^{90}Sr, ^{137}Cs, ^{106}Ru and ^{95}Zr concentration in plants SA (Bq kg^{-1}) on their concentration in soil D (kBq m^{-2}): $SA = k \cdot D$ (1), was established by Klechkovskij et al. [4,5] and confirmed by experiments carried out under him in field conditions in the East-Ural area by Prister et al. [6].

The linearity of the $SA = f(D)$ relationship in the period after the Chernobyl NPP accident is disputed by some scientists such as Knatko [7], Grodzinskij and Gudkov [8], Gaponenko [9] in the contaminated territories of Ukraine and Belarus and Hall [10] near a NPP in Great Britain. Beresford et al. [11] summarised data on TF_{ag} in a soil-milk chain, obtained in the CIS countries at various territories remote from NPP. They concluded that TF decreased from n*E+01 to n*E-01 - as D grows from dozens to thousands of kBq m^{-2}. The problem is very important for this project because the levels of soil contamination used by various authors differ over a wide range, from 1–100 to 2000–147600 kBq m^{-2}.

However, the direct relationship $SA = k \cdot D$ has been confirmed by our data obtained over several years. The range of observations has been extended since 1987 to include a wide range of soils from automorphous to hydromorphous, including peats. Contamination of crops in the year of the accident was predominantly by aerial deposition but since 1987 by root uptake. Therefore, only data obtained since 1987 were considered. For example the $SA = f(D)$ relationship for graminaceous plants is presented in Fig. 2. Essentially the parameter k is TF, depending on type of soil, crop and period of

^{137}Cs incubation in the soil. High values of R^2 (≈0.90) for all combinations soil-crops were observed, which confirms the hypothesis of a linear relationship between SA and D [12]. Hence data on TF values obtained by all participants of the project can be compared without accounting for the level of soil contamination.

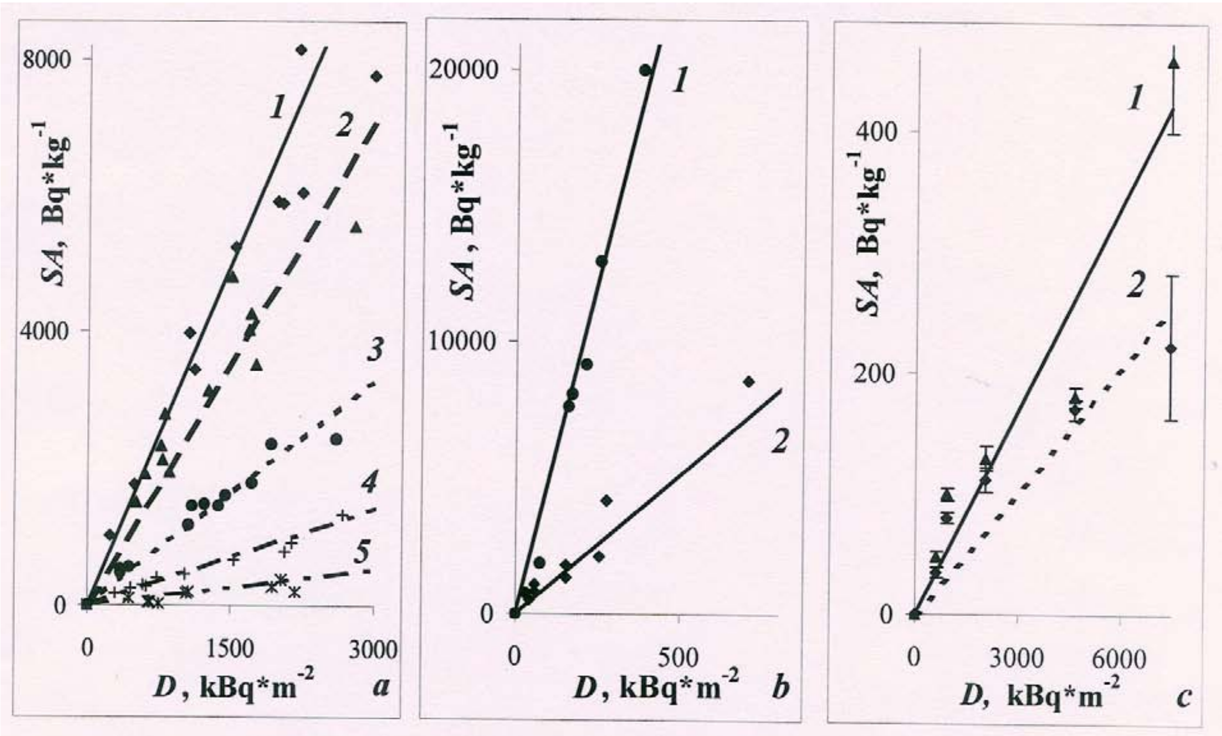

FIG. 2. Linear dependence between ^{137}Cs specific activity in grasses SA (dry weight) and density of soil contamination D.

a) in cereal grasses on a sod-podzolic soil in different years after fallout:
1-1987, 2-1989, 3-1992, 4-1995, 5-2000

b) in natural grasses on different soil: 1-peaty non-drained, 2-peaty drained

c) in *Sonchus* (*1*) and *Agropyron repens L.* (*2*) on a sod-podzolic soil in 1999

5. CHANGES IN RADIONUCLIDE TFs FROM SOIL TO PLANTS WITH TIME FROM 1987 TO 2002

Data from radiation monitoring in Ukraine were used in studies of radionuclide TF dynamics [13].

5.1. Dynamics of ^{137}Cs TF

Food and forage crops of 14 species were studied and the data show that ^{137}Cs availability to plants decreases in time. The function $TF = f(t)$ is exponential (Fig. 3 a,b) and consists of two components—quickly (*q*) and slowly (*s*) (FIG. 4) [12]. We consider TF_{ag} dynamics because it does not differ from TF but is used for the prediction of radiation state in practice.

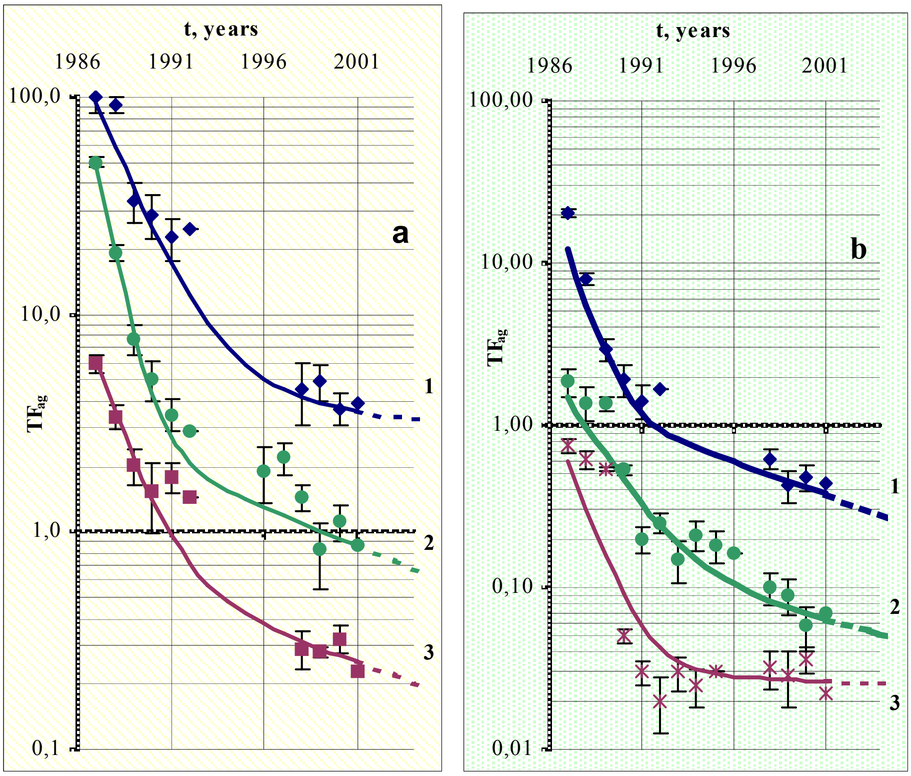

FIG. 3. The dynamics of TF_{ag} for ^{137}Cs.

a-on peat-bog soil for:
1-hay of natural grasses
2-hay of cereal sown grasses
3-beet

b-for green forage of maize on soil:
1-peat-bog
2-soddy-podzolic
3-chernozem

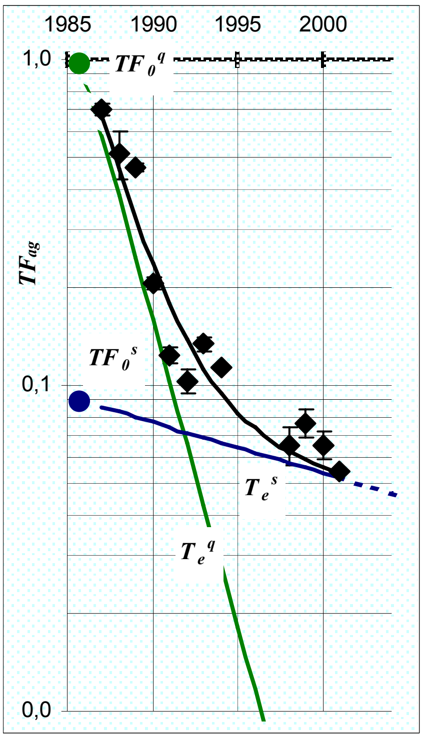

FIG. 4. The dynamics of ^{137}Cs TF_{ag} for winter wheat on sod-podzolic soil.

All relationships for ^{137}Cs can be approximated by the exponential function:

$$TF(t) = TF_0^{q+s} \cdot \left[a^q \cdot \exp\left(-0.693 \cdot \frac{t}{T_e^q} \right) + a^s \cdot \exp\left(-0.693 \cdot \frac{t}{T_e^s} \right) \right] \quad \textbf{(2)}$$

where $TF_0^{q+s} = TF_0^q + TF_0^s$ - extrapolated to 1986 (Table 9); a^q and a^s fraction of initial nuclide quantity in soil, periods of half-decrease for them T_e^q and T_e^s (Tables 10 and 11).

TABLE 9. AVERAGE VALUES OF TF_0^q AND TF_0^s FOR ^{137}Cs SINCE 1986 ($m^2\,kg^{-1}$)

Group of crops	Peat-bog		Sod-podzolic		Chernozem	
	TF_0^q	TF_0^s	TF_0^q	TF_0^s	TF_0^q	TF_0^s
			Hay			
Natural	152	4.7	28	1.8	-	-
Sown	141	2.9	6.8	0.43	3.1	0.063
Average±SD	**147±7.7**	**3.8±1.3**	**17±15**	**1.1±0.94**	**3.1**	**0.063**
			Green forage			
Maize	36	1.5	2.4	0.16	1.5	0.030
Lucerne	-	-	3.4	0.60	1.8	0.037
Clover	-	-	5.0	0.38	1.9	0.038
Average±SD	**36**	**1.5**	**3.6±1.3**	**0.37±0.20**	**1.7±0.20**	**0.035±0.004**
			Vegetables			
Cabbage	-	-	4.2	0.27	2.9	0.029
Tomato	-	-	3.5	0.15	1.5	0.030
Cucumber	-	-	3.1	0.17	2.3	0.023
Average±SD	**-**	**-**	**3.6±0.55**	**0.19±0.066**	**2.2±0.70**	**0.027±0.004**
			Roots, tubers			
Bulb onion	-	-	1.8	0.13	0.62	0.013
Beet	13	0.71	1.8	0.12	0.72	0.030
Potato	5.7	0.85	1.1	0.12	0.23	0.018
Average±SD	**9.6±5.5**	**0.78±0.098**	**1.6±0.42**	**0.12±0.009**	**0.52±0.26**	**0.020±0.009**
			Cereals			
Winter wheat	-	-	1.0	0.12	0.25	0.019
Winter barley	4.6	0.52	0.84	0.12	0.61	0.012
Winter rye	3.2	0.84	0.51	0.14	-	-
Average±SD	**3.9±1.0**	**0.68±0.23**	**0.80±0.26**	**0.12±0.012**	**0.43±0.25**	**0.016±0.005**
Scale of difference	***39***	***7.0***	***35***	***14***	***7.1***	***4.0***

Maximum differences between TFs were observed on chernozem and peat soil. Adsorption of the $^{137}Cs^+$ ion depends on the ion affinity of each adsorption site (energy of absorption) and their total capacity. Nuclides normally enter soil as water-soluble species WS, then come into equilibrium with regular exchangeable adsorbed species (RES) already present, over a period of several hours or days, a short time in relation to long-term experiments. The second stage of ion redistribution is the attainment of equilibrium with selectively bound and strongly adsorbed forms. The data of Konopljov [14] suggest that a part of the initial reduction of exchangeable nuclide in time with a period T_e^q results from *Cs* absorption on edge sites of layered clay minerals ('frayed' exchangeable species - FES). Desorption from these sites is slow and is followed by transfer to strongly adsorbing sites in the interlayers of clay minerals (high adsorbing species HAS), from where desorption is slow.

We propose that the first exponent in an equation of TF dynamics reflects the rapid transfer of initially exchangeable adsorbed ion (RES) into selective adsorbed one (FES). The second exponent reflects the slow ion transfer from selective adsorption sites FES to highly selective ones (HAS) from where remobilization is insignificant.

TABLE 10. AVERAGE VALUES OF T_e^q AND T_e^s FOR ^{137}Cs

Group of crops	Peat-bog		Sod-podzolic		Chernozem	
	T_e^q	T_e^s	T_e^q	T_e^s	T_e^q	T_e^s
			Hay			
Natural	1.4	35	1.8	12	-	-
Sown	0.69	8.7	1.8	12	1.5	8.7
Average	**1.1±0.53**[§]	**22±18**	**1.8**	**12**	**1.5**	**8.7**
			Green forage			
Maize	0.75	8.0	1.5	12	0.94	69
Lucerne	-	-	1.8	23	1.3	69
Clover	-	-	1.8	23	1.7	69
Average	**0.75**	**8.0**	**1.7±0.17**	**19±6.7**	**1.3±0.36**	**69**
			Vegetables			
Cabbage	-	-	1.4	12	0.71	69
Tomato	-	-	1.0	5.8	0.89	23
Cucumber	-	-	1.1	9.9	0.85	12
Average	**-**	**-**	**1.2±0.21**	**9.1±3.0**	**0.82±0.091**	**35±31**
			Roots, tubers			
Bulb onion	-	-	1.8	23	1.3	23
Beet	1.1	12	1.9	17	1.4	69
Potato	1.1	6.0	1.8	12	1.3	69
Average	**1.1**	**8.9±3.7**	**1.8±0.076**	**17±5.8**	**1.4±0.072**	**54±27**
			Cereals			
Winter wheat	-	-	1.4	23	0.87	35
Winter barley	1.2	12	2.3	69	0.75	35
Winter rye	1.4	12	1.3	14	-	-
Average	**1.3±0.18**	**12**	**1.7±0.54**	**35±30**	**0.81±0.080**	**35**
Average for all crops	**1.1±0.11**	**13±3.7**	**1.6±0.096**	**19±4.2**	**1.1±0.095**	**46±7.4**
Scale of difference	**2.1**	**5.5**	**2.4**	**12**	**2.3**	**8.0**

§ - average±SD

Comparison of T_e^q and T_e^s in Table 10 shows, that the rate of the second process is significantly lower then the first and there is a tendency for T_e^q to decrease in in the order sod-podzolic >chernozem > peat soil. The average rate of adsorbed ^{137}Cs transfer from RES to FES, T_e^q, is estimated as 1.3 years, and transfer from FES to HAS, T_e^s, 29 years.

The fraction of exchangeable adsorbed available ^{137}Cs initially, a^q, is 0.91–0.96 on average, i.e., the initial content of exchangeable adsorbed species is about 10 times higher than the selectively adsorbed one (Table 11). Variation of ^{137}Cs distribution between these species does not depend significantly on type of soil but the value of a^s for rye on peat and soddy-podzolic soil is significantly lower than for other crops, i.e., from 0.82 to 0.67, possibly due to experimental variation.

TABLE 11. AVERAGE VALUES OF a^q FOR ^{137}Cs

Group of crops	Peat-bog	Soddy-podzolic	Chernozem
		Hay	
Natural	0.97	0.94	-
Sown	0.98	0.94	0.98
Average	**0.98±0.007**[§]	**0.94**	**0.98**
		Green forage	
Maize	0.96	0.94	0.98
Lucerne	-	0.86	0.98
Clover	-	0.93	0.98
Average	**0.96**	**0.91±0.044**	**0.98**
		Vegetables	
Cabbage	-	0.94	0.99
Tomato	-	0.96	0.98
Cucumber	-	0.95	0.99
Average	**-**	**0.95±0.010**	**0.99±0.010**
		Roots, tubers	
Bulb onion	-	0.93	0.98
Beet	0.95	0.94	0.96
Potato	0.87	0.90	0.93
Average	**0.91±0.057**	**0.92±0.080**	**0.96±0.025**
Cereals			
Winter wheat	-	0.90	0.93
Winter barley	0.90	0.88	0.98
Winter rye	0.79	0.79	-
Average	**0.85±0.078**	**0.86±0.059**	**0.96±0.035**
Average for all crops	**0.92±0.026**	**0.91±0.012**	**0.97±0.006**
Scale of difference	***1.2***	***1.2***	***1.1***

[§] **-** average±SD

The ratio of easily to difficultly available species of ^{137}Cs reaches equilibrium after more then five T_e^q periods, i.e., the contribution of these species to plant uptake becomes comparable in 3–5 years after contamination and nuclide accumulation from FES centres predominates after 5–6 years and determines the radiation situation. Probably the contribution of the physical decay of ^{137}Cs ($T_{1/2}$=30 years) to the T_e^q value is too small to be taken into account but T_e^s values are comparable with $T_{1/2}$. Values of T_{ef}^s calculated as $T_{ef}^s = \frac{T_{1/2} \cdot T_e^s}{T_{1/2} + T_e^s}$ are presented in Table 12.

TABLE 12. AVERAGES OF T_{ef}^{s} FOR ^{137}Cs

Group of crops	Peat-bog	Sod-podzolic	Chernozem
		Hay	
Natural	16	8.3	-
Sown	6.7	8.3	6.7
Average	**11±6.6[§]**	**8.3**	**6.7**
		Green forage	
Maize	6.1	8.3	21
Lucerne	-	13	21
Clover	-	13	21
Average	**6.1**	**11±2.7**	**21**
		Vegetables	
Cabbage	-	8.3	21
Tomato	-	4.8	13
Cucumber	-	7.4	8.3
Average	**-**	**6.9±1.8**	**14±6.4**
		Roots, tubers	
Bulb onion	-	13	13
Beet	8.3	11	21
Potato	5.2	8.3	21
Average	**6.8±2.2**	**11±2.4**	**18±4.6**
		Cereals	
Winter wheat	-	9.5	16
Winter barley	8.3	13	16
Winter rye	8.3	21	-
Average	**8.3**	**14±5.9**	**16**
Average for all crops	**8.5±1.4**	**11±1.1**	**17±1.3**
Scale of difference	***3.1***	***4.3***	***3.1***

[§] - average±SD

5.2. Dynamics of ^{90}Sr TF.

The dynamics of ^{90}Sr availability for plants differs from that of ^{137}Cs to some extent in that TFs changed more slowly in the early years. Reliable conclusions on ^{90}Sr TF dynamics in the 15 years after the accident were only possible when the tendency to decrease was established. For example, data on ^{90}Sr TF from chernozem to four crops averaged for six farms are presented in Table 13. As for ^{137}Cs, the error *m* for ^{90}Sr TF is equal to 10–20% of the average and does not exceed 30%. This allows the use of TFs for prediction and creation of a soil classification. Analogous information is available for peat and sod-podzolic soils.

^{90}Sr TF changes with time are shown graphically for potatoes and winter wheat in FIG. 4. The relationship $TF = f(T)$ can be approximated, as for caesium, by an exponential function (R^2>0.8). The half-decrease period T_e values are shown in the Table 13. They range from 6.2 years for potatoes to 24 years for tomatoes and are close to 12 years on average from long-term observations (15–20 years or more).

TABLE 13. DYNAMICS OF AVERAGE ^{90}Sr TF VALUES FROM CHERNOZEM, Bq kg^{-1} AIR-DRIED MATTER/Bq kg^{-1} AIR-DRIED SOIL (AVERAGE OF 6 FARMS)

Year	Years after accident,	**Potato**		Tomato		Cabbage		Winter wheat	
		TF	±m[§]	*TF*	±m	*TF*	±m	*TF*	±m
1987	1	0.090	0.015	0.39	0.031	1.1	0.21	0.11	0.016
1988	2	0.13	0.025	0.37	0.019	2.2	0.22	0.083	0.010
1989	3	0.98	0.019	0.37	0.018	0.79	0.055	0.073	0.007
1994	8	0.073	0.007	0.43	0.035	1.7	0.92	0.073	0.018
2000	14	0.023	0.006	0.24	0.029	0.44	0.065	0.053	0.012
2001	15	0.027	0.006	0.28	0.020	1.4	0.95	0.027	0.003
T_e (years)		6.2		7.8		24		9.8	

§ - 68% confidence interval

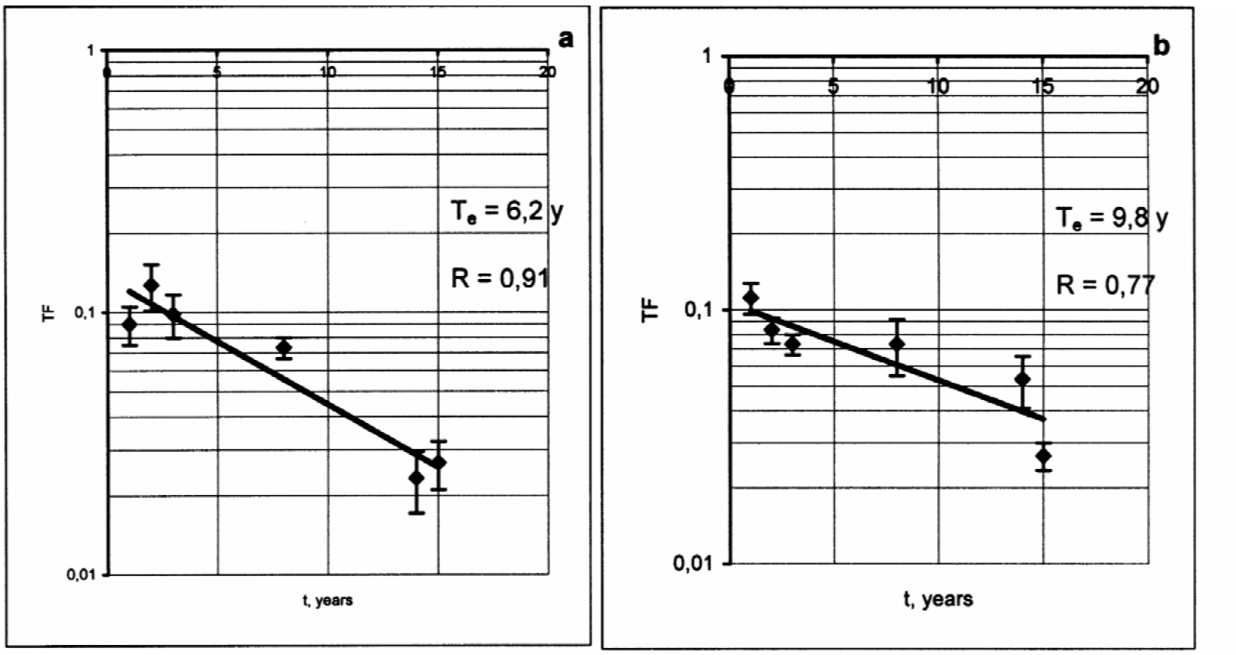

FIG. 4. Dynamics of ^{90}Sr TF (m^2 kg^{-1}) to agricultural crops grown on chernozem.

a - potatoes, b - winter wheat

Differences between ^{137}Cs and ^{90}Sr fixation rates in soil are the result of specific mechanisms of ^{137}Cs adsorption. Thus, comparison of caesium and strontium TFs should take account of the period of radionuclide incubation in the soil. It should be noted that accuracy of estimation of parameters determining the rate of strong selective absorption ought to increase as the period of observations approaches T_e.

6. RATIO OF RADIONUCLIDE TFs FOR DIFFERENT CROPS

As noted in the Introduction, TF values estimated for a single crop can not be used for comparing radionuclide uptake for the whole range of world soils. We tried to study the ratio between ^{137}Cs and ^{90}Sr TFs in various crops using the database of radionuclide behaviour collected after the Chernobyl NPP accident.

It has been shown above that TF values decreases exponentially with time. For ^{90}Sr the decrease follows a single component curve and that for ^{137}Cs follows a two-component curve. It is reasonable to suppose, that absolute TF values depend on crop species, and changes represent alterations of radionuclide availability in the soil solution. Therefore, the ratio between TFs of two crops should be constant in time within experimental error. Analysis of T_e values for all crops (TABLE 14) supports this suggestion. Some of the differences of T_e can be considered as experimental error caused by many factors, such as variation of experimental conditions in different regions, heterogeneity of radionuclide distribution in soil, changes in plant varieties and so on.

TABLE 14. AVERAGE VALUES OF CONVERSION RATIOS OF TFs OF ^{137}Cs AND ^{90}Sr OF AGRICULTURAL CROPS TO GRAIN OF WINTER WHEAT ON DIFFERENT SOIL TYPES (RATIOS OF TF VALUES OBTAINED IN EXPERIMENT AND FITTED BY EXPONENTIAL MODEL OF TF DYNAMICS)

Crops	Chernozem		Grey forest		Sod-podzolic	
	experimental	fitted	experimental	fitted	experimental	fitted
			^{137}Cs			
Beet	4.0	2.9	1.4	1.3	2.5	1.8
Cabbage	4.6	2.9	2.1	2.1	3.1	2.6
Potato	1.2	1.0	1.4	1.3	1.6	1.2
			^{90}Sr			
Beet	3.9	3.8	2.1	2.3	2.2	1.9
Cabbage	1.5	1.6	1.6	1.5	2.6	2.1
Potato	1.7	1.3	1.4	1.3	1.1	1.2

Ratios between TFs for several crops and TF to grain of winter wheat are presented in Fig. 5 and in Table 14. The directions of TF deviations for each crop change in different years, hence ratios between TF of two crops vary significantly, up to 3 times. Average ratios of TF for these crops over the period 1987–2001 vary up to 20% from average. Variability the of average ratios calculated with the exponential model is significantly lower than those obtained with raw experimental data.

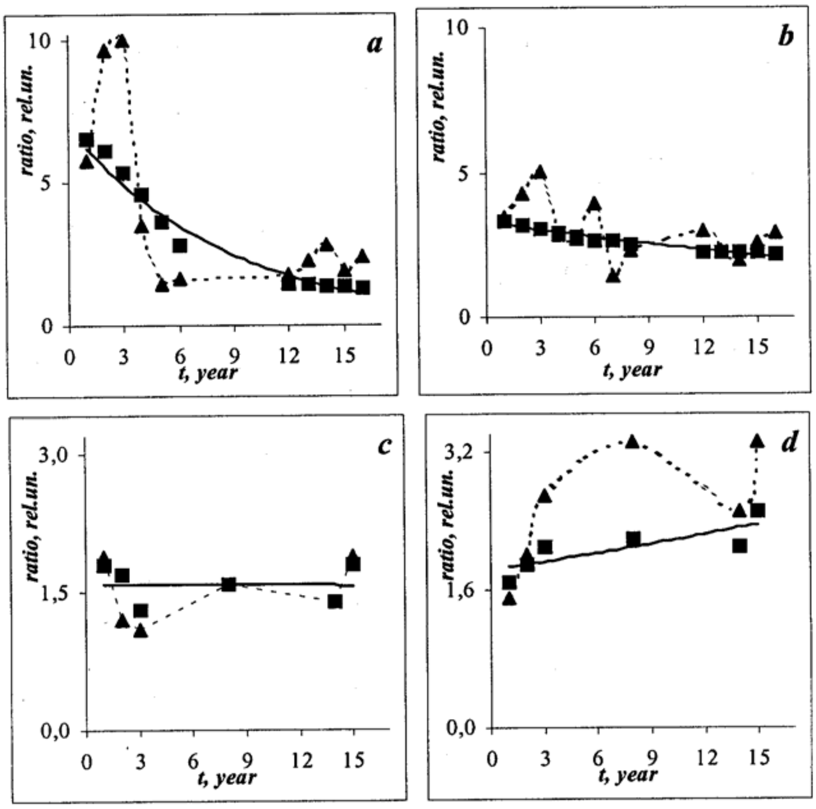

FIG. 5. Ratios of ^{137}Cs (a, b) and ^{90}Sr (c, d) TFs of heads of cabbage to grain of winter wheat on different soil types

a, c - chernozem

b, d - sod-podzolic soil

These data allow the conclusion that reference ratios between ^{90}Sr and ^{137}Cs TFs for different crops on the same soil can be used for combining the data obtained in various regions. Thus, TF ratios of ^{90}Sr and ^{137}Cs for various crops on the same soil permit comparison of the data obtained in different regions and with various crops adapted to local conditions. Accuracy of determination of TF reference ratios can be improved significantly using averages of ratios obtained over several years.

7. DEPENDENCE OF RADIONUCLIDE TF ON SOIL PROPERTIES

The relation between TF and soil properties has to be established to develop a soil classification based on TF values. This problem cannot be resolved without the complete quantitative estimation of soil properties (CESP) influencing radionuclide accumulation in plants. The data in Table 1 show the complexity of the problem. Each agrochemical characteristic varies in a wide range within a single type of soil depending on its location in the landscape and other factors. Even for such contrasting soils as chernozem and sod-podzol ranges of pH values overlap significantly.

Analysis of the large amount of data on radionuclide TFs accumulated after the Chernobyl accident in sub-project SP-3a of the Franco-German Initiative 'Database Chernobyl', has shown that the relationship between ^{137}Cs TF and soil properties is largely controlled by a few characteristics — pH of soil solution, cation adsorption capacity (CEC), organic matter content (OM), exchangeable K and Ca.

Sorption is an initial process of controlling the distribution of ions between the solid and liquid phases of soil. The adsorption of macro-quantities of ions depends on the capacity of the soil absorption complex (SAC) being filled so that ions, compete for adsorption sites. Klechkovsky, the founder of Soviet radioecological school, noted the specific character of radionuclide absorption at extremely low concentrations was in contrast to the classic theory of soil absorbing capacity. He proposed and demonstrated experimentally that at very low concentrations ions do not compete for adsorption sites because there are sufficient free sorption sites for total absorption of radionuclides even with very low absorption capacity [5].

