



Final Report on a Field Study of Soil-to-Plant Transfer of Radioactive  
Caesium, Strontium and Zinc in Tropical Northern Australia

to the

**IAEA/FAO/IUR CRP on “Classification of Soils Systems on the Basis of  
Transfer Factors of Radionuclides from Soil to Reference Plants”**

by

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# Final Report on a Field Study of Soil-to-Plant Transfer of Radioactive Caesium, Strontium and Zinc in Tropical Northern Australia

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## Abstract

Soil-to-plant radionuclide transfer factors for cesium (<sup>134</sup>Cs), strontium (<sup>85</sup>Sr) and zinc (<sup>65</sup>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 zinc (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.

## 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 cooperative 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 study was initiated as Australia's contribution to the CRP entitled "Classification of soil systems on the basis of transfer factors of radionuclides from soil to reference plants". 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 Twining *et al* (2001).

In summary, 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.

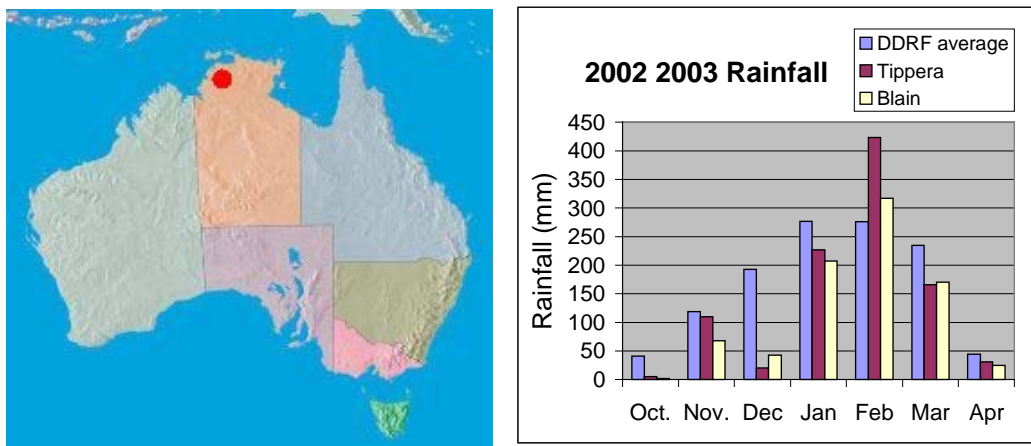
## Materials and Methods

### 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 what factors relating to soil type were most influential in affecting soil to plant transfer of radioactivity. A standard protocol to achieve this was prepared (FAO/IAEA/IUR, 1998) 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 Figure 1 together with rainfall data for the selected study sites. The Douglas Daly Research Farm is approximately 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.



**Figure 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 (eg Corbett, 1969; Jenny, 1980). Classification details for these specific soils are described in Lucas *et al.*, 1987. Under the Soils World Reference Base (WRB) (ISSS Working Group RB. 1998), 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 50m x 50m 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 active 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.

#### Addition of Radioactivity, Sowing of Crops and Sampling

Radioactivity ( $^{134}\text{Cs}$ ,  $^{85}\text{Sr}$  and  $^{65}\text{Zn}$ ) was initially added to the soils in October 1999. Due to its short half-life (65 days)  $^{85}\text{Sr}$  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 out 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 99) or 20 mm diameter sampler (April 00 and subsequently). In the first year, at least 6 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 3 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 (ie 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.

## Measures

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  $^{85}\text{Sr}$ .

Particle size distributions for each depth in each soil were performed by sedimentation and using a hydrometer (eg Corbett, 1969). Major clay mineralogy was evaluated using X-ray diffraction on the sub-2  $\mu\text{m}$  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, 1994) or inductively couple plasma mass spectrometry (Agilent 4500 ICP-MS; USEPA method 200.8, 1994). Total concentrations were evaluated following microwave digestion of 0.2g of sample in 10 mL concentrated  $\text{HNO}_3$  within a TFM pressure vessel (ANSTO modified USEPA method 3051, 1994). Estimates of the exchangeable fraction of the same elements in soils were determined after extraction with 1M  $\text{NH}_4\text{Cl}$  (Rayment and Higginson, 1992). 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 and pH (at a 1:5 ratio in water and in  $\text{CaCl}_2$ ) (Rayment and Higginson, 1992), and organic matter content using the Walkley-Black method (Allison, 1965).

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 60-70g soil samples, involving alkaline fusion, are given in Tagami and Uchida (2001). The water-soluble fraction was determined by shaking 50g of soil in 250mL deionised water over an 8hr period. The resultant soil solution was then obtained using a glass fibre filter (GF/A) followed by a 0.45 $\mu\text{m}$  membrane filter. Re in plants was determined as a total concentration using 20-25g samples. The analytical details are given in Tagami and Uchida (in press).

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 Twining *et al* (2003). 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 Zaw *et al* (2002). 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 Twining *et al.* (submitted).