Cation exchange between soil solution and soil can be described by:

$$SAC - M + m \Leftrightarrow SAC - m + M \qquad (3)$$

Only that part of radionuclide or nutrient ions which can desorb from the solid soil phase is available to plant roots because uptake of ions directly from adsorption sites (contact exchange) may be discounted as shown Sutkliff. Therefore, radionuclide uptake by plant roots is considered to be a function of ion flow from adsorption sites into soil solution.

De-sorption from SAC and, hence, a micro-component concentration in solution $[m]$ depends on a macro-component concentration $[M]$ and equilibrium constant K_e:

$$[m] = K_e \cdot \left[\frac{(SAC - m)}{(SAC - M} \right] \cdot [M] \qquad (4)$$

The amount of the absorbed macro-component $(SAC - M)$ does not change practically in an exchange process:

$$[m] = K_e \cdot (SAC - m) \cdot [M] \qquad (5)$$

Analysis of (5) shows that a micro-component distribution between SAC and a solution depends on the concentration of a macro-component $[M]$ competing with a micro-component for surface adsorption site. Therefore, radionuclide behaviour in a soil depends on the ion composition of macro-components and their distribution between solid and liquid phases.

The basis of the complete estimation of soil properties (CESP) is that soil can be considered as a three-phase system, solid (mineral), liquid (soil solution) and an intermediate quasi-crystalline or quasi-liquid phase (organic matter and its complexes with mineral matter).

What parameters and how many of them should be used for CESP? Adsorption capacity (E) includes the total number of sorption sites on both mineral and organic colloids but probably should be considered as mostly significant property of the solid phase. Extent of base saturation, sum of adsorbed bases (SAB) and and their cationic composition are important. SAB is determined in most experiments and is practically equal to the absorption capacity.

Organic matter (OM) influences soil structure, total sorbing surface and colloid fraction content. It also has a capacity for specific absorption and can form low solubility or weakly dissociating complex compounds. Hence, OM content should be regarded as the second important determinant of ion distribution between soil phases.

Soil solution reaction (pH) is an integral part of the soil-solution equilibrium system and can be regarded as the third important factor controlling ion behaviour in the soil.

Root uptake of a substance is proportional to its concentration in the soil solution. At ionic equilibrium the sorption (fixation) rate V_s and de-sorption (remobilization) rate V_d are equal and the fraction of ions de-sorbed (W) constant (6).

$$W = \frac{V_d}{V_s + V_d} = const \qquad (6)$$

Parameter W is thus the fraction of an ion in soil available for uptake by plants. Desorption of ions F_d is proportional to de-sorption rates of different reaction surfaces and their relative contributions to the total number of sorbing sites Sk_{di} (7)

$$F_d = \sum_{i=1}^{n} N_{si} \cdot k_{di} \cdot Sk_{di} \qquad (7),$$

where: N_{si} – equilibrium amount of ions sorbed on $i-th$ type of absorption sites in SAC;
Sk_{di} – partial sorption area of SAC, where de-sorption is characterised by a rate constant k_{di}.

F_d determines an ion concentration in soil solution and is proportional to $\sum Sk_{di}$ at equilibrium and other equal conditions. We propose to consider Sk_{di} as effective part of the exchange reaction depending on $i-th$ soil properties for a particular radionuclide.

Experimental estimation of $\sum Sk_{di}$ is very complicated therefore we have considered a simple empirical method. In a three-dimensional space (FIG. 6) where the dimensions are presented by inter perpendicular vectors of soil properties determining the completeness and tightness of ion sorption, the effective section of reaction for $i-th$ soil can be represented by area Sef, of the triangle with apexes lying in points respective to the vectors on axes pH_i, OM_i and E_i or SAB_i. At equilibrium the fraction of ion sorbed by various soils would be proportional to sorbing surface of solid phase as given by the effective section Sef_i. To make this model easier to understand, the analogy of the capture reaction section of an atom nucleus for a bombarding nuclear particle, e.g., a neutron, by may be used: the probability of particle capture by the nucleus is measured by an area of nuclear reaction section presented in barns with units of cm^2.

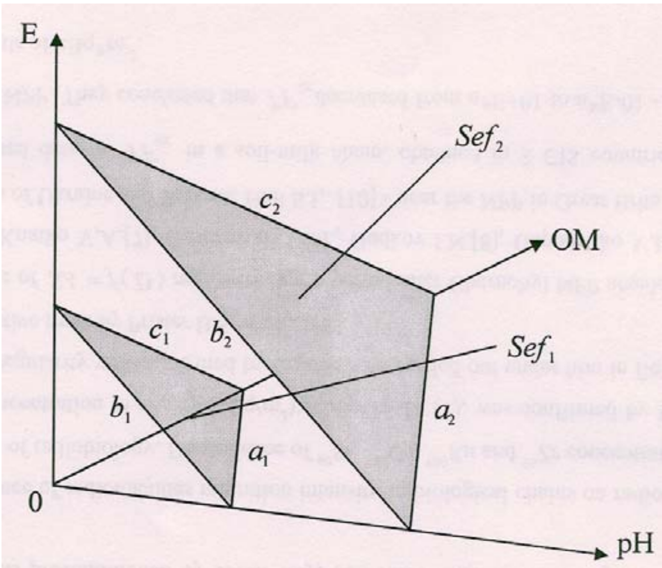

FIG. 6. Graphic presentation of the way of obtaining cesp as the area of section of three-phase system in three-dimensional space: SEF_1-sod-podzolic soil; SEF_2 – Chernozem where

E – absorbing capacity
OM – organic matter
pH - soil acidity
Sef – effective section of ion exchange
1 – sod-podzolic soil
2 - chernozem

The parameters used for Sef estimation have different units; consequently their values should be normalised. pH value is normalised by the number corresponding with a neutral reaction of soil solution pH=7; OM and SAB are normalised by the highest values of the range of soils under study, i.e. 6% and 40 meq/100 g of soil, respectively. The principle of normalising allows the data of various authors to be compared If soil characteristics exceed fixed values the normalised value of the parameter would be over 1. Sef is measured in squares of relative units.

Figure 6 shows the soil presented as a three-phase system in three-dimensional space. The area of section (triangle) is determined as follows:

$$Sef = \sqrt{p(p-a)\cdot(p-b)\cdot(p-c)} \qquad (8)$$

where p – half-perimeter of triangle $p = \frac{a+b+c}{2}$;

$a = (pH;OM) = \sqrt{N^2 pH + N^2 OM}$;

$b = (pH;E) = \sqrt{N^2 pH + N^2 E}$;

$c = (OM;E) = \sqrt{N^2 OM + N^2 E}$;

$N_{pH} = \frac{pH_i}{pH_{max}}$; $N_{OM} = \frac{OM_i}{OM_{max}}$; $N_E = \frac{E_i}{E_{max}}$.

Assessment of a large database [12,15,16] on ^{137}Cs accumulation in main crops, sown and natural grasses on various soils from peat to chernozem (FIG. 7 and 8) shows that dependence of TF_{ij} on section Sef can be approximated by the power law:

$$TF_{ij} = TF_{0j}(Sef = 1)\cdot Sef_i^{-\lambda_j} \qquad (9)$$

where: TF_{ij} – radionuclide transfer factor from $i-th$ soil to $j-th$ crops,

λ_j – degree index of dependence (9) for $j-th$ crops.

The regression coefficient of the ^{137}Cs TF on Sef is over 0.80 for most of the crops studied at different times after radionuclide entry in soil in 1986, which strongly supports the approximation in equation (9). Values of the parameters for equation (9) are presented in TABLE 15.

TABLE 15. PARAMETERS OF EQUATION $TF_{ij} = TF_{0j}(Sef_i = 1)\cdot Sef_i^{-\lambda_j}$ FOR AGRICULTURAL CROPS AND NATURAL GRASSES GROWN ON VARIOUS TYPES OF SOIL AND LANDSCAPES

Landscape	Crops	Number of observations	$TF_{0j}(Sef = 1)$, $m^2\,kg^{-1}$	Degree index λ_j	R^2
Automorphous	Natural grasses	9	0.47	0.96	0.80
	Lucerne	25	0.41	0.77	0.79
	Clover	49	0.27	0.81	0.74
	Cabbage	26	0.28	0.86	0.95
	Tomato	29	0.080	1.2	0.91
	Potato	46	0.022	1.1	0.73
	Wheat	16	0.011	1.7	0.86
Hydromorphous	Natural grasses	13	0.21	1.6	0.61
	Sown grasses	48	0.32	0.77	0.84
Scale of differences, times			43	2.2	

The parameter $TF_{0j}(Sef = 1)$ is the TF value from a soil with normalised values of parameters equal to 1, that is the standard soil of: pH=7.0; E=40 mg-eq/100 g; OM content =6.0%. The $TF_{0j}(Sef = 1)$ value can be used as the criterion of the species affinity for the element under study. The area of section Sef is a universal characteristic of soil properties so it can be used for soil classification not just for radionuclide behaviour in a soil-plant system.

An advantage of the use of CESP is that it does not require the very complicated mathematics needed for multi-dimensional spaces. The influence of other characteristics on TF can be estimated by calculation of the addition pyramid volume. The base is the triangle (pH, E, OM) and the height is the normalised value of an additional parameter, e.g. mobile potassium content in the prediction of ^{137}Cs behaviour, exchangeable calcium in the case of ^{90}Sr and mobile species of nitrogen or phosphorous in the prediction of soil fertility.

Grouping of soils by characteristic *Sef* can be useful in classification of soils by TF, which is a task of this given CRP. An analogous approach could be used to estimate of soil fertility.

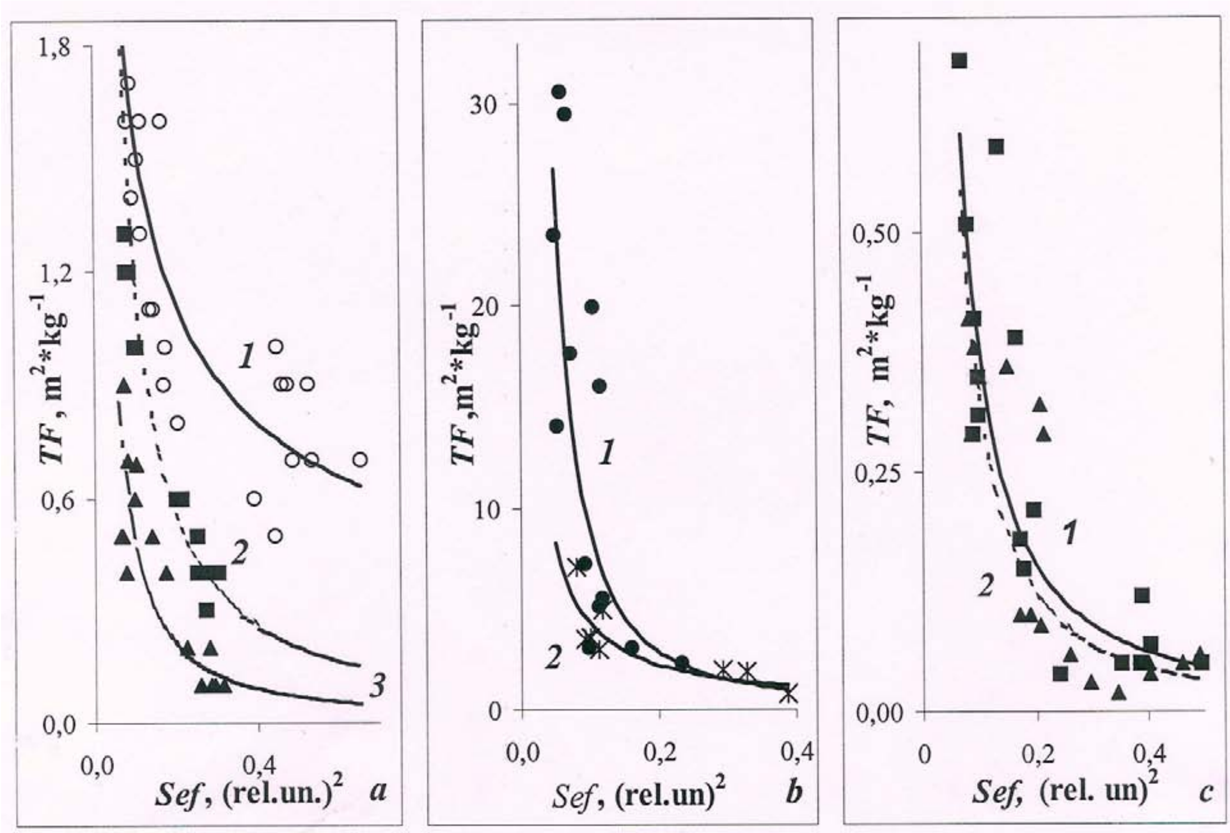

FIG. 8. Relationship between radionuclide transfer factor (TF) in plants and integrated assessment of soil properties Sef

(a) ^{137}Cs TF in*: 1*-green mass of lucerne, *2*- tomato fruit, *3*- potato tubers (1989)
(*b*) ^{137}Cs TF in natural grasses on: *1*-automorphic, *2*-hydromorphic soils (1999)
(c) ^{90}Sr TF in: *1*- tomato fruit*, 2*- potato tubers (1992)

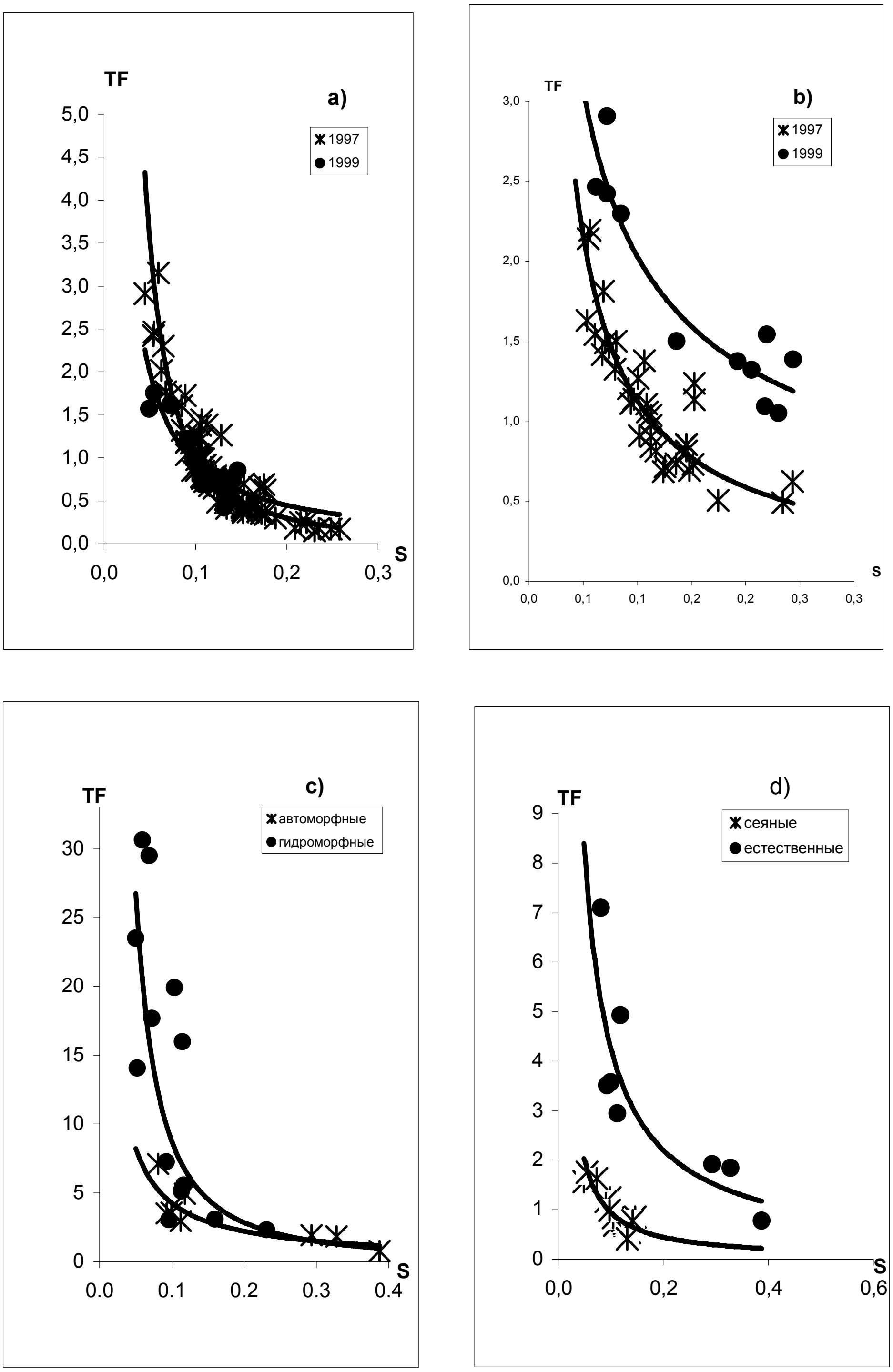

FIG. 9. Relationship between ^{137}Cs TF (Bq kg^{-1}/kBq m^{-2}) in grasses and integrated assessment of soil properties, Sef (rel.un.)[2]

a – sown grasses on automorphous soils; *b* - sown grasses on hydromorphous soils ; *c* - natural grasses on auto-(and hydromorphous soils (1999); *d* - sown and natural grasses on automorphous soils (1999)

8. CONCLUSIONS

(1) The coefficient of variation of radionuclide TF values obtained in field experiments from sod-podzolic, chernozem and peat-bog soils to oats, potatoes and beetroot is usually 10–20% and does not exceed 30% of the average in any case, which allows them to be used for prediction and the creation of analytical functions.

(2) The specific activity of plants depends linearly on the level of ^{137}Cs contamination of soil but the TF in specific conditions does not.

(3) Comparison of caesium and strontium TFs should take account of the period of their incubation in a soil in each specific case. Parameterised models, one-component for strontium and two-component for caesium, can be used for comparison. Experiments should be continued with aim of defining parameters, such as time to 50% reduction in T_e^s, to characterise the accumulation of strongly absorbed caesium in soil.

(4) The ratio of ^{90}Sr and ^{137}Cs TF values for various crops on the same soil permits the comparison of data obtained in different regions and with various crops. The accuracy of determination of the TF ratio can be improved significantly by using averages of the ratio over several years.

(5) Transfer factors are related to soil properties. The method proposed for the complete estimation of soil properties (CESP) could provide a basis for a soil classification grouping soils by their radioecological characteristics.

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UPTAKE OF CAESIUM-137 BY LEAFY VEGETABLES AND GRAINS FROM CALCAREOUS SOILS

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Abstract

^{137}Cs was deposited on Bikini Island at Bikini Atoll in 1954 as a result of nuclear testing and has been transported and cycled in the ecosystem ever since. Atoll soils are of marine origin and are almost pure $CaCO_3$ with high concentrations of organic matter in the top 40 cm. Data from previous experiments with mature fruit trees show very high transfer factors (TFs), [Bq kg^{-1} plant/Bq kg^{-1} soil, both in dry weight] into fruits from atoll calcareous soil. These TFs are much higher than reported for continental, silica-based soils. In this report TFs for five types of leafy vegetable crops and two types of grain crops are provided for use in predictive dose assessments and for comparison with other data from other investigators working with other types of soil in this CRP. Transfer factors for plants grown on calcareous soil are again very high relative to clay-containing soils and range from 2339 for grain crops and 21–113 for leafy vegetables. Results from these experiments, in this unique, high pH, high organic content, low potassium (K) soil, provide a boundary condition for models relating soil properties to TF.

1. INTRODUCTION

The United States conducted nuclear tests at Bikini Atoll from 1946 until 1958. Bikini, the main residence island, was contaminated by fallout on March 1, 1954 as a result of test Bravo that was part of the Operation Castle series of tests. Other atolls down wind of Bikini were also contaminated. About 90% of the currently estimated dose for people returning to live on Bikini is from ^{137}Cs accumulated by plants from soil [1, 2]. Detailed radioecology studies have been conducted for 25 years to determine general transport of ^{137}Cs at the atoll and uptake of ^{137}Cs from coral soil to major tree fruits, coconut (*Coco nucifera*), breadfruit (*Actocarpus altilis*), Pandanus (*Pandanus* spp.), papaya (*Carica papaya*), and banana (*Musa* spp.) that are prevalent in the local diet and contribute much of the estimated dose to returning residents [3–5].

Transfer factors (TFs) developed from these studies indicate a much higher uptake in plants of ^{137}Cs from coral soil [2, 6] than reported for continental, silica-based soils [7]. Presented in this report are TFs for ^{137}Cs for a variety of leafy vegetables and grains to supplement predictive dose assessments for atoll living and to supply comparative data fort his Coordinated Research Programme.

2. ATOLL SOIL

Atoll soils are of marine origin and consist primarily of calcium carbonate, some magnesium carbonate, organic carbon, nitrogen, and phosphorus mixed in new surfaces or older buried surface layers. There are essentially no clay minerals. Organic matter (OM) is very low in new soil or sand at the edge of an island but ranges between 5–15% in the dark surface soils in island interiors. Deeper organic layers are found in interior regions of larger islands. The OM concentration is high in the surface but decreases abruptly through a narrow transition zone about 25–30 cm deep. Observable organic matter is essentially absent below depths of 40–50 cm. Almost all roots involved in absorption of nutrients are in the top 40–50 cm organic-containing layer.

The phosphorus (P) content of atoll soil ranges from 2–6 g kg^{-1} but is quite variable and can be as high as 5–15 g kg^{-1} in some areas as a result of past guano deposits from nesting birds. Atoll soil is very low in total K and concentrations range from 0.2–0.4 g kg^{-1} [3, 8]. Extractable K ranges between 20 and 80 mg kg^{-1} in the top 30 cm and 1–10 mg kg^{-1} below 30 cm. Sixty to 75% of the extractable K is water-soluble [r^2 = 0.72–0.98 in various soils] (Stone and Robison, unpublished data). Potassium concentrations are highest in the 0–10 cm layer and originate from sea spray and decaying vegetation.

The pH of water-soil slurries ranges from 7.3 to as high as 8.8 [3] and cation exchange capacity from 12–24 cmoles kg^{-1}. More detail on elemental content of soil in test plots in this study is listed in Table 1.

Because clay minerals are non-existent or present in extremely small amounts and Ca is available in large amounts, relative uptake into plants of ^{137}Cs and ^{90}Sr in atoll soils is reversed from that observed in continental soils. ^{137}Cs uptake is high in atoll soils, while ^{90}Sr uptake is very low [3, 6].

3. EXPERIMENTAL DESIGN AND METHODS

The experimental site is located near the middle of Bikini Island in a relatively clear area about 30 m from a coconut grove. This area is within a region of the island with the highest ^{137}Cs concentrations in soil. A backhoe was used to excavate a 1 m deep trench, near the coconut grove boundary and around the entire plot area, to prevent coconut roots from penetrating the study site. A four-block design was used with 12 individual plots in each block (Fig. 1).

Chinese cabbage (Wong Bok, *Brassica pekinensis*), sorghum (*Sorghum bicolor*, cv. Cargill #40), and a hybrid cabbage (KK Cross, *Brassica oloeracea* var. *capitata* L.), were initially selected for random planting in each of 24 plots in Blocks 1 and 2 (Fig. 2a).

Subsequent to harvest of this first crop, a second crop was planted where corn (*Zea mays*, Hawaiian hybrid) replaced sorghum, Amaranth (*Amaranth tricolor* L.), sometimes referred to as Chinese spinach Yin Choi, replaced Wong Bok, and Kai Choi (*Brassica juncea* L.), sometimes referred to as leaf mustard, replaced KK Cross (Fig. 2b). Blocks 3 and 4 were planted with corn (*Zea mays*, cv. Silver Queen), Amaranth, and Mizuna (Brassica, campestris L.), all randomized among the 24 plots (Fig. 2c) immediately after second planting of Blocks 1 and 2.