### **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.

### **Results**

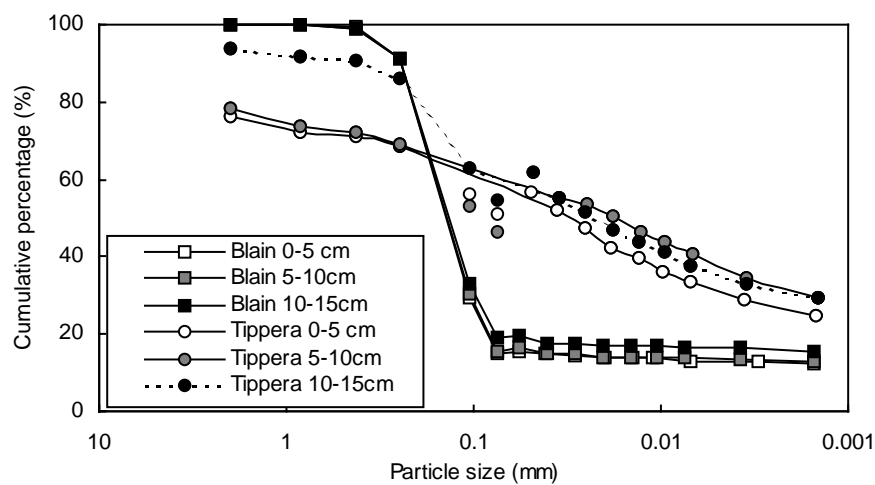
The acceptable results of the analyses and measurements carried out are summarised in the following text, tables and figures. Collated data are presented in Appendix A. More detailed data are available from the lead author.

### **Soil Properties**

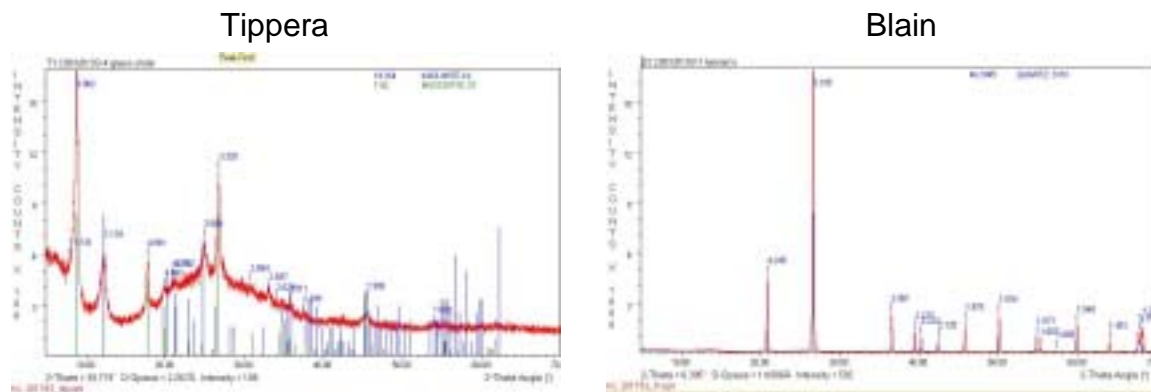
The values for particle size distributions, charge concentrations of exchangeable cations, cation exchange capacity (CEC), equilibrium pH and organic carbon content for each soil type at each depth are given in Table 1. The particle size distribution (also shown in Figure 2) confirms the greater clay 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 Lucas *et al.* (1987) 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 Figure 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. Note that the concentration values for cations have been charge compensated.

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 <sub>2</sub>		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 (mmol/kg) (averaged over 2000-2003)	Ca <sup>2+</sup>	19.7	13.4	11.6	25.2	17.0	19.1
	K <sup>+</sup>	3.1	2.9	3.6	6.0	5.3	5.3
	Mg <sup>2+</sup>	5.4	3.1	2.1	7.6	5.0	5.5
	Na <sup>+</sup>	5.6	5.5	6.3	5.5	6.1	7.5
Sum of exchangeable cations (effective CEC)		<b>33.8</b>	<b>24.9</b>	<b>23.6</b>	<b>44.3</b>	<b>33.3</b>	<b>37.4</b>
Organic carbon content (%)		0.14	0.02	0.00	1.34	0.54	0.42



**Figure 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)



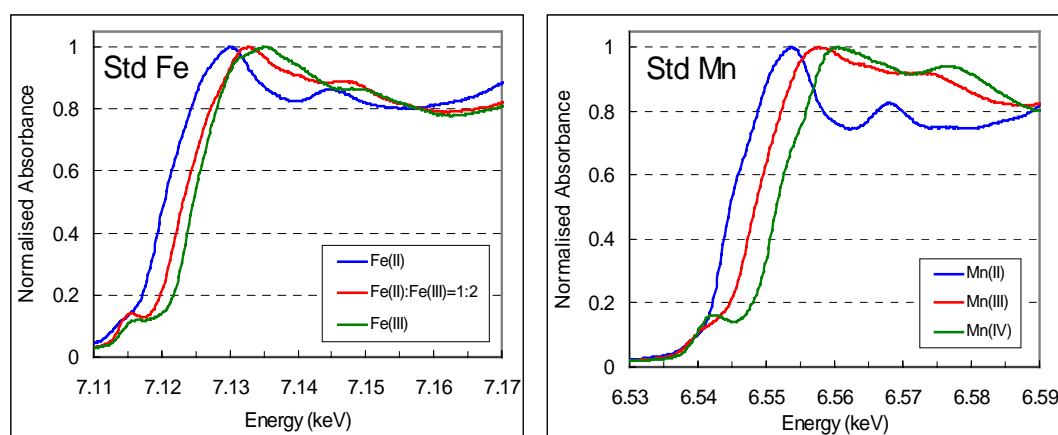
**Figure 3.** Graphs of XRD results for clay fraction mineralogy in each soil.

The pH values in water (Table 1) were found to be slightly acidic and a comparable result was observed in CaCl<sub>2</sub>. The pH values are similar (if slightly lower) to other results for these soils. Lucas *et al.* (1987) reported 5.9 to 6.1 for Tippera, and Eastik (NT DBIRD, pers. comm., 2001) determined 6.6 to 6.8 for Blain.

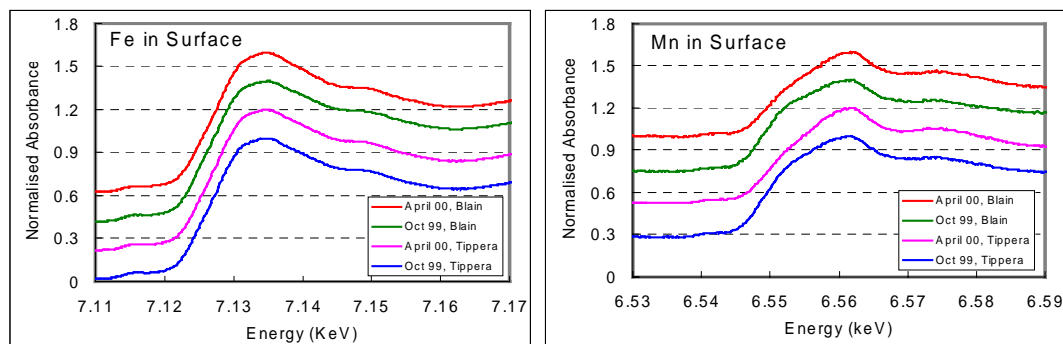


## XANES

Figure 4 shows the XANES spectra for the reference standards prepared for the study whilst Figure 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 (Figure 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 (ie. Mn(IV) from 66 % to 76 % in Tippera sample and Mn(IV) from 65 % to 71 % in Blain sample).



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



**Figure 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

Results for Fe in soil samples indicated that Fe species are very similar for both soils and sampling periods [Fe(II) ~ 10 %] (Figure 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

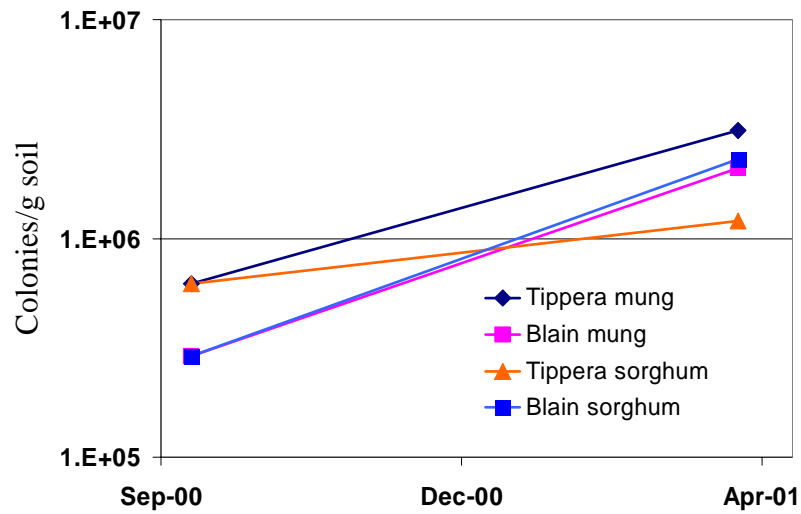
would be appropriate to consider the consequential possibility of seasonal changes in plant availability of these elements in future radiological dose assessment studies. These results also have relevance to the use of results from pot studies for soil-to-plant transfer compared with those obtained from 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.

## 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 (Figure 6).

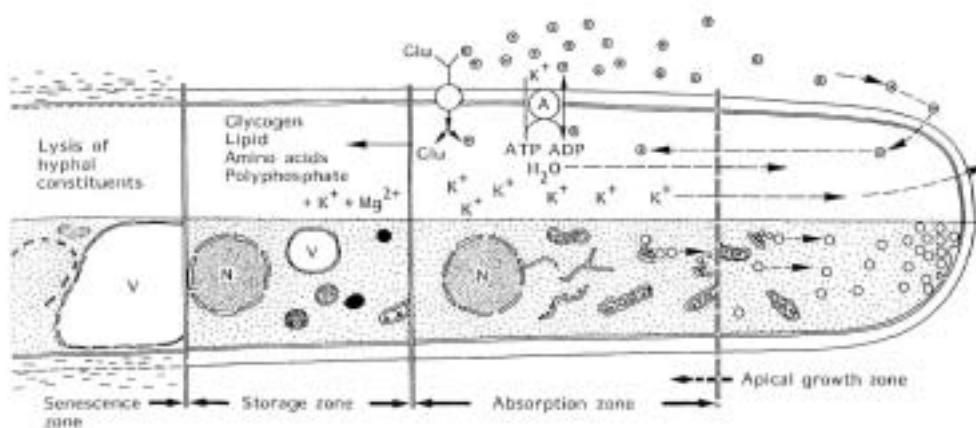
**Table 2.** Viable bacterial and fungal colony counts on nutrient agar (NA) and malt extract agar (MEA). N.B. Initial April samples were unreliable and these results were after storage at 4°C for 3 weeks. No loss of viability was observed.