Table1. Composition of Bikini Island soil in the test-plot area

Soil depth cm	Block	Plot	pH	EC µs/cm	$CaCO_3$ (%)	Inorg. C (%)	KCl extract NH4-N mg/kg	KCl extract NO3-N mg/kg	Total Cu mg/kg	Total Fe mg/kg	Total K mg/kg	Total Mg g/kg	Total Mn mg/kg	Total Na g/kg	Total P g/kg	Total Zn mg/kg	Exch. Al mg/kg	Exch. Ca mg/kg	Exch. Cd mg/kg	Exch. Co mg/kg	Exch. Cr mg/kg	Exch. Cu mg/kg	Exch. Fe mg/kg	Exch. K mg/kg	Exch. Mg mg/kg	Exch. Mn mg/kg	Exch. Na mg/kg	Exch. Ni mg/kg	Exch. P mg/kg	Exch. Pb mg/kg	Exch. Zn mg/kg
0 to 40	2	C	7.31	292	87.8	10.5	4.99	11.4	4.30	142	607	16.2	69.7	2.66	3.07	80.8	0.86	355	0.053	0.007	0.041	0.360	3.20	14.5	19.9	7.3	13.4	0.037	14.5	0.080	53.3
0 to 40	2	D	7.28	353	89.2	10.7	5.64	8.27	1.90	87.4	469	14.6	36.0	2.81	2.58	263	0.80	450	0.059	0.007	0.041	0.355	4.00	16.8	41.7	7.0	15.2	0.033	21.7	0.104	84.4
0 to 40	2	E	7.24	325	84.3	10.1	6.58	4.76	1.74	110	402	13.2	9.2	3.03	2.09	14.0	0.93	506	0.059	0.003	0.037	0.212	2.55	18.3	42.7	2.7	17.3	0.042	19.2	0.058	14.1
0 to 40	2	F	7.32	296	87.6	10.5	10.4	10.7	1.34	46.9	392	13.8	7.1	2.52	1.85	13.8	1.0	536	0.079	0.004	0.045	0.167	3.36	21.0	61.8	3.6	29.8	0.033	36.7	0.067	28.0
0 to 40	2	G	7.3	310	86.0	10.3	13.9	13.2	1.52	93.8	363	14.0	6.2	2.65	3.46	8.6	1.3	546	0.148	0.003	0.055	1.315	5.33	22.2	35.7	4.3	18.4	0.030	32.7	0.126	17.7
0 to 40	2	H	7.41	331	84.7	10.2	16.2	16.6	1.25	81.6	354	16.5	21.3	2.90	2.80	228	1.1	441	0.108	0.009	0.052	0.240	5.91	21.7	56.6	10.0	21.0	0.037	34.4	0.115	279
0 to 40	2	I	7.37	316	84.3	10.1	6.05	11.6	2.32	417	481	14.6	45.6	2.84	1.78	56.7	0.84	401	0.047	0.008	0.036	0.304	4.18	17.8	43.6	13.4	19.6	0.044	37.9	0.177	127
0 to 40	2	J	7.28	320	84.7	10.2	4.56	7.60	1.17	90.7	354	12.8	4.5	2.62	2.76	9.2	0.87	442	0.052	0.003	0.046	0.270	2.27	16.9	48.4	2.6	22.3	0.036	50.4	0.126	10.4
0 to 40	2	K	7.34	482	81.8	9.8	11.5	7.91	1.43	63.6	358	14.0	8.8	2.66	5.84	17.1	1.1	519	0.102	0.002	0.047	0.130	4.12	21.1	50.6	3.0	29.4	0.033	32.3	0.138	13.7
0 to 40	2	L	7.31	346	83.1	10.0	15.9	23.4	1.64	53.6	311	11.7	10.3	2.54	3.12	94.6	1.2	543	0.114	0.005	0.050	0.199	5.00	21.1	30.5	5.4	28.6	0.039	25.2	0.124	29.0
0 to 20	1	B	7.38	325	83.9	10.1	14.8	12.8	1.41	33.5	316	13.8	12.7	2.49	5.67	18.6	1.2	580	0.142	0.003	0.049	0.113	6.00	25.4	42.1	5.2	43.0	0.048	45.4	0.072	21.5
0 to 20	1	C	7.44	373	83.0	10.0	19.7	12.6	1.11	28.1	321	13.1	16.7	2.54	4.42	46.4	1.1	532	0.112	0.006	0.261	0.135	5.40	25.0	88.2	10.5	25.4	0.104	74.0	0.070	151
0 to 20	1	E	7.39	324	82.7	9.9	12.7	7.51	0.83	22.4	292	10.9	19.3	2.35	2.91	41.6	1.0	537	0.122	0.006	0.063	0.100	6.76	22.6	83.6	10.6	24.2	0.039	59.2	0.067	120
0 to 20	1	F	7.43	354	85.5	10.3	19.2	10.6	1.03	27.8	297	14.2	17.1	2.49	3.55	47.1	1.1	567	0.140	0.005	0.060	0.110	6.35	26.7	94.2	8.6	26.5	0.037	60.2	0.077	62.3
0 to 20	1	G	7.34	303	84.3	10.1	11.3	17.4	1.03	29.8	297	14.4	16.4	2.71	4.24	24.7	1.2	532	0.151	0.004	0.065	0.142	5.63	22.8	34.1	9.7	24.1	0.040	43.8	0.100	39.8
0 to 20	1	I	7.45	302	84.7	10.2	14.1	8.14	0.981	28.9	295	14.6	37.4	2.54	4.20	38.8	1.1	557	0.108	0.005	0.046	0.117	4.34	23.7	110	10.5	24.8	0.026	121	0.068	60.3
0 to 20	1	K	7.4	308	78.1	9.4	13.6	8.56	0.820	23.3	304	13.6	9.8	2.64	3.50	12.9	1.1	551	0.118	0.003	0.053	0.109	4.97	25.0	96.6	6.8	25.7	0.027	65.9	0.059	18.4
0 to 20	1	L	7.44	342	83.5	10.0	22.8	9.87	1.12	31.6	306	11.5	16.6	2.35	3.84	44.4	1.0	557	0.152	0.005	0.061	0.139	5.68	25.2	85.2	7.8	22.2	0.035	44.7	0.066	31.5
0 to 20	2	A	7.35	313	86.6	10.4	19.1	10.2	2.76	41.1	256	12.2	56.3	2.43	4.05	75.6	1.3	535	0.168	0.006	0.067	0.153	6.54	22.3	45.3	12.5	16.7	0.030	48.8	0.057	82.7
0 to 20	2	D	7.37	329	87.1	10.5	9.79	13.3	1.20	24.2	220	9.9	14.5	2.30	2.86	41.7	0.95	496	0.088	0.006	0.071	0.088	4.71	19.5	72.1	6.6	17.5	0.041	46.0	0.045	40.0
0 to 20	2	E	7.35	306	85.1	10.2	12.4	11.7	0.962	27.7	157	9.4	17.7	2.37	2.76	39.6	1.2	512	0.132	0.008	0.068	0.134	5.87	21.3	59.5	10.4	19.1	0.036	43.1	0.085	56.0
0 to 20	2	F	7.33	402	78.6	9.4	21.4	17.0	1.31	34.8	180	11.4	16.4	2.14	3.60	20.6	1.3	605	0.201	0.006	0.079	0.246	5.73	31.2	96.2	8.6	46.1	0.040	86.2	0.104	21.4
0 to 20	2	G	7.35	356	77.7	9.3	18.2	13.1	1.32	33.5	148	11.7	16.1	2.26	3.55	28.1	1.5	583	0.199	0.006	0.088	0.211	7.31	22.4	39.9	9.1	17.3	0.033	52.4	0.123	40.5
0 to 20	2	H	7.36	358	82.1	9.9	14.2	10.4	0.890	26.1	141	10.5	15.2	2.23	2.96	30.6	1.2	546	0.151	0.007	0.103	0.150	5.43	23.9	60.2	10.5	32.6	0.040	47.9	0.096	46.7
0 to 20	2	J	7.32	392	82.6	9.9	12.2	18.2	1.16	21.9	141	10.7	11.8	2.17	3.89	23.9	1.2	546	0.136	0.005	0.111	0.117	3.87	24.1	83.9	7.1	33.4	0.033	97.5	0.137	19.9
0 to 20	2	K	7.33	433	81.8	9.8	19.6	9.54	1.15	30.7	145	10.5	25.4	2.51	3.88	43.0	1.3	547	0.182	0.008	0.095	0.149	6.62	25.9	65.4	10.3	42.7	0.038	60.6	0.090	63.1
0 to 20	4	A	7.2	710	80.4	9.6	7.52	60.7	1.16	28.5	124	9.0	9.6	2.44	3.23	10.6	1.0	530	0.108	0.008	0.155	0.134	3.93	22.4	113	3.4	65.1	0.050	31.5	0.098	10.2
0 to 20	4	B	7.29	519	84.4	10.1	12.2	53.1	1.68	75.6	129	9.3	8.7	2.47	4.42	6.8	1.2	461	0.104	0.011	0.144	0.193	2.19	20.9	60.1	2.9	45.3	0.047	42.2	0.116	2.7
0 to 20	4	C	7.24	583	68.8	8.3	7.75	59.3	3.41	62.6	157	9.7	38.4	2.35	5.02	84.4	0.89	218	0.073	0.020	0.164	0.136	1.37	15.8	31.8	2.9	20.1	0.044	0.3	0.023	0.0
0 to 20	4	D	7.29	614	68.6	8.2	8.84	49.2	2.42	87.2	162	8.7	10.1	2.46	4.30	11.5	1.3	564	0.126	0.009	0.140	0.359	2.25	28.4	91.2	5.3	48.3	0.053	73.6	0.123	5.0
0 to 20	4	F	7.24	632	77.1	9.3	6.83	78.0	1.88	50.9	155	9.0	16.2	2.48	6.45	20.9	2.1	612	0.202	0.005	0.086	0.304	6.95	21.5	77.9	6.9	60.0	0.086	49.6	0.093	14.8
0 to 20	4	G	7.3	423	79.6	9.6	5.64	26.1	1.86	47.8	165	9.3	12.3	2.42	4.54	17.9	0.22	86	0.000	0.000	0.000	0.063	0.00	14.5	21.7	0.0	21.4	0.000	0.2	0.000	0.0
0 to 20	4	H	7.26	717	77.3	9.3	9.01	60.2	1.46	32.7	142	8.2	5.8	2.29	2.88	3.9	2.2	690	0.178	0.008	0.052	0.196	5.21	55.4	134	3.2	82.2	0.056	52.5	0.078	2.2
0 to 20	4	I	7.18	1110	78.1	9.4	8.65	164	1.83	63.1	126	9.1	10.7	2.42	4.27	10.0	2.2	689	0.157	0.003	0.073	0.216	5.71	33.7	125	3.0	82.7	0.056	51.8	0.077	2.2
0 to 20	4	K	7.23	713	74.5	8.9	12.5	84.0	2.12	52.4	154	9.5	13.0	2.45	5.34	10.9	1.6	452	0.142	0.003	0.068	0.194	6.04	24.4	95.6	3.9	67.9	0.049	40.5	0.060	4.8
0 to 20	4	L	7.26	628	79.6	9.5	6.98	64.2	1.80	35.5	128	11.2	8.4	2.53	4.26	10.0	1.3	475	0.143	0.000	0.054	0.142	4.70	54.5	82.7	3.5	70.9	0.023	50.9	0.043	5.3
0 to 20	3	A	7.24	751	80.7	9.7	6.73	113	1.28	24.1	122	9.0	5.4	2.47	2.96	9.0	1.6	447	0.094	0.001	0.054	0.138	4.16	22.3	78.3	2.4	62.4	0.043	46.3	0.049	1.7
0 to 20	3	B	7.26	686	80.8	9.7	7.38	87.6	1.50	27.7	139	10.7	7.5	2.60	3.81	7.6	1.2	510	0.083	0.005	0.052	0.128	3.82	19.6	77.2	2.4	54.2	0.049	44.9	0.063	4.5
0 to 20	3	D	7.24	683	79.9	9.6	6.78	111	1.63	51.1	129	9.2	8.8	2.28	3.17	5.2	1.8	792	0.097	0.005	0.063	0.217	5.06	22.3	82.2	3.4	60.8	0.051	58.6	0.117	2.4
0 to 20	3	E	7.22	949	78.2	9.4	9.26	117	2.05	33.7	129	7.2	8.7	2.12	4.15	4.9	1.4	301	0.020	0.007	0.038	0.121	4.08	16.4	34.6	3.6	37.5	0.041	0.0	0.000	0.0
0 to 20	3	F	7.28	683	76.2	9.1	11.59	61.3	2.43	45.6	141	7.9	13.3	2.18	5.86	10.1	1.9	603	0.193	0.003	0.077	0.248	8.36	23.4	93.0	5.5	70.6	0.060	42.1	0.135	7.4
0 to 20	3	G	7.28	708	95.6	11.5	5.10	110	1.31	22.3	102	8.5	10.6	2.37	3.63	9.2	1.9	640	0.107	0.004	0.058	0.232	5.81	22.5	90.7	4.7	69.0	0.051	60.5	0.119	10.6
0 to 20	3	I	7.2	613	82.7	9.9	7.62	75.0	1.33	20.1	140	12.3	17.0	2.67	3.89	15.1	1.9	521	0.182	0.003	0.076	0.280	0.95	63.1	85.6	6.9	69.8	0.054	55.9	0.235	14.7
0 to 20	3	J	7.26	615	72.1	8.7	4.40	92.5	2.29	27.5	141	12.2	13.0	2.35	2.78	9.8	3.6	516	0.158	0.004	0.077	0.148	0.47	54.4	104	4.5	70.0	0.045	50.0	0.266	7.3
0 to 20	3	K	7.3	492	88.2	10.6	9.01	119	2.10	44.6	145	9.7	9.5	2.07	4.07	7.1	0.84	189	0.044	0.000	0.021	0.067	2.16	15.3	38.8	1.4	19.7	0.012	15.6	0.012	1.3
0 to 20	3	L	7.3	745	69.6	8.3	12.49	107	2.32	58.6	147	10.0	27.4	2.27	6.28	35.4	0.55	191	0.068	0.000	0.026	0.096	2.72	15.6	29.3	3.1	19.6	0.015	21.3	0.041	7.3
0 to 40	4	D	7.4	653	85.0	10.2	8.29	58.8	1.76	42.7	149	12.7	8.5	2.60	5.49	9.7	1.3	539	0.211	0.000	0.063	0.146	1.50	57.6	171	3.3	77.0	0.015	64.4	0.151	3.3
0 to 40	4	F	7.38	473	100.3	12.0	7.74	45.7	1.06	22.9	109	11.9	5.9	2.68	4.54	15.2	1.4	518	0.181	0.003	0.060	0.113	0.51	52.2	80.8	1.9	67.0	0.034	26.6	0.231	4.5
0 to 40	4	I	7.38	734	82.3	9.9	6.85	101	0.643	17.4	104	12.7	5.8	2.50	2.27	5.6	0.99	527	0.165	0.000	0.055	0.105	0.69	52.9	111	2.9	75.8	0.030	28.5	0.169	3.6
0 to 40	3	D	7.33	533	119.3	14.3	5.79	85.3	0.717	24.1	97.7	13.8	5.2	2.76	2.77	43.0	1.6	598	0.070	0.002	0.054	0.134	3.73	18.8	93.6	2.0	56.8	0.051	34.0	0.110	1.7
0 to 40	3	G	7.34	557	74.9	9.0	5.94	93.2	0.636	26.3	104	11.0	5.1	2.58	1.55	4.1	2.3	544	0.066	0.000	0.042	0.115	3.65	54.1	87.3	2.8	71.8	0.026	39.0	0.050	4.2
0 to 40	3	I	7.25	858	78.2	9.4	7.49	85.9	1.52	45.8	129	10968	8.5	2577	5.29	7.1	2.3	633	0.132	0.000	0.074	0.150	6.51	54.1	80.1	3.9	92.2	0.044	43.9	0.078	2.3

6 soil cores composited

Note: Soils provided in "Un-ball milled" form.

The Hawaiian hybrid corn died in the second planting of blocks 1 and 2 and a third planting of corn (*Zea mays*, cv. Silver Queen) was required. The planting and harvesting schedule is shown in Table 2.

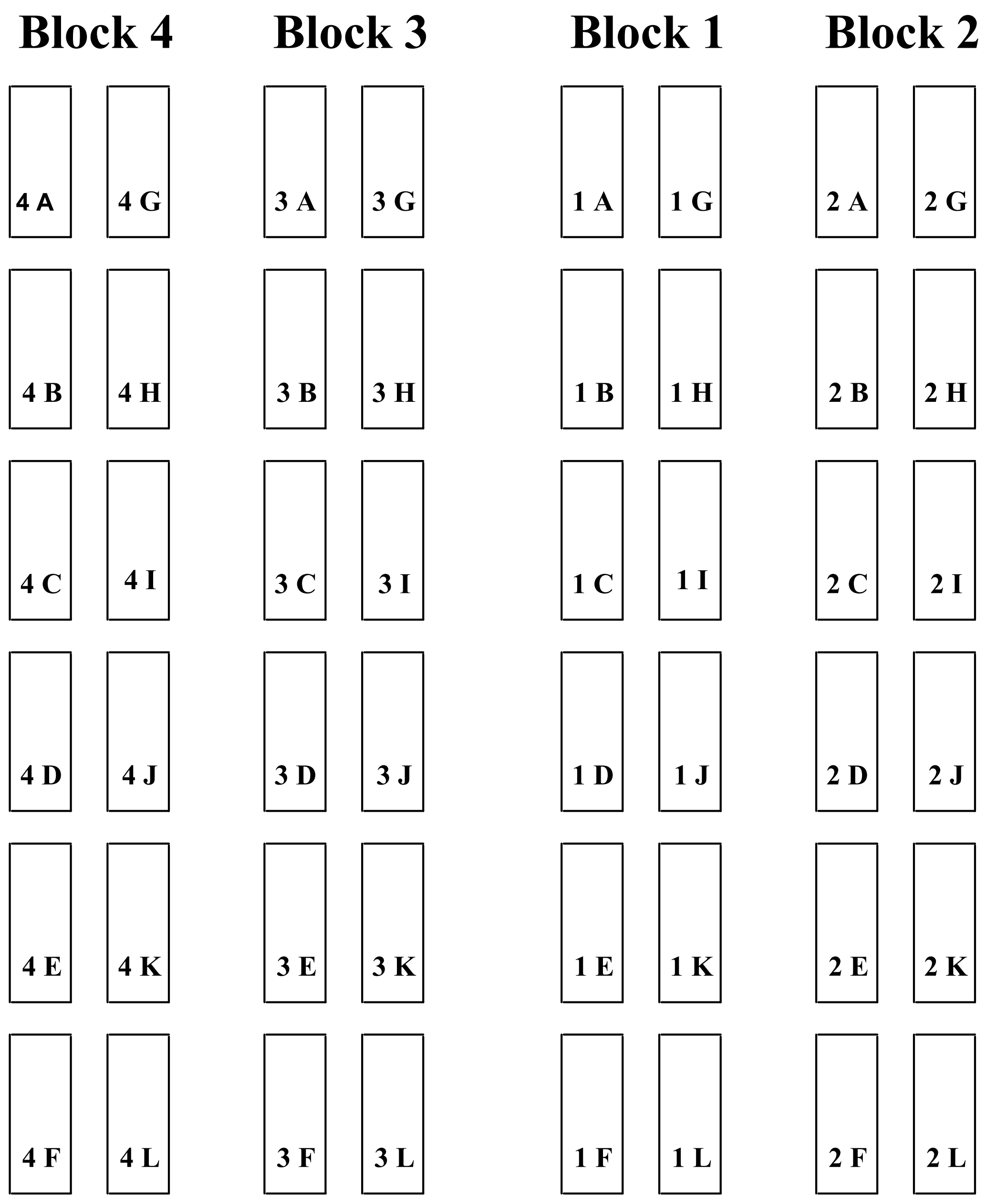

FIG. 1. The layout of 4 blocks and 48 plots at Bikini Island.

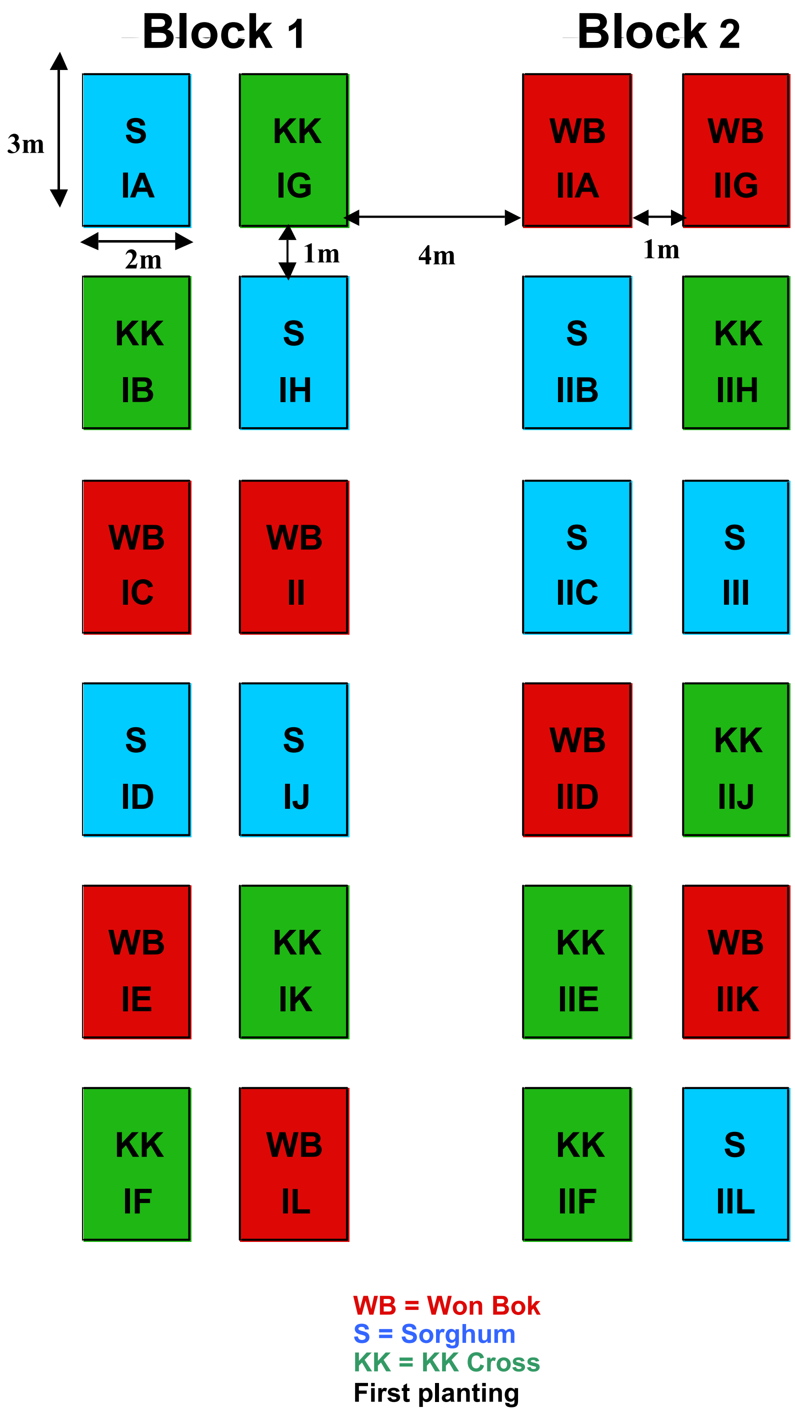

FIG. 2a. Distribution of crops in 24 plots in blocks 1 and 2 for the first planting.

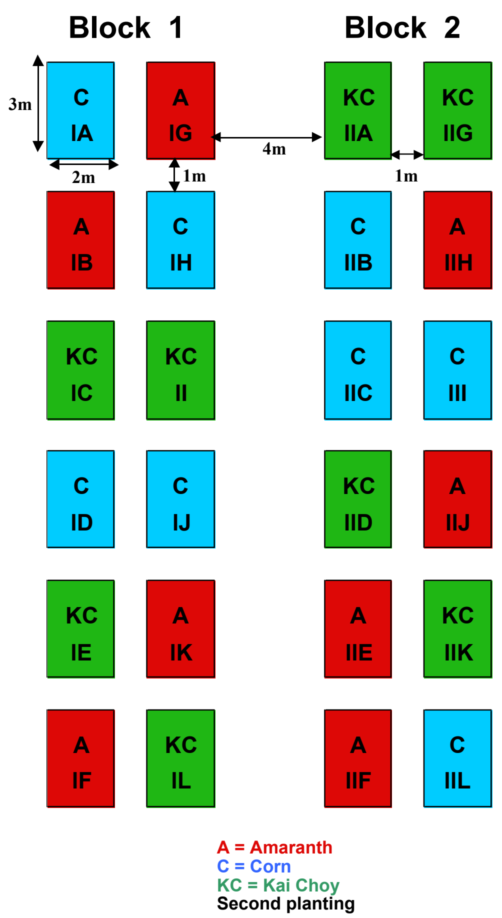

FIG. 2b. Distribution of crops in 24 plots in blocks 1 and 2 for the second planting.

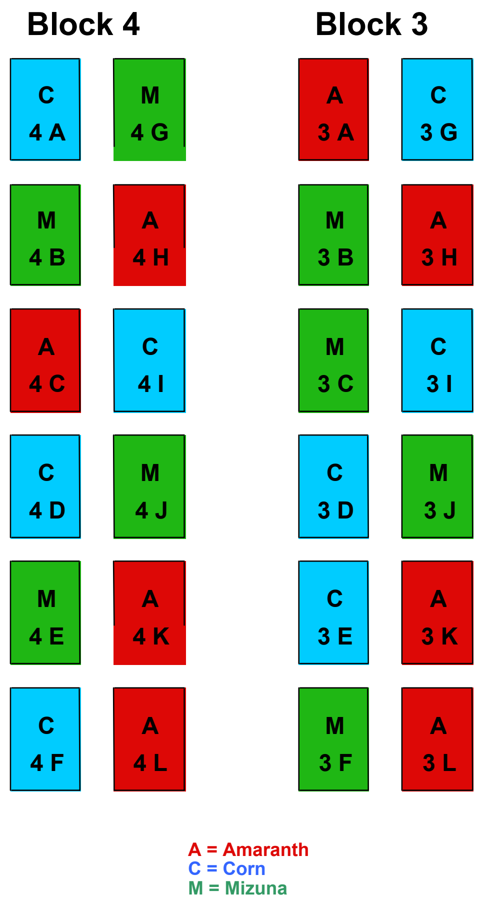

FIG. 2c. Distribution of crops in 24 plots in blocks 3 and 4.

TABLE 2. PLANTING AND HARVEST SCHEDULE FOR BLOCKS 1, 2, 3 AND 4

Blocks 1 & 2		Blocks 3 & 4	
First planting	*Dates*	*First planting*	*Dates*
Sorghum	05 May 1999	Amaranth	06 November 1999
Won Bok	15 May 1999	Corn	05 November 1999
KK Cross	29 May 1999	Mizuna	06 November 1999
First harvest		*First harvest*	
Sorghum	22-25 July 1999	Amaranth	11 December 1999
Won Bok	22-25 July 1999	Corn	22 January 2000
KK Cross	22-25 July 1999	Mizuna	Feb/March 2000
Second planting			
Corn (Hawaiian hybrid)	27 July 1999		
Amaranth	September 1999		
Kai choy	September 1999		
Second harvest			
Corn (Hawaiian hybrid)	All died		
Amaranth	November 1999		
Kai choy	November 1999		
Third planting			
Corn (Silver Queen)	04 November 1999		
Third harvest			
Corn (Silver Queen)	January 2000		

Nitrogen (N), P, and trace minerals (TM) were placed in the furrow at the time of planting. Plants were watered if rainfall was inadequate for good plant growth and N and TM were applied as required for quality growth and productivity.

Four soil profiles were collected in the 6 m space between plots 3 G and I A, 3 I and 1 C, 3 J and 1 E, and 3 L and 1 F. The increments were: 0–5 cm, 5–10 cm, 10–15 cm, 15–25 cm, 25–40 cm, and 40–60 cm. In addition, six 0–20 cm punch-tube core soil samples were collected from each plot that contained leafy vegetables. Two samples were taken at each end of a plot, 50 cm in from the end and 65 cm in from both sides of the plot. The other two samples were taken at the plot centre 65 cm from the sides. The root zone of the leafy vegetables was within the top 20 cm. of soil. Six 0–40 cm punch-tube core samples were taken as described above in all plots containing sorghum or corn. Corn and sorghum have a rooting zone to about 40 cm.

Sorghum seed and corn ears were harvested, double bagged in plastic bags, and placed in freezers on the island. Sorghum and corn stovers were cut about 8 cm above ground level, double bagged and placed in freezers. All leafy vegetables were cut just above ground level. They were then rinsed a leaf at a time to ensure there was no residual soil on the organic sample and then double bagged and frozen. All samples were shipped in Matson freezer vans to Lawrence Livermore National Laboratory (LLNL).

At LLNL the vegetation samples were dried by lyophilization to constant weight, ground to fine consistency in a Waring blender, and canned for gamma spectrometry analysis. All soil samples were sifted through a 2 mm screen to remove large pieces of coral that have little ^{137}Cs activity or impact on ^{137}Cs uptake from soil by plants. They were then oven dried at low temperature, ball-milled to a homogeneous powder, and canned for gamma analysis. All samples were packed into steel cans 231 cm^3 in volume for gamma analysis using 20 high-resolution, intrinsic, solid-state, germanium detectors.

4. RESULTS

Distribution of ^{137}Cs in soil between Blocks 2 and 3 after some 25 years of transport and redistribution is shown in Fig. 3. The cumulative distribution is shown in Fig. 4. Distribution of ^{137}Cs in the experimental plots is exponential to a depth of about 30–40 cm. At depths below about 40 cm, where organic content of soil is very low or non-existent, ^{137}Cs concentration is very low and forms an exponential distribution with a different slope. In island interior locations the organic layer can extend to 40–50 cm deep. In such cases ^{137}Cs distribution is a single exponential to 40–50cm. More than 90% of ^{137}Cs activity in the test plots is retained in the top 30 cm of soil. The ^{137}Cs concentration for the 0–20 cm or 0–40cm layers for each of the plots is listed in Table 3.

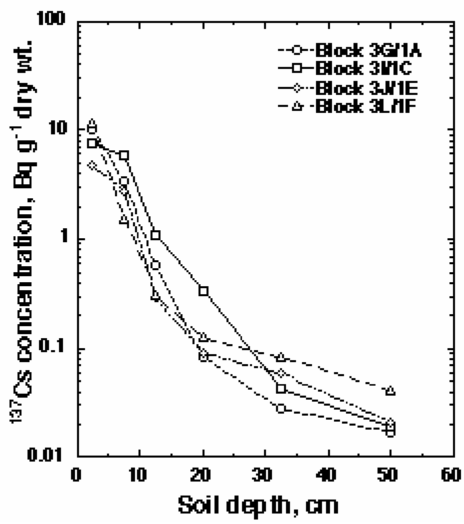

FIG. 3. The distribution of ^{137}Cs to a depth of 50 cm in soil in the test area.

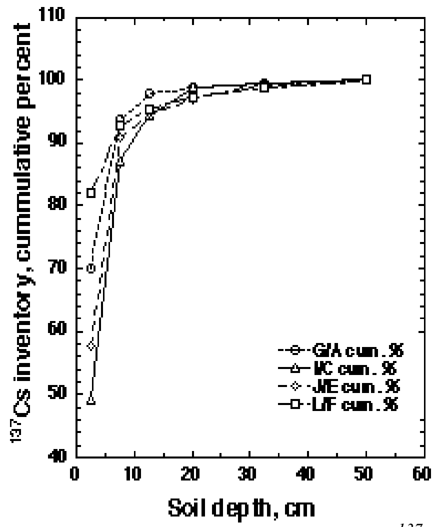

FIG. 4. The cumulative distribution of ^{137}Cs to 50 cm depth in soil in the study area.

TABLE 3. ^{137}Cs CONCENTRATION FOR 0–20 cm AND 0–40 cm IN PLOTS IN ALL BLOCKS

^{137}Cs concentration in the root zone Bq/g dry weight									
Block No	Plot No	0–40 cm	0–20 cm	Ratio 0–20/0–40	Block No	Plot No	0–40 cm	0–20 cm	Ratio 0–20/0–40
1	A	0.85			3	A		1.1	
	B	0.68	1.4	2.1		B		0.86	
	C	1.1	2.6	2.3		C		0.64	
	D	1.3				D	0.47	0.98	2.1
	E	0.59	1.1	1.9		E	0.49	1.5	3.1
	F	1.04	2	1.9		F		1.4	
	G	0.47	1.1	2.3		G	0.5	1.1	2.3
	H	0.51				H		1	
	I	1.2	2.7	2.2		I	0.96	0.8	0.83
	J	1.2				J		0.92	
	K	1.1	2.6	2.4		K		3.6	
	L	1.2	1.7	1.4		L		2.3	
2	A	0.46	1.1	2.4	4	A	0.61	1.1	1.9
	B	0.28				B		1.6	
	C	0.38				C		1.8	
	D	0.45	0.88	2		D	1	2.4	2.3
	E	0.79	1.6	2		E		2.2	
	F	1.1	2.7	2.5		F	0.51	0.81	1.6
	G	0.78	1.4	1.8		G		1.1	
	H	0.95	1.3	1.4		H		1.5	
	I	0.42				I	0.7	1.7	2.4
	J	0.4	1.3	3.3		J		2.9	
	K	0.81	1.8	2.2		K		1.6	
	L	0.81				L		1.5	
			Mean	2.1				Mean	2.1
			SD	0.45				SD	0.67
			No	16				No	8
			SE	0.11				SE	0.17

TABLE 4. ^{137}Cs CONCENTRATION IN PLANTS AND SOIL AND TFs IN BLOCKS 1 AND 2

		^{137}Cs concentration Bq/g dry weigh									
Block	Plot	Plant	In plant	0–40 cm	0–20 cm	TF	Plant	In plant	0–40 cm	0–20 cm	TF
1	A	Sorg seed	11	0.85		13	Sorg stover	15	0.85		18
	D	Sorg seed	24	1.3		18	Sorg stover	50	1.3		38
	H	Sorg seed	18	0.51		35	Sorg stover	23	0.51		45
	J	Sorg seed	44	1.2		37	Sorg stover	57	1.2		48
2	B	Sorg seed	16	0.28		57	Sorg stover	14	0.28		50
	C	Sorg seed	26	0.38		68	Sorg stover	22	0.38		58
	I	Sorg seed	30	0.42		71	Sorg stover	33	0.42		79
	L	Sorg seed	8	0.81		10	Sorg stover	13	0.81		16
					Mean	39				Mean	44
					SD	24				SD	21
					N	8				N	8
					SE	8 6				SE	7.3
1	1C	Won Bok	198		2.6	76	Kai Choy	41		2.6	16
	1E	Won Bok	210		1.1	191	Kai Choy	32		1.1	29
	1I	Won Bok	229		2.7	85	Kai Choy	61		2.7	36
	1L	Won Bok	178		1.7	105	Kai Choy	37		1.7	22
2	2A	Won Bok	90		1.1	82	Kai Choy	28		1.1	25
	2D	Won Bok	223		0.88	253	Kai Choy	51		0.88	57
	2G	Won Bok	66		1.4	47	Kai Choy	24		1.4	17
	2K	Won Bok	114		1.8	63	Kai Choy	29		1.8	16
					Mean	113				Mean	27
					SD	71				SD	14
					N	8				N	8
					SE	25				SE	4.9
1	1B	KK Cross	68		1.4	49	Amaranth	29		1.5	19
	1F	KK Cross	96		2	48	Amaranth			2	
	1G	KK Cross	49		1.1	45	Amaranth			1.1	
	1K	KK Cross	139		2.6	53	Amaranth			2.6	
2	2E	KK Cross	148		1.6	93	Amaranth			1.6	
	2F	KK Cross	74		2.7	27	Amaranth	41		2.7	15
	2H	KK Cross	89		1.3	68	Amaranth	29		1.3	22
	2J	KK Cross	83		1.3	64	Amaranth	35		1.3	27
					Mean	56				Mean	21
					SD	19				SD	5.1
					N	8				N	4
					SE	6.9				SE	2.5
1	A	Corn ear	3.7	0.85		4.4	Corn stovr	8.6	0.85		10.1
	D	Corn ear	8.1	1.3		6.2	Corn stovr	15	1.3		11.5
	H	Corn ear	4.5	0.51		8.8	Corn stovr	9.7	0.51		19
	J	Corn ear	50	1.2		42	Corn stovr	44	1.2		36.7
2	B	Corn ear		0.28			Corn stovr		0.28		
	C	Corn ear	25	0.38		66	Corn stovr	19	0.38		50
	I	Corn ear	4.6	0.42		11	Corn stovr	8.8	0.42		21
	L	Corn ear	14	0.81		17	Corn stovr	10	0.81		12.3
					Mean	22				Mean	23
					SD	23				SD	15
					N	7				N	7
					SE	8.7				SE	5.7

TABLE 5. ^{137}Cs CONCENTRATION IN PLANTS AND SOIL AND TFs IN BLOCKS 3 AND 4

	^{137}Cs concentration Bq/g dry weight											
Block	Plot	Plant	In plant	0–40 cm	0–20 cm	TF	Plot	Plant	In plant	0–40 cm	0–20 cm	TF
3	D	corn ears	13	0.47		27	D	corn stovr	18	0.47		38
	E	corn ears	7.6	0.49		16	E	corn stovr	12	0.49		25
	G	corn ears	8.5	0.5		17	G	corn stovr	13	0.5		26
	I	corn ears	18	0.96		18	I	corn stovr	15	0.96		16
4	A	corn ears	14	0.61		23	A	corn stovr	23	0.61		37
	D	corn ears	19	1		19	D	corn stovr	24	1		24
	F	corn ears	12	0.51		24	F	corn stovr	10	0.51		20
	I	corn ears	8.1	0.7		12	I	corn stovr	14	0.7		20
					Mean	19					Mean	26
					SD	5.1					SD	8
					N	8					N	8
					SE	1.8					SE	2.8
3	B	Mizuna	53.9		0.86	63	A	Amaranth	27.7		1.1	25
	C	Mizuna	51.6		0.64	81	H	Amaranth	18.1		1	18
	F	Mizuna	73.1		1.4	52	K	Amaranth	35.8		3.6	29
	J	Mizuna	LOST		0.92		L	Amaranth	52.6		2.3	23
4	B	Mizuna	58.4		1.6	37	C	Amaranth	20.7		1.8	12
	E	Mizuna	58.7		2.2	27	H	Amaranth	21.2		1.5	14
	G	Mizuna	68.6		1.1	63	K	Amaranth	32		1.6	20
	J	Mizuna	62.4		2.9	22	L	Amaranth	37.6		1.5	25
					Mean	49					Mean	21
					SD	22					SD	5.9
					N	7					N	8
					SE	8.8					SE	2.4

TF results for each plot, and therefore each planted crop, in blocks 1 and 2, are listed in Table 4 for crops from both the first and second plantings. The results from blocks 3 and 4 are listed in Table 5. Transfer factors (Bq g^{-1} dry plant/Bq g^{-1} dry soil) for all crops range from about 20–110.