			NA (bacteria)	MEA (fungi)
	Soil	Crop	colonies/g soil	
October-00	Tippera	nil	8.0E+06	6.2E+05
	Blain	nil	2.0E+07	2.9E+05
April-01	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



**Figure 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 (Figure 7). Several elements are more soluble at lower pH values, including Zn, P and Sr (see section on  $K_d$ s 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.



**Figure 7.** Diagram showing proton pumping from a fungal mycelium into the local soil micro-environment (taken from Garrett, 1981).

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 (IAEA/FAO/IUR, 1998).

#### Distribution coefficients ( $K_d$ values)

The results of batch  $K_d$  experiments for  $^{134}\text{Cs}$  are shown in Figure 8a. The retention of Cs is very strong, indicated by  $K_d$  values of approximately  $10^3$  mL/g. The adsorption of  $^{134}\text{Cs}$  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 (Cornell, 1993). The equilibrium pH values for Blain soils are higher than the Tippera soils. However, the experiments in which the pH was varied by  $\pm 1$  unit indicate that there is no significant pH dependence of Cs sorption in either soil.

The adsorption of  $^{85}\text{Sr}$  is much weaker than for  $^{134}\text{Cs}$ , with  $K_d$  values of approximately 30 - 60 mL/g being measured at the equilibrium pH of the samples (Figure 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).

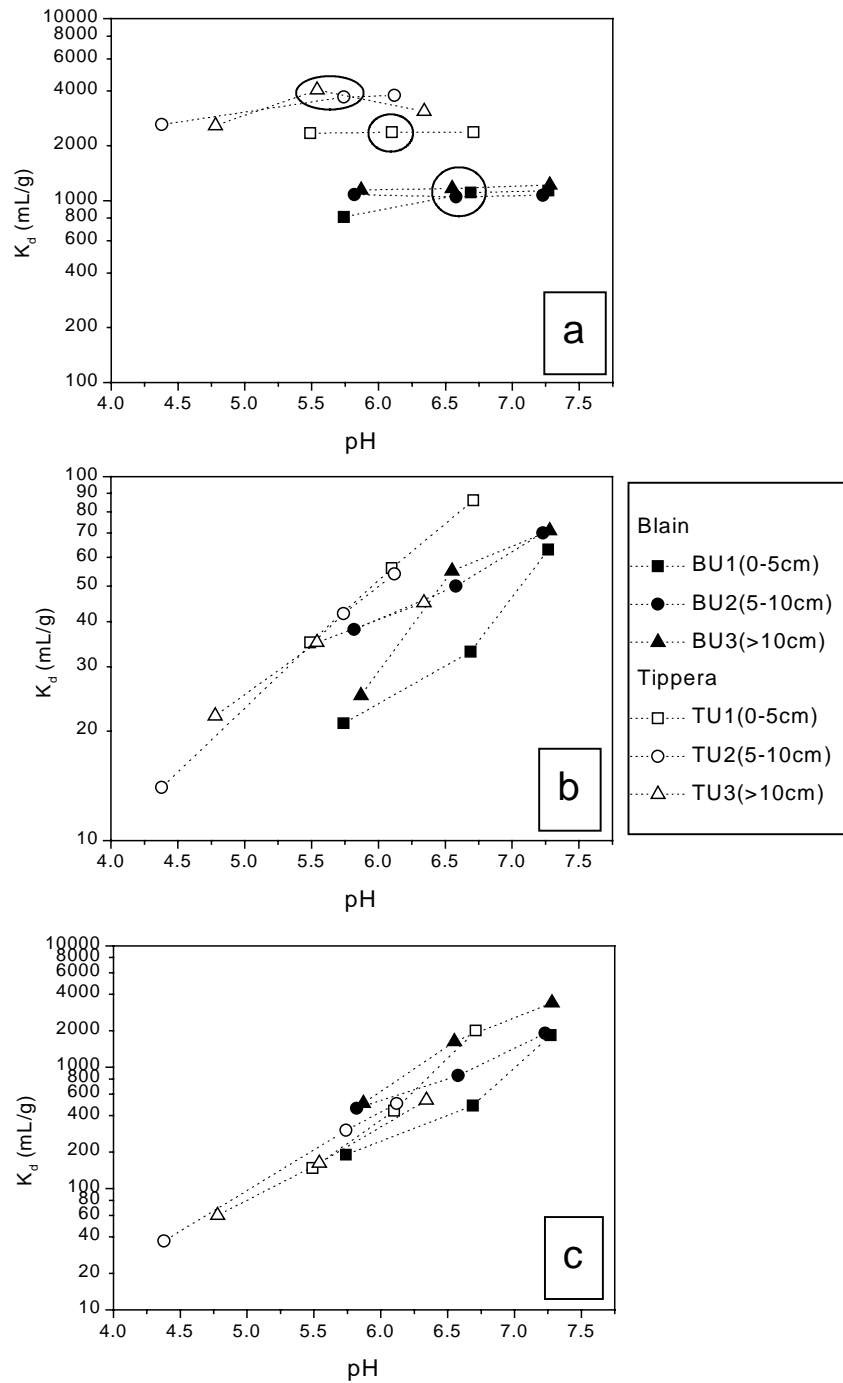
The  $K_d$  data for Zn show the strongest pH dependence of the studied radionuclides, increasing by more than an order of magnitude (from about 40 mL/g to 3000 mL/g) between pH of 4.4 and 7.3 (Figure 8c). Because of this strong pH dependence, the  $K_d$  values for Blain soils at their equilibrium pH (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 Sheppard and Thibault (1990). In general, the values are consistent with those reported for the generic soil types as categorised by Sheppard and Thibault (1990).

**Table 3.** Comparison of average distribution coefficients (standard error,  $n = 3$ ), measured at equilibrium pH values, with geometric mean values of Sheppard and Thibault (1990). 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



**Figure 8.** Distribution coefficient ( $K_d$ ) values for adsorption of radionuclides (a- $^{134}\text{Cs}$ , b- $^{85}\text{Sr}$ , c- $^{65}\text{Zn}$ ) 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.

## Soil Chemistry – elements

Details of semi-quantitative XRF analyses on the two soils were given earlier (Twining *et al* 2001). The results showed that oxides of Si, Al and Fe predominate, comprising approx. 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 (Cornell, 1993; Payne *et al.*, 2001). Results of ICP analyses on total digests (eg as reported in Twining *et al.* (2001) for year one. Detailed data for later years is available from the lead author. The best results were obtained for the first season samples with limit of detection problems hampering interpretation in later years.) 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 <sup>65</sup>Zn from both soils (refer to later results from gamma spectrometry).

In relation to the stable element analogues for the radioisotopes added into the system, the values for Cs, Sr and Zn are reported for each depth on both soils in Table 4. The data for Sr are similar between 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<sub>4</sub>Cl (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 (Sparks, 1995). All three trace elements in Tippera and Blain are towards the lower end of the global distributions. This observation is particularly true for Sr and Zn, both being 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<sub>4</sub>Cl) elemental content of unlabelled soils for Cs, Sr and Zn compared with median and range of measured values of global soils as reported by Sparks (1995).

		Cesium		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	-

In relation to the Tc analogue, Re, concentrations in soils were reported in detail elsewhere (Tagami *et al.*, 2003). Average total concentrations in Tippera and Blain soils were  $21.1 \pm 2.0$  µg/g and  $4.9 \pm 0.9$  µg/g respectively, for the samples collected in 2000. The percentages of water-soluble Re in these soil samples depended on soil type, e.g.,  $3.6 \pm 2.5\%$  for Tippera and  $12 \pm 8\%$  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 µg/g). Detectable levels in these samples range from 0.07 – 0.6 µg/g in sorghum and 0.06 – 0.6 µg/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.

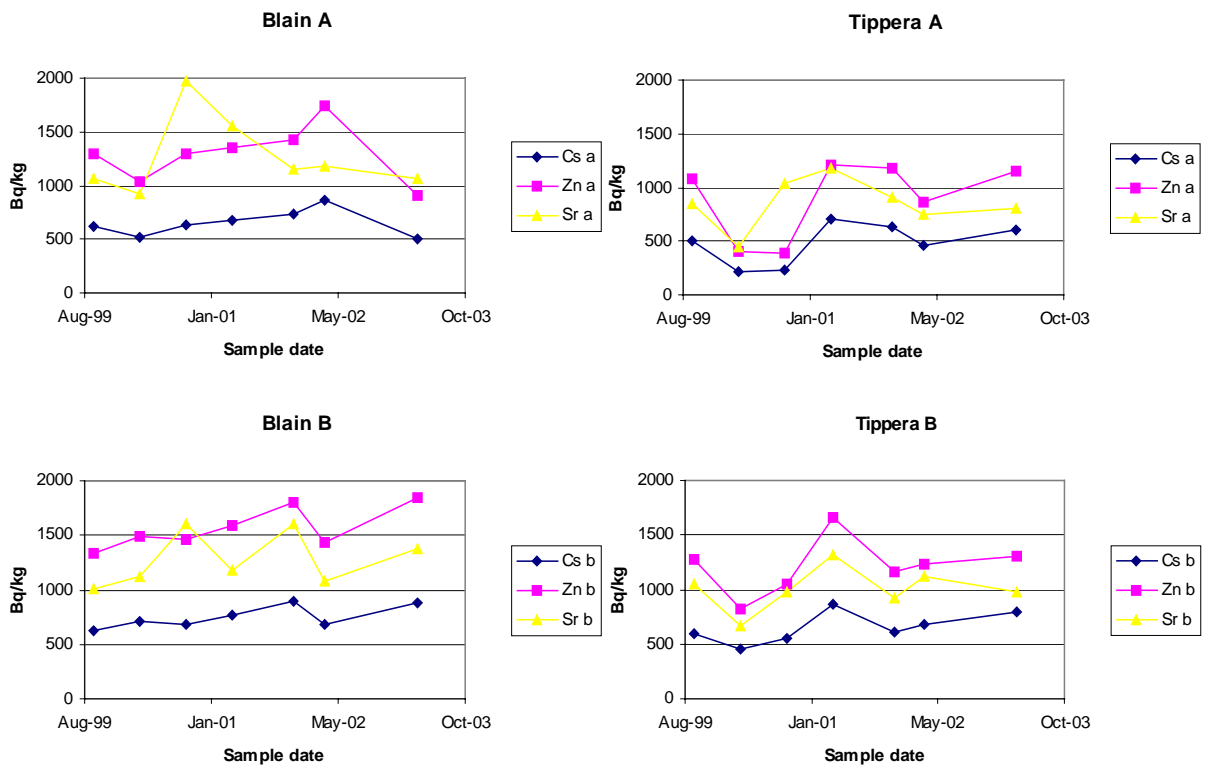
**Table 5.** TFs for Re from Tippera and Blain soils to sorghum grain and mung beans over two years. The values derive from duplicate or triplicate analyses on individual soil samples. Errors are  $\pm 1$  s.d. of the individual values above detection limit.