The difference in means between plots containing the same crop in blocks 1 and 2 and in blocks 3 and 4 was analysed using the nonparametric Mann-Whitney U test. A nonparametric method was used because of the small sample size (4 plots of each type of crop per block) for which statistical tests based on normal distributions are inappropriate. Nonparametric methods are more efficient and powerful than parametric methods under such conditions.

There was no statistical difference between crops grown in blocks 1 and 2 or between crops grown in blocks 3 and 4. There was no statistical difference between the combined data in blocks 1 and 2 and the combined data from blocks 3 and 4 for crops common to both. The lowest observed probability was 0.11 while most others exceeded 0.34. The statistical summary is listed in Table 6. As a result, data for a specific crop from all plots may be combined in calculating mean TF values. A summary of means (± 1 standard error) for all plots that contained the same crop is given in Table 7.

TABLE 6. MANN WHITNEY U-TEST PROBABILITIES FOR DIFFERENCE IN MEANS

Plant	Probabilities Blocks 1 verses 2	Blocks 3 verses 4	Blocks 1+2 verses 3+4
Sorghum seed	0.34		
Sorghum stover	0.34		
Corn ears	0.22	0.88	~1
Corn stover	0.4	0.69	0.57
Won Bok	0.49		
Kai Choy	0.98		
Amaranth	~1	0.2	0.53
KK Cross	0.34		
Mizuna		0.11	

TABLE 7. SUMMARY OF MEAN TFs AND ASSOCIATED STANDARD ERRORS

Plant	TF	Std error
Sorghum seed	39	8.6
Sorghum stover	44	7.3
Corn ears	23	4.3
Corn stover	27	3.5
Won Bok	113	25
Kai Choy	27	4.9
Amaranth	56	6.9
KK Cross	21	1.6
Mizuna	49	8.8

5. DISCUSSION AND CONCLUSIONS

It is clear that transfer factors for all annual food crops and long-lived tree food crops grown in coral soil are very high relative to those reported for continental silica-based soils. The range in TF for ^{137}Cs in coral soil for fruit trees and annual crops is 0.8–113 based on this report and [6]. The range in TF for ^{137}Cs in continental, silica-based soils ranges from 0.004–0.5 for fruits and vegetables [7]. The low end of the TF range for carbonate soils does not overlap the high end of the range from silica soil. On the other hand, TFs for ^{90}Sr are very low in Ca-rich coral soil (range 0.006–0.37) [6] while they are relatively high in silica soils (range 0.02–3.0) [7]. Thus, the ^{137}Cs and ^{90}Sr uptake from coral soil to plants is totally reversed from that observed in silica soils. Unique properties of the carbonate soil, and resulting TFs, provide an upper boundary condition for general models relating soil properties to TF.

There is a significant difference in TFs among various leafy vegetables. Mean value of TFs range from 21 for Amaranth to 113 for Won Bok cabbage with Mizuna, Kai Choi cabbage, and KK Cross cabbage in between. Based on these results it would be imprudent to assign a single TF to a category labelled leafy vegetables unless one is willing to accept such a range within a dose assessment. Also, these results indicate a level of information required for more specific application of TFs to dose assessments. There is not nearly as much difference between the two types of grain crops. Mean TFs for sorghum seed and stover are 39 and 44, respectively, and for corn kernels and stover 23 and 27, respectively. There is no statistically significant difference between the TF for sorghum seed and the stover (p=0.51) or between corn ears and the stover (p=0.38). So either part of these grain-producing plants will provide appropriate information.

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THE CLASSIFICATION OF SOIL SYSTEMS ON THE BASIS OF TRANSFER FACTORS OF RADIONUCLIDES FROM SOIL TO REFERENCE PLANTS

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Abstract

Transfer values of ^{134}Cs and ^{85} Sr to cabbage and rice from three soil types were measured in pot experiments over four seasons. TF values for both radionuclides were higher on an Acrisol than on two Fluvisols and were higher for cabbage than rice but the ratios of values to the two crops varied from soil to soil. Multiple regression analysis showed that soil cation exchange capacity, organic matter content, pH, period of contamination and radionuclide concentration in the soil could account for some 80% of the variation in TFs but the relative contributions were different for the two crops. The relationships were non-linear.

1. MATERIALS AND METHODS

Three types of soil: Ferric Acrisol, Orthi-thionic Fluvisol and Eutric Fluvisol were collected from areas representative of these types of soil in Vietnam. The soil samples were taken from the cultivated surface layer, i.e., not deeper than 20 cm., each type of soil was labelled with ^{85}Sr and ^{134}Cs following the FAO/IAEA/IUR protocol [1] and then dispensed into six pots each contained 10 kg soil (three pots with ^{85}Sr labeling and other three pots with ^{134}Cs labelling), so that for each type of soil and each type of radionuclide, there were three replicates for each reference crop.

The reference crops chosen for the study were cabbage as a broad leaf vegetable and rice as the cereal. Each year the cabbage was planted in October and harvested in January and the rice was planted in March and harvested in June. The pots with planted crops were kept in a greenhouse.

Both crop and soil samples were collected at harvest. For cabbage, only the edible part was taken, for the cereal only seed was taken. The crop samples were washed carefully to eliminate possible contamination from soil splashes adhered onto the sample surface. Then the crop samples were dried at 100°C to constant weights that were recorded. The dry crop samples then were ashed prior to determination of radioactivity. The soil samples were dried at 100°C to constant weight. Then the samples were ground and mixed well before determination of radioactivity. The contents of ^{85}Sr and ^{134}Cs in crops and soils were determined by using a gamma spectrometer with a HP Ge detector (resolution 1.9 keV and relative efficiency 41% at gamma line 1.33MeV) and reported in the units of radioactivity per unit of mass dry crop and mass dry soil. Based on these data, the soil-to-plant TF values of ^{85}Sr and ^{134}Cs were determined. The properties of soil such as pH, exchangeable K and Ca, CEC (Cation Exchangeable Capacity) and OM (Organic matter) were determined for each soil sample by appropriate techniques [2]. The contamination time is defined as the time interval between when the soil was labelled with the radionuclides to crop harvest. The planting time is the period from when crop was planted to harvest.

2. RESULTS AND DISCUSSION

All data of soil-to-plant transfer factors of ^{85}Sr and ^{134}Cs as well as the soil properties were reported the data sheets according to the protocol. The textures of soils are given in. Table 1.

TABLE 1. THE TEXTURES OF THE SOILS

Soil type	Clay (%)	Silt (%)	Sand (%)
Ferric Acrisol	8.8	3.8	87.4
Orthi-thionic Fluvisol	28.7	37.6	33.7
Eutric Fluvisol	18.0	22.6	59.4

Table 2 summarizes the statistical data on soil-to-plant TF values of ^{85}Sr and ^{134} Cs for each type of soil and for each type of crop planted. Among the three types of soil, the Ferralic Acrisol shows the highest TF value. For all types of soil and both radionuclides the TF values for cabbage are higher than those for rice. However, according to [3, 4] a higher or lower uptake capacity is not crop specific. If one crop appears to have high or low uptake from a certain type of soil then another crop will show the same behaviour.

TABLE 2. THE SOIL-TO-PLANT TF VALUES OF ^{85}Sr AND ^{134}Cs FROM DIFFERENT SOILS TO CABBAGE AND RICE

Nuclide	Crop class*	Data	Soil type			Grand Total†
			Eutric Fluvisol	Ferralic Acrisol	Orthi-thionic Fluvisol	
^{134}Cs	C	Average of TF	0.056	0.37	0.003	0.17
		SD of TF	0.11	0.47	0.003	0.34
		No of TF	14	14	8	36
	V	Average of TF	0.15	2.8	0.40	1.1
		SD of TF	0.26	2. 5	0.79	1. 9
		No of TF	21	20	20	61
^{85}Sr	C	Average of TF	0.24	0.44	0.20	0.31
		SD of TF	0.21	0.38	0.14	0.28
		No of TF	9	9	6	24
	V	Average of TF	9.2	18	5. 8	11
		SD of TF	3.0	10.	1.9	7. 9
		No of TF	9	8	9	26

* C and V represent for rice and cabbage, respectively

† Grand Total represents for all types of soils

This trend is clearly observed in the case of the Ferric Acrisol but it seems not to be true for Eutric Fluvisol and Orthi-thionic Fluvisol. For the former soil, TF values of both ^{85}Sr and ^{134}Cs to both cabbage and rice are always higher than those for the latter soils. However, the TF of ^{85} Sr from the Eutric Fluvisol to rice (0.24) is the same as it is from the Orthi-thionic Fluvisol (0.20), whereas the TF of ^{134}Cs from the former soil (9.2) is almost twice as high as that (5.8) from the latter (Table 2). In addition, the standard deviations of TF are rather wide even though on the same type of soil.

To find out the effects on TF variation of soil properties; pH, exchangeable K (cmol/kg), exchangeable Ca (cmol/kg), CEC (meq/100g), OM (%) and environmental conditions; concentration of radionuclide in soil (Bq/kg), contamination time (days), planting time (days), all the parameters were included as independent variables for regression analysis. The stepwise multiple regression method (SPSS Software Version 7.5) weighted by TF error was applied to identify which parameters are most significant in explaining the variations of TF. The criterion for selection is that at each step, the independent variable having the smallest probability of F but not in the equation will be included if that probability is sufficiently small (less than 0.05). Variables already in the regression equation are removed if their probability of F becomes sufficiently large (larger than 0.1). The method terminates when no more variables are eligible for inclusion or removal. The result is presented in Table 3. In general, the parameters, the constant, CEC, OM, pH, contamination time and concentration of radionuclides in soil can explain more than 80% of TF variance. For ^{134}Cs, the parameters involved to explain the TF variation are a little different from crop-to-crop. For cabbage, these parameters are CEC, pH, OM whereas for rice, they are pH, concentration of radionuclide in soil, contamination time.

TABLE 3. MODEL PREDICTIONS OF SOIL-TO-PLANT TF VALUES

Nuclide	Crop class	Parameters involving in explaining the variations of TF values	R^2
^{134}Cs	V (Cabbage)	Constant : 16.0±1.2 CEC : -0.182±0.044 pH : -1.392±0.176 OM : -1.296±0.309	0.801
^{134}Cs	C (Rice)	Constant: 4.150±0.226 pH : -0.467±0.037 Concentration of radionuclide in soil: -2.01E-5±3.56E-6 Contamination time: -8.67E-4±2.05E-4	0.893
^{85}Sr	V (Cabbage)	Constant : 48.0±4.4 CEC : -0.350±0.065 OM : -0.943±0.402 Contamination time: -4.28E-2±7.0 E-3 Concentration of radionuclide in soil: -0.119±0.020	0.901
^{85}Sr	C (Rice)	Constant: -0.348±0.230 CEC : -3.38E-2±4.0E-3 Contamination time: 2.78E-3±4.1E-4 OM : -0.133±0.033 Concentration of radionuclides in soil: 5.3E-04±2.4E-04	0.881

The correlation between the experimentally determined ^{134}Cs TF values and those predicted by corresponding models is give in Fig. 1 which shows that the models describe the dispersal of ^{134}Cs TF for ferralic acrisol soil rather well but not for the other types of soil (Orthi-thionic Fluvisol and Eutric Fluvisol). In addition, the correlation between the experimentally determined ^{134}Cs TF values and those predicted by the models are not quite linear.

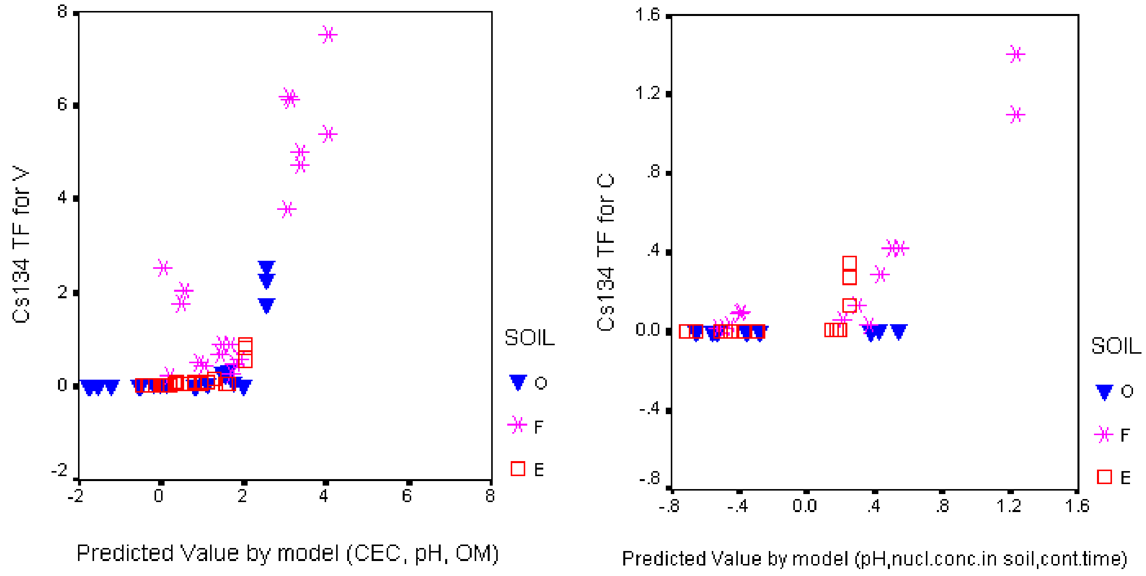

FIG. 1. The correlation between the experimentally determined ^{134}Cs TF values and those predicted by corresponding models.

For ^{85}Sr, the parameters involved in explaining the variation of TF are the same for both rice and cabbage. They are CEC, OM, contamination time, and concentration radionuclide in soil. Fig. 2 depicts the correlation between the experimentally determined ^{85}Sr TF values and those predicted by corresponding models. As seen from Fig. 2 the correlation is better than that of ^{134}Cs.

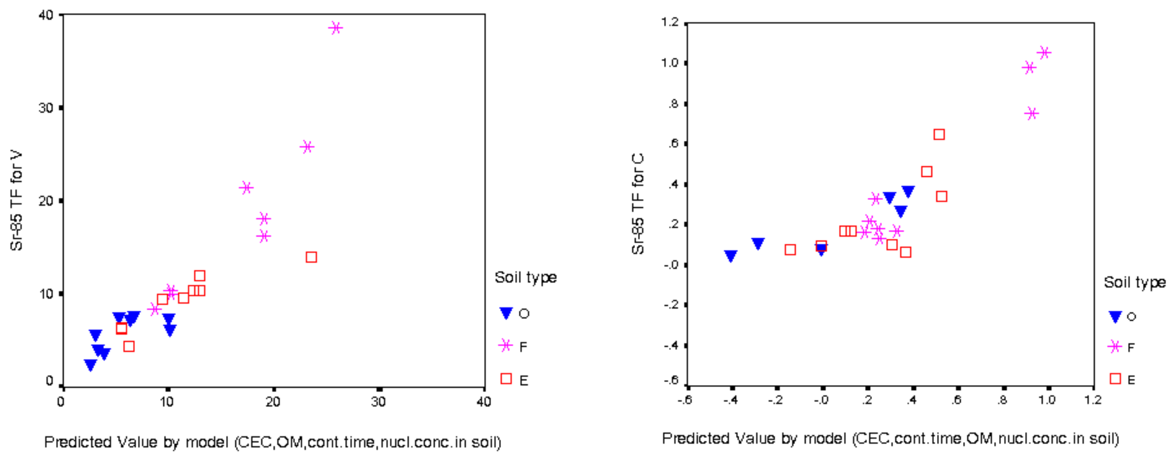

FIG. 2. The correlation between the experimentally determined ^{85}Sr TF values and those predicted by corresponding models.

To investigate the nonlinear dependence of TF values on the mentioned parameters, the TF values were transformed into logarithms and the same regression method was applied to identify the parameters involved in explaining the variation of logarithmic TF value. Table 4 summarizes the models describing the variation of logarithmic TF values. In this case, the models obtained are a little changed from those of the direct TF values, but the correlation between the experimentally determined logarithmic TF values and those predicted by corresponding models is better than the correlation between the measured TF values and their predicted values (Figs 3 and 4).

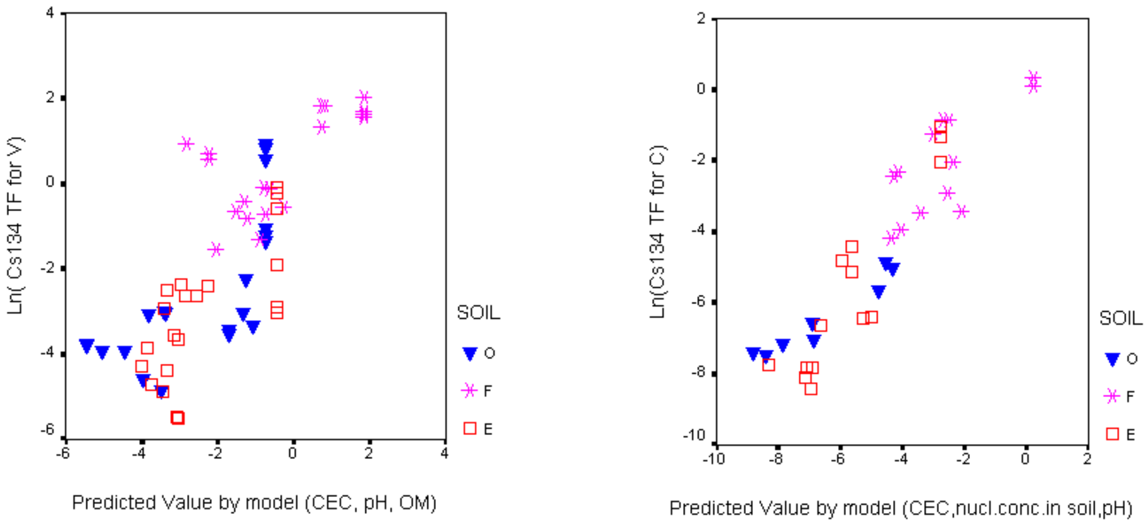

FIG. 3. The correlation between the logarithmic ^{134}Cs TF values and those predicted by corresponding models.

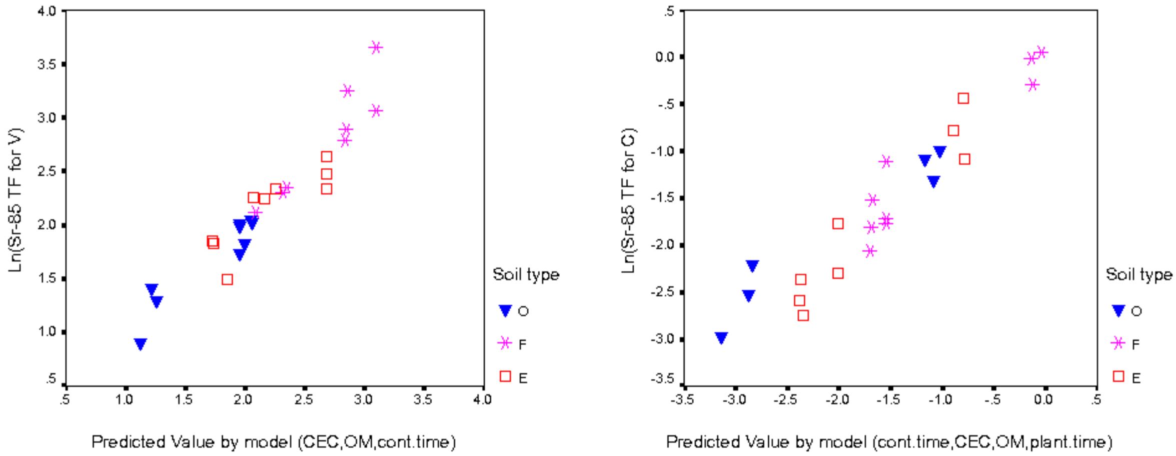

FIG. 4. The correlation between the logarithmic ^{85}Sr TF values and those predicted by corresponding models.

Table 5 shows the Pearson correlation coefficients between the experimentally determined TF values and the TF values predicted by both TF and logarithmic TF models. The logarithmic TF models describe the measured TF values better than the TF models. This proves that the dependence of TF values on the parameters is nonlinear. In [5] it states that the soil characteristics such as clay, OM, pH, exchangeable K and other environmental parameters such as concentration of radionuclide in soil and contamination time would govern the variation of ^{134}Cs TF values. However, this study found exchangeable K is not significant in governing the variation of ^{134}Cs TF value. Similarly, the role of exchangeable Ca is not significant in governing the variation of ^{85}Sr TF value. As seen in Table 4, the CEC is a factor in all models to explain the variation of logarithmic TF value.

TABLE 4. MODEL PREDICTIONS OF LOGARITHMIC TF VALUES

Radionuclide	Crop class	Parameters involving in explaining the variations of logarithmic TF values	R^2
^{134}Cs	V (Cabbage)	Constant : 5.9±1.4 CEC : -0.184±0.018 pH : -0.575±0.182 OM : -0.281±0.122	0.736
^{134}Cs	C (Rice)	Constant: 6.3±2.2 CEC : -0.232±0.023 Concentration of radionuclide in soil: -8.1E-5±1.3E-6 pH : -0.93±0.31	0.859
^{85}Sr	V (Cabbage)	Constant : 3.39±0.14 CEC : -0.047±.006 OM : -0.213±0.029 Contamination time: -3.7E-4±1.8 E-4	0.900
^{85}Sr	C (Rice)	Constant: -4.9±1.2 Contamination time: 0.005±0.001 CEC : -0.06±0.01 OM : -0.218±0.056 Planting time: 0.026±0.011	0.890

TABLE 5. PEARSON CORRELATION COEFFICIENTS BETWEEN THE MEASURED TF VALUES AND THEIR PREDICTED VALUES BY TF AND LOGARITHMIC TF MODELS

TF	^{134}Cs TF for rice (C)	^{134}Cs TF for cabbage (V)	^{85}Sr TF for rice (C)	^{85}Sr TF for cabbage (V)
TF model	0.776	0.733	0.858	0.885
Logarithmic TF model	0.944	0.840	0.965	0.897

3. CONCLUSIONS

The soil-to-plant transfer factor depends on the type of soil and has a wide range of variation. The soil-to-plant transfer factor also depends on the type of crop and type of radionuclide. The soil characteristics such as CEC, OM, pH could explain the variation of soil-to-plant transfer factor of Sr and Cs. In some cases the concentration of radionuclide in soil and the contamination time could also be involved in explaining the variation of soil-to-plant transfer factor of Sr and Cs.

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APPENDIX I

FAO/IAEA/IUR PROTOCOL FOR EXPERIMENTAL STUDIES ON THE UPTAKE OF RADIONUCLIDES FROM SOILS BY PLANTS

1. GENERAL

The aim of this protocol is to ensure that studies on the soil to plant transfer of radionuclides provide reliable data, which are suitable for radiological dose assessment calculations. The protocol should be used in combination with ISO Standard 17025 [1] and good laboratory practice.

Earlier versions of the protocol were developed by working groups of IUR and ESNA, and a more recent version was developed and used in the FAO/IAEA/IUR coordinated research project "Transfer of Radionuclides from Air, Soil and Fresh Water to the Food Chain of Man in Tropical and Subtropical Environments" These studies were focussed on equilibrium transfer parameters. They provided the data for the IUR databank [2].

The protocol should be reviewed and incorporate necessary requirements to meet specific research objectives and to comply with radiation safety considerations. The use of radionuclides for research (i.e. treatment of soil in field lysimeters or pots, soil sampling and analysis) adds a radiation exposure above that normally incurred from background radiation or increase the likelihood of exposure. Thus the principles of radiation protection and safety apply as defined in the IAEA Basic Safety Standards 115. These include: justification, dose limitation and ALARA. Safety requirements are documented in the study plan, including dose control and waste management. These should be commensurate with the use and shall conform to any requirement specified by the relevant national regulatory authority.

2. CROP AND SOIL TYPE SELECTION

2.1. Choice of crop

The choice of the crop depends entirely on the purpose of the study. Criteria used for the selection of a crop may include the following: high production, area sown to the crop, importance in the diet and amount of processing before human consumption. For most studies, suitable reference crops are grass, cereals and broad-leaved crops (e.g. cabbage). It is highly recommended that local agricultural expertise is used to assist in choosing a crop variety and that consideration of regional agricultural practice should dominate the selection process. It should be kept in mind that the uptake of radionuclides shows less crop specificity than soil specificity. If uptake is high for one crop in a particular soil system then it will be high for all crops.

The choice of the varieties should always reflect normal agricultural varieties and be representative of the region. This is essential where data are to be used for assessment calculations. Although arguments may be put forward for a group of investigators to use the same crop or variety for reasons of scientific curiosity, comparison or better replication -in the context of an assessment of the agricultural habits of a region- must dominate the selection of both the crop and the variety. If a project is limited to monitoring the same criteria for crop selection should apply.

Note that normal agricultural practice may involve a crop rotation. Consideration may therefore have to be given to implementing a similar scheme. Some crops cannot be cultivated for two or more years at the same location because of the build up of pests and diseases or a gradual decline in yield.

2.2. Choice of soil

Soils should be selected according the purpose of the study. Selected soils should be representative for the cultivated area and selected crop. Contacts with a soil extension service are recommended.

Soils should be described in two ways.

(a) According to their development using the FAO *World Reference Base for Soil Resources* (WRB) [3] (previous versions of this protocol used earlier systems). If a country does not use this system, it is advised to indicate the nearest WRB name after consulting a soil scientist.

(b) According to their texture and composition, e.g. clay, sandy loam, peat, etc. These data are relatively easy to determine, and provide a basis for a statistical analysis. The nomenclature in the WRB is insufficient on its own to allow a statistical evaluation of the effects of texture and soil composition.

It is recommended that the textural class of soil be determined by using the triangular diagram of the USDA system [4]. A hydrometer should be standardised as per USDA specification for determining the particle size distribution. The triangular diagram is available in most under-graduate textbooks on soil science.

3. TYPE OF EXPERIMENT

In a representative study of the influence of experimental variables on a terrestrial ecosystem, the system should not be disturbed by other factors. Soil-plant transfer experiments have used field conditions, lysimeter or pot experiments. There are sufficient indications that field experiments are more reliable than the other options. There is also evidence that the physical and chemical characteristics of a disturbed soil profile differ from those found in an undisturbed situation. Therefore in soil-plant studies where field experiments are not suitable, undisturbed soil monoliths are preferred to lysimeters filled with disturbed soil. However, the expense of collecting and transporting undisturbed soil monoliths may be prohibitive and disturbed soil may have to be installed in large outdoor lysimeters or large pots.

The experimental design can affect the interpretation of results. As an example, crops grown in a lysimeter can be contaminated by wind-blown soil originating from areas outside the lysimeter which contain no, or less, radioactivity than the soil within the lysimeter. Thus an underestimate of the effect of adhering soil may result. Field experiments do however, require a contaminated area. If such an area does not already exist, most countries will not allow soil to be contaminated with radionuclides such as ^{90}Sr and ^{137}Cs. If this is the case, lysimeters are advised.

If the intention is to determine the contribution of both root uptake and soil splash it will be necessary to avoid contamination of crops by rain splash. In this case it is recommended to cover the soil with a thin layer of inert material such as *perlite.*

Pot experiments generally, but not always, lead to a higher uptake than field or lysimeter studies as a result of a smaller soil volume. Roots have to be very efficient in small pots to take up sufficient nutrients and the uptake of radionuclides will therefore be higher. Observations of root uptake using relatively small pots under glasshouse conditions should always be compared with values obtained in field or lysimeter studies. Pot studies are probably not representative of field conditions. Consequently, pots should contain 8–12 kg of soil as a minimum because larger pots provide more reliable data and water shortage and nutrient deficiency problems are less likely.

The water regime may need careful attention. In some regions rainfall may exceed evapo-transpiration and drainage may be a problem (e.g. contaminated surface run-off, flooding or excessive contaminated leachate) whereas simulated rainfall may be needed in drier areas.

Consideration may need to be given to protecting the crop with netting. For example in the UK an experimental area was covered by a ‘fruit cage’; in winter months a nylon net (8.0 cm^2 mesh) which prevented pigeon damage but allowed snow accumulation, while during summer months a finer nylon net (2.5 cm^2 mesh) protected crops from small birds. Protection may also be required below ground level against burrowing animals such as rabbits.