	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



## Soil Radioactivity

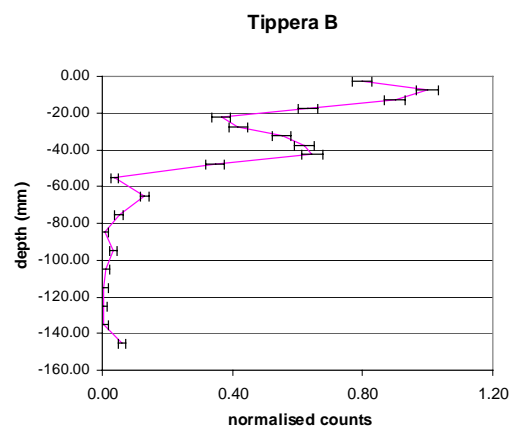
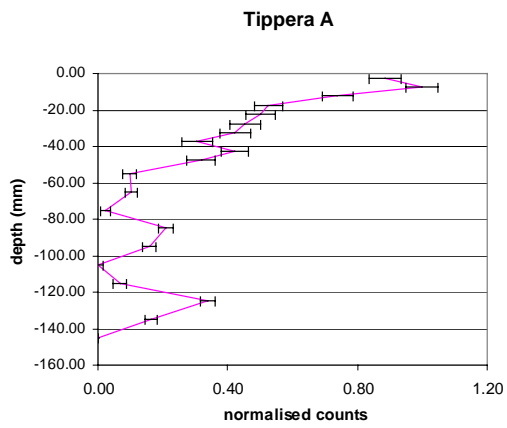
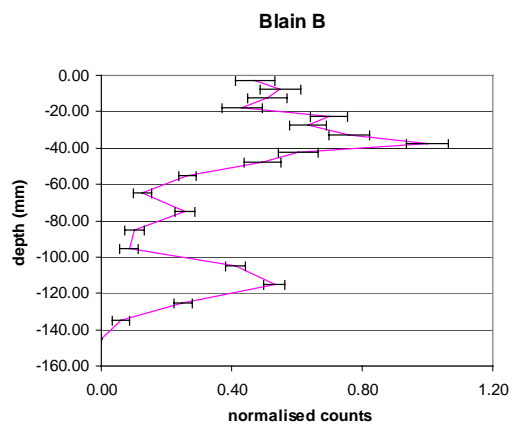
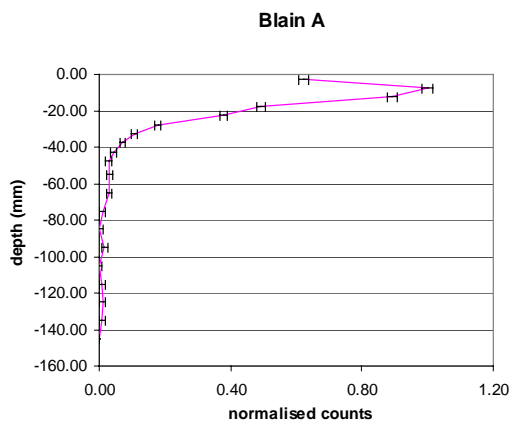
After correcting for radioactive decay, there was very little depletion of the added radioactivity from the sites over the 4.5 year experimental period. This is shown in Figure 9 which gives the average, decay corrected, activity for each nuclide in the top 20cm of the soil profile within each subplot.



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

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 4 or more over the entire period (Figure 9). This is despite the fact that each value is representative of at least 6 to 9 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 Figure 10 that shows the vertical profiles of  $^{134}\text{Cs}$  at the end of the experimental period. The profiles are based on individual cores that were separated into 5mm to 10mm sections (Given the time since addition,  $^{85}\text{Zn}$  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 4 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.



**Figure 10.** Depth profiles of  $^{134}\text{Cs}$  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.

### 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.

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

Radioactivity in soil is also defined as the average activity concentration in the top 20cm. 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 Figure 11. Strontium does not have the same pattern as the other radioisotopes because fresh  $^{85}\text{Sr}$  was added to the soils each year.

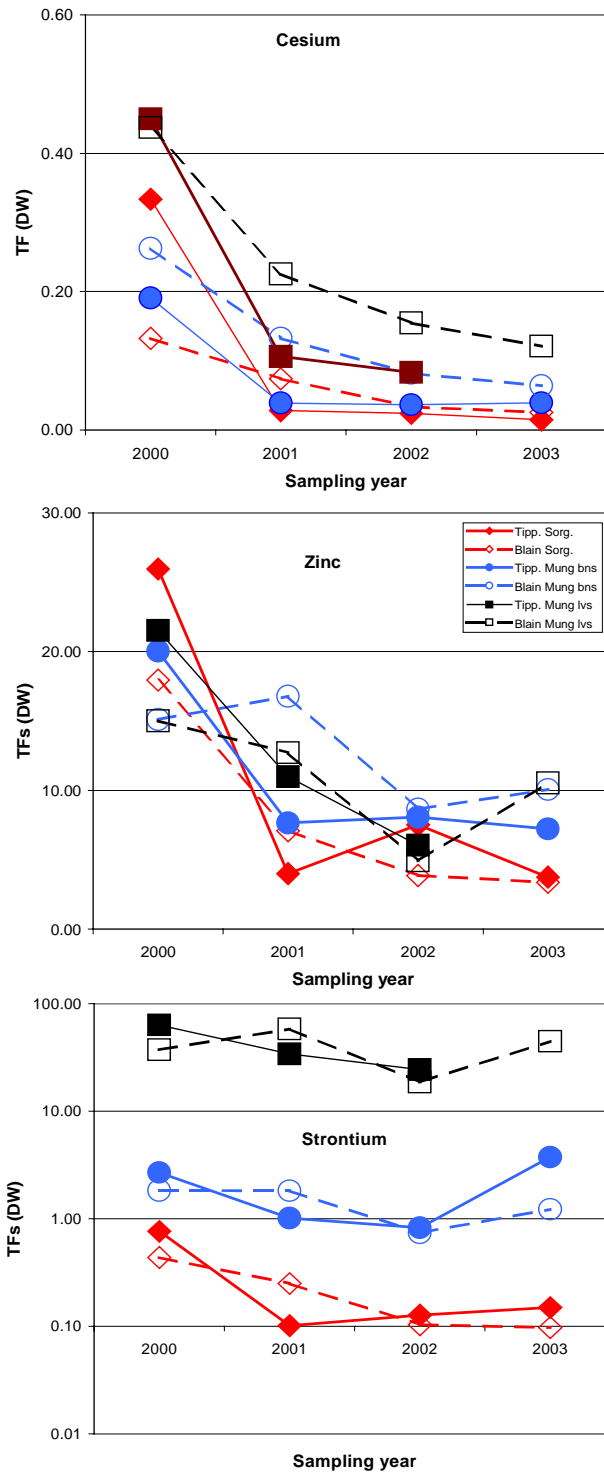
**Table 6.** Average transfer factors for artificial radionuclides between crops and soil (standard deviations in brackets, n = 3) in the samples from 2000 and 2003. The IAEA reference values are included for comparison. The Zn reference data were not specified to a soil type, hence the same reference values have been ascribed to both soils. The last mung leaves from Tippera were collected in 2002.

Soil Crop	BLAIN			TIPPERA		
	<sup>85</sup> Sr	<sup>134</sup> Cs	<sup>65</sup> Zn	<sup>85</sup> Sr	<sup>134</sup> Cs	<sup>65</sup> Zn
<b>Year 1</b>						
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)
<b>Year 4</b>						
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)
<b>Recommended values (95% confidence intervals) IAEA TECDOC 364 (1994)</b>						
	<b>Sand</b>	<b>Sand</b>		<b>Clay</b>	<b>Clay</b>	
<b>Grains</b>	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)
<b>Peas</b>	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)
<b>Leafy vegetable</b>	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)

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 a clay and a sand.

Firstly, 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.

Secondly, 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.



**Figure 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.

Lastly, 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, Figure 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).

### **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  $^{134}\text{Cs}$  compared to  $^{85}\text{Sr}$ . At any specific pH Tippera tended to bind  $^{85}\text{Sr}$  more strongly than did Blain, however, as there was a slight difference in pH between the two soils there was no apparent difference in  $^{85}\text{Sr}$  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.

The study highlighted very low mobility of the  $^{134}\text{Cs}$  and  $^{65}\text{Zn}$  in these soils. There has been no significant decline in the decay-corrected radioactivity in the top 20cm 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  $^{134}\text{Cs}$  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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Appendix A.

Collated data comprising IUR data sheets for TFs and associated information over each year of the study