4. EXPERIMENTAL DESIGN

4.1. Replication

A minimum of three replicates is required. If more than one parameter is tested more than three replicates are required; advice on statistical methods is recommended in this case. It is generally preferred to have uptake data for three successive growing periods. Experiments should be continued as long as possible if decreases in availability with time are studied. Five years is suggested as a minimum. If crop rotation is a common practice, then it should be included in the experimental design.

4.2. Crop production

In studies to determine site-specific transfer data the selection of varieties, fertilisation, irrigation and pesticide treatments should be according to local agricultural practice. The nutrient status of a soil influences radionuclide uptake, therefore experiments should not be better fertilised than local farmer’s fields. Should manure be required, it may be necessary to test for radionuclide content and apply a correction to the radionuclide content of the soil.

5. CHOICE OF RADIONUCLIDES AND APPLICATION

5.1. Choice of radionuclide

The choice of radionuclides depends on the purpose of the investigation. It should be kept in mind that there exist no differences of ecological behaviour between ^{133}Cs, ^{137}Cs and ^{134}Cs or between ^{90}Sr and ^{85}Sr. All reported cases of different behaviour appear to be artefacts of the experiment. Consequently, from an experimental point of view there is no preference as to which of the isotopes is used for either element. Because of the relatively short half-life of ^{134}Cs (2.06 years), the initial amount of ^{134}Cs should be higher for experiments that cover more than one year. For ^{85}Sr the half-life (64.8 days) is so short that it can only be used for one growing period. Because of recoil effects different nuclides of Am and Pu may differ in behaviour

5.2. Quantity of radionuclide

The amount of radioactivity applied should be calculated from an estimate of the uptake and yield, number of samples required, analytical sensitivity, etc. ALARA (As Low As Reasonable Achievable) dose considerations should be followed. Radionuclide concentrations that are too low can spoil a complete experiment and would certainly not be a good application of ALARA. Experience is that most people are too optimistic with respect to the counting times required.

Iodine poses a specific problem because of its volatility. Losses up to 65 percent have been noticed during uptake experiments.

5.3. Two or more radionuclides within the same experiment

Resolution of the detecting equipment may dictate how many radionuclides can be used in the same experiment. Sr and Cs can certainly be mixed. Initial use of only one of the available Sr and Cs radionuclides is recommended, because this provides an opportunity to use another radionuclide to check differences of availability with time (e.g. label first with ^{137}Cs, after two years with ^{134}Cs). Mixing of Am and of Pu with other radionuclides complicates the chemical separation. One may want to do Pu and Am studies in separate experiments.

5.4. Contamination of the soil

Almost all radionuclides are retained by soils. Pragmatically, soil scientists distinguish between adsorption and fixation. In this context adsorption is defined as a rather fast process that causes a pseudo-equilibrium between radionuclides in the soil-solution phase and solid phase within one to three months after application of the nuclides. In fact adsorption occurs continuously, long term adsorption is called fixation. Fixation may continue for many years and is the reason why the contact time, i.e. the time that the radionuclide is in contact with the soil, must always be carefully recorded. Sometimes, the term ageing is used to describe long-term decreases in the availability of radionuclides with time. As a rule of thumb, fixation is about 15% per year for Cs and 3% per year for Sr. However, for many systems these percentages may be much higher or lower.

Note that uptake experiments allowing only a very short contact time between soil and radionuclide (a few weeks or less) are in reality not very useful. In such cases direct contamination of the crop during deposition would probably have occurred and direct uptake always dominates over indirect uptake from soil.

Most assessment models consider an existing contamination of the soil, i.e., a pseudo-equilibrium situation. Radionuclides, and in particular Cs, should therefore be applied at least two months before sowing or planting. For Sr one month is sufficient.

APPENDIX II

TRANSFER FACTOR (TF) RESULTS

TABLE I. TF RESULTS Cs (Bq/kg dry crop)/(Bq/kg dry soil in 20 cm layer)

Nu..	*C*	*Crop, part*	***TF***	***TF***	***Exp.no***	*Country*	*Texture*	*Details soil*	*Soil Unit*	*Fertilizer*	*Time*	*Ex-K*	*CEC*	*pHKCl*	*OM*	*t-Co*	*t-Ex*	*Domin. clay min.*
		Al-Oudat			**A**													
Cs	C	cereals, gr	0.0019		A1	Syria	L	aridisol, Yermosol	Yermosol	non-fertile	7–30 m	0.8	20	7.9	1.3	A	F	high illite
Cs	F	alfafa	0.025		A2	Syria	L	aridisol, Yermosol	Yermosol	non-fertile	7–30 m	0.8	20	7.9	1.3	A	F	high illite
Cs	T	potato, tubers	0.027		A5	Syria	L	aridisol, Yermosol	Yermosol	non-fertile	7–30 m	0.8	20	7.9	1.3	A	F	high illite
Cs	V	cabbages	0.009		A6	Syria	L	aridisol, Yermosol	Yermosol	non-fertile	7–30 m	0.8	20	7.9	1.3	A	F	high illite
Cs	V	vegetables	0.012		A7	Syria	L	aridisol, Yermosol	Yermosol	non-fertile	7–30 m	0.8	20	7.9	1.3	A	F	high illite
Cs	X	cucumber	0.0096		A8	Syria	L	aridisol, Yermosol	Yermosol	non-fertile	7–30 m	0.8	20	7.9	1.3	A	F	high illite
Cs	C	cereals, gr	0.0016		A9	Syria	C	insiptisol, Xerosol	Xerosol	fertile	7–30 m	1.2	39	8.1	0.7	A	F	
Cs	F	alfafa	0.015		A10	Syria	C	insiptisol, Xerosol	Xerosol	fertile	7–30 m	1.2	39	8.1	0.7	A	F	
Cs	P	broad beans, pods	0.005		A11	Syria	C	insiptisol, Xerosol	Xerosol	fertile	7–30 m	1.2	39	8.1	0.7	A	F	
Cs	T	potato, tubers	0.01		A13	Syria	C	insiptisol, Xerosol	Xerosol	fertile	7–30 m	1.2	39	8.1	0.7	A	F	
Cs	V	cabbage	0.0071		A14	Syria	C	insiptisol, Xerosol	Xerosol	fertile	7–30 m	1.2	39	8.1	0.7	A	F	
Cs	V	vegetables	0.01		A15	Syria	C	insiptisol, Xerosol	Xerosol	fertile	7–30 m	1.2	39	8.1	0.7	A	F	
Cs	X	cucumber	0.0073		A16	Syria	C	insiptisol, Xerosol	Xerosol	fertile	7–30 m	1.2	39	8.1	0.7	A	F	
		Schuller			C													
Cs	C	Maize, grain	0.13		C1	Chile	L		Fluvisol, Dystric	none	5–37m	0.30	17.5	4.5	4.8	A	F	
Cs	C	Maize grain	0.03		C2	Chile	L		Fluvisol, Dystric	norm K	5–37m	0.70	17.5	4.5	4.8	A	F	
Cs	V	Chard, lvs	0.32		C3	Chile	L		Fluvisol, Dystric	none	5–37m	0.20	17.5	4.5	4.8	A	F	
Cs	V	Chard lvs	0.11		C4	Chile	L		Fluvisol, Dystric	norm K	5–37m	0.50	17.5	4.5	4.8	A	F	
Cs	V	Cabbage lvs	0.66		C5	Chile	L		Fluvisol, Dystric	none	24–37m	0.02	17.5	4.5	4.8	A	F	
Cs	V	Cabbage lvs	0.26		C6	Chile	L		Fluvisol, Dystric	norm K	24–37m	0.50	17.5	4.5	4.8	A	F	
Cs	C	Maize, grain	0.03		C7	Chile	L		Andosol, Umbric	none	5–37m	0.50	40	4.9	17.9	A	F	alophane & imogolite
Cs	C	Maize grain	0.02		C8	Chile	L		Andosol, Umbric	norm K	5–37m	1.00	40	4.9	18	A	F	
Cs	V	Chard lvs	0.25		C9	Chile	L		Andosol, Umbric	none	5–37m	0.30	40	4.9	19	A	F	
Cs	V	Chard, lvs	0.17		C10	Chile	L		Andosol, Umbric	norm K	5–37m	0.60	40	4.9	18.8	A	F	alophane & imogolite
Cs	V	Cabbage lvs	0.78		C11	Chile	L		Andosol, Umbric	none	24–37m	0.30	40	4.9	19	A	F	
Cs	V	Cabbage lvs	0.19		C12	Chile	L		Andosol, Umbric	norm K	24–37m	0.60	40	49	19	A	F	
Cs		**Djingova**			D													
Cs	C	wheat	0.018		D1	Bulgaria	L	chromic Luvisol	Luvisol		9–22 m	7.8	22	4.7	1.1	A	P	
Cs	V	cabbage	0.095		D3	Bulgaria	L	chromic Luvisol	Luvisol		9–22 m	7.8	22	4.7	1.1	A	P	
Cs	C	wheat	0.023		D4	Bulgaria	L	Eutric Fluvisol	Fluvisol		9–22 m	0.6	10	6.1	1	A	P	
Cs	V	cabbage	0.054		D6	Bulgaria	L	Eutric Fluvisol	Fluvisol		9–22 m	0.6	10	6.1	1	A	P	
		Sachdev			H													
Cs	C	mixed grain	0.0078		H1	India	S	Red Soil, Dharwar	Ferralsol	norm..dr.	6–60 m	0.36	8.66	5.5	0.66	A	P	
Cs	V	cabbage	0.0061		H2	India	S	Red Soil, Dharwar	Ferralsol	norm..dr.	6–60 m	0.347	8.66	5.5	0.66	A	P	
Cs	C	mixed grain	0.0066		H3	India	S	Alluvial soil, New Delhi	Fluvisol	norm..dr.	6–60 m	0.33	7.5	7.4	0.47	A	P	
Cs	V	cabbage	0.0098		H4	India	S	Alluvial Soil, New Delhi	Fluvisol	norm..dr.	6–60 m	0.372	8.3	7.4	0.87	A	P	
Cs	C	mixed grain	0.0086		H5	India	C	Black Soil, Indore	Vertisol	norm..dr.	6–60 m	0.459	46.1	6.4	1.82	A	P	
Cs	V	cabbage	0.0700		H6	India	C	Black soil,Indore	Vertisol	norm..dr.	6–60 m	0.52	46.1	6.4	1.81	A	P	
		LiJianguo			L													
Cs	C	wheat, grain	0.0011		L6	China	L	calcaric purplish soil	Cambisol	med. fert.	12–36m	25.9	41.1	7.4	4.3	A	F	
Cs	C	maize,grain	0.003		L7	China	L	calcaric purplish soil	Cambisol	med. fert.	8–36m	25.9	41.1	7.4	4.3	A	F	
Cs	P	broadbean,beans	0.015		L8	China	L	calcaric purplish soil	Cambisol	med. fert.	30–36m	16	27.5	7.5	3.6	A	F	
Cs	R	radish,root	0.009		L9	China	L	calcaric purplish soil	Cambisol	med. fert.	10–12m	25.9	41.1	7.4	4.3	A	F	
Cs	V	Chinese cabbage	0.05		L10	China	L	calcaric purplish soil	Cambisol	med. fert.	18–36m	17.9	23.3	7.5	5.7	A	F	
Cs	X	tomato, fruit	0.022		L11	China	L	calcaric purplish soil	Cambisol	med. fert.	15–36m	17.9	23.3	7.5	5.71	A	F	
Cs	V	Spinach, leaves	0.124		L12	China	L	calcaric purplish soil	Cambisol	med. fert.	8–9m	25.9	41.1	7.4	4.3	A	F	
Cs	V	Lettuce	0.021		L13	China	L	calcaric purplish soil	Cambisol	med fert	24–36m	20.5	37.2	7.5	6.9	F	F	
Cs	R	Potato	0.002		L14	China	L	calcaric purplish soil	Cambisol	med fert	4–12m	25.9	41.1	7.4	4.3	A	F	

TABLE I. TF RESULTS Cs (Bq/kg dry crop)/(Bq/kg dry soil in 20 cm layer)

Nu..	*C*	*Crop, part*	*TF*	*TF*	*Exp.no*	*Country*	*Texture*	*Details soil*	*Soil Unit*	*Fertilizer*	*Time*	*Ex-K*	*CEC*	*pHKCl*	*OM*	*t-Co*	*t-Ex*	*Domin. clay min.*
		Topcuoglu			O													
Cs	V	bl.cabb.lvs	0.36		O1	Turkey	S	calcic cambisol	Cambisol	productive	16 y	0.8	18	4.7	3.8	C	F	
Cs	C	maize,grain	0.0015		O2	Turkey	L_S	Rendzina, Rendzic Leptoso	Leptosol	low prodve	27–41m	1.7	47	6.3(w)	4.2	A	F	
Cs	V	bl.cabb. lvs	0.0047		O3	Turkey	L_C	Rendzina, Rendzic Leptoso	Leptosol	productive	4–16m	0.7	49	6.9	2.6	A	F	
Cs	V	bl.cabb. lvs	0.0056		O4	Turkey	L_S	Rendzina, Rendzic Leptoso	Leptosol	productive	4–39m	1.6	57	6.7	3.7	A	F	
Cs	C	maize,grain	0.0013		O5	Turkey	S	Rendzina, Rendzic Leptoso	Leptosol	low prodve	27–41m	1.4	44	6.5(w)	3.4	A	F	
Cs	V	bl. cabb. lvs	0.0038		O6	Turkey	S	Rendzina, Rendzic Leptoso	Leptosol	low prodve	20–39m	1.4	44	6.5(w)	3.4	A	F	
		Prister			P													
Cs	C	oats, grain	0.008		P1	Ukraine	C	Chernozem	Chernozem	norm. dr.	14–16 y	0.50	34.1	6.5	3.8	C	F	
Cs	C	oats, grain	0.433		P2	Ukraine	P	Histosol peat-bog	Histosol	norm. dr.	14–16 y	0.23	9.8	4.7	11.8	C	F	
Cs	C	oats, grain	0.151		P3	Ukraine	S	PodzoLuvisol	Podzol	norm. dr.	14–16 y	0.42	3.6	5.2	1	C	F	
Cs	R	beet, roots	0.042		P4	Ukraine	C	Chernozem	Chernozem	norm. dr.	14–16 y	0.53	34.7	6.5	4	C	F	
Cs	R	beet, roots	0.818		P5	Ukraine	P	Histosol peat-bog	Histosol	norm. dr.	14–16 y	0.24	10	4.8	11.95	C	F	
Cs	R	beet, roots	0.224		P6	Ukraine	S	PodzoLuvisol	Podzol	norm. dr.	14–16 y	0.43	3.8	5.2	1	C	F	
Cs	T	potatoes, tubers	0.028		P7	Ukraine	C	Chernozem	Chernozem	norm. dr.	14–16 y	0.50	32.8	6.5	3.77	C	F	
Cs	T	potatoes, tubers	0.528		P8	Ukraine	P	Histosol peat-bog	Histosol	norm. dr.	14–16 y	0.22	9.7	4.7	11.75	C	F	
Cs	T	potatoes, tubers	0.142		P9	Ukraine	S	PodzoLuvisol	Podzol	norm. dr.	14–16 y	0.40	3.6	5.2	1.05	C	F	
		Quang			Q													
Cs	C	Rice, Cereals	0.0041		[a] Q1	Vietnam	L		Fluvisols, Eutric		6–36 m	0.72	23.57	6.8	1.1	A	P	
Cs	V	Cabbage,leaves	0.0472		[a] Q2	Vietnam	L		Fluvisols, Eutric		6–36 m	1.11	20.0	7.4	2.4	A	P	
Cs	C	Rice, Cereals	0.144		[a] Q3	Vietnam	S		Acrisols, Ferralic		6–36 m	0.34	10.72	6.6	1.14	A	P	
Cs	V	Cabbage,leaves	3.80		[a] Q4	Vietnam	S		Acrisols, Ferralic		6–36 m	0.82	16.5	7.4	1.7	A	P	
Cs	C	Rice, Cereals	0.0016		[a] Q5	Vietnam	C		Fluvisols, orthi-Thionic		6–36 m	1.1	26.45	6.4	3.24	A	P	
Cs	V	Cabbage,leaves	0.2430		[a] Q6	Vietnam	C		Fluvisols, orthi-Thionic		6–36 m	1.50	29.4	6.9	4.3	A	P	
Cs	C	Rice, Cereals	0.0012		[b] Q7	Vietnam	L		Fluvisols, Eutric		16–36 m	0.72	23.57	6.8	1.1	A	P	
Cs	V	Cabbage,leaves	0.0051		[b] Q8	Vietnam	L		Fluvisols, Eutric		16–36 m	1.11	20.0	7.4	2.4	A	P	
Cs	C	Rice, Cereals	0.0457		[b] Q9	Vietnam	S		Acrisols, Ferralic		16–36 m	0.34	10.72	6.6	1.14	A	P	
Cs	V	Cabbage,leaves	2.08		[b] Q10	Vietnam	S		Acrisols, Ferralic		16–36 m	0.82	16.5	7.4	1.7	A	P	
Cs	C	Rice, Cereals	0.0016		[b] Q11	Vietnam	C		Fluvisols, orthi-Thionic		16–36 m	1.1	26.45	6.4	3.24	A	P	
Cs	V	Cabbage,leaves	0.0221		[b] Q12	Vietnam	C		Fluvisols, orthi-Thionic		16–36 m	1.50	29.4	6.9	4.3	A	P	
		Robison			R													
Cs	C	sorghum seed	15		R1	Marshall I	S	albic Arenosol (atoll soil)	Arenosol, albic		46 y	0.04		8.1	5.3	F	F	
Cs	V	KK Cross	52		R3	Marshall I	S	albic Arenosol (atoll soil)	Arenosol, albic		46 y	0.04		8.1	5.3	F	F	
Cs	V	amaranth	21		R4	Marshall I	S	albic Arenosol (atoll soil)	Arenosol, albic		46 y	0.04		8.1	5.3	F	F	
Cs	V	won bok	99		R5	Marshall I	S	albic Arenosol (atoll soil)	Arenosol, albic		46 y	0.04		8.1	5.3	F	F	
Cs	V	kai choi	24		R6	Marshall I	S	albic Arenosol (atoll soil)	Arenosol, albic		46 y	0.04		8.1	5.3	F	F	
Cs	V	mixed vegs	44		R7	Marshall I	S	albic Arenosol (atoll soil)	Arenosol, albic		46 y	0.04		8.1	5.3	F	F	
		Skarlou			S													
Cs	C	Maize, seeds	0.043		S1	Greece	L	Andosol, low productive	Andosol		8–30 m	0.4	3	5.2	0.6	A	P	
Cs	C	Maize,seeds	0.025		S2	Greece	S	Andosol, med. productive	Andosol		8–30 m	1	9	6.3	2.7	A	P	
Cs	C	Maize,seeds	0.0013		S3	Greece	C	Fluvisol, med. productive	Fluvisol		8–30 m	0.4		6.8	2	A	P	
Cs	C	Maize,seeds	0.009		S4	Greece	S	Luvisol, low productive	Luvisol		8–30 m	0.2		5	0.5	A	P	
Cs	V	mixed vegs	0.359		S5	Greece	L	Andosol, low productive	Andosol		8–30 m	0.39	3	5.2	0.6	A	P	
Cs	V	mixed vegs	0.198		S6	Greece	S	Andosol, med. productive	Andosol		8–30 m	1.06	9	6.3	2.7	A	P	
Cs	V	mixed vegs	0.021		S7	Greece	C	Fluvisol, med. productive	Fluvisol		8–30 m	0.42		6.8	2	A	P	
Cs	V	mixed vegs	0.086		S8	Greece	S	Luvisol, low productive	Luvisol		8–30 m	0.23		5	0.5	A	P	
		Twining			T													

TABLE I. TF RESULTS Cs (Bq/kg dry crop)/(Bq/kg dry soil in 20 cm layer)

Nu..	*C*	*Crop, part*	***TF***	***TF***	***Exp.no***	*Country*	*Texture*	*Details soil*	*Soil Unit*	*Fertilizer*	*Time*	*Ex-K*	*CEC*	*pHKCl*	*OM*	*t-Co*	*t-Ex*	*Domin. clay min.*
Cs	C	sorghum, grain	0.042		T1	Australia	C_L	arenic Acrisol	Acrisol	norm. dr.	5–42m	0.5	3.7	4.8	1.1	A	F	Kaolinite
Cs	C	sorghum, grain	0.049		T2	Australia	C_S	arenic Acrisol	Acrisol	norm. dr.	5–42m	0.3	2.6	5.8	0.5	A	F	Kaolinite
Cs	P	mung, seed	0.060		T3	Australia	C_L	arenic Acrisol	Acrisol	norm. dr.	5–42m	0.5	3.7	4.8	1.1	A	F	Kaolinite
Cs	P	mung, seed	0.112		T4	Australia	C_S	arenic Acrisol	Acrisol	norm. dr.	5–42m	0.3	2.6	4.8	0.5	A	F	Kaolinite
		Uchida			U													
Cs	C	rice	0.0020		U1	Japan	L	Gleysol	Gleysol	norm. dr.	short	5.5	11	5.1	1.9	F	F	
Cs	R	radish	0.0800		U2	*	L	Gleysol	Gleysol	norm. dr.	short	0.19	6.9	5.5	1.36	A	P	
Cs	C	rice	0.0017		U3		L	Andosol	Andosol	norm. dr.	short	0.6	24	4.7	3.6	F	F	
Cs	R	radish	0.1000		U4	*	L	Humic Andosol	Histosol gr.	norm. dr.	short	0.30	20	4.8	4.27	A	P	
Cs	V	mixed, spinach negl.	0.7300		U5		L	Humic Andosol	Histosol gr.	norm. dr.	short	0.3	16.6	4.7	4.27	A	P	
Cs	C	rice	0.0017		U6		L	Fluvisol	Fluvisol	norm. dr.	short	0.1	9.4	4.8	1.4	F	F	
Cs	R	radish	0.0360		U7	*	L	Eutric Fluvisol	Fluvisol, Eutric	norm. dr.	short	0.19	22.6	5.4	2.84	A	P	
Cs	R	radish	0.0190		U8	*	L	Yellow soils	Acrisol	norm. dr.	short	1.06	7.6	5.9	1.06	A	P	
		Wasserman			W													
Cs	C	maize, grain	0.38	0.38	W1	Brazil	S	Acrisol	Acrisol	norm. dr.	20 m	0.02	2.3	5.4		0.5	A	
Cs	V	cabbage	1.34	1.34	W2	Brazil	S	Acrisol	Acrisol	norm. dr.	14 m	0.02	2.3	5.4		0.5	A	
Cs	V	cabbage	0.21	0.42	W3	Brazil	L	Ferralsol	Ferralsol	norm. dr.	15 y	0.09	12	8.2		1.9	O	
		maize, grain		0.11				Ferralsol										
Cs	C	maize, grain	0.069	0.069	W4	Brazil	L	Ferralsol , Ga	Nitisol	n.dr.,pests	15 y	0.22	6.4	8.5	8.5	2.59	O	
Cs	V	cabbage	1.28	1.28	W5	Brazil	L	Ferralsol, Ga	Nitisol	n.dr.,pests	15 y	0.22	6.4	8.5		2.5	O	
Cs	C	maize, grain	0.093	0.037	W6	Brazil	L	Nitisol	Nitisol	n.dr.,pests	8 y	0.16	3.3	6.7	7.3	2.16	A	
Cs	V	cabbage	0.94	0.15	W7	Brazil	L	Nitisol	Nitisol	n.dr.,pests	9 y	0.16	3.3	7.1		2.2	O	
		Sanzharova			Z													
Cs	C	mixed cereals	0.030		Z1	Russia	L	albic Luvisol	Luvisol	med. dress.	15 y	0.70	12.3	5.6	0.9	C	F	
Cs	V	cabbage	0.091		Z2	Russia	L	albic Luvisol	Luvisol	med. dress.	15 y	0.84	18.1	5.5	1.1	C	F	
Cs	C	mixed cereals	0.005		Z3	Russia	C	calcaric Chernozem	Chernozem	high dress.	40 y	0.74	26.5	7.1	2.74	F	F	
Cs	V	cabbage	0.021		Z4	Russia	C	calcaric Chernozem	Chernozem	high dress.	40 y	1.11	38.8	7.3	3.72	F	F	
Cs	C	mixed cereals	0.007		Z5	Russia	C	chernic Chernozem	Chernozem	high dress.	15 y	1.00	41.0	6.3	3	C	F	
Cs	T	potato	0.011		Z6	Russia	C	chernic Chernozem	Chernozem	high dress.	15 y	1.10	40.0	6.5	2.8	C	F	
Cs	V	cabbage	0.024		Z7	Russia	C	chernic Chernozem	Chernozem	high dress.	15 y	1.10	40.0	6.5	2.8	C	F	
Cs	C	mixed cereals	0.031		Z8	Russia	P	Dystric Histosols	Histosol	low dress.	15 y	0.27	43.0	4.8	18.3	C	F	
Cs	V	cabbage	0.057		Z9	Russia	P	Dystric Histosols	Histosol	low dress.	15 y	0.13	17.1	4.6	6.67	C	F	
Cs	C	mixed cereals	0.077		Z10	Russia	P	histic Gleysols	Histosol gr.	low dress.	15 y	0.18	34.7	4.8	16.8	C	F	
Cs	C	mixed cereals	0.013		Z11	Russia	C	grey-luvic Phaeozem	Phaeozem	med. dress.	15 y	0.10	11.0	5.9	1.5	C	F	
Cs	V	cabbage	0.023		Z12	Russia	L	grey-luvic Phaeozem	Phaeozem	high dress.	15 y	0.50	18.0	5.5	0.7	C	F	
Cs	C	mixed cereals	0.014		Z13	Russia	C	haplic Chernozem	Chernozem	high dress.	15 y	0.90	26.0	6.3	2.1	C	F	
Cs	V	cabbage	0.041		Z14	Russia	C	haplic Chernozem	Chernozem	high dress.	15 y	1.40	42.0	6.3	3.1	C	F	
Cs	C	mixed cereals	0.008		Z15	Russia	C	haplic Kastanozems	Kastanozem	high dress.	40 y	1.56	24.9	6.5	1.9	F	F	
Cs	V	cabbage	0.023		Z16	Russia	C	haplic Kastanozems	Kastanozem	high dress.	40 y	1.70	27.0	7.6	2.1	F	F	
Cs	C	mixed cereals	0.015		Z17	Russia	L	luvic Chernozem	Chernozem	high dress.	15 y	0.70	27.0	5.8	2	C	F	
Cs	T	potato	0.130		Z18	Russia	L	luvic Chernozem	Chernozem	high dress.	15 y	0.80	33.0	5.5	2.6	C	F	
Cs	V	cabbage	0.035		Z19	Russia	L	luvic Chernozem	Chernozem	high dress.	15 y	0.90	33.0	5.3	2.6	C	F	
Cs	V	cabbage	0.165		Z20	Russia	C	molli-gleyic Fluvisol	Fluvisol	high dress.	15 y	1.77	32.6	7.2	1.9	C	F	
Cs	C	mixed cereals	0.027		Z21	Russia	S	Umbric podsols	Podzol	low dress.	15 y	1.70	14.0	5.8	1.8	C	F	
Cs	T	potato	0.053		Z22	Russia	S	Umbric podsols	Podzol	low dress.	15 y	1.20	12.0	7.1	1	C	F	
Cs	V	cabbage	0.043		Z23	Russia	S	Umbric podsols	Podzol	low dress.	15 y	0.50	11.0	7.0	0.9	C	F	

[a] Complete sampling period [b] First year not considered [c] Fertilizer gifts are expressed as dressing, to avoid confusing with fert., which has been used for fertile

TABLE II. TF RESULTS Sr (Bq/kg dry crop)/(Bq/kg dry soil in 20 cm layer)

Nu	*C*	*Crop, part*	*TF*	*Exp. no*	*Country*	*Text.*	*Details soil*	*Soil Unit*	*Time*	*Ca-ex*	*CEC*	*pHKCl*	*OM*	*t-Co*	*t-Ex*
		Al-Oudat		A											
Sr	C	cereals	0.027	A17	Syria	L	Yermosol	Yermosol	7–30 m	12	20	7.9	1.3	A	F
Sr	F	alfafa	2.4	A18	Syria	L	Yermosol	Yermosol	7–30 m	12	20	7.9	1.3	A	F
Sr	P	br beans, pods	0.053	A19	Syria	L	Yermosol	Yermosol	7–30 m	12	20	7.9	1.3	A	F
Sr	T	potato tubers	0.23	A21	Syria	L	Yermosol	Yermosol	7–30 m	12	20	7.9	1.3	A	F
Sr	V	cabbage	2.3	A22	Syria	L	Yermosol	Yermosol	7–30 m	12	20	7.9	1.3	A	F
Sr	V	mixed vegs	1.6	A23	Syria	L	Yermosol	Yermosol	7–30 m	12	20	7.9	1.3	A	F
Sr	X	cucumber	0.49	A24	Syria	L	Yermosol	Yermosol	7–30 m	12	20	7.9	1.3	A	F
Sr	C	cereals	0.014	A25	Syria	C	Xerosol	Xerosol	7–30 m	19	39	8.1	0.7	A	F
Sr	F	alfafa	0.09	A26	Syria	C	Xerosol	Xerosol	7–30 m	19	39	8.1	0.7	A	F
Sr	P	br beans pods	0.048	A27	Syria	C	Xerosol	Xerosol	7–30 m	19	39	8.1	0.7	A	F
Sr	T	potato tubers	0.079	A29	Syria	C	Xerosol	Xerosol	7–30 m	19	39	8.1	0.7	A	F
Sr	V	cabbage	2.4	A30	Syria	C	Xerosol	Xerosol	7–30 m	19	39	8.1	0.7	A	F
Sr	V	mixed vegs	0.8	A31	Syria	C	Xerosol	Xerosol	7–30 m	19	39	8.1	0.7	A	F
Sr	X	cucumber	0.41	A32	Syria	C	Xerosol	Xerosol	7–30 m	19	39	8.1	0.7	A	F
		Schuller		C											
Sr	C	maize, grain	0.03	C13	Chile	L		Dystric Fluvisol	5–37m	1.26	19	4.5	4.8	A	F
Sr	V	chard, lvs	6.5	C14	Chile	L		Dystric Fluvisol	5–37m	1.43	19	4.5	4.8	A	F
Sr	V	Cabbage lvs	3.8	C15	Chile	L		Dystric Fluvisol	25–37m	0.2–0.5	19	4.5	4.8	A	F
Sr	C	maize, grain	0.04	C16	Chile	L		Umbric Andosol	5–37m	0.3–0.6	40	4.9	19	A	F
Sr	V	chard, lvs	2.6	C17	Chile	L		Umbric Andosol	5–37m	0.3–0.6	40	4.9	19	A	F
Sr	V	cabbage lvs	7.3	C18	Chile	L		Umbric Andosol	25–37m	0.3–0.6	40	4.9	19	A	F
		Sachdev		H											
Sr	C	wheat grain	0.298	H21	India	S	Fluvisol	Fluvisol	7–60 m	6.4	8.3	7.2	0.87	A	P
Sr	C	wheat grain	0.69	H22	India	C	Vertisol	Vertisol	7–60 m	20.9	46.1	6.4	1.82	A	P
Sr	C	wheat grain	0.50	H23	India	S	Ferralsol	Ferralsol	7–60 m	5.2	8.7	5.5	0.66	A	P
Sr	V	cabbage	0.39	H24	India	S	Fluvisol	Fluvisol	7–60 m	6.4	8.3	7.2	0.87	A	P
Sr	V	cabbage	0.49	H25	India	C	Vertisol	Vertisol	7–60 m	20.1	46.1	6.4	1.82	A	P
Sr	V	cabbage	0.045	H26	India	S	Ferralsol	Ferralsol	7–60 m	5.2	8.7	5.5	0.66	A	P
		LiJianguo		L											
Sr	C	wheat, grain	0.014	L1	China	L	calcaric purplish soil	Cambisol	12–36m	25.9	41.1	7.4	4.30	A	F
Sr	C	maize,grain	0.0	L2	China	L	calcaric purplish soil	Cambisol	4–36m	25.9	41.4	7.4	4.30	A	F
Sr	P	broadbean	0.11	L3	China	L	calcaric purplish soil	Cambisol	30–36m	16.0	27.5	7.5	3.60	A	F
Sr	R	radish, root	0.780	L4	China	L	calcaric purplish soil	Cambisol	10–12m	25.9	41.1	7.4	4.30	A	F

TABLE II. TF RESULTS Sr (Bq/kg dry crop)/(Bq/kg dry soil in 20 cm layer)

Nu	*C*	*Crop, part*	*TF*	*Exp. no*	*Country*	*Text.*	*Details soil*	*Soil Unit*	*Time*	*Ca-ex*	*CEC*	*pHKCl*	*OM*	*t-Co*	*t-Ex*
Sr	V	Chinese cabbage	1.07	L5	China	L	calcaric purplish soil	Cambisol	18–36m	17.9	23.3	7.5	5.70	A	F
Sr	X	tomato, fruit	0.03	L6	China		calcaric purplish soil	Cambisol	15–36m	17.9	23.3	7.5	5.70	A	F
Sr	V	spinach, leaves	0.42	L7	China	L	calcaric purplish soil	Cambisol	8–9m	25.9	41.1	7.4	4.30	A	F
Sr	V	lettuce	0.06	L8	China	L	calcaric purplish soil	Cambisol	24–36m	20.5	37.2	7.5	6.90	A	F
Sr	R	potato, root	0.01	L9	China	L	calcaric purplish soil	Cambisol	4–12m	25.9	41.1	7.4	4.30	A	F
		Topcuoglu		O											
Sr	V	bl. cabb. lvs	0.99	O11	Turkey	L	Rendzic Leptosol	leptosol	4–20 m			5.7	4.2	A	F
Sr	V	bl. cabb. lvs	0.84	O12	Turkey	S	Rendzic Leptosol	leptosol	4–20 m			5.9	3.4	A	F
		Prister		P											
Sr	C	oats, grain	0.102	P10	Ukraine	C	Chernozem	Chernozem	14 –16 y	23.3	34.1	6.5	3.8	C	F
Sr	C	oats, grain	0.123	P11	Ukraine	P	Histosol peat-bog	Histosol	14 –16 y	5.2	9.8	4.7	11.8	C	F
Sr	C	oats, grain	0.696	P12	Ukraine	S	PodzoLuvisol	PodzoLuvisol	14 –16 y	2.9	3.6	5.2	1.0	C	F
Sr	R	beet, roots	0.556	P13	Ukraine	C	Chernozem	Chernozem	14 –16 y	24.0	34.7	6.5	4.0	C	F
Sr	R	beet, roots	0.427	P14	Ukraine	P	Histosol peat-bog	Histosol	14 –16 y	5.3	10.0	4.8	12.0	C	F
Sr	R	beet, roots	1.340	P15	Ukraine	S	PodzoLuvisol	PodzoLuvisol	14 –16 y	3.1	3.8	5.2	1.0	C	F
Sr	T	potatoes, tubers	0.082	P16	Ukraine	C	Chernozem	Chernozem	14 –16 y	22.8	32.8	6.6	3.8	C	F
Sr	T	potatoes, tubers	0.103	P17	Ukraine	P	Histosol peat-bog	Histosol	14 –16 y	5.2	9.7	4.7	11.8	C	F
Sr	T	potatoes, tubers	0.417	P18	Ukraine	S	PodzoLuvisol	PodzoLuvisol	14 –16 y	2.9	3.6	5.2	1.1	C	F
		Quang		Q											
Sr	C	rice	0.17	Q21	Vietnam	L		Eutric Fluvisols	3–20 m	18.8	22.9	6.8	0.83	A	P
Sr	V	cabbage	12	Q22	Vietnam	L		Eutric Fluvisols	3–20 m	18.8	22.9	7.3	1.61	A	P
Sr	C	rice	0.32	Q23	Vietnam	S		Ferralic Acrisols	3–20 m	5.6	8.72	6.7	0.92	A	P
Sr	V	cabbage	29	Q24	Vietnam	S		Ferralic Acrisols	3–20 m	5.6	8.72	5.2	0.61	A	P
Sr	C	rice	0.16	Q25	Vietnam	C		Thionic Fluvisols	3–20 m	10.2	19.2	6.2	2.41	A	P
Sr	V	cabbage	6.8	Q26	Vietnam	F		Thionic Fluvisols	3–20 m	10.2	19.2	4.1	2.91	A	P
		Twining		T											
Sr	C	sorghum, gr.	0.20	T5	Australia	CL	arenic Acrisol	Acrisol	5–42m	2.0	3.7	4.8	1.1	A	F
Sr	C	sorghum, gr.	0.17	T6	Australia	CS	arenic Acrisol	Acrisol	5–42m	1.4	2.6	5.8	0.5	A	F
Sr	P	mung, seed	1.7	T7	Australia	CL	arenic Acrisol	Acrisol	5–42m	2.0	3.7	4.8	1.1	A	F
Sr	P	mung, seed	1.3	T8	Australia	CS	arenic Acrisol	Acrisol	5–42m	1.4	2.6	5.8	0.5	A	F
		Uchida		U											
Sr	C	Rice	0.020	U10	Japan	L	Gleysol	Gleysol	short		11	5.1	1.9	F	F
Sr	R	radish	0.260	U11	Japan	L	Gleysol	Gleysol	short	3.8	6.9	5.6	1.36	A	P
Sr	C	Rice	0.007	U12	Japan	L	Andosol	Andosol	short	3.2	22.0	4.9	6.4	F	F

TABLE II. TF RESULTS Sr (Bq/kg dry crop)/(Bq/kg dry soil in 20 cm layer)

Nu	*C*	*Crop, part*	*TF*	*Exp. no*	*Country*	*Text.*	*Details soil*	*Soil Unit*	*Time*	*Ca-ex*	*CEC*	*pHKCl*	*OM*	*t-Co*	*t-Ex*
Sr	R	radish	0.390	U13	Japan	L	Humic Andosol	Histosol gr	short	9.9	7.6	5.9	1.06	A	P
Sr	V	Lettuce	1.790	U14	Japan	L	Humic Andosol	Histosol gr	short	2.4	16.6	4.7	4.27	A	P
Sr	C	Rice	0.011	U15	Japan	L	Fluvisol	Fluvisol	short	6.5	9.4	4.8	1.4	F	F
Sr	R	radish	0.180	U16	*	L	Eutric Fluvisol	Eutric Fluvisol	short	16.3	22.6	5.4	2.84	A	P
		Wasserman		W											
Sr	C	maize gr.	0.078	W11	Brazil	S		Acrisol	4–20 m	1.0	6.32	5.40	0.5	A	L
Sr	V	cabbage	2.0	W12	Brazil	S		Acrisol	4–20 m	1.0	6.32	5.4	0.5	A	L
Sr	C	Maize,gr	0.020	W13	Brazil	S		Ferralsol	4–20 m	4.4	5	7.1	2.2	A	L
Sr	V	cabbage	0.57	W14	Brazil	S		Ferralsol	4–20 m	3.3	5	5.5	2.1	A	L
Sr	C	Maize,gr	0.0050	W15	Brazil	S		Nitisol	4–20 m	4.0	12	4.2	2.1	A	L
Sr	V	cabbage	0.13	W16	Brazil	S		Nitisol	4–20 m	4.0	12	4.2	2.1	A	L
		Sanzharova		Z											
Sr	C	w. wheat, grain	0.19	Z31	Russia	L	albic Luvisols	Luvisol	50 y	12.0	16.0	5.5	0.7	F	F
Sr	V	cabbage	0.92	Z32	Russia	L	albic Luvisols	Luvisol	15 y	10.0	17.0	5.5	0.9	C	F
Sr	C	mixed cereals	0.038	Z33	Russia	C	Calcaric Chernozem	Chernozem	40 y	18.6	26.5	7.1	2.74	F	F
Sr	V	cabbage	0.14	Z34	Russia	C	Calcaric Chernozem	Chernozem	40 y	22.8	38.8	7.3	3.72	F	F
Sr	C	mixed cereals	0.051	Z35	Russia	C	chernic Chernozem	Chernozem	50 y	23.0	34.0	5.6	2.8	F	F
Sr	T	potato	0.052	Z36	Russia	C	chernic Chernozem	Chernozem	50 y	24.0	38.0	6.3	2.8	F	F
Sr	V	cabbage	0.34	Z37	Russia	C	chernic Chernozem	Chernozem	50 y	32.0	40.0	6.4	2.8	F	F
Sr	C	mixed cereals	0.39	Z38	Russia	P	Dystric Histosols	Histosol	15 y	11.4	43.0	4.8	18.3	C	F
Sr	V	cabbage	2.1	Z39	Russia	P	Dystric Histosols	Histosol	15 y	7.0	17.1	4.6	6.67	C	F
Sr	C	mixed cereals	0.14	Z40	Russia	L	grey-luvic Phaeozem	Phaeozem	15 y	13.0	18.0	5.4	1	C	F
Sr	V	cabbage	0.78	Z41	Russia	L	grey-luvic Phaeozem	Phaeozem	50 y	12.0	18.0	5.5	0.7	F	F
Sr	C	mixed cereals	0.077	Z42	Russia	C	haplic Chernozem	Chernozem	15 y	18.8	31.3	6.3	2.1	C	F
Sr	T	potato	0.051	Z43	Russia	C	haplic Chernozem	Chernozem	50 y	32.0	42.0	6.6	3.1	F	F
Sr	V	cabbage	0.50	Z44	Russia	C	haplic Chernozem	Chernozem	15 y	30.4	42.1	6.3	3.1	C	F
Sr	V	cabbage	0.30	Z45	Russia	C	haplic Kastanozems	Kastanozem	40 y	20.0	27.0	7.6	2.1	F	F
Sr	C	mixed cereals	0.21	Z46	Russia	P	Eutri-Histic Gleysols	histic Gleysol	15 y	5.9	34.7	4.8	16.8	C	F
Sr	C	mixed cereals	0.10	Z47	Russia	L	luvic Chernozem	Chernozem	15 y	18.0	27.0	5.8	2	C	F
Sr	V	cabbage	0.69	Z48	Russia	L	luvic Chernozem	Chernozem	15 y	20.0	33.0	5.3	2.6	C	F
Sr	V	cabbage	1.3	Z49	Russia	C	molli-gleyic Fluvisols	Fluvisol	15 y	15.0	33.0	7.2	1.9	C	F
Sr	C	mixed cereals	0.14	Z50	Russia	S	Umbric Podzols	Podzol	15 y	4.0	11.0	5.9	1.5	C	F
Sr	T	potato	0.12	Z51	Russia	L	Umbric Podzols	Podzol	15 y	6.0	10.0	7.3	1.8	C	F
Sr	V	cabbage	0.84	Z52	Russia	L	Umbric Podzols	Podzol	15 y	6.0	8.0	4.6	1.3	C	F

TABLE III. TF values of Cs for cereals derived from IUR data bank and earlier CRP (Bq/kg dry crop)/(Bq/kg dry soil in 20 cm layer)

Nu		crop, part	TF	*Exp.no*	*Country*	Text-1	Text-2	Soil unit	Source	Ex-K	pH(KCl)	Investigator
Cs	C	Rice	0.093	n1	Vietnam	P	S	Ferralsol	earlier CRP		4.9	Ngo, Vietnam
Cs	C	Rice	0.018	u1	Japan	L	C	Andosol	earlier CRP	0.28	5.8	Uchida, Japan
Cs	C	Rice	0.011	u2	Japan	L	C	Eutric Gleysol	earlier CRP	0.29	5.5	Uchida, Japan
	C											
Cs	C	barley	0.270	e1	Sweden	P		Histosol	IUR		5.5	Eriksson, Sweden
Cs	C	barley	0.018	e2	Sweden	P		Histosol	IUR		5.8	Eriksson, Sweden
Cs	C	wheat	0.055	e3	Sweden	P		Histosol	IUR		5.5	Eriksson, Sweden
Cs	C	barley	0.08	e4	Sweden	S	P	Histosol	IUR		5.0	Eriksson, Sweden
Cs	C	wheat	0.0012	s1	Germany	L		Luvisol	IUR		5.9	Steffens, Germ.
Cs	C	wheat	0.042	s2	Germany	S		Podzol	IUR		4.7	Steffens, Germ.
Cs	C	barley	0.0054	f1	Netherl.	C		Eutric Fluvisol	IUR		6.8	Frissel, Neth.
Cs	C	barley	0.0039	f2	Netherl.	L		orthic Luvisol	IUR		6.8	Frissel, Neth.
Cs	C	barley	0.031	f3	Netherl.	S		Eutric Fluvisol	IUR		5.6	Frissel, Neth.
Cs	C	Hyp. value	0.012	f4	Netherl.	S		gleyic Podzol	IUR		5.8	Frissel, Neth.
Cs	C	Hyp. value	0.0063	f5	Netherl.	C		Eutric Fluvisol	IUR		7.4	Frissel, Neth.
Cs	C	Hyp. value	0.0055	f6	Netherl.	S	P	Dystric Histosol	IUR		5.5	Frissel, Neth.
Cs	C	Hyp. value	0.0042	f7	Netherl.	S	P	Eutric Histosol	IUR		5.6	Frissel, Neth.
Cs	C	Hyp. value	0.0097	f8	France	L		orthic Luvisol	IUR		6.5	Frissel, France
Cs	C	Hyp. value	0.017	f9	France	L		Rendzina	IUR		7.3	Frissel, France
Cs	C	Hyp. value	0.023	f10	Germany	S		humic Podzol	IUR		5.9	Frissel, Germ.
Cs	C	Hyp. value	0.011	f11	UK	L		Rendzina	IUR		7.3	Frissel, UK

Text-1 is main texture

Text-2 is additional texture

The hypothetical values are derived from grass values using a conversion ratio of 5.

TABLE IV. TF results of Co, I, K-40, Mn, Pb, Po, Th, U, Zn (Bq/kg dry crop)/(Bq/kg dry soil in 20 cm layer)

Nucl.	C	Crop, part	TF	exp no	Country	Text.	Remarks	Soil unit	ex-Ca	ex-K	CEC	pHKCl	OM	t-Co	t-Ex	t-Fa	Time
		TF RESULTS Co															
		Djingova															
Co	C	wheat	0.005	D7	Bulgaria	L	norm. dr.	Chromic Luvisol	9.8	7.8	22	4.7	1.1	A	P	I	9–22 m
Co	S	straw	0.012	D8	Bulgaria	L	norm. dr.	Chromic Luvisol	9.8	7.8	22	4.7	1.1	A	P	I	9–22 m
Co	V	cabbage	0.048	D9	Bulgaria	L	norm. dr.	Chromic Luvisol	9.8	7.8	22	4.7	1.1	A	P	I	9–22 m
Co	C	wheat	0.017	D10	Bulgaria	L	norm. dr.	Eutric Fluvisol	7.1	0.6	10	6.1	1	A	P	I	9–22 m
Co	S	straw	0.018	D11	Bulgaria	L	norm. dr.	Eutric Fluvisol	7.1	0.6	10	6.1	1	A	P	I	9–22 m
Co	V	cabbage	0.036	D12	Bulgaria	L	norm. dr.	Eutric Fluvisol	7.1	0.6	10	6.1	1	A	P	I	9–22 m
		Wasserman															
Co	C	maize, grain	0.021	W30	Brazil	S		Acrisol, ultisol	0.8	0.02	2.3	4.0	0.5	A	L		1-2 y
Co	V	cabbage	0.049	W31	Brazil	S		Acrisol, ultisol	0.8	0.02	2.3	4.0	0.5	A	L		1-2 y
Co	V	cabbage	0.250	W32	Brazil	C		Ferralsol	0.8	0.04	5.2	4.5	1.6	A	L		1-2 y
Co	C	maize, grain	0.012	W33	Brazil	C		Nitisol, oxisol	0.4	0.01	5.2	4.5	1.6	A	L		1-2 y
Co	V	cabbage	0.073	W34	Brazil	C		Nitisol, oxisol	0.4	0.01	5.2	4.5	1.6	A	L		1-2 y
		Uchida															
Co	R	radish	0.0014	U201	Japan	S	high dr.	Acrisol	9.9	1.06	7.6	5.91	1.06	A	P	I	1 y
Co	R	radish	0.0034	U202	Japan	S	high dr.	Eutric Fluvisol	16	0.19	23	5.4	2.84	A	P	I	2 y
Co	R	radish	0.0110	U203	Japan	S	high dr.	Gleysol	3.8	0.19	6.9	5.6	1.36	A	P	I	2 y
Co	R	radish	0.0110	U204	Japan	L	high dr.	Humic Andosol	2.4	0.3	20	4.8	4.27	A	P	I	1 y
Co	V	Cabbage	0.1600	U205	Japan	L	high dr.	Humic Andosol	2.4	0.3	16.6	4.7	4.27	A	P	I	3 y
		TF RESULTS I															
		Uchida															
I	C	wheat	0.0002	U207	Japan		high dr.	Humic Andosol			37	4.9	9	A	P	I	8 y
I	R	radish	0.026	U208	Japan		high dr.	Humic Andosol			37	4.9	9	A	P	I	1 y
I	V	lettuce	0.0041	U209	Japan		high dr.	Humic Andosol			37	4.9	9	A	P	I	3 y
		TF RESULTS K-40															
		Wasserman															
Nucl.	C	Crop, part	TF	exp no		text.	remarks	soil unit	ex-Ca	ex-K	CEC	pHKCl	OM	t-Co	t-Ex		time
K-40	V	cabbage	22	W30	Brazil	L		Ferralsol	11.1	0.09	12	8.2	1.9	O	L		
K-40	C	maize, grain	4	W31	Brazil	L		Nitisol, oxisol	2.3	0.16	3.3	6.7	2.155	A	L		8 y
K-40	F	maize, leaves	8	W32	Brazil	L		Nitisol, oxisol	1.8	0.09	2.9	6.2	2	A	L		8 y
K-40	V	cabbage	23	W33	Brazil	L		Nitisol, oxisol	2.3	0.16	3.3	7.1	2.2	O	L		9 y
K-40	C	maize, grain	3	W34	Brazil	L	Goiânia	Nitisol, oxisol	4.9	0.22	6.4	8.5	2.586	O	L		13 y
K-40	F	maize, leaves	5	W35	Brazil	L	Goiânia	Nitisol, oxisol	4.9	0.22	6.4	8.5	1.948	O	L		13 y

TABLE IV. TF results of Co, I, K-40, Mn, Pb, Po, Th, U, Zn (Bq/kg dry crop)/(Bq/kg dry soil in 20 cm layer)

Nucl.	C	Crop, part	TF	exp no	Country	Text.	Remarks	Soil unit	ex-Ca	ex-K	CEC	pHKCl	OM	t-Co	t-Ex	t-Fa	Time
		TF RESULTS Mn															
Nucl.	C	Crop, part	TF	exp no		text.	remarks	soil unit	ex-Ca	ex-K	CEC	pHKCl	OM	t-Co	t-Ex	t-Fa	Time
		Djingova															
Mn	C	wheat	0.038	D13	Bulgaria	L	norm dr	Chromic Luvisol	9.8	7.8	22	4.7	1.1	A	P	I	9–22 m
Mn	S	straw	0.037	D14	Bulgaria	L	norm dr	Chromic Luvisol	9.8	7.8	22	4.7	1.1	A	P	I	9–22 m
Mn	V	cabbage	0.14	D15	Bulgaria	L	norm dr	Chromic Luvisol	9.8	7.8	22	4.7	1.1	A	P	I	9–22 m
Mn	C	wheat	0.11	D16	Bulgaria	L	norm dr	Eutric Fluvisol	7.1	0.6	10	6.1	1	A	P	I	9–22 m
Mn	S	straw	0.115	D17	Bulgaria	L	norm dr	Eutric Fluvisol	7.1	0.6	10	6.1	1	A	P	I	9–22 m
Mn	V	cabbage	0.099	D18	Bulgaria	L	norm dr	Eutric Fluvisol	7.1	0.6	10	6.1	1	A	P	I	9–22 m
		Uchida															
Mn	R	radish	0.0039	U211	Japan		high dr.	Acrisol	12	0.67	8	5.7	1.32	A	P	I	2 m
Mn	R	radish	0.0080	U212	Japan		high dr.	Eutric Fluvisol	16	0.19	23	5.4	2.84	A	P	I	2 m
Mn	R	radish	0.054	U213	Japan		high dr.	Gleysol	3.79	0.19	6.9	5.6	1.36	A	P	I	2 m
Mn	R	radish	0.10	U214	Japan	L	high dr.	Humic Andosol	2.4	0.3	20	4.8	4.27	A	P	I	2 m
Mn	V	Cabbage	3.8	U215	Japan	L	high dr.	Humic Andosol	2.4	0.3	16.6	4.7	4.27	A	P	I	2 m
		TF RESULTS Pb															
		Uchida															
Pb	C	Rice	0.0014	U217	Japan	L	high dr.	Andosol	15	0.5	22	5.4	4.5		F	F	
Pb	C	Rice	0.0014	U218	Japan	C	high dr.	Fluvisol	12	0.3	25	4.9	2.4		F	F	
Pb	X	mixed fruit	0.014	U219	Japan	L	high dr.	Acrisol	10	1.8	20	5.6	1.0		F	D	
Pb	X	mixed fruit	0.00042	U220	Japan		high dr.	Andosol	10	1.5	10	5.6	1.7		F	D	
Pb	X	mixed fruit	0.0063	U221	Japan	L	high dr.	Cambisol	4.4	0.70	38	4.1	5.1		F	D	
Pb	X	mixed fruit	0.00060	U222	Japan	L	high dr.	Fluvisol	9.5	1.8	7.8	5.9	1.3		F	D	
		TF RESULTS Po															
		Topcuoglu															
Po	V	bl.cabb. lvs	0.067	O22	Turky	L	r. leptosol	Rendzina	36	0.7		6.3	2.6	On	Fs		
Type of contamination:On = natural																	
Type of experiment: Fs = Sheltered area																	
		TF RESULTS Th															
		Uchida															
Th	C	Rice	0.000053	U224			high dr.	Andosol	15	0.5	22	5.4	4.5		F	F	
Th	C	Rice	0.000042	U225		C	high dr.	Fluvisol	12	0.3	25	4.9	2.4		F	F	
Th	X	mixed fruit	0.000015	U226		L	high dr.	Acrisol	10	1.8	20	5.6	1		F	D	
Th	X	mixed fruit	0.000028	U227		S	high dr.	Andosol	10	1.5	10	5.6	1.7		F	D	
Th	X	mixed fruit	0.000033	U228		L	high dr.	Cambisol	4.4	0.7	38	4.1	5.1		F	D	
Th	X	mixed fruit	0.000017	U229		L	high dr.	Fluvisol	9.5	1.8	7.8	5.9	1.3		F	D	

TABLE IV. TF results of Co, I, K-40, Mn, Pb, Po, Th, U, Zn (Bq/kg dry crop)/(Bq/kg dry soil in 20 cm layer)

Nucl.	C	Crop, part	TF	exp no	Country	Text.	Remarks	Soil unit	ex-Ca	ex-K	CEC	pHKCl	OM	t-Co	t-Ex	t-Fa	Time
		TF RESULTS U															
		Uchida															
U	C	Rice	0.000047	U231		L	high dr.	Andosol	15	0.5	22	5.4	4.5		F	F	
U	C	Rice	0.000041	U232			high dr.	Fluvisol	12	0.3	25	4.9	2.4		F	F	
U	X	mixed fruit	0.000034	U233		L	high dr.	Acrisol	10	1.8	20	5.6	1		F	D	
U	X	mixed fruit	0.000034	U234			high dr.	Andosol	10	1.5	10	5.6	1.7		F	D	
U	X	mixed fruit	0.000052	U235		L	high dr.	Cambisol	4.4	0.7	38	4.1	5.1		F	D	
U	X	mixed fruit	0.00003	U236			high dr.	Fluvisol	9.5	1.8	7.8	5.9	1.3		F	D	
		TF RESULTS Zn															
		Twining															
Zn	C	Sorghum, grain	10.1	T9		C_L	normal dr.	Ferric Acrisol	0.7	0.7	1	4.7	4.6	A	F	D	5–17 m
Zn	C	Sorghum, grain	10.7	T10		C_S	normal dr.	Ferric Acrisol	0.6	0.4	2.3	5.2	2	A	F	D	5–17 m
Zn	P	Mung, seed	14.5	T11		C_L	normal dr.	Ferric Acrisol	0.7	0.7	1.5	4.7	4.6	A	F	D	5–17 m
Zn	P	Mung, seed	15.8	T12		C_S	normal dr.	Ferric Acrisol	0.7	0.4	2.2	6.1	2	A	F	D	5–17 m
		Uchida															
Zn	R	radish	0.15	U238		L	high dr.	Acrisol	9.9	1.1	7.6	5.9	1.06	A	P	I	1
Zn	R	radish	0.52	U239		L	high dr.	Eutric Fluvisol	16	0.19	23	5.4	2.84	A	P	I	2 y
Zn	R	radish	0.6	U240		L	high dr.	Gleysol	3.8	0.19	6.9	5.6	1.36	A	P	I	2 y
Zn	R	radish	0.53	U241		L	high dr.	Humic Andosol	2.4	0.3	20	4.8	4.27	A	P	I	1 y
Zn	V	Cabbage	2.5	U242		L	high dr.	Humic Andosol	2.4	0.3	17	4.7	4.27	A	P	I	3 y

APPENDIX III

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Results of the Proficiency Test for the IAEA CRP: Classification of Soil Systems based on Transfer Factors of Radionuclides from Soil to Reference Plants

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http:/www.iaea.org/programmes/aqcs

Objectives of the Study

- To assess the performance of laboratories measuring strontium-90 and gamma-emitting radionuclides in natural samples.

- To provide analysts with a means whereby they can check their methods, identify sources of bias and, where necessary, take corrective action to improve their level of performance.

Matrix & Sample Sets

Matrix	Soil	Cabbage
Spiked sample size (No of samples)	γ – 100 g (1) ^{90}Sr – 10 g (3)	γ – 200 g (1) ^{90}Sr – 50 g (3)
Sample blank	γ – none ^{90}Sr – 30 g	γ – none ^{90}Sr – 150 g
Standard solution	γ – 2.5 ml ^{90}Sr – 2.5 ml	

Choice of Radionuclides and Activity Levels

Radionuclides of interest	γ ^{54}Mn, ^{57}Co, ^{60}Co, ^{65}Zn, ^{88}Y, ^{134}Cs, ^{137}Cs, ^{152}Eu β ^{90}Sr
Activity Levels in spiked samples	γ 10 - 400 Bq/kg d.w. β 1 - 2.2 Bq per sample
Activity Levels in the standard solution	γ 20 - 150 Bq/g β 7.72 Bq/g
Manufacturers of the standard solutions	NIST (USA), CERCA LEA (France), POLATOM (Poland)

Origin and Characteristics of Samples

- Two natural samples, soil from China and cabbage from Austria were used to prepare samples for analysis.

- Three 10g soil samples (coded from B1 to B3) and three 50g cabbage samples (coded from B5 to B7) were spiked with known amounts of a certified standard solution containing ^{90}Sr radionuclide. No attempt was made to perfectly homogenize the samples after spiking, therefore analysts were requested to perform measurements on the entire sample which had to be transferred quantitatively from the bottle.

- Small batches of two selected materials (100g of soil and 200g of cabbage) were spiked separately with known amounts of a certified standard solution containing a mixture of gamma-emitting radionuclides. After spiking the samples were homogenized. Each sample was measured to ensure that the material could be considered homogeneous for the purpose of this exercise. Samples were coded G1 (spiked soil) and G2 (spiked cabbage).

- Two customized standard solutions containing ^{90}Sr and mixed gamma-emitting radionuclides (^{54}Mn, ^{57}Co, ^{60}Co, ^{65}Zn, ^{88}Y, ^{134}Cs, ^{137}Cs and ^{152}Eu) were prepared.

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Radionuclide Standards

NIST SRM 4234A

- ^{90}Sr radioactivity standard (2.494 MBq g^{-1}; u = 0.28 %)

CERCA LEA

- MN54-ELSA50, ^{54}Mn radioactivity standard (7.23 MBq g^{-1}; u = 0.5 %)
- Y88-ELSA50, ^{88}Y radioactivity standard (10.2 MBq g^{-1}; u = 0.75 %)
- EU152-ELSB45, ^{152}Eu radioactivity standard (624 kBq g^{-1}; u = 0.75 %)

POLATOM

- SRCo-66, ^{57}Co radioactivity standard (53.8 kBq g^{-1}; u = 0.6 %)
- SRCo-43, ^{60}Co radioactivity standard (53.4 kBq g^{-1}; u = 0.4 %)
- SRZn-6, ^{65}Zn radioactivity standard (46.2 kBq g^{-1}; u = 1.8 %)
- SRCs-21, ^{134}Cs radioactivity standard (53.1 kBq g^{-1}; u = 0.5 %)
- SRCs-23, ^{137}Cs radioactivity standard (41.1 kBq g^{-1}; u = 1.5 %)

Performance Indicators

Relative bias and ratio

$$Bias \quad [\%] = \frac{x_{lab} - X_{ref}}{X_{ref}} \times 100$$

$$Ratio = \frac{x_{lab}}{X_{ref}}$$

where x_{lab} is a value reported by the analyst; X_{ref} is a target value assigned by the IAEA

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Performance Indicators: *z-Score*

The z-Score value is calculated from to the following equation:

$$z - Score = \frac{x_{lab} - X_{ref}}{\sigma}$$

where the target value for the standard deviation (σ) is assigned by the organizer according to the concentration level of the analyte.