Data for mung leaves 2000-2003

Sr	V	Mung, leaf	32.9	3.3	C* S	1158	0.5	1.5	21.0	6.2	6.7	0.3	A	F	D	02	10	02	12	03	4	15	99	Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Sr	V	Mung, leaf	44.7	4.5	C* S	1108	0.7	2.2	25.2	6.2	6.6	0.2	A	F	D	02	10	02	12	03	4	15	100	Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Zn	V	Mung, leaf	24.1	2.4	C* L	822	0.7	0.7		4.7	5.4	4.6	A	F	D	99	10	00	1	00	4	15	94	Ferric acrisol	Kaolinite	N, P, K, S added	Tw d3342
Zn	V	Mung, leaf	19.5	1.9	C* L	822	0.7	0.7		4.7	5.4	4.6	A	F	D	99	10	00	1	00	4	15	93	Ferric acrisol	Kaolinite	N, P, K, S added	Tw d3343
Zn	V	Mung, leaf	21.0	2.1	C* L	822	0.7	0.7		4.7	5.4	4.6	A	F	D	99	10	00	1	00	4	15	93	Ferric acrisol	Kaolinite	N, P, K, S added	Tw d3344
Zn	V	Mung, leaf	17.2	1.7	C* S	1486	0.4	0.7		6.1	6.4	2.0	A	F	D	99	10	00	1	00	4	15	92	Ferric acrisol	Kaolinite	N, P, K, S added	Tw d3346
Zn	V	Mung, leaf	11.5	1.2	C* S	1486	0.4	0.7		6.1	6.4	2.0	A	F	D	99	10	00	1	00	4	15	93	Ferric acrisol	Kaolinite	N, P, K, S added	Tw d3347
Zn	V	Mung, leaf	16.3	1.6	C* S	1486	0.4	0.7		6.1	6.4	2.0	A	F	D	99	10	00	1	00	4	15	94	Ferric acrisol	Kaolinite	N, P, K, S added	Tw d3348
Zn	V	Mung, leaf	20.5	2.0	C* L	538	1.8	4.7	38.5	4.6	5.4	1.0	A	F	D	99	10	00	12	01	4	15	93	Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Zn	V	Mung, leaf	45.6	4.6	C* L	260	2.3	4.1	37.5	4.6	5.5	2.2	A	F	D	99	10	00	12	01	4	15	91	Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Zn	V	Mung, leaf	26.9	2.7	C* L	483	3.1	4.6	42.8	4.6	5.4	1.0	A	F	D	99	10	00	12	01	4	15	97	Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Zn	V	Mung, leaf	35.2	3.5	C* S	545	1.3	2.0	19.7	4.8	5.4	0.4	A	F	D	99	10	00	12	01	4	15	93	Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Zn	V	Mung, leaf	30.0	3.0	C* S	489	1.2	2.0	20.3	4.8	5.5	0.4	A	F	D	99	10	00	12	01	4	15	93	Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Zn	V	Mung, leaf	42.4	4.2	C* S	406	1.4	2.9	25.9	5.0	5.7	0.4	A	F	D	99	10	00	12	01	4	15	92	Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Zn	V	Mung, leaf	2.7	0.3	C* L	1840	2.9	4.9	43.5	5.4	6.1	0.8	A	F	D	99	10	01	12	02	4	15	97	Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Zn	V	Mung, leaf	8.6	0.9	C* L	815	2.7	4.4	39.1	5.4	6.0	0.7	A	F	D	99	10	01	12	02	4	15	60	Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Zn	V	Mung, leaf	6.9	0.7	C* L	1035	2.0	4.6	38.3	5.6	6.2	0.8	A	F	D	99	10	01	12	02	4	15	73	Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Zn	V	Mung, leaf	3.9	0.4	C* S	1555	1.5	2.6	24.0	6.0	6.5	0.3	A	F	D	99	10	01	12	02	4	15	86	Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Zn	V	Mung, leaf	4.3	0.4	C* S	1405	1.5	3.3	28.4	6.2	6.7	0.3	A	F	D	99	10	01	12	02	4	15	74	Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Zn	V	Mung, leaf	6.6	0.7	C* S	1359	1.5	2.8	25.0	6.2	6.6	0.3	A	F	D	99	10	01	12	02	4	15	78	Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Zn	V	Mung, leaf	ns		C* L	945	2.3	4.5	43.8	5.4	6.1	0.5	A	F	D	99	10	02	12	03	4	15		Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Zn	V	Mung, leaf	ns		C* L	1330	1.9	3.4	36.4	5.4	6.0	0.5	A	F	D	99	10	02	12	03	4	15		Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Zn	V	Mung, leaf	ns		C* L	1196	1.4	4.4	51.0	5.6	6.2	0.4	A	F	D	99	10	02	12	03	4	15		Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Zn	V	Mung, leaf	12.9	1.3	C* S	773	0.5	1.9	24.3	6.0	6.5	0.2	A	F	D	99	10	02	12	03	4	15	99	Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Zn	V	Mung, leaf	11.3	1.1	C* S	965	0.5	1.5	21.0	6.2	6.7	0.3	A	F	D	99	10	02	12	03	4	15	99	Ferric acrisol	Kaolinite	N, P, K, S added	Tw
Zn	V	Mung, leaf	7.4	0.7	C* S	978	0.7	2.2	25.2	6.2	6.6	0.2	A	F	D	99	10	02	12	03	4	15	100	Ferric acrisol	Kaolinite	N, P, K, S added	Tw

a All counting errors have been propagated through to give the TF st d.

\* Soil type - All soils could be classified in the major class F. Oxides of Si, Al and Fe predominate, comprising approx. 90% of the total mineralogy.

Of these, silicates were the major component and were higher in Blain (CS) (~32%) than in Tippera (CL) (~28%).

Aluminium oxides were the next most abundant but in this case they were more abundant in the Tippera soil (12% cf 8%). Iron oxides were approximately equivalent (~5%) in both soils.

\*\* Conc of nucl in soil is corrected back to date of contamination (columns T&U)

ns - No mung leaves were collected from the Tippera site in 2003.