Performance Indicators: z-Score

The target values for the standard deviation (σ) have been assigned on the basis of the Horwitz function[1],[2]. This formula allows estimation of the reproducibility standard deviation expected in a collaborative trial (the standard deviation of the robust mean which expresses inter-laboratory precision).

The values for the constants A and B were derived on the basis of data collected during worldwide interlaboratory studies organized by the IAEA during 1999 – 2003.

[1] W. Horwitz, L. R. Kamps and K. W. Boyer, J. Assoc. Off. Anal. Chem., 1980, **63**, 1344.
[2] M. Thompson, Analyst, 2000, **125**, 385

Performance Indicators

The target values for the standard deviation calculated for this exercise

Analyte	Sample G1	Sample G2	Sample G3
^{54}Mn	14%	-	6%
^{57}Co	15%	9%	5%
^{60}Co	12%	8%	4%
^{65}Zn	16%	9%	6%
^{88}Y	8%	-	4%
^{134}Cs	14%	8%	5%
^{137}Cs	12%	8%	5%
^{152}Eu	-	-	7%
Analyte	**Samples B1, B2, B3**	**Samples B5, B6, B7**	**Sample B9**
^{90}Sr	34%	30%	10%

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How Can Interlaboratory Studies Contribute Towards Quality of Analytical Data?

Interlaboratory studies are primarily designed to test the accuracy of a laboratory's performance.

The evaluation of measurement uncertainty and the judgment of participants uncertainty statements represent an untapped source of valuable information from interlaboratory studies.

Uncertainty evaluation in proficiency tests

Currently test results are at best, accompanied with an indication of their repeatability only and usually provide no indication of their analytical uncertainty. However, new requirements coming into force (ISO 17025) will oblige laboratories to express their measurement uncertainty.

Consequently, PT evaluation procedures will have to be redesigned to incorporate uncertainty statements provided by the PT organizer and as reported by participating laboratories.

Performance Indicators: u-Score

The value of the u-test score is calculated according to the following equation[1]:

$$u \quad Score = \frac{x_{lab} - X_{ref}}{\sqrt{u_{lab}^2 + u_{ref}^2}}$$

where x_{lab} is a value reported by the analyst; X_{ref} is a target value and u_{lab} and u_{ref} are the corresponding combined standard uncertainties

[1] Brookes, C.J., Betteley, I.G. and Loxton, S.M.; Fundamentals of Mathematics and Statistics, Wiley, UK 1979.

u-Score Acceptance criteria

Comparison of calculated u-test value against the critical value:

Condition	Probability	Status
$\|t_{crit}\| < 1.64$	Greater than 0.1	The reported result does not differ significantly from the expected value
$1.95 > \|t_{crit}\| > 1.64$	Between 0.1 and 0.05	The reported result probably does not differ significantly from the expected value
$2.58 > \|t_{crit}\| > 1.95$	Between 0.05 and 0.01	It is not clear whether the reported result differs significantly from the expected value
$3.29 > \|t_{crit}\| > 2.58$	Between 0.01 and 0.001	The reported result is probably significantly different from the expected value
$\|t_{crit}\| > 3.29$	Less than 0.001	The reported result is significantly different from the expected value

For this proficiency test we have set the limiting value for the u-test parameter to 2.58 to determine if a result passes the test ($u \leq 2.58$).

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Performance Indicators

Acceptance Criteria

(I) Accuracy: the result passes if

$$\left| x_{lab} - X_{ref} \right| \leq t_{crit} \sqrt{u_{lab}^2 + u_{ref}^2}$$

(II) Precision: the result passes if

$$RSD_{\max}[\%] \geq \sqrt{\left(\frac{u_{lab}}{X_{ref}}\right)^2 + \left(\frac{u_{ref}}{X_{ref}}\right)^2} \times 100\%$$

where the tolerance (RSD_{max}) is set for each analyte as a function of its concentration level and ease of determination.

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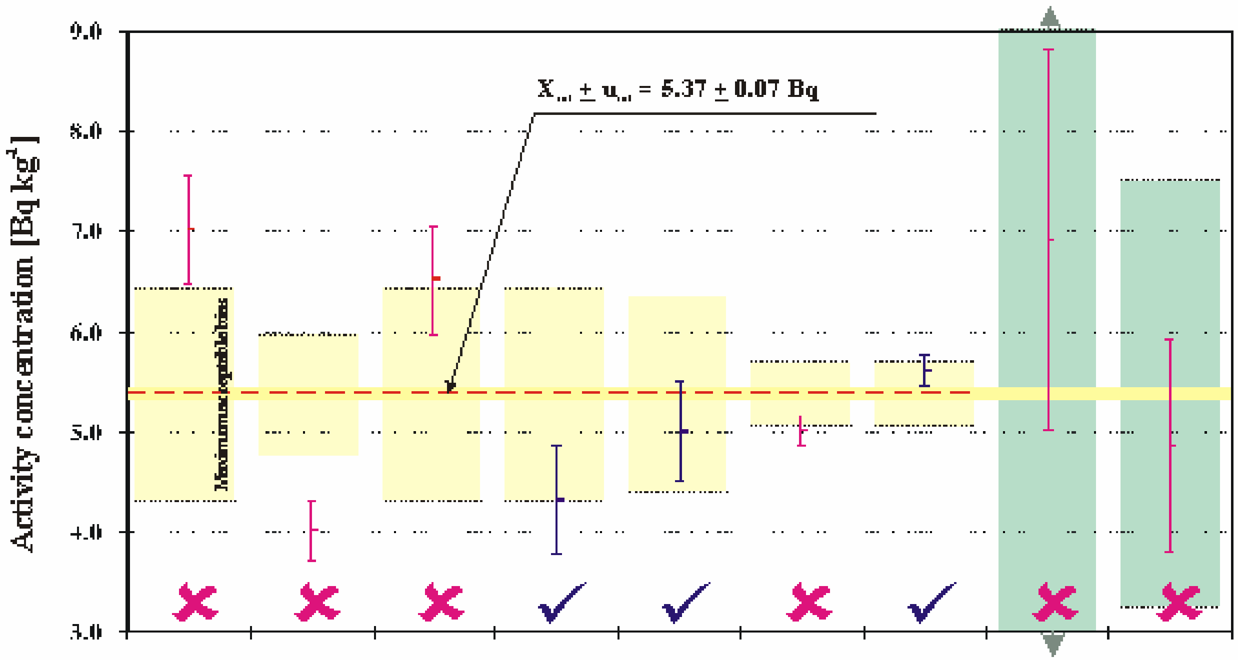
Performance Indicators

Acceptance Criteria – How does it work?



Criterion failed	I	I	I	none	none	I	none	II	II
u-Score	3.01	-4.45	2.09	-1.95	-0.73	-2.24	1.39	0.80	-0.50
z-Score	3.04	-2.25	2.10	-1.97	-0.69	-0.69	0.43	2.85	-0.98

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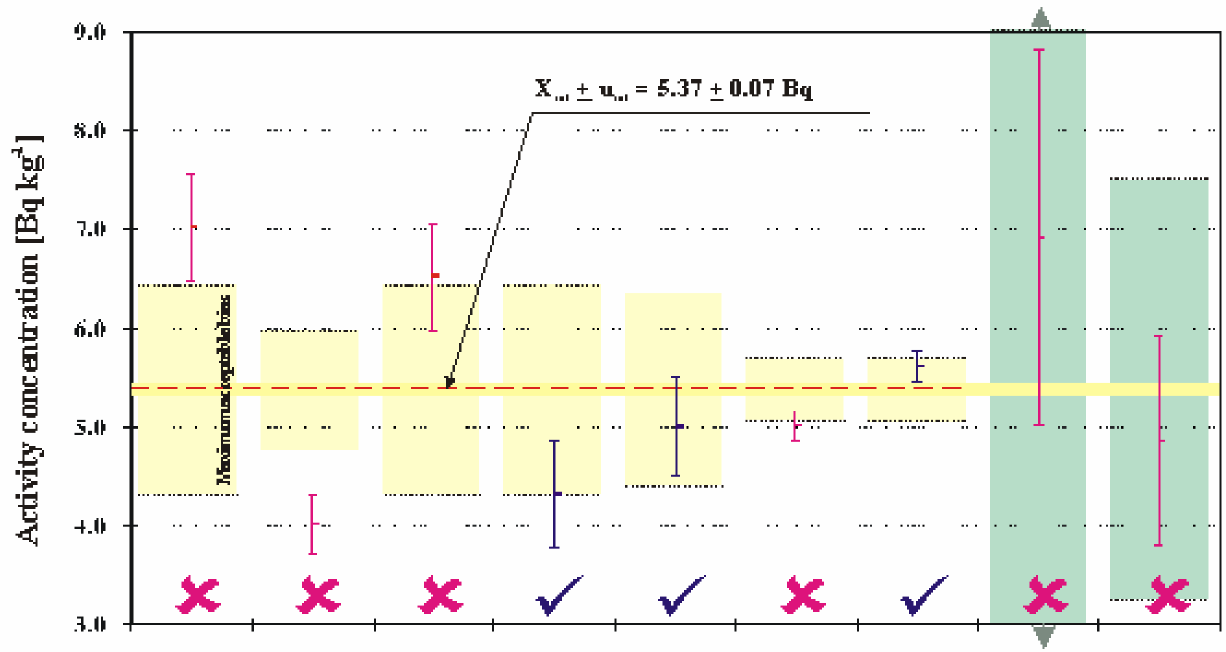
Performance Indicators

Acceptance Criteria – How does it work?



Criterion failed	I	I	I	none	none	I	none	II	II
u-Score	3.01	-4.45	2.09	-1.95	-0.73	-2.24	1.39	0.80	-0.50
z-Score	3.04	-2.25	2.10	-1.97	-0.69	-0.69	0.43	2.85	-0.98

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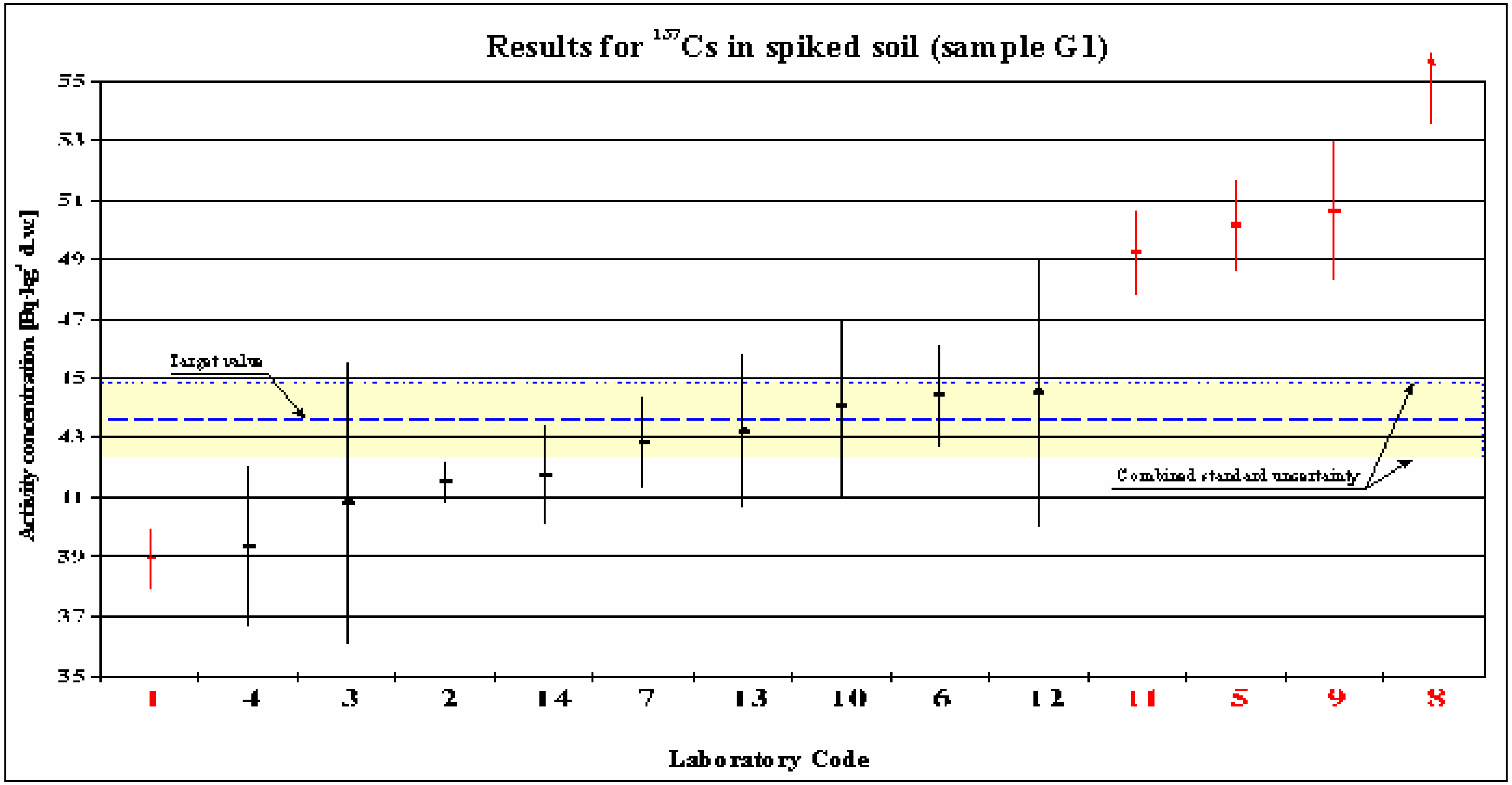
Participants' results for ^{137}Cs



Target Value ± u: 44 ± 1.3 Bq·kg⁻¹ d.w.
Total number of laboratory means submitted: 14
Number of results passing acceptance criteria: 9

Red - denotes a result which failed acceptance criteria

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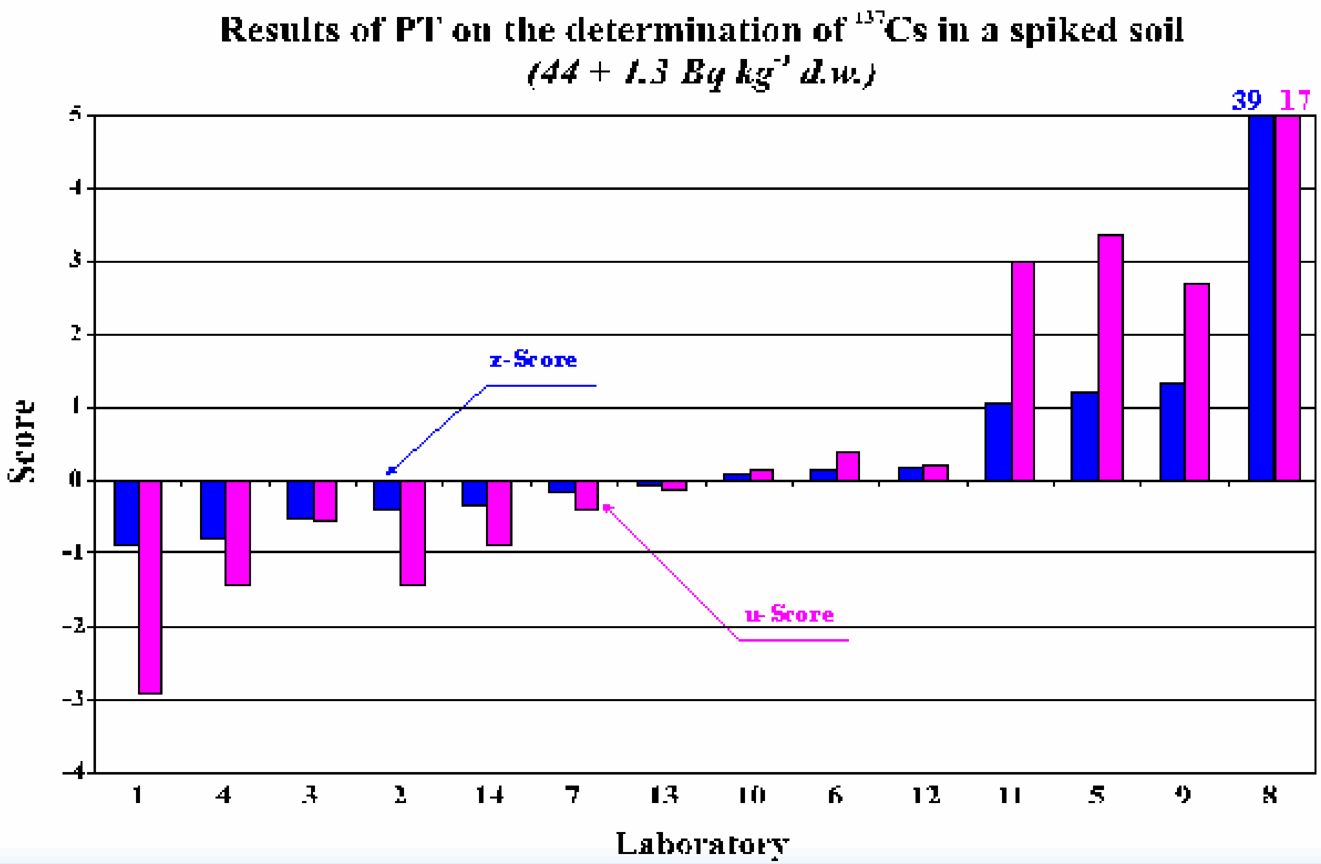
Participants' results for ^{137}Cs



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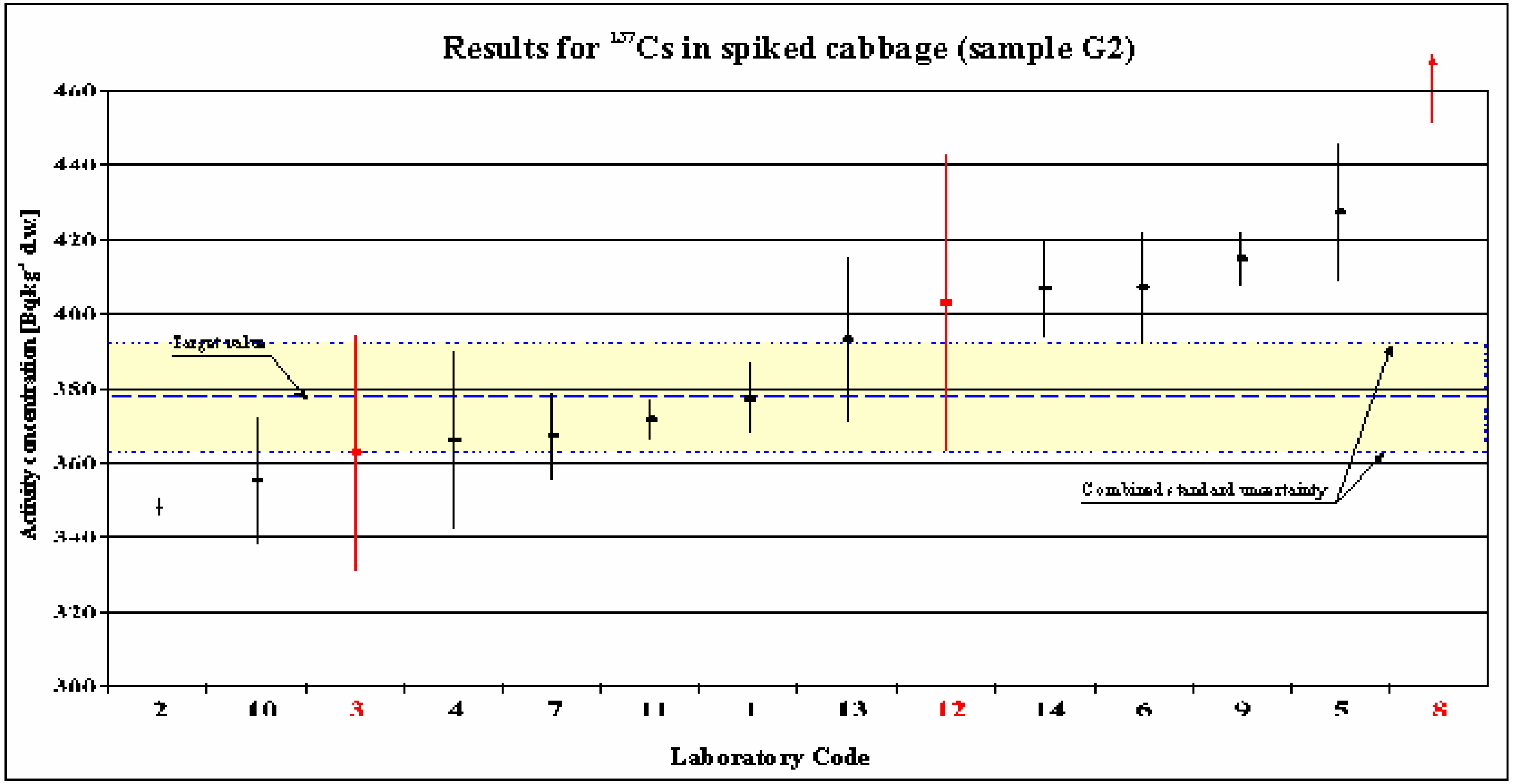
Participants' results for ^{137}Cs



Target Value + u: 380 + 15 Bq kg^{-1} d.w.
Total number of laboratory means submitted: 14
Number of results passing acceptance criteria: 11

Red - denotes a result which failed acceptance criteria

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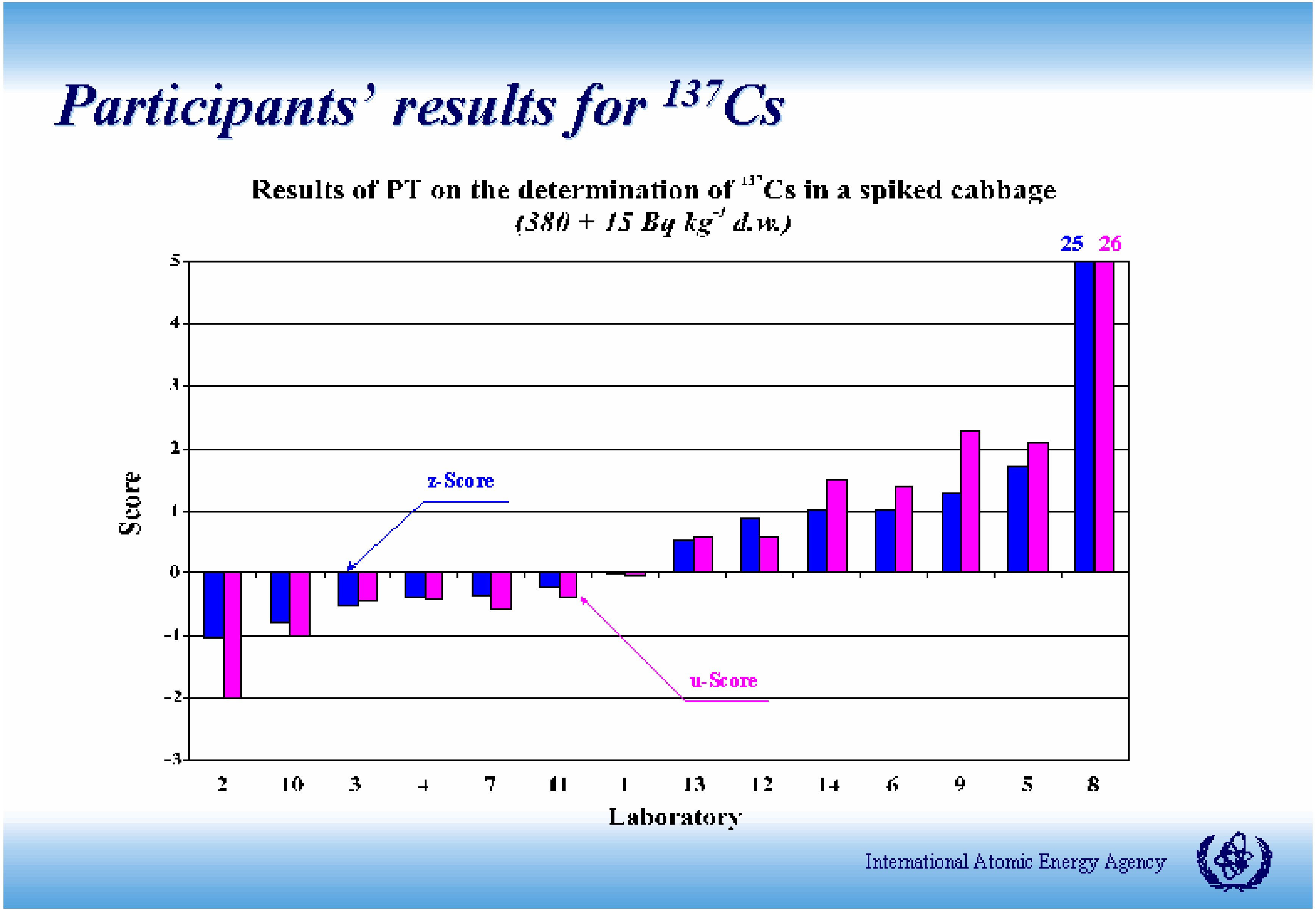
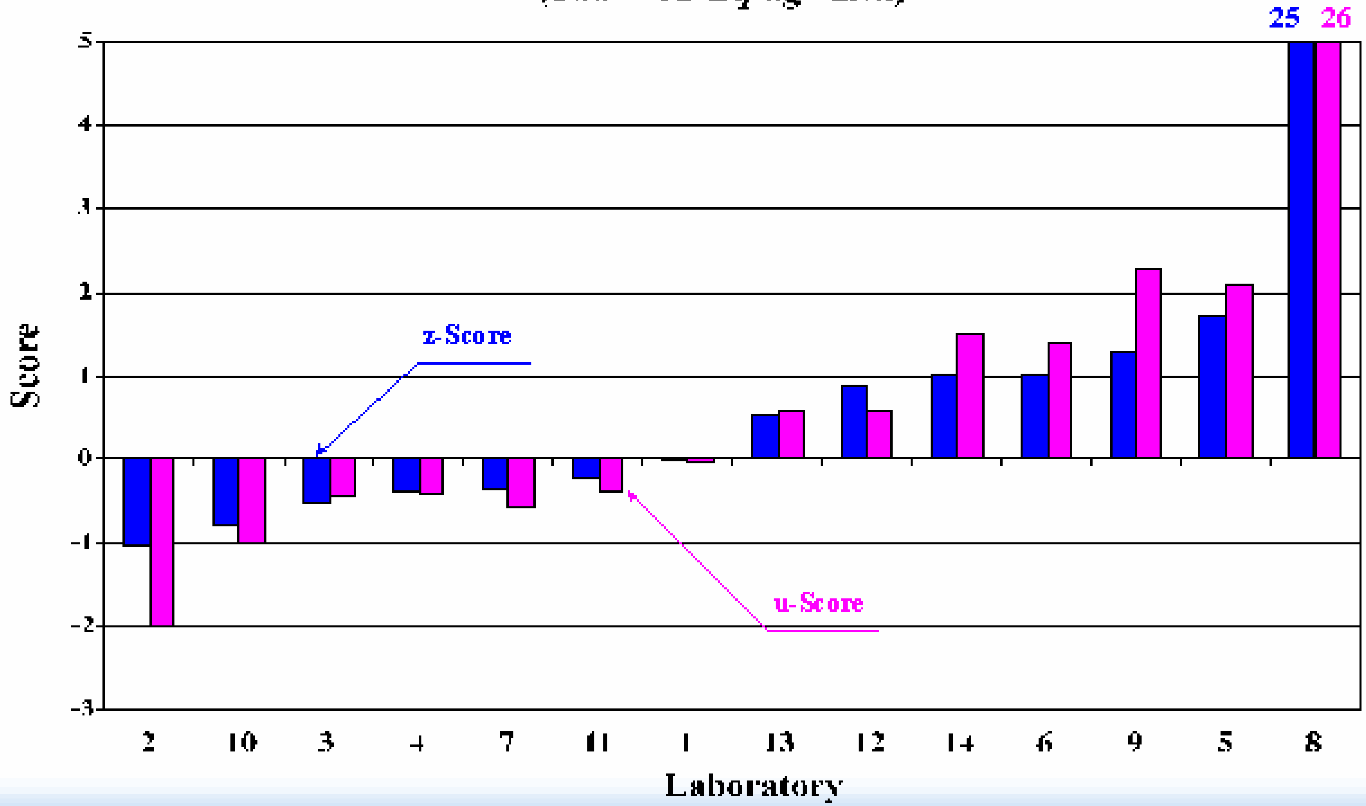

Participants' results for 137Cs
Results of PT on the determination of 137Cs in a spiked cabbage
(380 + 15 Bq kg-1 d.w.)
25
26
5
4
3
2
1
0
-1
-2
-3
Score
z-Score
u-Score
2
10
3
4
7
11
1
13
12
14
6
9
5
8
Laboratory
International Atomic Energy Agency

Participants' results for ^{137}Cs

Results of PT on the determination of ^{137}Cs in a spiked cabbage
(380 + 15 Bq kg^{-1} d.w.)



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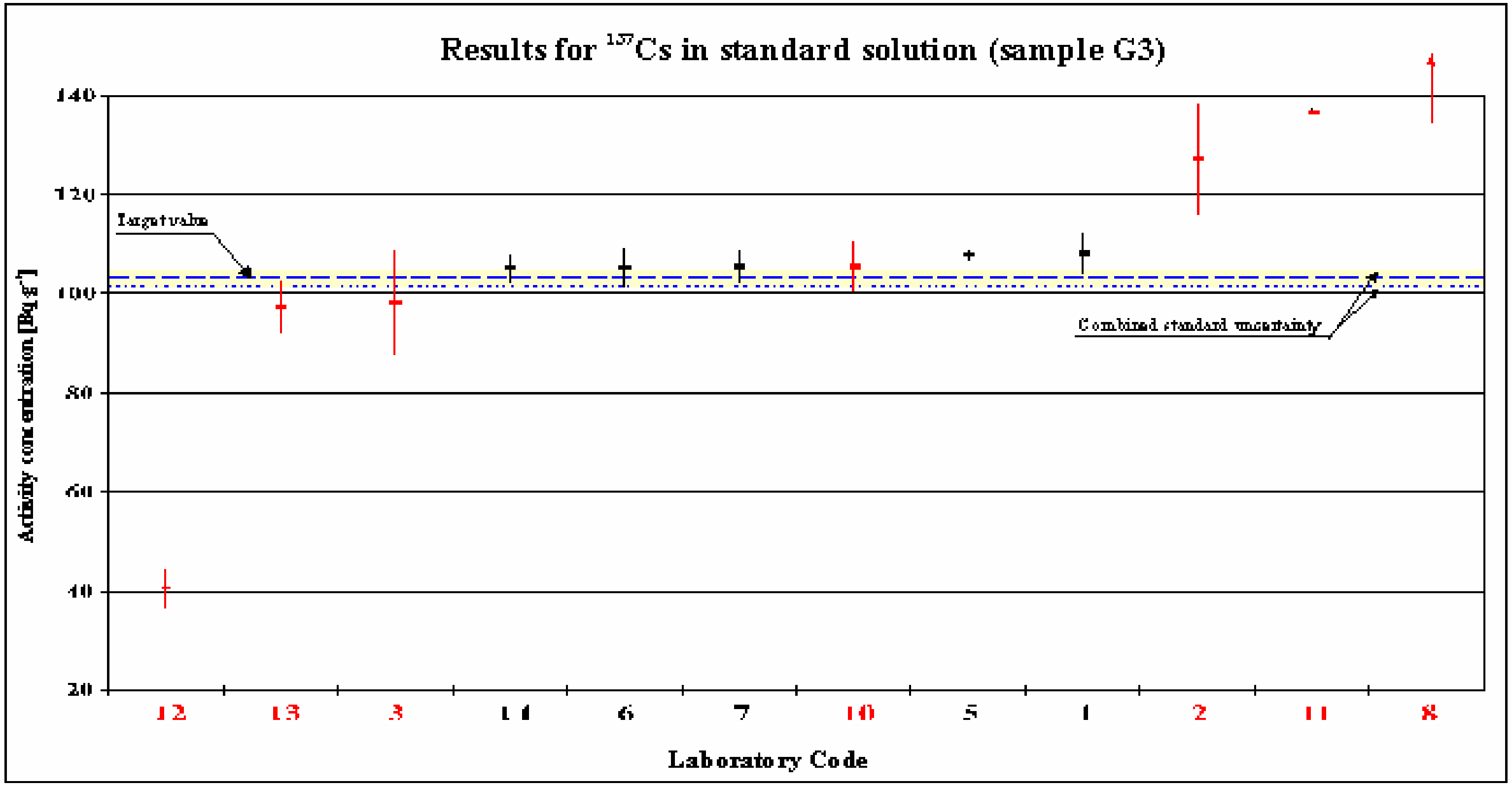
Participants' results for ^{137}Cs



Target Value + u: *103 + 1.6 Bq g⁻¹*
Total number of laboratory means submitted: *12*
Number of results passing acceptance criteria: *5*

Red - denotes a result which failed acceptance criteria

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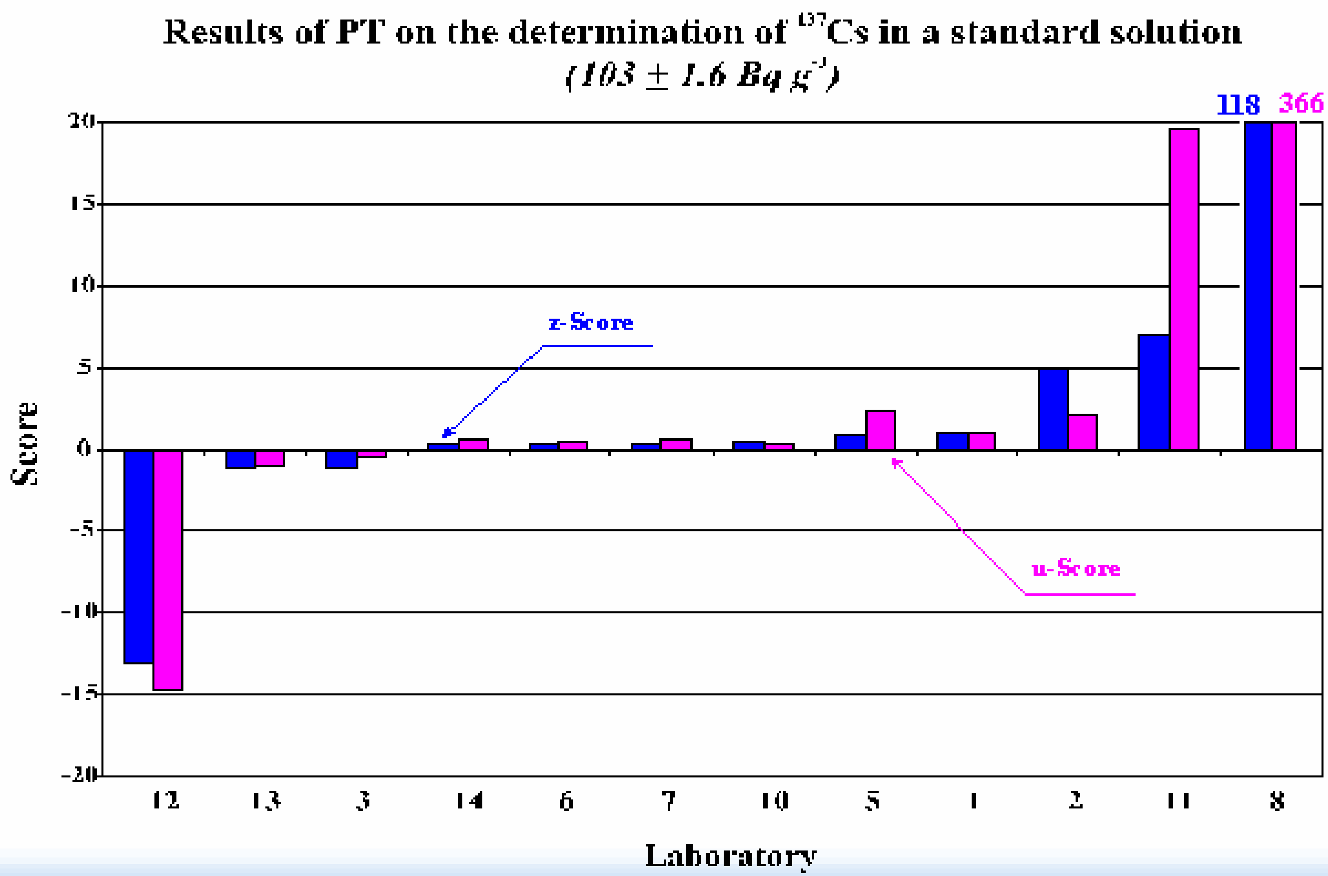
Participants' results for ^{137}Cs



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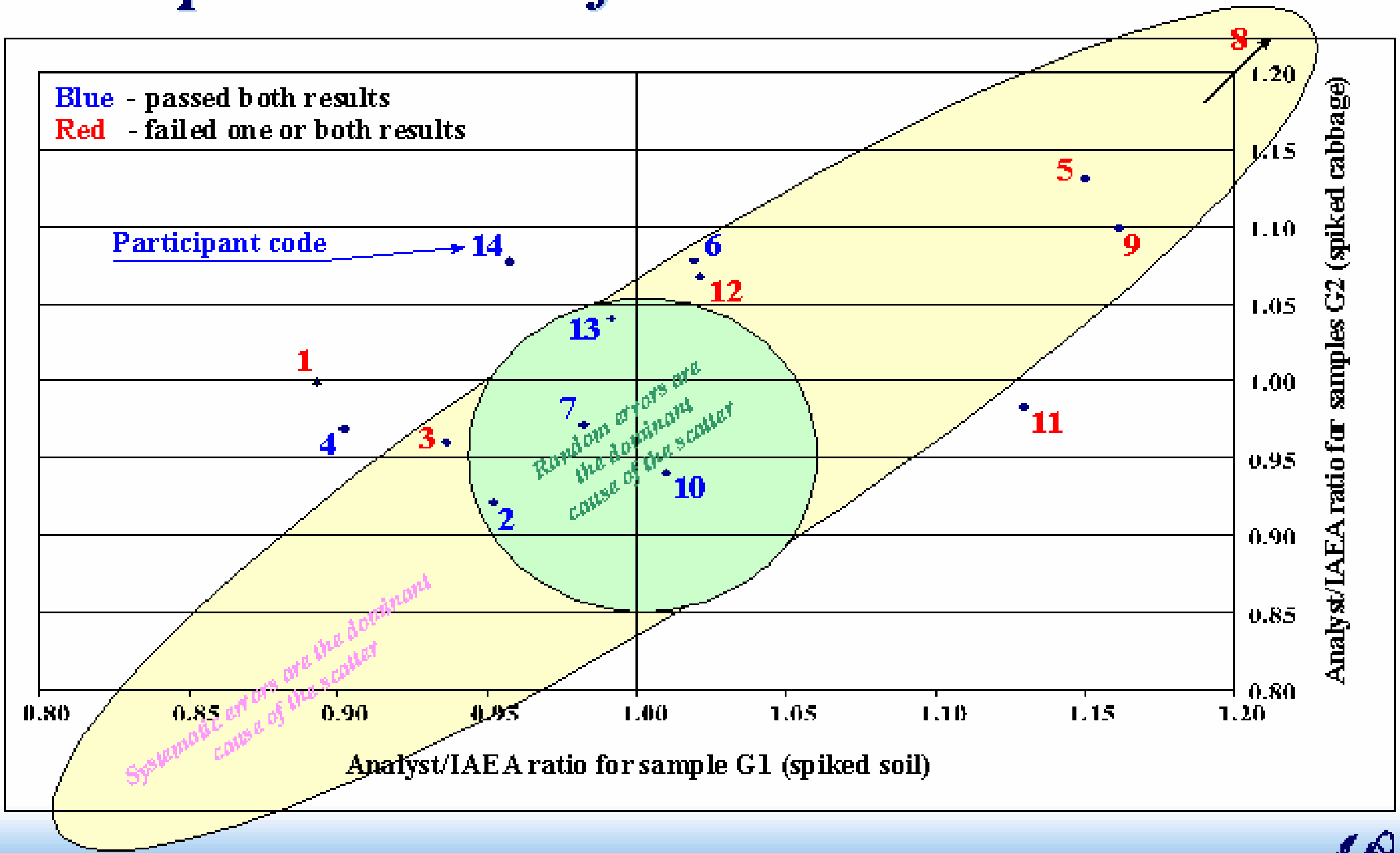
Participants' results for ^{137}Cs



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Distribution of Results (1)

Results for determination of γ-emitting radionuclides in spiked soil

	No of results				
	Reported	Not reported	Rejected (Accuracy)	Rejected (Precision)	Accepted
^{54}Mn	11	3	0	1	10 (91%)
^{57}Co	10	4	3	0	7 (70%)
^{60}Co	11	3	0	0	11 (100%)
^{65}Zn	7	7	1	3	3 (43%)
^{88}Y	11	3	1	5	5 (45%)
^{134}Cs	12	2	4	0	8 (67%)
^{137}Cs	14	0	5	1	9 (64%)

Distribution of Results (2)

Results for determination of γ-emitting radionuclides in spiked cabbage

	No of results				
	Reported	Not reported	Rejected (Accuracy)	Rejected (Precision)	Accepted
^{57}Co	11	3	2	2	7 (64%)
^{60}Co	11	3	0	2	9 (82%)
^{65}Zn	10	4	0	3	7 (70%)
^{134}Cs	12	2	4	2	6 (50%)
^{137}Cs	14	0	1	2	11 (78%)

Distribution of Results (3)

Results for determination of γ-emitting radionuclides in standard solution

	No of results				
	Reported	Not reported	Rejected (Accuracy)	Rejected (Precision)	Accepted
^{54}Mn	12	2	5	2	5 (42%)
^{57}Co	12	2	6	4	2 (17%)
^{60}Co	12	2	4	4	4 (33%)
^{65}Zn	12	2	3	2	7 (58%)
^{88}Y	12	2	5	4	4 (33%)
^{134}Cs	12	2	9	2	1 (8%)
^{137}Cs	12	2	3	4	5 (42%)
^{152}Eu	12	2	5	2	5 (42%)

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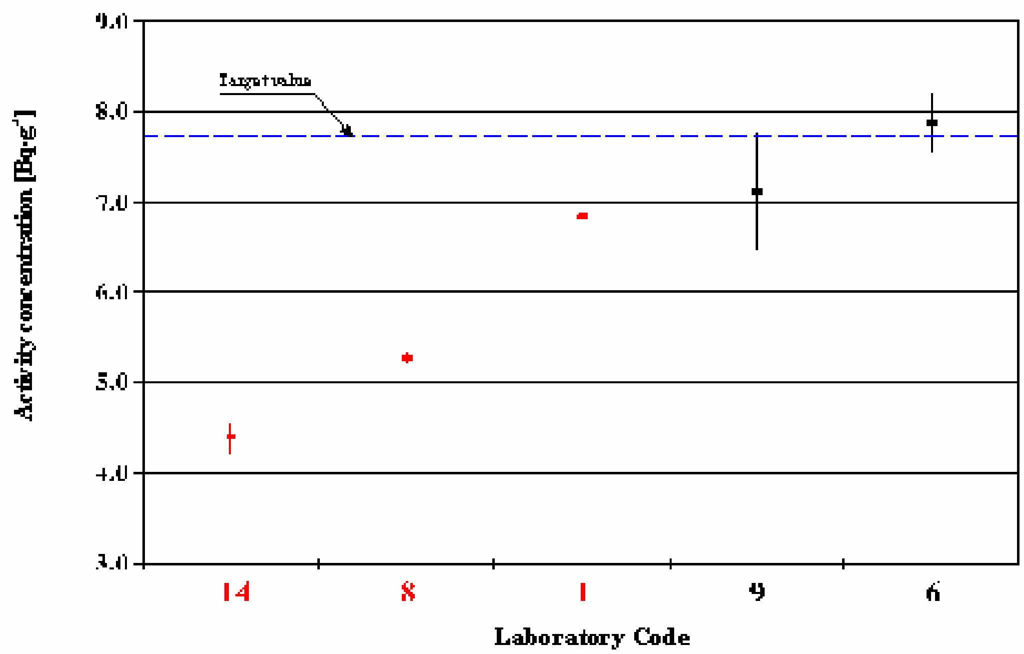
Participants' results for ^{90}Sr

Results for ^{90}Sr in standard solution (sample B9)



Target Value + u: 7.72 + 0.022 Bq g⁻¹
Total number of laboratory means submitted: 5
Number of results passing acceptance criteria: 2

Red - denotes a result which failed acceptance criteria

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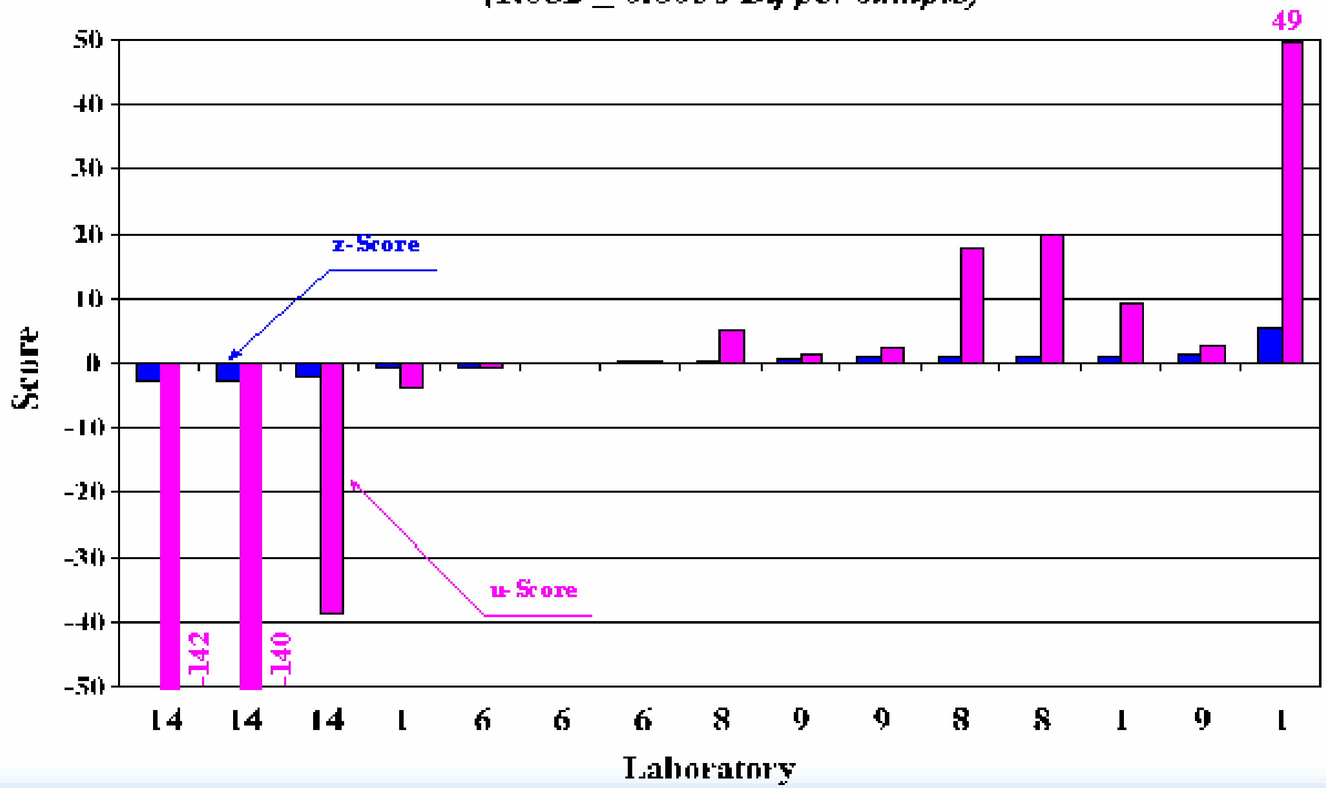
Participants' results for ^{90}Sr

Results of PT on the determination of ^{90}Sr in a spiked soil
(1.082 ± 0.0056 Bq per sample)



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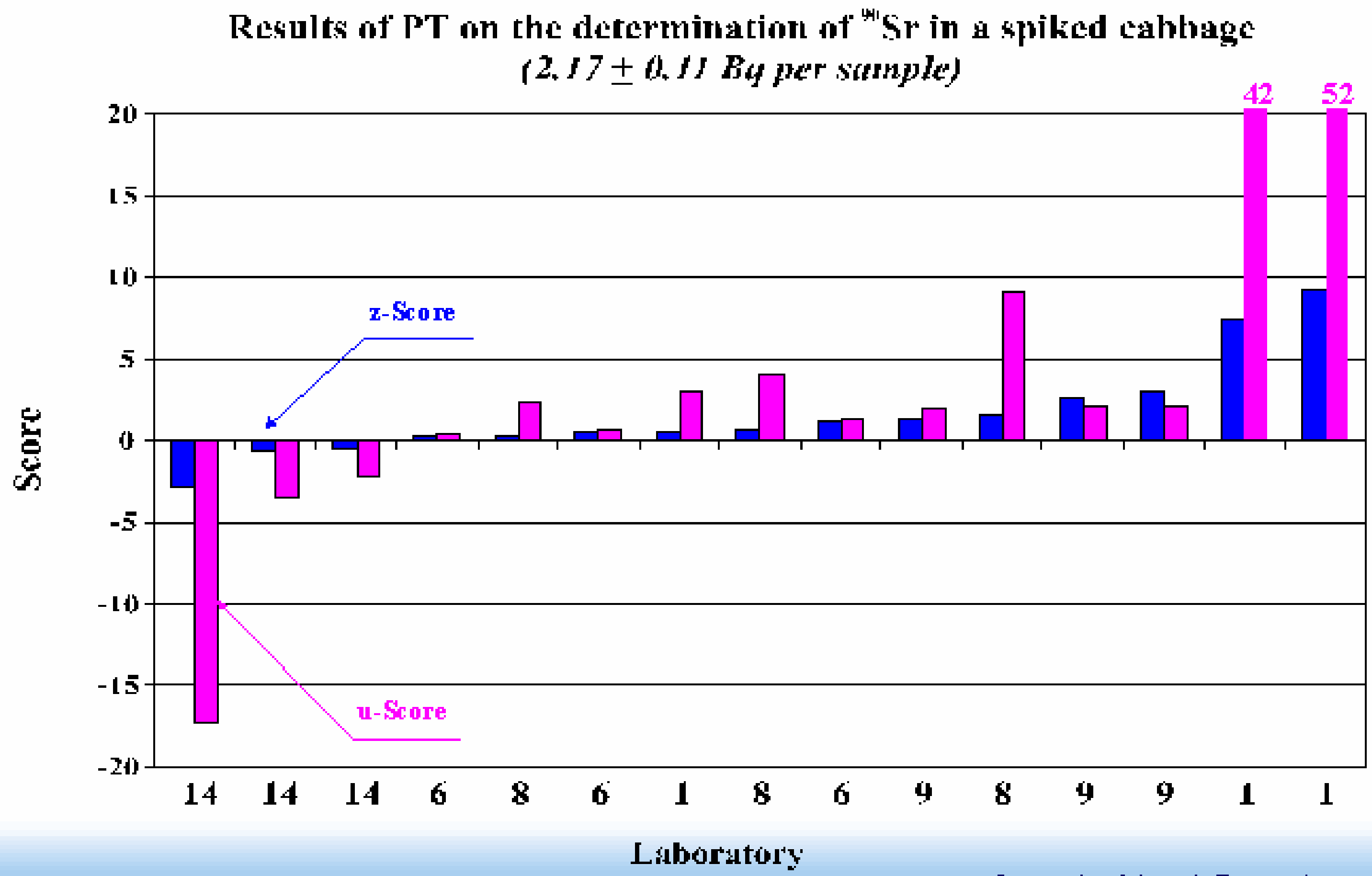
Participants' results for ^{90}Sr



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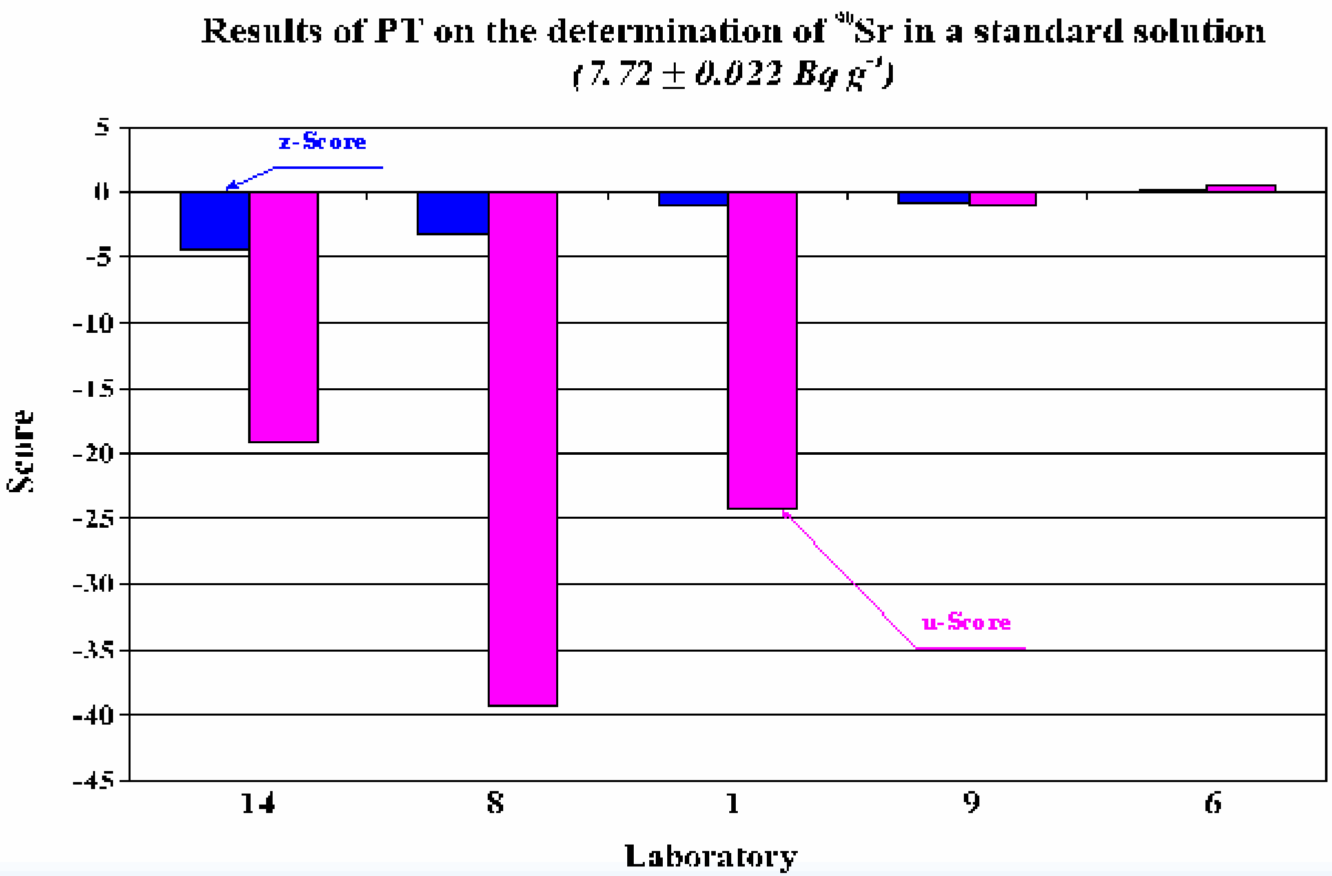
Participants' results for ^{90}Sr



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Distribution of Results (4)

Results for determination of ^{90}Sr

	No of results				
	Reported	Not reported	Rejected (Accuracy)	Rejected (Precision)	Accepted
Spiked soil	15	27	9	0	6 (40%)
Spiked cabbage	15	27	8	2	6 (40%)
Standard solution	5	9	3	0	2 (40%)

Distribution of Results (5)

Results for determination of γ-emitting radionuclides

In addition, the Analysts reported results for the following radionuclides:

- *in spiked soil – ^{109}Cd (Lab 5 and 7),*
  ^{155}Eu (Lab 7)
  ^{203}Hg (Lab 7)

These radionuclides were not present in the samples.

Distribution of Results - Summary

Analyte	Sample	Number of results reported	Failed z-Score |z| > 2	Failed Accuracy Criterion	Failed Precision Criterion	Number of laboratories with final status "Passed"
^{54}Mn	G1 G3	11 12	Lab 2 Lab 4, 11, 12	- Lab 1, 4, 5, 11, 12	Lab 2 Lab 2, 3	10 5
^{57}Co	G1 G2 G3	10 11 12	- Lab 6 Lab 4, 5, 6, 11, 12, 14	Lab 1, 6, 14 Lab 1, 6 Lab 4, 5, 6, 11, 12, 14	- Lab 3, 12 Lab 1, 2, 3, 13	7 7 2
^{60}Co	G1 G2 G3	11 11 12	- - Lab 2, 4, 11, 12, 13	- - Lab 4, 5, 11, 12	- Lab 3, 12 Lab 2, 3, 10, 13	11 9 4
^{65}Zn	G1 G2 G3	7 10 12	Lab 5 Lab 12 Lab 2, 3, 4, 11, 12	Lab 1 - Lab 4, 11, 12	Lab 5, 7, 10 Lab 3, 12, 13 Lab 3, 13	3 7 7
^{88}Y	G1 G3	11 12	Lab 1 Lab 2, 4, 11, 12, 13	Lab 1 Lab 4, 5, 11, 12, 13	Lab 2, 3, 10, 12, 13 Lab 2, 3, 10, 13	5 4
^{134}Cs	G1 G2 G3	12 12 12	- Lab 2, 12 Lab 1, 4, 11, 12, 13	Lab 1, 2, 11, 12 Lab 1, 2, 10, 11 Lab 1, 4, 5, 6, 7, 11, 12, 13, 14	- Lab 3, 12 Lab 2, 3	8 6 1
^{137}Cs	G1 G2 G3	14 14 12	Lab 8 Lab 8 Lab 2, 8, 11, 12	Lab 1, 5, 8, 9, 11 Lab 8 Lab 8, 11, 12	Lab 8 Lab 3, 12 Lab 2, 3, 10, 13	9 11 5
^{152}Eu	G3	12	Lab 4, 12	Lab 1, 4, 5, 12, 14	Lab 2, 3	5
^{90}Sr	B1 - B3 B5 - B7 B9	15 15 5	3 (Lab 1, 14) 5 (Lab 1, 5, 14) 2 (Lab 8, 14)	9 (Lab 1, 8, 14) 8 (Lab 1, 8, 14) 3 (Lab 1, 8, 14)	- 2 (Lab 9) -	2 2 2

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ABBREVIATIONS

Nu: Radionuclide: Chemical symbol
C: Crop class

C = Cereals (wheat, rice, maize, sorghum, millet (grain and/or flour))
F = Fodder (maize without grain, clover, alfalfa)
G = Grass, vegetation of pastures
L = Leaves of root crops (generally non-edible parts)
P = Pods and seeds of beans, peas and nuts
R = Roots of root crops (beet, carrot, radish)
S = Stems and shoots of cereals, peas, beans, potato, etc. (generally non-edible parts)
T = Tubers (potato, swedes)
V = Green vegetables (cabbage, spinach, leek, lettuce, onions, garlic)
X = Miscellaneous (fruits, tomato, cucumber, etc., coconuts)
H = Herbs (tea, thyme, spices)

TF: TF-value expressed as (Bq/kg dry weight crop)/(Bq/kg dry weight soil top 10 cm layer) for grass, and (Bq/kg dry weight crop)/(Bq/kg dry weight soil top 20 cm layer) for all other crops).

Ex K.: Exchangeable K in soil (cmol(+)/kg) or (milli-equivalents/100 g)
Ex Ca.: Exchangeable Ca in soil (cmol(+)/kg) or (milli-equivalents/100 g)
CEC: Cation exchange capacity (cmol(+)/kg) or (milli-equivalents/100 g)
pH_{KCl}: pH value observed in 0.1 M KCl or 0.01 $CaCl_2$
pH_w: pH value observed in water without addition of any salt
OM: Organic matter percentage (not C, but OM)

t-Co.: Type of contamination
Contamination type codes:

A = Artificially added
C = Chernobyl contamination
F = Nuclear weapons fallout
O = Other (sea spray, contaminated sludge)

t-Ex.: Experiment type
Experiment type codes:

F = Field experiments
L = Lysimeter experiments
P = Pot or container experiments

Time: Time elapsed between contamination of the soil and harvest, only.

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